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Search for x-ray induced decay of the 31-yr isomer of ^{178}Hf at low x-ray energies

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Upper limits for the energy-integrated cross section for the decay of the 31-yr ^{178}Hf isomer, induced by x-ray irradiation, are extended to lower x-ray energies than we reported previously. Over the range of incident x-ray energies $6 < E_x$ (keV)<20, we report the energy-integrated cross section as less than 1×10^{-26} cm^2 keV, with less stringent limits set down to 4 keV. These limits are at least 3 orders of magnitude below values for which enhancements to the isomer decay rate have been reported by other workers.

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The nucleus ^{178}Hf has a long-lived isomeric state ($t_{1/2} = 31$ yr; J^π, $K = 16^+, 16$ at an excitation of 2.446 MeV). This isomer has been the object of several studies for possible mechanisms that might trigger its decay. The potential to control nuclear energies (MeV) with atomic energies (keV) is the driving interest. Isomeric ^{178}Hf is a favorite nucleus to study because it is long lived, available in research quantities, has a well known decay scheme, high excitation energy, and targets of the isomeric Hf can be fabricated. Collins *et al.* [1] reported the accelerated emission of γ rays from this isomer when it was irradiated with photons produced by a dental x-ray machine. The "triggering" x-ray energy was at first claimed to be within the 20–60 keV regime. Further publications by this group [2] suggest that the phenomenon may be induced by lower x-ray energies ($E_x < 20$ keV). In contrast, using synchrotron radiation at the advanced photon source (APS) we reported [3] limits on such accelerated emission some 5 orders of magnitude lower than those of Ref. [1]. Very recently, a new measurement using monochromatic x rays from the SPring-8 synchrotron light source has been reported by Collins *et al.* [4]. They report enhancement in the decay of the Hf isomer for x-ray energies in the region between 9 and 13 keV. The purpose of the present measurement is to extend our previous upper limits to lower x-ray energies.

The present experiment was similar to that of Ref. [3], modified to increase the sensitivity for low-energy x rays. The primary change was replacement of the mixed Al and HfO_2 powder target of our previous work with thin electroplated Hf. This change drastically reduces the absorption in the x-ray flux incident on the Hf and thus also reduces the uncertainties associated with the large corrections for absorption in the mixed Al and HfO_2 powder at the lowest incident x-ray energies. The experiment was arranged as is shown in Fig. 1. The "white" beam from a tapered undulator [5] at the SRI-CAT 1-ID beamline of the APS at Argonne National Laboratory was used. The photons were collimated to a beam 1.4×2 mm^2, and the target was placed at 45° with respect to the incident beam. The beam entered the target chamber (which was flushed with helium and maintained at slightly above atmospheric pressure) through a 0.25 mm thick Be window.

The target assembly was composed of two 0.5 mm thick, 2.54 cm diameter Be disks, each with a 2 mm diameter HfO_2 deposit at the center of one side and clamped together with the HfO_2 deposits facing each other. The HfO_2 target deposits were prepared by electroplating techniques, similar to those described in Ref. [6]. The Hf target material was obtained from a LAMPF beam stop, purified chemically, and then electroplated onto the Be disks, which had been coated with a (~1000-Å thick) evaporated Ti layer, and the plating surface was masked to a 2 mm diameter area. One plating cycle produced a layer of HfO_2 nominally 100 μg/cm^2 thick, with the Hf rapidly converting to HfO_2. Cycles were repeated to buildup target thickness. The total amount of isomeric ^{178}Hf was obtained by γ-ray counting, and the ratio of the isomer mass to the HfO_2 mass was independently determined to be 3.8×10^{-4}. The total ^{178}Hf target activity was measured to be 0.35 μCi, and the total thickness of the two HfO_2 deposits in the clamped assembly was deduced to be ≈ 0.5 mg/cm^2. (The target also contained ^{172}Hf activity of comparable intensity.) The target was then mounted with

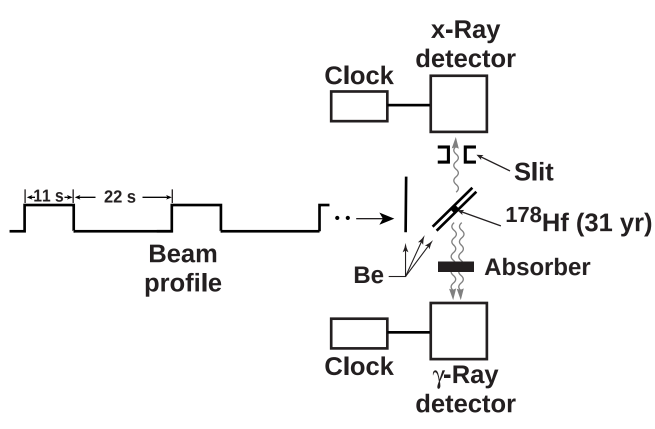

FIG. 1. Schematic of the experimental arrangement.

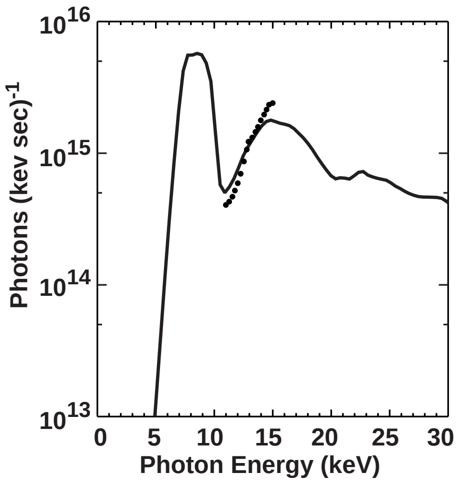

FIG. 2. Comparison of the calculated (line) and measured (dots) photon intensity from the advanced photon source incident on the HfO_2 target.

the center of rotation at the mid point of the two HfO_2 layers so that the square, 2×2 mm^2, projection of the beam profile on the target closely matched the 2 mm diameter deposit size.

A single Ge detector (50 mm diameter×15 mm thick, located at 90° and at a distance of 22.9 cm) viewed the target through a 6.3 mm thick Plexiglas window. A composite (Pb, Ta, Cd, Cu) absorber was placed in front of the Ge detector to limit the counting rate from scattered low-energy radiation. On the other side of the chamber, a Si(Li) detector was located at 90° and at a distance of 47.6 cm, viewing the target through a 0.5 mm thick Be window. A 0.05 $\times$0.05-mm^2 collimator in front of the Si(Li) detector was required to reduce the counting rate to a manageable level. The purpose of the Si(Li) detector was to monitor the fluorescent x-rays and thus the beam luminosity. The target was aligned with the beam by scanning the target through the beam, both horizontally and vertically, while monitoring the (beam-on) yield of characteristic x-rays.

Tapered gap settings were used for the 1-ID undulator to smooth out the effect of harmonics in the energy spectrum of the incident synchrotron x-ray flux: two average gap settings of 15 mm and 20 mm were used with a 5-mm taper. A 1.5-mm graphite absorber was used with the 15 mm gap setting to reduce the higher beam intensity. As a check on the calculated energy profile produced with the tapered undulators, measurements were made after the data collection with a monochromator and ionization chamber. This was done by scanning a Si(311) monochromator in energy, between 11 and 15 keV, and recording the transmitted intensity with a N_2 ionization chamber. The ionization chamber current was corrected for ion-chamber efficiency, the monochromator band width from perfect crystal diffraction theory, and for heat load and/or mounting-induced lattice strain. Figure 2 shows the calculated and measured energy distributions of the x-ray beam profile at the target position, with the data representing the weighted average of the measured energy profiles for the two gap settings, and with appropriate corrections for absorbers.

As a check of the transparency of the system to low-energy x rays, a measurement was made with a thin Cu target evaporated onto Be. Because the K x rays in Cu are low in energy, their measured yields are sensitive to the low-energy flux in the beam: the flux below 10 (20) keV is responsible for half (86%) of the K x-ray yield. The observed and calculated Cu K x-ray yields are consistent to within a factor of 2, satisfactory agreement given the uncertainties from the very tight collimation of the Si(Li) detector.

Data were collected with the radioactive HfO_2 target in the time sequence described in Ref. [3]: with the beam cycled on-target for 11 s, followed by two 11-s counting periods with the beam off target, but this time without any moving shutters in front of the detectors. The γ- and x-ray detector events were clocked with respect to the beam cycle. The total measurement time with the HfO_2 target was about 20 h for each gap setting. The number of Hf K x-rays per second, normalized to the beam current, remained constant to within $\pm$3% for all the data runs, indicating the stability of

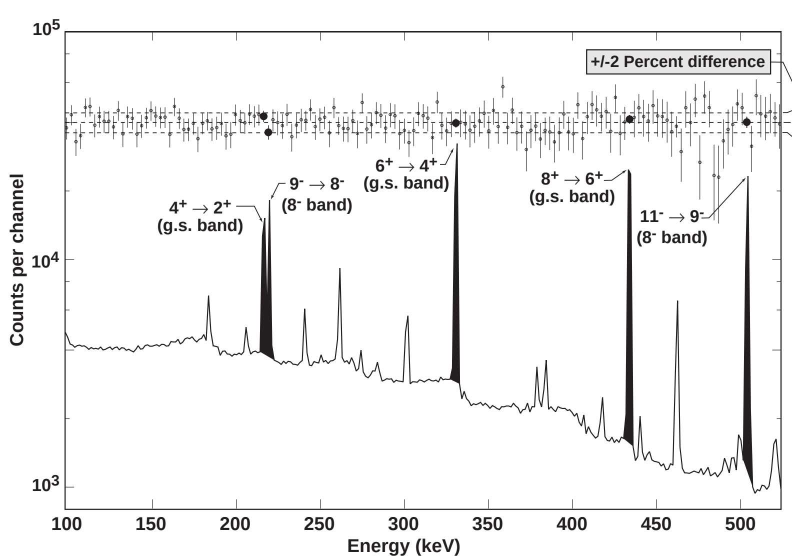

FIG. 3. The γ-ray spectrum for $100<E_\gamma$ (keV)<500. The filled photopeaks correspond to the transitions at 213, 217, 326, 426, and 495 keV reported to be enhanced [1,2]. The spectrum represents an accumulation of $\sim$22 s counting periods, immediately following the 11-s irradiations of the target. The channel widths are $\sim$0.15 keV/channel. The upper points show the percent difference between the first half and the second half of this interval on a linear scale. 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 solid points in the difference are at the location of the supposed enhanced transitions. The dashed lines indicate $\pm$2% limits in the difference.

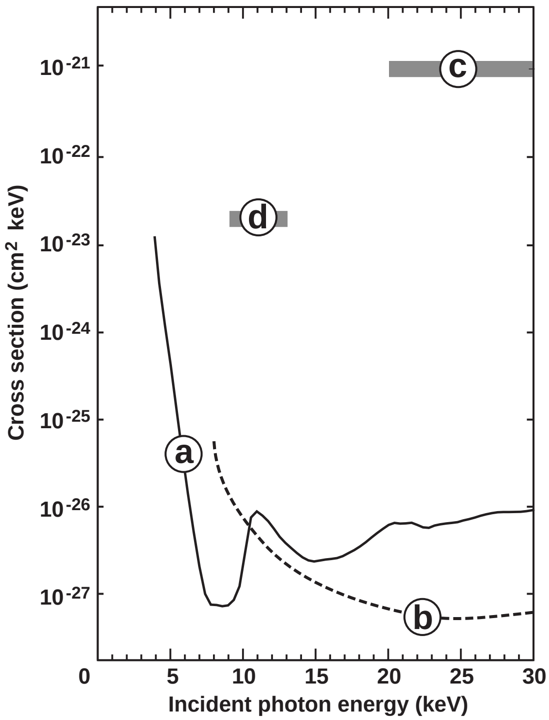

FIG. 4. Upper limit of the integrated cross section for photon-induced deexcitation of the 31-yr ^{178}Hf isomer for incident x-ray energies between 4 and 30 keV from the present measurement is shown as a solid line (a). The limit is based on a 99% confidence limit in the statistical uncertainty in the measurement with systematic errors included. The limit from Ref. [3] is shown as a dashed line (b), with the cross sections implied by the measurements of Collins *et al.* in Ref. [1] (c), and Ref. [4] (d) as cross hatched bars. [For (d), the integrated cross section was estimated from the data and discussion in Ref. [4].]

the beam on the target. Comparisons of the γ-ray intensities from the isomeric ^{178}Hf decays in these 11 s intervals (combining all the data from the two settings of the undulator gap) are illustrated in Fig. 3. They show no increase in the yield of γ rays for which such enhancement had been claimed [1,2]. We find no enhancement within the statistical errors of the measurement. Also, no evidence was found for the 129-keV γ ray reported in Ref. [7].

To obtain cross section limits we also have to estimate systematic errors in converting the results to an absolute limit. Systematic errors include uncertainties in the target thickness, the photon flux (where the uncertainties are indicated by the comparison in Fig. 2), absorbers, and detector efficiencies. The absolute cross sections are estimated to be uncertain by a factor of 2 due to such systematic errors. The cross section limits shown in Fig. 4 correspond to a 99% confidence limit from counting statistics, and then multiplied by a factor of 2 for the systematic uncertainty. For x-ray energies below $E_x = 13$ keV, the present limits are lower than those in our previous work [3] because of absorption effects. Above $E_x = 13$ keV our previous experiment set tighter limits because of the larger amount of Hf isomer exposed to the beam in that measurement. The cross section limits shown in Fig. 4 are for decay either through the $J^\pi = 8^-$ isomer, which is by far the most plausible path, or for decay that somehow bypasses the 8^- isomer.

In summary, we have established an upper limit to the energy-integrated cross section for x-ray induced decay of the 31-y ^{178}Hf isomer that is less than 1×10^{-26} cm^2 keV over the incident photon energy range $6 < E_x$ (keV) < 20. This limit, compared to our previous effort [3], is significantly lower in magnitude at the lowest incident x-ray energies and free of large corrections due to absorption in the Al+HfO_2 target mixture in that experiment. In the low-energy region, the upper limits reported here are at least three orders of magnitude below the recent values of Ref. [4], where regions of 1–3% enhancement over intervals of 50–100 eV are reported for monochromatic beams in the energy interval $9 < E_x$ (keV) < 13. Finally, combining our present and earlier [3] results, stringent limits on enhanced decay of isomeric 31-yr Hf induced by x-ray irradiation from $6 < E_x$ (keV) < 100 are well below all reported positive results. A full paper, including the details of the measurements reported here, as well as those in Ref. [3], is in preparation.

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