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Abstract

This document contains a comprehensive literature review in support of the theoretical assessment of the ^{178m2}Hf de-excitation, as well as a rigorous description of controlled energy release from an isomeric nuclear state.

Contents

1 Executive Summary

We have completed a theoretical assessment of the state of science and technology regarding ^{178m2}Hf de-excitation, especially its use as an energetic material for storing energy and the controlled release of that energy. While there are many outstanding nuclear physics questions regarding nuclear isomers, we would assess the prospects for the application of nuclear isomers to energy storage technologies to be poor. The reason for this conclusion has to do with the relative probability of the specific nuclear reactions "releasing" the energy to all possible nuclear reactions. In all the cases studied, the nuclear reactions which "release" the energy are not sustained by the subsequent nuclear reactions, requiring a source external to the assembly to cause the "release." The energy cost of the external source greatly exceeds the energy gained in the "release" for these nuclear isomer systems.

Nuclear isomers exist as a consequence of the "structure" of nuclei, including specific energy levels and the quantum numbers of the quantum mechanical arrangement of the individual nucleons (protons and neutrons) in the nucleus. Some of these quantum numbers are well understood *e.g.* angular momentum. Others like K are known, but the detailed dependence of reaction rates, *etc.* on K is only empirically determined, as no general theory exists. The extent of nuclear isomerism in the table of isotopes is an exploration that is ongoing in experimental nuclear physics. This exploration is not likely to ever be complete given the number of isotopes and the extraordinarily large number of possible nuclear states.

Common to all of these nuclear isomer systems is the fact that the transitions from the isomer states are greatly inhibited, usually forbidden by quantum number conservation. The 31-year half life of ^{178m2}Hf is roughly 10^9 seconds, while an "allowed" nuclear transition would take place in something of order 10^{-22} seconds. The conservation of K is responsible for this stability. This responsibility is not rigid, as the ^{180m}Ta half-life limit is $> 10^{16}$ years. The actual ground state of ^{180m}Ta is unstable, providing a test case for "controlled energy release" because the natural abundance of ^{180m}Ta, while small (0.0012%), makes creating targets for experiments possible. Another feature of ^{180m}Ta is the fact that there are no background radiations due to decay, as there is in the ^{178m2}Hf case, which has a molar decay rate of roughly 10^{15} decays per second (or 3 kCi). By exciting the ^{180m}Ta level with a relatively high energy beam of photons (>1 MeV) with a broad spectrum, it is possible to connect the isomer state to other states which do sometimes decay to the ground state. This so-called *K-mixing* band (a set of states) is inferred to exist where the nuclear level densities are high. Once this happens, and the ^{180}Ta nucleus decays by γ-cascade to the ground state, the decay of the ground state can be observed, verifying the "de-excitation" of the isomer. Since the isomer state is only about 77 keV above the ground state, little additional energy is available. That is, it is necessary to put 1 MeV in, to get out 1.077 MeV, for example.

The actual *K-mixing* mechanism has not been identified explicitly in ^{180m}Ta de-excitation, but is the subject of an ongoing series of experiments. However, the picture that begins to emerge is that any *K-mixing* relevant to isomer decay must occur in states of high rotational energy, such as occur high in the ground state and other known excitation bands, as these have states with the largest angular frequency at energies close to the isomer. The identification of the likely location of *K-mixing* provides important constraints in other nuclear isomer systems.

Another possibility for de-exciting nuclear isomer states is by particle interactions, particularly neutron interactions. Neutron capture and re-emission is one possible nuclear reaction. An isomer like ^{178m2}Hf could capture an energetic neutron, to form a compound nucleus ^{179}Hf in a highly excited state. The *K-mixing* takes place in the compound nucleus, which emits a neutron becoming ^{178m2}Hf again, but now it can also decay to lower states, maybe even the ground state. These type of reactions have been studied in ^{177}Lu where the neutron can be accelerated (emitted with a higher energy than was absorbed) from the isomer de-excitation in the compound state. Assuming that the *K-mixing* is complete in the compound (a plausible theoretical *ansatz*), detailed calculations of the neutron interactions in both ^{177}Lu and ^{178m2}Hf show that while the neutron reaction (n,n$'$) increases with decreasing neutron energy, the competing reaction (n,γ) also increases the same way, with the ratio of the two reactions remaining the same: roughly 1 to 1000 for (n,n$'$) to (n,γ).

Excitation to the *K-mixing* band by electromagnetic transition has been investigated in ^{178m2}Hf in a number of experiments. The theoretical considerations lead to the expectation that the cross section is small for incident x-ray energies < 100 keV. With respect to the reported cross sections, these theoretical expectations are 3 to 6 *orders of magnitude* lower, and in agreement with the limits placed by experiments reporting null effects (that is, not observing any de-excitation). In most of these estimates, work to identify the *K-mixing* mechanism is abandoned, having made only the assumption that it exists.

Largely because of the discrepancy between expectations and observations, the possibility that atomic electrons might play a role was explored. The physical mechanism is referred to as NEET, Nuclear Excitation through Electron Transition. Reactions were explored using very narrow bandwidth beams of x-rays available at synchrotron light sources. The energy regions of the Hf L-shell electrons were scanned with a beam and positive results reported for de-exciting ^{178m2}Hf. The only other system where NEET has been observed is ^{197}Au for which the probability of NEET was measured to be 5×10^{-8} per incident x-ray. Theoretical expectations are 1.3×10^{-7}, within a factor of two, which given the difficulties is a remarkable agreement. The observations of NEET reported for ^{178m2}Hf de-excitation is something like $2{\times}10^{-3}$, many of orders of magnitude greater than typical values calculated for the corresponding transitions ($\leq 10^{-6}$). The possibility of *coherent* effects on the Hf inner shell electrons was explored theoretically but failed to explain the large observed values. Finally, there are a number of other experiments observing null results, in agreement with the calculations.

Though many aspects of the nuclear science related to ^{178m2}Hf and other isomer "de-excitation" mechanisms are not entirely understood, the known theory is able to calculate the rates for the various nuclear reactions to at least a factor of 2 or 3, and within a factor of 10. This theory is consistent with experiments in other nuclei that measure the same kind of reactions. Thus, while "de-excitation" of nuclear isomers had been observed, it is unlikely to be true in the cases that these observations vary greatly with theoretical expectation, especially where other experiments measuring the same rates observe null results.

Setting aside the nuclear physics, the various schemes for utilizing nuclear isomers for energy storage can be further investigated. In the case of nuclear fluorescence reactions, symbolized by the shorthand notation (γ,γ'), it is instructive to consider the condition for

"criticality" to create a self-sustaining reaction (a *chain reaction*). These reactions require that the ratio of the specific (γ,γ') to all reactions, (γ,X) is larger than 1. This condition is not met by many orders of magnitude even given the largest reported cross sections for the nuclear fluorescence reactions.

These reactions in nuclei are complicated by the very narrow width of nuclear states, for which even the very slight motion of the atom at temperatures greater than 1 K is enough to Doppler shift the nuclear level energy away from narrow γ-ray sources. A broad energy band is required, however this presents another problem: the (γ,X) is dominated by photo-ionization in the atomic medium at these energies. The flux of the γ-rays and x-rays causing the "de-excitation" are also producing an electron-ion plasma which have very different transmission characteristics than the initial cold matter.

If, instead, an external source of x-ray is used to "burn" the isomer, the small ratio of (γ,γ') to (γ,X) makes the energy recovery very inefficient. Further, the same material problems of heating the energetic medium with an effectively high black-body temperature source complicates the analysis of such energy storage schemes.

Considering γ-ray laser ("graser") schemes as a variant of x-ray induced energy release from ^{178m2}Hf and other nuclear isomers, the laser "pump" puts so much energy into the material that the conditions required for lasing cannot be maintained. This is also because of the dominance of the x-ray photo-ionization cross sections.

In the case of neutrons, the (n,n$'$) reaction on ^{178m2}Hf could be used to "de-excite" the nuclear isomer. In proposals using a neutron "multiplication" reaction in conjunction, *e.g.* ^{9}Be(n,2n) to construct a "chain reaction" the competing channels, mostly (n,γ) are a few orders of magnitude larger. Consequently, the neutron flux rapidly declines and the "chain reaction" extinguished. In fact, in the particular case of ^{178m2}Hf (n,n$'$) combined with ^{9}Be(n,2n), any concentration of ^{9}Be effectively poisons the "reactivity" of the material.

Once again, if neutrons are used to "burn" the nuclear isomer, the efficiency of the mechanism is very low because of the high energy expense of generating neutrons.

In conclusion, there does not seem to be any realistic scheme to utilize the stored energy represented by the nuclear isomer level, independent of the nuclear science governing the transition of that level to the nuclear ground state. The prospects are much less likely given the understanding of the nuclear physics of this transition.

On a final note, the production of nuclear isomers, even in the small amounts required for research, is a daunting process. The initial discovery of ^{178m2}Hf irradiated 100 mg of HfO_2 for *two years* in a high neutron flux reactor facility and required an additional three years to process, resulting in an estimated 25 picograms of ^{178m2}Hf. Considerations of large scale processing with reactor irradiation conclude that it is impractical to produce even gram quantities in this manner.

Accelerator production is only slightly more efficient. However, estimates are based on notional facilities for which the technologies have not yet been invented. Even in this case, huge capital costs ($\approx$ \$1billion *e.g.* the class of accelerator similar to the Spallation Neutron Source, SNS, at ORNL) would be required to produce grams per year.

Technologies beyond our imagination may exist to harness unique physical systems in

the future, so we cannot conclude that such technologies could not exist. However, the underlying science describing the particular physical systems we have studied in this report, nuclear isomers, seems well understood. This scientific understanding lead us to judge that nuclear isomers are not suitable for the practical applications proposed: energy storage. To the extent that the de-excitation of nuclear isomers can be induced in a controlled manner, it is unlikely that this fact has any practical application.

2 Introduction

This report provides an assessment of the state of research on the nuclear isomer ^{178m2}Hf and the potential for controlled energy release. Among the elements of the assessment are an extensive literature review of the state of nuclear isomer research, a quantitative description of 'controlled energy release' and a description of the state of theoretical and experimental understanding of nuclear isomers, particularly ^{178m2}Hf.

A specific goal of this report was to provide a critical assessment of the claims made in recent Russian literature by Muradian [109] that neutron induced stimulated de-excitation through inelastic scattering produces enhanced decay of the nuclear isomer ^{178m2}Hf.

The energy storage capacity of nuclear isomers is compared with other 'energy storage' systems in Table 1. The highest energy density system is ordinary matter, which if totally converted to energy as in the expression $E = mc^2$ sets the maximum limit of energy density.

Table 1: The energy density of various physical systems.

System	Energy Density (MJ/kg)
mass-energy equivalence	9×10^{10}
d-T fusion	5×10^{8}
^{235}U fission	8×10^{7}
^{64}Zn(n,γ)	3×10^{7}
^{178m2}Hf de-excitation	3×10^{6}
Radioisotope thermoelectric generator	1×10^{5}
hydrogen combustion	1×10^{2}
propane	5×10^{1}
gasoline	5×10^{1}
body fat	4×10^{1}
wood	2×10^{1}
TNT	4×10^{0}
Li Battery	1×10^{0}
lead-acid battery	1×10^{-1}

Nuclear fusion and fission both have large energy density, followed in the list by neutron capture on specific nuclei (such as happens in supernova explosions). The de-excitation of ^{178m2}Hf to the ground state of ^{178}Hf is shown in comparison.

The interest in nuclear processes for energy storage is the fact that the 'energy densities' of such systems are 4 to 6 *orders of magnitude* larger than chemical systems. Practical energy producing systems have been realized using nuclear fission and the heat generated from nuclear decay.

Nuclear fission works because of the nuclear properties of the excited nucleus ^{236}U, which is formed by the capture of a neutron on ^{235}U. The subsequent fission of ^{236}U produces multiple neutrons along with the decay daughter nuclei. A property of ^{235}U is that the

probability of capturing a neutron is high, and does not vary over the energy spectrum of the neutrons, at least in the range of neutron energies produced in the fission. Thus it is possible to arrange a concentration of ^{235}U such that the *net* number of neutrons produced *increases* in time through multiple captures, fissions and neutron production: the so-called 'chain reaction.' Large energy release is possible here because the system will 'run away' exponentially if the neutrons are not taken out of the chain of reactions. Controlled energy release is possible because the reaction can be controlled by regulating the number of removed neutrons.

This example is instructive because it illustrates the requirements of an energy release system with a practical realization as a power source. The system is designed so that neutron production balances neutron loss. Once this is done, the system is configured to minimize the loss of neutrons through diffusion. In an unregulated system, the reaction runs away until the energy produced by the sum of the reactions destroys the conditions required to sustain the reaction. However, introducing elements in the system to regulate the number of neutrons provides a means of controlling the energy release.

A similar scenario must exist for all means of controlled release of energy stored in nuclear states.

This report first reviews the physics of isomers, particularly ^{178m2}Hf (see Section 3). The reactions leading to a transition of the nuclear isomer state to some lower energy state can proceed through a number of channels. In Section 4, we review the possibilities of de-exciting the isomer via reactions with photons or particles such as neutrons, as well as via electric interactions. The physics of controlled and explosive energy release is developed in Sections 5.1–5.2. In Section 5.3, we explore the criticality conditions of admixtures of ^{178m2}Hf and ^{9}Be with a Monte-Carlo simulation; such a mixed system was proposed by Muradian [109] as a possible avenue for achieving a self-sustaining process of energy release from an isomeric state.

Because some of the experimental results on ^{178m2}Hf are in disagreement with theoretical expectations from nuclear and atomic physics, we have presented the analysis of experiments on other nuclear isomers in comparison with theoretical calculation. This provides a test of the degree to which theory can explain the various physical phenomena relevant to ^{178m2}Hf.

3 The physics of hafnium isomers

In this section, we review the nuclear structure ingredients that lead to nuclear isomers in general and to the isomer of interest, ^{178m2}Hf, in particular. We explain the physics reasons leading to the remarkably long life time of ^{178m2}Hf and discuss the structural changes that are required to de-excite this state. We summarize the present knowledge of the structure of ^{178}Hf and provide an overview over the research contributions from the past 40 years that have resulted in a well-established and fairly comprehensive picture of the structure of this nucleus.

3.1 Basics of nuclear isomers

3.1.1 Classification of isomers

Atomic nuclei, like all quantum systems, when excited, will eventually decay to the lowest energy configuration. An isomer (or isomeric state) is an excited state in the nucleus whose decay is generally suppressed (most often due to nuclear structure effects) leading to an excited system with an uncharacteristically long lifetime. For systems where the total excitation energy is below the threshold for particle emission, decay generally proceeds through a chain of electromagnetic transitions until the ground-state configuration is reached. Typically, electromagnetic transitions between nuclear states occur quickly, with half-lives, $t_{1/2}$, of the order of picoseconds (1 ps = 10^{-12}s). However, for some excited nuclear states, their structure inhibits normal electromagnetic-transition mechanisms, slowing down the decay considerably. While experimental constraints generally establish $t_{1/2}$= 1 ns (1 nanosecond = 10^{-9}s) as an effective lower lifetime limit for the purpose of defining an isomeric state, many isomers have much longer lifetimes. For example, ^{180m}Ta has a half-life of over 10^{15} years. The isotope detailed in this report, hafnium-178 (^{178}Hf), has several isomeric states: ^{178m1}Hf at 1.147 MeV with a lifetime of 4s and ^{178m2}Hf at 2.446 MeV with a lifetime of 31 years (here $m1$, $m2$, ... denote the first, second, etc., metastable states of the isotope under consideration). While electromagnetic processes, such as γ-ray emission and internal conversion are still the most common decay mechanisms for isomers, their long lifetimes may permit the state to decay through other slower channels, such as α-emission, β-decay, and spontaneous fission (see Walker and Dracoulis [155]).

The enhanced lifetime of isomeric states is generally due to the fact that the electromagnetic decay of the excited state requires a significant change in shape, spin, and/or spin orientation, thus suppressing the decay. According to the mechanism that hinders their decay, isomers are classified into *shape isomers*, *spin isomers*, and *K-isomers*.

Shape isomers occur when the surface that determines the energy as a function of nuclear deformation for a given isotope exhibits not only a primary minimum, which corresponds to the ground-state configuration, but also a secondary minimum, which often leads to an isomeric state of different deformation. In this case, the energy barrier between the two minima leads to distinct and disconnected configurations, where the de-excitation of the excited state through gamma-decay is highly suppressed. An example of such a shape isomer is the 14 ms (1 millisecond = 10^{-3}s) state ^{242m}Am, which has an energy of 2.2 MeV and

decays by spontaneous fission.

Spin isomers occur when the decay of the metastable state requires gamma radiation of high multipolarity λ[1], where λ also determines the maximum change in the nuclear spin (also referred to as angular momentum) that is permitted in the transition. Typical electromagnetic decays, with lifetimes of the order of picoseconds, occur with $\lambda = 1$ or 2. In general, since large changes in the intrinsic structure are required, high multipolarity decreases the decay rate. In addition, the decay rate is also determined by the gamma-ray energy, E_γ, and is proportional to $E_\gamma^{2\lambda+1}$. A combination of high multipolarity and low transition energy leads to a highly suppressed decay probability, and, thus, a long lifetime. Typically, spin isomers occur when the only option for the decay of a high-spin state is a transition to lower states that require a change in spin greater than three units. For instance, the ^{180m}Ta state mentioned above, has spin $J = 9\hbar$ and lies only 75 KeV above the $J = 1\hbar$ Ta ground state. The decay by gamma emission requires a spin change of $8\hbar$, which is highly suppressed; this, in combination with the small energy difference between the states accounts for the isomer's anomalously long lifetime.

K-isomers can occur in axially-symmetric deformed nuclei. Such nuclei (most of which are prolate, *i.e.* cigar shaped), can undergo collective rotation about an axis [see A. Bohr and B.R. Mottelson, *Nuclear Structure* (World Scientific, Singapore, 1998), vol. 2]. For even-even nuclei, such as ^{178}Hf, the starting point is for all the constituent nucleons to couple their intrinsic spins to a total spin of zero. Quantum-mechanically, collective rotation about an axis of symmetry is forbidden, and, thus, the collective nuclear spin is perpendicular to the symmetry axis. The energy spectrum for collective rotation follows the simple pattern $E = J(J+1)/2\mathcal{I}$, where $\mathcal{I}$ is the moment of inertia and only even values for J are allowed, *i.e.* $J = 0, 2, 4, ...$ The nucleus, however, is not a rigid object, and it is possible for the constituent particles to realign, giving an intrinsic spin that then couples with the collective rotational motion. In this case, the intrinsic spin, $\vec{K}$, is aligned along the symmetry axis; it is added vectorialy to the collective spin, $\vec{I}$, to produce the total angular momentum vector $\vec{J}$. The energy spectrum in this case follows the pattern $E = [J(J+1) - K^2]/2\mathcal{I}$, where J now assumes all values starting from K. For a given projection K of the total angular momentum $\vec{J}$ onto the symmetry axis, a set of levels with $J = K,\ K+1,\ K+2, \ldots$ exists. Each set is characterized by strong electromagnetic transitions among its members and is referred to as a K band. There are two commonly occurring mechanisms for generating K bands. The first involves an intrinsic motion caused by a surface vibration that distorts the nuclear quadrupole shape; this vibration has $K = 2$. The second mechanism is the one that is responsible for the long-lived isomers in ^{178}Hf: individual single-particles detach from the collective rotation and couple their intrinsic spins (with amplitude K) to the angular-momentum vector of the collective motion. Overall, the projection K is very nearly conserved and introduces an additional selection rule: electromagnetic transitions between states of different K are allowed when the change in K is less or equal to the multipolarity λ of the transition: $\Delta K \equiv |K_{final} - K_{initial}| \leq \lambda$. In addition, due to the fact that changing K

[1]Electromagnetic transitions are characterized by two quantities. First, is their type: electric (E), which involves changes in the charge distribution, and magnetic (M), which involves changes in the internal distribution of magnetic moments. Second, is their multipolarity λ. The labeling convention is then $E\lambda$ and $M\lambda$. Thus, an electric-quadrupole transition is labeled as $E2$, while a magnetic-dipole transition is $M1$.

requires a realignment of individual particles or the dissipation of shape vibrations coupled to the collective motion, transitions that change K, even if allowed, tend to be suppressed relative to transitions within the K-band, *i.e.* those with $\Delta K = 0$. For example, often $\lambda = 2$ transitions within the γ-band ($\Delta K = 2 - 2 = 0$) are preferred relative to transitions into the ground-state band ($\Delta K = 2 - 0 = 2$). Transitions that violate the K-selection rule are called *K-forbidden*. However, since K is approximately conserved, such transitions are only severely hindered, rather than strictly forbidden. Excited states of axially-symmetric deformed nuclei that require K-forbidden transitions in order to decay can be classified as K-isomers. The states ^{178m1}Hf and ^{178m2}Hf mentioned above are such K-isomers, with K=8 and K=16, respectively (the ground-state band of ^{178}Hf has K=0).

A classical picture that illustrates K alignment and its conservation is the precession observed in a rotating symmetric top, such as a gyroscope or the Earth. The top is spinning about a symmetry axis, which is tilted. To an observer sitting on the top, it is spinning at a constant rate about its symmetry axis, which is analogous to K in the nuclear system. In the external reference frame, the tilted spinning top then precesses about a vertical axis. The two angular momenta then add to the total angular momentum, which is aligned with a vertical axis.

3.1.2 K mixing and the decay of K isomers

The fact that K isomers can decay by K-forbidden transitions indicates that the initial and/or final states contain some mixture of K values. A measure of the goodness of the K quantum number is the *reduced hindrance*, $f_\nu \equiv (T^\gamma_{1/2}/T^W_{1/2})^{1/\nu}$, where $T^\gamma_{1/2}$ is the partial γ-ray half life of the transition, $T^W_{1/2}$ is the corresponding Weisskopf single-particle estimate, and $\nu \equiv |\Delta K| - \lambda$ is the degree of forbiddenness [155]. The quantity f_ν provides insight into how the hindrance scales with the degree of forbiddenness. An early study by Löbner [100] found $f_\nu \approx 100$ to be a reasonable value. While this value is often used as a rule of thumb, subtle nuclear structure effects, which are responsible for K mixing in the states under consideration can cause large variations in f_ν. Several mechanism are known to induce K mixing in nuclear states: (i) Coriolis effects, which induce an alignment of the orbital angular momenta of the individual nucleons with the angular momentum characterizing the collective rotation of the nucleus; (ii) small admixtures of triaxial shapes in the wave function, and (iii) statistical mixing with neighboring levels in regions of high level density. An important aspect of all three of these mechanisms is that mixing will occur only if the states that are being mixed are relatively close in excitation. This is largely deduced from first-order perturbation theory where the mixing amplitude between two states is proportional to $V/\Delta E$, where V is the interaction matrix element (of the fundamental nuclear Hamiltonian) between the two states and ΔE is their energy difference. In general, K mixing becomes more prominent at higher excitation energies where the density of states is high and states of different K can be nearly degenerate. Past experiments have examined all three mechanisms to analyze K-forbidden transitions and to understand the systematic behavior of observed reduced hindrance factors (For more details, see the articles by P. Walker [154, 155] and the thesis of Gareth Jones [73]).

3.2 The structure of ^{178}Hf

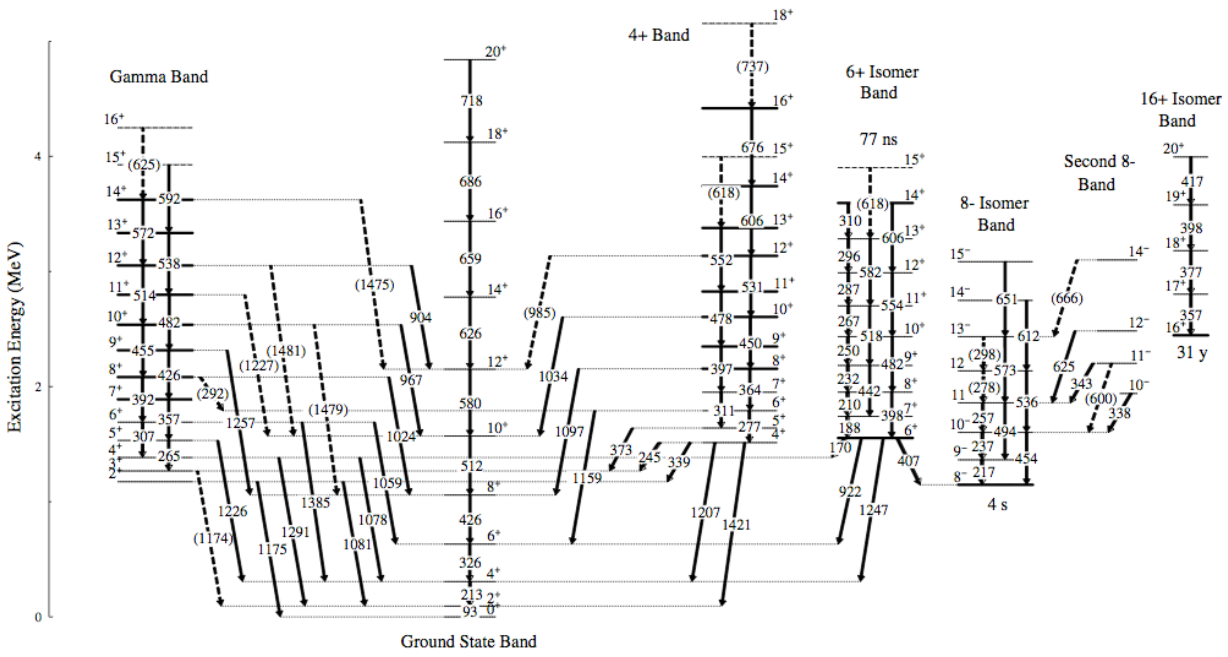

Figure 1: Levels and decay scheme for ^{178}Hf showing the observed band structure (from Ref. [69])

To date, 134 discrete levels up to an excitation energy of 5.388 MeV have been identified in ^{178}Hf. Shown in Fig. 1 is a level diagram showing most of the levels with spin and parity and their excitation energy. In addition, the decay paths (arrows) are also shown with the gamma-ray energies tabulated. The decay scheme outlines the structure of seven bands, including the ground-state band, which exhibits the typical structure of a rotating deformed nucleus, i.e., even spins with a quadratic dependence in the excitation energy on spin as well as a strong de-excitation exclusively within the band. Also typical is the γ-band shown to the left; it has $K = 2$ and exhibits a characteristic set of rotational levels built on top of a 2^+ bandhead state that is associated with an intrinsic surface vibration along the quadrupole gamma degree of freedom. Shown in the right side of the figure are several bands built on the quasiparticle excitations of the ground state. There are two low-lying $K = 8^-$ bands built on neutron and proton two-quasiparticle states, respectively, as well as two other two-quasiparticle $K = 4^+$ and $K = 6^+$ bands. The 4 s 8^- state (the lowest two-quasiparticle state) preferentially decays by K-forbidden electric-dipole ($\lambda = 1$) emission to the 8^+ state in the ground-state band at 1.059 MeV. Additionally, there is the very long-lived four-quasiparticle $K = 16^+$ band, which is an excitation constructed by combining both of the lower $K = 8^-$ two-quasiparticle excitations. The decay of the 16^+ state is highly suppressed (with a 31 yr half-life) not only because a change of K by at least 8 units is required, but also because the transition has to have a high multipolarity λ, due to large angular-momentum differences between the initial ($J = 16^+$) and energetically feasible final states. Electromagnetic decay of this state occurs to three levels in the $K = 8^-$ isomer band,

Table 2: Multi-quasiparticle structure of isomeric states in ^{178}Hf. Shown are the K^π=J^π quantum numbers of several isomeric states in ^{178}Hf, their energies, life times, and quasiparticle structure, deduced from experimental and theoretical observations. The K^π=6^+ state can be explained as a two-quasiparticle proton excitation, while the K^π=8^- isomer is a mixed proton-neutron two-quasiparticle state. The K^π=14^- and K^π=16^+ states, which are at higher energy, are 4-quasiparticle states. For details, see References [15, 108, 44, 66].

K^π	E [MeV]	Life time	Quasiparticle structure
6^+	1.554	78 ns	$p(5/2^+[402])p(7/2^+[404])$
8^-	1.147	4 s	$\alpha p(7/2^+[404])p(9/2^-[514]) + \beta n(9/2^+[624])n(7/2^-[514])$
14^-	2.574	68 μs	$p(5/2^+[402])p(7/2^+[404])n(9/2^+[624])n(7/2^-[514])$
16^+	2.446	31 y	$p(7/2^+[404])p(9/2^-[514])n(9/2^+[624])n(7/2^-[514])$

First, there is the electric $\lambda = 3$ transition to the 13^- state with a transition energy of only 12.7 keV. The low transition energy not only significantly suppresses the decay, but also causes the transition to occur via internal conversion, rather than gamma emission. The two other γ-decay modes that have been identified are the magnetic $\lambda = 4$ transition to the 12^- state in the 8^- band with a γ-ray energy of 309.5 keV, and the electric $\lambda = 5$ decay to the 11^- state with a transition energy of 587 keV. Overall, the combination of the high multi-polarity and change in K by eight units accounts for the extraordinarily long half-life of the 16^+ state.

3.2.1 Quasiparticle excitations and high-spin isomers in the hafnium region

Deformed shell-model calculations can predict energies for single-particle (neutron or proton) orbitals as a function of nuclear deformation. The orbitals are generally labeled by their *asymptotic Nilsson quantum numbers* $\Omega^\pi[Nn_z\Lambda]$, where Ω denotes the single-nucleon spin projection on the symmetry axis (here taken to be the z-axis), π is the parity of the state, N is the principal quantum number associated with the major shell to which the orbital belongs, n_z gives the number of oscillator quanta in the z-direction (and thus the number of nodes in the wave function along the z axis), and Λ is the projection of the single-particle orbital angular momentum onto the z-axis. For the well-deformed hafnium isotopes, such calculations find that orbits with large values for Ω cluster around the Fermi level [163]. These high-Ω orbitals play an important role in explaining the existence of isomeric states in the hafnium region: When neutron or proton pairs are broken, these orbitals can be occupied and nuclear states with large K values (resulting from a combination of the large single-particle Ω values) and long lifetimes are produced.

The configurations that are obtained by breaking nucleon pairs and occupying excited

single-nucleon orbitals are referred to as *quasiparticle excitations*. For ^{178}Hf, several high-Ω orbitals have been identified as building blocks for quasiparticle states: $\Omega^{\pi}[Nn_z\Lambda]$ = $7/2^+[633]$, $5/2^+[512]$, $7/2^-[514]$, $9/2^+[624]$, $9/2^-[505]$, and $11/2^+[615]$ neutron orbits cluster around the Fermi level for quadrupole deformations around β(neutrons)=0.2; the relevant proton orbits for β(protons)$\approx$0.25 are $\Omega^{\pi}[Nn_z\Lambda] = 5/2^+[402]$, $7/2^+[404]$, $9/2^-[514]$. Isomeric states in ^{178}Hf have been explained in terms of 2-quasiparticle or 4-quasiparticle states that can be constructed from these single-nucleon states, as Table 2 indicates.

Rotational bands, built on the quasiparticle configurations, have been identified. Since quasiparticles in large-Ω orbits tend to resist the rotational alignment of their angular momenta induced by the Coriolis force, high-K isomeric bands are expected to have good K values. Experiments testing the K-band mixing of the rotational bands in ^{178}Hf seem to confirm this [68, 66] (also: see below).

The location of the deformed single-particle states with large Ω-values close to the Fermi level is responsible for the extremely low value of the excitation energy of the 4-quasiparticle state that is known as the ^{178m2}Hf isomer. The fact that it lies below any states of spin 14 or higher prevents it from decaying via a low-mulitpole gamma transition and contributes directly to the remarkably long lifetime of the isomer.

More generally, the hafnium region near A=178, with its well-deformed, axially symmetric isotopes, provides particularly favorable conditions for the existence of K isomers. Multi-quasiparticle isomers have been found there systematically and calculations predict the existence of further isomeric states with very large K values [155, 163].

3.3 Summary of key publications on the structure of ^{178}Hf

Over the forty years or so since the discovery of the 31-year ^{178m2}Hf isomer, a wealth of information about the properties of ^{178}Hf and its isomers have been obtained from a wide variety of experiments, which has also lead to theoretical interpretations. For the most part, the experiments have focused on tracking the complex network of γ-transitions to build a detailed picture of the level structure. Structure information has been obtained from the β-decay of the ground-state and isomeric states of ^{178}Ta, neutron capture on ^{177}Hf, and Coulomb excitation to high-spin states in heavy-ion experiments. In addition, laser-spectroscopy has been employed to measure static properties, such as the charge radius and electric quadrupole and magnetic dipole moments of the isomer. This section summarizes some of the principal works that have shed light onto the structure of ^{178}Hf .

Decay of an isomeric state in ^{178}Hf with $K \geq 16$; Helmer and Reich (1968) [70]. The first observation of the ^{178m2}Hf state was reported in 1968, when Helmer and Reich [70] published their observation of a long-lived isomeric state in ^{178}Hf at about 2.5 MeV excitation energy. The state (^{178m2}Hf) was produced by neutron irradiation of hafnium oxide of natural isotopic abundance. Chemical purification and isotopic separation of the irradiated samples allowed the authors to identify the observed transitions with the relevant isotopes. A decay scheme was proposed for ^{178m2}Hf and a conservative lower limit of 10 years was placed on the half-life of the isomer. The observed decay pattern, half-live and energy considerations, and an examination of the arrangement of single-particle configurations in the mass region led to

the conclusion that the isomeric state had to be one of very high intrinsic spin, with the most reasonable configuration being a 4-quasiparticle state with $K^\pi = 16^+$, which predominantly decays via a 5-times K-forbidden, highly electron-converted, E3 transition to the $I = 13$ member of the $K^\pi = 8^-$ band (and an alternative, less likely, configuration being a 4-quasiparticle state with $K^\pi = 17^+$, which decays via a M4 transition to the $I = 13$ member of the $K^\pi = 8^-$ band).

Half life of ^{178m2}Hf and its neutron capture production; Helmer and Reich (1973)[71]. In 1973, the half life of the ^{178m2}Hf isomeric state was determined by the same authors [71]. They compared γ-ray spectra of ^{178m2}Hf samples, produced via neutron irradiation of ^{177}Hf, taken over the course of six years and determined the half life of ^{178m2}Hf to be 31$\pm$1 years.

The four quasiparticle ^{178}Hf isomeric state; De Boer et al. (1976)[15]. The energy of the isomeric state was determined more precisely by De Boer *et al.* [15] in 1976. The spin, parity, and K-value of the isomer were were established by measuring conversion lines of its de-exciting transitions; the level scheme was verified by measuring γ-rays in single and coincidence experiments. They found the excitation energy of the isomeric state to be 2.4475$\pm$0.0025 MeV, determined the spin and K-quantum numbers as $(I^\pi, K) = 16^+, 6$, and assigned the four-quasiparticle configuration $p(7/2^+[404])$ $p(9/2^-[514])$ $n(9/2^+[624])$ $n(7/2^-[514])$ to the level. Decay of the isomer via a 5-times K-forbidden E3 transition was found to be in agreement with findings for the decay of other isomers.

Structural changes in the yrast states of ^{178}Hf; Khoo and Lovhoiden (1977)[90]. Further evidence for the quasiparticle nature of several rotational bands in ^{178}Hf was presented by Khoo and Lovhoiden in 1977 [90]. The ^{176}Yb$(\alpha, 2n)^{178}$Hf reaction was employed to produce the isotope of interest and γ-ray spectra and angular distributions were observed. The known level scheme of ^{178}Hf was enhanced and probable structures for the band heads in terms of quasiparticle configurations were identified. The K=16 isomer was singled out as a particularly striking example of an *yrast trap*, a high-spin state lying so low in energy that its decay is strongly hindered.

K-forbidden decays in ^{178}Hf; Van Klinken et al. (1980) [94]. In 1980, Van Klinken *et al.* [94] studied K-forbidden decay modes in ^{178}Hf. They carried out electron and γ-ray spectroscopy and identified an M4 branch in the decay of the ^{178m2}Hf isomer, which allowed them to place the ^{178m2}Hf isomer at an excitation energy of 2.4460 keV. They also determined hindrance factors for several K-forbidden transitions and compared them to similar transitions in neighboring nuclei. The dominant decay mode of the ^{178m2}Hf isomeric state, by an $E3$ transition to the 13^- state of the $K^\pi = 8^-$ band was found to have a hindrance factor of 66 per unit of forbiddeness, and the $M4$ transition to the 12^- state of the band was found to have a hindrance factor of 64. Decays from members of the $K^\pi = 8^-$ band to the ground state band ($K^\pi = 0^+$) were studied as well.

Isomeric trapping following Coulomb excitation of high spin states in ^{178}Hf; Hamilton et al. (1982)[61]. Hamilton *et al.* [61] were the first to populate the 4s $K = 8^-$ isomer at 1147.4 keV in ^{178}Hf in a Coulomb-excitation experiment that was designed to investigate band crossing effects for $K = 0^+$ bands. The observation of transitions in the $K = 8^-$ band was unexpected, as Coulomb excitation primarily populates states that are connected to the ground-state band by collective electric quadrupole (E2) transitions and no branching from

members of the ground-state band to the $K = 8^-$ was observed in the experiment. The mechanism for populating this isomer could not be explained.

Study of the low-lying states in ^{178}Hf through the (n,γ) reactions; Hague et al. (1986) [60]. Hague *et al.* [60] carried out a detailed study of the structure of ^{178}Hf below about 2.1 MeV employing thermal- and resonance-neutron capture processes. They constructed an extensive level scheme, containing 65 levels, most of which were ordered into rotational bands, and provided information on the location and intensity of a very large number of γ-transitions and branching ratios.

Coulomb excitation of the $K^\pi = 8^-$ isomer in ^{178}Hf; Xie et al. (1993) [161]. This work investigated the reaction mechanism for populating the $K^\pi = 8^-$ isomer in ^{178}Hf using Coulomb excitation. The experiment used a ^{178}Hf target bombarded by a ^{130}Te beam at beam energies of 560, 590, and 620 MeV. Both prompt and delayed γ-rays were analyzed using the Crystal Ball. The 4s $K^\pi = 8^-$ isomer at 1.1474 MeV was populated with a cross section reported to be $2.7\pm^{1.9}_{1.4}$, $4.3\pm^{3.4}_{2.0}$, and $7.5^{6.1}_{3.2}$ mb for each of the three bombarding energies, respectively. Analysis showed that the population of the $K^\pi = 8^-$ isomer could be explained by a direct excitation with an electric $\lambda = 3$ transition from excited states in the ground-state band to odd-spin states in the $K^\pi = 8^-$ band. However, this experiment could not rule out other mechanisms such as Coulomb excitation of high-spin states in the ground-state band, with higher K-mixing, followed by γ-decay into states in the $K^\pi = 8^-$ band.

Nuclear properties of the exotic high-spin isomer ^{178m2}Hf from collinear Laser Spectroscopy; Boos et al. (1994) [16]. This paper examines the hyperfine spectrum in atomic transitions in the optical spectrum to determine nuclear properties of the 31-yr ^{178}Hf isomer. By measuring small shifts in the hyperfine spectrum where the hafnium nucleus was in the ground-state configuration or the 31-yr isomer they performed a precise determination of the difference in the mean-charge radius between the ground state and the isomer ($\delta\langle r^2\rangle = -0.059(9)$ fm^2) as well as the magnitude and sign of quadrupole moment ($Q^{178m2} = +6.00(7)$ b) and the magnetic dipole moment ($\mu^{178m2} = +8.16(4)$ nuclear magnetons) for the isomer.

Rational band on the 31 year 16^+ isomer in ^{178}Hf; Mullins et al. (1997) [108]. Mullins *et al.* [108] employed an incomplete fusion reaction in order to populate high-spin states in several hafnium isotopes and study their decay via electromagnetic transitions. The authors were able to extend previously known bands in ^{177,178,179}Hf. In ^{178}Hf, they added a $J^\pi = 18^+$ and a tentative $J^\pi = 20^+$ level to the ground state band and measured rotational bands built on the 16^+ ^{178m2}Hf isomer and the 14^- isomer at 2.574 MeV. They presented further evidence for the four-quasiparticle structure of ^{178m2}Hf and for the two-quasiparticle structure of the two lower lying 8^- states.

Limit to high-spin isomerism in hafnium isotopes; Xu et al. (2000) [163]. This paper presents a theoretical examination of the properties of hafnium isotopes to determine the potential existence of high-spin isomers. The authors achieved a realistic treatment of multi-quasiparticle states by performing configuration-constrained calculations of the potential energy surface within a deformed Woods-Saxon basis with a Lipkin-Nogami treatment for pairing and the Stutinsky averaging method. Calculations were performed for a variety of

hafnium isotopes. Overall good agreement was achieved in describing the $K^\pi = 16^+$ isomer in ^{178}Hf. Further calculations were reported for ^{182}Hf and ^{186}Hf to investigate the lowest spin occurring for three distinct types of excitation to generate high spin: 1) prolate collective rotations, 2) oblate collective rotations, and 3) multi-quasiparticle excitations. They report that with increasing neutron number, the multi-quasiparticle states become yrast (*i.e.* they become the lowest states of a given angular-momentum value in the spectrum of the nucleus) at higher angular momentum, leading to the possible existence of high K isomers. In general, the multi-quasiparticle states are energetically favored over oblate collective rotations up to spins of $35\hbar$.

Gamma rays emitted in the decay of 31-yr ^{178m2}Hf ; Smith et al. (2003) [137]. Smith *et al.* investigated the spontaneous decay of the ^{178m2}Hf isomer by coincidence γ-ray spectroscopy. They were able to observe direct, high-multipolarity, strongly K-forbidden, M4 and E5 γ-ray emission associated with the transition from the 16^+ isomer to the 12^- and 11^- states, respectively, in the rotational band built on the $K^\pi = 8^-$ 4s isomer. They also measured low-intensity transitions between members of the two $K^\pi = 8^-$ bands, which allowed them to obtain an estimate of the mixing strength between the two bands. They determined reduced hindrance factors for transitions between the K^π=16^+ and K^π=8^+ bands to be of the order of about 100, in agreement with earlier systematics and measurements. They were not able to detect any direct γ-ray transition to the ground-state band; nor did they observe the 129 keV line that both Collins *et al.* [37] and Rusu [131] had reported earlier as associated with stimulated decay of ^{178m2}Hf.

Coulomb excitation paths of high-K isomer bands in ^{178}Hf; Hayes et al. (2002) [69]. Hayes *et al.* carried out Coulomb excitation experiments with a 650 MeV ^{136}Xe beam impinging on an enriched ^{178}Hf target. They observed prompt γ-ray yields of high spin states in the rotational bands built on on the K^π=6^+ (77 ns), K^π=8^- (4s) and K^π=16^+ (31 y) isomeric states in ^{178}Hf. They employed different models for K-mixing in the rotational bands in order to fit the measured γ-yields and extract information on the mechanisms for populating the different rotational bands. They concluded that three different mechanisms are responsible for populating the isomer bands: The K^π=6^+ appears to be primarily populated by multistep allowed and forbidden excitations through the γ and 4^+ bands, while the K^π=8^- band appears to be populated via E3 excitation from the ground-state band. While Coulomb excitation of the high-K band built on ^{178m2}Hf is in apparent violation of the K-conservation rule, Hayes *et al.* observed the population of that band, as had Hamilton *et al.* [61] and Xie *et al.* [161] in earlier experiments. Hayes *et al.* deduce that the population of this band occurs via direct feeding from the ground-state band, but at very high levels. They conclude that K mixing must increase rapidly with increasing spin.

Study of Isomers using Reactions with a ^{178}Hf beam - Ph.D. Thesis by Gareth Jones, Surrey (2006) [73]. The objective of this thesis work was to search for theoretically predicted high-K multi-quasiparticle states in ^{177,178}Hf and to examine their decays. Blocked BCS calculations, which have successfully reproduced the energies of experimentally observed multi-quasiparticle states in the hafnium region motivated and guided the search. Deep-inelastic collisions between 1.15 GeV ^{178}Hf projectiles and the nuclei in a thick ^{208}Pb target produced a range of hafnium isomers, populated to high spins. The γ-rays from the decays of the excited nuclei were detected with the GAMMASPHERE array. Measured transitions and

half lives for ^{177,178,179}Hf were found to be consistent with previous measurements. A new, weak M3 decay branch for a known isomer in ^{177}Hf was discovered, but the predicted high-K states were not identified. The $K^\pi = 19^+$ and 22^- 6-quasiparticle states predicted to exist in ^{178}Hf, were not observed either, despite a careful consideration and search for possible decay paths of such states. The absence of experimental evidence for the predicted states could not be sufficiently explained. Possible explanations for the outcome of the experiment included limited accuracy of the predicted energies, difficulties in the experimental identification of low-multiplicity γ cascades by which the states may decay, the possibility of very long life times (> 1 yr) of the predicted states, or some combination of these factors. The thesis work also identified new spin-trap isomers in antimony and molybdenum isotopes, produced via fusion-fission reactions of the 1.15 GeV ^{178}Hf beam with an ^{27}Al target.

Breakdown of K selection in ^{178}Hf; Hayes et al. (2006) [68]. Hayes *et al.* carried out an activation experiment to measure Coulomb excitation of the ^{178m2}Hf isomer as a function of collision energy. They irradiated natural Ta targets with a ^{178}Hf^{24+} beam at safe Coulomb excitation energies, collected the scattered Hf ions, and counted the activities five months later. These measurements, in conjunction with a new analysis of their earlier ^{178}Hf(^{136}Xe,^{136}Xe)^{178}Hf Coulomb excitation experiment, were employed to probe the goodness of the K quantum number as a function of spin. The authors observed a systematic decrease with increasing spin of the hindrance of K-forbidden transitions from the ground state band to rotational bands with $K \geq 4$. The rapid breakdown of the goodness of the K quantum number as low-K bands are excited to higher rotational levels indicated that higher-K components are admixed in these bands with increasing spin. The observed (large) hindrance factors for the decays of the high-K isomer bands, on the other hand, indicate that high-K bands remain very pure, even for states at higher spin.

Spin dependence of K mixing, strong configuration mixing, and electromagnetic properties of ^{178}Hf; Hayes et al. (2007) [66]. This paper is a long article that details the work summarized in the earlier Physical Review Letter [68] by the same authors (see *Breakdown of K selection in ^{178}Hf*, above). The purpose of the work was to shed light on the K-forbidden population of the K=8$^-$ isomer that had been seen in early Coulomb excitation experiments [61, 161] and to investigate in detail the unexpectedly large population of the K^π=16$^+$ band the authors had observed in their previous ^{178}Hf(^{136}Xe,^{136}Xe)^{178}Hf Coulomb excitation experiment. A re-analysis of that experiment, in conjunction with activation data obtained from counting the decays of ^{178m2}Hf, produced via irradiation of natural Ta targets with a ^{178}Hf^{24+} beam, made it possible to determine a set of matrix elements for electromagnetic operators causing the transitions between the bands. From this, the authors were able to identify three distinctly different paths for populating the dominant rotational bands in Coulomb excitation of ^{178}Hf, and to infer information about the amount of K mixing in the different bands, as a function of spin. The authors concluded that the K^π=6$^+$ isomer band is populated via a multi-step process, while the K^π=8$^-$ bands are excited directly from the ground state band and the γ band by highly K-forbidden $E3$ transitions. The K^π=16$^+$ isomer was found to be populated. No evidence of direct excitation from the ground-state band to the $J^\pi = 16^+$ isomer state is reported nor substantiated by the data. The exact mechanism for populating the 16$^+$ state has not been identified. Two possible paths exist based on first exciting the nucleus to high spin-states (probably $J > 20\hbar$) in the ground-state band

via multi-step Coulomb excitation: 1) multiple-step transitions (either Coulomb excitation in the reaction or in the subsequent γ-decay) to the $K^\pi = 16^+$ band through a network of K-allowed transitions excitations (based on the possibility that there may be more K bands above 3-4 MeV in excitation); and/or 2) single-step, K-forbidden transitions (Coulomb excitation or γ-decay) from states in the ground-state band into the $K^\pi = 16^+$ band, (mediated by K-mixing). Overall, the authors found substantial K-forbidden Coulomb excitation of several rotational bands. The observations are consistent with a rapid breakdown of the K quantum number as the low-K bands are excited to higher energy, *i.e.* higher K components are admixed in the low-K bands (such as the ground state band) with increasing spin, while the high-K bands (such as the K^π=16^+ isomer band) remain relatively pure for all known states. The authors also considered various de-excitation paths for the ^{178m2}Hf isomeric state: Coulomb depopulation was calculated to be possible (at the 1% level), but no intermediate states that might aide the de-excitation of the ^{178m2}Hf state via photons were found. The work also provides a large amount of new nuclear structure information, including improved information on the quasiparticle nature of the various isomeric bands.

Projected shell-model description for nuclear isomers; Sun (2008) [142]. This unpublished reprint (on the nuclear theory archive) describes calculations for isomeric states in several nuclei using the formalism of the so-called projected shell model. The projected shell model is a configuration interaction method utilizing a deformed Nilsson basis, where the residual interaction is general taken to be of the form quadrupole-plus-pairing. The projected shell model derives its name from projecting angular momentum onto the deformed basis while diagonalizing the residual interaction within the configuration basis. Calculations are reported for ^{178}Hf demonstrating overall agreement between theory and experiment for the ground-state band as well as the $K^\pi = 6^+$ band, the first and second $K^\pi = 8^+$ bands, and the $K^\pi = 14^-$ and $K^\pi = 16^+$ bands. Note that counter to experiment, in the calculations, the $K^\pi = 14^-$ band lies slightly below the $K^\pi = 16^+$ band. Projected shell-model calculations were also reported to examine the existence of a $K^\pi = 6^+$ bands in $N = 104$ isotones. A discussion on the existence of shape isomers for nuclei in the A=70 region based on the projected shell model is also given.

3.4 Section summary

Past experiments have provided extensive detail of the structure of ^{178}Hf. One of the most powerful tools employed has been Coulomb excitation, a multi-step process used to excite the nucleus to angular momenta greater than 20 units or so. In addition, it has also permitted a probe of the "goodness" of the K quantum number at high spin. While levels have been analyzed and identified up to 5 MeV, the spectrum is by no means complete up to this excitation energy. Indeed, many of the known bands have not been extended up to this energy. However, following extensive analysis of observed γ-transitions, the spectrum is fairly complete up to an energy of approximately 2.7 MeV. Additional K bands built on quasiparticle excitations are possible, and have even been predicted. From the Couomb-excitation experiments it can be concluded that these are either weakly populated, so it is not possible to pick them out of the background, or that they have long half lives which have hindered their observation in the experiments carried out so far. It also has to be remarked

that because of the extreme sensitivity of modern γ-detectors, such as GAMMASPHERE, Coulomb excitation has been very successful in populating a wide range of states, including the $K^\pi = 16^+$ isomer. However, these experiments have not been able to identify a state with mixed K that could serve as a doorway state to de-excite the 31-yr isomer following the absorption of the low-energy (10 keV) photon. It is important to note that for such a state to act as a "trigger" it would also have to have an allowed γ-decay to a state lying significantly below the isomer that continues to decay to the ground-state band, otherwise the phase-space factors for electromagnetic decay would dictate that it decay back to the isomeric state. The γ-decay of such a state should have been observable in the γ-coincidence experiments following Coulomb excitation, but has not been seen thus far.

4 Potential energy release mechanisms

As discussed in the introduction, there is energy stored in an isomeric state, and, if pure ^{178m2}Hf could be obtained, the energy-density would be extremely high compared with chemical methods of energy storage. In order to decide if there is a here a practical device, we have first to see if there are systematic methods of *releasing* that isomer energy. Is there a triggering mechanism for releasing that potential energy?

Some experiments of the last 10 years appear to suggest that the isomer energy of ^{178m2}Hf can be released by incident X-rays of energies in the range 5–50 keV, and these experiments are reviewed in detail in Section. 4.1. More recently, it has been conjectured [109] that neutrons may be an suitable incident projectile for a reaction that releases the excitation energy, and this prospect is examined in Section. 4.2. Finally, in Section. 4.3 we consider how nuclear excitations may be prompted by electron transitions, a process which, if it occurred, could amplify the triggering induced by X-rays.

4.1 Reactions with photons

It is certainly possible to de-excite isomers by reactions with photons. One physical mechanism is a direct absorption of a photon by the nucleus in an isomeric state leading to its excitation to an intermediate level that will then decay bypassing the isomeric state either by γ-emission to the ground state or by a particle emission or fission. Another mechanism is coupling of atomic interactions to the nucleus in a process like the nuclear excitation by electron transition (NEET) described in the Sect. 4.3. In this case, a photon excites the atomic electrons and their excitation energy is transferred in a resonant process to the isomeric nucleus exciting it to an intermediate state that then decays bypassing the isomeric state.

The main focus of this section is a description and discussion of X-ray experiments aimed at inducing de-excitation of the K-isomer ^{178m2}Hf. First, however, we briefly review established de-excitations of the K-isomer ^{180m}Ta and the spin isomer ^{68m}Cu triggered by reactions with photons. The nuclear photo-absorption is the de-excitation mechanism in these two cases. We then proceed to the ^{178m2}Hf case where presumably the NEET mechanism should play a role if a de-excitation induced by X-rays with energies ≤ 90 keV occurs.

4.1.1 Triggered de-excitation of ^{180m}Ta

The isotope ^{180m}Ta is the rarest stable isotope occurring in nature and it is the only naturally occurring isomer. The ground state of ^{180}Ta is 1^+ with half life of 8.1 h. The 9^- isomeric state at excitation energy of 75.3 keV has a half-life greater than 1.2×10^{15} yr.

Depopulation of the isomeric state ^{180m}Ta by the reaction ^{180m}Ta $(\gamma,\gamma')^{180}$Ta was first reported in Ref. [33]. Bremsstrahlung with end-point energy of 6 MeV was used to irradiate enriched ^{180m}Ta target. The γ radiation excited the isomer to intermediate states of at least 2 MeV excitation energy that subsequently decayed through unobserved cascades to the 1^+ ground state of ^{180}Ta. The electron capture and a beta decay of the ^{180}Ta ground state then produced ^{180}Hf and ^{180}W. The $K\alpha$ and $K\beta$ fluorescence of the ^{180}Hf was then observed. The reported integrated cross section was 4.8×10^{-25} cm^2 keV which was two orders of magnitude greater than typical (γ,γ') reactions that produce isomers of other species.

In a subsequent experiment [27], the energy of the bremsstrahlung was varied between 2 to 5 MeV. This allowed the identification of the energy range of intermediate states through which the isomer depopulation proceeds. Two energy regions were identified, one at 2.8 MeV and the other at 3.6 MeV. The reported integrated cross section was 1.2×10^{-25} cm^2 keV, which exceeds by an order of magnitude known cross sections that produce isomers of other species.

The large magnitude of the integrated cross section reported in Ref. [27] was disputed in a comment [111]. An independent measurement using 1.3 MeV and 4 MeV γ-radiation found upper limit of 14 nb cross section at 1.33 MeV and finite value of 0.52(20) mb at 4 MeV. It was claimed that the 4 MeV result considered together with data obtained for ^{115}In$(\gamma,\gamma')^{115m}$In did not support the large integrated cross section of Ref. [27]. Also, the bremsstrahlung calibration in the commented upon experiment was questioned.

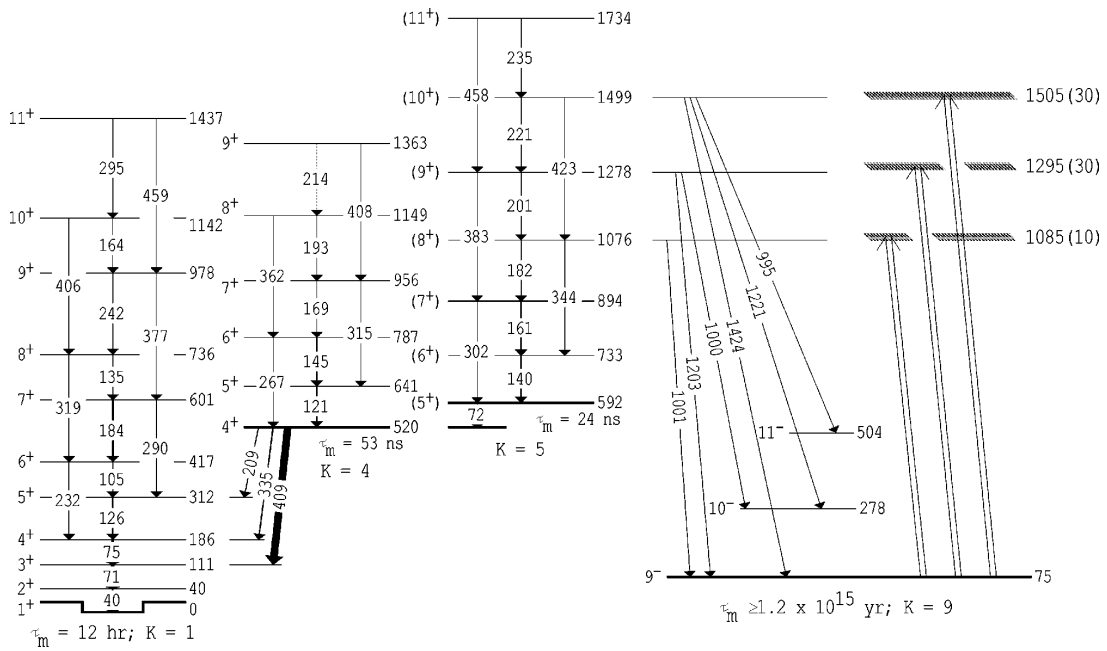

Figure 2: Partial level scheme for ^{180}Ta with energies in keV. Photoactivation transitions are shown on the right. Also shown are the predicted $E1$ transitions decaying from the $K^\pi = 5^+$ band back to the $K^\pi = 9^-$ isomer band. Adopted from Ref. [156].

In the reply [21], the authors of the original paper defended their calibration of the bremsstrahlung spectra and cast doubt on the Ta/In data connection.

Another experiment using bremsstrahlung with end-point energies in the range of 5.4 - 7.6 MeV was reported in Ref. [85]. The goal of this study was a determination of the depletion probability of the ^{180m}Ta isomer after γ-ray absorption. Under the assumption that the cross section for the isomer depletion can be taken as a product of the giant electric dipole resonance cross section and the probability for decay of the compound nucleus to the ground state and with the possibility to determine the absolute cross section of the (γ,γ') reaction using a monitoring reaction ^{232}Th(γ,f) allowed to deduce the probability of the ground-state population. It was found that this probability increases with γ-ray energy and reaches about 15% at the highest measured energy of 7.6 MeV. Therefore, there is 85% probability that the compound-nucleus excited state will decay back to the isomer. This result suggests only a modest K-mixing at the excitation energies studied in ^{180}Ta.

Astrophysics consequences of the depopulation of the ^{180m}Ta isomer by resonant photoabsorption were investigated in Ref. [12]. The irradiation of the isomer was performed for bremsstrahlung endpoint energies from 0.8 to 3.1 MeV. The depopulation of the isomer was observed down to about 1 MeV. An intermediate state as low as 1.01 MeV was suggested. This implies a reduction of the isomer lifetime in a photon bath accompanying the s-process.

A theoretical investigation of the ^{180m}Ta isomer depopulation by resonant photoabsorption was performed in Ref. [156]. Experimental results reported in Ref. [12], in particular the suggestion of an intermediate state at 1.01 MeV excitation energy above the isomer energy, were interpreted. It was proposed that the deexcitation proceeds through the $K = 5$ band

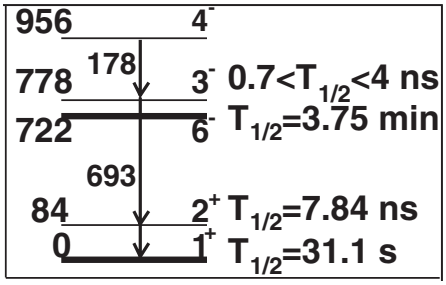

Figure 3: Partial level scheme for ^{68}Cu and deexcitation γ-rays with energies in keV. Adopted from Ref. [140].

8^+, 9^+ and 10^+ states. The isomer is a $K = 9$, 9^- state. $E1$ radiation excites each of the three $K = 5$ states, which subsequently decay either back to the isomer or to the 1^+ $K = 1$ ground state, see Fig. 2. The 8^+, $K = 5$ state with excitation energy of 1076 keV is identified with the 1.01 MeV state (measured from the isomer energy) of Ref. [12]. A substantial K-mixing of the non-yrast states is suggested in order to explain experimental observations.

4.1.2 De-excitation of ^{68m}Cu by Coulomb excitation at ISOLDE

In a recent letter [140] a pioneering study was reported on induced isomeric deexcitation of ^{68m}Cu nucleus leading to triggered γ-ray emission. The experiment was performed at ISOLDE, CERN. Coulomb excitation was used with a post-accelerated beam of ^{68m}Cu isomerically purified through selective laser ionization. The isomeric state has $J^\pi = 6^-$, excitation energy of 722 keV and a half-life of 3.75 min. The beam was used to bombard a ^{120}Sn target and the detection of the γ-rays was performed with the MINIBALL array. It was established that the isomeric state is excited into an intermediate 4^- state at 956 keV, which then promptly decayed to a 3^- state at 778 keV that further deexcites via $3^- \rightarrow 2^+ \rightarrow 1^+$ sequence, see Fig. 3. The 1^+ state is the ground state with a half-life of 31.1 s. It is interesting to note that by the Coulomb excitation of the 6^- isomer in ^{68}Cu, the induced instantaneous depopulation of a nuclear isomer was demonstrated. Unlike in the case of the K-isomers, e.g. ^{180m}Ta or ^{178m2}Hf where the isomer depopulation can only proceed through weak transitions arising from K-mixing, here an alternative scheme is revealed. The E2 Coulomb excitation feeds a member of the multiplet which deexcites faster through M1 than E2 transitions, eventually bypassing the isomer.

4.1.3 ^{178m2}Hf X-ray experiments

Experiments involving the ^{178m2}Hf isomer are more complex and challenging compared to ^{180m}Ta investigations. First, the 31-year half-life of ^{178m2}Hf compared to the extremely long half-life of ^{180m}Ta implies a much stronger background from the natural decay of the ^{178m2}Hf. Second, a significantly higher excitation energy of the ^{178m2}Hf isomer, 2.446 MeV, compared to 75.3 keV of ^{180m}Ta , means a much higher level density in the vicinity of the isomeric state, making interpretation of measurements, as well as the theoretical understanding, more challenging. Third, the decay of the short-lived ground state of ^{180}Ta was successfully used

as a signal for the ^{180m}Ta induced deexcitation. This cannot be done in the ^{178}Hf case as it is stable. Finally, a production of the ^{178m2}Hf target is a complicated task in itself.

Bremsstrahlung X-ray experiments by Collins et al.

The first report of accelerated γ-ray emission from the ^{178m2}Hf irradiated with X-rays was published in Ref. [29]. X-ray pulses from a device typically used in dental medicine with an end point energy set to be 70 or 90 keV were used in the experiment. Intensities of several γ transitions were found to increase by about 4%. An integrated cross section of 10^{-21} cm^2 keV for the resonant absorption of X-rays to induce γ-decay was deduced. An excess of 6%±2% and 2%±1% was reported for the 495 keV line corresponding to the $11^- \rightarrow 9^-$ transition in the $K^\pi = 8_1^-$, 4 s isomer band and for the 426 keV line corresponding to the $8^+ \rightarrow 6^+$ transition in the ground-state band, respectively. On the contrary, no enhancement was found for the 574 keV line of the $13^- \rightarrow 11^-$ transition in the $K^\pi = 8_1^-$ 4 s isomer band that feeds the 495 keV transition during the spontaneous decay. The low-energy 93 keV ($2^+ \rightarrow 0^+$) and 213 keV ($4^+ \rightarrow 2^+$) lines were blocked by a Pb filter. The ground-state band $6^+ \rightarrow 4^+$ 326 keV transition was not discussed.

A separate paper describing the same experiment was published in Laser Physics journal [28]. In this paper, an enhancement was reported for the $8^+ \rightarrow 6^+$ 426 keV transition and, in addition, for the $6^+ \rightarrow 4^+$ 326 keV transition in the ground-state band. However, the enhancement of the 495 keV line $11^- \rightarrow 9^-$ transition was not confirmed.

Comments on these experiments

Three published comments followed the paper [29].

In the first one [118], it was suggested that the reported enhancements are due to statistical fluctuations. It was argued that the both measured and calculated (γ, γ') integrated cross sections are several orders of magnitude smaller than that reported in Ref. [29]. At the same time, there were inconsistensies in the enhancements reported in Refs. [29] and [28], i.e. no mentioning of the 495 keV line enhancement in the latter, was pointed out. Further, it was noticed that some of the ^{178}Hf lines show negative variations.

In the second comment [105], the large integrated cross section reported in [29] was shown to represent about 38% of the E1 energy-weighted sum rule. Taking into account the 90 keV energy of the X-ray photons, the excitation energy of the intermediate state that facilitates deexcitation of the isomer cannot be higher. It is physically unacceptable for such a low-lying state to carry such a large portion of the E1 sum rule. Consequently, nuclear absorption is ruled out as the physical process that can explain accelerated γ-ray emission reported in Refs. [29] and [28].

The third comment [152] argued along similar lines as the previous comment [105]. Its conclusion was that a resonant excitation by the X-rays into an intermediate state is excluded as an explanation of the data obtained in [29]. Atomic interactions coupling strongly to the nucleus were suggested as a speculative explanation to be explored.

In their reply [30], the authors of the experiment disputed the statistical fluctuation explanation of their data as suggested in the first comment [118]. Figures showing enhancements for three ground-state band transitions were presented. The 426 keV $8^+ \rightarrow 6^+$ transition discussed in the original paper was shown together with the 326 keV $6^+ \rightarrow 4^+$ transition

and the 213 keV $4^+ \rightarrow 2^+$ transition. The latter two transitions were not discussed in the letter [29] except for a statement that the 213 keV $4^+ \rightarrow 2^+$ transition was blocked by a Pb filter. The most strongly enhanced transition reported in the original letter, the 495 keV line corresponding to the $11^- \rightarrow 9^-$ transition in the $K^\pi = 8_1^-$ 4 s isomer band, was not mentioned in the reply.

Subsequent bremsstrahlung experiments by Collins et al.

Results of a subsequent bremsstrahlung experiment performed by Collins' group was published in Ref. [32]. The X-ray end point energy was 63 keV. A 1.6% enhancement was reported in the 213 keV $4^+ \rightarrow 2^+$ ground-state band transition. The 326 keV $6^+ \rightarrow 4^+$ ground-state band transition was also found enhanced by irradiation and integrated cross section of 2.2×10^{-22} cm^2 keV was deduced. Further, it was stated that the resonant absorption takes place below 20 keV as indicated by the use of selective absorption filters in the irradiating beam. Counts were summed for the 213 keV $4^+ \rightarrow 2^+$ ground-state band transition and the 217 keV $9^- \rightarrow 8^-$ 4-s isomer band transition. No other 4-s isomer band transitions were reported. The 426 keV, $8^+ \rightarrow 6^+$ ground-state band transition so prominently discussed in Ref. [29] was not mentioned in this work. It was proposed that the induced transitions bypass the 4-s isomer band through which the ^{178m2}Hf spontaneously decays. This was in contradiction to the claims made in the original Ref. [29], where the 495 keV line corresponding to the $11^- \rightarrow 9^-$ 4-s isomer band was reported to be the most significantly enhanced.

More results obtained with bremsstrahlung X-rays were reported in Ref. [31]. The end-point energy was 90 keV. The 326 keV $6^+ \rightarrow 4^+$ ground-state band transition was found enhanced by irradiation. Integrated cross section of 3×10^{-23} cm^2 keV was reported.

Coincident measurements of the ^{178m2}Hf γ-transitions either from spontaneous decay or from an induced decay by bremsstrahlung X-rays with end point energy set between 60 and 90 keV were reported in Ref. [37]. A new 129.5 keV line was found during X-ray irradiation in coincidence with the 213 keV $4^+ \rightarrow 2^+$ ground-state band transition (see also the discussion of the thesis by C. Rusu [131] below). When gated on this new line, only the 213 keV ground-state band transition was seen. On the contrary, when gated on the 88.8 keV, $8^- \rightarrow 8^+$ transition, all the ground-state band cascade was observed. The conclusion was that the induced decay bypasses the 4-s isomer band, in contradiction to the original claims in [29], through unidentified cascades that include the 129.5 keV transition. The transitions connecting the lower members of the ground-state band were found more enhanced.

Argonne measurements

Independent experiments seeking to confirm the X-ray induced acceleration of the ^{178m2}Hf isomer were performed at Argonne National Laboratory with an intense white X-ray beam from the Advanced Photon Source [4]. The X-ray intensity was 4 orders of magnitude larger than those in [29]. The explored X-ray energy range was 20-60 keV. Using a tungsten shutter, the beam was on the target for 11 s and off the target for 22 s during runs over a period of 10 hours. If the claims in [29] concerning the enhancement of the $11^- \rightarrow 9^-$ transition in the $K^\pi = 8_1^-$, 4 s isomer band were correct, the measured γ-rays in the ground-state band during the beam-off period would show an enhanced counting rate in interval the first 11 s interval as compared to the second 11 s interval. No statistically significant difference in

the γ-ray intensities between the two periods was found for any γ-ray line in the spectrum. Further, an attempt was made to search for changes in intensities due to a hypothetical prompt deexcitation bypassing the 4 s isomer band. No significant increase in activity for any transition was seen, although the background with the beam on was high and the Pb absorbers were in place. The obtained data were consistent with an integrated cross section less than 2×10^{-27} cm^2 keV for decays that would go through the 4-s isomer and a value less than 2×10^{-26} cm^2 keV for decays that would populate the ground-state band directly, more than three orders of magnitude below the valuse reported by Collins *et al.*

Collins' experiments with tuned beam at SPring-8

Tunable synchrotron radiation from the SPring-8 facility was used to study induced γ-emission of ^{178m2}Hf [41]. Energy range of 9 to 13 keV was explored in steps of 5 eV. The ^{178m2}Hf target irradiation lasted several tens of second at each energy. The energy range was selected because it contains much of the transition strength for photoionization of the L-shell electrons of Hf. A 4 σ enhancement of the summed 213 keV $4^+ \rightarrow 2^+$ ground-state band transition and 217 keV $9^- \rightarrow 8^-$ transition in the $K^\pi = 8_1^-$ 4 s isomer band was reported with cross section of $\approx 2 \times 10^{-22}$ cm^2. The nuclear excitation by electron transition (NEET) process was identified as the physics mechanism of the induced transitions. The NEET branch was reported to be 2×10^{-3} relative to L-shell photoionization. The null result of the Argonne measurement [4] was attributed to lack of optical transparency at the resonant energies for NEET. In addition to the ^{178m2}Hf transitions, transitions due to the ^{172}Hf impurity were also measured. Fractional increases in Figs. 4 and 5 of Ref. [41] demonstrate large statistical fluctuations. It is not clear why there was no attempt to increase irradiation time in the regions of interest (e.g. near L_1 and L_3 edges) in order to improve statistics of the alleged transition enhancements.

Second Argonne experiment

Responding to the new experiments by Collins' group concluding that the induced γ-emission of ^{178m2}Hf was triggered by X-rays not in the energy range of 20-60 keV as reported in Ref. [29] but rather at lower 9 to 13 keV range [32, 41] that was not explored in the first Argonne measurement [4], a new experiment was performed at the Advanced Photon Source using a thin electroplated Hf target [3]. An increased sensitivity to the low-energy X-rays was achieved. The same 11 s illumination of the target and two 11 s counting periods were used as in the original experiment. No enhanced γ-emission was observed for any ^{178}Hf line. An upper limit to the energy-integrated cross section for X-ray induced decay of the 31-y ^{178}Hf isomer was established that was less than 1×10^{-26} cm^2 keV over the incident photon energy range of 6-20 keV, three orders of magnitude below the values reported by Collins' group.

Further Collins' measurements at SPring-8 and Paul Scherrer Institute

Monochromatic synchrotron radiation X-rays with tunable energy were also used by the Collins' group in follow up experiments to the 2001 SPring-8 measurements [41]. Results obtained in 2002 at the SPring-8 and in 2003 at the Paul Scherrer Institute SLS synchrotron radiation source were reported in Ref. [40]. Enhancement of the ground-state band transitions was reported for X-ray energies around 9.56 keV near the Hf(L_3) edge. Further, a new 130 keV line not seen in spontaneous decay was reportedly seen when the ^{178m2}Hf target was

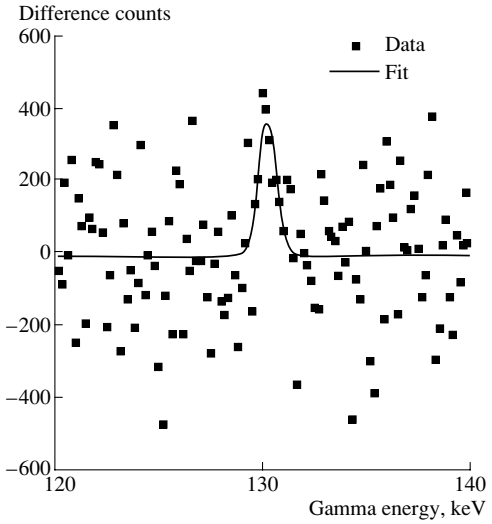

Figure 4: Differences in the number of counts from γ photons collected with the synchrotron radiation incident and blocked. Quoted from Ref. [40].

irradiated. This line was identified as the same one that was observed in coincidence with the $4^+ \rightarrow 2^+$ ground-state band transition in measurements discussed in Refs. [37] and [131]. The second Argonne experiment was criticized for having lower energy resolution and lower sensitivity to photons near the 130 keV γ-line. A 12σ enhancement for the ground-state band transitions was reported. However, an examination of Figure 7 of Ref. [40] (130 keV line), reproduced here in Fig. 4, and Figure 8 of Ref. [40] reveals large statistical fluctuations that make enhancement claims unconvincing. The fit revealing the 130 keV line in Fig. 4 is rather arbitrary. It should also be noted that there is 125.8 keV γ-transition in ^{172}Hf, which is present due to target impurity, that makes identification of the proposed weak 130 keV ^{178}Hf transition challenging.

Results obtained with the tunable monochromatic X-rays from SPring-8 and Paul Scherrer Institute SLS synchrotron were discussed and summarized in Ref. [38] in a similar way as was done in Ref. [40].

Another experiment was performed by the Collins' group at SPring-8 in 2004 [39]. This time, a discovery of a trigger level that serves as an intermediate state for the induced deexcitation of the isomer was reported. It was claimed that the trigger level lies at 2457.20(22) keV and that a direct deexcitation of this level to the ground state by a single γ-transition was observed. It should be noted that this is by far the strangest claim made by Collins *et al.* In order for a prompt transition to occur to the ground state, this intermediate state must have a spin-parity of 1^+, 1^-, or 2^+. Which means a change in spin of 14-15 units from the isomer to the intermediate state. At the same time, a state with such a spin and parity would decay to the first excited 2^+ state of ^{178}Hf at 93 keV. So, not just the 2457 keV transition but also a 2364 keV transition should have been seen simultaneously. The intermediate state, or trigger level, excited either by X-ray resonance absorption or by the NEET mechanism is expected to have spin 15^- as the most probable transition induced by the synchrotron X-rays would be electric dipole ($E1$). A transition of the 15^- state to the ground state has a

probability of 10^{-49}. We note that this work makes use of a 2447.86 keV γ-ray coming from the beta-decay of ^{214}Bi for calibration purposes, which has a half life of 19.4 m, and is part of the ^{232}Th alpha-decay chain. It is likely to be observed because of Rn in the air. These γ-rays actually follow the beta-decay of ^{214}Bi and are from ^{214}Po. This particular γ-ray is from a 2.4% branch in the beta-decay of ^{214}Bi. The sum of this background line and the source X-ray is 2447.86 keV + 9.56 keV = 2457.41 keV, which is within the error for the 2457.20(22) keV line observed. Thus, it is quite possible that the observed 2457.2 line is due to a pile up effect where scattered 9.5 keV photons coincide with the 2447.86 keV ^{214}Bi line in the detector. Given the high irradiation rate, 10^{12} photons cm^{-2} s^{-1} this is reasonable. It is interesting to note that in this paper the authors admitted criticism of their previous results. In particular, it was admitted that measurements of increases of spontaneous decay transition intensities were subject to fluctuations that may have resulted from the difficulties in stabilizing experimental alignments. Further, the observation of new lines such as the one at 130 keV that appeared in the γ-spectra of the induced decay had difficulties because of its proximity to a line from the spontaneous decay of a contaminant isotope, ^{172}Hf.

Carroll's independent experiments

Independent experiments by Carroll and collaborators were described in Refs. [20] and [127]. In the former, a search for low-energy ($\leq$20 keV) triggered γ-emission from ^{178m2}Hf was performed using the YSU MINIBALL detector array. Irradiations of samples were performed using a radiographic X-ray tube with a rotating anode operated in a pulse mode. Bremsstrahlung was produced with photon endpoint of 100 keV. The isomeric sample contained 3×10^{13} ^{178m2}Hf atoms. A dummy sample containing only the ground-state Hf isotopes was also used. No induced de-excitation was observed. An upper limit on the integral cross section for triggering on the order of 10^{20} cm^2 keV for incident photons near 10 keV was obtained. In the latter paper, an experiment using intense monochromatic synchrotron radiation from the X15A beamline at the National Synchrotron Light Source at Brookhaven National Laboratory was described. Studies were performed to probe incident photon energies over the L_1, L_2, and L_3 X-ray edges of Hf and the 1213 keV range. No enhancement of the ^{178m2}Hf γ-lines was observed and a limit on the integrated cross section of the order of 1×10^{-25} cm^2 keV was set.

Sandia group experiment at CAMD and Stone's independent analysis and report

The Sandia Group of P. McDaniel performed an experiment at the CAMD Facility, Baton Rouge, in November 2003 with the aim to test claims of triggering the decay of the 31 y isomer ^{178m2}Hf by suitable tuned X-rays. The experiment was performed in four stages. The system set up comprised two Ge detectors, a ^{178m2}Hf source (with some ^{172}Hf and its daughter ^{172}Lu), and a ^{137}Cs source. The more intense transitions in the decay of ^{172}Lu, and the ^{137}Cs, acted as invariant standards to monitor the behaviour of the detection system. The X-ray flux at the beam line used at CAMD was given as approximately 3×10^{13} photons keV^{-1} cm^{-2} s^{-1} in a line width of about 1 eV. This flux was lower than the fluxes used in Collins' group experiments at Spring-8 and at the SLS where positive results of X-ray triggering of ^{178m2}Hf decay have been reported. The data were taken in 30-minutes intervals with the X-rays on the Hf target at a fixed X-ray energy. The beam-off data were taken with beam in the synchrotron but with the X-ray line gate closed. The energy range of

9509-9595 eV in steps of 1 eV was explored. In total, eleven intense γ-peaks were analysed: The 213 keV, 326 keV and 426 keV transitions in the ground-state band and five 8^-, 4-s isomer band transitions of ^{178}Hf, three transitions in ^{172}Lu and one in ^{137}Cs. The Sandia group has published results in the CAMD 2003 Annual Report. Their conclusion was that based on the data from all ^{178m2}Hf transitions triggering at the level of 0.5 % over a wide X-ray energy range between 9560 and 9570 eV was observed. This claim was based on results obtained during the first and the longest stage of the experiment called "Sweeps". The stage named "Triples" gave according to the Sandia group an inconclusive triggering upper limit of 0.05%. The last two stages had lower statistical quality and gave no results to support observation of triggering. An independent analysis of the raw data collected by the Sandia group in all four stages of the experiment was performed by J. R. Stone and N. J. Stone [141]. They found a presence of a slow drift in the detector response. When this was modeled and taken into account no evidence remained for triggering by X-rays in the energy range 9509-9595 eV at the 0.2% level and no evidence at 9563 keV at the 0.05% level. It was suggested that reports of evidence for triggering made by the Sandia group based on the same data are in error through having ignored, or failed to recognize, the presence of the slow drift during the "Sweeps" stage of the experiment. The drift in the detector response had impact on the analysis because majority of the background runs were performed at the beginning of the "Sweeps" stage while most of the beam-on data were taken at later times when the detector counts were increasing for both the background and the beam-on data in all observed transitions. It should be noted that the result of the analysis of the "Sweeps" stage by Stones is in agreement with the Sandia results and conclusions from the three other stages. In those stages, the background was taken at times close to the beam-on measurement, so no time-drift in the detectors could influence the analysis.

Coincidence measurement reported in the thesis by C. Rusu

In another experiment by the Dallas group performed by a Ph.D. student C. Rusu, nuclear spectroscopy was used to study photoexcitation of ^{178m2}Hf [131]. Coincident detection of the Hf γ-photons was achieved with four HPGe detectors. A bremsstrahlung X-ray generator was used as excitation source, which covered a continuous energy-range from 0 to 60 keV. An observation of X-ray triggered isomer deexcitation was reported. In particular, an analysis of the γ-γ coincidence data revealed that during X-ray irradiations, a line of 129.5 keV was found in coincidence with the 213.4 keV $4^+ \rightarrow 2^+$ ground state band transition. The confidence level reported was 5σ. The 129.5 keV line is not a known transition of the spontaneous decay of ^{178m2}Hf. The 129.5 keV γ-transition was interpreted as a member of a sequence of γ-transitions that bypasses most of the normal decay cascade populated by spontaneous decay. It should be noted that results of this experiment were also reported in Ref. [37]. In the same paper and also in Ref. [40] an observation of a 130 keV line was reported when ^{178m2}Hf was irradiated by synchrotron radiation at SPring-8 facility although the evidence for a signal was marginal due to low statistics. It should also be noted that two 129 keV transitions were identified in ^{178}Hf in (n,γ) activation experiments [60]. The first corresponds to a transition from the 1513.8 keV 4^+ state to 1384.5 keV 4^+ γ-band state. The second corresponds to a transition from the 1538.8 keV 4^- state to 1409.4 keV 4^- state. The 129.5 keV transition was observed when gated on the 213.4 keV transition together with other ground-state band transitions and with the 88.8 keV $8^- \rightarrow 8^+$ transition. When gated

on the 129.5 keV transition, only the 213.4 keV transition was seen. These coincidences were observed only during 2.5 ms pulse X-ray irradiation. As described in the thesis in detail, effort has been made to subtract background carefully.

TRIP measurement

An attempt to reproduce coincidence measurements by C. Rusu [131] was made by the Sandia group of P. MacDaniel. The experiment, named TRiggered Isomer Proof (TRIP) Test, was performed at the National Synchrotron Light Source at Brookhaven National Laboratory in 2005 [104]. Since bremsstrahlung was used in experiment [131], it was not known at which energy the isomer triggering (if any) occurred. Based on the claims of Ref. [40] that the same 130 keV line was observed at the SPring-8 facility experiment at the X-ray energy around 9567 eV and based on the Sandia group earlier experiment [141] claiming isomer triggering at the level of 0.5 % near the 9567 eV X-ray energy, it was decided to perform the coincidence measurements at this X-ray energy. As a result of the experiment a 4σ effect was reported for the 213 keV - 129 keV coincidence peak in the beam-on vs. beam-off difference spectrum. Next, the decay path involving the 129 keV transition was suggested comprising the 1513.8 keV 4^+ state and 1384.5 keV 4^+ state in the γ-band of ^{178}Hf. Finally, a suppression of the 426 keV and 326 keV ground state band transitions relative to the lower 213 keV and 93 keV ground state band transitions was observed.

Comments on TRIP report

By examining the coincidence spectra, reproduced here in Fig. 5, we conclude that the peaks were due to statistical fluctuations. The raw beam-on minus beam-off coincidence spectrum in the 129 keV region was strongly negatively biased. Compton plateau from the strong 326 keV and 426 keV ground state band peaks was blamed for this issue. The Compton background due to the 326 keV, 426 keV, 495 keV and 574 keV transitions was modeled and the whole coincidence spectrum corrected. The net result was basically a shift of the difference coincidence spectrum upwards. The question is, why the Compton background was not subtracted directly from the original coincidence peaks similarly as done, e.g. in Ref. [131]. Concerning the suppression of the 426 keV and 326 keV ground-state band transitions presented as a clear proof of bypassing the higher ground-state band states and feeding directly the 4^+ state from a prompt triggered decay, it should be noted that the 495 keV and 574 keV 4-s isomer band transitions were actually seen enhanced with the beam on, but the enhancement was blamed on random coincidences. The 129 keV transition was suggested to originate from the decay of the 1513.8 keV 4^+ level. It is known that there are other five transitions out of this level, all of which have a significantly (more than a factor of ten) higher branching ratio. The measurements should have seen some of these transitions. For example in a comparable energy range, there is a 245 keV transition to the 3^+ γ-band state that would also decay to the 307 keV 4^+ ground state band state [60]. The most important objection concerns the X-ray energy selection in the TRIP experiment. First, by examining Fig. 7 of Ref. [40], reproduced here Fig. 4, we conclude that no 130 keV peak was observed in the SPring-8 measurement near the 9567 eV X-ray energy. More significantly, the Sandia group data from the CAMD measurement analysed by J. R. Stone and N. J. Stone [141] proved that there was no evidence of triggering at this X-ray energy at the 0.05% level. If the coincidence observation reported in Ref. [131] was real, the induced

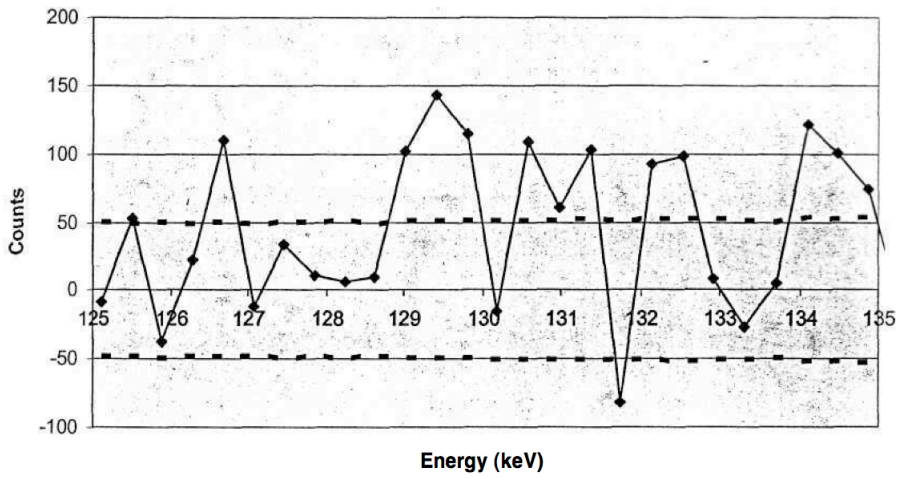

Figure 5: Differences in the number of counts of transitions in coincidence with the 213.4 keV transition with the beam on and the beam off. Adopted from Ref. [104].

isomer decay occurred at a different X-ray energy.

Tkalya's NEET calculations and assessment of Collin's experiments

Theoretical assessment of the X-ray induced decay of ^{172}Hf^{m2} was presented by E. Tkalya in Refs. [146, 147]. Two mechanisms for the induced decay were considered: (1) direct interaction of the incident X-rays with the nucleus and (2) the nucleus-X-ray interaction proceeding via atomic shells. It was establish that the absence of K forbiddenness for all transitions to a hypothetical mixed K intermediate level cannot explain the cross sections reported by Collins *et al.* even if collective nuclear matrix elements or resonant conditions were assumed. Tkalya also tested, and rejected, the hypothesis that the enhancement is due to normal nuclear transitions in the inverse nuclear excitation by electron transition process. The conclusion was that there is no explanation of the Collins' group experimental results within quantum electrodynamics and the contemporary concepts of atomic nuclei. In Ref. [147], responding to the report of the intermediate state at 2457.20(22) keV decaying directly to the ^{178}Hf ground state [39], it was sarcastically suggested that the intermediate level is a mixed J state, i.e. one would have to assume that the angular momentum is not conserved in order to explain this experimental claim. It should be pointed out that the NEET calculations by Tkalya agree or slightly over predict the experimental observations in ^{197}Au [49]. The most optimistic calculations for the ^{172}Hf^{m2} isomer underpredict integrated cross sections reported by Collins *et al.* by orders of magnitude. At the same time, they are consistent with the limits established by the Argonne measurements and the experiments by Carroll and Roberts.

Conclusions

Overall, the X-ray ^{172}Hf^{m2} experiments by Collins *et al.* are statistically marginal and inconsistent. None of the reported positive triggering results were confirmed by independent groups, including those exeriments performed by former collaborators (Carroll). The reported cross sections and integrated cross sections strongly exceed theoretical expectations. The only measurement that shows statistical significance is the coincidence experiment de-

scribed in the thesis by C. Rusu [131]. The Sandia group experiments failed to confirm this measurement, but only a very small X-ray energy region was explored. The experimental results related to the 129 keV - 213 keV coincidence in the induced isomer decay could be strengthened by higher statistics. In addition, background reduction might be achieved by using anti-coincidences *e.g.* with the 326 keV $6^+ \rightarrow 4^+$ ground-state band.

4.2 Reactions with particles

4.2.1 Introduction

While much effort has been devoted to the search for γ+^{178m2}Hf triggering, it is worthwhile to speculate about triggering being induced by neutrons incident on ^{178m2}Hf. One possible outcome of such a reaction is 'superelastic scattering', or 'inelastic scattering by neutron acceleration' (INNA) [95], whereby the neutron is re-emitted with some part of the isomer energy, and could perhaps continue to trigger neighboring isomers, gaining energy all the time. This is a 'thin chain', and not a multiplying chain reaction as in fission reactors, but may yet prove useful. The process would end either when the neutron escapes from the isomer bulk, or is captured in an (n,γ) reaction to produce ^{179}Hf decaying by emitting γ rays. More γ energy would be produced in a single (n,γ) reaction than in a single INNA reaction, but capture will terminate the thin chain of isomer triggering. Some proposals [109] using mixtures of other elements, to be reviewed below, have been made for generating a multiplying chain reaction.

An analysis of neutron-isomer reactions should predict the relative amounts of inelastic and INNA reactions (which are endothermic and exothermic reactions, respectively), and also the competition with the (n,γ) capture reaction. The competition between these three processes will depend on whether there is K-hindrance or K-mixing in the decay of the ^{179}Hf compound nucleus, and also on level densities, transmission coefficients and γ strengths as are used in all statistical-model calculations. If there should be large K-mixing, then normal statistical models should be usable immediately to give good predictions, whereas if there are large K-hindrances, then we should expect the compound nucleus to decay back to the m2 isomer, which is that no triggering is taking place.

4.2.2 Level densities for ^{178}Hf and ^{179}Hf

The structure of levels in the region of the ^{178}Hf isomer is relatively well known, and the level density around the isomer state is about 100 MeV^{-1} (summed over all J^π). At high energies where not all levels are known (above e.g. 2.5 MeV in ^{178}Hf and above 1 MeV in ^{179}Hf) we assume a Gilbert and Cameron model [53] of the level densities as tuned to $D_0 = 57 \pm 6$ eV, the spacing of resonances for thermal neutrons.

If we add a neutron to ^{178m2}Hf to make ^{179}Hf* excited states, then we are at an excitation energy of 6.099 MeV (the neutron separation energy in ^{179}Hf) plus the isomer energy, or 8.55 MeV excitation energy. An interesting theoretical question is whether the observed level density in ^{179}Hf* agrees with Fermi-gas formulae for 6.099 MeV, or for 8.55 MeV. That is, whether the isomer structure is preserved (or not) within the ^{179}Hf* compound-nucleus system.

4.2.3 Experiments

Experiments which scatter neutrons on isomers have been performed for the isomer states of ^{177m}Lu [128], ^{178m2}Ta [110], and ^{180m}Ta [85], as well as most recently for ^{68,70m}Cu [140].

The earliest neutron experiments[110] measured total and (n,γ) cross sections, and, with low-energy s-wave neutrons, were able to probe the resonant structure of ^{179}Hf* excited states at spin states of $J = 16 \pm \frac{1}{2}$. These experiments revealed a mean level spacing of about 0.9 eV, corresponding to a single-J level density of about 5.5×10^5. It was pointed out this is within 10% of the density expected from the Ignatyuk approach, and within a factor of 2 expected from the Fermi gas model for an excitation energy of 8.55 MeV. The Fermi gas model for 6.1 MeV of excitation predicts a level density of only 1.1×10^4. The fact that this is much further from experiment than the Ignatyuk predictions suggests that there is indeed considerable K-mixing within the ^{179}Hf* compound nuclear states.

There were also a series of experiments [69, 68, 66] in which ^{178}Hf was excited in multiple Coulomb excitation by a Xenon projectile. These experiments were below the Coulomb barrier, and so were direct reactions, and not compound-nucleus reactions. That is, they probed the structure of ^{178}Hf itself, and they revealed that the 16^+ isomer state *could* be populated by some of the decays from high-lying states in the ground-state band. The fractional population was small (the $19^+_{K=16} \rightarrow 18^+_{K=16}$ transition in the four-quasiparticle $K^\pi = 16^+$ band were measured to be $\approx 10^{-3}$ of the $8^+_{\rm GSB} \rightarrow 6^+_{\rm GSB}$ transition), but do reveal that there is a non-zero but small degree of K-mixing within the ground-state band of ^{178}Hf. This comes about from the stronger Coriolis forces from the high angular frequencies in the high-spin states induced by Coulomb excitation. The degree of K-mixing is however estimated as less than one Weisskopf unit, or one nucleon's worth of rearrangement. It is much less than a collective effect of all 178 nucleons.

4.2.4 Theory

The question now arises as to whether we have all the pieces to construct a realistic theory of neutron interactions with ^{178}Hf, or whether the K-selection and K-mixing phenomena are too idiosyncratic for us to make a theory with good predictive power. We have seen weak K-mixing in ^{178}Hf, and strong K-mixing in ^{179}Hf. To answer, we conclude that the strong K-mixing in ^{179}Hf, suggested by experiments and advocated by Oganessian [114], Collins [26], Karamian[84] and Muradian[109], is precisely that assumption which allows us to make a good theory.

Assuming strong K-mixing in the compound-nucleus states in ^{179}Hf, we can now use standard statistical (Hauser-Feshbach) methods without any further K-hindrance factors. As is standard in the field, we fine-tune parameters to known properties of the nuclei.

We performed Hauser-Feshbach calculations, using TALYS (www.talys.eu), for neutrons incident on the 16^+ isomer state of ^{178}Hf at 2.45 MeV, and predicted the energies and intensities of outgoing neutrons to produce ^{178}Hf, and of outgoing gamma-rays to produce ^{179}Hf. The calculations used the global neutron optical developed by Koning and Delaroche, applied to all levels here without any collective couplings in the entrance channel, and used for both ground and isomer initial states. This optical potential gives the transmission coefficients $\mathcal{T}_L$ shown in Fig. 6.

The level density scheme used the known discrete levels for the first 130 levels, where the isomer is level 98. Above that energy we used the scheme of Gilbert and Cameron [53], with

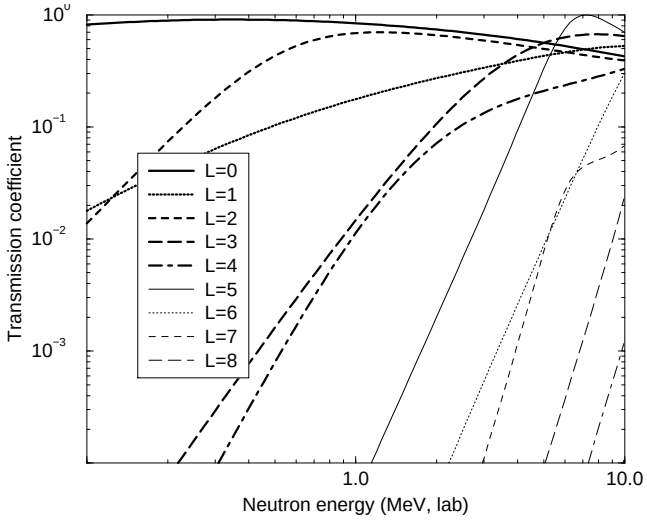

Figure 6: Transmission coefficients for neutrons on ^{178}Hf as a function of neutron energy, for partial waves $L = 0 \ldots 10$. Calculated from the global Koning-Delaroche optical potential (Nuclear Physics A **713**(2003) 310), neglecting the spin-orbit component.

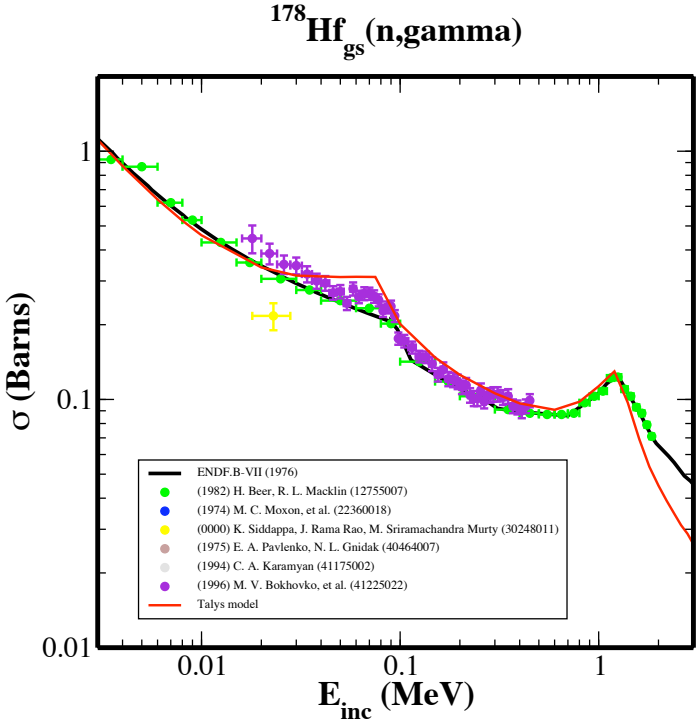

Figure 7: The cross sections for ^{178}Hf (n,γ) for neutrons incident on the ^{178}Hf ground state. We show 6 experimental data sets, the ENDF.B-VII evaluation (black line), and the results of our present TALYS statistical model (red line). We see excellent agreement for the cross sections in this case.

parameter a fitted to the observed $D_0 = 57$ eV for neutrons incident on the ^{178}Hf ground state. We used spin distribution parameter $\sigma = 6.61$ at the neutron separation energy,

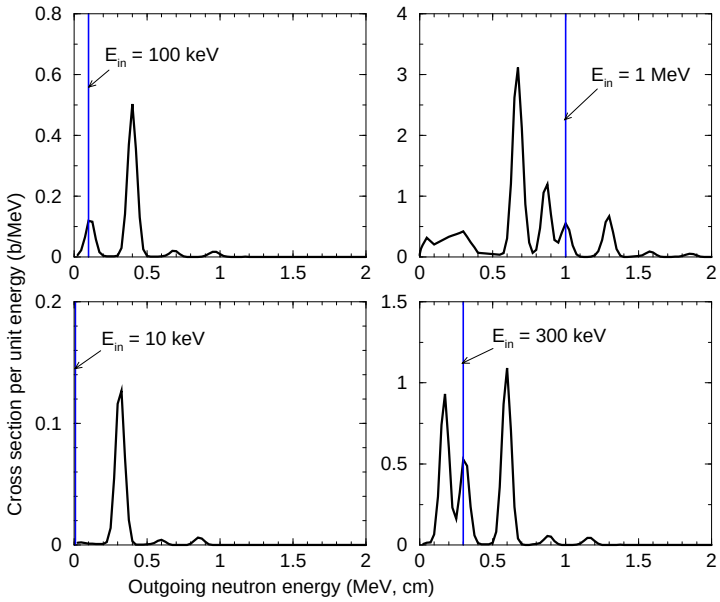

Figure 8: Energy spectra of outgoing neutrons for four beam energies of neutrons incident on the ^{178m2}Hf isomeric state, calculated by a statistical Hauser-Feshbach model assuming complete K-mixing in ^{179}Hf*.

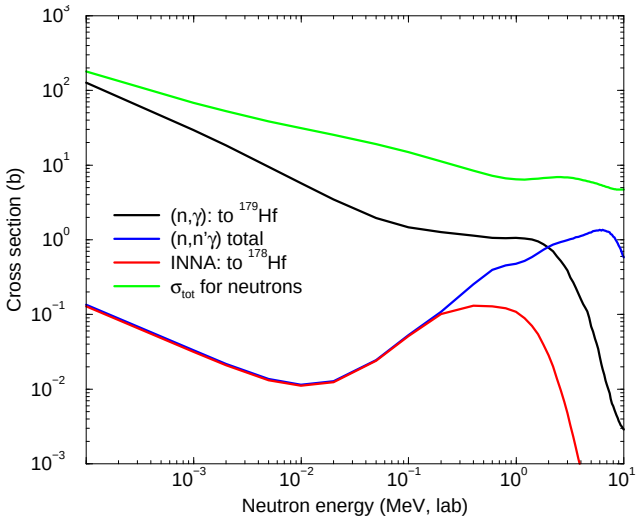

Figure 9: INNA (superelastic) cross section (red line) for neutrons incident on ^{178m2}Hf, from the Hauser-Feshbach calculation using TALYS, using complete K-mixing in ^{178}Hf*. The blue line shows the total triggering cross section, including also the emission of neutrons with less than the incident energy. For comparison, the black line shows the (n,γ) cross section to ^{179}Hf, and the green line the total neutron cross section (for all reaction and elastic processes).

and matched to the constant temperature formula at 6.49 MeV excitation energy. These level parameter assignments predict a resonance spacing for s-wave neutrons incident on the isomer of $D_{0m} = 0.64$ eV, in good agreement with the Muradian et al [110] observation of $0.9^{+0.6}_{-0.3}$ eV. The gamma strength function followed the form of Kopecky-Uhl, with GDR

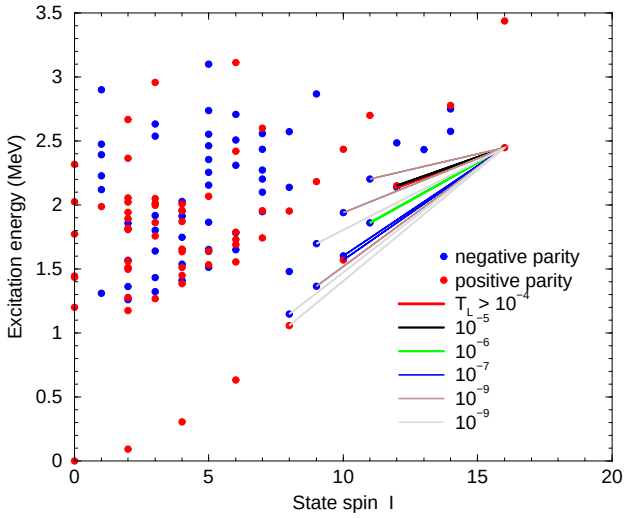

Figure 10: The points show the spin, parity and energy of levels in ^{178}Hf. The lines connect the 16^+ isomer state to states that could be reached after fusion of a low-energy s-wave neutron to make ^{179}Hf*, and then the evaporation of a neutron. The lines are color-coded according to the transmission probabilities according to Fig. 6. All the other final states have $\mathcal{T}_L < 10^{-9}$.

parameters of $E = 14.2$ MeV, $\Gamma = 4.14$ MeV and $\sigma_0 = 476$ mb. The gamma strength function was tuned so that neutrons incident on the ground state have the experimental total radiative width of $\Gamma_\gamma = 0.054$ eV. There is a theoretical uncertainty arising from this last approximation, which may affect the overall (n,γ) cross sections by up to a factor of 2 or 3. We can check the overall accuracy of the model only for neutrons incident on the ^{178}Hf ground states, and we see in Fig. 7 that the agreement there is excellent.

These Hauser-Feshbach calculations, for each incident neutron energy, predict the energies and cross sections of outgoing neutrons, as shown in Fig. 8 for energies of 10, 100, 300 keV and 1 MeV (shown by the blue lines). The cross sections to the residual nuclei and isomeric levels are shown in Fig. 9, as a function of incident neutron energy from 100 eV to 20 MeV (lab). Not all the triggering cross section for producing low-lying levels of ^{178}Hf can count as INNA, however: only that part where the neutron outgoing energy is greater than the ingoing energy. For large neutron energies, it turns out the there are larger cross sections for the emission of a lower-energy outgoing neutrons ('normal' inelastic reactions), followed by gamma-decays that bypass the isomer level. The total triggering cross section for the production of de-excited ^{178}Hf nuclei is shown by the blue curve in Fig. 9.

These results show that INNA does occur, but that for neutron energies up to 10 keV it is about 10^{-3} of the (n,γ) capture cross section. The small INNA component, moreover, does *not* give energetic neutrons with all of the isomer energy. This can be seen from the curves in Fig. 6 and from the ^{178}Hf level diagram: a (say) 2 MeV outgoing neutron could be in *at most* a $L = 6$ partial wave by the transmission coefficient being at least 10^{-4}, but from the level diagram must be *at least* in a $L = 12$ partial wave to carry sufficient angular momentum away from the isomer compound. Fig. 10 shows lines connecting to the final states ^{178}Hf that can be reached by evaporation of neutrons with various threshold values of

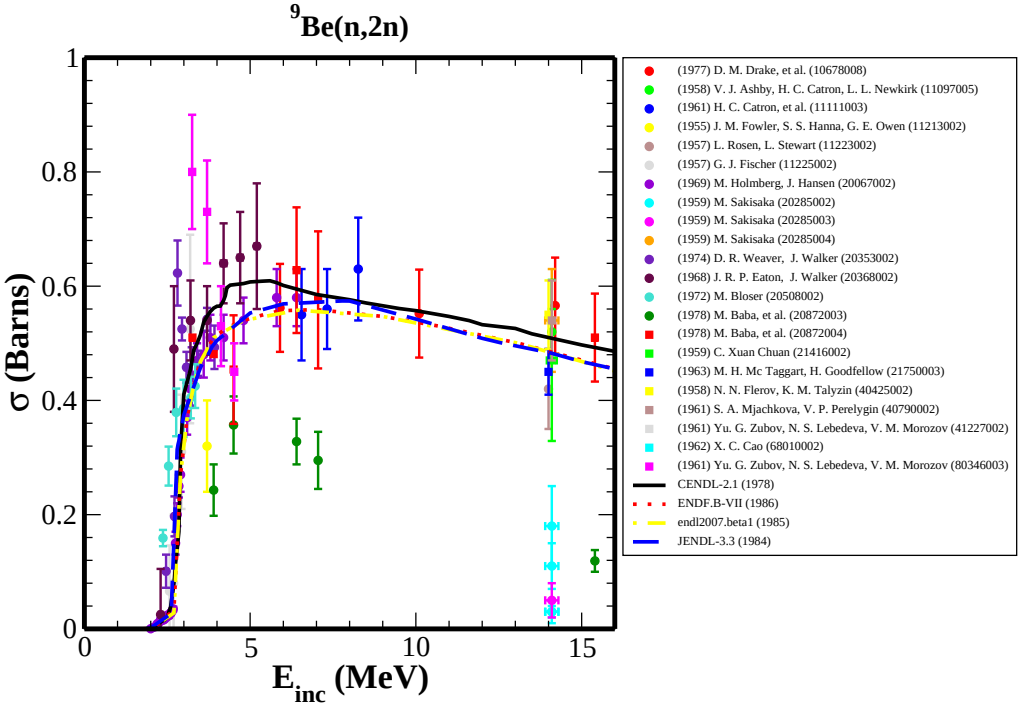

Figure 11: Experimental cross sections for the reaction ^{9}Be(n,2n) as a function of incident neutron energy. The threshold is 1.84 MeV, but the (n,2n) cross section is only significant for neutrons with more than 3 MeV.

the transmission coefficients of Fig. 6. These contradictory requirements mean that INNA is a very weak process compared with (n,γ) capture processes, in which up to 8.5 MeV of energy is released by γ-emissions as the ^{179}Hf decays to its ground state[2].

Similar calculations are also feasible for the INNA reaction on the ^{177}Lu isomer, and we have compared our results (not shown here) to the recent French experiments [128]. Our calculations for neutron capture and decay to the ground state of ^{178}Lu agree well with experiment, but there is a small discrepancy concerning the decay to the ^{178}Lu(9^{-}) isomer. The decay to that isomer was not seen experimentally (it would give a decay component with a specific lifetime), yet is predicted by Hauser-Feshbach models to be about 70% of the ground-state decay. The the overall (n,γ) cross sections are thus still uncertain by a factor of up to 2.

4.2.5 Recent Russian Proposals

In a recent speculative note, Muradian [109] suggested several points that may yet enable an enhanced INNA (superelastic) scattering, and the triggering of isomer energy by neutrons.

[2]We note ironically that this is much more energy than hoped to have been released by the isomer triggering, but only occurs once per neutron.

His points are

1. *That* ^{178}Hf(n,γ) *cross section is uncertain.* We agree, but only within a factor of 2 or 3 when we compare where experiments are possible. This is far from a factor of 10^3.

2. *That K-mixing in* ^{179}Hf *is large.* We agree, and use this as the basis of our calculations. If K-mixing were yet still not 100% complete, then our INNA predictions must be regarded as an upper limit.

3. *That the fraction of high-K states decreases with increasing excitation energy in* ^{178}Hf*, so that INNA should increase for higher-energy initial states.* The first part is not true for the observed level scheme. And the INNA rate depends moreover on K-mixing in ^{179}Hf rather than ^{178}Hf, since low-energy neutrons produce compound-nucleus rather than direct reactions.

4. *That neutron cross sections are large at low energies such as 1 eV.* This is true: both (n,n$'$) and (n,γ) cross sections rise as $1/v$ at low energies. However, they do so with fixed ratio, so the INNA/capture ratio is constant.

5. *That including a mixture of* ^{9}Be *in the* ^{178m2}Hf *bulk may produce a multiplying chain because of the* ^{9}Be(n,2n) *reaction.* However, the ^{9}Be(n,2n) reaction has a threshold of 1.84 MeV (see Fig. 11), and even a full-2.445 MeV incident neutron will yield outgoing neutrons of only 0.3 MeV: a rather low energy where (n,γ) capture on ^{178m2}Hf is more than 10 times the cross section for INNA and further neutron production.

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4.2.6 Summary

Under the optimistic but well-founded assumption that complete K-mixing occurs within the compound-nucleus states, we are able to formulate a good theory of neutron+isomer reactions. This theory has been tested for neutrons incident on the ^{177}Lu $23/2^-$ isomer, and so can be applied for our n + ^{178m2}Hf reaction.

We conclude that inelastic neutron acceleration (INNA) does occur, and that neutrons can release the energy of some fraction of the ^{178m2}Hf isomers, but that for neutrons below 10 keV incident energy, this fraction is about 10^{-3}. For higher neutron energies the fraction does increase to 10^{-2} of $\sigma_{\rm tot}$, but even at 1 MeV most of the energy is carried away by photons rather than neutrons. This is because of the spin-trap form of the ^{178}Hf spectrum, which means that is rather difficult to emit neutrons with any good fraction of the isomer excitation energy. Injecting neutrons into a target of even pure ^{178m2}Hf isomer will therefore hardly yield any even non-multiplying chain reactions. Including a mixture of ^{9}Be in the target will not produce a multiplying chain, because even a full-2.445 MeV incident neutron will yield outgoing neutrons of only 0.3 MeV, at which energy the triggering probability is 10^{-3}.

4.3 Nuclear excitation by electron transition (NEET)

Nuclear excitation by electronics transition (NEET) is a way to enhance the decay of nuclear states via excitation of the atomic states. The attractiveness of this process is that the atomic excitation has, in general, higher cross section. If one can manipulate at the atomic level and cause the enhancement of de-excitation of nuclear levels, then there may be a way to control energy releasing in nuclei. However, to be able to meet the NEET, certain conditions need to be satisfied: such as energy degeneracy between the atomic and nuclear states, and the same transition multipolarity between the states. Therefore, the NEET probability is several orders of magnitude smaller than atomic de-excitation by x-ray emission.

4.3.1 Physics of NEET

When a hole is produced in an inner atomic shell, the electron in the outer shell will like to fill in the vacancy thus producing x-rays or Auger electrons. Morita [107] first suggests the possibility of NEET via absorbing virtual photons. One can imagine the process is quite similar to the inverse internal conversions. The internal conversion process which associated with the gamma-ray decay strongly depends on the energy of the gamma-ray, and the multipolarity of the gamma-ray. The probabilities of radioactive transitions drop quickly with the power of multipole order of the transition. Isomer states in general arise as a consequence of certain hindrances when the radioactive transitions are considerable reduced.

4.3.2 Experiments

Otozai et al. [121] investigated 70-keV states in ^{189}Os and obtained a NEET probability of $(1.7 \pm 0.2) \times 10^{-7}$, and Fujioka et al. [49] for 78-keV state in ^{197}Au with a probability of $(2.2 \pm 1.8) \times 10^{-4}$, both cases using electron beams to produce holes in the K-shells. However, the measured ^{189}Os NEET probability becomes lower and lower as time goes by. Saito et al. [133] measured $(4.3 \pm 0.2 \times 10^{-8})$; Shinohara et al. measured $(5.7 \pm 1.7) \times 10^{-9}$ and Ahmad et al. [5] measured $< 9 \times 10^{-10}$ using synchrotron x-rays. For ^{197}Au, Kishimoto et al. [93] measured $(5.0 \pm 0.6) \times 10^{-8}$ using synchrotron x-rays. These recent results are few orders of magnitude lower than the results from the first measurements. It is possible that the earlier measurements results are due to effects of statistical fluctuations. The ^{237}Np NEET results from Saito et al. [133] $(2.1 \pm 0.6) \times 10^{-4}$ is way too large compared to the theoretical calculations by 8 orders of magnitude (see next section), it possible the measurement result suffers from similar spurious effects as in the earlier ^{189}Os and ^{197}Au measurements.

4.3.3 Theory

Morita calculated NEET probability in ^{235}U via a virtual photon absorption, Using the perturbation theory in his calculations,and suggested using NEET as a possibility of ^{235}U and ^{235m}U separation. Few years later mentioned by Grechukin and Soldatov [57] that Morita uses an inaccurate expression of the electron-nucleus interaction that led significant overestimation of the NEET probability. Pisk et al. [124] assumes that the NEET occurs

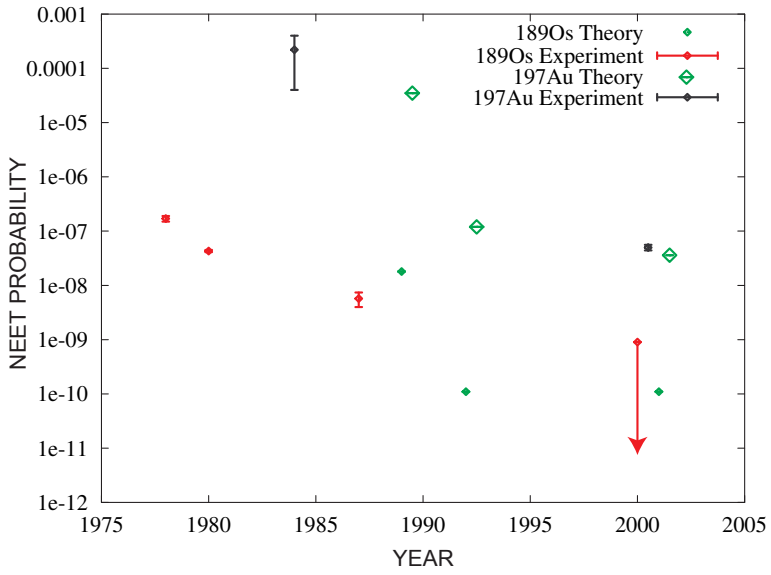

Figure 12: The probabilities for NEET processes have decreased, as experiments and theory have both improved.

during the ionization process with nuclear and electron transitions being simulataneous even if the resonance conditions are not met. They calculated the NEET probabilities of ^{189}Os (1.8×10^{-8} (E2), 2.3×10^{-7} (M1)), ^{197}Au(3.5×10^{-5} (M1), ^{237}Np(1.5E-7(E1) and ^{192}Ir(4.6E-6(M1)). Tkalya [144] revisited Os, Au, Np by using assumptions as in Pisk et al.(89) with a Soldatove code [57] to calculate mean field and electronics wave functions. The calculated the NEET transition probabilities are: ^{189}Os(1.1×10^{-10}), ^{197}Au(1.2×10^{-7}), ^{237}Np(3.1×10^{-12}), ^{192}Ir(2×10^{-11}), and ^{161}Dy(6.6×10^{-11}). These calculation results are low compared to the experimental measurements due to the inability in the experimental setup performed to distinguish between the true NEET process to the inelastic electron-nucleus collisions, direct photo-absorption, and Compton excitation of nucleus level. He suggested that a special coincidence measurement is needed to obtain the true NEET component. In a more recent updated paper by Harston [62], the NEET transition probabilities have been recalculated: ^{189}Os(1.1×10^{-10}), ^{197}Au(3.6×10^{-8}), ^{237}Np(2×10^{-12}).

Apart from ^{197}Au results which have good agreements between the experimental measurements and theoretical calculations, both ^{189}Os and ^{237}Np showed quite large discrepancies. Early measurements seem to have issues with observing spurious signals from statistical fluctuations. Because of these discrepancies, several other nuclei such as ^{229m}Th, ^{235m}U, ^{178m}Ta, etc. have been proposed for clear investigation of the NEET process. Figure 12 shows a plot of the measured NEET probabilities and theoretical results for both ^{197}Au and ^{189}Os with respect to time. As we can see from the plot, earlier measurements suffer from spurios effects and earlier theories overestimate the NEET probability.

4.3.4 ^{178m2}Hf

Recent results by Collins et al. [41] using the Spring-8 facility in Osaka to investigate isomeric ^{178m2}Hf with monochromatic beams of 9-13 keV reported enhanced decay of the isomers at three energies. They ascribed the enhanced decay to the L-shell NEET process with a probability of 2×10^{-3}. This conclusion is also orders of magnitude greater than any reasonable theoretical estimation. For example, detailed calculations by Tkalya [145] demonstrate a range of maximum L-shell NEET probabilities 1×10^{-4} to 1×10^{-11} for nuclear mutipolarities of E1, E2 and M1. More recent theoretical estimation by Karpeshin et al. [89] published in Chinese Physics Letters add the consideration of resonance internal conversion processes, the resulting transition probability could increase by a factor of 800 for E3 transition. However, this additional boost still could not account for the enhancement observed by Collins et al. [41]. A careful review of Collins' results suggesting that summing effects between the baseline ^{214}Bi gamma-ray source used in the experiment that emits gamma-ray at 2447 keV and the monochromatic 9-13 KeV beams may produce a summing peak at 2457.2-keV and was mistakenly interpreted as the direct transistion of the ^{178m2}Hf to the ground state.

5 Controlled and Explosive Energy Release

Here we develop a framework for assessing the performance of systems designed to release energy in a controlled or explosive way. As a starting point we first define a simple metric that can be used to judge a broad variety of systems. This in Section 5.2 is then applied to the particular class of systems that rely on energy stored in nuclear states.

Combining these general considerations with the detailed cross sections from Section 4.2, we are in a position to use standard Monte Carlo transport codes to examine the possible existence of chain reactions wherein a neutron may trigger the release of energy from a whole series of isomeric nuclei, acquiring more energy at each step. In Section 5.3, with its numerical tables, we show the detailed results of such computer simulations. Finally, in Section 5.4 we address some of basic physical requirements that must be satisfied of isomers were to be hypothetically used in gamma-ray lasers.

5.1 Motivation

Our basic goal is to relate the performance of nuclear storage systems to the microscopic (nuclear and atomic) characteristics of these systems. The two are related through details of particle transport. Though nuclear and atomic structure can be complicated, the aspects of particle transport important for energy release depend only on a few fairly simple quantities. Table 3 gives references to equations that describe the relation between performance and microscopic characteristics of systems that rely on energy stored in nuclear isomers.

Figure 13 shows a generic energy release system. Some energy E_{in} is sent in, and corresponding to this some energy E_{out} comes out. A basic metric for the system performance is the gain:

$$G = \frac{E_{\text{out}}}{E_{\text{in}}}. \tag{1}$$

For practical applications one can't usefully use all of the energy that comes out. Here we will ignore the efficiency for recovering useful energy. However, it is important to note that by the same token one can't usefully use all of the energy that is sent *in*. This efficiency cannot be ignored. And for this reason the energy E_{in} is defined to include all of the energy it costs to drive the release system.

As an example of why it is important to consider the total energy input into the system (and not just the part that is useful), consider the simple system shown in figure 14. In this system an accelerator is used to make neutrons. These neutrons hit a big block of copper (the kind found in common pennies) and through absorption release nuclear energy. The energy released by a neutron capturing on copper is

$$E_{\text{out}} \approx 10\,\text{MeV}. \tag{2}$$

This implies that ordinary copper (and many other common materials) contain a tremendous nuclear energy that could potentially be exploited. Ten thousand pounds of copper (costing about $40K at today's prices) hold enough energy to power the United States for about a day.

In principle one could bombard the copper with neutrons of almost arbitrarily low energy. However, this isn't really the issue. The real question has to do with how much energy it costs to make a neutron. With the exception of systems that achieve fusion, the most efficient neutron generators today use about 1000 MeV to make a single neutron. So the gain of our

Table 3: Microscopic Quantities Influencing the Performance of Energy Release Systems

Type	Trigger Particle	Quantity	Note
non-multiplying	neutron	eq. 6	
non-multiplying	photon	eq. 9	eq. 15 for simple absorption
multiplying	neutron	eqs. 23 and 6	transport simulation for accurate estimate
multiplying	photon	eqs. 29 and 9	

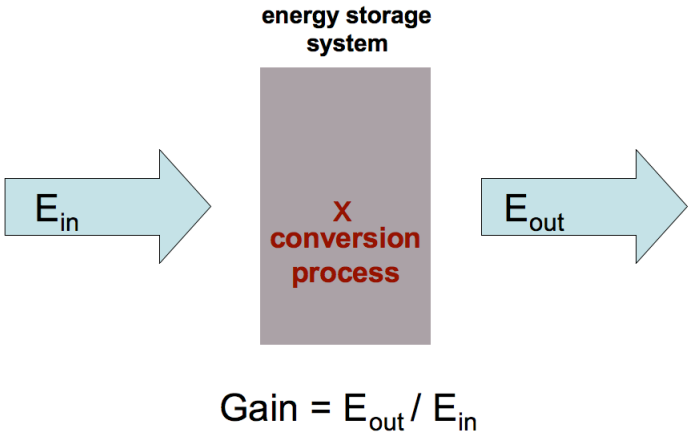

Figure 13: A generic energy release system. An energy E_{in} is sent into the system. This results in an energy E_{out} leaving the system. A basic measure for the efficiency of the system is the gain $G = E_{\text{out}}/E_{\text{in}}$. An efficient system must have a gain larger than one. Note that E_{in} is defined to include all of the energy that it costs to drive the system, and not just the portion that results in useful energy conversion.

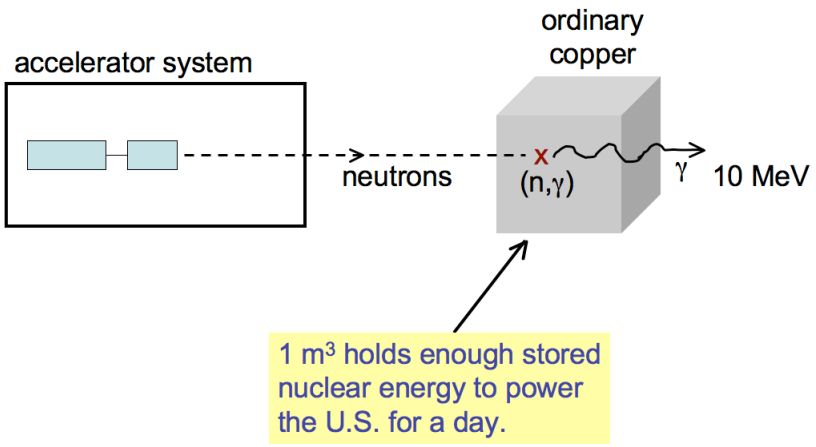

Figure 14: Illustration of an energy release system that exploits the nuclear energy stored in ordinary copper. Though the nuclear energy that could be released from copper and other systems is enormous, current technology cannot be used to efficiently extract this energy.

copper system is

$$G \approx \frac{10\,\text{MeV}}{1000\ \text{MeV}} \approx 0.01, \tag{3}$$

which makes it unsuited for energy production. As far as we know, though, there is no fundamental limit on the cost of making a single neutron. If some method for making neutrons without fission at a cost of a few MeV per neutron were discovered the world's energy problems would be solved in a simple way.

5.2 Microphysics Impacting Energy Release for Nuclear Storage Systems

It is convenient to divide a discussion of nuclear energy release systems according to the role of particles in prompting energy release. There are a few basic cases:

- Spontaneous emission. At this extreme energy release does not depend on sending particles into the system storing the nuclear energy. Energy release may still depend on the ambient conditions. These include density and temperature of the material, electron density, an applied electric field, and so on. We will not consider this case here.
- Particle-induced energy release without particle multiplication. In these systems "trigger" particles are used to induce energy release. Daughters of interactions involving these trigger particles do not play a crucial role.
- Particle-induced energy release with particle multiplication. In these systems the daughters of interactions involving trigger particles go on to produce more energy. The distinction between the multiplying and non-multiplying cases is to some extent artificial. The non-multiplying case is just the limit where multiplication is inefficient, and the two systems are described by the same formalism. But from the perspective of understanding the important physics it is convenient to divide the two.

Laser systems are not described by the cases above and are not considered here.

5.2.1 Energy Release Systems Relying on Particle-Induced Energy Release without Multiplication

In this type of system a trigger particle is sent in to induce energy release. Following a single interaction event (which may itself involve a complicated multi-step microphysics process) the particle is effectively lost. The mean energy released per particle sent in is

$$\langle e_{\rm out}\rangle = \Sigma\langle \epsilon_{\rm interact} e_i \times \frac{\sigma_i}{\sigma_{\rm tot}}\rangle. \tag{4}$$

Here $\sigma_i/\sigma_{\rm tot}$ is the probability for a reaction of type i and e_i is the energy released in a reaction of type i. Angle brackets in this equation denote an average over the distribution of incident particles. This is needed because the incident beam may be characterized by a broad energy distribution, so that different particles in the beam are characterized by different interaction cross sections. The probability for an incident particle to interact with the energy storage medium is given by

$$\epsilon_{\rm interact} = 3\cdot 10^{-3}\left(\frac{\rho L}{1\,{\rm g/cm^3}}\right)\left(\frac{\sigma_{\rm tot}}{1\,{\rm barn}}\right)\left(\frac{200}{A}\right), \tag{5}$$

where ρ and L are the density and linear dimension of the energy storage material and A is the atomic mass of the isotope comprising this material. To maximize the gain one would

want to design a system so that the interaction efficiency is near or larger than unity. For neutron induced reactions with cross sections in the range of a few barns this implies a system with areal density larger than about 300 g/cm^2 (or a linear dimension near 30 cm for material with a density of 10g/cm^3). For photon-induced de-excitation with photons that have energies in the 10 keV range and total interaction cross sections near 10^5 barns a system with areal density near 0.003 g/cm^2 is ideal. For photon-based systems without multiplication an interaction efficiency much larger than unity is not useful.

A discussion of how eq. 4 is applied to energy release from isomeric de-excitation is given below.

(n,n')-induced de-excitation of an isomer The average energy released from a single interaction between neutrons and a target in an excited isomeric state is

$$\langle e_{\rm out} \rangle = E_{\rm exc} \left(\frac{f_n \sigma_{nn'}}{\sigma_{\rm tot}} \right) \tag{6}$$

where $E_{\rm exc}$ is the excitation energy of the isomer, f_n is the fraction of (n,n') reactions that lead to de-excitation of the isomer, and $\sigma_{\rm nn'}$ is the cross section for (n,n') reactions. In the best case every neutron interaction leads to de-excitation and the release of an energy $E_{\rm exc}$. If a way to make neutrons at an energy cost of less than $E_{\rm exc}$ were discovered we could use isomer systems to generate power. If such a discovery were made, we could use the same neutron sources to more efficiently produce energy through (n,γ) reactions on any one of dozens of common stable materials.

Photo-induced de-excitation of an Isomer For these systems there are three important reaction channels:

- Atomic scattering and absorption of incident photons. To a good approximation these processes just deplete otherwise useful photons and do not lead to appreciable energy release.
- Nuclear photo-absorption followed by decay back to the isomer. This is also not useful for energy release.
- Nuclear photo-absorption followed by decay to the ground state. This releases an energy

  $$e_{\rm out} = e_{\rm res} + E_{\rm exc} \approx E_{\rm exc}, \tag{7}$$

  where $E_{\rm exc}$ is the excitation energy of the isomer above the ground state and $e_{\rm res}$ is the energy of the absorbed photon. We will denote the cross section of this process of absorption followed by de-excitation to the ground state by

  $$f\sigma_\gamma, \tag{8}$$

  where f is the fraction of the time that nuclear photo-absorption leads to decay back to the ground state.

Neglecting energy released in the first two types of interactions gives for the average energy release (eq. 4)

$$\langle e_{\rm out}\rangle \approx E_{\rm exc}\langle\frac{\sigma_\gamma f}{\sigma_{\rm tot}}\rangle. \tag{9}$$

This equation applies both to the case of simple photon absorption and to the case where de-excitation is mediated by electron processes resulting from the interaction between incident photons and atomic electrons. To give some insight into the role of beam properties in energy gain systems we will consider the simple absorption case in some detail below.

For the case where direct photon absorption is responsible for de-excitation, the cross section σ_γ for a nucleus in state i to capture a photon and be directly excited to state j can be estimated in a model independent way through appeal to detailed balance:

$$\sigma_\gamma(E = E_{\rm res}) = 2.5\cdot 10^3{\rm b}\left(\frac{1\,{\rm MeV}}{E_{\rm res}}\right)^2\left(\frac{2J_j+1}{2J_i+1}\right). \tag{10}$$

Here J denotes spin and $E_{\rm res}$ represents the excitation energy of the state i relative to the state j.

Only those photons with the right energy to excite the nucleus out of the isomeric state to some higher lying state can be absorbed. At zero temperature the range of photon energies that can be absorbed is given by

$$\Gamma_0, \tag{11}$$

the intrinsic width of the resonantly excited state for decay to the isomeric state. Atoms in material at finite temperature have non-zero velocities and so "see" Doppler shifted photons. The typical shift δE of the apparent photon energy is given by

$$\frac{\delta E}{E_{\rm res}} \approx \frac{v_{\rm thermal}}{c} \approx 10^{-6}\sqrt{\frac{T}{T_0}\frac{240}{A}}, \tag{12}$$

where $v_{\rm thermal}$ is the average thermal velocity, $T_0 = (1/40)$eV is a typical room temperature, and A represents the atomic mass number of the absorbing nucleus. For simplicity we are assuming that the Debye and Einstein temperatures of the material are not greater than about twice the ambient temperature. Under these conditions the behavior of nuclei in the solid with respect to absorption of MeV photons is close to that of atoms in a gas at the same temperature. We note that measured Debye temperatures for actinides are typically in the range 100-200 K and that only a handful of pure materials have a Debye temperature larger than about 600K. Equation 12 suggests the definition of an effective thermal absorption width

$$\Gamma_{\rm thermal} \approx \Gamma_0 + 1\,{\rm eV}\left(\frac{E_{\rm res}}{1\,{\rm MeV}}\right). \tag{13}$$

The thermal average of the resonant scattering cross section is given by

$$\sigma_{\rm NRF} = \sigma_\gamma\frac{\Gamma_0}{\Gamma_{\rm thermal}} \tag{14}$$

for photons with an energy within $\Gamma_{\rm therm}$ of the resonance energy. Photons outside of this window are not efficiently absorbed. The average energy release is then

$$\langle e_{\rm out}\rangle = E_{\rm exc}\left(\frac{f\sigma_\gamma(\Gamma_0/\Gamma_{\rm thermal})}{\sigma_{\rm tot}}\right)\left(\frac{\Gamma_{\rm thermal}}{\Gamma_{\rm beam}}\right). \quad (15)$$

The first term in parenthesis represents the fraction of time that in interaction involving a resonant photon leads to de-excitation of the isomer. The second term in parenthesis represents the fraction of photons in the incident beam that have have the right energy to de-excite the isomer. Both terms are smaller than or equal to one.

The largest possible gain for systems like these occurs if incident photons can be made with perfect efficiency. In this case

$$G^{\rm MAX} = \frac{E_{\rm exc}}{E_{\rm res}}. \quad (16)$$

For an efficient system this gain must be larger than one.

Using the equations above one can relate the requirements for an efficient energy release system to the characteristics of the light source and the nucleus being de-excited. For the sake of illustration let's suppose that the isomer lies at an energy $E_{\rm exc} = 1$ MeV and that $E_{\rm res}$=10 keV. The maximum possible gain is then a factor of 100. If we want to address what nuclear and beam characteristics are needed for an efficient system it is convenient to treat the two terms in parenthesis in eq. 15 separately. The first term describing the fraction of interacting photons that de-excite the isomer is

$$\frac{f\sigma_\gamma(\Gamma_0/\Gamma_{\rm thermal})}{\sigma_{\rm tot}} \approx f\frac{\Gamma_0}{\Gamma_{\rm thermal}}\left(\frac{2.5\cdot 10^7{\rm barns}}{\sigma_{\rm atomic}+2.5\cdot 10^7{\rm barns}\Gamma_0/\Gamma_{\rm thermal}}\right). \quad (17)$$

Here $\sigma_{\rm atomic}$ is the atomic interaction and we have taken the spin factor to be about 1. For hafnium the atomic interaction cross section (not counting elastic coherent scattering) for 10 keV photons is about $6.7{\cdot}10^4$ barns. The requirement that most resonant photons undergo nuclear photo-absorption rather than atomic absorption is then $\Gamma_0 > 0.01$ meV, which is not at all implausible from the perspective of nuclear structure.

If we suppose for our example case that most resonant photons induce de-excitation of the isomer, then the requirement for an efficient system ($G > 1$) is just determined by the condition that the fraction of incident photons with the right energy to be resonantly absorbed is not too small:

$$\frac{\Gamma_{\rm thermal}}{\Gamma_{\rm beam}} > \frac{1}{100}. \quad (18)$$

To gain insight into this equation we need to consider characteristics of real photon beams. Bremsstrahlung sources (common x-ray machines) are characterized by an approximately flat power spectrum and a fractional resolution near unity. If such a source were used to excite a 10 keV resonance, the effective beam width would have to be near 10 keV. Of course it would be possible to use attenuation of the beam to filter out lower energy photons, but this doesn't help the overall energy budget. For a bremsstrahlung machine, then, the requirement for efficient energy gain is

$$\Gamma_0 > \frac{10{\rm keV}}{100} = 100{\rm eV}, \quad (19)$$

which is really implausibly large.

The world's most advanced light sources achieve energy resolutions near 0.1%. For these, efficient gain requires a nuclear state with a partial width

$$\Gamma_0 > 100\,\mathrm{meV} \tag{20}$$

which is still quite large but not implausible. It is worth noting that the current high resolution light sources are very inefficient at making monochromatic photons. Sources relying on the collision between relativistic electrons and laser light to make x-rays, for example, use about 100 MeV to make a single 10 keV photon. With such sources efficient gain is not possible for any nucleus.

5.2.2 Energy Release Systems Relying on Particle Multiplication

In these systems a single incident particle can lead to many reactions. The energy gained per incident particle is

$$E_{\mathrm{out}} = M\langle e_{\mathrm{out}}\rangle \tag{21}$$

where M is the effective multiplication describing the number of reactions induced by a single incident particle and

$$\langle e_{\mathrm{out}}\rangle = \Sigma e_i \langle \frac{\sigma_i}{\sigma_{\mathrm{tot}}}\rangle \tag{22}$$

is again the mean energy released per reaction. Note that in general the spectrum of daughter particles will change from generation to generation. For this reason an accurate solution of eq. 21 typically involves a transport calculation.

The formalism describing particle multiplication in complicated systems is well developed. For our purposes it will suffice to consider the case of an infinite medium, for which the multiplication is as large as possible. The ratio of the number of particles present in a given generation to that present in the previous generation is commonly written as k. In terms of the scattering properties

$$k = \Sigma \nu_i \frac{\sigma_i}{\sigma_{\mathrm{tot}}}, \tag{23}$$

where ν_i is the number of particles emitted in a reaction of type i (zero for absorption, two for an (n,2n) reaction, and so on). The total number of particles created per incident particle is

$$M = \Sigma k^n = \frac{1}{1-k}, \tag{24}$$

for $k < 1$. For $k > 1$ the particle population grows without bound.

(n,n') induced de-excitation of an isomer Nuclear properties determining the efficiency of these systems are described by eqs. 22 and 24. Though the solution of a transport equation is needed to accurately estimate performance, we can make some basic observations with simple arguments. First, it is clear that with (n,n') reactions alone one can't make a critical system. There will always be some contribution from neutron absorption, so that the

number of neutrons in the next generation will be smaller than the number in the current generation.

To give a very schematic sense of the kinds of multiplication possible consider again the example of an isomer with an excitation energy of 1 MeV. Purely for the sake of example, we'll suppose that neutrons with an energy of 1 MeV are sent into the system and that every (n,n') reaction leads to de-excitation of the isomer. For nuclei with mass number near 200 (platinum for example) a typical total (n,n') cross section is about 1 barn, a typical absorption cross section is about 80 mb, and a typical total scattering cross section is about 5 barns. With these numbers

$$k \approx 1 - \frac{\sigma_{\mathrm{n}\gamma}}{\sigma_{\mathrm{tot}}} \approx 0.98 \tag{25}$$

and the multiplication is about 50. We should note that this is a gross overestimate of the multiplication because of the degradation of the spectrum with generation. Both elastic and (n,n') reactions lead to daughter neutrons with less energy than their parents, and the absorption cross section strongly increases with decreasing neutron energy. Solution of the transport equations for an infinite platinum medium give a $k < 0.1$. Assuming though that the spectrum remains constant at 1 MeV gives a mean energy per reaction of

$$\langle e_{\mathrm{out}} \rangle = E_{\mathrm{exc}} \left(\frac{\sigma_{\mathrm{nn'}}}{\sigma_{\mathrm{tot}}} \right) + Q_{\mathrm{n}\gamma} \left(\frac{\sigma_{\mathrm{n}\gamma}}{\sigma_{\mathrm{tot}}} \right) \approx 0.2 + 0.16\,\mathrm{MeV} = 0.36\,\mathrm{MeV}, \tag{26}$$

where we have assumed a 10 MeV q value for absorption. With the numbers above the mean energy released per incident neutron is

$$E_{\mathrm{out}} = M \langle e_{\mathrm{out}} \rangle \approx 18\,\mathrm{MeV}, \tag{27}$$

which is not much better than a simple block of copper.

photo-induced de-excitation of an isomer Stimulated de-excitation of an isomer generally produces a cascade of discrete daughter photons. An estimate for the k eigenvalue is found by averaging eq. 24 over the photons produced following de-excitation

$$k = \frac{1}{\nu} \Sigma_{j=1}^{\nu} \frac{\sigma_{j,\gamma} f_j}{\sigma_{j,\mathrm{tot}}} \nu_j \tag{28}$$

where j identifies a photon in the cascade and ν represents the total number of photons in the cascade. This equation neglects the contribution of elastic and inelastic scattering to the criticality. The different ν_j correspond to the number of photons produced when a photon with energy e_j induces de-excitation. Typically $\nu_j \approx \nu$ and

$$k \approx \Sigma_{j=1}^{\nu} \frac{\sigma_j}{\sigma_{j,\mathrm{tot}}}. \tag{29}$$

Eq. 29 can be used to estimate the criticality of photon-based energy storage systems.

5.3 Monte Carlo Simulations Describing Neutron Transport and Growth in Systems made from Beryllium and the Hafnium Isomer

To quantify energy release from super-elastic (n,n′) de-excitation of the Hf isomer in macroscopic systems we developed a set of Monte Carlo simulations. Monte Carlo simulations were done with Mercury, a 3D particle transport code [User Guide version b.15, LLNL Technical manual, UCRL_TM-204296, Rev.1 (2006)]. Cross section and outgoing particle distribution data used in the simulations was taken from Hauser-Feshbach based evaluations described in Section 4.2.

These provide a reaction-by reaction account of neutron scattering, absorption, and production. A description of the models and data used for the simulations is given below. For the purpose of understanding whether or not one can build an efficient energy generation system or weapon from Hafnium we want to know the effective multiplication of neutrons in the Hafnium assembly. Alternatively, since (n,n′) reactions are responsible for generating energy, we want to know how many inelastic collisions result from each neutron put into the system. This gives a measure of the gain, and is in close analogy with the basic consideration for conventional fission weapons. When a uranium or plutonium assembly becomes super-critical a single neutron will result in a great many fission events and the release of a large amount of energy.

Systems containing beryllium were considered to address the suggestion by [109] that (n,2n) reactions on the beryllium would act as a catalyst for isomer de-excitation. Results of simulations for systems made of beryllium and ^{178m2}Hf are given in Table 4. This shows the number of reactions of different types induced per incident neutron. A range of geometries, incident neutron energies, and compositions was considered. The basic conclusions from this table are:

1. Systems made of ^{178m2}Hf and beryllium, and that rely on super-elastic scattering, cannot be used for weapons or energy release. These systems are calculated to be quite sub-critical. In no case is the number of (n,n′) reactions per incident neutron larger than 1.5.Note that our simulations describe very large Hf assemblies (1 meter radius for the homogeneous mixtures of beryllium and hafnium). More reasonably-sized systems would have smaller efficiencies than calculated here.

2. For the homogeneous mixtures of beryllium and hafnium the efficiency for energy release via super-elastic scattering is a decreasing function of beryllium concentration. This implies that beryllium acts as a poison rather than a catalyst.

To test the sensitivity of our results to assumptions about cross sections for the hafnium isomer we ran a second set of simulations. These take the excited hafnium to be in the 12^+ excited state at 2.15 MeV rather than the 31 y isomeric state. Since this 12^+ state has a much larger super-elastic (n,n′) cross section than the isomer it should represent a sort of upper bound on the possible efficiency of energy generation systems built from hafnium. Note that one couldn't actually build a system from the 12^+ state, since it has a sub-picosecond lifetime.

Results from studies of the 12^+ state are given in table 5. The conclusions are the same as for the isomeric state, although the efficiency of inducing (n,n′) reactions is a few times larger in the case of the 12^+ state. As for the isomer, this efficiency is a decreasing function of the beryllium concentration.

Results given in tables 7 and 8 were obtained for a spherical assembly made of a ^{178m2}Hf core and a ^{9}Be reflector. They show that for 1 MeV neutrons, an energy at which superelastic scattering is the most likely to happen, very few (n,2n) reactions occur in the beryllium reflector. They are several orders of magnitude too low to be used for weapons or energy release. The same is true for a 14 MeV neutron source, where a larger number of (n,2n) reactions occurs in both hafnium and beryllium through neutron downscatter.

5.3.1 Description of Monte Carlo Models

Several models were developed. As a simple check we first modeled a 'broomstick' (very long cylinder with small radius) made of ^{178m2}Hf hit by a pencil beam of mono-energetic neutrons. Results of these validation simulations are given in table 6. Because the cylinder is assumed to have very small radius any neutron that interacts within the cylinder will escape. This implies that the ratio of the number of events for two different reaction types is simply proportional to the ratio of the two reaction cross sections. We checked this by comparing the values in table 6 to the results from the Hauser-Feschbach calculations and found it to be good. We also checked the superelastic cross-sections and energy spectrum of outgoing neutrons and verified consistency with results from the Hauser-Feschbach evaluations. To quantify various reactions in a BeHf mixture, we modeled a 10 cm-radius and a 100 cm-radius sphere of pure ^{178m2}Hf of density 13.31 g/cm^3 and tallied the number of reactions/source neutron occurring in the medium for five source neutron energies ranging from 1 to 14 MeV. The point source was located at the center of the sphere. The pure Hf was then replaced by a mixture of ^{9}Be and ^{178m2}Hf, where the ^{9}Be atom fraction was varied from 0.001% to 95%. The density of the mixture was modified accordingly. We considered two ^{178}Hf states: the 31 y 16^+ isomeric state and the 12^+ excited sate at 2.15 MeV. Results are given in tables 4 and 5.

We also considered heterogeneous assemblies of beryllium and hafnium consisting of a pure sphere of hafnium surrounded by a 50 cm thick beryllium reflector. We modeled four sphere sizes, 10, 30, 50 and 100 cm radius respectively, for the ^{178m2}Hf core. The neutron source was isotropic and monoenergetic, with an energy of either 1 or 14 MeV. We defined two source geometries: a volumetric source with the dimensions of the Hf sphere, or a point source located at the center of the Hf sphere. Results for the 1 and 14 MeV neutron source are given in tables 7 and 8 respectively.

Table 4: Number of reactions/per source neutron in a 100cm radius sphere made of ^{178m2}Hf and ^{9}Be mixture. Column 1 refers to the concentration in atom fraction of ^{9}Be.

	1Mev			2MeV			5 MeV			10MeV			14MeV		
atfr	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)
$1{\cdot}10^{-3}$	4.95	0.470	0.00	6.21	0.713	0.00	7.41	1.34	$2.61{\cdot}10^{-5}$	9.58	1.29	0.391	12.5	1.16	0.823
$1{\cdot}10^{-4}$	4.94	0.470	0.00	6.21	0.712	$2.00{\cdot}10^{-7}$	7.41	1.34	$2.43{\cdot}10^{-4}$	9.59	1.29	0.391	12.5	1.16	0.824
0.01	4.98	0.469	0.00	6.35	0.703	$4.70{\cdot}10^{-6}$	7.70	1.32	0.0120	9.94	1.29	0.398	13.0	1.15	0.828
0.05	5.16	0.464	0.00	6.24	0.711	$1.60{\cdot}10^{-6}$	7.46	1.34	$2.34{\cdot}10^{-3}$	9.65	1.29	0.393	12.6	1.16	0.824
0.10	5.41	0.457	$1.00{\cdot}10^{-7}$	6.50	0.693	$1.07{\cdot}10^{-5}$	8.03	1.30	0.0250	10.3	1.28	0.405	13.4	1.14	0.833
0.15	5.69	0.449	$1.00{\cdot}10^{-7}$	6.86	0.671	$2.22{\cdot}10^{-5}$	8.39	1.28	0.0390	10.8	1.27	0.413	14.0	1.13	0.839
0.20	6.01	0.440	$1.00{\cdot}10^{-7}$	7.07	0.658	$2.78{\cdot}10^{-5}$	8.80	1.26	0.0542	11.3	1.25	0.423	14.5	1.12	0.845
0.25	6.37	0.429	$1.00{\cdot}10^{-7}$	6.67	0.682	$1.66{\cdot}10^{-5}$	9.26	1.24	0.0706	11.8	1.24	0.433	15.2	1.10	0.852
0.30	6.78	0.416	$1.00{\cdot}10^{-7}$	7.32	0.644	$3.10{\cdot}10^{-5}$	9.78	1.21	0.0886	12.5	1.22	0.445	16.0	1.09	0.860
0.40	7.79	0.386	0.00	7.94	0.613	$3.99{\cdot}10^{-5}$	11.0	1.15	0.130	14.0	1.19	0.475	17.8	1.05	0.881
0.50	9.13	0.348	$2.00{\cdot}10^{-7}$	8.80	0.573	$4.55{\cdot}10^{-5}$	12.7	1.07	0.179	16.1	1.13	0.515	20.2	1.00	0.910
0.60	10.9	0.300	$2.00{\cdot}10^{-7}$	10.1	0.521	$4.85{\cdot}10^{-5}$	15.0	0.968	0.240	18.9	1.06	0.571	23.4	0.940	0.952
0.70	13.4	0.243	$1.00{\cdot}10^{-7}$	12.1	0.451	$4.60{\cdot}10^{-5}$	18.2	0.834	0.316	23.1	0.962	0.654	28.1	0.854	1.02
0.80	16.8	0.175	0.00	15.3	0.352	$3.75{\cdot}10^{-5}$	23.1	0.652	0.412	29.7	0.802	0.779	35.4	0.723	1.12
0.90	22.3	0.0944	0.00	21.4	0.210	$2.27{\cdot}10^{-5}$	31.8	0.393	0.537	42.1	0.529	0.985	49.3	0.494	1.32
0.95	27.1	0.0493	0.00	27.2	0.115	$1.14{\cdot}10^{-5}$	40.1	0.219	0.613	54.5	0.313	1.14	63.2	0.301	1.48

Table 5: Number of reactions/source neutron in a 100 cm radius sphere made of a ^{178}Hf and ^{9}Be mixture. Column 1 refers to the concentration in atom fraction of ^{9}Be. Hafnium is here assumed to start in the 12^{+} excited state at 2.15 MeV, imagining this to be an isomer.

	1Mev			2MeV			5 MeV			10MeV			14MeV		
Be-9 atfr	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)	(n,el)	(n,n′)	(n,2n)
$1{\cdot}10^{-4}$	8.28	2.56	$1.00{\cdot}10^{-7}$	10.0	2.99	$4.00{\cdot}10^{-7}$	10.1	3.43	$2.66{\cdot}10^{-5}$	15.5	4.52	0.652	18.4	5.10	0.905
$1{\cdot}10^{-3}$	8.28	2.56	$1.50{\cdot}10^{-6}$	10.1	2.99	$2.80{\cdot}10^{-5}$	10.4	3.42	0.0121	15.6	4.52	0.652	18.5	5.10	0.906
0.010	8.32	2.55	$1.26{\cdot}10^{-5}$	10.0	2.99	$3.10{\cdot}10^{-6}$	10.1	3.43	$2.45{\cdot}10^{-4}$	15.5	4.52	0.652	18.4	5.10	0.905
0.050	8.52	2.53	$6.78{\cdot}10^{-5}$	10.3	2.97	$1.46{\cdot}10^{-4}$	10.1	3.43	$2.36{\cdot}10^{-3}$	16.0	4.50	0.655	18.9	5.07	0.910
0.100	8.78	2.51	$1.31{\cdot}10^{-4}$	10.5	2.94	$2.73{\cdot}10^{-4}$	10.8	3.41	0.0252	16.5	4.47	0.659	19.5	5.04	0.915
0.150	9.09	2.49	$1.93{\cdot}10^{-4}$	10.8	2.91	$4.13{\cdot}10^{-4}$	11.3	3.40	0.0393	17.0	4.44	0.663	20.1	5.00	0.921
0.250	9.81	2.43	$3.00{\cdot}10^{-4}$	11.2	2.87	$5.30{\cdot}10^{-4}$	11.8	3.39	0.0545	17.7	4.41	0.667	20.9	4.97	0.928
0.200	9.43	2.46	$2.47{\cdot}10^{-4}$	11.5	2.83	$6.35{\cdot}10^{-4}$	12.4	3.37	0.0710	18.4	4.37	0.673	21.7	4.92	0.936
0.300	10.3	2.39	$3.50{\cdot}10^{-4}$	12.0	2.79	$7.49{\cdot}10^{-4}$	13.0	3.36	0.0890	19.3	4.34	0.680	22.7	4.88	0.944
0.400	11.4	2.31	$4.28{\cdot}10^{-4}$	14.5	2.57	$1.12{\cdot}10^{-3}$	14.7	3.31	0.130	21.4	4.24	0.697	25.2	4.77	0.965
0.500	13.0	2.20	$4.58{\cdot}10^{-4}$	13.0	2.69	$9.74{\cdot}10^{-4}$	17.1	3.24	0.180	24.4	4.13	0.721	28.6	4.63	0.994
0.600	15.3	2.07	$4.40{\cdot}10^{-4}$	16.6	2.42	$1.20{\cdot}10^{-3}$	20.6	3.13	0.241	28.9	3.99	0.757	33.7	4.46	1.03
0.700	19.0	1.91	$3.93{\cdot}10^{-4}$	19.9	2.22	$1.18{\cdot}10^{-3}$	26.0	2.98	0.316	36.0	3.80	0.813	41.8	4.25	1.09
0.800	25.2	1.71	$2.62{\cdot}10^{-4}$	25.5	1.96	$9.89{\cdot}10^{-4}$	35.4	2.75	0.412	48.3	3.55	0.903	55.8	3.97	1.19
0.900	36.7	1.47	$1.11{\cdot}10^{-4}$	36.5	1.60	$6.07{\cdot}10^{-4}$	53.6	2.40	0.537	73.1	3.18	1.06	84.1	3.57	1.36
0.950	47.8	1.31	$4.23{\cdot}10^{-5}$	47.5	1.36	$3.28{\cdot}10^{-4}$	71.9	2.13	0.614	98.9	2.89	1.18	114.	3.28	1.51

Table 6: Number of reaction/source neutrons in a thin cylinder made of Hf, for several neutron energies E_n.

Hf state	E_n [MeV]	Elastic	(n,n′)	(n,2n)	(n,p)	(n,γ)	(n,α)
	1	0.7498	0.08480			0.1654	
	2	0.7073	0.1497			0.1179	$2.000{\cdot}10^{-7}$
^{178m2}Hf	5	0.5930	0.3952		$4.000{\cdot}10^{-7}$	0.01178	$3.300{\cdot}10^{-6}$
	10	0.4878	0.3031	0.2077	$3.698{\cdot}10^{-4}$	$6.139{\cdot}10^{-4}$	$2.881{\cdot}10^{-4}$
	14	0.5414	0.08279	0.3695	$2.251{\cdot}10^{-3}$	$3.539{\cdot}10^{-7}$	$1.426{\cdot}10^{-3}$
	1	0.6749	0.2279				
	2	0.7073	0.2642			0.028510	$2.000{\cdot}10^{-7}$
Hf 12^+	5	0.5929	0.4047			$2.441{\cdot}10^{-3}$	$2.100{\cdot}10^{-6}$
	10	0.4878	0.1726	0.3386	$2.935{\cdot}10^{-4}$	$4.534{\cdot}10^{-4}$	$2.144{\cdot}10^{-4}$
	14	0.5414	0.05032	0.3965	$2.012{\cdot}10^{-3}$	$2.971{\cdot}10^{-4}$	$1.171{\cdot}10^{-3}$

Table 7: Reaction/source neutron in the ^{178m2}Hf core and ^{9}Be reflector for four core sizes, and two 1 MeV isotropic neutron source geometries. The volumetric source has the dimensions of the Hf core, and the point source is located at the center of the Hf sphere.

		Volumetric source				Point source			
Reaction	Material	r= 10 cm	r= 30 cm	r= 50 cm	r= 100 cm	r= 10 cm	r= 30 cm	r= 50 cm	r= 100 cm
Elastic	Hf core	3.04	4.34	4.61	4.78	3.61	4.89	4.94	4.94
	Be Reflector	172.	51.7	28.5	13.1	117.	3.64	0.0860	$5.28{\cdot}10^{-6}$
(n,n′)	Hf core	0.219	0.368	0.408	0.439	0.304	0.464	0.470	0.470
	Be Reflector	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
(n,2n)	Hf core	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Be Reflector	$3.30{\cdot}10^{-7}$	$1.70{\cdot}10^{-7}$	$1.50{\cdot}10^{-7}$	$7.00{\cdot}10^{-8}$	$3.40{\cdot}10^{-7}$	$3.00{\cdot}10^{-8}$	0.00	0.00
(n,g)	Hf core	0.703	0.923	0.960	0.983	0.798	0.995	1.00	1.00
	Be Reflector	0.162	0.0462	0.0250	0.0112	0.110	$3.26{\cdot}10^{-3}$	$7.51{\cdot}10^{-5}$	0.00

Table 8: Reaction/source neutron in the ^{178m2}Hf core and ^{9}Be reflector for four core radii r, and two 14 MeV isotropic neutron source geometries. The volumetric source has the dimensions of the Hf core, and the point source is located at the center of the Hf sphere.

		Volumetric source				Point source			
Reaction	Material	$r = 10$ cm	$r = 30$ cm	$r = 50$ cm	$r = 100$ cm	$r = 10$ cm	$r = 30$ cm	$r = 50$ cm	$r = 100$ cm
Elastic	Hf core	5.20	9.77	10.9	11.8	7.12	12.3	12.5	12.5
	Be Reflector	565.	199.	114.	53.4	407.	16.9	0.473	$2.79{\cdot}10^{-5}$
(n,n′)	Hf core	0.407	0.842	0.967	1.06	0.604	1.13	1.16	1.16
	Be Reflector	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
(n,2n)	Hf core	0.445	0.673	0.732	0.777	0.618	0.818	0.823	0.823
	Be Reflector	0.799	0.317	0.193	0.0964	0.452	0.0137	$3.05{\cdot}10^{-4}$	0.00
(n,3n)	Hf core	$1.90{\cdot}10^{-3}$	$2.68{\cdot}10^{-3}$	$2.87{\cdot}10^{-3}$	$3.01{\cdot}10^{-3}$	$2.55{\cdot}10^{-3}$	$3.15{\cdot}10^{-3}$	$3.16{\cdot}10^{-3}$	$3.16{\cdot}10^{-3}$
	Be Reflector	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
(n,g)	Hf core	1.01	1.60	1.71	1.78	1.21	1.80	1.82	1.82
	Be Reflector	0.534	0.182	0.103	0.0476	0.384	0.0153	$4.16{\cdot}10^{-4}$	$5.00{\cdot}10^{-8}$

5.4 Gamma-ray lasers

A simple treatment of the γ-ray laser appears in the article by V. Vali and W. Vali [151]. While more complex γ-ray laser schemes have been proposed since this paper appeared, the fundamental challenges of realizing a γ-ray laser are illustrated in the considerations presented in this paper.

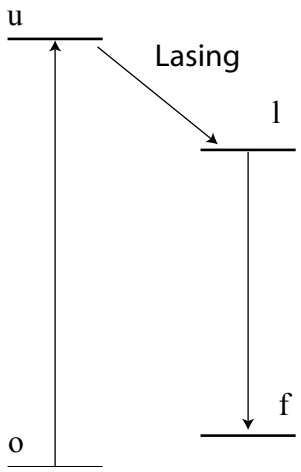

Figure 15: Basic energy levels and state label definitions.

A simple "two-state" laser is assumed. The schematic level diagram is shown in Fig. 15. The system is in the initial state designated "o." It is pumped to the upper state "u" which undergoes a transition to the lower state "l" through spontaneous emission. The system further relaxes to state "f." The wavelength of the laser light is λ_{ul} and the laser state populations are designated: N_o, N_u, N_l, N_f, the number of atoms in the respective states.

The cross section for induced emission of a γ-ray from the nuclei in the excited state "u" is given by the expression:

$$\sigma_0 = \frac{\lambda_{ul}^2}{2\pi} \frac{\gamma_u}{\gamma_u + \gamma_l} \frac{2I_l + 1}{2I_u + 1} \tag{30}$$

where γ_u and γ_l are the transition widths and I_u and I_l are the angular momentum of the upper and lower states. The process by which mono-chromatic γ-rays induce emission from nuclei in the excited state "u" competes with all the other γ-ray scattering processes. The intensity I of γ-rays falls off exponentially with the distance x traveled in side the medium.

$$I = I_0 e^{-N\sigma_e x} \tag{31}$$

where N is the number density of atoms and σ_e is the electronic scattering cross section. This is the sum of the Rayleigh, Compton, photoelectric and pair production cross sections.

The decay rate of the nuclei in the state "u" is governed by its spontaneous decay rate

$$\frac{dN_u}{dt} = -\lambda N_u \tag{32}$$

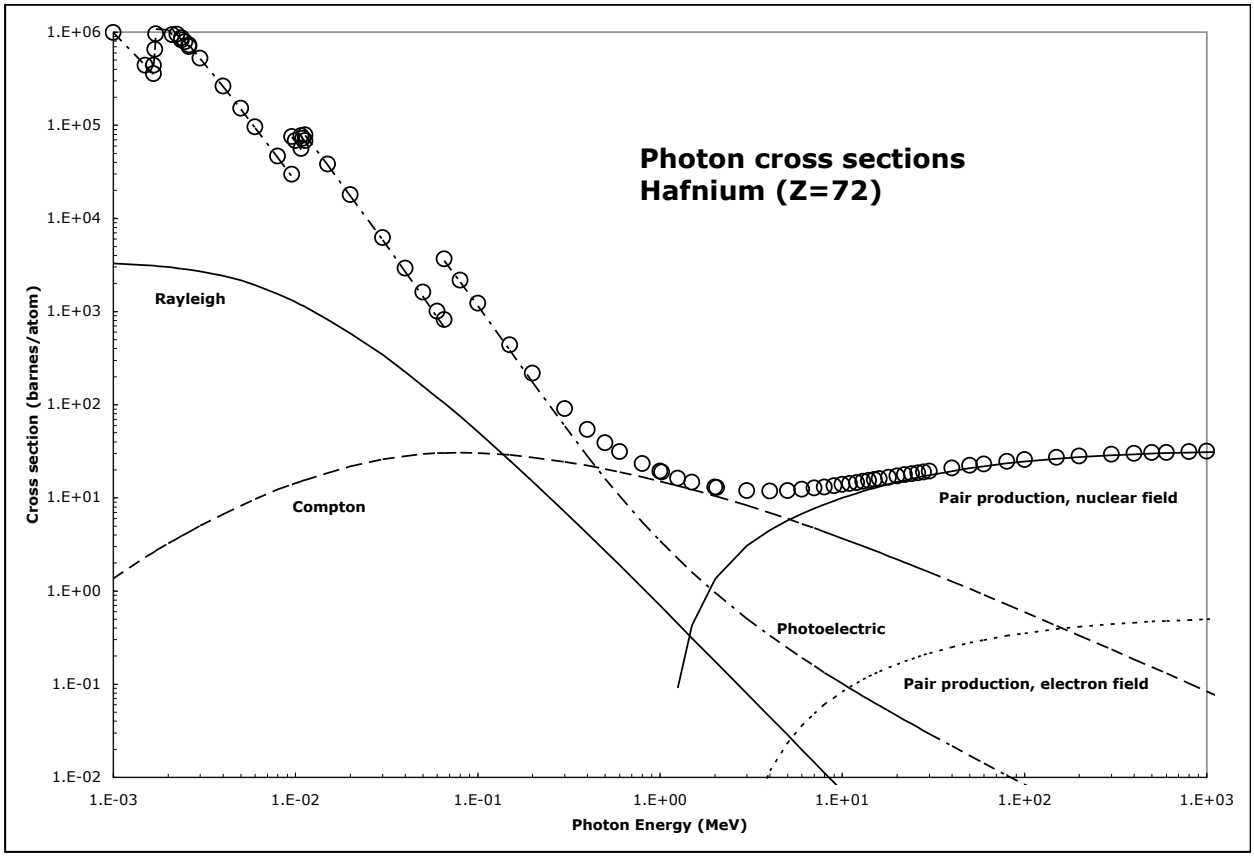

Figure 16: Photon cross section for Hf as a function of photon energy showing the contributions of different processes. The data for this plot was generated from `physics.nist.gov/PhysRefData`.

where λ is the decay constant. If induced emission occurs with some probability p, the decay constant is increased by the factor $(1+p)$ where p is defined by the ratio of cross sections:

$$p = \frac{N_u \sigma_0}{N_l \sigma_l + N\sigma_e} \tag{33}$$

where N_l is the number density of nuclei in state "l" and σ_l is the nuclear resonant absorption cross section from state "l" to state "u." Assuming a high density of excited nuclei "u" and a low density of nuclei "l,"

$$N_l \sigma_l < N_u \sigma_0 \tag{34}$$

and,

$$N_l \sigma_l < N\sigma_e \tag{35}$$

we have

$$p = \frac{N_u \sigma_0}{N\sigma_e}. \tag{36}$$

The property of the system to "lase" is to have more photons participating in the stim-

ulated emission scattering than all other electronic scattering processes, *i*.e.:

$$\frac{N_u \sigma_0}{N_l \sigma_l + N \sigma_e} > 1 \tag{37}$$

which is the condition of criticality. This leads to the requirement on the number density of excited nuclei to all nuclei:

$$\frac{N_u}{N} > \frac{\sigma_e}{\sigma_0}. \tag{38}$$

Because $N_u/N \leq 1$ we find the condition $\sigma_0 > \sigma_e$.

Consider the $\lambda = 3$ electric transition of the 16^+ state to the 13^- state in the 8^--band of the Hf nucleus discussed above. The energy separation is 12.7 keV, or $\lambda_{ul} = 1.5$ nanometer. Assuming the 13^- state undergoes a low multipole electromagnetic transition to the state "f," it would have a half-life of roughly 1 ps, $\gamma_l = 7 \times 10^{11}\ s^{-1}$. The 16^- state has a 31 year half life leading to a transition rate of $\gamma_u = 7 \times 10^{-10}\ s^{-1}$. If there were no other suppression factors (such as line broadening due to nuclear recoil on decay, or line width broadening) the cross section for stimulated emission would be $\sigma_0 = 1.2 \times 10^{-14}$ barns.

The Hf photon scattering cross section for a 12.7 keV γ is $\sigma_e = 6 \times 10^4$ barns. The ratio $\sigma_e/\sigma_0 = 5 \times 10^{18}$ which is much larger than unity. The criticality condition is not met in this case, that is, the probability that the γ-ray undergoes an electronic scatter is much greater than the probability that it would induce a transition of the "u" state to the "l" state with the addition of a second γ-ray.

Even under this very optimistic scenario, it is unlikely that the conditions for producing a γ-ray laser could be met utilizing the ^{178m2}Hf isomer.

More complicated schemes for realizing γ-ray lasers have been proposed (see, for example [160]). Using nuclear isomers as the energetic material requires overcoming the fundamental problem that a long lived isomer state has a very narrow line width. The emitted γ causes the nucleus to recoil, thus Doppler shifting energy of the γ. The line width is so narrow that the emitted γ is "de-tuned." Many schemes involve creating an environment for the nucleus that reduces the effect of the recoil, such as the creation of a Mössbauer crystal. Additional cooling of such crystals is necessary to reduce the Doppler broadening due to the thermal motion of the atoms in the lattice. Finally, some widths are so narrow that the γ's motion in the gravitational potential will de-tune them from resonance, a point noticed in the earliest considerations [151].

A review of the status of γ-ray laser research is written by Elton [47] refers to the article by Collins [34] which applies ideas from an earlier review [10], and other work [35, 36, 120, 119]. The main thrust of this work is to overcome the difficulty presented by the very narrow nuclear state line widths. A review of many proposed γ-ray laser schemes was extensively reviewed by Baldwin and Solem [9]. The difficulty of creating an environment uniform enough so that the nuclei were not subject to energy level shifting effects (*e*.g. strong local magnetic fields) led the authors to conclude that no scheme would work.

Further, they went on to formulate the 'graser dilemma:' that the laser 'pump can destroy the conditions essential to gain.' The conditions required to resolve the graser dilemma can place limits on the characteristics of the candidate nuclear systems proposed as the laser medium.

In this schemes described below, the nuclear isomer is produced separately from the actual time at which it will be used for the laser. In the case of ^{178m2}Hf the concept allows for the production of the laser medium which involves an irradiation of an appropriate feed stock nuclei to produce the isomer, its separation from the stock, and the formation of an appropriate crystal. Assuming concentrations of 10^{22} cm^{-3} it should be noted that the crystal itself is radioactive, roughly 13 mCi.

The first scheme proposed in [34] uses a long lived nuclear isomer which is pumped by a coherent source of optical radiation, Figure 17.

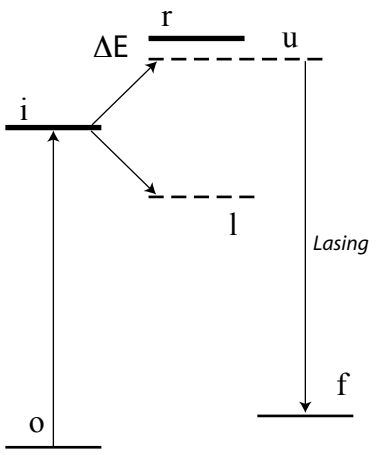

Figure 17: Level scheme for a proposed γ-ray laser with a nuclear isomer "storage" level 'i' and a coherent pump to virtual levels 'u' and 'l'. The frequency of the pump is tuned so that the level 'u' is within a small ΔE of a real state 'r' so that the transition rate from 'i' to 'u' is resonantly enhanced. The state 'u' then undergoes stimulated emission to 'f.'

The population of the 'u' and 'l' virtual states occur through scattering the coherent radiation from 'i.' Choosing an energy value of that coherent radiation such that 'u' is close to a real level in the nucleus, 'r' will enhance the 'i' to 'u' transition and inverts the level population, which then lases from the 'u' to 'f' levels. Level 'r' must exist so that E_i-E_i can be pumped with a laser where ΔE is of order neV (nano-electron-volt) or meV (milli-electron-volt) to benefit from the resonant enhancement. The state 'r' must also have the correct quantum numbers as does the virtual state 'u,' a photon quanta difference from 'i.'

If the coherent pump photon energy is much above 10 eV (soft x-rays) photo-ionization of the medium takes place. The creation of the plasma changes the opacity of the material, usually increasing it and shortening the mean-free path of the pump photons preventing the creation and subsequent inversion of the virtual states 'u' and 'l.' The time over which this happens depends on many factors, but the photon-ionization cross sections are usually much larger than the excitation cross sections of the level 'i' (a factor of 1000 is typical). In the "high energy" pump case, the plasma is being created much faster than the inverted state population.

Keeping the pump photon energy below 10 eV is also problematic, as solid state factors (such as the opacity of metals to near optical photons). The issue for the nuclear structure is the level density near the isomer level 'i,' where the state 'r' is required for the resonant enhancement of the virtual state 'u.'

The incoherent pump scheme described in [34] is shown in Figure 18.

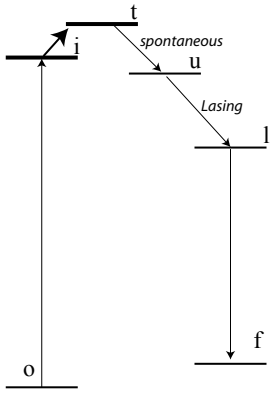

Figure 18: Level scheme for a proposed γ-ray laser with a nuclear isomer "storage" level 'i'.

This sample is then "pumped" from the isomer level 'i' to a presumed band of levels designated "t", referred to as the "K-mixing" band. It is assumed that this band exists close in energy to the isomer level. This band couples the isomer band to some other band (perhaps the ground state band) and the level subsequently decays spontaneously (with a lifetime of 1 to 10 ps) to the upper laser level "u." The properties of this level must inhibit its decay to a length of time comparable to the laser pump time, of order 1 ns. The transition from "u" to "l" proceeds by the induced decay by stimulated emission. Finally, the level "l" must undergo a spontaneous decay with a time short compared to the pumping time to the level "f."

An estimated intensity of the pump is roughly 10 kJ/cm^2 in 10 keV photons. This corresponds to the intensity in that spectral band of a 300 eV black body radiator. These photons have a mean free path in Hf metal of roughly 4 μm. Only 1 in 1000 pump "i" to "t", the remaining photons ionize the Hf atoms creating a hot electron plasma, which shuts off the illumination of the remaining Hf atoms. The pump power then goes into heating the plasma, launching a shock wave into the metal resulting in the subsequent hydrodynamic expansion. This takes place in a time scale short compared to the proposed pump durations of 100ps. This scheme fails to resolve the "graser dilemma."

One final scheme proposed for a graser [165] attempts propose to overcome these problems by using a laser cooled gas of nuclear isomers. Cooling the atoms to μK temperatures is possible using modern atomic physics techniques. At these temperatures, and in a gaseous phase, the nuclei are assumed free from the problems of band shift and spreading. However, *any* laser scheme pumping the nuclear isomer increases the temperature of the gas. In the case of pump photon energies exceeding the ionization potential, there is the additional opacity problems and the radiation-hydrodynamic behavior of a mixed neutral gas - plasma

system. Keeping the atoms cool would require at least as much cooling power as pump power, and would have to establish the low required temperatures for the *duration* of the pump and stimulated decay.

All of these proposed schemes find some possible set of mechanisms for establishing the conditions for producing a γ-ray laser. However, in most cases, the important effects of the reactions of pump photons *other* than the pumping transitions cause a failure to establish the conditions required for lasing.

6 Conclusions and remarks

Our over-arching conclusion from the assessment of the information we reviewed is that the use of ^{178m2}Hf nuclear isomer for energy storage with controlled energy release is improbable. The origins of the stability of nuclear isomers provides the first challenge to the notion. While a precise theory of nuclear structure is lacking, the physical picture which has been built up by the collaboration of theory and experiment can explain the observed phenomena *e.g.* of ^{180}Ta. Reaction and decay pathways which would lead to the rapid decay of this state would prevent any amount of it surviving its synthesis, yet there is a measurable cosmic abundance.

The level schemes of nuclei, of ^{178m2}Hf in particular, are very complex and composed of a huge number of states, yet the structure is well described and the physical character of those states can be explained by the current physical picture. The proposed existence of "new" states with specific quantum numbers can always be verified by careful experimental work.

The exploration of nuclear structure as well as the description of transition probabilities between the various excitation bands also provides a way of estimating the size of decay probabilities and reaction rates. Thus nuclear fluorescence can be used as a tool along with *e.g.* Coulomb excitation, and should present a consistent picture of inter-band and intra-band transitions. In ^{178m2}Hf this is true only by setting aside the experimental results observing large transition rates with small excitation energies.

Ingenious schemes can be imagined for enhancing the transition rates, *e.g.* resonant scattering, or electronic interactions. However, these schemes can be calculated and, in instances where other experiments have been done, compared to those experiments. In no case does the theory support the very large cross sections required to explain the "high rate" triggering results.

To test the ideas of creating a situation were energy release is self-sustaining by the products of the de-excitation reactions, it is possible to use the measured cross sections, or where there is no data, calculate cross sections needed to evaluate the reactivity of the material. We find that, using the best information from nuclear data, no scheme provides a practical energy release process. This is true for all the variety of release mechanisms proposed: neutron scattering, photon scattering, or various laser schemes. The fundamental problem with all these schemes is the relative probability of the relevant reaction to occur for the release is much smaller than the probability of something else happening, and that there is no situation that satisfies the relevant "criticality" conditions that would sustain the various reaction schemes. This is true even if the unexplained large cross sections observed by some groups are taken at face value.

Our conclusion is that the utilization of nuclear isomers for energy storage is impractical from the the points of view of nuclear structure, nuclear reactions, and of prospects for controlled energy release. We note that the cost of producing the nuclear isomer is likely to be extraordinarily high, and that the technologies that would be required to perform the task are beyond anything done before and are difficult to cost at this time.

While additional research in the nuclear physics of isomers would be a worthy academic endeavor, the usefulness of this research for a practical energy source is doubtful.

Acknowledgments

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A Glossary of Concepts

Collective rotation: Due to the location of individual single-particle levels and the presence of a quadrupole-quadrupole interaction, any nuclei exhibit deformed shapes as their lowest energy configuration; most often axially-symmetric prolate shapes. Much like di-atomic molecules, these deformed nuclei can rotate collectively about an axis perpendicular to their symmetry axis (for quantum systems rotation about a symmetry axis is forbidden). The spectrum for collective rotation as a function of total angular momentum J follows the pattern $E = J(J+1)/2\mathcal{I}$, where $\mathcal{I}$ is the moment of inertia and only even values of J are allowed. A schematic of collective rotation is shown in Fig. 19.

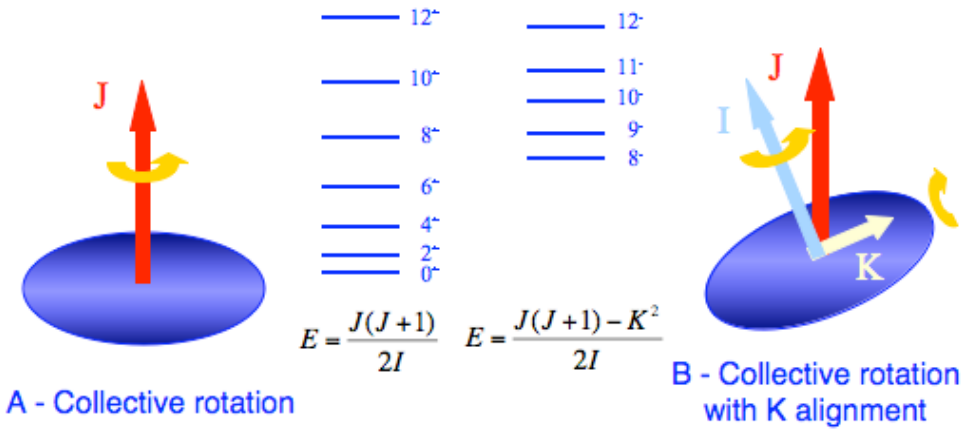

Figure 19: Schematic describing: a) collective rotation of a prolate nucleus and b) collective rotation with K alignment (for illustrative purposes a $K^\pi = 8^-$ band is shown). The overall behavior of the energy levels is also shown.

Compound nucleus: A concept originally introduced by Niels Bohr to describe nuclear reactions when a projectile is absorbed into the target. In general, the "compound" state is excited, and involves many complicated configurations in the nucleus. Because of the very high density of states, the nucleus is not in any one particular state, but posses properties that are statistically distributed across the broad spectrum of states. An important feature of the compound nucleus is that once formed, it loses all memory of its formation.

Decay probabilities: The standard statistical (Hauser-Feshbach) method allocates decay probabilities to each final state in proportion to its transmission coefficient $\mathcal{T}_L$, after scaling so the total probability is unity.

Electromagnetic decay: Excited states in a nucleus lying below the threshold for particle emission will generally decay by emitting electromagnetic radiation (in the form of photons) to reach the lowest energy configuration. Two types of radiation occur, electric, usually denoted by E and is due to re-arrangments in the charge distribution in a nucleus, and magnetic, denoted by M is caused by changes in the distribution of magnetic moments in the nucleus. In addition, radiation is also characterized by its multipolarity, λ, e.g., dipole ($\lambda = 1$), quadrupole ($\lambda = 2$, etc. The multipolarity λ determines the maximum change in the angular

momentum permitted in the transition, e.g., $|J_f - J_i| \leq \lambda$, where here J_i and J_f denote the angular momentum for the initial and final states. In addition, the type, i.e., electric or magnetic, and multipolarity determine the change in parity required in the transition. For electric transitions odd multipolarities require a change in parity, while even multipolarities occur only between states of the same parity. This is reversed for magnetic transitions. General features for electromagnetic decay are that the parity changing electric-dipole ($E1$) transitions are the fastest, followed by the parity conserving electric-quadrupole ($E2$) and magnetic-dipole ($M1$) transitions. In general, decays involving higher multipolarites are suppressed as is also seen in the section defining the Weisskopf unit.

Isomer: Excited, metastable state of an atomic nucleus. The life times of isomeric states can vary over many orders of magnitude; experimental constraints establish $t_{1/2}$= 1 ns (1 nanosecond = 10^{-9}s) as an effective lower life time limit for the purposes of defining an isomeric state, while some isomers with much longer life times also exist (^{180}Ta has a half live of over 10^{15} years). Isomers can decay via α, β, γ-ray emission, or internal conversion. There are three types of isomeric states, *shape isomers*, *spin isomers* and *K isomers*, defined according to the mechanism that hinders their decay. *Shape* isomers can occur when there are two (or more) stable mean-field configurations that represent a local minima in the energy surface. Due to the significant differences in the low-lying structure, for example prolate and oblate shapes, transitions between the lowest states with these configurations tend to be suppressed. *Spin* isomers occur when a high-spin state lies below states of lower angular momentum that would permit an "allowed" electromagnetic decay, i.e., a multipolarity $\lambda = 1, 2$. A K isomer can occur when a given level has alignment K of its total angular momentum along its symmetry axis and all the states lower in excitation energy that are accessible to decay require a large change in K. See e.g.,Walker and Dracoulis [155].

Level density: The level density, or also density of states, is the number of levels per unit energy, for each spin J and parity π. We can also present the total level density, after summing over all J and π. In general, the density of states is known experimentally in two regions of excitation energy. First, at low energies, the level density is determined by counting the observed discrete levels. Second, for excitations just at the neutron separation energy, neutron scattering on stable targets provides the density of states through neutron S-wave resonances, D_0. For much of the spectrum, however, the density of states is modeled with a modified form of the Fermi-gas model, generally attributed to Gilbert and Cameron [53]]. The parameters in the model are tuned to experimental data and extrapolated to other nuclei.

K quantum number: In general, nuclei are not rigid objects, often intrinsic degrees of freedom will couple with the collective rotation. Often, individual particles will couple their intrinsic spins to a value K and align it along the symmetry axis of the rotating prolate nucleus. The intrinsic spin $\vec{K}$ is then coupled with the rotational angular momentum, $\vec{I}$ to form the total angular momentum $\vec{J}$. The spectrum as a function of angular momentum J follows the pattern $E = [J(J+1) - K^2]/2\mathcal{I}$, with $J = K,\ K+1,\ K+2, ...$ The component K along the symmetry axis is very nearly conserved, in a manner very similar to the classical symmetric top. The K quantum number in comparison with collective rotation is shown in Fig 19.

K-hindrance and K-mixing: Strictly speaking, K is not a symmetry of the underlying Hamiltonian, like parity or angular momentum, and thus is not an absolute quantum number to be preserved. However, because of the explicit structure of states with different K, the interaction matrix element, V, connecting them is weak; typically it is of the order 10 eV to

100 eV. Applying first-order perturbation theory, the mixing amplitude between two states is proportional to $V/\Delta E$, where ΔE is the difference in energy between the two states. Thus, when the level density in a system is much less than 10^3 MeV^{-1}, states should be relatively pure, and transitions to different K states will be severely hindered. At higher excitation energies, when the level density reaches 10^{4-6} MeV^{-1} or more, multiple K values can be mixed within each energy eigen-state of the nucleus. In this regime, we have strong K-mixing.

Neutron transmisson coefficient $\mathcal{T}_L$: This, for each angular momentum partial wave L, is the probability from the exterior of populating a compound-nuclear state. And also the time-reversed process: the probability of escape from a initial CN state. The $\mathcal{T}_L$ for particles are calculated from the optical potential for neutron-nucleus scattering, and are essentially proportional to the imaginary parts of these optical potentials. They increase slowly with energy for low L, and are very small but increasing as $\sim E^5$ for large L: see Fig. 6.

Neutron inelastic scattering, direct and compound: When a neutron reacts with a nucleus in its ground state, it can either scatter elastically and keep the same centre-of-mass energy, or it can give energy to the nucleus and come away with less of its own. This second outcome is called 'inelastic scattering', and occurs when the neutron has energy at least equal to the excitation energy of the final state of the nucleus.

When neutrons react with isomers, however, there is a third possible outcome: the nucleus could instead give energy to the neutron. The nucleus would then be left in an energy level below that of the isomer, and then decay more rapidly, whereas the outgoing neutron will have a larger kinetic energy. This processes is called 'superelastic scattering', or 'inelastic scattering by neutron acceleration' (INNA). In contrast to normal inelastic scattering, INNA can occur for any incident neutron energy, no matter how low. If there are large K-hindrances, however, then we should expect the compound nucleus to decay back to the isomer (an example of 'compound elastic scattering').

Parity: A symmetry property exhibited in quantum systems when the Hamiltonian describing the motion is invariant to reflections about the origin. The parity quantum number is often denoted by the symbol π and takes the values $+1$ (even) and -1 (odd). For even-parity states, the wave function has the behavior $\psi(x) = \psi(-x)$ while for odd-parity states, we have $\psi(x) = -\psi(-x)$.

Yrast state: Technical term to define the state of a given angular momentum that has the lowest excitation energy. Derived from Swedish. The root is *yr* which is an adjective meaning to 'whirl'. *Yrast* is the superlative of *yr*, literally meaning "dizziest".

Weisskopf unit: Measure of Electromagnetic transition strength corresponding to the rearrangement of just one nucleon. This is a useful estimate to giving an overall scale to base the strength of a particular transition. In particular, highly collective transitions will have transition rates substantially larger than the Weisskopf estimate, while transition rates lower than the Weisskopf estimate are generally hindered. For a transition of multipolarity λ and γ-ray energy E_γ (measured in MeV), the decay rate, T is

$$T = \frac{B(\lambda+1)}{\lambda[(2\lambda+1)!!]^2}\left(\frac{3}{\lambda+3}\right)^2\left(\frac{E_\gamma}{197\text{ MeV}}\right)^{2\lambda+1}\left(\frac{R}{\text{fm}}\right)^C 10^{21}s^{-1}, \tag{39}$$

where $B = 16$ and $C = 2\lambda$ for Electric transitions and 1.9 and $2\lambda - 2$, respectively, for magnetic transitions, and R is the nuclear radius (often approximated for a nucleus with A

nucleons as $1.2A^{1/3}$ fm). Three important features are evident: 1) electric transitions tend to have a larger decay rate than magnetic transitions; 2) the decay rate increases substantially with increasing γ-ray energy, and 3) for nominal γ-ray energies of the order 1-5 MeV, the decay rate decreases substantially with increasing multipolarity.

B Physical Properties of Hafnium

Symbol: Hf

Atomic Number: 72

Atomic Mass: 178.49

Density: 13,310 kg/m^3

$T_m = 2233°$C

$T_b = 4603°$C

$c_p = 0.144$ J/g K

$C_p = 25.73$ J/mol K

Ground state electron configuration: [Xe]4f^{14}5d^{2}6s^2

Solar abundance Hf/H: 6×10^{-12}

Terrestrial abundance: 5.3 p.p.m.

Seawater: 7×10^{-6} p.p.m.

Table 9: Hafnium isotopes, see the *Nuclear Wallet Cards* distributed at `www.nndc.bnl.gov`

A	Jπ	Δ (MeV)	$T_{1/2}$, Γ or Abundance	Decay mode
153		-27.302	60 ns	
154	0+	-32.733	2 s	ϵ100.00%, α0.00%
154m	(10+)	-30.062	9 μs	IT100.00%
155		-34.102	0.89 s	ϵ100.00%
156	0+	-37.8522	23 ms	α100.00%
156m	8+	-35.8932	0.52 ms	α100.00%
157	7/2-	-38.754	110 ms	α86.00%, ϵ14.00%
158	0+	-42.1041	2.85 s	ϵ55.70%, α44.30%
159	7/2-	-42.8535	5.6 s	ϵ65.00%, α35.00%
160	0+	-45.9372	13.6 s	ϵ99.30%, α0.70%
161		-46.3187	18.2 s	$\epsilon > 99.87\%$, $\alpha < 0.13\%$
162	0+	-49.1731	39.4 s	$\alpha 8.0 \times 10^{-3}\%$, ϵ99.99%
163		-49.2863	40.0 s	ϵ100.00%, $\alpha < 1.0 \times 10^{-4}\%$
164	0+	-51.8215	111 s	ϵ100.00%
165	(5/2-)	-51.6355	76 s	ϵ100.00%
166	0+	-53.859	6.77 m	ϵ100.00%
167	(5/2)-	-53.4678	2.05 m	ϵ100.00%
168	0+	-55.3605	25.95 m	ϵ100.00%
169	(5/2)-	-54.7169	3.24 m	ϵ100.00%
170	0+	-56.2539	16.01 h	ϵ100.00%
171	7/2(+)	-55.4313	12.1 h	ϵ100.00%
171m	1/2(-)	-55.4094	29.5 s	IT100.00%, ϵ
172	0+	-56.4035	1.87 y	ϵ100.00%
173	1/2-	-55.4118	23.6 h	ϵ100.00%
174	0+	-55.8466	2.0×10^{15} y	α100.00%
			0.16%	
175	5/2(-)	-54.483	70 d	ϵ100.00%
176	0+	-54.577	**5.26%**	
177	7/2-	-52.8896	**18.60%**	
177m	23/2+	-51.5741	1.09 s	IT100.00%
177m	37/2-	-50.1496	51.4 m	IT100.00%
178	0+	-52.4443	**27.28%**	
178m	8-	-51.2968	4.0 s	IT100.00%
178m	16+	-49.9982	31 y	IT100.00%
179	9/2+	-50.4719	**13.62%**	
179m	1/2-	-50.0969	18.67 s	IT100.00%
179m	25/2-	-49.3661	25.05 d	IT100.00%
180	0+	-49.7884	**35.08%**	
180m	8-	-48.6469	5.47 h	IT99.70%, $\beta -$ 0.30%
181	1/2-	-47.411	42.39 d	$\beta -$ 100.00%
181m	(25/2-)	-45.6691	1.5 ms	IT100.00%
182	0+	-46.0586	8.90×10^{6} y	$\beta -$ 100.00%
182m	8-	-44.8856	61.5 m	β^-58.00%, IT42.00%
183	(3/2-)	-43.2861	1.067 h	β^-100.00%
184	0+	-41.5013	4.12 h	β^-100.00%
184m	8-	-40.229	48 s	β^-100.00%
185		-38.359	3.5 m	β^-100.00%
186	0+	-36.431	2.6 m	β^-100.00%
187		-32.984	30 s	β^-?
188	0+	-30.879	20 s	β^-

C Production of ^{178m2}Hf

Producing the nuclear isomer ^{178m2}Hf has been the subject of many studies. The initial discovery of ^{178m2}Hf by Helmer and Reich [70] followed a two year irradiation of 100-200 mg of hafnium oxide in reactors with thermal neutron fluxes $> 4 \times 10^{14}$ n/cm^2/s. The samples were allowed to decay for three years. The hafnium was then chemically separate from the sample. In part of the subsequent spectroscopic analysis of the samples, ^{178}Hf was isotopically separated. No estimate of the amount of produced ^{178m2}Hf was provided by the authors.

In a recent paper by Karamian, *et al.* [76] the production cross section for ^{178m2}Hf was measured (along with other isotopes of Hf). From that paper the production of ^{178m2}Hf can be estimated by the expression:

$$N_{178m2} = N_{177} \frac{\sigma_{\text{production}}}{\sigma_{\text{total}} - \sigma_{\text{burnup}}} \left(e^{-\sigma_{\text{burnup}}\Phi} - e^{-\sigma_{\text{total}}\Phi} \right) \tag{40}$$

where Φ is the neutron flux, N_{177} is the amount of ^{177}Hf which serves as the "feed stock" for the production and N_{178m2} is the amount of ^{178m2}Hf produced. The cross sections reported by Karamian, *et al.* provide an estimate for the production.

Table 10: Cross sections relevant to ^{178m2}Hf production

σ_{total} ^{177}Hf(n,X)	$\sigma_{\text{production}}$ ^{177}Hf(n,γ)^{178m2}Hf	σ_{burnup} ^{178m2}Hf (n,X)
373 b	2.6×10^{-6} b	2×10^{-4} b

It is instructive to calculate the total quantity of ^{178m2}Hf that Helmer and Reich would have produced. Starting from 100 mg of HfO_2, with ^{177}Hf at 18.6% abundance the initial amount of "feed stock" would be roughly 16 mg. Estimating the reactor flux for 2 years of running to be $\Phi = 6.3 \times 10^{21}$ n/cm^2 yields roughly 0.075 ng of ^{178m2}Hf.

To obtain gram quantities of ^{178m2}Hf it would require processing 10 metric tonnes of HfO_2.

Accelerator production might be possible *via* the reaction ^{179}Hf(n,2n)^{178m2}Hf. This cross section was evaluated by Chadwick for an unpublished report (Hermannsfeldt, private communication). The cross section at 18 MeV incident neutron energy is calculated to be 10 mb. The shape of the cross section above 18 MeV is uncertain. The total neutron cross section ^{179}Hf(n,X) is approximately 2.5 b. Each incident neutron incident on the ^{179}Hf target makes 0.004 ^{178m2}Hf nuclei, or 250 incident neutrons to make a single ^{178m2}Hf.

Neutrons would be made by accelerating deuterons to high energy and directed onto a Li target to produce neutrons in the appropriate energy range. A thick Li target would yield roughly 1/3 of a neutron out in the energy range of interest. A high intensity machine would accelerate 6×10^{18} deuterons/s/Ampere. The neutron yield would be 2×10^{18} neutrons/s/A. Assuming that 120 MeV deuteron accelerator can be designed and built with roughly 100 mA beam currents, the neutron yield would be 2×10^{17} neutrons/s.

The ^{178m2}Hf production for one year of running would be 2×10^{22} atoms, or roughly 6 g.

Additional issues with accelerator production of ^{178m2}Hf are the enrichment of ^{179}Hf from natural stock, and the processing of the irradiated target to recover the ^{178m2}Hf. There are also considerable technical challenges regarding the accelerator, the Li target and processing the ^{178m2}Hf from the ^{179}Hf target. Finally, not all the cross sections relevant for the estimating production are known.

D Summary of discussions with visitors

The following visitors came to the Lawrence Livermore Laboratory, gave specific isomer presentations to the Assessment Group, and participated in wide-ranging discussions. Sometimes they also gave general seminars to the Livermore nuclear division.

Nicholas and Jirina Stone

Drs Nicholas and Jirina Stone, from Oxford University and University of Tennessee, visited May 20-22, 2008. They discussed with us many details of their report on Mcdaniel's experiments in at the CAMD Facility, Baton Rouge, La., in November 2003.

Nicholas Stone also presented a general seminar 'Short-lived excited state g-factor measurements with Radioactive Ion Beams [RIBs]: new opportunities and limitations of the Recoil-in-Vacuum (RIV) method', and Jirina gave a seminar entitled 'Nuclear Equation of State of High Density Matter'.

William Herrmannsfeldt

Dr Herrmannsfeldt from SLAC, Stanford University, visited June 18, 2008. He summarised his previous work on HIPP, the 'Hafnium Isomer Production Panel' that met during 2002-03, and its as-yet-unpublished report.

Olivier Roig

Dr Roig's permanent affiliation is CEA/DIF/DPTA Service de Physique Nucléaire – Bruyères-le-Châtel, France, but was visiting Los Alamos National Laboratory for February–July 2008. He visited us July 21-22, 2008.

He presented a general seminar 'Experiments with a high spin K isomeric target of $^{177}\mathrm{Lu}^{m}$' to the division, covering his published research in isomer de-excitation by thermal neutrons, and well as a second seminar 'Cross section measurements and more of the INNA reaction on ^{177}Lu' concerning his recent and unpublished attempted to probe the INNA process in more detail.

Ching-Yen Wu

Dr Wu, from the experimental group at LLNL, talked about his collaboration's previous experiments that produced isomers, including ^{178m2}Hf, by Coulomb excitation. He also talked out his group's new experiments to be performed at Argonne Laboratory in August 2008.

Phillip Walker

Professor Walker, of the University of Surrey, United Kingdom, visited July 28-29, 2008. He gave a talk 'Isomers at the interface between atomic and nuclear physics' on the first day, and subsequently joined extensive discussions about isomers and K-mixing.

Yuri Oganessian

Dr Oganessian visited Livermore on September 29 - October 1, 2008. On the first day he gave a general talk 'Heavy ion physics at Dubna', and on the Tuesday he showed slides and discussed his work. He talked about the production and isolation of isomers, laser spectroscopy, K-mixing, and neutron-capture experiments.

James Carroll

Professor Carroll, from Youngstown State University Ohio, gave on Thursday Oct 2 a general talk entitled 'Studies of Nuclear Structure related to an Induced Depletion of Isomers'. Before that, he discussed with us a range of questions about isomer depletion, and also his recently submitted paper [22] about his last experiment at Spring8.

List of Figures

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[24] D. Cline, A. B. Hayes, and C. Y. Wu, *Nuclear structure implications for controlled energy release of isomers*, Journal of Modern Optics, **52** (2005), pp. 2411–2422. **Abstract:** Heavy-ion induced Coulomb excitation has been used to determine level energies and Eλ matrix elements for rotational bands built

on isomeric states in ^{178}Hf. The Eλ matrix elements are consistent with a dramatic increase in K mixing with increasing spin in the ground band, presumably due to Coriolis induced rotation alignment. The spin dependence of the K-distribution for the mixed K ground band states has been deduced. The large K mixing at higher spin provides viable Coulomb excitation paths for stimulated depopulation of high-K isomers in ^{178}Hf.

[25] C. B. Collins and J. Carroll, *Progress in the pumping of gamma-ray laser*, Hyperfine Interactions, **107** (1997), pp. 3–41. **Abstract:** A gamma-ray laser would stimulate coherent emission of radiationat wavelengths below 1 from excited states of nuclei. However, the difficulties in realizing such a device were considered insurmountable when the first cycle of study ended in 1981. Since then, research on the feasibility of a gamma-ray laser has taken on a completely new character. A nuclear analog of the ruby laser has been proposed and many of the component steps for pumping the nuclei have been demonstrated experimentally. A quantitative model based upon the new data and the concepts of this decade shows the gamma-ray laser to be feasible if some real isotope has its properties sufficiently close to the ideals. The greatest positive impact has come from the discovery of giant resonances for pumping nuclei with photons that greatly reduce the levels of input power needed. Most recently, attention has been focused upon efforts to demonstrate prelasing levels of fluorescence from simulation nuclides and actual gamma-ray laser candidates. Problems being addressed are the acquisition of macroscopic samples of the best nuclei for testing and the demonstration of appropriate instrumentation.

[26] C. B. Collins, J. Carroll, Y. Oganessian, and S. Karamian, *Evidence for K mixing in ^{178}Hf*, Hyperfine Interactions, **107** (1997), pp. 141–147. **Abstract:** Systematics for the appearance of K-mixing levels for the pumping or spontaneous decay ofmulti-quasiparticle isomers inHf isotopes are detailed in this letter. The possible location of such a level in the nuclide ^{178}Hf is discussed and an experiment is proposed to investigate its existence.

[27] C. B. Collins, J. J. Carroll, T. W. Sinor, M. J. Byrd, D. G. Richmond, K. N. Taylor, M. Huber, N. Huxel, P. v. Neumann-Cosel, A. Richter, C. Spieler, and W. Ziegler, *Resonant excitation of the reaction $^{180}Ta^m(\gamma,\gamma')^{180}Ta$*, Phys. Rev. C, **42** (1990), pp. R1813 – R1816. **Abstract:** Irradiation of Ta with a superconducting linear accelerator has provided data for the characterization of the reaction ^{180}Ta^m(γ,γ')^{180}Ta. The depopulation of the isomer ^{180}Ta^m via an intermediate state or narrow band of states near 2.8 MeV has been found with an integrated cross section of 1.2×10^{-25} cm^2 keV. This large value exceeds, by nearly an order of magnitude, known cross sections for (γ,γ') reactions producing isomers of other species. Another intermediate state or narrow band is also indicated by the data at an energy 0.6 MeV higher.

[28] C. B. Collins, F. Davanloo, M. C. Iosif, R. Dussart, J. M. Hicks, S. A. Karamian, C. A. Ur, V. I. Kirischuk, J. J. Carroll, H. E. Roberts, P. McDaniel, and C. E. Crist, *Evidence for the Forced Gamma Emission from the 31-Year Isomer of Hafnium-178*, Laser Physics, **9** (1999), pp. 8–11. **Abstract:** A sample containing $0.63*10^{15}$ nuclei of the 4 quasiparticle isomer of ^{178}Hf having a half life of 31 years and excitation energy of 2.45 MeV was irradiated with X-ray pulses derived from a commercial X-ray unit typically used in dental medicine. It was operated at 15 mA to produce Bremsstrahlung radiation with an endpoint energy of 90 keV. With a high purity Ge detector the number of gamma photons collected from transitions between the lower members of the ground state band was found to increase by about 2% above that emitted by spontaneous decay of the isomer. Such an enhancement is consistent with an integrated cross section of $2*10^{-21}$ cm^2 keV, a value in agreement with results scaled from comparable measurements for ^{180}Ta. However, this is believed to be the first observation of photon initiated gamma emission, with a substantial gain in energy. This result has considerable significance to the development of a gamma-ray laser.

[29] C. B. Collins, F. Davanloo, M. C. Iosif, R. Dussart, J. M. Hicks, S. A. Karamian, C. A. Ur, I. I. Popescu, V. I. Kirischuk, J. J. Carroll, H. E. Roberts, P. McDaniel, and C. E. Crist, *Accelerated Emission of Gamma Rays from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation*, Phys. Rev. Lett, **83** (1999), p. 695. **Abstract:** A sample of $6.3*10^{14}$ nuclei of the 4-quasiparticle isomer of ^{178}Hf having a half-life of 31 yr and excitation energy of 2.446 MeV was irradiated with x-ray pulses from a device typically used in dental medicine. It was operated at 15 mA to produce bremsstrahlung radiation with an end point energy set to be 70 or 90 keV. Spectra of the isomeric target were taken with a high

purity Ge detector. Intensities of selected transitions in the normal decay cascade of the ^{178}Hf isomer were found to increase by about 4%. Such an accelerated decay is consistent with an integrated cross section of 1*10^{-21} cm^2 keV for the resonant absorption of x rays to induce gamma decay.

[30] ——, *Reply to Comments* , Phys. Rev. Lett, **84** (2000), p. 2544.

[31] C. B. Collins, F. Davanloo, M. C. Iosif, R. Dussart, J. M. Hicks, S. A. Karamian, C. A. Ur, I. I. Popescu, V. I. Kirischuk, J. M. Pouvesle, P. McDaniel, and C. E. Crist, *Study of the Gamma Emission from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation*, Physics of Atomic Nuclei, **63** (2000), pp. 2067–2072. **Abstract:** A sample containing 6.3×10^{14} nuclei of the 16^+ isomer of ^{178}Hf having a half-life of 31 yr and an excitation energy of 2.446 MeV was irradiated with x-ray pulses from a device operated at 15 mA to produce bremsstrahlung with an endpoint energy of 90 keV. The gamma spectra of the isomeric target were taken with a Ge detector. The intensity of the 325.5-keV (6^+ to 4^+) transition in the ground-state band of ^{178}Hf was found to increase by about 2%. Such an enhanced decay of the ^{178}Hf isomer is consistent with an integrated cross section value of 3×10^{-23} cm^2 keV if resonance absorption occurs within energy ranges corresponding to the maxima of the x-ray flux, either near 20 keV or at the energies of the characteristic emission lines of W.

[32] C. B. Collins, F. Davanloo, A. C. Rusu, M. C. Iosif, N. C. Zoita, D. T. Camase, J. M. Hicks, S. A. Karamian, C. A. Ur, I. I. Popescu, R. Dussart, J. M. Pouvesle, V. I. Kirischuk, N. V. Strilchuk, P. McDaniel, and C. E. Crist, *γ-emission from the 31-yr isomer of ^{178}Hf induced by x-ray irradiation*, Phys. Rev. C, **61** (2000), p. 054305. **Abstract:** A sample containing 6.3*10^{14} nuclei of the 16 isomer of ^{178}Hf having a half life of 31 years and excitation energy of 2.446 MeV was irradiated with x-ray pulses derived from a device operated at 15 mA to produce bremsstrahlung radiation with an end point energy set to be 63 keV. Gamma spectra of the isomeric target were taken with two independent Ge detectors. Intensities of the 213.4 keV (4 →2) and 325.5 keV (6→4) transitions in the ground state band of ^{178}Hf were found to increase when irradiated. The largest enhancement was 1.6±0.3% measured in the 213.4 keV transition. Such an accelerated decay of the ^{178}Hf isomer is consistent with an integrated cross section exceeding 2.2*10^{-22} cm^2 keV if the resonant absorption takes place below 20 keV as indicated by the use of selective absorption filters in the irradiating beam.

[33] C. B. Collins, C. D. Eberhard, J. W. Glesener, and J. A. Anderson, *Depopulation of the isomeric state $^{180}Ta^{m}$ by the reaction $^{180}Ta^{m}(\gamma,\gamma\prime)^{180}Ta$*, Phys. Rev. C, **37** (1988), p. 2267. **Abstract:** The irradiation of an enriched sample of ^{180}Ta^m with bremsstrahlung from a linear accelerator having an end-point energy of 6 MeV has excited a strong channel for the reaction ^{180}Ta$^m(\gamma,\gamma\prime)^{180}$Ta, which requires a total spin change of 8. An integrated cross section of 4.8×10^{-25} cm^{2}keV has been found. This is a large value exceeding by two orders of magnitude known cross sections for (γ,$\gamma\prime$) reactions producing isomers of other species.

[34] C. B. Collins, F. W. Lee, D. M. Shemwell, B. D. DePaola, S. Olariu, and I. I. Popescu, *The coherent and incoherent pumping of a gamma ray laser with intense optical radiation*, J. Appl. Phys, **53** (1982), p. 4645. **Abstract:** Both the coherent and incoherent pumping with intense optical radiation of electromagnetic transitions in nuclei were modeled in this work. In the first case the anti-Stokes upconversion of conventional laser radiation was shown to be a potentially viable means for pumping a γ-ray laser. With the most favorable possible arrangement of nuclear energy levels, the threshold for stimulated output at 10 keV might be reached at a few tens of Joules for isomeric media with lifetimes of the order of tens of years. Whether such an arrangement of excited states actually exists will require additional experimentation. In the case of the incoherent pumping of nuclear material, the resonant excitation of energetic states of nuclei by x radiation from laser plasmas or exploding wires was modeled with results similar to those for the case of coherent pumping. Thresholds for stimulated output could be as low as a few hundred Joules into the primary laser pulse producing the x-ray flash. Again the possibilities were found to be critically dependent upon unknown parameters, particularly upon the efficiency for the conversion of input energy into x rays. The overall possibilities for the excitation of a γ-ray laser with optical pumping were concluded to be reasonably encouraging and the need for additional spectroscopic data was clearly defined.

[35] C. B. Collins, S. Olariu, M. Petrascu, and I. Popescu, *Enhancement of γ-Ray Absorption in the Radiation Field of a High Power Laser*, Physical Review Letters, **43** (1979), pp. 1397–1400. **Abstract**:

This paper reports an analytic estimate of the cross section for the absorption of a γ-ray photon when the nuclear recoil is compensated by the simultaneous absorption of an optical photon from the radiation field of a high-power laser. A multiphoton transition model is developed and cross sections of the order 1 fm^2 are obtained when nearly resonant intermediate states of nuclear excitations are assumed to lie within a few tens of eV of the transition energy.

[36] ——, *Laser-induced resonant absorption of γ radiation*, Phys. Rev. C, **20** (1979), pp. 1942–1945. **Abstract:** This paper reports an analytic estimate developed from a multiphoton model of the cross section for the absorption of a γ-ray photon when the nuclear recoil is compensated by the simultaneous absorption of an optical photon from the radiation field of a high power laser. Tabulated data show 22 experimentally accessible nuclear transitions which might be expected to demonstrate this effect.

[37] C. B. Collins, A. C. Rusu, N. C. Zoita, M. C. Iosif, D. T. Camase, F. Davanloo, C. A. Ur, I. I. Popescu, J. M. Pouvesle, R. Dussart, V. I. Kirischuk, N. V. Strilchuk, and F. J. Agee, *Gamma-Ray Transitions Induced in Nuclear Spin Isomers by X-Rays*, Hyperfine Interactions, **135** (2001), pp. 51–70. **Abstract:** Because of the high density of energy storage and the large cross section for its release, nuclear spin isomers have attracted considerable recent interest. The triggering of induced gamma emission from them has encouraged efforts to develop intense sources of short-wavelength radiation. One of the more interesting examples is the 16^+ 4-qp isomer of ^{178}Hf which stores 2.445 MeV for a half-life of 31 years meaning that as a material, such isomeric ^{178}Hf would store 1.3 GJ/g. Recently, a sample containing 6.3×10^{14} nuclei of the isomer of ^{178}Hf was irradiated with X-ray pulses derived from a device operated at 15 mA to produce bremsstrahlung radiation with end point energies set to values between 60 and 90 keV. Emission of gamma radiation from the sample was increased by 1-2% above the quiescent value of spontaneous emission. Such an accelerated decay of the ^{178}Hf isomer is consistent with an integrated cross section of 2.2×10^{-22} cm^2 keV if the resonant absorption of the X-rays takes place below 20 keV as indicated by the use of selective absorbing filters in the irradiating beam. The work reported here describes the current experimental focus and results recently obtained with the use of coincident detection of emitted gamma photons by several detectors.

[38] C. B. Collins, N. Zoita, F. Davanloo, S. Emura, Y. Yoda, T. Uruga, B. Pattersond, B. Schmittd, J. Pouveslee, I. Popescuf, V. Kirischukg, and N. Strilchuk, *Accelerated γ-emission from isomeric nuclei*, Radiation Physics and Chemistry, **71** (2004), pp. 619–625. **Abstract:** A nuclear-XAFS effect occurs during the scattering of electrons or X-ray photons when the excitation of inner shell electrons couples both energy and angular momentum into nuclear channels of excitation. Nuclear transitions can be induced if the density of excited states of the nucleus enclosed by the electrons is high enough. The 31-yr isomeric state, ^{178m2}Hf satisfies the condition for excitation by monochromatic X-rays from synchrotron sources. Strong effects upon such nuclei are studied.

[39] C. B. Collins, N. Zoita, F. Davanloo, Y. Yoda, T. Uruga, J. Pouvesle, and I. Popescu, *Nuclear resonance spectroscopy of the 31-yr isomer of Hf-178*, Laser Phys. Lett, **2** (2005), pp. 162–167. **Abstract:** Induced release of the high energy densities stored in isomeric nuclear states may be important in the development of ultrashort wavelength lasers. Such a release could compensate the spontaneous power density radiated from the laser medium at threshold. The most promising candidate for such a role seems to be the 31-yr isomeric nucleus of Hf-178 that stores 1.3 GJ/g in the electromagnetic excitation of its constituent protons and neutrons. Successful studies of the induced release of energies from such isomeric states have required an extension of techniques for nuclear resonance spectroscopy using synchrotron radiation (SR) that had previously been applied only to ground state materials. In 2004, monochromatic X-rays from the SPring-8 SR source were used to identify one of the excited nuclear states that mediates the induced decay of the 31-yr isomer of Hf-178. That trigger level was found to lie at 2457.20(22) keV. It was excited when an isomeric nucleus absorbed an incident X-ray photon. We found that one branch of its subsequent decay consisted of a strong electromagnetic transition to the ground state of the nucleus. The energy of the γ -photon emitted was equal to the energy of the trigger level. Proximity in energy of that level to the energy of 2446.06 keV stored by the isomer makes it easy to induce a release of the stored energy and encourages prospects for the development of a gamma ray laser.

[40] C. B. Collins, N. C. Zoita, F. Davanloo, S. Emura, Y. Yoda, T. Uruga, B. Patterson, B. Schmitt, J. M. Pouvesle, I. I. Popescu, V. I. Kirischuk, and N. V. Strilchuk, *Accelerated Decay of the 31-yr Isomer of Hf-178 Induced by Low-Energy Photons and Electrons*, Laser Physics, **14** (2004), pp. 154–165. **Abstract:** A process for transferring energy from electron shells into nuclear excitation (NEET) was first proposed by Masato Morita in 1973. It was generalized to a broader class of electron bridging mechanisms (EBM) by Matinyan. These proposals offered an exciting promise for modulating nuclear properties with X-ray photons at accessible levels of power. The development of 3rd generation synchrotron radiation (SR) sources made it possible to conclusively demonstrate NEET/EBM in two recent experiments by exciting nuclear levels of ground state ^{197}Au and isomeric ^{178}Hf^{m2}, respectively, with SR X-rays from SPring-8. X-ray energies incident upon the isomeric target were tuned from 9 to 13 keV, and an excitation branch attributed to NEET/EBM was found to have a probability of 1.6×10^{-3} relative to L-shell photoionization of the surrounding electrons. The rate for the resulting emission of exoergic γ photons approached the theoretical maximum for such processes. These landmark results were reconfirmed and extended in a series of experiments at SPring-8 conducted in May 2002. Increases in the rates of spontaneous decay of the isomeric nuclei, ^{178}Hf^{m2}, were studied when targets were irradiated with monochromatic SR X-rays tuned over hundreds of eV with spectral widths of both 1.0 and 0.1 eV. A close correlation of accelerated gamma emission with rates of ionization of the L-shell electrons in the target was observed. The frequent appearance of a triplet structure in the excitation spectrum for induced gamma emission resembled the additional structure seen in XAFS studies of the inner-shell electrons at energies near the ionization edges. In addition to the accelerated emission of some gamma lines present in spectra of spontaneous decay, new lines were also observed. Features in the excitation functions for such lines suggested that electrons as well as X-ray photons could initiate the accelerated decay of isomeric nuclei. In 2003, measurements of the temporal correlations between the pulses of SR stimulation and the fast response of the resulting gamma emission were studied successfully at the SR sources (SLS) at the Paul Scherrer Institute and at SPring-8. Results encourage prospects for applications depending on the control of nuclear properties with X-rays.

[41] C. B. Collins, N. C. Zoita, A. C. Rusu, M. C. Iosif, D. T. C. F. Davanloo, S. Emura, T. Uruga, R. Dussart, J. M. Pouvesle, C. A. Ur, I. I. Popescu, V. I. Kirischuk, N. V. Strilchuk, and F. J. Agee, *Tunable synchrotron radiation used to induce γ -emission from the 31 year isomer of ^{178}Hf*, Europhys. Lett, **57** (2002), pp. 677–682. **Abstract:** A process for transferring energy from electron shells into nuclear excitation, NEET, has offered the promise for modulating nuclear properties at accessible levels of power. It had been proven recently by exciting a nuclear level of 197 Au with synchrotron radiation, but measured couplings were far below theoretical ob jectives. Reported here is an extension of that approach for excitation to ^{178}Hf^{m2} isomeric nuclei. Isomeric targets were irradiated with X-rays in the beamline BL01B1 at the synchrotron radiation source SPring-8. Energies were tuned from 9 to 13 keV. In this range an excitation branch attributed to NEET was found to have a probability of $2*10^{-3}$ relative to L-shell photoionization. The resulting emission of exoergic γ-photons was observed from the target at a rate approaching the theoretical maximum.

[42] R. D'Alarcao, P. Chowdhury, E. H. Seabury, P. M. Walker, C. Wheldon, I. Ahmad, M. P. Carpenter, G. Hackman, R. V. F. Janssens, T. L. Khoo, C. J. Lister, D. Nisius, P. Reiter, D. Seweryniak, and I. Wiedenhoever, *High-K isomers in neutron-rich hafnium nuclei at and beyond the stability line* , Phys. Rev. C, **59** (1999), pp. R1227 – R1231. **Abstract:** Pulsed ^{238}U and ^{208}Pb beams have been used to populate multi-quasiparticle high-K isomers in neutron-rich hafnium isotopes at and beyond the line of -stability, via inelastic excitation and transfer. Spectroscopic properties and configuration assignments of several new high-K isomers are compared with earlier theoretical predictions. A striking example of the robustness of the K quantum number is demonstrated by the observed competition between E1 and E3 decay modes in ^{180}Hf, the heaviest stable isotope of the element.

[43] S. Deylitz, B. D. Valnion, K. E. Abiary, J. de Boer, A. Gollwitzer, R. Hertenberger, G. Graw, R. Kulessa, C. Briançon, D. L. Du, R. Meunier, M. Hussonnois, O. Constantinescu, S. Fortier, J. B. Kim, L. H. Rosier, G. Rotbard, Y. T. Oganessian, S. A. Karamian, H. J. Wollersheim, H. Folger, J. Gerl, T. Happ, and C. Hategan, *Inelastic deuteron scattering from the high-spin isomer ^{178}Hf^{m2} (16^{+})*, Phys. Rev. C, **53** (1996), pp. 1266–1272. **Abstract:** In inelastic scattering of 22 MeV deuterons from

an isotope-separated target of ^{178}Hf containing about 2×10^{13} nuclei in the isomeric 16^+ state we observed rotational excitation to the 17^+ state at an excitation energy with respect to the isomeric state of 356.5±0.4 keV and weak evidence for the 18^+ state at 737±2 keV. We compared the differential cross sections with coupled-channel calculations and with scattering from ^{178}Hf (0^+) and ^{177}Hf ($7/2^-$).

[44] G. Dracoulis, *High-K Isomerism*, Nuclear Physics A, **752** (2005), pp. 213–222. **Abstract:** Recent results on the identification of high-K isomers in the Yb-Lu-Hf-Ta region populated using deep-inelastic reactions are presented in the context of the expectations for the formation of multi-quasiparticle states close to and to the right of the stability line. Factors that affect the purity of the K-quantum number include chance degeneracies, which provide an opportunity to extract mixing-matrix elements, and statistical mixing due to the presence of high level densities above the yrast line.

[45] V. Dzjamko, I. Sokolyuk, and T. Zajac, *A mechanism for excitation of metastable levels by (γ,γ') reactions*, Hyperfine Interactions, **107** (1997), pp. 175–183. **Abstract:** The analysis of the cross-section of 77Se and 87Sr isomeric states excitation in the (γ,γ') reaction, was carried out using neutron pick-up and stripping reaction results. It allows the shell configurations of the transitions to the activation states to be determined. To check thus obtained conclusions the energies of $J^\pi = 1^-$ excited states for 90Zr and 138Ba were calculated, which are in good agreement with the resonance structure at the γ-quanta elastic scattering in the 5 to 10 MeV energy range.

[46] A. Y. Dzyublik, V. Méot, and G. Gosselin, *Decay of Isomers Stimulated by Laser Radiation*, Laser Physics, **17** (2007), pp. 760–764. **Abstract:** A theory is presented for the induced decay of isomer levels for nuclei exposed to a laser field, which couples the initial isomer level to intermediate higher lying levels with short lifetimes. We took into account the electric dipole transitions between these levels in a one-photon approximation. The time-dependent Schrödinger equation with a periodic Hamiltonian is solved by the method of the composite Hilbert space. A simple expression for the broadening of the isomer level ΔΓ, caused by the laser radiation, is derived. As an example, we considered the decay of the isomer level 970.17 keV with the spin $I^\pi = 23/2^-$ of ^{177} Lu, coupled by the laser wave to the virtual level 1352.33 keV with $I^\pi = 21/2^+$. The ratio of the broadening ΔΓ to the natural width of the isomer level Γ is found to be about 40% for a 10^{17} W/cm^2 laser. Any influence of the electronic environment is not taken into account.

[47] R. C. Elton, *X-Ray Lasers*, Academic Press, Inc., 1990. ISBN: 0-12-238080-0.

[48] J. P. Farrell, V. Dudnikov, J. J. Carroll, and G. Merkel, *Beam-Based Production of ^{178m2}Hf*, Hyperfine Interactions, **143** (2002), pp. 55–61. **Abstract:** In this study, the production yield for the reaction ^{176}Yb(9Be, a3n)^{178}Hf was explored using the FN tandem injected superconducting LINAC at SUNY at Stony Brook at a ^{9}Be energy of 65 MeV. By comparing the experimental yield of ^{178}Hf ground state γ rays with those of 180 W as a function of energy, the cross section for production of the incomplete fusion γ rays in ^{178}Hf was evaluated. Coincidence measurements were made to get information about the population strength of the high spin states in ^{178}Hf. From these measurements, the maximum cross section for the reaction ^{176}Yb(^{9}Be, a3n)^{178m2}Hf is estimated to be no larger than 5 mb.

[49] H. Fujioka, K. Ura, A. Shinohara, T. Saito, and K. Otozai, *Observation of nuclear excitation by electron transition (NEET) in ^{197}Au*, Z. Phys. A, **315** (1984), pp. 121–122. **Abstract:** Nuclear excitation was observed in ^{197}A when the Au atom was ionized by pulsed electrons of 100 keV. It is attributable to NEET of Ml interaction. The NEET probability was found to be $(2.2\pm1.8)\times10^{-4}$.

[50] J. P. Gangrskii, *Charge radii of high-spin isomers measured by laser spectroscopy methods*, Physics of Particles and Nuclei, Letters, **4** (2007), p. 518. **Abstract:** It is presented the review of experimental data about the charge radii differences between the ground and high spin isomeric states. The methods of high resolution laser spectroscopy were used for the measurements of the differences. There are compared charge radii differences obtained by two ways: from the measurements of atomic levels isomeric shifts and from measurements of quadrupole moments for both levels under the assumption that the charge radii differences are induced by the difference of their quadrupole deformations. There are considered the quasiparticle isomer obtained at the rapture of the nucleon pairs, 1h11/2 isomers in the region of Cd-Ba nuclei and 1i13/2 isomer in the Hg-Pb region. The various behavior of obtained charge radii differences for different nuclei is discussed.

[51] D. S. Gemmell, *Nuclear Transitions Induced by Synchrotron X-rays*, AIP Conf. Proc, **652** (2003), pp. 239–249. **Abstract:** We discuss two rare but interesting processes by which synchrotron x-rays with energies up to about 100 keV may be used to induce nuclear transitions. In the NEET (Nuclear Excitation by Electronic Transition) process, an intense x-ray beam is employed to make vacancies, e.g. K-holes, in the atoms of a specific nuclear isotope. When a vacancy is filled by an electronic transition from a higher atomic level, there is some probability that instead of the usual x-ray or Auger emission, the nucleus of the atom itself will be excited. This is then followed by a nuclear decay exhibiting characteristic gamma-rays or other types of radiation, with time delays typical of the nuclear states involved. The probability for NEET increases when the energies of the atomic and the nuclear transitions become close. We address some theoretical aspects of the process and describe experimental efforts to observe it in ^{189}Os and ^{197}Au. The second process to be discussed is the possibility of "triggering" the decay of a nuclear isomer by irradiation with an x-ray beam. We focus on the case of the 31-year, 2.4-MeV, 16^+ isomer of ^{178}Hf. There has been speculation that if one could isolate gram quantities, say, of this isomer and then have the capability to accelerate its decay in a controlled way, one would have a powerful triggerable source of enormous energy. This could be used to generate explosions, for rapid irradiations, or for more general energy-storage applications, depending on the rate of energy release. We describe attempts to observe this process.

[52] E. Geva and R. Kosloff, *Three-level quantum amplifier as a heat engine: A study in finite-time thermodynamics*, Phys. Rev. E, **49** (1994), pp. 3903 – 3918. **Abstract:** The finite-rate performance of a quantum heat engine, constructed from a three-level amplifier, is analyzed. Consistent definitions of thermodynamical quantities in terms of quantum observables are postulated. The performance is analyzed in steady state, where the operation of the amplifier only influences the surroundings. Quantum master equations describe the irreversible dynamics induced by the coupling of the working medium to the reservoirs. It is shown that the standard assumption of field-independent dissipation is inconcistent with thermodynamics. Field-dependent relaxation equations, based upon the semigroup approach, and consistent with thermodynamics, are formulated. These equations are valid if the time scale of the external field is slow compared to that associated with the bath fluctuations. The steady-state values of the thermodynamical quantities are evaluated. The power is found to have maxima as a function of important controls, such as the field amplitude, frequency, and the coupling with the baths. The existence and locations of these maxima differ from those obtained in the standard treatment, where the dissipation is field independent. The irreversible nature of engine operation is due to the finite rate of heat transfer and a genuine "quantum-friction'' loss term due to dephasing.

[53] A. Gilbert and A. Cameron, *A composite nuclear-level density formula with shell corrections*, Can. J. Physics, **43** (1965), p. 1446.

[54] I. Goncharov, A. Dovbnya, Y. Ranyuk, and O. Shevchenko, *Longliving ^{178m2}Hf, ^{172}Hf, ^{150}Eu and ^{133}Ba Isotopes Photoproduction Study*, Problems Of Atomic Science And Technology, N5. Series: Nuclear Physics Investigations, **48** (2007), pp. 22–25. **Abstract:** The yield of the ^{178m2}Hf (T = 31y), ^{172}Hf(1.87), ^{150}Eu (36.9) and ^{133}Ba (10.5) isotopes was measured after irradiation of a tantalum target with 1.0 GeV Kharkiv linear accelerator electron beam. The integral and average cross-sections are obtained. The results are discussed and compared with another data. The number of produced ^{178m2}Hf atoms in tantalum target is $(2.7 \pm 0.3)\ \ 10^{11}$.

[55] B. Gorski, S. A. Karamian, Y. T. Oganessian, and S. L. Bogomolov, *Determination of Microquantities of Hafnium in Rees*, Journal of Radioanalytical and Nuclear Chemistry, **170** (1993), pp. 353–358. **Abstract:** A method for the determination of Hf in an Yb matrix using the reaction 176Hf(α,3n)177W is described. The excitation function of this reaction was measured and the 4He ion energy of 36 MeV was chosen. 177W was determined by γ-spectrometry after chemical isolation by means of extraction chromatography with tri-n-butyl phosphate. Calibration measurements were carried out with known quantities of ^{nat}Hf. The sensitivity limit obtained was at the level of 3*10^{-8} g/g. The Hf admixtures in ^{nat}Yb and 176Yb samples were estimated quantitatively.

[56] C. Granja, J. Kuba, A. Haiduk, and O. Renner, *Survey of nuclei for low-energy nuclear excitation in laser-produced plasma* , Nuclear Physics A, **784** (2007), pp. 1–12. **Abstract:** We present a survey of stable and long-lived nuclei as well as nuclear isomers looking for candidates of studies of low-energy

(1-30 keV) nuclear excitation by laser-produced plasma radiation. We concentrate on medium-size high-power lasers with pulse duration of hundreds of ps providing energy up to 1000 J and subrelativistic intensity of 10^{16} -10^{17} W cm^{-2} . Screening criteria are primarily the transition energy and the half-life, spin and parity of nuclear levels. ^{181}Ta is suggested as first candidate for which an estimation of reaction efficiency is included.

[57] D. Grechukhin and A. A. Soldatov, , Institute of Atomic Energy report, **2706** (1976).

[58] O. Grudzevich, A. Zelenetskij, A. Ignatyuk, and A. Pashchenko, *Neutron Reactions with Nuclei in Isomeric States*, International Atomic Energy Agency INDC(CCP)-357, (1993). **Abstract:** The authors give theoretical calculation results for the cross-sections of reactions (n,n′) and (n,2n) with high-spin target nuclei in the excited state. It is shown that an increase in the target nucleus excitation energy shifts the cross-section curve to the left along the energy axis, while an increase in the ground state spin considerably reduces the (n,2n) reaction cross-section in the threshold region of the reaction. In the authors' opinion, this change in the excitation functions is due to a sharp rise in the role of gamma competition because of the effect of the spin conservation law on neutron emission.

[59] N. A. Guardala, J. L. Price, and J. H. Barkyoumb, *Preparation and De-excitation Measurements of ^{177m}Lu Using Fast Neutrons*, Hyperfine Interactions, **143** (2002), pp. 63–68. **Abstract:** The $t_{1/2}$ = 160.4 d isomer 177m Lu has many attractive features related to inducing gamma-ray decay to the ground state of the $t_{1/2}$ = 6.4 d 177Lu. Also, as opposed to other isomers it maybe possible to produce it in quantities without the need for extravagant or costly methods. An experimental approach is outlined for both studies using fast neutron inelastic scattering and short-pulse bremsstrahlung radiation to produce the rapid release of gamma rays from this isomer. Furthermore, cross sections for production of the isomer using fast neutrons with energies between 10-500 keV will also be studied.

[60] A. Hague, R. Casten, I. Förster, A. Gelberg, R. Rascher, R. Richter, P. V. Brentano, G. Berreau, H. Börner, S. Kerr, K. Schreckenbach, and D. Warner, *A study of the low-lying states in ^{178}Hf through the (n,γ) reaction*, Nuclear Physics A, **455** (1986), pp. 231–293. **Abstract:** The nucleus ^{178}Hf was studied through thermal neutron and averaged resonance neutron capture reactions. The γ-ray and conversion electrons were measured with high resolution spectrometers. A level scheme up to an excitation energy of ∼ 2.1 MeV was constructed. It includes ∼ 65 levels, most of which are ordered into 18 rotational bands. The level scheme is complete up to about 1800 keV for spins between 2 and 5. The neutron binding energy was established to be at 7626.3 (3) keV. The consistent Q form of the IBA-1 (CQF) was used to describe the low-lying collective γ and $K^\pi = 0^+$ bands. The agreement with the data was found to be excellent for the energies and B(E2) ratios of the ground and γ bands, whereas the agreement was poor for the $K^\pi = 0^+$ bands.

[61] J. H. Hamilton, A. V. Ramayya, R. M. Ronningen, R. O. Sayer, H. Yamada, C. F. Maguire, P. Colombani, D. Ward, R. M. Diamond, F. S. Stephens, I. Y. Lee, P. A. Butler, and D. Habs, *Isometric trapping following Coulomb excitation of high spin states in ^{178}Hf*, Physics Letters B, **112** (1982), pp. 327–330. **Abstract:** Coulomb excitation of states in 178, 180Hf has been observed to 16^+ and 12^+, respectively, with Kr and Xe ions. A strong up bend in the moment of inertia above the 12^+ level is seen in ^{178}Hf and is well reproduced by cranked shell model calculations. For the first time in Coulomb excitation, population of the lowest K[pi] = 8- isomer in ^{178}Hf was observed.

[62] M. R. Harston, *Analysis of probabilities for nuclear excitation by near-resonant electronic transitions*, Nuclear Physics A, **690** (2001), pp. 447–455. **Abstract:** Theoretical results are presented for nuclear excitation of low-lying isometric states of ^{197}Au, ^{189}Os and ^{237}Np by a near-resonant electronic deexcitation process known as NEET. A detailed comparison is made between current and previous theoretical results in order to clarify a number of anomalies. For ^{197}Au, the only case in which the current experimental precision appears to be sufficient to provide a stringent test of theory, the theoretical result for the NEET probability is P_{NEET}=3.6×10^{-8}. This is a factor of three lower than previous theoretical results and reduces the difference between theory and the recent experimental result, P_{NEET}=(5.0±0.6)×10^{-8}, to a level of approximately 2σ.

[63] M. R. Harston and J. J. Carroll, *Nuclear Excitation and Deexcitation in Resonant Electronic Transitions*, Laser Physics, **14** (2004), pp. 1452–1456. **Abstract:** Two nuclear processes involving a nuclear

transition that is resonant with an electronic transition are discussed. In one, known as BIC, the nucleus de-excites by excitation of an electron to a bound orbital, and thus this process constitutes a subthreshold contribution to the nuclear decay rate. In the second process, known as NEET, nuclear excitation occurs by de-excitation of an initially excited electronic state. The current status of experiment and theory for these two processes is reviewed. Theoretical results for NEET for different electronic transitions in several nuclei are presented in order to consider upper limits on the NEET probability.

[64] ——, *Limits on Nuclear Excitation and Deexcitation of $^{178}Hf^{m2}$ by Electron-Nucleus Coupling*, Laser Physics, **15** (2005), pp. 487–491. **Abstract:** Nuclear excitation in transitions that are resonant with electronic deexcitations are discussed in the context of recent claims of the triggering of the decay of $^{178}Hf^{m2}$ by irradiation with low-energy photons (C. Collins et al., Laser Phys. 14, 154 (2004)). The upper limits for the NEET cross sections are $< 10^{-31}$ cm^2 for transitions filling L-shell holes. These results are significantly below the values of 10^{-22} cm^2 reported by Collins et al. and confirm the recent results by Tkalya (E. Tkalya, Phys. Rev. C. 68, 064 611 (2003)). These calculations suggest that NEET is unlikely to be the mechanism responsible for the reported apparent enhancements of ^{178}Hf nuclear line intensities by X-ray irradiation.

[65] T. Hayakawa, T. Shizuma, S. Miyamoto, S. Amano, K. Horikawa, K. Ishihara, M. Mori, K. Kawase, M. Kando, N. Kikuzawa, S. Chiba, T. Mochizuki, T. Kajino, and M. Fujiwara, *Half-life of the ^{164}Ho by the (γ, n) reaction from laser Compton scattering γ rays at the electron storage ring NewSUBARU*, Phys. Rev. C, **77** (2008), p. 068801. **Abstract:** The half-life of an isomer in ^{164}Ho was measured by using photodisintegration reactions. The photons were generated by Compton scattering of laser photons and relativistic electrons at the electron storage ring NewSUBARU in the super photon ring 8-GeV (SPring-8) facility. The half-life is 36.4±0.3 min. This value is about 3% shorter than the previous value 37.5±1.5 min reported in 1966.

[66] A. B. Hayes, D. Cline, C. Y. Wu, H. Ai, H. Amro, C. Beausang, R. F. Casten, J. Gerl, A. A. Hecht, A. Heinz, H. Hua, R. Hughes, R. V. F. Janssens, C. J. Lister, A. O. Macchiavelli, D. A. Meyer, E. F. Moore, P. Napiorkowski, R. C. Pardo, C. Schlegel, D. Seweryniak, M. W. Simon, J. Srebrny, R. Teng, K. Vetter, and H. J. Wollersheim, *Spin dependence of K mixing, strong configuration mixing, and electromagnetic properties of ^{178}Hf*, Phys. Rev. C, **75** (2007), p. 034308. **Abstract:** The combined data of two Coulomb excitation experiments has verified the purely electromagnetic population of the K=4^+,6^+,8^-, and 16^+ rotational bands in ^{178}Hf via $2 \leq \nu \leq 14$ K-forbidden transitions, quantifying the breakdown of the K-selection rule with increasing spin in the low-K bands. The γ^-, 4^+, and 6^+ bands were extended, and four new states in a rotational band were tentatively assigned to a previously known K=0^+ band. The quasiparticle structure of the 6^+ (t=77 ns) and 8^- (t=4 s) isomer bands were evaluated, showing that the gyromagnetic ratios of the 6+ isomer band are consistent with a pure +[404],+[402] structure. The 8- isomer band at 1147 keV and the second 8^- band at 1479 keV, thought to be predominantly $^-$[514],$^+$[624] and $^-$[514],$^+$[404], respectively, are mixed to a degree approaching the strong-mixing limit. Based on measured <K=16^+||E2|| K=0^+> matrix elements, it was shown that heavy-ion bombardment could depopulate the 16^+ isomer at the ∼1% level, although no states were found that would mediate photodeexcitation of the isomer via low-energy x-ray absorption.

[67] ——, *Spin dependence of K mixing, strong configuration mixing, and electromagnetic properties of ^{178}Hf*, Physical Review C, **75** (2007), p. 034308. **Abstract:** The combined data of two Coulomb excitation experiments has verified the purely electromagnetic population of the K=4+,6+,8-, and 16^+ rotational bands in ^{178}Hf via 214 K-forbidden transitions, quantifying the breakdown of the K-selection rule with increasing spin in the low-K bands. The -, 4+, and 6+ bands were extended, and four new states in a rotational band were tentatively assigned to a previously known K=0^+ band. The quasiparticle structure of the 6+ (t=77 ns) and 8- (t=4 s) isomer bands were evaluated, showing that the gyromagnetic ratios of the 6+ isomer band are consistent with a pure +[404],+[402] structure. The 8- isomer band at 1147 keV and the second 8- band at 1479 keV, thought to be predominantly -[514],+[624] and -[514],+[404], respectively, are mixed to a degree approaching the strong-mixing limit. Based on measured K=16^+E2K=0^+ matrix elements, it was shown that heavy-ion bombardment

could depopulate the 16^+ isomer at the ∼1% level, although no states were found that would mediate photodeexcitation of the isomer via low-energy x-ray absorption.

[68] A. B. Hayes, D. Cline, C. Y. Wu, J. Ai, H. Amro, C. Beausang, R. F. Casten, J. Gerl, A. A. Hecht, A. Heinz, R. Hughes, R. V. F. Janssens, C. J. Lister, A. O. Macchiavelli, D. A. Meyer, E. F. Moore, P. Napiorkowski, R. C. Pardo, C. Schlegel, D. Seweryniak, M. W. Simon, J. Srebrny, R. Teng, K. Vetter, and H. J. Wollersheim, *Breakdown of K Selection in ^{178}Hf*, Phys. Rev. Lett, **96** (2006), p. 042505. **Abstract:** Coulomb activation of the four quasiparticle K=16^+ ^{178}Hf isomer (t1/2=31 y) has led to the measurement of a set of E matrix elements coupling the isomer band to the ground band. The present data combined with earlier ^{178}Hf Coulomb excitation data have probed the K components in the wave functions and revealed the onset and saturation of K mixing in low-K bands, whereas the mixing is negligible in the high-K bands. The implications can be applied to other quadrupole-deformed nuclei.

[69] A. B. Hayes, D. Cline, C. Y. Wu, M. W. Simon, R. Teng, J. Gerl, C. Schlegel, H. J. Wollersheim, A. O. Macchiavelli, K. Vetter, P. Napiorkowski, and J. Srebrny, *Coulomb Excitation Paths of High-K Isomer Bands in ^{178}Hf*, Phys. Rev. Lett, **89** (2002), p. 242501. **Abstract:** Three distinctly different mechanisms are shown to populate the K^p=6^+ ($t_{1/2}$=77ns), 16^+ (31 yr), and 8^- (4 s) isomer bands of ^{178}Hf by Coulomb excitation. High spin states of the three isomer bands were populated by Coulomb excitation of a hafnium target with a 650 MeV ^{136}Xe beam. Although direct population of high-K bands is highly K-forbidden, isomer bands in ^{178}Hf were populated up to spins $13^+_{K=6}$, $20^+_{K=16}$, and $14^-_{K=8}$ with in-band γ yields of $\sim 10^{-4}$ of the ground state band. The data are consistent with a rapid increase in K mixing with increasing spin in the isomer bands.

[70] R. Helmer and C. Reich, *Decay of an isomeric state in ^{178}Hf with $K \geq 16$*, Nuclear Physics A, **114** (1968), pp. 649–662. **Abstract:** A long-lived isomeric activity in ^{178}Hf has been produced by neutron irradiation of hafnium. The isomeric state occurs at an energy of ≈2.5 MeV and has a half-life >10 y. The gamma-ray transitions associated with the decay of the isomer have been studied by means of a Ge(Li) spectrometer and a NaI(Tl) gamma-gamma coincidence system. The resulting decay scheme includes the following known levels of the $K^\pi=0^+$ ground-state rotational band: 93.181 (I=2), 306.625 (4), 632.19 (6), and 1058.56 keV (8). This scheme also includes the previously reported $K^\pi=8^-$ level at 1147.44 keV along with the following members of its rotational band: 1364.11 (I=9), 1601.50 (10), 1859.12 (11), 2136.50 (12), and 2433.32 keV (13). The M1 transition probabilities of these intraband transitions were obtained from the observed gamma-ray intensities assuming the validity of the Alaga rules for the rotational E2 transitions. These probabilities provide an estimate of the amount of mixing between this band that built on the $K^\pi=8^-$ state at 1480 keV. Although a configuration assignment for the isomeric state cannot be made with certainty on the basis of the present data, the most plausible interpretation is that it is a four-quasi-particle state with $K^\pi=16^+$.

[71] R. G. Helmer and C. W. Reich, *Half-life of ^{178m2}Hf and its neutron capture production*, Nuclear Physics A, **211** (1973), pp. 1–6. **Abstract:** The half-life of the high-spin isomeric state in ^{178}Hf at > 2.43 MeV has been measured to be 31 ± 1 y. The cross section for production of this isomer in a thermal-reactor neutron spectrum has been measured to be (2 ± 1) × 10^{-7} b. The computation of this value assumes that the burn-up cross section of the isomer is < 20 b, an assumption which is supported by the experimental data.

[72] I. N. Izosimov, *Triggering of nuclear isomers via decay of autoionization states in electron shells (NEET)*, Laser Physics, **17** (2007), p. 755. **Abstract:** Nuclear excitation by an electron transition (NEET) may be used for triggering the decay of nuclear isomers only when there are compensations between energies (ΔE) and multipolarities (ΔL) of the nuclear transition and the transition in an electron shell. It is shown that using the autoionization states (AS) allows one to compensate for the ΔE and ΔL differences. Laser radiation may be used for the excitation of AS with energies up to 10-15 eV and 229m Th (3.5 eV) nuclear isomer excitation by NEET via AS decay. Ion beams, electron beams, and X rays may be used for the excitation of the trigger nuclear levels with energies up to 150 keV by NEET via AS and for the triggering of the nuclear isomer decay. For excitation of AS with the energies up to 150 keV, two or more hole states in deep inner electron shells must be excited. The

cross section for such two-hole state excitation in electron shells by ion beams may be sufficiently high. The possibilities of NEET via AS for the triggering of nuclear isomer decay are discussed.

[73] G. Jones, *Study of Isomers using Reactions with a ^{178}Hf Beam*, University of Surrey Ph.D. Thesis, October (2006) 1-172. **Abstract:** A pulsed ^{178}Hf beam at an energy of 1150 MeV was used to initiate deep-inelastic reactions in a thick ^{208}Pb target, in order to study high-K isomers in ^{177,178}Hf. Blocked BCS calculations predict high-K multi-quasiparticle states at $I^{\pi} = 43/2^{-}$ and $45/2^{\pi}$ in ^{177}Hf and $I^{\pi} = 19^{+}$ and 22^{-} in ^{178}Hf. These isomers were not observed in the experiment, however limits have been placed on the lifetime of the $K^{\pi} = 19^{\pi}$ isomer for given excitation energies. The quality of the blocked BCS calculations has been evaluated and compared with configuration-constrained energy-surface calculations. Previously unobserved decay branches from high-K isomers in ^{177,179}Hf have been identified, including the first highly K-forbidden M3 transition to be observed from a multi-quasiparticle state. Spin-trap isomers in near-spherical Z =51, Sb isotopes have been populated for the first time using fusion-fission reactions with a pulsed 178 Hf beam at an energy of 1150 MeV , impinging upon a ^{27}Al target. Gamma-rays were observed from the decay of $T_{1/2} = 200(30)$ μs and 52(3) μs isomers with spins and parities $I^{\pi} = (25/2^{+})$ and $(27/2^{+})$ in ^{121,123}Sb respectively. These states are proposed to have aligned ν $(h11/2)^2$ configurations coupled to an odd proton (πd5/2 or πg7/2 in ^{121,123}Sb respectively). Intermediate isomers were also identified at $I^{\pi} = 19/2^{-}$ ($T_{1/2} = 8.2(2)$ ns) and $(15/2^{-})$ ($T_{1/2} = 40(2)$ ns) in ^{121,123}Sb respectively. Spins and parities of the states in these nuclei were obtained using a combination of angular correlation and internal conver- sion measurements. The configurations of states in these nuclei are compared with the systematics of neighbouring Sn isotones and other Sb isotopes. In a survey of other fusion-fission products, an $T_{1/2} = 18(5)$ ns isomer has been identified in ^{99}Mo. The long lifetime of this level is ascribed to a hindered E1 transition from a state with configuration $\pi(g9/2)^2$ ν g7/2 .

[74] P. Kálmán and T. Keszthelyi, *Resonant electronic-bridge process of the isomeric transition of ^{235m}U induced by intense laser fields*, Phys. Rev. A, **44** (1991), pp. 4761 – 4764. **Abstract:** The effect of intense laser fields on the resonant electronic-bridge process involving the isomeric E3 transition of ^{235m}U, which has a photon energy of 73.5 eV, and an intermediate excitation of an electron from the P1(6s1/2) electronic shell of binding energy 71 eV is investigated for lasers of photon energies 1.16 and 2.32 eV within the intensity range 10^{10}-10^{13} W/cm^2. It is found that in spite of the hindering effect of power broadening, the ratio of the transition probability per unit time in the laser-assisted process to that of the laser-free γ decay can be enhanced by the intense laser field if the resonance condition is met.

[75] S. Karamian, J. Carroll, J. Adam, , and N. Demekhina, *Production of the ^{178m2}Hf isomer using a 4.5-GeV electron accelerator*, Nuclear Instruments and Methods in Physics Research A, **530** (2004), pp. 463–472. Received 22 March 2004. **Abstract:** High-productivity methods are required for the accumulation of long-lived isomers in amounts that are sufficient for the creation of experimental targets. A tantalum sample was activated with the Yerevan synchrotron using 4.5-GeV bremsstrahlung and the presence of ^{178m2}Hf was detected with good statistical accuracy by γ-activity measurements. The integrated and mean cross-section values were deduced from the experiment. The isomer-to-ground-state ratio was then estimated and compared with that known for the p+ Ta reaction studied at 660 MeV. In the present experiment, both converter and target were relatively thin for better definition of the experimental conditions. However, an assembly designed for high-productivity irradiations should be thick and then the converter can also serve as the target sample when irradiated with a high-energy electron beam. The optimization of the isomer production was solved analytically and the largest estimated yield was determined as calibrated to the experimental yield. The maximum yield of ^{178m2}Hf was found to be of about 310^9 nuclei/s using an electron beam current of 100 mA. This is lower than the yield achieved with proton beams, although for a practical comparison the total cost and radiation safety conditions should be considered. The present results provide a basis for numerical estimations.

[76] S. Karamian, J. Carroll, J. Adam, E. Kulagin, and E. Shabalin, *Production of long-lived hafnium isomers in reactor irradiations*, High Energy Density Physics, **2** (2006), pp. 48–56. **Abstract:** Experiments on production of long-lived ^{178m2}Hf isomer in reactor irradiations are described. Properties of this nuclide are promising for its potential application as a relatively safe power source characterized

by high density of accumulated energy. Metal natHf samples were activated in the Dubna IBR-2 reactor at positions corresponding to different neutron fluxes. Samples were bare or shielded by Cd and B4C layers. The gamma activity of the samples was analyzed with Ge gamma spectrometers during a two-year period following their irradiation. In the presence of dominant activation products 175Hf and 181Hf, the high-spin isomers 178m2 Hf and 179m2 Hf were also detected despite relatively low levels. The isomer-to-ground state ratios and cross-sections were determined from the measured yields. For ^{178m2}Hf, the cross-section for burnup (destruction) by neutron capture after its production was also estimated, clarifying the results from earlier experiments. In the context of suggestions for use of 178m2 Hf for applications, the results confirm that large-scale production of this isomer by reactor irradiations is not feasible. In contrast, the efficiency of production of ^{179m2}Hf is much higher and an amount of about 10^{16} atoms may be produced in standard reactor irradiations. For 178m2 Hf , more productive methods are known, in particular fast neutron irradiations at E_n=14 MeV and spallation reactions at intermediate energies. Neutron cross-sections for isomers may also be significant in astrophysics.

[77] S. Karamian, J. de Boer, Y. Oganessian, A. B. I, Z. Szeglowski, B. Markov, j. Adam, V. Stegailov, C. Briançon, O. Constantinescu, and M. Hussonnois, *Observation of photonuclear reactions on isomeric targets: $^{178}Hf^{m2}(\gamma,n)^{177}Hf^{m2}$, $^{180}Ta^{m}(\gamma,2n)^{178}Ta^{m,g}$ and $^{180}Ta^{m}(\gamma,p)^{179}Hf^{m2}$*, Z. Phys. A, **356** (1996), pp. 23–29. **Abstract:** Photonuclear reactions on isomeric and ground state Hf and Ta targets were studied systematically by activation technique. The reaction yields were measured and isomer-to-ground state ratios Y_m/Y_g were deduced. The systematics of the Y_m/Y_g values versus spin difference of initial and final states in the photonuclear reaction shows that the structure selectivity for the population of the levels does not survive at high excitation energies.

[78] S. A. Karamian, *Comparative Analysis of the ^{178m2}Hf Yield at Reactions with Different Projectiles*, Physics of Atomic Nuclei, **68** (2005), pp. 1765–1776. **Abstract:** The long-lived high-spin ^{178m2}Hf K isomer can be produced in nuclear reactions with different projectiles. The reaction yields and cross sections have been measured in a series of experiments and the results are now reviewed. The systematics of isomer-to-ground-state ratios are drawn and real production capabilities are estimated for the best reactions. Such a summary is relevant to the significance of isomer studies, both for nuclear-science knowledge and for possible applications. Potential isomer appli- cations have been stressed previously in popular publications with probably overestimated expectations. The real possibilities are restricted in part by the production yield and by other shortcomings as well.

[79] S. A. Karamian, J. Adam, , D. V. Filossofov, D. Henzlova, V. Henzl, V. G. Kalinnikov, N. A. Lebedev, A. F. Novgorodov, C. B. Collins, I. I. Popescu, and C. A. Ur, *Accumulation of the ^{178m2}Hf isomeric nuclei through spallation with intermediate-energy protons of tantalum and rhenium targets*, Nuclear Instruments and Methods in Physics Research Section A, **489** (2002), pp. 448–468.

[80] S. A. Karamian and J. Adam, *Estimation of the spallation-residue angular momentum in p+Ta, Re reactions*, Czechoslovak Journal of Physics B, **53** (2003), p. 381. **Abstract:** Yields and cross-sections for the radioactive isotope production have been, determined via gamma-activity measurements after irradiation of the Ta and Re targets by protons with energies from 100 to 660 MeV. Mass-distribution of the reaction products contains two wide peaks corresponded to the spallation and fission product nuclei, respectively. High-spin Hf and Lu isomers are among the detected nuclides. The spallation-residue angular momentum, I, has been estimated basing on the determined isomer-to-ground state ratio, sigma(m)/sigma(g), and using, known systematics of sigma(m)/sigma(g), versus I. Non zero value of spin I should increase a probability of fission. Measured fission-to-spallation ratio is indeed higher than predicted by the Monte Carlo code simulation without angular momenta in account.

[81] S. A. Karamian, J. Adam, P. Chaloun, D. V. Filossofov, V. Henzl, D. Henzlova, V. G. Kalinninkov, N. A. Korolev, N. A. Lebedev, A. F. Novgorodov, C. B. Collins, I. I. Popescu, and C. A. Ur, *Yield of Radionuclides and Isomers Measured in Fragmentation of the natW and 186W (97%) Targets with Protons at 630, 420 and 270 MeV*, Nuclear Instruments and Methods in Physics Research A, **527** (2004), pp. 609–623. **Abstract:** Yields and cross-sections of the radioactive nuclides produced after the irradiation of natural composition W and enriched 186W targets at the Dubna synchrocyclotron were measured using the γ-ray spectroscopy methods with high-resolution Ge detectors. Among the

detected nuclides we identified the spallation and fission products. High-spin isomeric states in the Hf and Lu nuclides were populated and the isomer-to-ground state ratios could be estimated. The nuclide yields were calculated using the LAHET code at six different values of the proton energy in the range from 100 to 800 MeV both for the natW and enriched 186W targets. The measured isotope yields are in general good agreement with the calculations. A shortcoming of the code is the inability to predict isomer-to-ground state ratios. The experimental data show that the 177mLu, 178m2Hf and 179m2Hf high-spin isomers are produced with a 2.5 times higher yield in the 97% enriched 186W target as compared to the natW target under identical irradiation conditions. This makes significance for the creation of high-activity isomeric sources. The mass-distribution of the products and the fission-to-spallation ratio were also deduced and compared with theory prediction.

[82] S. A. Karamian and J. J. Carroll, *Possibility of Combining Nuclear Level Pumping in a Plasma with Lasing in a Solid*, Hyperfine Interactions, **143** (2002), pp. 69–78. **Abstract:** Nuclear isomers can be used for the storage and release of "clean" nuclear energy and several triggering schemes have been discussed. Here the possibility to utilize resonance between atomic and nuclear transitions in the form of a hybridization of atomic-nuclear excitation is considered. Several isotopes and specific nuclear levels are identified as candidates for triggering via atomic transitions. A variety of ionization states and atomic-shell configurations arises in a hot plasma generated by short high-power pulses of laser light. The non-radiative conversion of the ionization energy within an atom can be suppressed in the hot plasma surroundings. The time scales of different processes in nuclear, atomic and condensed-matter subsystems are compared and the fast ionization in a solid, X-ray radiance in a plasma, and sample melting and recrystallization may precede nuclear fluorescence. A time scale shorter than 0.1 ns makes this sequence promising for the collective excitation of short-lived modes in a nuclear subsystem.

[83] ——, *Prospects for Coherently Driven Nuclear Radiation by Coulomb Excitation*, Laser Physics, **17** (2007), pp. 80–8. **Abstract:** Possible experiments are discussed in which Coulomb excitation of nuclear isomers would be followed by sequential energy release. The possibility of coherent Coulomb excitation of nuclei ensconced in a crystal by channeled relativistic heavy projectiles is considered. The phase shift between neighbor-nuclei excitations may be identical to the photon phase shift for emission in the forward direction. Thus, the elementary string of atoms may radiate coherently with emission of characteristic nuclear γ rays, and the intensity of the radiation would be increased due to the summation of amplitudes. Mossbauer conditions should be important for this new type of collective radiation, which could be promising in the context of the γ-lasing problem.

[84] S. A. Karamian, J. J. Carroll, S. Iliev, and S. P. Tretyakova, *Weak K hindrance manifested in decay of the ^{178}Hfm2 isomer*, Phys. Rev. C, **75** (2007), p. 057301. **Abstract:** An experiment has been performed to detect the emission mode in ^{178}Hfm2 isomer decay and a partial half-life of $(2.5\pm0.5)\times10^{10}$ y was measured. It was concluded that decay is strongly retarded by the centrifugal barrier arising due to the high spin of this isomeric state. Additional analysis shows, however, that the K-hindrance in this decay is relatively weak, despite the strong manifestation of spin-hindrance.

[85] S. A. Karamian, C. B. Collins, J. J. Carroll, and J. Adam, *Isomeric to ground state ratio in the $^{180}Ta^{m}(\gamma,\gamma\prime)^{180}Ta^{g}$ reaction*, Phys. Rev. C, **57** (1998), pp. 1812–1816. **Abstract:** The yield Y of the ^{180}Ta$^{m}(\gamma,\gamma\prime)^{180}$Ta^{g} ($T_{1/2}$=8.15 h) reaction has been measured as a function of the bremsstrahlung end-point energy Ee in the range of 2.5-7.6 MeV. An activation technique was used for the yield measurement which was calibrated by monitoring the ^{232}Th(γ,f) reaction yield. The Y(Ee) function was numerically simulated and the probability $\sigma_g/(\sigma_g+\sigma_m)$ for the depletion of the ^{180}Ta^{m} isomer after γ-ray absorption was deduced. This probability is important for astrophysical questions and schemes for producing induced γ emission and perhaps a γ-ray laser.

[86] S. A. Karamian, C. B. Collins, J. J. Carroll, J. Adam, A. G. Belov, and V. I. Stegailov, *Fast neutron induced depopulation of the $^{180}Ta^{\mathrm{m}}$ isomer*, Physical Review C, **59** (1999), p. 755. **Abstract:** Fast neutron-induced depopulation of the ^{180}Ta^{m} (I, $K\pi$=9, 9-) isomer to the 1^{+} ground state was detected by activation techniques. Natural Ta foils together with a ^{232}Th monitoring target were activated using the neutron flux produced from a 7.3 MeV electron beam by a special choice of converters and shields. The Ka_2 x-ray line (54.6 keV) of Hf was successfully observed in the γ-ray spectrum from activated Ta and attributed to the decay of ^{180}Ta^{g} ($T_{1/2}$ = 8.15 h) . The yields of three reactions ^{232}Th(n,f),

^{181}Ta(n,γ) and ^{180}Ta^{m}(n,n$'$)^{180}Ta^{g}, were measured in the same conditions and compared. Finally, the mean probability for ^{180}Ta^{m} depopulation after MeV neutron scattering was deduced to approach about 0.4. This establishes almost complete K mixing above the neutron binding energy.

[87] F. Karpeshin, Z. Jing-Bo, and Z. Wei-Ning, *Resonance Conversion as a Catalyzer of Nuclear Reactions*, Chinese Phys. Lett, **23** (2006), pp. 2391–2394. **Abstract:** It is shown that resonance internal conversion offers a feasible tool for mastering nuclear processes with laser or synchrotron radiation. The physics of the process is discussed in detail in a historical aspect. Possible experimental application is shown in the case of the M1 70.6-keV transition in nuclei of ^{169}Yb. The nuclear transition rate in hydrogen-like ions of this nuclide can be enhanced by up to four orders of magnitude.

[88] F. F. Karpeshin, *Electron Shell as a Resonator*, Hyperfine Interactions, **143** (2002), pp. 79–96. **Abstract:** Main principles of the resonance effect arising in the electron shells in interaction of the nuclei with electromagnetic radiation are analyzed and presented in the historical aspect. Principles of NEET are considered from a more general position, as compared to how this is usually presented. Characteristic features of NEET and its reverse, TEEN, as internal conversion processes are analyzed, and ways are offered of inducing them by laser radiation. The ambivalent role of the Pauli exclusion principles in NEET and TEEN processes is investigated.

[89] F. F. Karpeshin, M. B. Trzhaskovskaya, and Z. Jing-Bo, *Resonance Conversion as the Effective Way of Triggering the ^{178m2}Hf Isomer Energy*, Chinese Phys. Lett, **23** (2006), pp. 2049–2051. **Abstract:** Experiments on photo-induced de-excitation triggering of ^{178m2}Hf are revisited. We present an alternative and more effective way of triggering by exploiting the resonance internal conversion. Full theoretical description of a possible channel is presented. It is therefore revisited the impact itself produced by those experiments.

[90] T. Khoo and G. Lovhoiden, *Structural Changes in the Yrast states of ^{178}Hf*, Physics Letters B, **67** (1977), pp. 271–274. **Abstract:** High-spin levels in ^{178}Hf* including the ground-state band, several 2-quasiparticle (qp) bands, and two 4-qp isomers have been identified from (α, 2n) studies. Changes in the structure of the yrast line are observed. The 4-qp isomers with K^{π} = 16 + and 14- are yrast traps.

[91] J. B. Kim, J. Morel, M. Etcheverry, N. Coursoi, D. Trubert, O. Constantinescu, S. A. Karamian, Y. T. Oganessian, C. Briançon, and M. Hussonnois, *Precise measurement of gamma-ray energies and gamma-ray emission probabilities in ^{178m2}Hf decay*, Journal of Radioanalytical and Nuclear Chemistry, **215** (1997), pp. 229–233. **Abstract:** Nowadays large interest is attached to the production of exotic beams and targets for nuclear stmcture and reaction studies.The nucleus ^{178}Hf, with its long-lived ($T_{1/2}$=31 years) high-spin isomeric state $I^{\pi} = 16^{+}$ at a relatively low-excitation energy (2.446 MeV), is indeed a unique probe for studying phenomena in a new way. A wide research program has been underway in the framework of the "Hafnium Collaboration" which now concerns around 80 scientists. Various targets have been plepared and adapted to different types of experiments. Irradiation processes with high-intensity beams, high-purity chemistry methods and isotopic separations have been developed. The obtaining of appreciable amounts of very high purity ^{178m2}Hf by isotopic separation, has made it possible for us to carry out γ-ray measurements with good statistics and high energy resolution with a γ-spectrometer, in order to use this isomer as a calibration standard in the 200 to 600 keV energy range. For example, relative to the main γ-rays, energies have been determined with an absolute uncertainty of about 2 eV and photon emission probabilities with a relative uncertainty better than 1%.

[92] J. B. Kim, D. Trubert, O. Constantinescu, S. A. Karamian, Y. T. Oganessian, C. Briançon, and M. Hussonnois, *Purity determination of super-enriched ^{176}Yb by neutron activation and thermal neutron capture cross section measurement of the high-spin (16^{+}) isomer ^{178m2}Hf* , Journal of Radioanalytical and Nuclear Chemistry, **215** (1997), pp. 219–222. **Abstract:** Neutron activation analysis has been applied to determine the purity of super-enriched ^{176}Yb as well as a diagnostic to optimize yields from the P.A.R.I.S. mass separator at Orsay. In 25 mass separations carried out during 3 months, 910 mg of super-enriched $^{176}Yb_2O_3$ were recovered from the original 7 g of $^{176}Yb_2O_3$ (97.5% enrichment). Neutron activation analysis has shown that only two per thousand of the initially present ^{174}Yb remained

in the super-enriched product. Using this ^{176}Yb super-enriched (99.998%) target, micro-quantities of the high spin (16^+) isomer ^{178m2}Hf were produced by the (α,2n) reaction at the U-200 cyclotron in Dubna. Quantities of about $2*10^{12}$ atoms of this isomer have been irradiated by thermal neutron at the ORPHEE Reactor, Saclay. Only the neutron capture giving rise to the isomeric state $25/2^-$ of ^{179}Hf has been measured, by detection γ-rays of this 25.1 days isomer. A cross section of 45 $\pm$ 5 barns was obtained for ^{178m2}Hf(n,γ)^{179m2}Hf.

[93] S. Kishimoto, Y. Yoda, M. Seto, Y. Kobayashi, S. Kitao, R. Haruki, T. Kawauchi, K. Fukutani, and T. Okano, *Observation of Nuclear Excitation by Electron Transition in ^{197}Au with Synchrotron X Rays and an Avalanche Photodiode*, Phys. Rev. Lett, **85** (2000), pp. 1831–1834. **Abstract:** We have succeeded in observing nuclear excitation by electron transition (NEET) in ^{197}Au by a new method. Monochromatic x-rays of synchrotron radiation were used to ionize the K shell of gold atoms in a target foil. The internal-conversion electrons emitted from excited nuclei were detected with a silicon avalanche photodiode. At a photon energy of 80.989 keV, the NEET probability in ^{197}Au was determined to be $(5.0\pm0.6)\times10^{-8}$ from a comparison of the event number per photon between NEET and the nuclear resonance at 77.351 keV.

[94] J. V. Klinken, W. Z. Venema, R. V. F. Janssens, and G. T. Emery, *K-forbidden decays in ^{178}Hf; M4 decay of an Yrast state*, Nuclear Physics A, **339** (1980), pp. 189–204. **Abstract:** The decays of a 16^+ four-quasiparticle state and an 8- two-quasiparticle state in ^{178}Hf have been studied with emphasis on K-forbidden decay modes. An M4 branch in the decay of the 16^+ state was identified, locating the isomer at an excitation energy of 2446.0 keV. Upper limits were found for the intensity of the 8- to 6+ transition and for other possible decay modes of the 16^+ state. Hindrance factors for the K-forbidden transitions are compared with those for similar transitions in neighbouring nuclides.

[95] I. A. Kondurov, E. M. Korotkikh, Y. V. Petrov, and G. I. Shuljak, *Acceleration of thermal neutrons by isomeric nuclei (^{180}Hf^m)*, Physics Letters B, **106** (1981), pp. 383–385. **Abstract:** Inelastic accelaration of neutrons by ^{180}Hf^m has been observed. The experimental cross section for acceleration of thermal neutrons is σ_{in} = 52 $\pm$ 13 b. The K-hindrance factor determining the long lifetime of the isomer is absent in this reaction.

[96] U. Köster, O. Arndt, R. Catherall, J. Correia, B. Crepieux, R. Eichler, P. Fernier, L. Fraile, S. Hennrich, K. Johnston, S. Lukic, J. Neuhausen, B. N. Singh, B. Pfeiffer, and E. Siesling, *ISOL beams of hafnium isotopes and isomers*, Eur. Phys. J. Special Topics, **150** (2007), pp. 293–296. **Abstract:** The production of ISOL beams of hafnium is described. Radioactive Hf isotopes were produced at ISOLDE by 1.4 GeV proton-induced spallation in Ta and W foils. Chemical evaporation in form of HfF_4 and mass separation in the molecular sideband HfF_3^+ after electron impact ionization provided intense and pure beams. Beams of $^{158-185}$Hf and short-lived isomers down to 1.1 s ^{177m}Hf were observed, but the method could be extended to reach even more exotic isotopes: down to about ^{154}Hf (N = 82) on the neutron-deficient side and up to neutron-rich ^{188}Hf.

[97] Y. kun Ho, Z. shu Yuan, B. hui Zhang, and Z. ying Pan, *Self-consistent description for x-ray, Auger electron, and nuclear excitation by electron transition processes*, Phys. Rev. C, **48** (1993), p. 2277. **Abstract:** The physical processes about x-ray emission, Auger electron emission, and nuclear excitation by electron transition (NEET) induced by deexcitations of the excited atomic states, e.g., the inner shell hole states, are studied self-consistently in the unified nonrelativistic quantum mechanics framework. The emphasis is on exploring the NEET process and its interference with the other two processes. The transition probabilities of these deexcitation channels are given in analytical forms. The numerical calculations for $^{189}_{76}$Os, $^{197}_{79}$Au, and $^{237}_{93}$Np have been done and compared with the available experimental results.

[98] L. Lakosi, N. C. Tam, and I. Pavlicsek, *Photoexcitation of ^{189}Os^m and ^{193}Ir^m. I. Excitation of ^{189}Os^m by low-energy x rays*, Phys. Rev. C, **52** (1995), pp. 1510–1515. **Abstract:** Production of 5.84 h ^{189}Os^m by 200 through 300 kV bremsstrahlung has been measured and attributed to population of four nuclear states between 69.54 and 275.91 keV. From the measurements and known level parameters, contributions to isomer production from individual levels have been separated, and the half-life of the 216.66 keV level has been determined as 77$\pm$10 ps. Besides nuclear resonance absorption (NRA), the

first level has long been assumed to be also excited by another process called nuclear excitation by electron transition (NEET), via virtual photon exchange due to recombination of vacancies produced in the K shell by incident x rays. However, contribution from competing NRA cannot be separated, and the claimed dominance or even the existence of the NEET process has remained unconfirmed.

[99] L. Lakosi, N. C. Tam, I. Pavlicsek, and A. Petö, *Photoexcitation of $^{189}Os^m$ and $^{193}Ir^m$. II. Excitation by ^{137}Cs and ^{60}Co γ rays*, Phys. Rev. C, **52** (1995), pp. 1516–1519. **Abstract:** Photoexcitation of 5.84 h $^{189}Os^m$ and 10.6 d $^{193}Ir^m$ was observed using high activity ^{137}Cs and ^{60}Co sources for irradiation. The highly converted weak isomeric transitions were detected by large area 2π multiwire proportional chambers. Determining resonance photon flux densities with Monte Carlo simulation, isomer excitation cross sections integrated over identified activation levels were evaluated and level half-lives were derived.

[100] K. E. G. Löbner, *Systematics of absolute transition probabilities of K-Forbidden gamma-ray transitions*, Physics Letters B, **26** (1968), pp. 369–370. **Abstract:** All available experimentally determined absolute γ-ray transition probabilities between different intrinsic states in odd- and even-mass nuclei of the rare earth region (with neutron numbers $89 \leq N \leq 114$ and proton numbers $62 \leq Z \leq 78$) and of the actinide region ($N \geq 134$) are compared to the Weisskopf estimate for different $|\Delta K|$ values.

[101] E. Lubkiewicz, H. Wollersheim, R. Kulessa, C. Briangon, W. Brüchle, O. Constantinescu, M. Debowski, E. Ditzel, H. Folger, J. Gerl, F. Hannachi, T. Happ, M. Hussonnois, E. Jäger, S. Karamian, M. Kaspar, T. Kröll, Y. Oganessian, I. Peter, H. Schaffner, S. Schremmer, R. Schubert, N. Trautmann, K. Vetter, and G. Zauner, *Coulomb excitation of the $K^\pi = 16^+$ rotational band in ^{178}Hf*, Z. Phys. A, **355** (1996), pp. 377–381. **Abstract:** A heavy-ion multiple Coulomb excitation experiment on a very exotic target containing microweight quantities of ^{178}Hf in the $K^\pi = 16^+$ isomeric state has been performed at 4.77 MeV/u ^{208}Pb beam energy. The first excited $I^\pi = 17^+$ state has been observed at an excitation energy of 357.4 ± 0.3 keV with respect to the isomeric state. The intrinsic electric quadrupole moment of $Q_0 = 8.2 \pm 1.1$ b has been derived from the experimental data within the rigid rotor model.

[102] M. MacGregor and G. Reffo, *Neutron Interactions with ^{178m}Hf*, Nuclear Science and Engineering, **114** (1993), pp. 124–134. **Abstract:** We have performed an analysis of the interaction of low energy neutrons with the 16^+ metastable state of ^{178}Hf at 2.45 MeV. We have discerned the results of cross section calculations in view of possible applications for energy gains from different neutron energy sources.

[103] S. Matinyan, *Lasers as a bridge between atomic and nuclear physics*, Physics Reports, **298** (1998), pp. 199–249. **Abstract:** This paper reviews the application of visible and ultraviolet laser radiation to several topics in low-energy nuclear physics. We consider laser-induced nuclear anti-Stokes transitions, laser-assisted and laser-induced internal conversion, and the electron bridge and inverse electron bridge mechanisms as tools for deexcitation and excitation of low-lying nuclear isomeric states. A study of the anomalously low-lying nuclear isomeric states (in the case of the ^{229}Th nucleus) is presented in detail.

[104] P. McDaniel, W. Kemp, C. Crist, M. Helba, J. Kirtland, and R. Tittsworth, *TRiggered Isomer Proof (TRIP) Test*, Sandia Report SAND2007, **2690** (2008), pp. 1–94. **Abstract:** The object of the TRiggered Isomer Proof (TRIP) Test was to demonstrate convincing proof that when a Hf-178m2 target was irradiated by low energy x-rays additional radiations occur that strongly indicate isomeric decay has been stimulated. A successful series of isomer triggering experiments was completed at the National Synchrotron Light Source in January, March, August and September of 2005. The experiments used Hf-178m2 targets containing $2xl0^{13}$ and 10^{14} isomers. All spectra were recorded in coincidence mode. Several phenomena were observed that very strongly indicate triggering. (1) A total of 359 counts were recorded in the 129.4 keV transition peak as observed in Figure No. 1. This greatly surpassed the planned minimum of 200 counts predicted a priori. This peak is significant at the 4.13 sigma level. (2) A decay path from near 2.0 MeV to the Ground State was observed as reported in theTable of the Isotopes. (Observed transitions are in italics and brackets.) 1942.0 keV(?) -> [306.4 keV]-> 1635.6 kev(4+) -> [121.8 keV] -> 1513.8 keV(4+) -> [129.4 keV] -> 1384.5 keV(4+) -> [1077.8 keV] -> 306.6 keV(4+) -> [213.4 keV] -> 93.2 keV(2^+) -> [93.2keV] -> 0.0 keV(0^+) (3) But the strongest proof of

all was the suppression of the upper two transitions in theGround State Band relative to the lower two transitions in the Ground State Band whenthe x-ray beam was turned on. This effect was significant at the 30.6 sigma level in thecase of the 93.2 keV transition.

[105] D. P. McNabb, J. D. Anderson, J. A. Becker, and M. S. Weiss, *Comment on "Accelerated Emission of Gamma Rays from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation"* , Phys. Rev. Lett, **84** (1999), p. 2542.

[106] P. Mohr, F. Käppeler, and R. Gallino, *Survival of nature's rarest isotope ^{180}Ta under stellar conditions*, Phys. Rev. C, **64** (2007), p. 012802. **Abstract:** The nucleosynthesis of nature's rarest isotope ^{180}Ta depends sensitively on the temperature of the astrophysical environment because of depopulation of the long-living isomeric state via intermediate states to the short-living ground state by thermal photons. Reaction rates for this transition have been measured in the laboratory. These ground state rates understimate the stellar rates dramatically because under stellar conditions intermediate states are mainly populated by excitations from thermally excited states in ^{180m}Ta. Full thermalization of ^{180}Ta is already achieved for typical s-process temperatures around kT=25 keV. Consequently, for the survival of ^{180}Ta in the s-process fast convective mixing is required which has to transport freshly synthesized ^{180}Ta to cooler regions. In supernova explosions ^{180}Ta is synthesized by photon- or neutrino-induced reactions at temperatures above T_9=1 in thermal equilibrium; independent of the production mechanism, freeze-out from thermal equilibrium occurs at kT$\sim$40 keV, and only 35$\pm$4% of the synthesized ^{180}Ta survive in the isomeric state.

[107] M. Morita, *Nuclear Excitation by Electron Transition and Its Application to Uranium 235 Separation*, Progress of Theoretical Physics, **49** (1973), pp. 1574–1586. **Abstract:** A new mechanism for nuclear excitation is studied theoretically by considering the deexcitation of electronic states of atom. When an electron of inner shells is kicked off by the bombarded electron or X ray, an electron of the adjacent shells immediately jumps into the vacancy. The energy corresponding to the difference of the binding energies for these two shells, E_1-E_2, is usually carried away by the emitted characteristic X ray or the Auger electron which is ionized from an outer shell. There is, however, another possibility of this energy release by exciting a nuclear state. That is, the nuclear ground state is excited to the higher energy level by receiving the excess energy of the electronic state by means of electromagnetic interactions between nucleus and electrons. A theory is made for this process, and it is found that the probability of this process is extremely small in most of the cases, but it will be appreciable, if the interaction energy is approximately equal to the energy mismatch E_N-(E_1-E_2), where E_N is the nuclear excitation energy. An example of ^{235}U is discussed in connection with a possible separation of ^{235}U from uranium isotopes.

[108] S. M. Mullins, G. D. Dracoulis, A. P. Byrne, T. R. McGoram, S. Bayer, W. A. Seale, and F. G. Kondev, *Rotational band on the 31 yr 16$^+$ isomer in ^{178}Hf*, Physics Letters B, **393** (1997), pp. 279–284. **Abstract:** High-spin states in ^{178}Hf have been identified using particle-γ-γ-time techniques and the incomplete fusion reaction, ^{176}Yb(^{9}Be,3n)^{178}Hf. The rotational band associated with the four-quasiparticle $T_{1/2}$ = 31 year, $K^\pi = 16^+$ isomer in ^{178}Hf has been established. The g_K-g_R values obtained from the in-band decay properties confirm the configuration of the isomer as does its alignment which matches the sum of the alignments of the two K^π = 8- two-quasiparticle bands that contain the components of the 16$^+$ configuration. New information has also been obtained on the four-quasiparticle 14- band on the two-quasiparticle 6+ and 8- bands.

[109] G. Muradian, *Decay of ^{178m2}Hf Isomer Nuclei during Neutron Inelastic Scattering*, Moscow, (2004). **Abstract:** It is shown that the neutron can initiate the chain self-sustaining process of ^{178m2}Hf isomeric energy release. The experiments on search for other isomers which are more suitable for the chain process are proposed.

[110] G. V. Muradian, O. Y. Shatrov, M. A. Voskanian, L. P. Yastrebova, V. L. Volkov, C. Briancon, M. Hussonnois, Y. T. Oganessian, S. A. Karamian, and O. Constantinescu, *Search for Neutron Resonances in the ^{178m2}Hf Isomer and Their Investigation*, Physics of Atomic Nuclei, **66** (2003), pp. 6–16. **Abstract:** Experiments aimed at detecting and investigating neutron resonances in the ^{178m2}Hf isomer are described, and the results obtained in these experiments are presented. The investigations in question

are of great interest since the structure of this isomer–it is interpreted as the ($\pi 7/2^+$, $\pi 9/2^+$, ν $7/2^+$, ν $9/2^+$) configuration–and its high spin of J = 16 differ significantly from the structure and spin of nuclei studied previously. The experiments performed at the Kurchatov Institute employed a neutron source based on the FAKEL linear electron accelerator and a multisection detector from NaI(Tl) crystals that was able to ensure a 4π coverage. This equipment made it possible to study gamma-ray cascades in radiative neutron capture versus neutron energy. Despite an extremely small number of isomer nuclei, a low content of the isomer in the target used, and its high radioactivity, resonances were discovered that arise upon neutron capture by a high-spin ^{178m2}Hf nucleus. The parameters of these resonances were found. The mean spacing between the revealed resonances is about 1 eV, which is consistent with calculations based on the Fermi gas model. This indicates that the Fermi gas model describes well the density of both low- and high-spin levels. At the same time, the above agreement suggests that, upon the formation of a compound nucleus, the structure of the isomeric state is destroyed completely. On the other hand, glaring discrepancies between experimental data and the predictions of the statistical model were found: gamma transitions from high-spin resonances (J = $31/2^+$, $33/2^+$) populate predominantly the low-spin ground state (J = $9/2^+$) rather than the high-spin state of the ^{178m2}Hf isomer (J = $25/2^-$); the radiative width is approximately one-third as great as that which is predicted by the statistical model; and the properties of gamma cascades are different for different resonances, this difference being beyond statistical fluctuations. The results of the present investigation make it possible to reveal special features in the behavior of the quantum number K at high excitation energies.

[111] Z. Németh, *Comment on "Resonant excitation of the reaction $^{180}Ta^m(\gamma,\gamma')^{180}Ta$"*, Phys. Rev. C, **45** (1992), pp. 467–469. **Abstract:** We point out that the large integral cross section values reported for the v reaction by Collins *et al* correspond to transition strengths exceeding the recommended upper limits. The small overall cross section ratio of the aforementioned and the ^{115}In(γ,γ')^{115}In^m reactions is shown to be an indirect piece of evidence against the enormous integral cross sections. Collins et al. used the ^{87}Sr(γ,γ')^{87}Sr^m reaction to verify their calculated photon flux, but did not identify the observed activation levels. We identify four photoactivation levels of ^{87}Sr at 1228.42 keV, $5/2^+$, 1770.46 keV, $5/2^+$, 1919.9 keV, $7/2^+$, 2706.5 keV, $9/2^+$, and deduce the existence of (at least) a further one around 2.6 MeV. These new data suggest that the calculated photon flux used by Collins *et al* should be rechecked.

[112] J. Odeurs, R. Coussement, K. Vyvey, H. Muramatsu, S. Gheysen, R. Callens, G. Neyens, I. Serdons, R. N. Shakhmuratov, Y. Rostovtsev, and O. Kocharovskaya, *Induced Transparency for Gamma Radiation via Nuclear Level Mixing*, Hyperfine Interactions, **143** (2002), pp. 97–110. **Abstract:** A significant reduction of absorption of single gamma photons has been observed using the Mössbauer spectra of ^{57}Fe in a FeCO3 crystal. The absorption deficit can be ascribed to partially destructive interference for absorption because of two indistinguishable absorption paths. The necessary coherence is created by means of level mixing produced by a suitable combination of a magnetic dipole and electric quadrupole interaction.

[113] Y. Oganessian, M. Hussonnois, C. Briançon, S. Karamian, Z. Szeglowski, D. Ledu, R. Meunier, M. Constantinescu, J. Kim, and O. Constantinescu, *Production, chemical and isotopic separation of the ^{178m2}Hf high-spin isomer*, Hyperfine Interactions, **107** (1997), pp. 129–139. **Abstract:** The ^{178m2}Hf with its long-lived ($T_{1/2}$ = 31 y), high-spin I^π 16^+ isomeric state, is a challenge for new and exotic nuclear physics studies. The ^{178m2}Hf isomer has been produced in microweight quantities using the 176Yb(a,2n) nuclear reaction, by irradiation with a high-intensity beam using the U-200 cyclotron in Dubna. Radiochemistry and mass separation methods have been developed, with the aim to separate and purify the produced Hf material. Thin targets of isomeric hafnium-178 on carbon backings have been prepared and used in experiments with neutron, proton and deuteron beams.

[114] Y. Oganessian and S. Karamian, *K-mixing in nuclear reactions*, Hyperfine Interactions, **107** (1997), pp. 43–56. **Abstract:** Isomer-to-ground-state ratios are measured for the group of photon- and - induced nuclear reactions. The photonuclear reactions on high-spin isomeric targets - ^{178}Hfm2(16^+) and ^{180}Tam (9-) - were detected successfully. Results are analysed from the point of view of the K-selectivity manifestation for the level population. The yields comparison with the regular values confirms strong K-mixing at high excitations.

[115] Y. T. Oganessian, S. A. Karamian, Y. P. Gangrski, B. Gorski, B. N. Markov, Z. Szeglowski, C. Briancon, D. Ledu, R. Meunier, M. Hussonnois, O. Constantinescu, and M. I. Subbotin, *Production, chemical and isotopic separations of the long-lived isomer ^{178m2}Hf ($T_{1/2}$=31 years)*, J. Phys. G, **18** (1992), pp. 393–399. **Abstract:** The nucleus ^{178}Hf, with its long-lived ($T_{1/2}$=31 y) high-spin isomeric state I^{π} =16^{+}, is a challenge for new and exotic nuclear physics studies. The authors describe the first experiments they have performed in order to produce a reasonable microweight quantity of this hafnium isomer with an isomeric to ground-state ratio as high as possible (here 5%). The reaction ^{176}Yb(^{4}He, 2n) using an enriched target has been studied by measuring the excitation functions and the isomeric to ground-state ratio. About 3*10^{14} isomeric atoms have been produced up to now in irradiations with high-intensity beams (approximately=100 mu A) at the U-200 cyclotron in Dubna. Chemical separation methods could be checked using about 10^{13} atoms of the isomer. Isotopic separation experiments have been performed in Orsay and preliminary results are given for the separation efficiency.

[116] M. Oi, P. M. Walker, and A. Ansari, *Shape coexistence and tilted-axis rotation in neutron-rich hafnium isotopes*, Physics Letters B, **505** (2001), pp. 75–81. **Abstract:** We have performed tilted-axis-cranked Hartree-Fock-Bogoliubov calculations for a neutron-rich hafnium isotope (^{182}Hf) whose proton and neutron numbers are both in the upper shell region. We study whether the shell effects play a role in producing high-K isomers or highly gamma-deformed states at high spin. In particular, the possibility of shape coexistence and the effect of wobbling motion are discussed.

[117] S. Olariu, J. Carroll, C. B. Collins, and I. Popescu, *Emission of gamma rays by electron-nuclear double transitions*, Hyperfine Interactions, **107** (1997), pp. 197–203. **Abstract:** TIn this paper we discuss the relevance of electron-nuclear double transitions for the problem of the amplification of gamma rays.We consider the electron-nuclear coupled system of 151Eu in a CaF2 lattice, in the presence of a strong static magnetic field. We analyze the possibility of pumping this electron-nuclear coupled system from the ground atomic electron Zeeman level to a higher electron level with the aid of amicrowave magnetic field, while the nucleus is simultaneously undergoing a gamma-ray transition to the nuclear ground state. It is shown that an overall inversion condition can be satisfied even in the absence of inversion of the nuclear populations, provided that the ratio of the electron populations in the excited and ground electron Zeeman states has a sufficiently small value.

[118] S. Olariu and A. Olariu, *Comment on "Accelerated Emission of Gamma Rays from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation"* , Phys. Rev. Lett, **84** (2000), p. 2541.

[119] S. Olariu, I. Popescu, and C. B. Collins, *Multiphoton generation of optical sidebands to nuclear transitions*, Phys. Rev. C, **23** (1981), pp. 1007–1014. **Abstract:** Recent calculations have shown that multiphoton transitions in a nucleus that involve a γ-ray photon could be induced by intense fields of optical radiation. The effect of such transitions was predicted to be a shift of the γ-ray energy by the addition or subtraction of the energy of the optical photon. A literature search has shown that such optical sidebands have been observed in certain instances on the ^{57}Fe Mössbauer line at 14 keV and not satisfactorily explained by conventional models. This work reports the formulation and evaluation of the multiphoton model for the values of experimental variables appropriate to those experiments. Using no fitted parameters, remarkable agreement was obtained between the predictions of the multiphoton theory and the previously reported results.

[120] ——, *Tuning of γ-ray processes with high power optical radiation*, Phys. Rev. C, **23** (1981), pp. 50 – 63. **Abstract:** This paper reports the examination of the general problem of multiphoton processes which might occur at the nuclear level when one of the photons belongs to an intense radiation field of optical frequencies. Transition probabilities are reported which relate the rates for γ-ray transitions induced by the optical radiation to the Breit-Wigner cross sections for the absorption or emission of single γ photons. In emission the induced processes of both anti-Stokes Raman scattering and two-photon emission are considered and both are found to lead to the generation of tunable γ radiation. Transition probabilities are calculated to be large enough to support the production of usable spectral sources that could be tuned over a range of γ-ray energies approximately 10^{3} times greater than can be realized by conventional Mössbauer techniques. Analogous possibilities for the detection of nuclear reactions induced by intense optical fields also have been evaluated in this work.

[121] K. Otozai, R. Arakawa, and T. Saito, *Nuclear excitation by electron transition in ^{189}Os*, Nuclear Physics A, **297** (1978), pp. 97–104. **Abstract:** A precise experiment on nuclear excitation by electron transition (NEET) was first performed on ^{189}Os. An osmium film was bombarded with electrons of 72-100 keV to measure the absolute values of the cross section for osmium isomer (^{189m}Os) production. The threshold energy for this process is 74 keV, and the cross section is, e.g., 1.1 nb at 100 keV. The NEET probability derived from the measured excitation function is $(1.7 \pm 0.2) \times 10^{-7}$.

[122] A. Pálffy, J. Evers, and C. H. Keitel, *Isomer Triggering via Nuclear Excitation by Electron Capture*, Phys. Rev. Lett, **99** (2007), p. 172502. **Abstract:** Triggering of long-lived nuclear isomeric states via coupling to the atomic shells in the process of nuclear excitation by electron capture (NEEC) is studied. NEEC occurring in highly charged ions can excite the isomeric state to a triggering level that subsequently decays to the ground state. We present total cross sections for NEEC isomer triggering considering experimentally confirmed low-lying triggering levels and reaction rates based on realistic experimental parameters in ion storage rings. A comparison with other isomer triggering mechanisms shows that, among these, NEEC is the most efficient.

[123] N. R. Pereira, G. M. Merkel, and M. Litz, *Economics of Isomeric Energy*, Laser Physics, **17** (2007), pp. 874–879. **Abstract:** A high energy density source based on nuclear isomers may be conceptually attractive, but it is unrealistic if the energy's price is too excessive. This paper estimates the price of isomeric energy, and shows that isomers can become practical only for low-energy applications.

[124] K. Pisk, Z. Kaliman, and B. A. Logan, *Calculation of nuclear excitation in an electron transition*, Nuclear Physics A, **504** (1989), pp. 103–108. **Abstract:** We have made a theoretical investigation of nuclear excitation during an electron transition (NEET). Our approach allows us to express the NEET probabilities in terms of the excited nuclear level width, the energy difference between the nuclear and electron transition, the Coulomb interaction between the initial electron states, and the electron level width. A comparison is made with the available experimental results.

[125] I.-I. Popescu, *New Perspectives for the Production and Accumulation of $^{178m2}Hf$ Isomers*, Report Number: A282524, Contract Number: FA8655-02-1-3069, 30 Sep 2003, (2003). **Abstract:** This report results from a contract tasking Induced Gamma Emission Foundation as follows: The contractor will investigate the production and accumulation of the nuclear isomer 178m2 Hafnium by spallation of Tungsten targets with high-energy protons. Targets made of the isotope 186 Tungsten will be designed using the LAHET computer code, and fabricated for experimental verification. The Hafnium yield will be assayed and compared to LAHET calculations.

[126] L. A. Rivlin, *Gamma-ray lasing by free nuclei and by matter-antimatter beams*, Hyperfine Interactions, **107** (1997), pp. 57–67. **Abstract:** I discuss the possibilities to induce the gamma-ray emission departing from attempts to use the Mössbauer effect. Three separate approaches are considered: (A) Stimulated radiativetransitions in deeply cooled nuclear beams with hidden inversion;(B) external two-photon ignition of nuclear lasing accompaniedby gamma-ray giant pulse emission; and (C) burst-like radiativeannihilation of relativistic beams of electrons and positrons orparapositronium atoms ignited by an external beam of soft photons.

[127] H. E. Roberts, M. Helba, J. J. Carroll, J. Burnett, T. Drummond, J. Lepak, R. Propri, Z. Zhong, and F. J. Agee, *Gamma Spectroscopy of Hf-178m2 Using Synchrotron X-Rays* , Hyperfine Interactions, **143** (2002), pp. 111–119. **Abstract:** Preliminary survey experiments have been performed to examine the triggering of gamma emission from the 31-year Hf-178m2 isomer using intense monochromatic synchrotron radiation from the X15A beamline at the National Synchrotron Light Source at Brookhaven National Laboratory. Initial studies were performed to probe incident photon energies over the L_1 , L_2 , and L_3 X-ray edges of Hf and the 12-13 keV range. Resonances larger than the experimental minimum detectable level of 10^{-25} cm^2 keV were not observed.

[128] O. Roig, G. Bélier, V. Méot, D. Abt, J. Aupiais, J.-M. Daugas, C. Jutier, G. L. Petit, A. Letourneau, F. Marie, and C. Veyssiére, *Evidence for inelastic neutron acceleration by the ^{177}Lu isomer*, Phys. Rev. C, **74** (2006), p. 054604. **Abstract:** The neutron burnup cross section on the long-lived metastable state of ^{177}Lu has been measured from a specially designed isomeric target. The Maxwellian averaged cross section obtained for this reaction on ^{177}Lu^m(J=23/2$^-$) is σ^m_{burnup}=626±45 b at the reactor

temperature T=323 K. The difference between the burnup cross section and the previously measured capture cross section $\sigma_{n,\gamma}$, clearly shows a possible existence of $^{177}Lu^{m}$ deexcitation via (n,n′) inelastic neutron acceleration channels. The results are interpreted in terms of a statistical approach using parameters from a deformed optical potential calculation.

[129] Y. Rostovtsev, R. Kolesov, and O. Kocharovskaya, *Laser-Mössbauer Spectroscopy as a New Tool for Nuclear Transitions* , Hyperfine Interactions, **143** (2002), pp. 121–131. **Abstract:** New spectroscopic technique based on modification of Mössbauer spectra via optical coherent driving of atomic transitions is suggested. Possible experimental realizations are discussed.

[130] G. Rotbard, G. Berrier-Ronsin, O. Constantinescu, S. Fortier, S. Galès, M. Hussonnois, J. B. Kim, J. M. Maison, L.-H. Rosier, J. Vernotte, C. Briançon, R. Kulessa, Y. T. Oganessian, and S. A. Karamian, *Blocking effect in the $16^+ \rightarrow 16^+$ (p,t) transition on the isomeric $^{178}Hf^{m2}$ target*, Phys. Rev. C, **48** (1993), pp. R2148–2150. **Abstract:** The availability of a target of ^{178}Hf in its aligned four-quasiparticle, $J^{\pi}=16^+$ isomeric state, has allowed to measure the effect of blocking of the aligned-quasineutron orbitals in the (p,t) reaction. The experimental ratio of cross sections $R=\sigma(16^+ \rightarrow 16^+)/\sigma(0^+ \rightarrow 0^+)$ agrees with a predicted value deduced from the known (p,t) cross section on odd Hf isotopes.

[131] C. Rusu, *Study of $^{178m2}Hf(\gamma,\gamma')^{178}Hf$ Reaction by Nuclear Spectroscopy Methods*, University of Texas at Dallas, Ph.D. Thesis, (2002). **Abstract:** Controlled release of the energy stored in nuclear isomers is very attractive for producing powerful sources of induced gamma-ray radiation. High-energy nuclear isomers can have very long lifetimes of the order of years and thus they can serve as good energy storage media. For instance, 1 mg of the ^{178m2}Hf isomer stores about 1 MJ as excitation energy of the isomeric state. One of the most promising ways to release the energy stored in the isomeric state involves photonuclear reactions in the energy range of soft X-rays. Photons with energies lower than 20 keV can excite the ^{178m2}Hf isomeric state to an intermediate level from which gamma transitions cascade to lower levels with a decay rate much faster than the rate of spontaneous decay of the isomeric state. In this work, the photoexcitation of ^{178m2}Hf by (γ,γ') reactions at low energies was studied by nuclear spectroscopy methods. The experimental arrangement was based on the coincident detection of gamma photons with four large HPGe detectors. A bremsstrahlung X-ray generator was used as excitation source, which covered a continuous energy-range from 0 to 60 keV. The interpretation of the experimental results shows that the decay of the ^{178m2}Hf isomer can be triggered by X-rays at a power on the order of only mW/cm^2. The analysis of the gamma- gamma coincidence data has revealed that during X-ray irradiations, a line of 129.5 keV was found in coincidence with the 213.4 keV ground state band (GSB) transition. The 129.5 keV line has not been previously observed and is not a known transition of the spontaneous decay of ^{178m2}Hf. The 129.5 keV gamma transition must be a member of a sequence of gamma transitions that bypasses most of the normal decay cascade populated by spontaneous decay. This result leads to a deeper understanding of the (γ,γ') reactions at low energies. These photonuclear reactions are of special interest because of the importance of possible practical applications. The studies of the decay of nuclear isomers induced by X-rays should be seen in a larger perspective of the development of new controlled sources of incoherent gamma-radiation and they could also mean a significant progress for the development of a gamma-ray laser.

[132] T. B. Ryves, *The Decay Scheme of ^{180m}Ta*, J. Phys. G, **6** (1980), pp. 763–769. **Abstract:** The β$^-$ and electron-capture branching ratios in the decay of ^{180m}Ta and the K internal conversion coefficient of the 93 keV γ-ray were deduced from coincidence measurements between β$^-$, γ and K x-rays and a measurement of the relative γ-ray intensities. The uncertainties in the branching ratios were estimated by a Monte-Carlo technique. The half-life of ^{180m}Ta was also measured as (8.152+/-0.006) h.

[133] T. Saito, A. Shinohara, and K. Otozai, *Nuclear excitation by electron transition (NEET) in ^{237}Np following K-shell photoionization*, Physics Letters B, **92** (1980), p. 293. **Abstract:** Deexcitation γ-rays of ^{237}Np were detected during irradiation of ^{237}Np with the γ-radiation from ^{57}Co. This is explained as a result of the nuclear excitation of the 103 keV level in ^{237}Np induced by the KL_3 electronic radiationless transition following K-shell photoionization. The probability for nuclear excitation is found to be 2.1×10^{-4} per created K-hole.

[134] H. E. D. Scovil and E. O. Schulz-DuBois, *Three-Level Masers as Heat Engines*, Phys. Rev. Lett, **2** (1959), pp. 262 – 263. **Abstract:** The purpose of this note is to demonstrate that three-level masers can be regarded as heat engines.

[135] E. Segre and A. Hemholz, *Nuclear Isomerism*, Rev. Mod. Phys, **21** (1949), pp. 271–305.

[136] V. M. Semkova, *Accurate Measurements Of Neutron Activation Cross Sections*, Bulgarian Joumal of Physics, **26** (1999), pp. 29–42. **Abstract:** The applications of some recent achievements of neutron activation method on high intensity neutron sources are considered frorn the view point of associated errors of cross sections data for neutron induced reaction. The important corrections in γ-spectrometry insuring precise determination of the induced radioactivity, methods for accurate determination of the energy and flux density of neutrons, produced by different sources, and investigations of deuterium beam composition are considered as factors determining the precision of the experimental data. The influence of the ion beam composition on the mean energy of neutrons has been investigated by measurement of the energy of neutrons induced by different magnetically analysed deuterium ion groups. Zr/Nb method for experimental determination of the neutron energy in the 13 -15 MeV energy range allows to measure energy of neutrons from D-T reaction with uncertainty of 50 keV. Flux density spectra from D(d,n) Ed = 9.53 MeV and Be(d,n) Ed = 9.72 MeV are measured by PHRS and foil activation method. Future applications of the activation method on NG- 1 2 are discussed.

[137] M. B. Smith, P. M. Walker, G. C. Ball, J. J. Carroll, P. E. Garrett, G. Hackman, R. Propri, F. Sarazin, and H. C. Scraggs, *Gamma rays emitted in the decay of 31-yr $^{178}Hf^{m2}$*, Phys. Rev. C, **68** (2003), p. 031302. **Abstract:** The spontaneous decay of the Kπ=16$^+$, 31-yr ^{178}Hf m2 isomer has been investigated with a 15-kBq source placed at the center of a 20-element γ-ray spectrometer. High-multipolarity M4 and E5 transitions, which represent the first definitive observation of direct γ-ray emission from the isomer, have been identified, together with other low-intensity transitions. Branching ratios for these other transitions have elucidated the spin dependence of the mixing between the two known Kπ=8- bands. The M4 and E5 γ-ray decays are the first strongly K-forbidden transitions to be identified with such high multipolarities, and demonstrate a consistent extension of K-hindrance systematics, with an inhibition factor of approximately 100 per degree of K forbiddenness. Some unplaced transitions are also reported.

[138] ——, *γ rays emitted in the decay of 31-yr $^{178}Hf^{m2}$* , Phys. Rev. C, **68** (2003), p. 031302. **Abstract:** The spontaneous decay of the K$^\pi$=16$^+$, 31-yr ^{178}Hf^{m2} isomer has been investigated with a 15-kBq source placed at the center of a 20-element γ-ray spectrometer. High-multipolarity M4 and E5 transitions, which represent the first definitive observation of direct γ-ray emission from the isomer, have been identified, together with other low-intensity transitions. Branching ratios for these other transitions have elucidated the spin dependence of the mixing between the two known K$^\pi$=8$^-$ bands. The M4 and E5 γ-ray decays are the first strongly K-forbidden transitions to be identified with such high multipolarities, and demonstrate a consistent extension of K-hindrance systematics, with an inhibition factor of approximately 100 per degree of K forbiddenness. Some unplaced transitions are also reported.

[139] F. Stedile, *Generating an Inversion on a Nuclear Transition - Photopumping of ^{103}Rh*, Hyperfine Interactions, **143** (2002), pp. 133–142. **Abstract:** Quite a number of proposals for a gamma-ray laser have been made over the years. One first step on the way to a gamma-ray laser is an inversion between nuclear states. For the natural isotope ^{103}Rh we have the favorable condition that there are two low-lying nuclear levels at energies of 357 keV and 295 keV with lifetimes of 107 ps and 9.7 ps, respectively. With two Nuclear Resonance Fluorescence (NRF) measurements the population of these low-lying levels via feeding from higher-lying levels was investigated. Altogether 26 higher-lying nuclear levels that show a branching to one or even both interesting low-lying levels have been found. Summing over all contributions from these feeding levels this results in a population inversion between the levels at 357 keV and 295 keV of ^{103}Rh.

[140] I. Stefanescu, G. Georgiev, F. Ames, J. Äystö, D. L. Balabanski, G. Bollen, P. A. Butler, J. Cederkäll, N. Champault, T. Davinson, A. D. Maesschalck, P. Delahaye, J. Eberth, D. Fedorov, V. N. Fedosseev, L. M. Fraile, S. Franchoo, K. Gladnishki, D. Habs, K. Heyde, M. Huyse, O. Ivanov, J. Iwanicki,

J. Jolie, B. Jonson, T. Kröll, R. Krücken, O. Kester, U. Köster, A. Lagoyannis, L. Liljeby, G. L. Bianco, B. A. Marsh, O. Niedermaier, T. Nilsson, M. Oinonen, G. Pascovici, P. Reiter, A. S. H. Scheit, D. Schwalm, T. Sieber, N. Smirnova, J. V. D. Walle, P. V. Duppen, S. Zemlyanoi, N. Warr, D. Weisshaar, and F. Wenander, *Coulomb Excitation of ^{68,70}Cu: First Use of Postaccelerated Isomeric Beams*, Phys. Rev. Lett, **98** (2007), p. 122701. **Abstract:** We report on the first low-energy Coulomb excitation measurements with radioactive I=6$^-$ beams of odd-odd nuclei ^{68,70}Cu. The beams were produced at ISOLDE, CERN and were post-accelerated by REX-ISOLDE to 2.83 MeV/nucleon. γ rays were detected with the MINIBALL spectrometer. The 6$^-$ beam was used to study the multiplet of states (3$^-$, 4$^-$, 5$^-$, 6$^-$) arising from the $\pi 2p_{3/2}$ $\nu 1g_{9/2}$ configuration. The 4$^-$ state of the multiplet was populated via Coulomb excitation and the B(E2;6$^-$→4$^-$) value was determined in both nuclei. The results obtained illustrate the fragile stability of the Z=28 shell and N=40 subshell closures. A comparison with large-scale shell-model calculations using the ^{56}Ni core shows the importance of the proton excitations across the Z=28 shell gap to the understanding of the nuclear structure in the neutron-rich nuclei with N∼40.

[141] J. R. Stone and N. J. Stone, *Analysis of the Experiment into Possible Tuned X-ray Triggering of the Decay of ^{178m2}Hf carried out in at the CAMD Facility, Baton Rouge, La., in November 2003.*, Department of Chemistry and Biochemistry, University of Maryland, MD 20742, USA. Department of Physics, University of Oxford, Oxford, OX1 3PU, UK., 2004.

[142] Y. Sun, *Projected shell model description for nuclear isomers*, http://arxiv.org/abs/0803.1700v1. **Abstract:** Nuclear isomer is a current research focus. To describe isomers, we present a method based on the Projected Shell Model. Two kinds of isomers, K-isomers and shape isomers, are discussed. For the K-isomer treatment, K-mixing is properly implemented in the model. It is found however that in order to describe the strong K-violation more efficiently, it may be necessary to further introduce triaxiality into the shell model basis. To treat shape isomers, a scheme is outlined which allows mixing those configurations belonging to different shapes.

[143] Y. Sun, X.-R. Zhou, G.-L. Long, E.-G. Zhao, and P. M. Walker, *Nuclear structure of ^{178}Hf related to the spin-16, 31-year isomer*, Physics Letters B, **589** (2004), pp. 83–88. **Abstract:** The projected shell model is used to study the multi-quasiparticle and collective excitations of ^{178}Hf. With an axially symmetric basis, the spin-16 isomer at 2.4 MeV appears to be well separated in energy/spin space from other configurations. However, projected energy surface calculations suggest that ^{178}Hf has significant softness to axially asymmetric shapes, which can strongly modify the level distribution. The implications for photodeexcitation of the isomer are discussed.

[144] E. V. Tkalya, *Nuclear excitation in atomic transitions (NEET process analysis)*, Nuclear Physics A, **539** (1992), pp. 209–222. **Abstract:** The paper is devoted to a detailed analysis of the process of nuclear excitation in atomic transitions. All calculations are carried out within the framework of QED. A comparison with experimental results on nuclear excitation of ^{189}Os, ^{197}Au and ^{237}Np is made. The substantial difference between observed probabilities and theoretical estimates is revealed. The possibility of a direct experimental measurement of NEET probability is discussed.

[145] E. V. Tkalya, *Probability of L -shell nuclear excitation by electronic transitions in ^{178}Hf^{m2}*, Phys. Rev. C, **68** (2003), p. 064611. **Abstract:** We calculate the cross section for the x-ray induced accelerated decay process of the isomer ^{178}Hf^{m2}(16$^+$, 2.446 MeV, 31 yr) via an unknown nuclear state (including the case of a "mixed-K" state) hypothesized to lie about 10 keV in energy above the isomer. The excitation of the 31 yr isomer to an intermediate level is assumed to proceed via an electronic transition between atomic shells as has been recently postulated to explain the observation of "triggering" of the decay of the isomer by irradiation with x rays. The accelerated decay cross section is calculated in the most favorable case to be 10^{-32} cm^2, a value many orders of magnitude below that needed to explain the results of the isomeric triggering experiments.

[146] ——, *Induced decay of ^{178}Hf^{m2}: Theoretical analysis of experimental results*, Phys. Rev. C, **71** (2005), p. 024606. **Abstract:** This article reviews experimental results obtained recently on the x-ray-induced acceleration of the decay of the long-lived isomer ^{178}Hf^{m2} . Two basic mechanisms for the induced decay

are considered: (1) direct interaction of the incident x rays with the nucleus and (2) the nucleus-x-ray interaction proceeding via atomic shells. We establish that the absence of K forbiddenness for all transitions to a hypothetical "mixed K" level cannot explain the measured cross sections even if collective nuclear matrix elements, resonant conditions, and so on, are assumed. We also tested, and rejected, the hypothesis that the enhancement is due to normal nuclear transitions in the inverse nuclear excitation by electron transition process. The possibility to make measurements with intense laser radiation is considered too. Thus, there appears to be no explanation of these experimental results within quantum electrodynamics and the contemporary concepts of atomic nuclei.

[147] ——, *Induced decay of the nuclear isomer ^{178m2}Hf and the 'isomeric bomb'*, Physics - Uspekhi, **48** (2005), pp. 525–531.

[148] M. J. Trbovich, D. P. Barry, R. E. Slovacek, Y. Danon, R. C. Block, J. A. Burke, N. J. Drindak, G. Leinweber, and R. V. Ballad, *Hafnium Resonance Parameter Analysis Using Neutron Capture And Transmission Experiments*, Report LM-04K119, October 13, (2004). **Abstract:** The focus of this work is to determine resonance parameters for stable hafnium isotopes in the 0.005 - 200 eV region, with special emphasis on the overlapping ^{176}Hf and ^{178}Hf resonances near 8 eV. The large neutron cross section of hafnium, combined with its corrosion resistance and excellent mechanical properties, make it a useful material for controlling nuclear reactions. Experiments measuring neutron capture and transmission were performed at the Rensselaer Polytechnic Institute (RPI) electron linear accelerator (LINAC) using the time of flight method. ^{6}Li glass scintillation detectors were used for transmission experiments at flight path lengths of 15 and 25 m. Capture experiments were done using a sixteen section NaI(Tl) multiplicity detector at a flight path length of 25 m. These experiments utilized various thicknesses of metallic and isotopically-enriched liquid samples. The liquid samples were designed to provide information on the 176Hf and ^{178}Hf contributions to the 8 eV doublet without saturation. Data analysis was done using the R-matrix Bayesian code SAMMY version M6 beta. SAMMY is able to account for experimental resolution effects for each of the experimental setups at the RPI LINAC, and also can correct for multiple scattering effects in neutron capture yield data. The combined capture and transmission data analysis yielded resonance parameters for all hafnium isotopes from 0.005 - 200 eV. Resonance integrals were calculated along with errors for each hafnium isotope using the NJOY [1] and INTER [2] codes. The isotopic resonance integrals calculated were significantly different than previously published values; however the calculated elemental hafnium resonance integral changed very little.

[149] H. E. Türeci, L. Ge, S. Rotter, and A. D. Stone, *Strong Interactions in Multimode Random Lasers*, Science, **320** (2008), p. 646. **Abstract:** Unlike conventional lasers, diffusive random lasers (DRLs) have no resonator to trap light and no high-Q resonances to support lasing. Because of this lack of sharp resonances, the DRL has presented a challenge to conventional laser theory. We present a theory able to treat the DRL rigorously and provide results on the lasing spectra, internal fields, and output intensities of DRLs. Typically DRLs are highly multimode lasers, emitting light at a number of wavelengths. We show that the modal interactions through the gain medium in such lasers are extremely strong and lead to a uniformly spaced frequency spectrum, in agreement with recent experimental observations.

[150] P. Ugorowski, R. Propri, S. Karamian, D. Gohlke, J. Lazich, N. Caldwell, R. Chakrawarthy, M. Helba, H. Roberts, and J. Carroll, *Design and characterization of a compact multi-detector array for studies of induced gamma emission: Spontaneous decay of ^{178m2}Hf as a test case*, Nuclear Instruments and Methods in Physics Research Section A, **565** (2006), pp. 657–676. **Abstract:** Reports that incident photons near 10 keV can induce the emission of gamma rays with concomitant energy release from the 31-year isomer of ^{178}Hf challenge established models of nuclear and atomic physics. In order to provide a direct and independent assessment of these claims, a multi-detector system was designed as a specialized research tool. The YSU miniball is unique in its combination of performance characteristics, compact size and portability, enabling it to be easily transported to and placed within the confines of beamline hutches at synchrotron radiation sources. Monochromatic synchrotron radiation was used in the most recent studies from which evidence of prompt triggering was claimed, suggesting similar sites for independent tests of these results. The miniball array consists of six high-efficiency BGO scintillators coupled with a single 65% Ge detector and provides time-resolved gamma-ray calorimetry

rather than purely spectroscopic data. The need to record high detected folds from the array (up to seven-fold gamma coincidences) makes this system different in practice from standard spectroscopic arrays for which data is typically restricted to triples or lower folds. Here the system requirements and design are discussed, as well as system performance as characterized using the well-known natural decay cascades of ^{178m2}Hf. This serves as the foundation for subsequent high-sensitivity searches for low-energy triggering of gamma emission from this isomer.

[151] V. Vali and W. Vali, *Induced γ-Ray Emission*, Proceedings of the IEEE, **51** (1963), pp. 182–184. **Abstract**: The extension of optical maser techniques to the γ-ray region is considered. It is shown that under certain conditions induced γ rays can be produced. The condition of criticality rather than that of oscillation is used because a γ-ray maser does not have a resonant structure. The main observable effects are the shortening of lifetimes in some γ excitations and the appearance of two or more coherent γ quanta. It appears to be difficult to prepare samples that will become critical. The principles are applied to a specific example.

[152] P. von Neumann-Cosel and A. Richter, *Comment on "Accelerated Emission of Gamma Rays from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation"* , Phys. Rev. Lett, **84** (1999), p. 2543.

[153] P. M. Walker, *Mechanisms of K Mixing: the Role of Octupole Vibrations in ^{180}Ta*, Hyperfine Interactions, **143** (2002), pp. 143–151. **Abstract:** K -mixing mechanisms are reviewed, with reference to isomers in the A $\approx$ 180 region of well deformed nuclei. The strong E1 photoactivation of ^{180}Ta^m, through states at 1.1, 1.3 and 1.5 MeV, provides evidence for the role of octupole vibrations in the conversion of the K = 9 isomer to the K = 1 ground state.

[154] P. M. Walker, D. M. Cullen, C. S. Purry, D. E. Appelbe, A. P. Byrne, G. D. Dracoulis, T. Kibédi, F. G. Kondev, I. Y. Lee, A. O. Macchiavelli, A. T. Reed, P. H. Regan, and F. Xu, *K-forbidden transitions from multi-quasiparticle states*, Physics Letters B, **408** (1997), pp. 42–46. **Abstract:** Four-quasiparticle, K$^\pi$ = 12$^+$ states in ^{172}Hf and ^{178}W, with half-lives less than 2 ns, are found to decay to their respective Kπ = 0$^+$ ground-state bands, in competition with transitions to intermediate-K, two-quasiparticle structures. All decay transitions are weakly hindered. When taken together with other K-isomer decay rates, an overall trend of decreasing hindrance with increasing excitation energy is evident. Estimates based on density-of-states considerations approximately reproduce the trend of the data, but also indicate the need to include other K-mixing effects.

[155] P. M. Walker and G. D. Dracoulis, *Exotic Isomers in Deformed Atomic Nuclei*, Hyperfine Interactions, **135** (2001), pp. 83–107. **Abstract:** Excited states of atomic nuclei can have long half lives, due to the angular-momentum couplings of unpaired nucleons. Such isomeric states provide opportunities for exploring novel nuclear physics, astrophysics and physics at the atomic/nuclear interface. This review focuses on the properties of isomers in deformed nuclei, and emphasises the importance of axial symmetry in preserving the integrity of the K quantum number. A region of neutron-rich nuclei around ^{188}Hf (Z = 72, N = 116) is predicted to have exceptional isomer properties, and experimental advances are now opening up this region to detailed investigation.

[156] P. M. Walker, G. D. Dracoulis, and J. J. Carroll, *Interpretation of the excitation and decay of ^{180}Ta^m through a K^π=5$^+$ band*, Phys. Rev. C, **64** (2001), p. 061302. **Abstract:** The production and survival of ^{180}Ta in stars presents a nuclear structure and astrophysics puzzle. From earlier work in the laboratory, photon scattering from the 9$^-$, 75 keV isomeric state has been shown to result in eventual de-excitation to the relatively short-lived 1$^+$ ground state, through intermediate states whose character is uncertain. It is suggested here that the lowest observed photon resonances match the energies of known states in the rotational band based on the recently assigned K$^\pi$=5$^+$ state at 592 keV, states which are accessible by E1 excitation from the 9$^-$ isomer. Analysis of the previously observed resonance strengths and the known in-band properties implies γ-ray E1 transition probabilities which are stronger than expected given the nominal K forbiddenness. Predicted widths for "back"-decay to the 9$^-$ isomer and its rotational band are calculated and shown to be significant. Their observation would have implications for the proposed associations and mixing effects above the yrast line.

[157] ——, *Interpretation of the excitation and decay of ^{180}Ta^m through a K^π=5$^+$ band*, Phys. Rev C, **64** (2001), p. 61302. **Abstract:** The production and survival of ^{180}Ta in stars presents a nuclear

structure and astrophysics puzzle. From earlier work in the laboratory, photon scattering from the 9-, 75 keV isomeric state has been shown to result in eventual de-excitation to the relatively short-lived 1+ ground state, through intermediate states whose character is uncertain. It is suggested here that the lowest observed photon resonances match the energies of known states in the rotational band based on the recently assigned Kπ=5+ state at 592 keV, states which are accessible by E1 excitation from the 9- isomer. Analysis of the previously observed resonance strengths and the known in-band properties implies γ-ray E1 transition probabilities which are stronger than expected given the nominal K forbiddenness. Predicted widths for "back"-decay to the 9- isomer and its rotational band are calculated and shown to be significant. Their observation would have implications for the proposed associations and mixing effects above the yrast line.

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prolate shapes. Highly excited, long-lived states are predicted. However, at angular momenta close to $40\hbar$, orbital alignment effects for collective oblate rotation lead to a deep potential minimum that competes energetically with the prolate multiquasiparticle states.

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[165] A. A. Zadernovsky, *Gamma-Ray Laser with Hidden Population Inversion of Nuclear States*, Laser Physics, **17** (2007), pp. 316–323. **Abstract:** A comparative analysis of various schemes of a gamma-ray laser is presented. All of the schemes employ a hidden inversion of nuclear states, which appears in cooled nuclear ensembles due to nuclear recoil in the emission and absorption of gamma photons.

[166] A. A. Zadernovsky and J. J. Carroll, *Non-Radiative Triggering of Long-Lived Nuclear Isomers*, Hyperfine Interactions, **143** (2002), pp. 153–174. **Abstract:** The triggering of long-lived isomeric nuclei by non-radiative excitation to a relatively short-lived mediating state is considered. Coulomb triggering in inelastic scattering of heavy ions, a transfer of triggering energy from resonant electron transitions of atomic shell (NEET) and triggering by capture of a free electron into a bound atomic state (NEEC) are discussed. Cross sections for the above processes of non-radiative triggering are presented and the relative efficiencies of these different triggering mechanisms are discussed. Numerical estimates are presented for the selected isomers.