Search for X-ray-induced Acceleration of the Decay of the 31-year Isomer of ^{178}Hf by Using Synchrotron Radiation

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Introduction

Releasing the energy stored in an isomeric nuclear state in a controlled way with an atomic or electromagnetic trigger is an attractive speculation: the energy gain may be on the order of the ratio of nuclear/atomic energies — MeV/keV. (Nuclear isomers are loosely defined as excited nuclear states with lifetimes longer than 10^{-9} s.) Nuclear isomers, therefore, represent an opportunity for a stand-alone energy source if suitable schemes for trigger and control of the energy release can be found. Potential applications include space drive as well as very bright γ-ray sources [1].

The nucleus ^{178}Hf has a nuclear isomer with excitation energy E_x = 2.447 MeV (Fig. 1). The 2.447-MeV isomeric state decays slowly ($t_{1/2}$ = 31 y) to the nearby state at 2.433 MeV. The J^π = 13^- state loses energy in a rapid ($t \sim 10^{-12}$ s) γ-ray cascade ending at the 8^- rotational band head, which, in turn, decays via the ground-state rotational band cascade. The γ-ray cascade is delayed at the 8^- state at 1.147 MeV, since the 8^- state is also isomeric, with $t_{1/2}$ = 4 s. Very scarce quantities of the 16^+, 31-yr isomer are available for research (~ 10^{15} atoms).

Reports of triggered decay of the ^{178}Hf isomer induced by x-rays delivered by a dental x-ray machine have been made [2-5]. Enhancements of ~ 1-2% in the isomer decay rate ($dN/dt = -(1+\varepsilon)N/\tau$) had been reported for various γ-rays in the cascade (distinguished by red and vertical lines in Fig. 1). The reported integrated cross section for triggering the decay is 10^{-21} cm^2 keV, so large as to demand new physics. We have sought to verify these reports by taking advantage of the intense photon flux available at beamline 1-ID at APS.

Methods and Materials

Samples of HfO_2 that included ~ 10^{15} atoms of isomeric 31-yr ^{178}Hf were irradiated at SRI CAT 1-ID in March 2001. Samples were fabricated at LANL by using material chemically extracted from the LANSCE/LAMPF

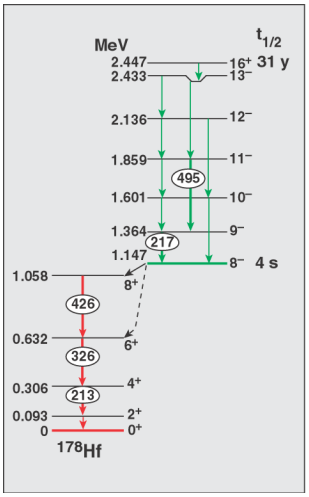

FIG. 1. Nuclear energy level diagram showing the decay of the 31-yr ^{178}Hf isomer. Transition energies are labeled in keV. Transitions reported as enhanced are highlighted.

target/beam stop. The undulator 1-ID was operated with maximum taper (5 mm) in the gap and two average gap settings: 15 mm and 20 mm. This arrangement generated a smooth "white" photon flux peaking at ≈ 2×10^{15} photons/keV-s at E_{ph} ≈ 16 keV and extending in energy to well over 100 keV (Fig. 2.) The 1-ID white beam was mechanically chopped to form a pulse train of 11-s beam-on and 22-s beam-off during the irradiation intervals, ≈ 8 h for each of three samples. Precision γ-ray spectrometers based on Ge detectors with energy resolution characterized by full-width, half-maximum (FWHM) of ≈ 1.0 keV at E_γ = 300 keV were used to count the sample during the irradiation cycles. Individual γ-rays characteristic of the ^{178}Hf isomer decay were easily identified in the γ-ray spectra, along with beam-induced florescent Hf K x-rays. These x-rays were used to monitor beam incident on the sample: The measured value agreed within a factor of two of the calculated beam flux. The

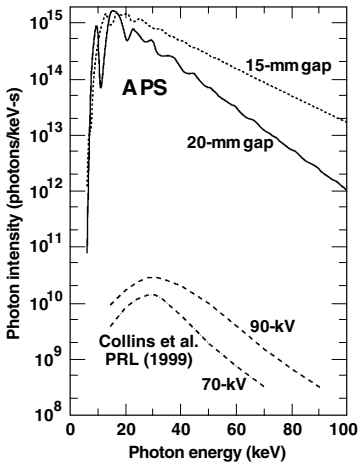

FIG. 2. The calculated photon intensity from the APS incident on the 2-mm-diameter area of the target material in the present experiment, for the two tapered settings of the undulator gap that were used. Photon intensities given in previous experiments for the 70- and 90-kV settings of the dental x-ray machine on a 1-cm-diameter target are also shown.

experimental signal of triggered isomer decay is an increase in the yield of characteristic ^{178}Hf γ-rays, above the background decay rate of the 31-yr isomer (Fig. 1). The experimental design gave the experimental team two chances to observe any increase in isomer depopulation: during the 11-s beam-on sample intervals or during the 22-s beam-off interval. Analysis of the spectra obtained in the beam-off interval is especially attractive because (1) there is no background from scattered beam photons, and (2) a fraction of the decay γ-rays are caught and held up in the 4-s, 8^- level midway down the decay chain and therefore have a characteristic 4-s half-life (Fig. 1). Counts due to isomer triggering will be in excess of the expected counts from the radioactive 31-yr isomer at the beginning of the 22-s counting interval and will decay away with a 4-s half-life during the 22-s counting interval, leaving the background of counts from the 31-yr isomer.

A straightforward procedure to isolate decays induced by triggering that cascade through the 4-s isomer is to divide the 22-s counting interval into two equal parts, integrate the γ-ray spectra of the corresponding halves $C(1)$ and $C(2)$, over time, form the difference spectrum $C(1) - C(2)$, and identify net counts correlated with the characteristic of the ^{178}Hf isomer decay γ-rays. The result for the HfO_2 sample R1 containing 7.3×10^{14} isomeric atoms is shown in the lower portion of Fig. 3, which expresses the count rate difference as $[C(1) - C(2)]/C(2)$, in percent. The upper portion of Fig. 3 illustrates the γ-ray spectrum associated with the decay of isomeric ^{178}Hf. Clearly there is no net positive enhancement for the characteristic γ-rays identified earlier (labeled in Fig. 1); instead, the decay rate is constant within 2%. The number of triggered events is given by the product of the cross section for triggering σ, with the incident photon flux and the number of isomeric atoms per unit area, $\phi\, N/A$, well-characterized quantities. Our experimental upper limit to the integrated cross section for isomer depopulation σ_{int} (after correction for self-absorption in the HfO_2 sample) is illustrated in Fig. 4. The limit is less than 2×10^{-27} cm^2 keV for incident photon energies between 20 and 60 keV,

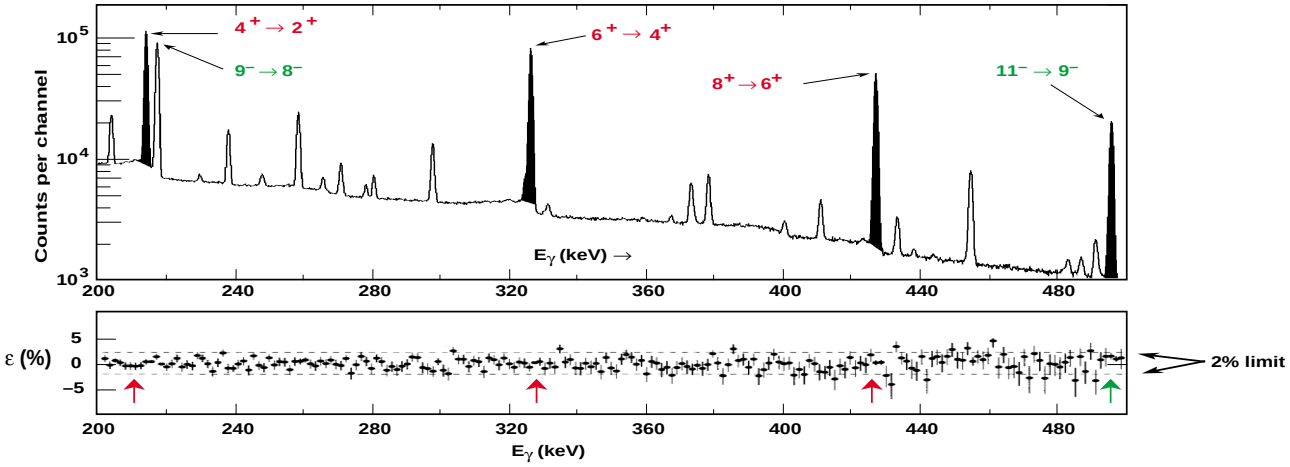

FIG. 3. A partial γ-ray spectrum of isomeric 31-yr ^{178}Hf. Photopeaks for the transitions at 213, 217, 426, 426, and 495 keV are filled in for the previously reported enhanced transitions. The spectrum is an accumulation of ~ 22-s counting periods, immediately following the 11-s irradiations of the R1 sample. The average undulator gap for these data was 15 mm, and the data were accumulated over 8.5 h. The points with error bars show the difference spectrum between the first half and the second half of the 22-s counting interval, in percent. This difference, with the points summed over an energy interval corresponding to the detector resolution, reflects any triggered excess in de-excitation through the 4-s isomer. The dashed lines indicate 2% limits in the difference.

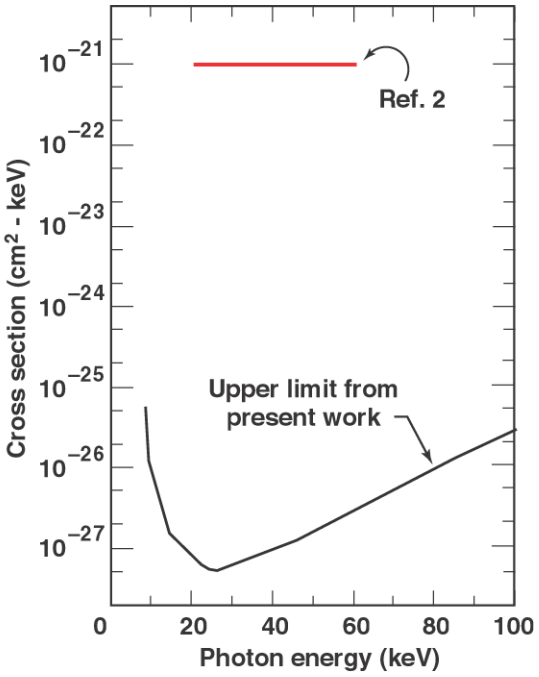

FIG. 4. Upper limit of the cross section for photon-induced de-excitation of the 31-yr ^{178}Hf isomer through the 4-s, 8$^-$ isomer on the basis of the measurements reported here. The value for this cross section reported in Ref. 2 is also shown.

more than five orders of magnitude below the previous positive reports. The present upper limit is clearly discrepant with earlier work. Finally, the limit discussed here is for triggered decay cascading through the 8$^-$ state; if the triggered decay mode bypasses the 8$^-$ state, the limits are a factor of ~ 10 higher. More details of this experiment and the results can be found in Ref. 6.

Discussion

The goal of the experiment was to verify claims in the literature of an extraordinary cross section for triggered decay of the 31-yr ^{178}Hf isomer, induced by x-ray bombardment. Our result sets an upper limit for the cross section over a wide range of incident photon energy that is consistent with nuclear physics estimates for the process and orders of magnitude below the previous work. Prospects for practical use of nuclear isomers as energy storage devices, triggered on demand, are still speculative.

Acknowledgments

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