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Tunable synchrotron radiation used to induce γ-emission from the 31 year isomer of ^{178}Hf

C. B. Collins[1], N. C. Zoita[1], A. C. Rusu[1], M. C. Iosif[1], D. T. Camase[1], F. Davanloo[1], S. Emura[2], T. Uruga[3], R. Dussart[4], J. M. Pouvesle[4], C. A. Ur[5,6], I. I. Popescu[6], V. I. Kirischuk[7], N. V. Strilchuk[7] and F. J. Agee[8]

[1] *Center for Quantum Electronics, University of Texas at Dallas Richardson, TX 75083-0688, USA*
[2] *The Institute of Scientific and Industrial Research, University of Osaka Osaka 567-0047, Japan*
[3] *SPring-8 / JASRI - Sayo-gun, Hyogo 679-5198, Japan*
[4] *GREMI, CNRS, Université d'Orléans - Orléans, France*
[5] *H. Hulubei National Institute of Physics and Nuclear Engineering Bucharest, Romania*
[6] *IGE Foundation - Bucharest, Romania*
[7] *Scientific Center "Institute for Nuclear Research" - Kiev, Ukraine*
[8] *Air Force Office of Scientific Research, AFOSR/NE 801 Randolph St., Arlington, VA 22203-1977, USA*

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Abstract. – A process for transferring energy from electron shells into nuclear excitation, NEET, has offered the promise for modulating nuclear properties at accessible levels of power. It had been proven recently by exciting a nuclear level of ^{197}Au with synchrotron radiation, but measured couplings were far below theoretical objectives. Reported here is an extension of that approach for excitation to ^{178}Hf^{m2} isomeric nuclei. Isomeric targets were irradiated with X-rays in the beamline BL01B1 at the synchrotron radiation source SPring-8. Energies were tuned from 9 to 13 keV. In this range an excitation branch attributed to NEET was found to have a probability of 2×10^{-3} relative to L-shell photoionization. The resulting emission of exoergic γ-photons was observed from the target at a rate approaching the theoretical maximum.

In 1993, Ho *et al.* [1] described how higher-order processes for non-resonant excitation (NEET) could couple energy into nuclei from the surrounding electronic shells. "Satellites" on absorption transitions between nuclear levels were predicted. Kishimoto *et al.* [2] have recently proven such NEET phenomenology by exciting the ground state of ^{197}Au with synchrotron radiation (SR). They observed the excitation and decay of the $(1/2^+ \rightarrow 3/2^+)$ (Mössbauer) transition of ^{197}Au at 77.351 keV by resonant absorption of SR at 80.989 keV. Although the absorption satellite for exciting the nuclear transition was not explored by tuning the SR

energy to other "off-resonant" values, it was reasonably concluded that excitation through K-shell photoionization was responsible with a branching probability for NEET of $5.0 \pm 0.6 \times 10^{-8}$.

As a vehicle for the further demonstration of NEET spectroscopy, the 16^+ state of $^{178}\mathrm{Hf}^{m2}$ is particularly attractive. It is a 4-quasiparticle state having 2.446 MeV of excitation and a half-life of 31 years. If NEET were excited by SR there would be the possibility for exoergic emission of γ photons with energies exceeding those of the irradiation. Then, the experimental arrangement could be facilitated by the use of layered absorbers to selectively remove X-rays scattered from the SR source. Reports of the use of X-rays with energies below 20 keV to accelerate the decay of $^{178}\mathrm{Hf}^{m2}$ spin isomers [3, 4] further encouraged planning for the use of that isomer in a NEET experiment.

Reported here are the first successful examples of the irradiation of samples of $^{178}\mathrm{Hf}^{m2}$ isomeric nuclei with synchrotron radiation. Moreover, in this work the SR was tuned over the range of energies 9–13 keV, selected because it contains much of the transition strength for photoionization of the L-shell electrons of Hf. A branching ratio of 2×10^{-3} was found for the excitation of nuclear transitions from absorption of SR by the L-shell electrons. Those events led to the emission of exoergic γ-photons.

Two targets were used in these experiments. Both were fabricated from material in which the isomeric fraction was about 10^{-3} of the total Hf content. One was composed of two separately encapsulated samples of 5.8×10^{12} $^{178}\mathrm{Hf}^{m2}$ isomeric nuclei each. The other target contained three sealed samples of 1.3×10^{13} each. They were irradiated with the tunable output from a bending magnet in the BL01B1 beamline at the synchrotron radiation source SPring-8 in two separate experimental series separated by weeks. Passed through a monochromator the output beam reaching the target was 2 mm $\times$ 5 mm in size in the first survey experiment. The flux was $3.0 \pm 0.6 \times 10^{10}$ photons cm^{-2} s^{-1} with a nominal spectral bandwidth of 0.5 eV. It was tuned from 9.0 to 13.0 keV in steps of about 5 eV, remaining 10 s at each energy. The basic scan was repeated four times. Of particular importance was that the first target was optically thin to SR between 9 and 13 keV when mounted at 45°. The target constructed for the second experimental series was designed to give higher signal rates. It was mounted in grazing incidence inclined 12° with respect to the line of the SR beam which was adjusted to have cross-sectional dimensions of 1 mm $\times$ 5 mm. Tuning range intervals found to be interesting in the first experiment were re-examined with 0.5 eV steps in SR energy. Data were collected for 50 s at each X-ray energy and each scan was repeated 3 or 4 times, depending upon limitations imposed by the periodic reinjection of the synchrotron current.

The performance of the target is of critical importance in such experiments. Spontaneous decay of the population of isomers produces energetic γ's able to escape from millimeter thicknesses of target structure and mounting. However, if not optically thin to the SR at the X-ray energies used, the spontaneous emission from the entire target can readily "drown" any component of NEET contributed by the outer micron thickness of sample exposed to the SR. Several attempts at demonstrating NEET with isomeric targets have recently failed for lack of optical transparency at the resonant energies for NEET [5]. In the work reported here particular attention was given to the properties of the target structure. In the second series of experiments the SR intensities were continuously measured in-line with ionization chambers placed before and after the isomeric target. Figure 1 shows the SR flux (corrected for attenuation caused by passage through the first ionization chamber) that was incident upon the target assembly for each of the several intervals of SR energy examined. Also shown in fig. 1 is the resulting attenuation coefficient of the target obtained from both ionization chambers. Despite the considerable changes in SR flux set during the experiment, agreement can be seen between the absorption coefficients extracted for the target for the different conditions, and with the attenuation calculated for the geometry and measured density of the

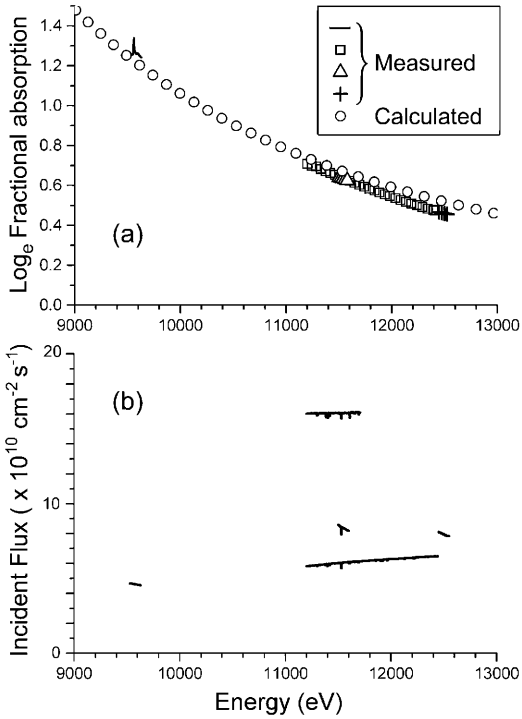

Fig. 1 – Irradiation environment. (a) Absorption coefficient of the target measured in-line as a function of SR energy during the different irradiations. (b) Measured values of SR flux.

plastic encapsulations of the samples stacked into the target. No fitted parameters were used in preparing fig. 1.

A Si drift detector (SDD) was used both to confirm the location of the target activity in the SR beam and to survey the level of scattered SR in the detector environment. Figure 2 shows the level of elastic scattering to be sufficiently low to facilitate gating upon photons selected to record the intensity of L_α fluorescence from the Hf component of the target. Figure 3

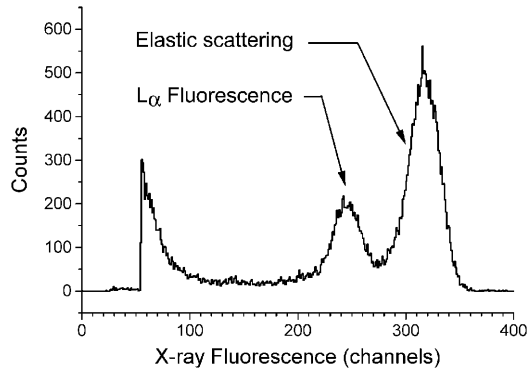

Fig. 2 – Spectrum of the low-energy X-rays scattered by the target from the SR beam.

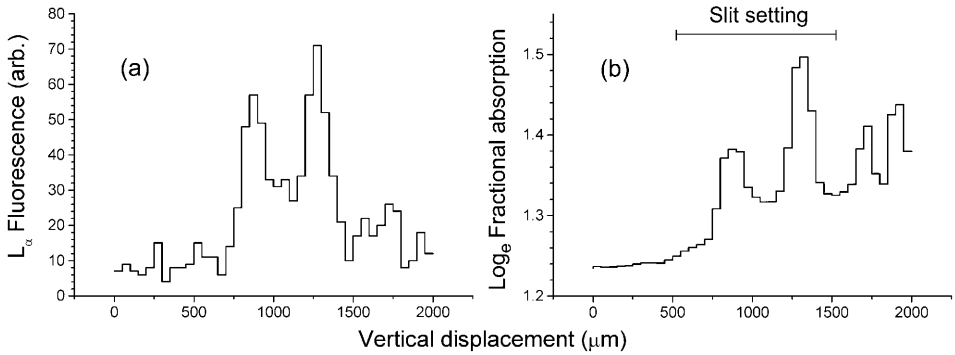

Fig. 3 – Target alignment. (a) Hf(L_α) fluorescence from the target as a function of vertical displacement. (b) Absorption coefficient of the target measured in-line with the SR beam narrowed to 0.1 mm vertical height.

shows the technique used to locate the activity of the target in the SR beam. The data were obtained by moving the target vertically with a stepping motor while the absorption coefficient was measured with the ionization chambers and the intensity of L_α fluorescence from the Hf component was monitored by the SDD. The slit limiting the vertical dimension of the SR beam was narrowed to 0.1 mm during this adjustment. After the activity was located, the slit was set as shown in fig 3. The process was repeated in the horizontal direction and showed the activity to extend 4.6 mm in that direction. The activity had the form of an elliptical annulus in each sample and the samples had been assembled into the target slightly out of register to prevent the Hf components from shadowing each other and that accounts for the morphology seen in fig. 3.

A Ge spectrometer 10 mm thick × 16 mm dia. was placed 25 mm from the target at right angles to the beam and was shielded with layered foils of W, Cd, and Cu, as seen by the beam. It was connected to an EG&G 673 spectroscopy amplifier from which the output was continuously digitized and recorded to permit subsequent re-examination of the electrical pulses describing the detected photons. The spectrum of the target has been described previously [3, 4] and was composed of only the well-known decays [6] of $^{178}\mathrm{Hf}^{m2}$ and of the ^{172}Hf impurity and its daughters. Of the former, only the $(4, 2)$, $(6, 4)$, and $(8, 6)$ members of the ground state band, GSB at 213.4, 325.6, and 426.4 keV, respectively, and the lowest member of the 8^- band at 216.7 keV were recorded at significant counting rates because of declining efficiency of the Ge-detector for higher energy γ's. Of the lines from the impurities, only two, 125.8 and 181.5 keV, could be recorded with significance. Data were analyzed with a mesh of 8.8 γ channels per keV. When the monochromator changed energies, a digital signal was sent to a second channel of the data logging system to correlate the signals from γ-photons with the energies of the irradiation. Regions of interest (ROIs) associated with each of the spectral features were identified in the 4096 channel histograms of the numbers of γ-photons detected during each period of irradiation with SR. For simplicity, in most cases the sums for the ROIs for 213.4 and 216.7 keV were used to study the excitation functions of the $^{178}\mathrm{Hf}^{m2}$ sample. The ROI for the 181.5 keV impurity line provided a convenient indication for the null effect.

The most pronounced evidence for NEET is shown in fig. 4. Raw data are shown in the form of counts collected in ROI(216.7) during 60 s periods of irradiation. The energy of the SR X-rays was incremented in 3 steps of 0.5 eV each during those periods. The intervals over which total counts were collected for entry into table I are indicated. The corresponding

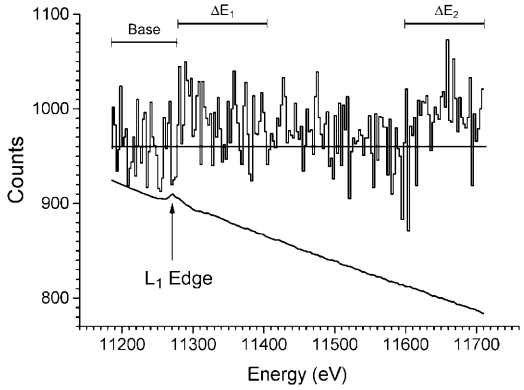

Fig. 4

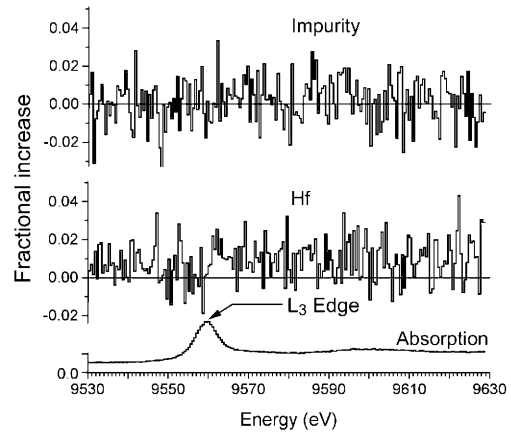

Fig. 5

Fig. 4 – Photon counts collected in the ROI(217), together with the fractional absorption showing the Hf(L_1) edge. Intervals of SR energy used in analysis are shown.

Fig. 5 – Photon counts collected in the ROI(181+126) for the impurities and ROI(213+217) for Hf, together with the fractional absorption showing the Hf(L_3) edge.

feature at the L_3 edge for photoionization was examined in 0.5 eV steps with 50 s duration repeated 4 times. Figure 5 shows the results in terms of fractional increases over the average counting rates observed at the lower X-ray energies just below the L_3 edge. Numerical values of the counts are reported in table I.

From the data of table I the cross-sections, σ_c, for excitation can be readily expressed as $\sigma_c = fA/F$, where f is the fractional enhancement over spontaneous emission, A is the rate coefficient for spontaneous emission ($A = 7.09 \times 10^{-10}$ s^{-1}), and F is the irradiating photon flux. Values of flux were taken from fig. 1(b) and were corrected for the absorption from the encapsulation. For the 3-sample target at 12° inclination, the effective average fluxes over the

Table I – *Comparison of the gamma-emission from isomeric* Hf *nuclei and from impurities in the target. Data are presented as counts collected when irradiated with X-rays having energies in the structure studied in comparison with results when irradiated with "Baseline" energies proximate to those in the structure.*

Feature	ROI	In structure	Adjacent baseline		Excess
		Counts	Counts	Norm.([a]) Counts	(counts)
Fig. 4-ΔE_1	ROI(181)	66175 (257)	50681 (225)	66609 (296)	−434 (392)
	ROI(217)	45476 (213)	33597 (183)	44156 (241)	1320 (322)([b])
Fig. 4-ΔE_2	ROI(181)	55737 (236)	50681 (225)	56473 (251)	−736 (344)
	ROI(217)	38662 (197)	33597 (183)	37437 (204)	1225 (284)([c])
Fig. 5	Area(126+181)	276333 (526)	112399 (374)	274753 (914)	1580 (1054)
	ROI(213+217)	433703 (659)	175385 (419)	428719 (1023)	4984 (1217)([d])

([a]) Multiplied by ratio of durations for counting.
([b]) Estimation of the effect: (3.0 ± 0.73)%.
([c]) Estimation of the effect: (3.3 ± 0.76)%.
([d]) Estimation of the effect: (1.2 ± 0.28)%.

TABLE II – *Integrated cross-sections obtained for the three structures for excitation of the* $^{178}\mathrm{Hf}^{m2}$ *isomeric population by* NEET.

Feature	Eff. photon flux ($\mathrm{cm}^{-2}\mathrm{s}^{-1}$)	Enhancement f	Cross-section (cm^2)
Fig. 4-ΔE_1	1.2×10^{11}	0.030 (0.0073)	1.77 (0.43) $\times 10^{-22}$
Fig. 4-ΔE_2	1.2×10^{11}	0.033 (0.0076)	1.95 (0.45) $\times 10^{-22}$
Fig. 5	2.7×10^{10}	0.012 (0.0028)	3.15 (0.74) $\times 10^{-22}$

activity for the SR energies shown in fig. 4 and fig. 5, respectively, were reduced from fig. 1 with multiplication by 0.72 and 0.58. Resulting fluxes and cross-sections are summarized in table II. As can be seen, the confidence factors exceed 4σ for all the 3 measurements reported in this work.

The photoionization cross-section at the L_1 edge is 7.5×10^{-20} cm^2 [7], so these excitation bands represent about 2×10^{-3} of the photoionization probability. Though large, such a value still conforms to the upper limits for NEET of a few $\times 10^{-3}$ calculated [1] for neighboring nuclei. The work reported here appears to extend the first proof of the NEET process for nuclear excitation [2] to the isomeric nucleus $^{178}\mathrm{Hf}^{m2}$. Particularly notable is that in this case of isomer excitation, probabilities approach the theoretical maxima of a few $\times 10^{-3}$ for L-shell photoionization.

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