Search for X-Ray Induced Acceleration of the Decay of the 31-Yr Isomer of ^{178}Hf Using Synchrotron Radiation

I. Ahmad,[1] J. C. Banar,[2] J. A. Becker,[3] D. S. Gemmell,[1] A. Kraemer,[3] A. Mashayekhi,[4] D. P. McNabb,[3] G. G. Miller,[2] E. F. Moore,[1] L. N. Pangault,[2] R. S. Rundberg,[2] J. P. Schiffer,[1] S. D. Shastri,[4] T. F. Wang,[3] and J. B. Wilhelmy[2]

[1]*Physics Division, Argonne National Laboratory, Argonne, Illinois 60439*
[2]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545*
[3]*Lawrence Livermore National Laboratory, Livermore, California 94550*
[4]*Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439*

(Received 18 April 2001; published 27 July 2001)

Enhanced decay of the 31-yr isomer of ^{178}Hf induced by x-ray irradiation has been reported previously. Here we describe an attempt to reproduce this result with an intense "white" x-ray beam from the Advanced Photon Source. No induced decay was observed. The upper limits for the energy-integrated cross sections for such a process, over the range of energies of 20–60 keV x rays, are less than 2×10^{-27} cm^2 keV, below the previously reported values by more than 5 orders of magnitude; at 8 keV the limit is 5×10^{-26} cm^2 keV.

DOI: 10.1103/PhysRevLett.87.072503 PACS numbers: 27.70.+q, 23.20.–g, 23.90.+w, 25.20.Dc

The 31-yr isomer of the nucleus ^{178}Hf, a K isomer with spin $J^\pi = 16^+$ at an excitation of 2.446 MeV, has been the subject of considerable speculation as to its possible use in gamma-ray lasers and other applications in which the energy of the isomeric state might be rapidly emitted [1]. Such use depends on finding a mechanism that could trigger the emission [2]. Recently, Collins *et al.* [3] reported the accelerated emission of gamma rays from a sample of this isomer when it was irradiated with photons from a dental x-ray machine. This development led to intensified conjectures about gamma-ray lasers and related phenomena [4]. However, it also prompted a set of criticisms [5] pointing out that the integrated cross section found in [3] for resonant absorption of x rays (1×10^{-21} cm^2 keV) was ~7 orders of magnitude too large to be consistent with values typical of photon absorption in this mass region. Collins *et al.* have published reports of further measurements [6], qualitatively consistent with their original result. We carried out an independent measurement designed to verify the results reported in [3,6].

Figure 1 shows the relevant energy levels and band structure in ^{178}Hf. In its normal decay mode, the 16^+ isomeric state decays through an $E3$ transition to the 13^- member of the $K = 8$ band whose $J^\pi = 8^-$ bandhead is itself an isomer decaying with a 4-s half-life to the 8^+ member of the ground-state band, followed by a cascade to the ground state. The hypothesis offered by [3,6] for their observation of accelerated decay postulates a state (or states) of mixed K, thought to be populated from the 16^+ isomeric state by resonant absorption of x-ray photons with energies in the range 20 to 60 keV. Such an intermediate state then may decay to a lower-lying level in the $K = 8$ band and thence to the 8^- bandhead, whose deexcitation with a 4-s half-life cascades in a well-known way, down through the ground-state band.

The x-ray machine used in [3,6] was operated at voltages in the range of 63–90 kV for irradiation of a target containing 6.3×10^{14} isomeric ^{178}Hf nuclei distributed over a 1-cm diameter area in a sealed plastic container. Gamma-ray spectra were measured with a Ge detector and it was found that selected transitions in the normal decay cascade of the isomer increased by about 4%, implying an integrated cross section of about 10^{-21} cm^2 keV for absorption into and decay from the intermediate K-mixed state(s). In other reports of the same series of experiments cross sections range from 2×10^{-22} to 2×10^{-21} cm^2 keV [and down to $(2–3) \times 10^{-23}$ cm^2 keV if the relevant x-ray energy

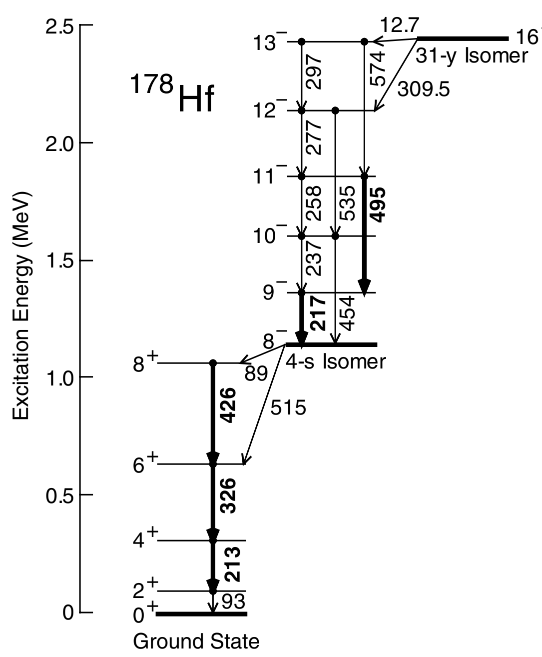

FIG. 1. Energy level diagram showing the decay of the 31-yr ^{178}Hf isomer. The transition energies are labeled in keV. Those transitions that were reported to be enhanced in [3,6] are highlighted.

happened to coincide with the energy of the tungsten K x-ray lines] [6].

In the present measurements we used a "white" beam from a tapered undulator insertion device [7] at the SRI-CAT 1-ID beam line of the Advanced Photon Source (APS) at Argonne National Laboratory with x-ray intensities that were over 4 orders of magnitude larger than those in [3,6] (see Fig. 2). The size of the beam spot at the target, 37 m from the undulator, was 2×2 mm. The power level of the x-ray beam incident on the targets was limited to a maximum of $\sim$210 W, corresponding to an average undulator gap of 15 mm, after the windows and absorbers used in the beam line. During the measurement, the stored electron beam at the APS was maintained at a steady 100 mA through the use of a continuous "top-up" mode of operation.

We employed three targets, designated $R1$, $R2$, and $R3$, containing 7.3×10^{14}, 3.0×10^{15}, and 6.4×10^{15} isomeric ^{178}Hf nuclei, respectively. The Hf was produced at Los Alamos National Laboratory using the LAMPF/LANSCE accelerator. The reaction mechanism was 800-MeV proton spallation on thick Ta targets/beam stops. The Hf was chemically extracted from the Ta target material, purified, precipitated and fired to produce HfO_2 for use in the current experiment. In addition to the 31-yr ^{178}Hf isomer, the target contained some stable Hf from impurities in the beam stop. The spallation reaction also produced other Hf isotopes, including trace amounts of ^{172}Hf ($t_{1/2} = 1.89$ yr), that proved useful in monitoring the target during the experiment.

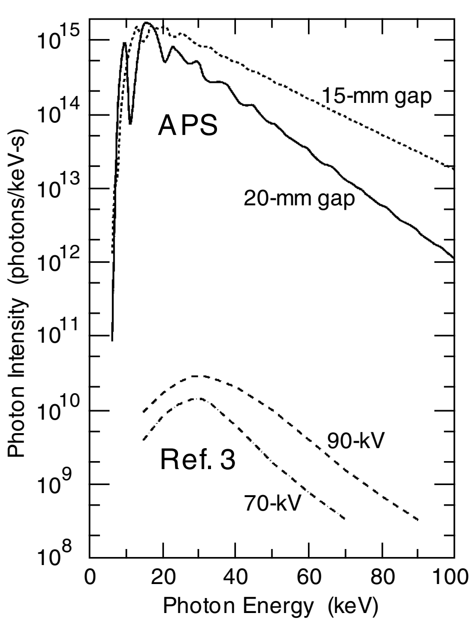

FIG. 2. The calculated photon intensity from the Advanced Photon Source, incident on the 2-mm-diameter area of the target material in the present experiment, for the two tapered settings of the undulator gap that were used. The photon intensities given in Refs. [3,6] for the 70- and 90-kV setting of the dental x-ray machine on a 1-cm diameter target are also shown.

Several windows and filters in the x-ray beam reduced the beam intensity, especially at energies below 10 keV. The target assembly was able to handle the power density and thereby preserve the integrity of the radioactive target material. For each of the three targets, the hafnium oxide was mixed with fine aluminum powder and then sealed by electron-beam welding into a 2-mm diameter by 1.6-mm deep cavity machined into a water-cooled aluminum block. The aluminum entrance window for each target was 0.15-mm thick.

The undulator was operated with maximum taper (a total of 5 mm) in the gap and at two average gap settings (15 and 20 mm). In this way, as is shown in Fig. 2, the sharp energy structure inherent in undulator radiation was smoothed out and the two gap settings filled in the remaining variations of photon intensity with energy. At these photon intensities, the few percent enhancement that had been seen in the earlier work [3,6] should become a manyfold increase in the gamma-ray intensity from induced decays.

X rays and gamma rays emitted from the target were measured using two planar Ge detectors, (A) and (B), mounted on opposite sides of the target in the horizontal plane and at 90° to the horizontally incident photon beam. The Ge crystals were both 50 mm in diameter and their thicknesses were 17 mm (A) and 15 mm (B). The distances from their front faces to the target were 36 and 39 cm, respectively. The detectors were collimated and heavily shielded with Pb. Absorbers were introduced as needed in front of the detectors to reduce counting rates to a few thousand per second. The target was turned 15° from normal incidence of the photon beam.

Each target was carefully aligned with the photon beam by maximizing the yield of beam-induced hafnium K x rays. These Hf K x rays were used as an in-beam monitor of the photon fluence on the target material in all measurements. This technique insured that the target integrity was maintained and that the experimental luminosity remained at the expected values.

The main measurements involved runs with three targets at two undulator gap settings, each accumulating data over a period of about 10 h. While data were accumulated continuously, the beam was cycled on and off the target by means of moving a tungsten shutter. In each cycle, the beam was on the target for 11 s (t_i) and then off for 22 s. For data-analysis purposes, the beam-off period was divided into two equal periods of slightly less than 11 s (t_1 and t_2). To protect the detectors, Pb shields 2.54-cm thick (A) and 1.27-cm thick (B) were moved in front of the detectors during the beam-on part of the cycle. In the case of detector A, this shield contained a 2.4-mm diameter collimating hole to permit the aforementioned monitoring of the beam-induced Hf x rays. The detected photons were recorded in event-by-event mode, with the time relative to the beginning of the cycle recorded for each event.

A beam-induced decay of the isomer would result in an increased production of the 4-s isomer, and the emission of the subsequent gamma rays in the ground-state band during the beam-off period. In particular, this would result in an enhanced counting rate in interval t_1 as compared to t_2.

The relevant portion of one of our spectra is presented in Fig. 3 in a format similar to that used in [3,6]. Plotted are the sum and the difference of spectra accumulated for one target and undulator setting. Similar results were found for all three targets, both average undulator gap settings, and both detectors. (Because of an unexpected accelerator shut-down only one undulator setting was used with the $R2$ target.) We found no statistically significant difference in the gamma-ray intensities between the periods t_1 and t_2 for any gamma-ray line in the spectrum.

This result is to be compared to what one would have expected, based on the values of [3,6], where it was found that the decay transitions were enhanced (accelerated) by 2%–6%. The enhancement factor ϵ ($\sim$ 0.04 in the case of [3]) can be estimated roughly (ignoring small factors due to absorption in the target, etc.) from the relation

$$\epsilon N/\tau = (N/A)\phi\sigma_{\rm int}\,, \tag{1}$$

where N is the number of isomeric target atoms, τ is the lifetime of the isomeric state (1.4×10^9 s), A is the area of the target, ϕ is the number of incident photons/s keV at the resonant absorption energy, and $\sigma_{\rm int}$ is the integrated cross section for the postulated absorption leading to deexcitation of the isomer. N/τ is the normal decay rate of the isomeric nuclei in the target. This then gives

$$\epsilon = (\tau/A)\ \phi\sigma_{\rm int}\,. \tag{2}$$

In [3,6] the value of $\epsilon = 0.04$ was reported. With their values of $A = 0.8$ cm^2 and $\phi = \sim 2 \times 10^{10}$ photons/s keV, a cross section of $\sigma_{\rm int} = 10^{-21}$ cm^2 keV was deduced.

If we take this value of $\sigma_{\rm int}$ and use our values of $A = 0.03$ cm^2 and $\phi = \sim 5 \times 10^{14}$ photons/s keV ($\sim$ the value in the 20–60 keV range as shown in Fig. 2), the decay rate (and the counting rate in gamma rays associated with the decay of the isomeric state) should be enhanced by a factor $\epsilon \sim 20\,000$. Instead, we see that the decay rate is constant to within our uncertainty of $\pm 2\%$, i.e., almost a millionfold smaller

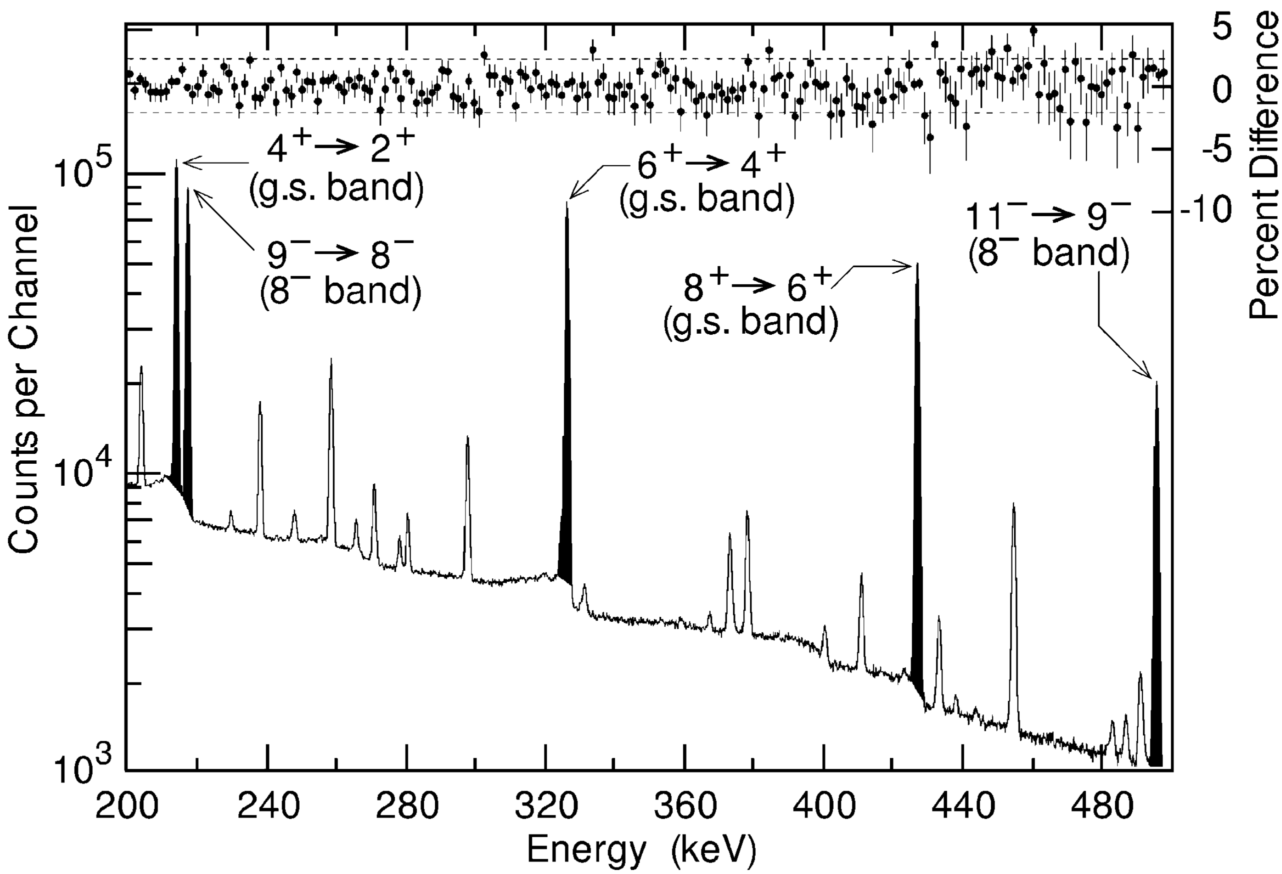

FIG. 3. The gamma-ray spectrum from 200–500 keV is shown, with the photopeaks for the transitions at 213, 217, 326, 426, and 495 keV, that were reported to be enhanced in [3,6], filled in. The spectrum is an accumulation of $\sim$22-s counting periods, immediately following the 11-s irradiations of the $R1$ target. The average undulator gap for these data was 15 mm, and the data were accumulated over 8.5 h. The channel widths are $\sim$0.15 keV/channel. The upper points with error bars show the difference spectrum between the first half and the second half of this interval. This difference, with the points summed over an energy interval corresponding to the detector resolution, should reflect any excess in deexcitation through the 4-s isomer. The dashed lines indicate $\pm 2\%$ limits in the difference.

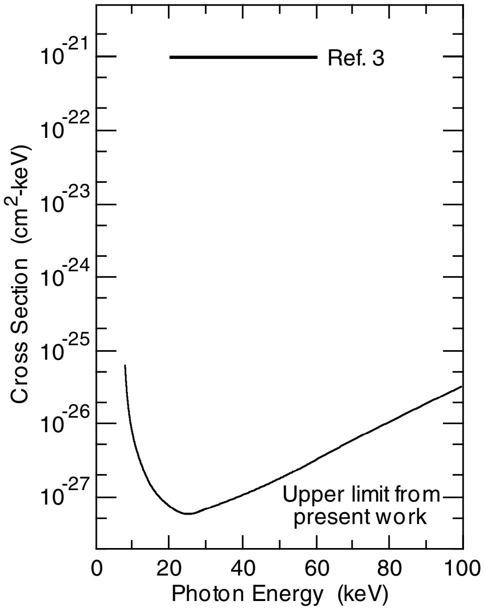

FIG. 4. Upper limit of the cross section for photon-induced deexcitation of the 31-yr ^{178}Hf isomer through the 4-s, 8^- isomer from the measurements reported here. The value for this cross section reported in Ref. [3] is also shown. Other values reported by this group in the references cited in Ref. [6] range from 2×10^{-22} to 2×10^{-21} cm^2 keV. The present limits are substantially below all previously reported values.

than that corresponding to the value of Refs. [3,6]. This implies $\sigma_{\mathrm{int}} < 2 \times 10^{-27}$ cm^2 keV, a value much more in keeping with the accumulated body of knowledge of gamma-ray transition strengths in this mass and energy region. Our limit is shown explicitly as a function of incident photon energy in Fig. 4, where the shape reflects the combined spectrum from the two undulator settings, and at the low energies is also limited by the rapid rise of absorption in the target material. At 15, 10, and 8 keV we find $\sigma_{\mathrm{int}} < 2 \times 10^{-27}$, 10^{-26}, and 5×10^{-26} cm^2 keV, respectively.

Another conceivable deexcitation scenario for the isomer is a hypothetical coupling transition that would bypass the $K = 8$, 4-s isomer, with prompt deexcitation occurring only during the irradiation period. We searched for this mode by looking for changes in the intensities of the transitions during the irradiation. For these tests we normalized the beam-on and beam-off spectra to the 1094-keV transition from the decay of ^{172}Hf, both taken with the Pb absorbers in place. Though the backgrounds were higher with the beam on, we observed no significant increase in activity for any transition in the ground-state band, or any other transition, though these limits are about an order of magnitude higher than those for the out-of-beam measurements. Using the data for target $R3$ for both undulator gap settings we obtain a limit of $\sigma_{\mathrm{prompt}} < 2 \times 10^{-26}$ cm^2 keV for this case.

In summary, we have repeated the experiment of Collins *et al.* [3,6] using three different targets and a much more intense x-ray beam. We see no evidence for an x-ray induced acceleration of the decay of the 31-yr isomer of ^{178}Hf, either delayed, through the 4-s isomer, or prompt during the irradiation. Our data are consistent with an integrated cross section of $\sigma_{\mathrm{int}} < 2 \times 10^{-27}$ cm^2 keV for decays that would go through the 4-s isomer and a value of $\sigma_{\mathrm{prompt}} < 2 \times 10^{-26}$ cm^2 keV for decays that would populate the ground-state band directly. These upper limits are somewhat higher than the highest observed values for $E1/M1$ photoabsorption reactions in this mass region, which might lead to observable effects with cross sections on the order of 10^{-28} cm^2 keV [5]. A more complete data analysis is under way and a full article will be published.

This work was supported in part by the U.S. Department of Energy (DoE) under Contract No. W-31-109-ENG-38 (ANL), in part by the DoE under Contract No. W-7405-ENG-48 (UC-LLNL), and No. W-7405-ENG-36 (LANL), and in part by the DoE Nuclear Energy Research Initiative (NERI). We are indebted to J. Greene, T. L. Khoo, C. J. Lister, R. Nelson, P. D. Palmer, W. Patterson, and K. Teh for their help in various aspects of these measurements, and to D. R. Haeffner, D. M. Mills, and G. Shenoy for their encouragement of this investigation at the SRI-CAT 1-ID beam line.

[1] J. Van Klinken *et al.,* Nucl. Phys. **A339**, 189 (1980); S. M. Mullins *et al.,* Phys. Lett. B **393**, 297 (1997); *Table of Isotopes,* edited by R. B. Firestone and V. Shirley (Wiley, New York, 1996), 8th ed.; P. Walker and G. Dracoulis, Nature (London) **399**, 35 (1999).

[2] See, for example, S. Olariu and A. Olariu, Phys. Rev. C **58**, 333 (1998); C. B. Collins and J. J. Carroll, Hyperfine Interact. **107**, 3 (1997).

[3] 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, Phys. Rev. Lett. **82**, 695 (1999).

[4] See, for example, Science **283**, 769 (1999).

[5] S. Olariu and A. Olariu, Phys. Rev. Lett. **84**, 2541 (2000); D. P. McNabb *et al.,* Phys. Rev. Lett. **84**, 2542 (2000); P. von Neumann-Cosell and A. Richter, Phys. Rev. Lett. **84**, 2543 (2000).

[6] C. B. Collins *et al.,* Laser Phys. **9**, 8 (1999); C. B. Collins *et al.,* Phys. Rev. C **61**, 054305 (2000); C. B. Collins *et al.,* Phys. At. Nucl. **63**, 2067 (2000).

[7] B. Lai *et al.,* Argonne National Laboratory Report No. ANL/APS/TB-3, 1993; R. J. Dejus *et al.,* Argonne National Laboratory Report No. ANL/APS/TB-3, 1993; Argonne National Laboratory Report No. ANL/APS/TB-17, 1994.