



## Accelerated $\gamma$ -emission from isomeric nuclei

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### Abstract

A nuclear-XAFS effect occurs during the scattering of electrons or X-ray photons when the excitation of inner shell electrons couples both energy and angular momentum into nuclear channels of excitation. Nuclear transitions can be induced if the density of excited states of the nucleus enclosed by the electrons is high enough. The 31-yr isomeric state,  $^{178m2}\text{Hf}$  satisfies the condition for excitation by monochromatic X-rays from synchrotron sources. Strong effects upon such nuclei are studied.

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### 1. Introduction

The motions of charges<sup>1</sup> within the nucleus are quantized and so are able to store electromagnetic excitation by promoting one or more of the constituent nucleons up the resulting ladders of excited states. Interactions leading to spontaneous emission of the excess energy are strong and lifetimes of excited states are short. In nuclei, the spins of the component particles are usually "paired" in antiparallel combinations. A spin isomer results when such a pair is "broken" and rearranged so that the two individual spins are parallel. Spontaneous emission may then be inhibited by selection rules limiting changes in total angular momentum and its projections and lifetimes against spontaneous decay reach values of years to millions of years.

Since excited states of nuclei can decay by emitting  $\gamma$ -photons during transitions to lower energy states, they must also be able to absorb resonantly X-ray photons of electromagnetic radiation, as has been known for over 60 years (Pontecorvo and Lazard, 1939). Although driven by potential applications ranging from high resolution lithography,  $\gamma$ -ray holography, and even the possibility of a  $\gamma$ -ray laser, developments were inhibited by the very short lifetimes of the nuclear excited states. Experimental difficulties were severe when attempting to separate a result, usually fluorescence, from its cause, which was often some accelerator product, the scattering of which tended to mask all desired effects. Because of the more convenient time scales, the involvement of nuclear spin isomers in such research and development appeared attractive.

The use of X-rays to excite a nuclear isomer was first reported in 1988 (Collins et al., 1988). Using the Bremsstrahlung from a target irradiated by a medical LINAC, the spontaneous decay of the isomer  $^{180m}\text{Ta}$  was accelerated. This is the only naturally occurring

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<sup>1</sup>Neutrons act as negative "holes" in the average positive fluid of the nucleus.

isomeric nucleus; 0.01% of all natural Ta is found in this excited state remaining from the original cosmic nucleosynthesis of the elements. It stores 75.3 keV per nucleus. The X-rays at 2.8 MeV were resonantly absorbed causing a transition to a highly excited state which then decayed back to the ground state of  $^{180}\text{Ta}$  that was unstable against particle processes of further decay to  $^{180}\text{Hf}$  or  $^{180}\text{W}$ . Evidence of the success of the manipulation of the lifetime of the isomeric  $^{180\text{m}}\text{Ta}$  was found in a residue of  $^{180}\text{Hf}$  produced by the X-ray irradiation. The actual electromagnetic transitions involved were masked by the intensities of the scattered X-rays used to irradiate the sample. However, the fact that products of the decay of the ground states were detected as a result of accelerating the decay of isomeric  $^{180}\text{Ta}$  nuclei meant that the total energy emitted per event exceeded the energy imparted by the incident photon by 75.3 keV, the energy originally stored in each isomeric nucleus. While not a practical increase in energy for any applications, acceleration of the decay of the isomeric  $^{180}\text{Ta}$  nuclei did prove the hypothesis that exoergic reactions of isomeric nuclei could be stimulated by incident X-rays. Motivation was created for a search for exoergic systems that resulted in much greater energy “gain,” when triggered by incident X-rays of more conveniently lower energies.

At energies below 100 keV, X-rays incident upon a solid medium do not simply penetrate in order to irradiate the nuclei in the sample. Photoionization of the surrounding electrons is the dominant effect and offers the opportunity to “leverage” the impact of incident radiation upon the nuclei to much greater magnitudes. A process for transferring energy from inner electron shell vacancies into nuclear excitation, was first proposed in 1973 (Morita, 1973). Called NEET (Nuclear Excitation by Electron Transition), it was developed further (Ho et al., 1993) and generalized to a broader class of electron bridging mechanisms, EBM (Matinyan, 1998). The cross sections for producing inner shell vacancies by incident X-rays are much larger than for directly exciting nuclear transitions and the various transfer mechanisms suggested that nuclear transitions might be excited with cross sections approaching 0.1–0.5% of the values characteristic of photoionization. Those proposals offered an exciting promise for modulating nuclear properties with X-ray photons at accessible levels of power, and even for facilitating the triggering of the decay of nuclear isomers.

Photoionization is a multi-order process in which the quantum interference between path options creates a rich phenomenology studied in the field of XAFS (Campbell and Gilfrich, 1970; Sayers and Bunker, 1988; Emura et al., 1993). An inner shell vacancy is not a quantum stationary state, but rather a quantum intermediate state in a scattering process. It arises from the summation of possibilities for inelastic scattering of an

incident photon or electron that has arrived by the several different (but indistinguishable) geometric paths that include elastic and inelastic scattering events with neighboring atoms. In short, the transient intermediate state vacancy retains “memory” of how it could have been produced and those possibilities sum to give constructive and destructive interferences. So, nuclear excitation caused by the incidence of photons or electrons at X-ray energies might reasonably be expected as another type of XAFS phenomena at levels of 0.1%. A first test would be to look for structured excitation functions for such Nuclear-XAFS.

## 2. Experimental results

As a vehicle for a test of nuclear-XAFS, the isomeric nuclei  $^{178\text{m}2}\text{Hf}$  is uniquely ideal. It stores 2.445 MeV per isomeric state for a half-life of 31 years. As a material this represents 1.3 GJ/g and so encourages applications. Small samples exist for experimentation that present no significant hazard to personnel because the slow rate of spontaneous decay results in activities below 10 kBq for practical arrangements. A plan to leverage the effect of X-ray irradiation upon nuclear states by using L-shell electrons would involve irradiation of a sample with X-rays having energies around 10 keV. Characteristic  $\gamma$ -rays emitted by excited states of  $^{178}\text{Hf}$  nuclei have energies in the range of 100's of keV so that the scattered X-rays that historically had masked the effects they caused could be simply removed by thin foils designed to absorb the lower energy X-ray photons.

First experiments were done with a small Bremsstrahlung generator familiar from dental examinations (Collins et al., 1999, 2000). Acceleration of the decay of the  $^{178\text{m}2}\text{Hf}$  isomer by about 2% was seen in the enhanced rate of emission of the  $\gamma$ -photons from some of the transitions shown in Fig. 1. With high-pass absorption filters, it was shown that it was the part of the X-ray continuum with energy below about 20 keV that accelerated (triggered) the decay (Collins et al., 2000). While only the bands of excited states involved in spontaneous decay are shown in Fig. 1, there are 17 other known bands which are not involved in spontaneous decay because of selection rules. Also shown in Fig. 1 is a K-mixing level (Collins et al., 2001a) which has been assumed to have energy close to that of the isomeric 16+ state and mixed values of the pure quantum states of the projections of angular momenta upon which the selection rules act to inhibit spontaneous decay. The usual path of decay is to cascade down the 8-band until the 8-state at the bandhead is reached. It has a 4 s half-life that introduces a statistical time lag in further cascading into the ground state band (GSB). In the pulsed Bremsstrahlung experiments with coincidence detection using 4 Ge detectors, we showed that the

accelerated decay benefited from a “short circuit” around the 8-level because the induced  $\gamma$ -photons from the GSB were detected less than 1 ms after the

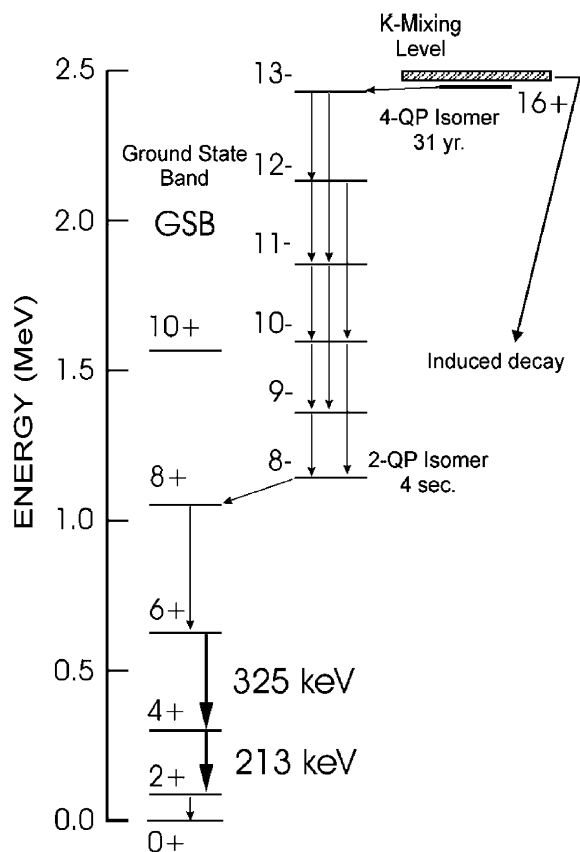


Fig. 1. Energy level diagram illustrating the spontaneous and induced decay of the 31-yr isomer of  $^{178m2}\text{Hf}$ . Shown by heavy arrows are two transitions in the ground state band, GSB that are particularly useful for detecting enhanced emission rates induced by X-rays.

application of the pulse of irradiation (Collins et al., 2001a). Evidently the involvement of inner shell electrons in the nuclear transitions relaxed the constraints imposed by the selection rules by making available for change the additional angular momentum of those electrons. It was also shown at those flux levels (around  $5 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1} \text{ keV}^{-1}$ ) the percentage by which the decay was accelerated was proportional to the applied flux (Collins et al., 2001b). That might not be expected to hold when higher-order effects become involved at much greater flux levels.

In 2000–2001, two experiments were done at the third generation synchrotron radiation (SR) source, SPring-8 in Japan. They definitively showed that the inner shell electrons could be manipulated to leverage the excitation of nuclear transitions. Kishimoto et al., (2000) excited the 77.351 keV level of  $^{197}\text{Au}$  with “off-resonance” monochromatic SR X-rays by using the inner shell electrons to bridge the gap in energy. Working in a broad international collaboration, we used tunable monochromatic X-rays to measure the excitation function for the accelerated decay of isomeric  $^{178m2}\text{Hf}$  nuclei (Collins et al., 2002). We found an excitation function that peaked near the  $L_{III}$  and  $L_I$  edges for photoionization in Hf. Additional evidence for the “short-circuit” around the delay associated with the 8-level was found in the emission induced by the X-rays of  $\gamma$ -photons in a line near 130 keV, not present in spontaneous decay (Collins et al., 2001a).

More recently, we demonstrated that the excitation function for accelerated decay of the isomeric  $^{178m2}\text{Hf}$  nuclei was a description of the XAFS phenomena in the target we were using and not some artifact from noise in the monochromator or elsewhere in a particular SR source. Shown in Fig. 2 are the results obtained in 2001–2003 with the SR sources at SPring-8 and at the Paul Scherrer Institute (PSI) in Villigen, Switzerland. Agreement is good between results taken with completely

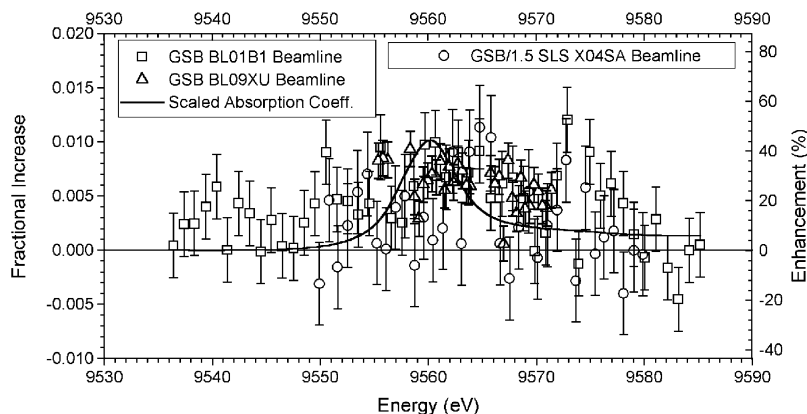


Fig. 2. Fractional increases in the number of  $\gamma$ -photons collected in the GSB of  $^{178m2}\text{Hf}$  together with the relative absorption coefficient scaled to fit the vertical range to show the Hf ( $L_{III}$ ) edge. Data from three beamlines are shown as marked. The scale to the right has been corrected for the duty cycle of the SR excitation.

different SR sources over the three year period. Such agreement removes any doubt that the structure found in the excitation function for enhanced GSB emission might have been an artifact of a particular X-ray source.

During two experiments in 2002, SPring-8 was operating in the multi-bunch mode meaning there were 11 repetitions of a “bunch” of 160 current pulses lasting 50 ps, each pulse being followed by a spacing gap of 2 ns. Between bunches, there were a few extra spaces and for stability a 12th bunch was left unfilled. Neither bending magnet nor undulator has significant  $Q$  so that the SR follows the same duty cycle. Thus the  $\gamma$ -emission from the GSB must originate from a source with a duty cycle of  $(11/12) \times (50/2000) = 1/43$ . When the duty cycle is considered, the acceleration or decay of the  $^{178m2}\text{Hf}$  isomeric population caused by the creation of  $2p_{3/2}$  holes in the electron shells of the atoms approaches 50% near the maxima seen at 9561 and 9567 eV in Fig. 2.

Most recently, focus upon the use of SR sources to accelerate the decay of the 31-yr isomer of  $^{178m2}\text{Hf}$  has been placed upon establishing the highest levels of statistical confidence for the excitation function of the effect and upon the benchmark “signature” of the triggered decay. To optimize performance, particular care in characterization of the physical target and its alignment is essential. Fig. 3 shows direct measurement of the physical characteristics of a typical target used at SPring-8 in 2002. It contained about  $1.8 \times 10^{13}$  isomeric nuclei. The isomeric fraction was about  $10^{-3}$  of the total Hf content in the form  $\text{HfCl}_x\text{O}_y$ . Comparable levels of activity from an impurity,  $^{172}\text{Hf}$  and its daughter were also included in the material from which the samples were fabricated. Each sample was encapsulated and mounted on a goniometer to be inclined about  $7^\circ$  above the plane of propagation of the incident X-rays. In this way the normal area presented by the target to the SR

flux was smaller than the transverse cross section of the SR beam. Fig. 3 shows the tomography of the target measured with X-rays at 9561 eV incident upon the tilted sample mounted upon the goniometer. The X-ray energy corresponds to the  $L_{\text{III}}$  edge in Hf for photoionization in order to maximize absorption by the Hf content. It is significant that the measured absorption coefficient for the Hf is comparable to the total losses for the substrate and window material and that the sum of all losses still leaves all of the activity exposed to useful fractions of the incident flux.

Two beamlines at SPring-8 were used for irradiation, BL01B1 and BL09XU in which the insertion devices consisted of a bending magnet and an undulator, respectively. Fluxes ranged from  $9.5 \times 10^{10}$  to  $2.0 \times 10^{12}$  photons  $\text{cm}^{-2}\text{s}^{-1}$  with nominal spectral bandwidths chosen between 1 eV and 100 meV, by adjustment of the monochromator. Spectra were examined by advancing the energy of the X-rays in steps with data being collected for 100–500 s at each step. Often data were collected for an additional 30–150 s during which the SR was blocked by a thick absorber. The SR intensities and sample absorption coefficients were continuously measured in-line with ionization chambers placed before and after the isomeric target. Also as before, 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 by observing the  $L\alpha$  fluorescence from Hf in the target.

Two opposed Ge detectors, 47 mm thick  $\times$  45 mm diameter and 43 mm thick  $\times$  50 mm diameter, respectively, were placed 22 mm from the target at right angles to the beam with the target between them. Each was shielded with layered foils of W, Cd, and Cu, as seen by the beam. They were connected to spectroscopy amplifiers from which the outputs were continuously digitized and recorded to permit subsequent reexamination of the electrical pulses describing the detected photons. Data were analyzed with a mesh of 6.8  $\gamma$ -channels per keV. When the SR monochromator changed energies, a digital signal was sent to a third channel of the data logging system to correlate the signals from  $\gamma$ -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  $\gamma$ -photons detected during each period of irradiation with SR.

For simplicity, in most cases in the work reported here, the sums of counts for the ROIs for the accessible members of the GSB at 213.4, 325.6, and 426.4 keV were used together with the “new”  $\gamma$ -line at 130 keV that is excited only during induced decay to study the excitation functions of the  $^{178m2}\text{Hf}$  sample. The ROI for the 125.8 and 181.5 keV impurity lines provided convenient indications for the null effect.

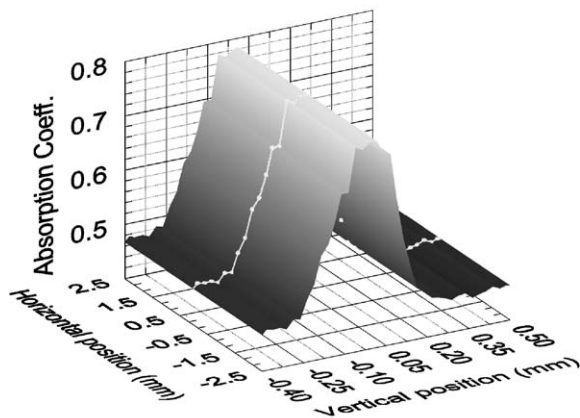


Fig. 3. Tomography of the absorption coefficient of the mounted target measured with SR X-rays at 9561 eV. Points locate discrete measurements.

Measurement of the excitation function of the new  $\gamma$ -line at 130.2 keV provided a significant supplement to the measurements of enhancements of members of the ground state band (GSB) previously reported (Collins et al. 2002). The upper panel of Fig. 4 shows the numbers of counts collected in the best measurements made in 2002. In the case of data for the 130 keV line made on the 09XU beamline, the scanning through the SR X-ray energies was made with 0.1 eV steps and the  $\sigma$  was calculated from the variance when 10 successive measurements were combined into one with width  $\Delta E=1.0$  eV. The GSB data were taken as described above. Summing the counts and combining  $\sigma$ 's according to statistical rules for the data between 9566 and 9572 eV gave a total accumulation of 6123 (970) extra photons over the course of our SR experiments in 2002. This indicates a confidence of  $6.3\sigma$  including all causes of variation, not just counting statistics as described above.

The lower panel of Fig. 4 shows the improvement obtained by remeasuring the pronounced feature for

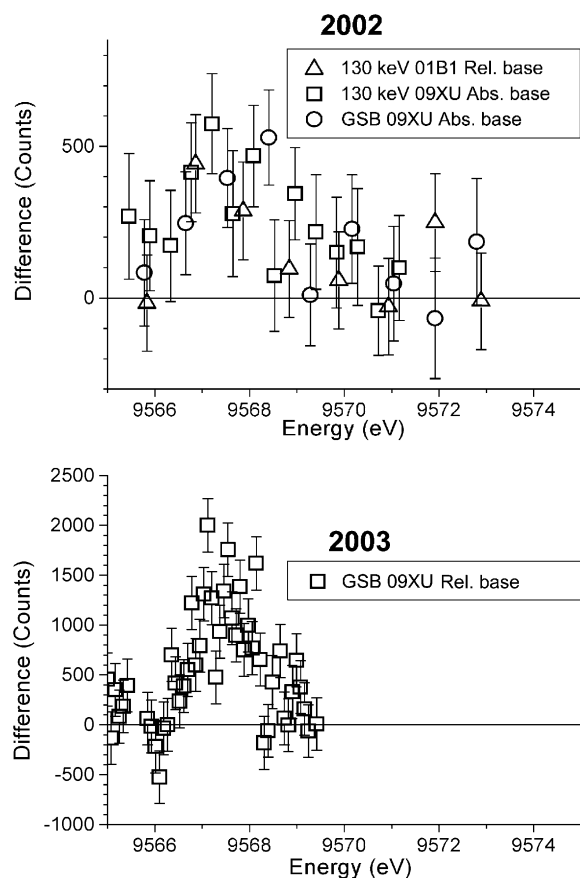


Fig. 4. Differences in the counts collected in the  $\gamma$ -lines indicated. “Abs” indicates that the baselines were obtained with the X-ray beam blocked as described in the text.

triggering near 9567 eV with monochromatic X-rays at the 09XU beamline at the SPring-8 source in 2003. Longer measurement times were used over reduced tuning ranges to obtain better statistics. A range of 7 eV was scanned with SR X-rays having width of 100 meV. Data were accumulated for 500 s at each X-ray energy. The total number of counts within the structure above the base shown in Fig. 4 is 11,954. Combining the  $\sigma$ 's for each corresponding point according to usual statistical practice gives an uncertainty for the total of 997. In summary, the number of extra counts induced in the GSB within the XAFS-like peak shown in Fig. 4 is 11,954 (997) giving a confidence of  $12\sigma$  that the number of  $\gamma$ -counts is increased by the X-ray irradiation. The combination of confidence of  $6.3\sigma$  established by measurements in 2002 and  $12\sigma$  achieved independently in 2003 appears to be a reproducible and compelling demonstration that monochromatic SR X-rays at energies 6 eV above the L3 edge in Hf will accelerate decay of  $^{178m2}\text{Hf}$  isomeric nuclei.

One of the important signatures of triggering the decay of the  $^{178m2}\text{Hf}$  isomeric nuclei is the emission of  $\gamma$ -photons in spectral lines not seen during spontaneous decay. Fig. 5 shows a comparison of the induced decay from the isomeric nuclei and the absence of such changes of counting rates of photons from the spontaneous decay of the impurity nuclides,  $^{172}\text{Hf}$  and its daughters. These data were obtained at SPring-8 by collecting counts from  $\gamma$ -photons detected during successive 200 s intervals of irradiation, separated by 35 s intervals during which the SR X-rays were blocked by a thick steel shutter so that only spontaneous emission was recorded. Multiplying the total counts collected during the time, the beam was blocked by the ratio (200/35) and subtracting the results from the

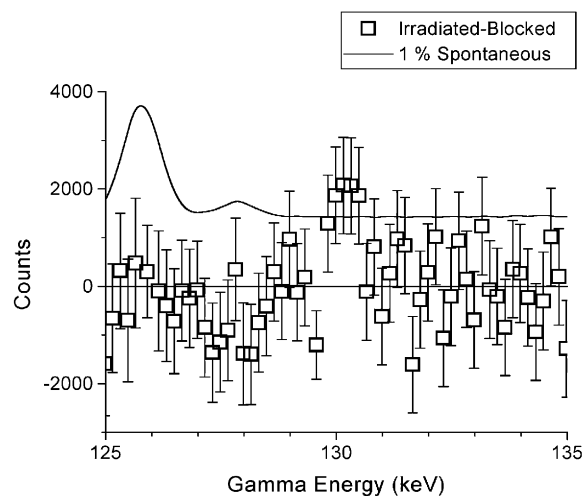


Fig. 5. Differences in the numbers of counts from  $\gamma$ -photons collected with SR incident and blocked.

spectra recorded during the time, the shutter was open yielded the results shown in Fig. 5. Statistical uncertainties were calculated from the numbers of counts and combined for differences according to accepted practice to obtain the error bars shown. Shown in Table 1 are numerical values for the counts from the induced decay of the isomer in comparison to the null result from the absence of any comparable change in the numbers of  $\gamma$ -photons from the ground state contaminant,  $^{172}\text{Hf}$  and daughters. Confidence that the results shown in Table 1 are not simply statistical fluctuations associated with the quantum nature of photon emission can be seen to be  $9.2\sigma$ . However, most recent measurements of the ambient levels of noise from electromagnetic interference that can be coupled into the  $\gamma$ -detection system suggest that the fluctuations seen in data such as shown in Fig. 5 exceed the minimums characteristic of quantum noise. While such additional noise could account for the rough appearance of the differences plotted in the figure, it could not account for the fact that enhancement during irradiation is found to the extent summarized in Table 1 for the 130 keV line while nothing is found in the reference line less than 5 keV different in  $\gamma$ -ray energy. Shielding for the detectors is being planned and will be constructed to reduce electromagnetic interference noise in future experiments.

Future research plans focus upon the time-dependence of the induced decay of the  $^{178\text{m}2}\text{Hf}$  isomeric nuclei. Prior to the use of SR irradiation, pulsed irradiation had already shown that any possible delay between irradiation and induced decay was less than 1 ms; a value precluding excitation of a cascade following the path of spontaneous decay because there was no statistical time lag associated with passage through the 4 s level at the head of the 8-band (Collins et al., 2001a). So it was perceived as an issue whether the induced decay was essentially “instantaneous,” meaning that applications requiring the control of the power, as opposed to the energy, from nuclear decay might be modulated at reasonable levels of stimulus applied externally.

As mentioned above, the electrons in the storage rings of SR sources are “bunched” into pulses of current of about 20–50 ps duration separated by about 2 ns in a pattern around the ring that is designed to meet user requirements. Insertion devices develop tangential X-ray beams from passage of the bunches and the temporal

pattern of X-ray pulses delivered to the users target reflects the bunch pattern in the ring. After a transit time around the ring the pattern repeats on a scale of  $\mu\text{s}$  for most 3rd generation SR sources. The collection of the  $\gamma$ -photons from the spontaneous decay is continuous while the accelerated emission is induced only during the times the X-rays are present. In Fig. 2 the ordinate to the left was the fractional increase of average  $\gamma$ -emission while the ordinate on the right was corrected for the 1/43 duty cycle of the X-ray irradiation.

The positioning of an avalanche photodiode (APD) at the end of the path of the X-ray beam after passing through the target activity enabled us to record the temporal structure of the X-ray pulses for comparison with the timings of the detection of the  $\gamma$ -photons from the decay they induced. Fig. 6 shows the first results obtained at the PSI source. Signals from the APD were conditioned with 1 GHz electronics so the apparent width of the X-ray pulses has been broadened to about 1 ns by instrumental limitations. The datum marked with the asterisk is an artifact found at all  $\gamma$ -energies and resulted from a “burst” of electromagnetic interference noise being coupled directly into the spectroscopy amplifiers. It exemplifies the difficulties of this type of

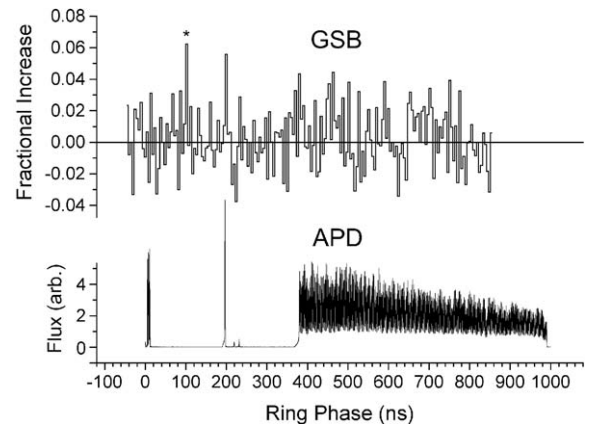


Fig. 6. Plot showing the induced decay of the isomeric nuclei as a function of the phase time of the electron bunches around the ring. The upper panel shows the fractional increase of  $\gamma$ -photons counted in the GSB when the target was irradiated at the phase times shown over the amount of spontaneous emission collected when the SR beam was blocked with a shutter.

Table 1

Summary of differences in the number of counts from  $\gamma$ -photons measured during irradiation and during the period the SR beam was blocked for data shown in Fig. 5

Nuclide	$\gamma$ Energy (KeV)	Data Channels No.	Counts (cts)	Error (cts)
$^{178\text{m}2}\text{Hf}$	$\sim 130$	5	9171	993
$^{172}\text{Hf}$	125.8	7	-786	1175

fluorescent detection mounted with relatively large (10%) Ge detectors and high resolution spectroscopy amplifiers.

While the first data shown in Fig. 6, seems to show a correlation of nuclear-XAFS fluorescence in the GSB with the pattern of the bunches in the ring to within a few ns, improved resolution is needed. There appears to be better correlation when irradiation returns immediately after a vacancy deliberately introduced in the bunch pattern with perhaps some indication of subsequent “fatigue” reducing the fluorescent yield of  $\gamma$ -fluorescence. The most recent measurements at SPring-8 at the BL09XU beamline confirm these indications raising the resolution of the correlation between cause and effect to  $< \sim 1$  ns. Since, the electrons are essential for leveraging the effect of the X-rays upon the nuclear levels, the evidence suggesting “fatigue” cannot be unexpected. There are many optical analogs such as damage and color centers that reduce the yield of various types of optical effects at very high levels of incidence flux, but future research is needed to consider other possible causes as well.

### 3. Conclusions

A current summary of this continuing research is that there is a nuclear-XAFS effect that uses the inner shell electrons in intermediate scattering states to leverage the coupling of electron energy and angular momentum into nuclear states. For  $^{197}\text{Au}$  the nuclear branch was small, being around  $10^{-8}$ . However, for isomeric  $^{178\text{m}2}\text{Hf}$  the branching ratio into nuclear excitation is about 0.16%, probably because of the more favorable ratio of interaction energy to nuclear level spacing. The effect of the nuclear-XAFS is to accelerate the decay of the 31-yr isomer  $^{178\text{m}2}\text{Hf}$  by broadening the paths of  $\gamma$ -cascades beyond the few included in spontaneous decay. By including states that effectively “short circuit” the statistical time lag associated with cascading through the 4 s 8-state, the application of pulsed X-rays causes the immediate decay of the energy stored by the isomer to within the 1 ns limits of current measurements. It seems that about 256x the energy successfully applied at the values corresponding to the L3 edge for photo-ionization is released by the resulting induced decay. Confidence limits are established to be  $12\sigma$  for observation of a maximum in the excitation function for nuclear-XAFS at 6 eV above the L3 edge for photo-ionization. Confidence that induced decay of the isomeric nuclei leads to the emission of a photons in a

new  $\gamma$ -line at  $\sim 130$  keV that is unique to induced decay is  $9.2\sigma$ .

Many potential applications are encouraged by this demonstration of a concentrated in situ source of “pump energy” that can be released by the creation of holes in the L-shells of the electrons surrounding the isomeric energy source. Since the induced “triggering” may be immediate, not only concentrated energy, but concentrated power can be developed as well. The latter is more important for the excitation of devices such as a gamma-ray laser.

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