Accelerated Emission of Gamma Rays from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation

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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. [S0031-9007(98)08333-1]

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The 4- and 5-quasiparticle isomers of Lu, Hf, and Ta are interesting because they have relatively long lifetimes for states with 2 to 3 MeV excitation energies. They are termed K isomers because spontaneous radiative decay is hindered by structural changes forbidden by K quantum numbers. In this mass region the nuclei are deformed, and the projection of the total angular momentum upon the symmetry axis contributes this quantum number K which should change during a radiative transition by no more than the multipolarity of the mediating moment. Transitions from the high-K isomer to the rotational states of a low-K band are "forbidden," and so relatively long lifetimes are inevitable. The most interesting example may be the 31-yr, 4-quasiparticle isomer ^{178}Hf having a 2.446 MeV excitation energy.

Proposals to trigger the release of the energy of a nuclear isomer by exciting it to some higher level associated with freely radiating states have been known for over a decade [1]. To be efficient such schemes require the existence at an energy near that of the isomer of a state of mixed K. It was proposed [1] to use the resonant absorption of x rays from a bremsstrahlung source to excite some fraction of a high-K isomeric population to the K-mixing level. From there some decay to one or more heads of low-K cascades could subsequently release the total energy of the isomer plus that of the absorbed trigger photon.

The types of K-mixing states needed in such schemes to induce the decay of nuclear isomers have been reported [2] in ^{180}Ta and described in ^{174}Hf and other isomers [3]. In the case of the Ta, the resonant absorption of x rays excited the 2-quasiparticle isomer of ^{180}Ta to a K-mixing level [4] at 2.8 MeV which then spontaneously decayed in part to the ground state through a gamma cascade. The integrated cross section for the resulting deexcitation of the isomer was 1.2×10^{-25} cm^2 keV. Studies of systematics have shown [5] that a similar K-mixing level could be reasonably expected in ^{178}Hf not more than 300 keV above the 2.446 MeV, 16^+ level of the 4-quasiparticle isomer. However, quantitative

estimates of the efficacy for inducing the emission of gamma decays have diverged with the most positive [6] predicting that the value for ^{180}Ta could be scaled by the square of the trigger wavelength up to 10^{-21} cm^2 keV for a 30 keV transition in ^{178}Hf. For contrast, in 1998 a more traditional computation reported that x-ray intensities at a sample of the isomer would have to exceed 3.9×10^{12} W cm^{-2} to obtain a 1% increase in the spontaneous decay rate of the ^{178}Hf isomer [7]. Between the two predictive extremes of the published Letters [6,7] lie the possibilities for a self-sustained photon chain reaction in an isomeric sample in the case of the larger estimate and for the impossibility of detecting the effect in the latter.

Described here is the first report of induced gamma emission from the 31-yr isomer of ^{178}Hf triggered by the resonant absorption of photons from bremsstrahlung radiation. In this experiment a sealed plastic target containing 6.3×10^{14} isomeric nuclei in a 1 cm diameter well was exposed to the bremsstrahlung radiation from a dental x-ray unit operating at 15 mA. The voltage applied to the anode of the x-ray tube was a half-wave rectification of 60 Hz ac. The end point varied during a half period up to the nominal maximum of 70 or 90 kV, as selected. A better signal-to-noise (S/N) ratio resulted from use of the 90 kV. The counting circuit was enabled for only the peak 48% portion of each radiation pulse so as to maximize the S/N. The Integrated TIGER Series Monte Carlo code was used to compute the irradiating spectrum at the target for these conditions incorporating the time dependence of the voltages and currents from the x-ray generator. The computed spectrum at the isomer location is presented in Fig. 1. It has been overlaid with a measured output spectrum from the x-ray device obtained through a pin hole at approximately a 5 m separation between source and detector.

The gamma spectrum was detected with a 10% coaxial Ge detector. Data acquisition was enabled only during the x-ray pulses as detected by a *p-i-n* diode. As normally used, the resulting duty cycle for the irradiation on the target was about 0.7%. The Ge detector was placed at about a 37 cm distance in a plane perpendicular to the axis of the x-ray beam. Careful beam collimation, detector shielding, and use of a 3 mm Pb + 3 mm Cu absorber in front of the detector provided sufficiently low rates of scattered x rays. Thus, the dead time, resolution, and pileup problems were insignificant even at the time of the maximum of the x-ray pulse. Portions of typical spectra are shown in Fig. 2, together with the differences obtained by subtracting raw data taken with and without x-ray irradiation at 90 and 70 keV.

The target contained an impurity of ^{172}Hf, principally radiating the well-known decay spectrum of ^{172}Hf and its daughters at a level comparable to the intensity of the 31-yr spontaneous decay of the ^{178}Hf isomer. The decay properties and level schemes of the ^{178}Hf and $A = 172$ nuclides are comprehensively reviewed in the literature [8,9]. A total of more than 100 lines was found experimentally, and each was identified with one of these two nuclides or with a natural background. There was no evidence of any previously unexpected nuclide. Components of the ^{172}Lu decay provided a convenient fiducial quantity against which to compare measured

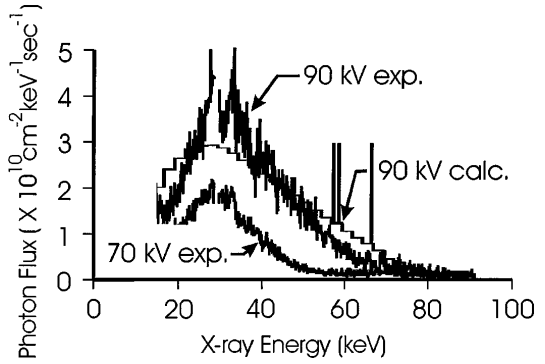

FIG. 1. Graph of the x-ray spectral flux expected in the position of the isomeric target. Rectangular steps plot calculated intensities normalized to the measured exposure. The experimental data for 70 and 90 keV end points were measured with a planar Ge detector from input attenuated with a pin hole and were normalized to the calculations.

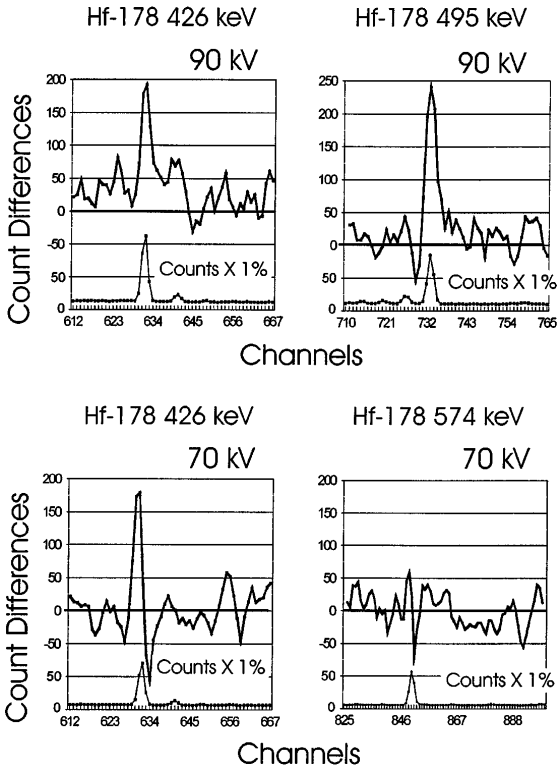

FIG. 2. Plot of the differences in counts obtained in the spectrum of the ^{178}Hf from the isomeric target when it was exposed to the x-ray beam and when it was not. A weighted smoothing procedure was applied. Also shown are the unsmoothed spectra from the unirradiated target scaled by 1% to facilitate comparison. In the upper row from left to right, the two lines correspond to the transitions $(8^+ \rightarrow 6^+)$ and $(11^- \rightarrow 9^-)$, shown by the heavy arrows in Fig. 3. The lower row from left to right presents again the $(8^+ \rightarrow 6^+)$ transition together with the $(13^- \rightarrow 11^-)$.

intensities of the emission from the isomer made with and without irradiation. The photon induced depopulation is possible for the isomeric state, but is not expected for e-capture radioactive nuclei, such as ^{172}Hf and its daughters. The relevant energy levels of the ^{178}Hf nuclide are shown in Fig. 3. The spontaneous decay of the 31-yr isomer shown by the thin arrows is entirely through the $K^\pi = 8^-$ band followed by cascade through the ground state band (GSB) beginning with the 8^+ member. Because of the Pb filter neither the $(2^+ \rightarrow 0^+)$ line at 93.2 keV nor the $(4^+ \rightarrow 2^+)$ line at 213.4 keV could be recorded with acceptable statistical accuracy. Particular attention was focused upon the $(8^+ \rightarrow 6^+)$ and $(11^- \rightarrow 9^-)$ lines at 426.4 and 495.0 keV, respectively, shown by the heavy arrows in Fig. 3 for which data are shown in Fig. 2. The 495.0 keV line showed the greatest enhancement when the target was irradiated with the bremsstrahlung. Particularly notable was the absence of enhancement of the 574.2 keV $(13^- \rightarrow 11^-)$ transition which normally feeds the 495.0 keV during the spontaneous decay of the ^{178}Hf isomer, as seen by the thin arrows in Fig. 3.

In this experiment the principal limitations were driven by the finite lifetime of the x-ray source. To acquire the in-beam spectra used for the differences in Fig. 2 required a nominal 24 hr of operation to accrue 546 sec of total exposure time. This was a sufficient duration to obtain about 6×10^4 counts in the prominent 1094 keV line of ^{172}Lu, used as a reference and corresponded to a consumption of about 10% to 15% of the projected service life of the x-ray source. A comparison spectrum was acquired without x rays by counting the target in the same physical place to get the same total counts in the fiducial line of the ^{172}Lu. Then it was normalized so that no Hf line in the in-beam spectrum would show statistically significant absorption and no Lu line would suggest gain because of an inequality in the durations for counting. The excess counts acquired when the target was irradiated are shown in Table I for prominent lines of the spectra, together with the standard deviations of the differences. In this particular case normalization of $-0.75\% \pm 0.38\%$ was required to meet the criterion for the last four lines tabulated. While statistical errors reported there were actually obtained from a peak fitting program, they can be understood in the case of major lines to accrue from a component of about 1% arising from the statistics of the total counts in the lines and the balance from the total counts in the areas needed for definition of the baselines. The latter factor was adversely affected by any proximity of Lu lines, such as in the case of the 495.0 keV line shown in Fig. 2. The further effect of the 0.35% error independently contributed by the normalization was generally neglected. For completeness, results are included for the $(10^- \rightarrow 8^-)$ line at 454.1 keV, apparently fed as part of the cascade from 11^-. Although not statistically significant, the apparent enhancement does not contradict a simple model that from 11^- the pattern of spontaneous decay is followed. The tabulated values indicate an enhancement of $6.3\% \pm 2.2\%$ for the 495.0 keV line and $2.0\% \pm 1.3\%$ for the 426.4 keV line.

During the spontaneous decay of the ^{178}Hf isomer the latter, being a member of the GSB, is fed only by cascade from the 4 sec, 8^- bandhead. Therefore, a counting system as used here with a 2 sec gate coincident with the irradiation should detect in the GSB intensities only about 30% of the additional intensity supplied to the 495.0 keV transition by the x-ray induced process. The relative enhancements shown in Table I agree with this ratio of about 30% and indicate that the induced decay observed in this experiment feeds into the GSB primarily through the 8^- level as in the spontaneous decay. In contrast,

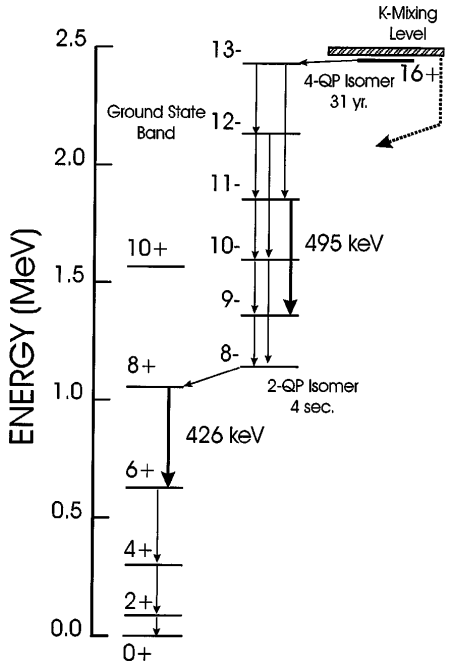

FIG. 3. Energy level diagram for ^{178}Hf showing a selection of levels relevant to this experiment. The spontaneous decay of the 31-yr, 16^+ isomer is shown by the thin arrows. The thick solid arrows show components studied in this work from the particular cascade from the decay forced by the x-ray irradiation. Heavy dotted arrows are inferred transitions needed to feed those shown.

TABLE I. Comparison between counts collected from gamma emission from the Hf target with and without x-ray irradiation by bremsstrahlung with an end point of 90 keV.

Line (keV)	Spontaneous (Counts)	Induced Enhancement (Counts)	Statistical ± (Counts)
Hf-426.4	21302	431	269
Hf-454.1	3326	384	358
Hf-495.0	15103	950	338
Hf-574.2	18681	−337	276
Lu-810.1	19481	−118	308
Lu-900.8	32274	−8	260
Lu-1093.6	59370	493	581

the scattering of statistically insignificant positive and negative differences tabulated for the lines of ^{172}Lu provide an illustration of comparable data for lines for which enhancements are not induced. It is interesting that the data for the 574.2 keV line of ^{178}Hf show the same null result as for the Lu lines. Unlike spontaneous decay of the isomer in which the 495.0 keV line is fed from the 574.2 keV line at a somewhat greater intensity, no detectable enhancement is shown in Table I for the 574.2 keV line, even when the 495.0 keV line is increased by 6%. Results were reproducible to within uncertainties indicated by Table I but generally favored values lower by 1σ. In conclusion we report enhancement caused by irradiation at 90 keV to be about $4\% \pm 2\%$ of the intensity of the 495.0 keV line emitted in the spontaneous decay of the isomer.

While the comparisons shown in Fig. 2 for 70 keV irradiation continue to indicate triggering at a lower voltage as well, the statistical significance was not sufficient to justify tabulation. Evidence was also found for the excitation of new lines feeding the 495.0 keV transition when induced, but statistics were inadequate to insure the reproducibility needed for definitive analysis.

In a simple model the yield of triggering events would equal the product of the number of isomeric atoms in a target, the spectral flux density from Fig. 1 at the appropriate energy, and the unknown integrated cross section for the branch of the excitation of a K-mixing level that ends in a state other than that of the initial isomer. Since each quantity is known except for the integrated cross section for the "triggering branch," that cross section can be obtained if the transition energy is estimated. As is clear from Fig. 1, the energy for the highest intensity of the incident photons is found below 40 keV, and this value was assumed for the cross section estimate to give an order of magnitude. At 40 keV an integrated cross section, $\sigma\Gamma$ was obtained,

$$\sigma\Gamma = 1 \times 10^{-21}\ \mathrm{cm}^2\,\mathrm{keV}, \qquad (1)$$

with an uncertainty of at least 25%. This estimate can be seen to be quite consistent with that reported [4] for ^{180}Ta, taking into account the lower triggering energy appearing in the Breit-Wigner cross section for the absorption in the present case with respect to the ^{180}Ta case. Bounds on the transition energy, E_k to the K-mixing level can be obtained from the low energy cutoff seen in the actual x-ray spectra of 20 keV and the substantial loss of intensity above 60 keV when the end point was changed from 90 to 70 keV. We report

$$E_k = 40 \pm 20\ \mathrm{keV}. \qquad (2)$$

For the case of isomeric ^{178}Hf we have demonstrated that the resonant absorption of an x-ray photon with the energy of the order of 40 keV can induce the prompt release of the 2.446 MeV stored by the isomer into freely radiating states. This is an energy gain of about 60. Only 9 $\mathrm{mW\,cm^{-2}}$ of total x-ray power in the bremsstrahlung were required at the target. Further research is needed to provide greater precision to the measurements of the transition energy to the K-mixing level needed to trigger the release of the stored isomeric energy. Such data will then facilitate a better understanding of these first evidences of the triggering of induced gamma emission from the 31-yr isomer of ^{178}Hf with very low energy x-ray photons through such large cross sections.

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