

# Accelerated Decay of the 31-yr Isomer of Hf-178 Induced by Low-Energy Photons and Electrons

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**Abstract**—A process for transferring energy from electron shells into nuclear excitation (NEET) was first proposed by Masato Morita in 1973. It was generalized to a broader class of electron bridging mechanisms (EBM) by Matinyan. These proposals offered an exciting promise for modulating nuclear properties with X-ray photons at accessible levels of power. The development of 3rd generation synchrotron radiation (SR) sources made it possible to conclusively demonstrate NEET/EBM in two recent experiments by exciting nuclear levels of ground state <sup>197</sup>Au and isomeric <sup>178</sup>Hf<sup>m2</sup>, respectively, with SR X-rays from SPring-8. X-ray energies incident upon the isomeric target were tuned from 9 to 13 keV, and an excitation branch attributed to NEET/EBM was found to have a probability of  $1.6 \times 10^{-3}$  relative to L-shell photoionization of the surrounding electrons. The rate for the resulting emission of exoergic  $\gamma$  photons approached the theoretical maximum for such processes. These landmark results were reconfirmed and extended in a series of experiments at SPring-8 conducted in May 2002. Increases in the rates of spontaneous decay of the isomeric nuclei, <sup>178</sup>Hf<sup>m2</sup>, were studied when targets were irradiated with monochromatic SR X-rays tuned over hundreds of eV with spectral widths of both 1.0 and 0.1 eV. A close correlation of accelerated gamma emission with rates of ionization of the L-shell electrons in the target was observed. The frequent appearance of a triplet structure in the excitation spectrum for induced gamma emission resembled the additional structure seen in XAFS studies of the inner-shell electrons at energies near the ionization edges. In addition to the accelerated emission of some gamma lines present in spectra of spontaneous decay, new lines were also observed. Features in the excitation functions for such lines suggested that electrons as well as X-ray photons could initiate the accelerated decay of isomeric nuclei. In 2003, measurements of the temporal correlations between the pulses of SR stimulation and the fast response of the resulting gamma emission were studied successfully at the SR sources (SLS) at the Paul Scherrer Institute and at SPring-8. Results encourage prospects for applications depending on the control of nuclear properties with X-rays.

## 1. INTRODUCTION

The need to compensate the spontaneous power density radiated at threshold continues to impede the development of ultrashort wavelength lasers. Plans for both hard X-ray and  $\gamma$ -ray lasers demand insertion into the working medium of power densities far beyond our present means at the instant threshold is to be reached. The inability to focus most of the pump sources being considered for such devices further exacerbates the problem. Quite early, it was recognized [1] that storing the requisite energy density *in situ* could be a solution if (1) a material for storing high energy densities could be found and (2) the stored energy could be promptly released upon application of a triggering pulse of much lower energy. Focus immediately fell upon the nuclear spin isomers.

At low energies, there are similarities between structures of the excited states of atoms and nuclei that help to explain the importance of nuclear spin isomers. Both systems involve the storage of energy in the quantized movement of the constituent charges. In atoms, electrons populate excited states, which can be conveniently described in terms of single-particle orbitals. In nuclei, the neutrons appear as relatively negative concentrations in the overall positive fluid of the nucleus; so there is the possibility to store energy by raising either protons or neutrons, or both, up distinctly separate “ladders” of excited single-particle states. Many interesting cases occur in nuclei deformed from the idealized spherical shape and the possibility for long-lived metastability is clear, just as it is in the atomic analogs. Both spins and projections of spins can be aligned to create particularly stable states of excitation. However,

while difficulties in arranging suitable lifetimes for populations of metastable states at the atomic level have limited the development of practical applications, at the nuclear level, spin isomers store excitation at densities of GJ/g for decades.

In either the case of atoms or nuclei, the energy stored in the population of excited states is often released by the spontaneous emission of electromagnetic radiation. Atoms emit photons of light and X-rays while nuclei emit gamma photons, but the principles and selection rules remain the same. In each system, spontaneous emission can be “forbidden” for simple dipole transitions because there is no lower energy state to which the excited particle could “fall” without overturning its total spin,  $J$ , or a projection onto the figure axis of  $J$  (termed  $K$ ). Either would be a step particularly forbidden by selection rules. More characteristic of just the nuclear systems is the formation of “mixed states,” which have been considered as important for the triggering of the release of the metastability. Besides a mixing of charge identities that has no analogy at the atomic level, there are possibilities for mixings of other types, such as those introducing properties resulting from the quantized rotation of the spherical nucleus as a whole or even from oscillation between prolate and oblate asymmetries. By introducing additional degrees of freedom from such mixings, more transitions may become allowed that would facilitate subsequent decay. With the demonstration that induced gamma emission is possible by scattering X-rays from such materials, considerable interest has been developed in the study of these processes because it seemed that the intermediate excitation of mixed states could be involved.

The types of  $K$ -mixing states needed in such schemes to induce the decay of nuclear isomers were first reported [2] in  $^{180}\text{Ta}$  and subsequently described in  $^{174}\text{Hf}$  and other isomers [3]. In the case of Ta, the resonant absorption of X-rays excited the 2-quasiparticle isomer of  $^{180}\text{Ta}^m$  to a  $K$ -mixing level 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 de-excitation of the isomer was  $1.2 \times 10^{-25} \text{ cm}^2 \text{ keV}$ . After 11 years of substantial controversy, independent confirmation was reported [4] that X-rays could accelerate (or “trigger”) the prompt decay of a nuclear spin isomer in the case of  $^{180}\text{Ta}^m$ . However, since trigger photons of MeV energy were required, this success had little direct application to the  $\gamma$ -ray laser problem.

Extensive searches for other attractive isomers have suggested as many as 32 possibilities [5]. However, the application of scaling studies [6] of the excitation energies of potential  $K$ -mixing states, together with actual measurements [7], and the need for reasonable shelf life of a practical material have reduced the list to one isomer,  $^{178}\text{Hf}^{m2}$ . As a vehicle for the further study, the  $16^+$  state of  $^{178}\text{Hf}^{m2}$  is particularly attractive. It is a 4-quasiparticle state having 2.446 MeV of excitation

and a half-life of 31 years, meaning that as a material it would store 1.3 GJ/g. This is an energy density of considerable significance to the development of ultrashort wavelength lasers.

In 1997, scaling studies [6] of the isomers of Hf presented “further evidence for a  $K$ -mixing level in  $^{178}\text{Hf}^{m2}$  lying no more than about 300 keV above the isomer.” Since the cross section for exciting a level depends upon  $\Delta E^{-2}$ , where  $\Delta E$  is the transition energy, the value measured for  $^{180}\text{Ta}^m$  could be scaled upward by four decades if the excitation energy required were 28 keV instead of 2.8 MeV, provided the width of the triggering process remained the same. However, as energies of nuclear transitions scale downward, lifetimes for spontaneous emission increase and widths reduce, unless the inner-shell electrons become involved. In actual cases, the rates for  $\gamma$  transitions between levels separated by small energies are usually accelerated further by internal conversion in which inner-shell electrons are coupled into the process and significantly increase the widths. For example, the spontaneous decay of the  $^{178}\text{Hf}^{m2}$  isomer begins with a radiative transition for which the rate (and width) is increased by some few  $\times 10^4$  because of internal conversion to the surrounding electrons in the L3 inner shell; in this case, the coupling between nucleons and the electrons in the L3 shell that surround them is particularly effective. Such couplings between nucleons and the inner-shell electrons might act to prevent the width of isomer triggering from decreasing so much as transition energies were reduced and then initial and final states could be reversed so that energy was transferred into the nucleus.

A process for transferring energy from electron shells into nuclear excitation was first proposed by Masato Morita in 1973 [8]. Called NEET (Nuclear Excitation by Electron Transition), it was developed further by Ho [9] and generalized to a broader class of electron bridging mechanisms (EBM) by Matinyan [10]. 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.

In 1998, samples of the 31-yr isomer of  $^{178}\text{Hf}^{m2}$  were exposed to Bremsstrahlung X-rays from a small source normally used in dental examinations and acceleration of the spontaneous decay was reported [11]. Operating parameters had been selected to provide maximum flux at the energies corresponding to transitions from L-shell electrons of Hf without allowing the end point of the Bremsstrahlung to reach the energy of the  $K$  edge. Filtering of the X-rays with Al foils showed that pulsed irradiation by the parts of the Bremsstrahlung with  $E < 20 \text{ keV}$  increased the rate of spontaneous emission of  $\gamma$  photons corresponding to some of the characteristic transitions found in spontaneous decay of  $^{178}\text{Hf}^{m2}$  by a few percent. Integrated cross sections of the order of  $2.2 \times 10^{-22} \text{ cm}^2 \text{ keV}$  were reported [12, 13]. The enhancement was found to scale linearly [14] with

flux at 20 keV around  $10^{10}$  photons  $\text{cm}^{-2}$   $\text{keV}^{-1}$   $\text{s}^{-1}$ . An intense round of Comments [15–17] and Reply [18] concerning those results established that scaling of the integrated cross section with  $\Delta E^{-2}$  while holding the width constant did not produce cross sections violating any fundamental upper limits (RULs) if the electrons were involved [17].

The quantitative model used by Ho [9] for the use of inner-shell electrons to enhance the acceleration of the decay of isomeric nuclei was limited by his assumption that the initial state of the process was a stationary state even though it included an inner-shell vacancy. He described the event as occurring in sequential steps, which is not a realistic view for solid media under laboratory conditions. Inner-shell ionization is a scattering process in which interferences between indistinguishable options for the trajectories of the incident projectile produce the rich phenomenology found in the X-ray absorption fine structure (XAFS) literature [19]. Excitation functions for higher order effects, such as visible or X-ray fluorescence, typically show [20] complex structure from resonances at energies displaced from the simple ionization edge by as much as tens of eV. These result from the scattering interferences in the projectile state and so reflect the geometry of the local potentials in the medium. As a result, sample preparation and thickness are key physical quantities affecting the excitation functions for higher order effects of photoionization [21]. Although neglecting the structure arising in the incident channel for the process, Ho's estimations for the output branch coupling the excitation into the nucleus should have been unaffected by his approximations. He reported that the branch into nuclear excitation could be as great as 0.4% of the number of inner-shell vacancies produced in the intermediate steps of the scattering [9]. Simply adding Ho's exposition of the branch for nuclear excitation to the XAFS perspective makes it reasonable to expect nuclear excitation to result from photoionization by X-rays. The excitation function for the nuclear branch would be expected to display the same complex structure typical of other higher order effects arising from photoionization.

Working at SPring-8 and KEK in 2000, Kishimoto *et al.* [22] demonstrated that synchrotron radiation (SR) scattered by inner-shell electrons did lead to  $\gamma$  fluorescence from  $^{197}\text{Au}$ , but the yield was only  $5 \times 10^{-8}$  at the single X-ray energy used. At SPring-8 in 2001 and 2002, our group extended the successful results of Kishimoto *et al.* to the 31-yr isomer of  $^{178}\text{Hf}^{m2}$  and reported [23] a yield for several X-ray energies of 0.2%, a value approaching the theoretical maximum. These landmark results were reconfirmed and extended in a series of experiments at SPring-8 conducted in May 2002. Increases in the rates of spontaneous decay of the isomeric nuclei,  $^{178}\text{Hf}^{m2}$ , were studied when targets were irradiated with monochromatic SR X-rays tuned over ranges of hundreds of eV and tens of eV with spec-

tral widths of 1.0 and 0.1 eV, respectively. A close correlation of accelerated gamma emission with rates of ionization of the L-shell electrons in the target was observed. The frequent appearance of a triplet structure in the excitation spectrum for induced gamma emission resembled the additional structure seen in XAFS studies of the inner-shell electrons at energies near the ionization edges. In addition to the accelerated emission of some gamma lines present in spectra of spontaneous decay, new lines were also observed. Features in the excitation functions for such lines suggested that electrons as well as X-ray photons could initiate the accelerated decay of isomeric nuclei. In 2003, measurements of the temporal correlations between the pulses of SR stimulation and the fast response of the resulting gamma emission were studied successfully at the SLS at the Paul Scherrer Institute and at SPring-8. Results encourage prospects for applications depending on the control of nuclear properties with X-rays.

In 2001, concurrently with our successful experiments at Spring-8, an attempt was made to excite the same isomer at the APS SR source at Argonne, which failed [24] to produce any evidence for an acceleration of the spontaneous decay. Despite the availability of published work [12] reporting that the X-ray energies important for exciting the accelerated decay of the isomer were  $E < 20$  keV, the design of the first failed experiment at APS included a target in which the isomeric activity was buried in a thick layer of Al that effectively excluded X-rays with  $E < 20$  keV. In 2002, the target was redesigned, but the experiment failed again at APS [25]. The authors attempted to interpret the negative results as “proving” an upper limit on the cross section for accelerated decay of the isomeric  $^{178}\text{Hf}^{m2}$  that was considerably smaller than the established value [25] of  $1.6$  to  $2.0 \times 10^{-3}$  of the photoionization cross section for L3 electrons. However, from the published data [25], it can be seen that the instrumentation used to search for  $\gamma$  photons from accelerated decay was totally blind at the energy of the “new” line reported [13] to be a unique result of triggered decay. Moreover, the  $\gamma$  spectra shown [25] were collected with a gate set to open seconds after the irradiating SR X-rays were moved off the target. Since the triggered decay is prompt, they were looking too late to collect any  $\gamma$  fluorescence that had been triggered several seconds previously. Nevertheless, the impression has remained that such persistent failures [24, 25] from attempts to accelerate the decay of isomers at APS somehow place further burdens of proof upon the successful works.

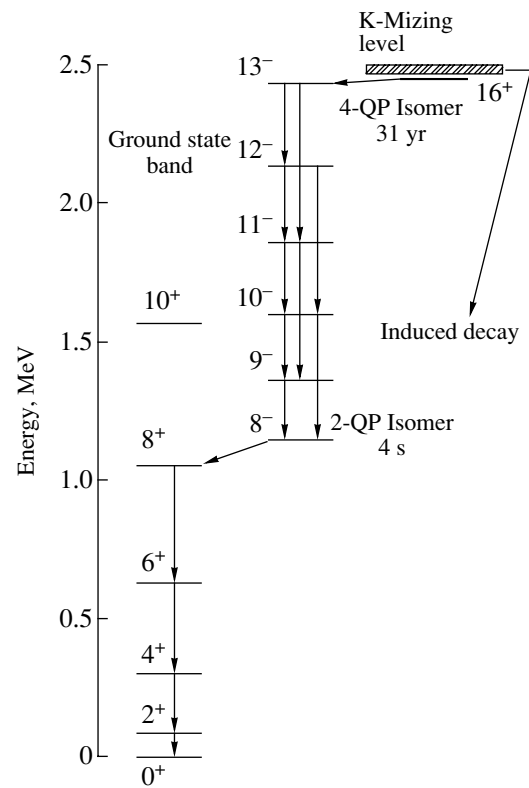
The purpose of the work reported here was to collect and extend our past measurements with a focus on the observations on which the highest confidence can be placed. As reported here, the confidence that an XAFS branch materially affects the decay of the 31-yr isomer of  $^{178}\text{Hf}$  reaches  $12\sigma$  when X-ray energies lie close to the L3 edge for photoionization. The effect most

readily observed is the emission of a new  $\gamma$  line near 130 keV and a prompt increase in the emission of  $\gamma$  photons belonging to transitions in the ground state band (GSB) of  $^{178}\text{Hf}$ . Such results encourage prospects for a  $\gamma$ -ray lasers, as well as set fiducial points with which to calibrate future experiments on isomer triggering.

## 2. EXPERIMENT

The design of an experiment to quantitatively measure the acceleration of the decay of isomeric nuclei is immediately confounded by the complexity of the bands of excited states. Figure 1 shows an energy level diagram of the excited states involved in the spontaneous decay. In that case, only two bands are involved, the GSB and the  $8^-$  band built upon the 4 s isomeric state. The corresponding spectrum of spontaneous decay from the targets used in our work has been described previously [11, 12] and was composed of only the well-known decays shown in Fig. 1 and of a  $^{172}\text{Hf}$  impurity and its daughters. Of the former, only the (4,2), (6,4), and (8,6) members of the GSB at 213.4, 325.6, and 426.4 keV, respectively, and the lowest member of the  $8^-$  band at 216.7 keV are recorded at significant counting rates. Of the lines from the impurities, only two, 125.8 and 181.5 keV, can be recorded with significance.

However, at least 17 other bands are known to have energy levels lying below that of the  $16^+$  isomeric level. There has been no reported cascading of spontaneous decay into those bands aside from the path shown in Fig. 1. However, when decay is induced by  $K$ -mixing, then “new”  $\gamma$  lines should be expected in the spectrum as larger changes of  $K$  are “mixed in” by the coupling to the electron transitions. One such line has been reported [13] by our group near 130 keV to arise from both triggering with Bremsstrahlung X-rays and with monochromatic X-rays from SR sources. Moreover, since the moment of inertia of the Hf nucleus is large and only little affected by quasiparticle excitation, the spacing of levels within bands often repeats. For example, within the  $1^-$  band with origin at 1310.1 keV, there are transitions at 129.4 and 216.4 keV that start at the same  $4^-$  level and which decay promptly with other transitions feeding the GSB starting with the (4,2) member at 213.4 keV. Another example is found in the  $2^-$  band originating at 1566.7 keV, with transitions from the  $4^-$  level being emitted at 180.4 keV in coincidence with other transitions too energetic to be detected with our Ge detectors, but which also feed the (4,2) member of the GSB. The two examples do not even approximately span the scope of complexity but already suggest that the observation of “prompt” triggering through 216 keV (not  $9^-$ ,  $8^-$  but  $4^-$ ,  $3^-$ ) in coincidence with the 213.4 keV component of the GSB would be possible, as would be the observation of triggering through  $\sim 181$  keV (not the impurity but the  $4^-$ ,  $2^-$  of  $^{178}\text{Hf}$ ) in coincidence with the same 213.4 keV GSB reference line. The pair of lines at 494.6 and 495.0 keV



**Fig. 1.** Energy level diagram illustrating the spontaneous and induced decay of the 31-yr isomer of  $^{178}\text{Hf}^{m2}$ .

from quite different bands and the pair 574.2 and 575.0 keV from different bands add the potentials for considerable uncertainty when the pattern of enhancements of spectral lines is not what is expected.

The design of an experiment to develop the highest possible confidence in the accelerated decay of the  $^{178}\text{Hf}^{m2}$  isomeric nucleus by X-rays should simply avoid, to the extent possible, any dependence upon  $\gamma$  transitions for which contributions to spontaneous decay could be enhanced by photons from transitions at nearly the same energies but arising from very different bands not normally observed. From this perspective, the (4,2) member of the GSB is satisfactorily isolated at 213.4 keV while the next (6,4) at 325.6 keV could be troubled by another transition at 324.9 keV.

The specific objectives of the experiments reported here were to demonstrate the following: (1) that the excitation function for observed enhancement of  $\gamma$  photons from  $^{178}\text{Hf}^{m2}$  isomeric nuclei contained XAFS-type structures that reproduced with high confidence, (2) that the enhancements observed included the excitation of at least one  $\gamma$  line not found in the spectrum of spontaneous decay, and (3) that the enhancement effect had a reproducible time-dependent relationship with the cause arising from the passage of electron bunches that generated X-rays within some insertion device in the SR source. The schematic design shown in Fig. 2

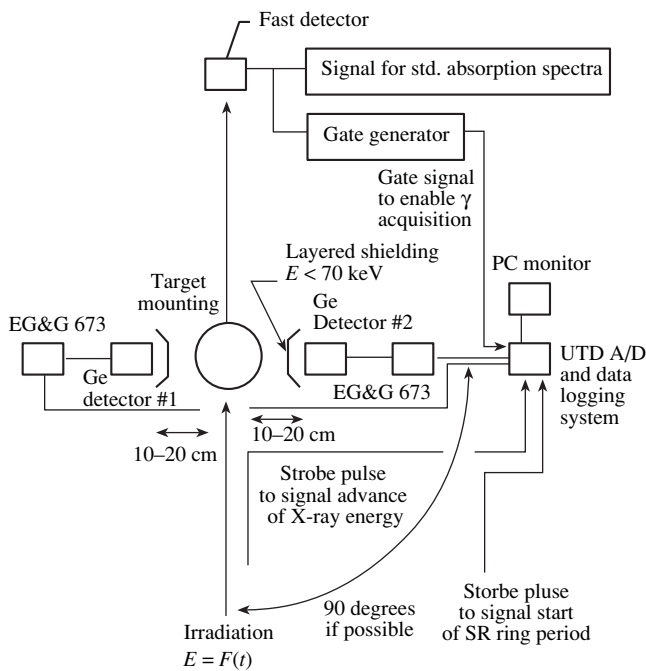


Fig. 2. Schematic drawing of the experimental arrangement.

was intended to support those objectives. It was used in all of the SR experiments to date, with small adaptations being made to conform to lessons learned and to physical constraints encountered at different facilities.

As a vehicle for the demonstration of NEET or EBM spectroscopy, the 31-yr  $16^+$  state of  $^{178}\text{Hf}^{m2}$  has been particularly attractive. When used with SR excitation through photoionization of the L shell, NEET and EBM offer the possibility for exoergic emission of  $\gamma$  photons with energies exceeding those of the irradiation. Consequently, experimental arrangements are facilitated by the use of layered absorbers to selectively remove X-rays scattered from the SR source [23]. That first report of the spectroscopy for the NEET or EBM excitation of  $^{178}\text{Hf}^{m2}$  suggested the existence of resonances in addition to those arising simply from the photoionization edges in the L shell. Reexamined in subsequent work reported here, the additional resonances were confirmed by tuning the incident monochromatic X-rays with high resolution and long dwell times through the L3 photoionization edge.

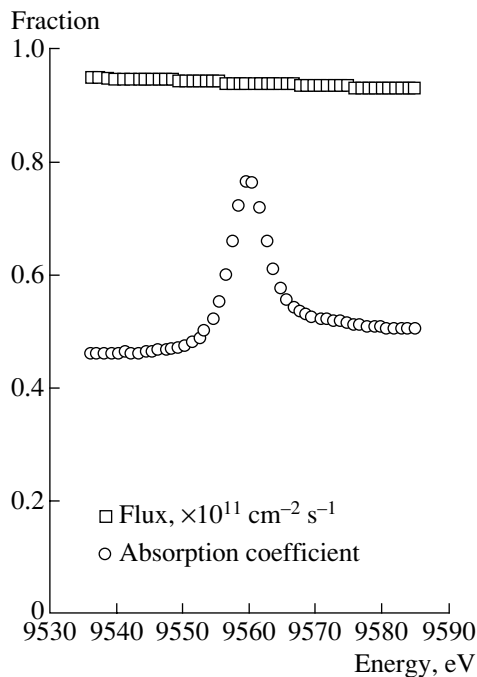
Four targets were used in these experiments. All were fabricated from the same material used in our previous work [23] and each contained about  $1.8 \times 10^{13}$   $^{178}\text{Hf}^{m2}$  isomeric nuclei. The isomeric fraction was about  $10^{-3}$  of the total Hf content. Two of the targets were fabricated in both 2002 and 2003 according to the same plan. In each target, the activity was deposited in a rectangular area nominally  $1.3 \times 5$  mm and the sample was encapsulated. In use, a target was mounted on a digitally controlled goniometer and inclined  $7^\circ$  to  $15^\circ$  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 cross section of the SR beam.

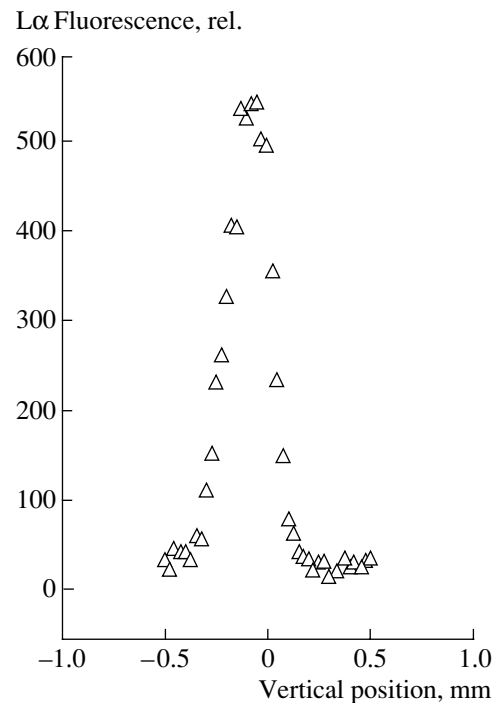
In 2002 two beamlines at SPring-8 were used for irradiation, BL01B1 and BL09XU, in which the SR was generated by a bending magnet and by an undulator, respectively. Fluxes were  $9.5 \pm 0.5 \times 10^{10}$  and  $5.0 \pm 1.0 \times 10^{11}$  photons  $\text{cm}^{-2} \text{s}^{-1}$  with nominal spectral bandwidths of 1 eV and 100 meV, respectively, after being passed through monochromatic filtration. In the first case, spectra were examined by advancing the energy of the X-rays in steps of about 1 eV with data being collected for 100 s at each step. Movement through a range of energies was repeated three times, and the results were summed. In the second case, the energies were advanced in steps of about 80 meV and remained at each for 200 s while data were collected and then for an additional 35 s during which the SR was blocked by a thick absorber. For comparison, the data were summed over five adjacent steps to improve statistics.

The performance of the target is of critical importance in such experiments. The first attempts elsewhere to excite isomeric targets with SR seem to have failed for lack of optical transparency at the resonant energies for NEET and EBM and because of ill-conditioning of the targets [24]. In the work reported here, the same attention was paid to the properties and mountings of the target structures as had been detailed previously [23]. The SR intensities and sample absorption coefficients were continuously measured in line with ionization chambers placed before and after the isomeric target. Overall absorption of the targets in these experiments was not significantly different from our previous experiments, except that uniformity of the distribution of the activity was improved. Values of flux and absorption coefficients are shown in Fig. 3 for a typical arrangement at the BL01B1 beamline. 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. An example is shown in Fig. 4. The data were obtained by moving the target vertically with a stepping motor while the intensity of  $L\alpha$  fluorescence from the Hf component was monitored by the SDD [23]. Because of the glancing angle of incidence, the contour seen is the near-Gaussian intensity distribution of the SR beam being “scanned” by the thinner projected dimension of the Hf content of the target. In vertical dimension, the beam can be seen to have been about 0.25 mm in the  $z$ -direction, and mechanical slits were set to trim the horizontal dimension to 7.0 mm. From the position seen in the figure, a final adjustment would have been made to center the peak at 0 mm.

Two opposed Ge detectors 47 mm thick and 45 mm in diameter and 43 mm thick and 50 mm in diameter, respectively, were placed 22 mm from the target at right angles to the beam with the target between them. Each



**Fig. 3.** Irradiation environment showing the absorption coefficient of the target measured in line as a function of SR energy during a typical irradiation at SPring-8 in 2002 together with measured values of SR flux.



**Fig. 4.** Profile of the SR X-ray beam scanned by the smaller dimension of the target as displayed by the counting rate for L $\alpha$  fluorescence.

was shielded with layered foils of W, Cd, and Cu, as seen by the beam. They were connected to EG&G 673 spectroscopy amplifiers, from which the output was continuously digitized and recorded to permit subsequent re-examination 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.

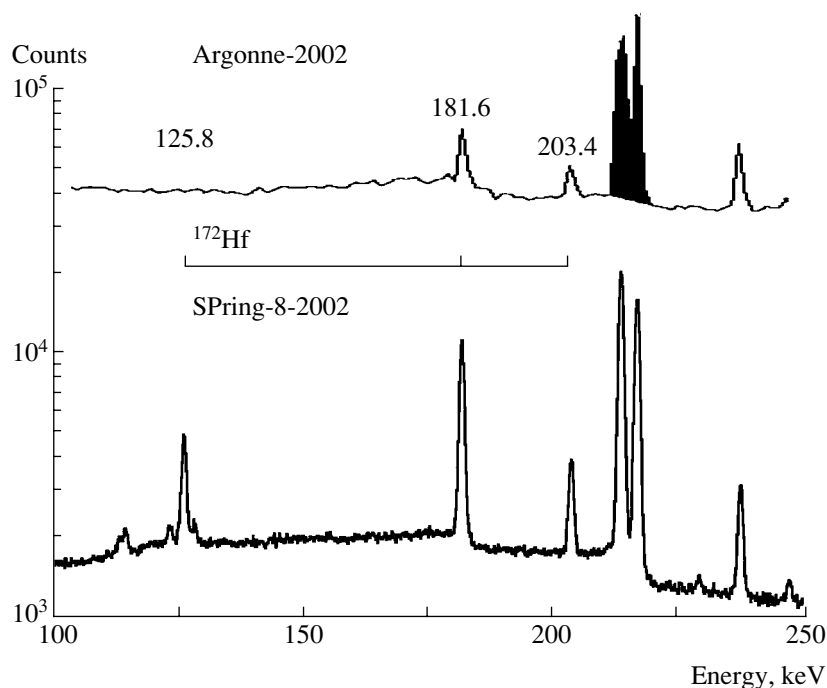
An illuminating comparison of the measurements systems used in the recently contradictory works is summarized in Fig. 5. Histograms of counts of  $\gamma$  photons detected during periods reported to be “typical” measurement series [25] are directly compared. Even though we collected only about 10% of the total number of photons, our histogram spectrum displays much better S/N, better energy resolution, and three-times better peak-to-Compton ratios. Most significant are the comparative sensitivities to lower energy photons. The three lines so marked arise from the decay of the impurity  $^{172}\text{Hf}$  and its daughters, so the relative strengths of the lines are fixed. The relative heights of the three lines in measurement histograms are proportional to the sensitivities of the detection system at those three photon energies. It can be seen that the APS (Argonne) system

had no sensitivity at 125.8 keV and so they could not have detected any photons from the important  $\gamma$  line near 130 keV that is only emitted by the accelerated decay of the isomers. In general, the performance of the measurement system used in the APS work was not functioning at modern norms.

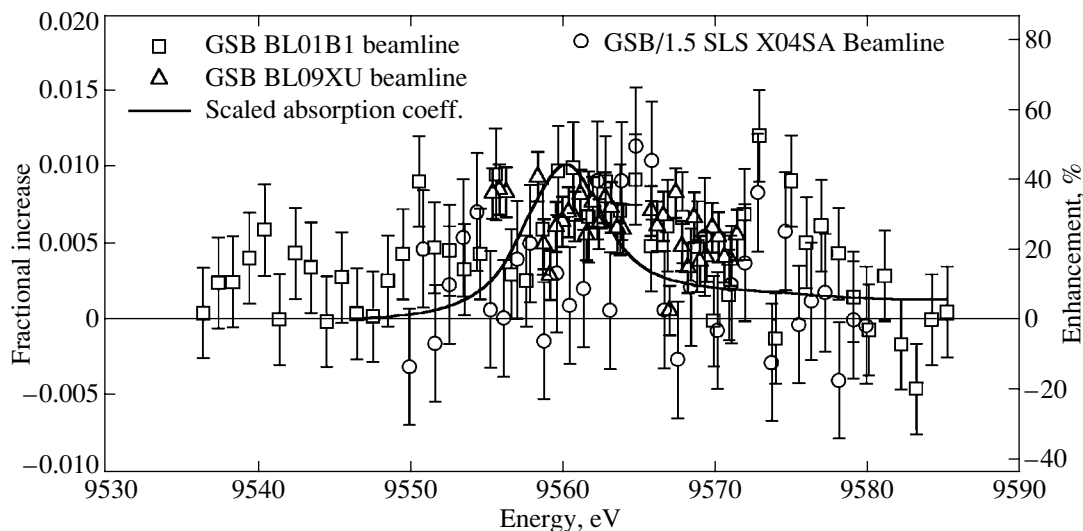
For simplicity, in most cases in the work reported here, the sums of counts for the ROIs for the accessible members of the GSB at 213.4, 325.6, and 426.4 keV were used to study the excitation functions of the  $^{178}\text{Hf}^{m2}$  sample. The ROI for the 181.5 keV impurity line provided a convenient indication for the null effect, provided it was not contaminated by contributions from one of the accidentally coincident lines of triggered isomeric nuclei.

### 3. RESULTS

The most remarkable evidence for excitation of the isomeric nuclei is shown in Fig. 6 in terms of fractional increases over the average counting rates observed at the lower X-ray energies below the L3 edge. Data identified in the legend box to the left were taken at SPring-8 in 2002. Plotted on the same figure are data identified in the right legend that were taken in 2003 at the SLS SR source of the Paul Scherrer Institute with essentially the same instrumentation, but while using the two targets fabricated in 2003. Because incident flux was somewhat greater, data have been reduced by a factor of 1.5 for comparison.



**Fig. 5.** Comparison of the energy resolution and amount of spectroscopic data reported by different groups for experiments performed in 2002 at the SR facilities shown.



**Fig. 6.** Fractional increases in the number of  $\gamma$  photons collected in the ROI for the GSB of  $^{178}\text{Hf}$  together with the relative absorption coefficient scaled to fit the vertical range to show the Hf(L3) 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.

Figure 6 confirms the suggestions of additional structure seen in Fig. 6 in our previous work [23] and corresponds to the general morphology of X-ray absorption fine structure (XAFS) measurements [19] made at the purely atomic level. Spectra for the excitation functions for higher order effects resulting from photoabsorption, such as visible or X-ray fluorescence [20], typically show additional resonances at energies

above and below the simple ionization edge seen in spectra of X-ray absorption; in those techniques, the sample preparation and thickness are the key physical quantities [21]. Main causes include transitions of inner-shell electrons to nonlocalized final states, such as the valence and conduction bands at X-ray energies below the edge. The involvement of two electrons can add structure above the edge. It seems reasonable to

conclude that such behavior has been found in these additional higher order effects of photoionization when the excitation is transferred into fluorescence from the nucleus.

The difficulty in extracting quantitative information from data such as that shown in Fig. 6 lies in determining the baseline above which the enhancement occurs without having data from a broader range of X-ray energies. For the first analyses, the baseline was selected to be a constant number shown by the line at zero in Fig. 6. It is not unique, but is one which is possibly consistent with the assumption that the effect of the SR irradiation is not to suppress the spontaneous decay of the isomeric nuclear state. Then, over the range of X-ray energies 9555.6 to 9568.9 eV corresponding to the main peak in photoionization at the L3 edge, the average enhancement in Fig. 6 for the 2002 data is 0.0068, corresponding to 10756 counts in excess of the 1587214 counts from spontaneous decay. The cross sections  $\sigma$  for excitation can be readily expressed as  $\sigma = fA/F$ , where  $f$  is the fractional enhancement over spontaneous emission (0.0068),  $A$  is the rate coefficient for spontaneous emission ( $A = 7.09 \times 10^{-10} \text{ s}^{-1}$ ), and  $F$  is the irradiating photon flux [23]. Values of flux were taken from Fig. 3 and were corrected for the mean of the absorption shown, giving for the resulting flux and cross section,  $6.5 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$  and  $7.4 \times 10^{-23} \text{ cm}^2$ , respectively. The photoionization cross section at the L3 edge is  $4.6 \times 10^{-20} \text{ cm}^2$ ; so this excitation in these targets represents about  $1.6 \times 10^{-3}$  of the photoionization probability. Though somewhat less than the value of 0.2% previously reported [23], this seems to be reasonable agreement, confirming again that this is a major effect.

During both experiments in 2002, SPring-8 was operating in the multibunch mode, meaning there were 11 repetitions of a “bunch” of 160 current pulses lasting 50 ps, with 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 a bending magnet nor undulator has significant Q; so 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 of decay of the  $^{178}\text{Hf}^{m2}$  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 9573 eV in Fig. 6.

Also significant among the results obtained in 2002 were the importance of “new”  $\gamma$  lines from transitions not seen in spontaneous decay. The number of counts from  $\gamma$  photons collected during the period the beam was blocked was multiplied by the ratio (200/35) and then was subtracted from the total number of counts collected during irradiation. Figure 7 shows an interval from the resulting difference spectrum without any further scaling. The Gaussian curve fitted to the data of

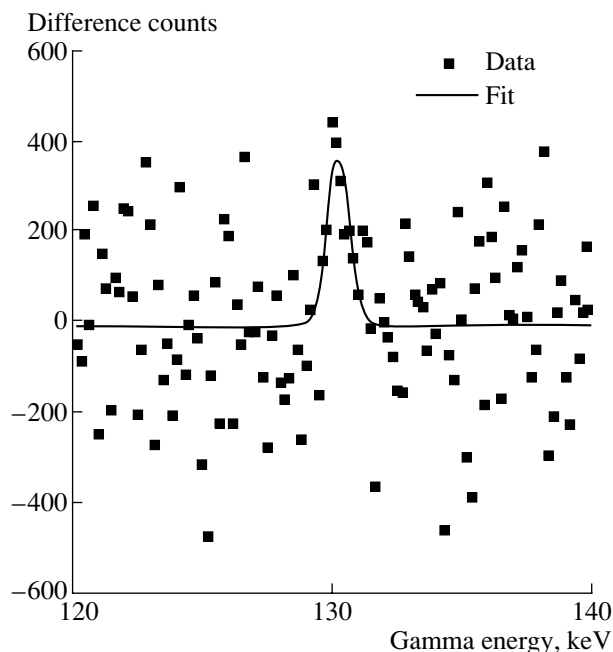


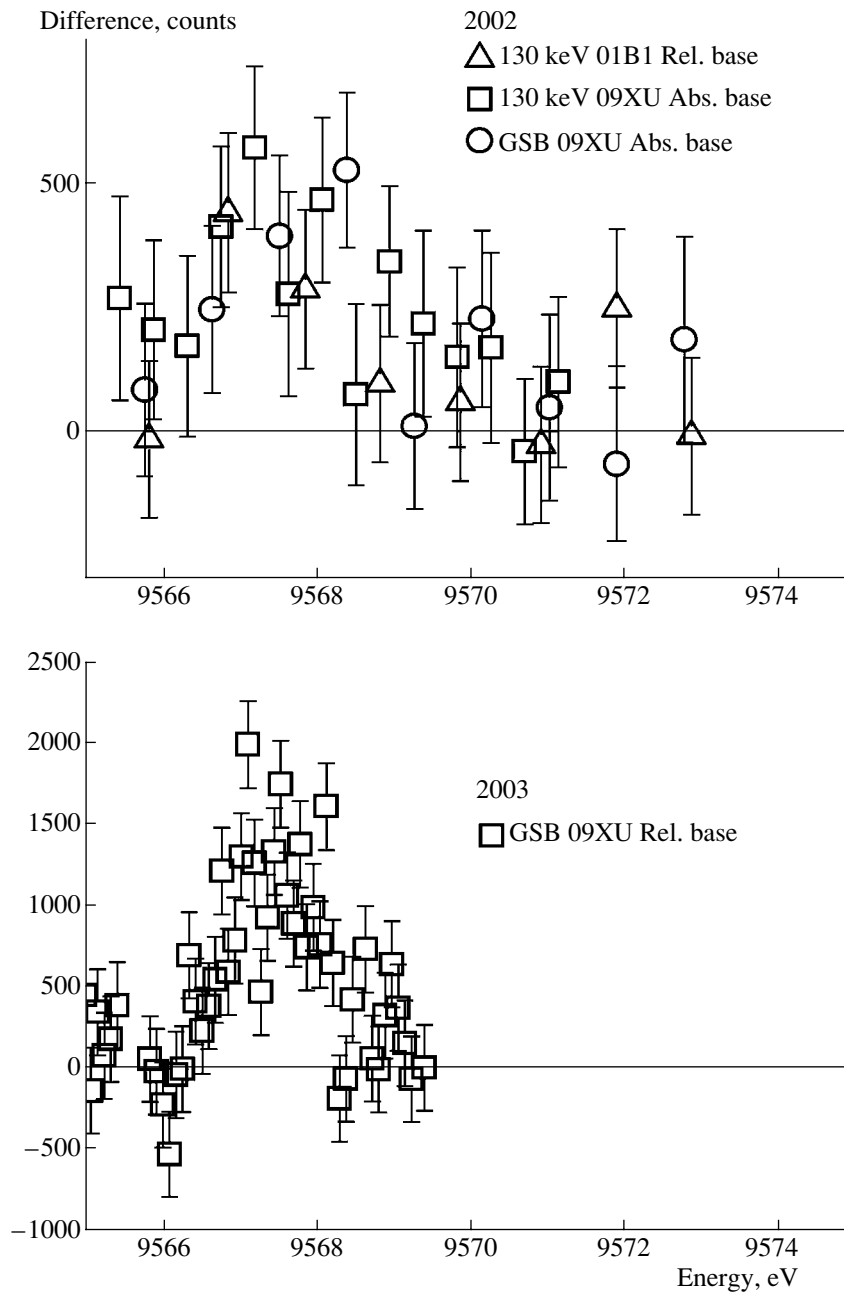
Fig. 7. Differences in the number of counts from  $\gamma$  photons collected with SR incident and blocked.

Fig. 7 gave an energy for the line of 130.23(14) keV. However, the uncertainty expressed in the centroid of the line is only statistical and does not reflect possible small shifts arising from the location of the line on the far wing of the larger impurity line at 125.8 keV. The value for the energy of the new line obtained as 130.23 keV is not significantly different from the value of 129.5 keV reported earlier [13]. The area under the curve in Fig. 7 was 405(118) counts. Coincidence measurements [13] had shown that this line was emitted only during the period of irradiation and was not present in delayed measurements [24, 25], such as those that had failed to show any enhanced decay.

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 GSB previously reported. The upper panel of Fig. 8 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 a step of 0.1 eV and  $\sigma$  was calculated from the variance when ten successive measurements were combined into one with a width  $\Delta E = 1.0$  eV. The GSB data were taken as described above. Summing the counts and combining  $\sigma$ 's in accordance with 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. 8 shows the improvement realized by remeasuring the pronounced feature for





**Fig. 8.** 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 a 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. 8 is 11954. 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. 8 is 11954 (997), giving a confidence of  $12\sigma$  that the number of  $\gamma$  counts is increased by the X-ray irradiation. The combination of the 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 of 6 eV above the L3 edge in Hf will accelerate decay of  $^{178}\text{Hf}^{m2}$  isomeric nuclei.

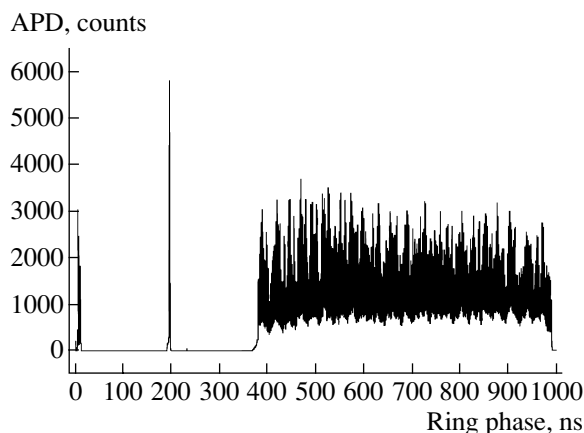
## 4. TIMING

An additional aim of the research on which this report is focused has been to measure the induced decay of the 31-yr isomer of Hf-178 while correlating the detection of the resulting gammas with the passage of an electron bunch. The most immediate objective was to confirm the efficacy of the measurement technique and to determine if the primary cascades of gamma transitions induced by the SR X-rays occurred promptly. Such knowledge would further illustrate the means through which reproducible measurements of isomer triggering could be made with high confidence by removing another possibly free parameter of experimentation.

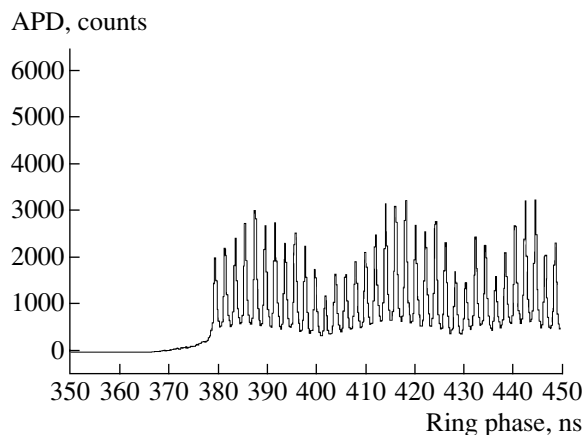
An avalanche photodiode (APD)  $0.2 \times 0.2$  mm was inserted at the position labeled “fast detector” in Fig. 2. It produced signal pulses that were conditioned with 1 GHz electronics. Standard nuclear instrumentation modules were used to develop a ramp voltage which increased until stopped by the arrival of a pulse supplied by SLS that was synchronized with the cycle period of the storage ring. Digitizing the final voltage reached by the ramp provided a measure of the “Phase time” at which the APD signal was detected with respect to the fiducial pulse available on each ring period and defined to be phase zero for reference. Also shown in Fig. 2 is that signals logged from detection of the gamma fluorescence by the Ge detectors were of two types. One was a standard semi-Gaussian analog pulse, the amplitude of which was proportional to the energy of the original gamma photon used to obtain the spectroscopy data reviewed above. The other was a standard NIM fast pulse containing the timing information obtained in the same way as was done with the APD signal. Both the energy and arrival (phase) time of each gamma photon were logged into a PC-based data acquisition system developed by us earlier.

Figure 9 shows the APD response measured as a function of the phase of the cycle of transit of the electron bunches around the storage ring at an average flux on the target of  $2.2 \times 10^{12}$  photons  $\text{cm}^{-2} \text{s}^{-1}$ . A vacancy was introduced into the pattern in order to isolate a single bunch to facilitate determination of the degree of rapidity with which the effect followed cause. With a combination of digital delay generators and software, the zero for the phase times was set as a point just before the onset of the vacancy in the pattern of bunches. Figure 10 shows a magnification of a part of the measured bunch pattern near the end of the vacancy. The actual duration of the passage of a current pulse is believed to be about 50 ps; so the temporal width seen in the figure is a measure of the limitation of the amplifier bandwidth to 1 GHz.

Figure 11 shows the induced decay of the isomeric nuclei as a function of the phase tune of the electron bunches around the ring. The data for the Ge detector was arranged to develop histograms of the numbers of gamma photons detected at the different phase times

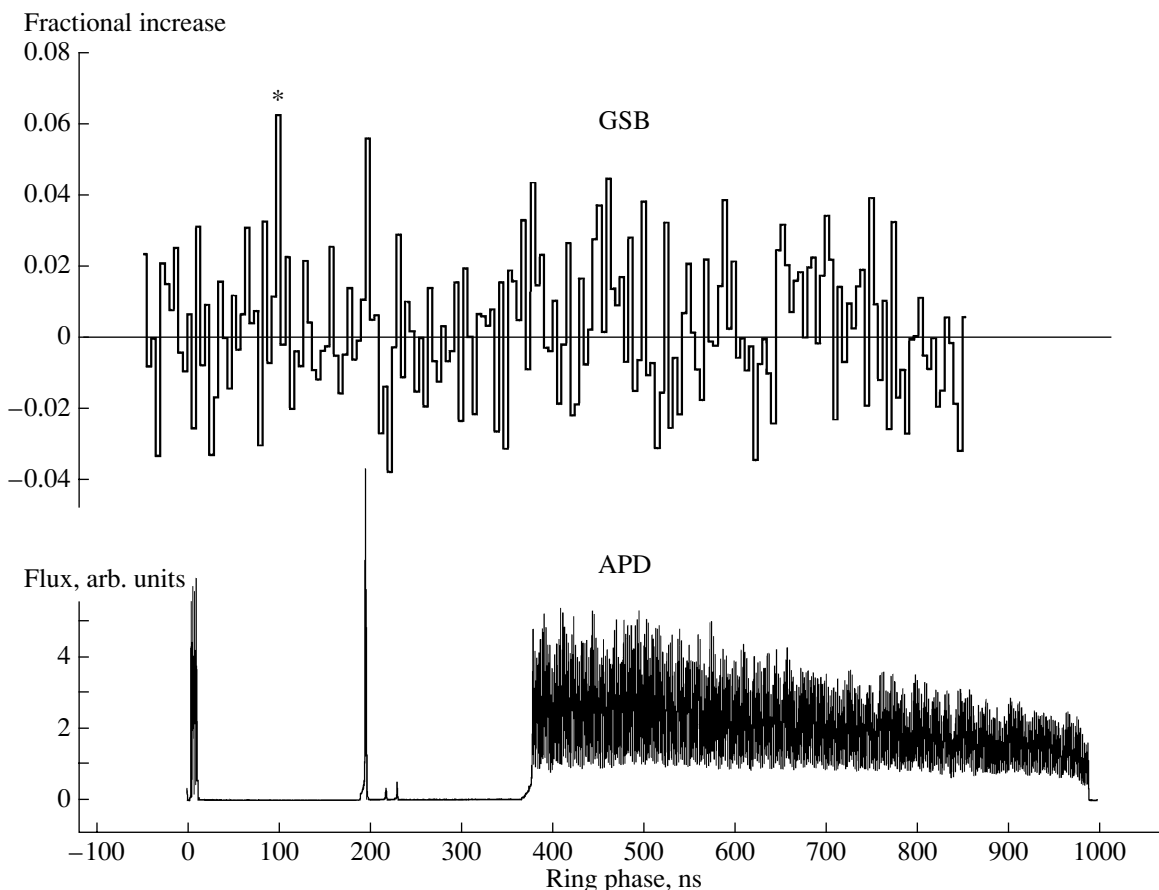


**Fig. 9.** Plot of the APD response measured as a function of the phase of the cycle of transit of the electron bunches around the storage ring at an average flux on the target of  $2.2 \times 10^{12}$  photons  $\text{cm}^{-2} \text{s}^{-1}$ .



**Fig. 10.** Magnification of a part of the measured bunch pattern near the end of the vacancy.

for each of the gamma energies corresponding to transitions between nuclear levels for either isomers or for impurities contained in the sample. During collection of the data, a digitally controlled beam shutter alternately opened and closed the beamline for a ratio of durations of 4:1. Photons collected while the shutter was closed were considered to represent the spontaneous emission from the sample. Data shown in Fig. 11 were the differences obtained by subtracting the number of counts collected for spontaneous emission and scaled by the relative times for collection from the data collected during irradiation. As mentioned above, the principle transitions in the GSB usually considered to show the induced emission and the rotational states of the nuclei to which the transitions correspond have been 426.4 keV (8,6), 325.6 keV (6,4), and 213.4 keV (4,2), respectively. In this work, we added the remaining member at 93.2 keV (2,0), respectively, so that the data marked GSB in Fig. 11 represents the entire GSB.



**Fig. 11.** 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.

The most significant result from Fig. 11 is the strong confirmation of the immediacy of the GSB cascade following irradiation with SR X-rays from each electron bunch. Also shown is one of the principal challenges to further work, SR noise. The peak marked with the asterisk is such an artifact. Unlike the rest of the structures, that peak shows in data for every gamma line. Further attention to electrical shielding is indicated in future experiments. The significance of the immediate timing of the induced decay is that it explains further why some competing experiments with the same material conducted at other SR facilities report failure to induce decay. A prevailing concept in other studies has been that induced decay should generally follow the decay cascades found for spontaneous emission, meaning that gamma fluorescence of the GSB should be delayed after irradiation with a statistical time lag of 4 s at the  $8^-$  level in between the isomeric state and the ground state. Because of artifacts such as those marked by the asterisk, it is an easier experiment to compare gamma counting rates being detected some seconds after the SR beam is blocked mechanically and then to look for a 4 s half-life for the decrease of that fluorescence. Since this current work has shown that the

gamma fluorescence follows SR irradiation by an interval smaller than the few nanosecond resolution of this experiment, the negative results in the simple measurements attempted elsewhere can be understood as waiting too long before opening the gate for data collection.

Figure 6 shows by open circles the excitation function for stimulating fluorescence through the GSB during phase time in the interval 350–550 ns. In order to make a direct comparison with the earlier results for this graph, the counts recorded in the GSB excluded contributions from the (2,0) transition.

## 5. CONCLUSIONS

In 2002 and 2003, four experiments conducted on three beamlines at two different monochromatic SR sources have produced a compelling set of results which affirm that the decay of the 31-yr isomeric  $^{178}\text{Hf}^{m2}$  nuclei can be accelerated by irradiation with low energy X-ray photons. Confidence of  $6.3\sigma$  was developed from data collected in 2002 and was increased to  $12\sigma$  by remeasurements conducted in 2003.

When examined as an incident monochromatic beam tuned through X-ray energies near the L3 photoionization edge of Hf, the excitation function measured for accelerated decay of the isomers in the sample displays structure resembling what is usually found in XAFS studies of inner-shell photoionization phenomena. Thus the induced emission of  $\gamma$  fluorescence can be perceived as a nuclear output channel of XAFS phenomena for which the branching ratio exciting it is  $1.6 \times 10^{-3}$ . The most pronounced structure in that excitation function for nuclear XAFS is a peak at 6 eV above the L3 edge at 9561 eV, which was been found in all measurements we made with SR sources.

For X-ray energies around the peak for nuclear XAFS, the induced decay of the isomeric nuclei is prompt. Any delay is shorter than we could measure with the timing system described above, meaning a few nanoseconds. When the complexity of the nuclear band structure of  $^{178}\text{Hf}$  is considered, it may not be surprising that the induced decay is prompt. After the  $K$  quantum number is mixed at the instant of triggering, there are considerably more cascades available that will avoid the statistical time lag associated with the  $8^-$  bandhead than there are cascades that feed the  $8^-$  level. Consistent with the observation that the  $\gamma$  fluorescence is promptly emitted after decay is accelerated is the observation of “new” lines in the  $\gamma$  spectrum of the induced decay. One such line lies near 130 keV. The excitation function for that line agrees with what is observed for the GSB and supports earlier published reports that the 130 keV line is excited in coincidence with the (4,2) member of the GSB at 213.4 keV [13]. A number of other new lines have been observed, but additional experimentation will be needed to establish reliability.

Confirmation of the efficiency with which monochromatic X-rays at fluxes between  $10^{10}$  and  $10^{12}$  photons  $\text{cm}^{-2}$   $\text{eV}^{-1}$   $\text{s}^{-1}$  will trigger the decay of the 31-yr isomer of  $^{178}\text{Hf}^{m2}$  can be obtained to a compelling degree of confidence by observing the prompt emission of  $\gamma$  photons in the GSB or around 130 keV when the X-ray energies are turned about 6 eV above the L3 edge for photoionization of Hf. Further work will be focused on the channels of cascades within the nuclei through which the fluorescent  $\gamma$  photons are emitted and on the spectra of the structures being found in the excitation functions for nuclear XAFS. Results to date provide strong encouragement for the development of a  $\gamma$ -ray laser by demonstrating the prompt release *in situ* of energy stored in spin isomers that is triggered by X-ray photons with quantum efficiencies of 256x at 9567 eV.

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