

Isomer Research: Energy Release Validation, Production, and Applications

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NUCLEAR ENERGY RESEARCH INITIATIVE

Isomer Research: Energy Release Validation, Production, and Applications

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Collaborators:

Project Start Date: April 2000 Projected End Date: September 2003

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Research Objectives

The goal of this applied nuclear isomer research program is the search for, discovery of, and practical application of a new type of high energy density material (HEDM). Nuclear isomers could yield an energy source with a specific energy as much as a hundred thousand times as great as that of chemical fuels. There would be enormous payoffs to the Department of Energy and to the country as a whole if such energy sources could be identified and applied to a range of civilian and defense applications. Despite the potential payoff, efforts in applied isomer research have been rather limited and sporadic. Basic research on nuclear isomers dates back to their discovery in 1935 with occasional hints to tantalize interest in HEDM. In most cases, these hints were refuted following careful examination by other groups.

The isomer research area is rich with possibilities: we prioritized several areas likely to be the most rewarding and fruitful for initial experimental investigation because these areas directly bear on important issues: Can the energy stored in nuclear isomers be released on demand? Is the size of the atomic-nuclear mixing matrix element large enough to be useful? Under what circumstances? Can we initiate quantal collective release of isomeric energy from a Mössbauer crystal?

Specific experiments targeted to provided some answers include:

- X-ray induced decay of isomeric Hf ($^{178m2}\text{Hf}$) with a sensitivity 10^5 times recent work
- NEET: A measurement of the atomic-nuclear mixing matrix element in ^{189}Os
- Stimulated Emission in isomeric (^{125m}Te)
- Superradiance in isomeric (^{93m}Nb)

Research Progress: Induced decay of Isomeric Hf-178

Triggered decay is key to extracting energy on demand from nuclear isomers. The issue of the cross section for x-ray induced decay of the isomeric state of ^{178}Hf ($E_x = 2.4$ MeV, $t_{1/2} = 31$ yr) has received significant attention and publicity in the past few years. Our research in the past 2 years focused on the question: What is the cross section for x-ray (keV) induced decay of the 31-yr isomer in the nucleus ^{178}Hf with nuclear excitation energy 2.4 MeV? The question is relevant because x-ray induced decay of isomeric ^{178}Hf had been reported [Collins, et al., Phys. Rev. Lett. 82, 695 (1999)], with the integrated cross section of 10^{-21} cm²-keV, and affirmed in an experiment with synchrotron radiation at SP-ring8 [Collins, et al., Europhys. Lett. 57, 677 (2002)]. This cross section is enormous, many orders of magnitude greater than nuclear cross sections. Our ANL, LANL, LLNL collaboration reported in 2001 [Ahmad, et al., Phys. Rev. Lett. 87, 072503, 2001]) an upper limit some 5

orders of magnitude below Collins 1999 for $E_x > 20$ keV. Nevertheless, agitation continued in the DOE community regarding the possibility of isomeric Hf-178 as an energy source.

In 2002, we designed and fielded a 2nd experiment at the ANL Advanced Photon Source (APS) with an experimental arrangement optimized for low energy x-ray bombardment, but still taking advantage of the intense “white” beam to include any possible x-ray triggering energy. Thin, electroplated Hf targets enriched in the 31-y isomer mounted on Be disks were used in the experiment. The result illustrated in Fig. 1 is an upper limit for the cross section $\sigma_{x\text{-ray}}$ for induced x-ray emission less than 10^{-27} cm²-keV at the 99% confidence limit for $E_x > 6.5$ keV. This result is orders of magnitude below other published claims (Ahmad, et al., UCRL-JC-150843. The manuscript with minor revisions has been accepted by Phys. Rev. C and publication is expected in April, 2003.)

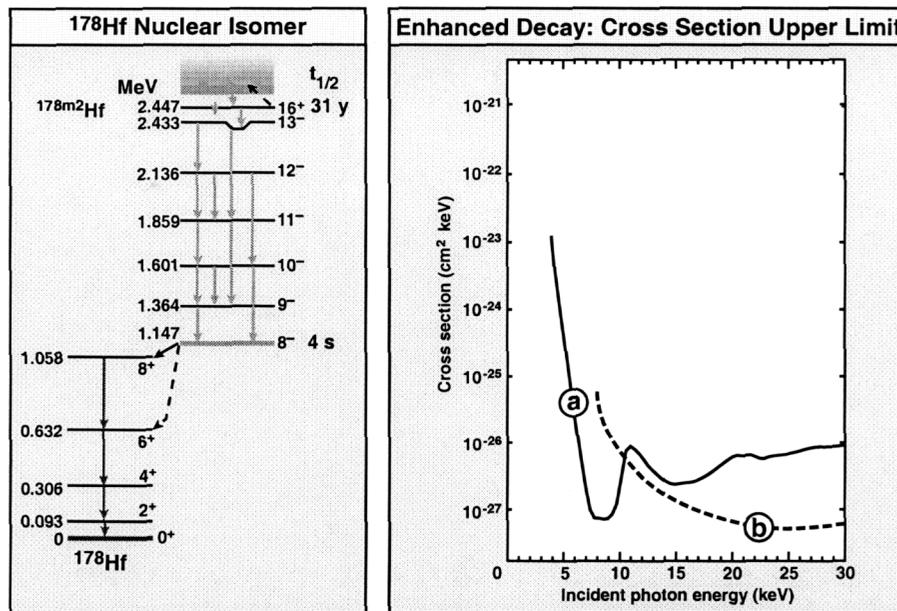


Figure 1. Decay scheme of 31-y isomeric Hf-178 (lhs). The cross section results of the ANL/LANL/LLNL collaboration are also illustrated (rhs). The results illustrated are from (a) the 2001 bombardment of the mixed HfO₂ (isomer enriched) + Al sample (Ahmad, et al. (2001) and (b) the 2002 bombardment with the thin electroplated sample (Ahmad, et al., UCRL-JC-150843). The thin target bombardment extends the upper limit for enhanced production to lower x-ray energies (6 keV). Results from the two experiments are consistent. The limit (b) is based on a 99 % confidence limit in the statistical uncertainty in the measurement together with a factor of 2 for estimated systematic errors.

Research Progress: Atomic-Nuclear Mixing in Os-189

An experiment has been designed to measure the atomic-nuclear mixing matrix element in ¹⁸⁹Os, by measuring the rate of inverse internal conversion process in ¹⁸⁹Os. The experiment takes advantage of the LLNL Electron Beam Ion Trap. The scheme is illustrated in Fig. 2. The capture of an electron of appropriate energy into an atomic state of He-like ¹⁸⁹Os would resonantly excite a nuclear level at 216.7 keV. This level decays about 22% of the time to a 5.7-hour, 31 keV metastable nuclear state. The experimental signature of inverse internal conversion is the decay of this metastable state.

He-like ^{189}Os (i.e., $^{189}\text{Os}^{74+}$) is made in EBIT by stripping neutral or low charge state Osmium in the EBIT ion trap of and successively stripping the electrons with a high-energy electron beam. The electron beam has two functions, 1) stripping the Osmium to the He-like state, and 2) producing 196 keV electrons. The capture of a 196-keV electron into the 2s state (l-shell) of He-like ^{189}Os produces an atomic nuclear degeneracy at 216.7 keV x-ray.

Research progress consisted of:

1. EBIT high voltage upgrade from low voltage mode (< 30 keV electron beam energy) to high voltage mode (up to ~220 keV electron beam energy).
2. Design and construction of a source to inject ^{189}Os into the EBIT
3. Design and construction of the detectors to measure the x-rays and electrons emitted by the decay of the 31 keV metastable nuclear state
4. Conditioning of EBIT for high voltage operation.
5. Certified safety reports for EBIT

Work in 2002 consisted of meeting requirements 1 – 5. We also made contact with 2 theoretical physicists (Harston, D. Gogny) to develop estimates of the expected cross section. Measurements will begin in 2003.

Research Progress: Superradiance in Isomeric ^{93m}Nb

Superradiance is an effect discovered by Robert Dicke in the 1950's that results in an enormous broadening of the photon channel through the cooperation of an ensemble of quantum excited states. The challenge of growing crystals that exhibit nuclear superradiance rests upon the limited availability of ^{93m}Nb enriched material. To date, two strategies for growing single crystals containing ^{93m}Nb have been employed. The first strategy is the epitaxial growth of lithium niobate (LiNbO_3) from lithium ethoxide solutions, and the second strategy is the growth of single crystals of potassium fluoroniobate (K_2NbF_7).

For the epitaxial growth of lithium niobate, our team has succeeded in the manufacture of thin LiNbO_3 films on substrates of both sapphire and lithium tantalate. However, epitaxial growth has yet to be verified via X-ray diffraction analysis (XRD). The difficulty of XRD in our case lies in the tiny thickness of the film compared to the underlying substrate crystal. Hence, observance of diffraction peaks caused by the additional thin crystal layer is non-trivial. Furthermore, a chemical procedure for the synthesis of lithium niobate has been developed, allowing very small Nb mass quantities in aqueous solutions as starting material to be converted into niobium chloride at the complete absence of moisture. For the second crystal growing strategy, the synthesis of crystalline potassium fluoroniobate has been successfully performed. The appearance and solubility of the crystals are consistent with that of potassium fluoroniobate.

Planned Activities

We are moving on from the isomeric Hf issue to attack the physics of nuclear isomers relevant to use as an energy source. During the coming year, we will particularly focus on:

- **NEET, Nuclear Excitation by Electronic Transition, in ^{189}Os .** The process has been demonstrated in ^{197}Au , in a recent synchrotron experiment in Japan. The matrix element is similar to the (inverse) internal conversion matrix element. Important conditions for NEET to compete with real photon emission to include an energy overlap of the atomic and nuclear states, and a common multipolarity of the atomic and nuclear transitions. We are developing an experiment to prepare ionized atomic ^{189}Os by bombardment with a

variable energy electron beam in an electron beam ion trap (EBIT) and pump a nuclear state in ^{189}Os at 216.6 keV. The energy of the electron beam is carefully controlled and tuned so that the sum of the energies of the bombarding electron beam and the L-shell ionized ^{189}Os (a free-bound transition) add up to the excitation of the nuclear ^{189}Os level at 216.6 keV. Trapped ions are periodically gathered up and counted. The signal is the energy and decay rate of the $J^{\pi} = 9/2^{-}$, $E_x = 30.814$ keV, $t_{1/2} = 5.7$ h state, populated in the decay of the 216.6-keV nuclear state. The experiment will be sited at LLNL's EBIT facility.

- **Superradiance in isomeric ^{93m}Nb .** During the next period, we will verify the epitaxial growth of small lithium niobate quantities on sapphire and tantalate substrates by means of XRD. XRD will also be used to confirm the structure and purity of the potassium fluoroniobate crystals. The synthetic procedure for the synthesis of niobium ethoxide, the chemical precursor to lithium niobate, will be optimized. The epitaxial growth of lithium niobate will continue to be optimized. The procedure for the growth of potassium fluoroniobate crystals will be scaled down to the 1 mg level, initially with non-radioactive Nb. Crystals will then be grown with trace amounts of ^{91m}Nb , which will be used to calculate the size of the crystals grown. Single crystals of potassium fluoroniobate enriched with ^{93m}Nb will then be grown and experiments to observe superradiance will be conducted.