Search for induced emission from the ¹⁷⁸Hf^{m2} isomer by low-energy x rays

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(Received 14 June 2012, revised manuscript received 22 January 2013, published 10 July 2013

Whether or not the ¹⁷⁸Hf^{m2} isomer has the characteristic of induced γ emission by low-energy x rays has been a focus of attention for many scientists and researchers in recent decades. In this paper, an experiment regarding triggering ¹⁷⁸Hf^{m2} emission decay is conducted at Shanghai Synchrotron Radiation Facility. An x-ray beam with 20.825-keV energy irradiates a rectangular sample containing about 10¹¹¹⁷⁸Hf^{m2} nuclides. By comparing the net gamma count rates during the irradiation with those after the irradiation, the data show that at the 426-, 495-, and 574-keV gamma lines, there is no significant enhancement, indicating that the induced γ decay of ¹⁷⁸Hf^{m2} has not been observed.

DOI: 10.1103/PhysRevC.88.014312

PACS number(s): 23.20.Lv, 23.35.+g, 25.20.Dc, 27.70.+q

I. INTRODUCTION

The study of the controlled release energy of an isomer has aroused great interest since the 1960s; the experiment of detecting gamma fluorescence through low-energy x-ray irradiation on 180m Ta is of great significance to this issue [1,2]. Owing to the high energy density and the long half-life of ${}^{178}\text{Hf}^{m2}$, it is considered as the most suitable subject for studying the induced release energy [3]. The partial level scheme of ¹⁷⁸Hf is listed in Fig. 1 and this isomer spontaneous decay path can be seen clearly. There are many research groups which conducted 178 Hf^{m2}-induced gamma ray emission experiments. However, the results obtained from different research institutions were contradictory. In 1999, the Collins team used a bremsstrahlung x-ray source to irradiate the sample containing 6.3×10^{14178} Hf^{m2} isomers [4,5]. They observed that the gamma counts of the 426-keV line increased when the end point energy of the x ray was 70 keV, and gamma counts of 426 and 495 keV increased when the end point energy of the x ray was 90 keV. After that, this team did a few triggering gamma decay experiments by applying the same x-ray source and then drew some positive conclusions. To validate Collins' results, the well-known research group from Argonne National Laboratory also did similar experiments at the Advanced Photon Source. Ahmad and his colleagues used two undulator gap settings and covered the 20-keV range of x-ray energy, but did not obtain the photon-induced deexcitation [6,7]. From 2000 to 2005, Collins et al. [8-15] studied further the controlled release energy of 178 Hf^{m2} isomer. One of the results indicated that a 2.457-MeV gamma photon was detected when an x ray of 9.567 keV irradiated the sample containing ¹⁷⁸Hf^{m2} isomers at Spring-8 Synchrotron Radiation Facility (SRF). They thought 1^{178} Hf^{m2} could decay directly to the ground state after absorbing an x-ray photon, but not go through the original spontaneous decay path. Another result obtained by this team was that the gamma counts of 495and 574-keV lines all increased, and the enhancement of the 495-keV line was more than that of the 574-keV line when a 20.825-keV x ray bombarded the 178 Hf^{m2} isomer. Ahmad *et al.*

have not continued to research the x ray from SRF triggering 178 Hf^{m2} isomer gamma emission. The Carroll research group has also conducted independent experiments with the highly sensitive detector array by using incidence energy 9.567-keV x ray to irradiate 178 Hf^{m2} isomer [16,17]. The result also disagreed with that of Collins.

At present, there are no reliable nuclear level experimental data to support the low-energy photon-induced deexcitation of 178 Hf^{m2}, but this does not mean it is certain that there is no "intermediate level." There is such a significant difference among the results of the 178 Hf^{m2}-induced gamma decay that more scholars and international research institutes are paying more attention to it and focusing on the study of the characteristics of the isomer. Under the support of the National Natural Science Foundation of China (NSFC) we have done some studies, such as production and chemical separation, about 178 Hf^{m2} isomer, which were published in Ref. [18]. An experiment of the low-energy x ray triggering 178 Hf^{m2} gamma decay is described here in detail.

II. EXPERIMENT

A. Sample preparation

Based on the 176 Yb(α ,2n) ${}^{178m^2}$ Hf reaction, some 178 Hf ${}^{m^2}$ isomers were produced at the CS30 Cyclotron. Because the natural Yb raw material was used to produce the isomer, many long-lived non- 178 Hf ${}^{m^2}$ nuclides including 175 Hf, 172 Hf, and 173 Lu were also created. Through chemical purification, these nuclides were extracted to a large extent and 1.2×10^{11178} Hf ${}^{m^2}$ isomer nuclei were firstly adopted to carry out the low-energy x ray triggering γ decay experiment [18].

The experiment was conducted in BL13W1 Hatch at Shanghai Synchrotron Radiation Facility (SSRF), which could provide an x-ray beam with the energy ranging from 8 to 72 keV. Because the beam section was a 45 mm × 5 mm rectangle, the sample was also made to be rectangular with an area smaller than the size of the x ray. Meanwhile, the LABSOCS code was used to calculate the absorption efficiency of the 50- μ m-thick polypropylene membrane for the low-energy x ray. The theoretical data indicated that the number of photons absorbed by the membrane was only 10%–30% of the total photons for the 10-keV x ray. Therefore, the sample containing

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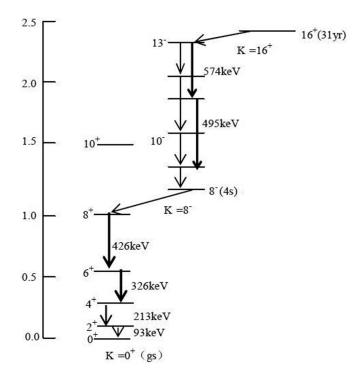


FIG. 1. The partial energy level scheme of ¹⁷⁸Hf.

¹⁷⁸Hf^{m^2} isomer was covered by a very thin polypropylene membrane to avoid external contamination. Figure 2 is a photograph of the sample before being covered. The bar cracked material is the sample containing ¹⁷⁸Hf^{m^2} isomers, which are disposed on the white filter paper and the sample thickness is no larger than 100 μ m. Lists of the sample specifications and details of sample preparation were described in another paper [18].

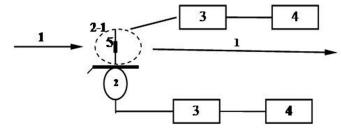


FIG. 3. The schematic diagram of the x ray irradiating 178 Hf^{m2} isomer experiment (1: x ray, 2: portable Ge detector, 2-1: Ge detector (dashed line), 3: MCD, 4: computer, 5: sample).

B. X-ray source

The BL13W1 beamline of Shanghai Synchrotron Radiation Facility is mainly used to research x-ray imaging and biomedical applications. The design specifications are described as follows:

Unfocused monochromatic beam

Imaging techniques

Photon energy range: 8-72.5 keV

Energy resolution ($\Delta E/E$): $<5 \times 10^{-3}$

Beam size: 45 mm (H) \times 5 mm (W) at 30 m at 20 keV

Flux output: 6×10^{10} photons/s/mm² at 20 keV at Si111 (a theoretical value)

 $7\times 10^8\ phs/s/mm^2$ at 70 keV at Si311 (a theoretical value)

Due to the appropriate energy and beam size, the BW13L1 beamline is chosen for the induced γ decay experiment.

C. Experiment arrangement

According to the results of Ref. [15], this experiment aims to confirm whether the gamma counts of energy 495-and 574-keV lines will increase and the enhancement of the former

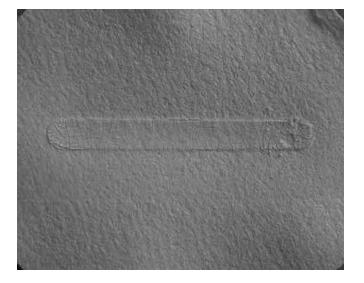


FIG. 2. The photograph of sample containing ${}^{178}\text{Hf}{}^{m2}$ isomer before being covered by polypropylene membrane. (This photograph is not enhanced.)

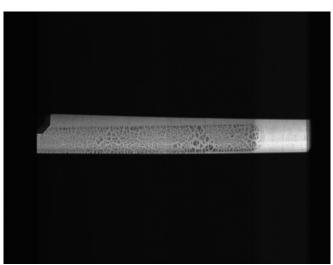


FIG. 4. The scan image of sample in the progress of x-ray flux irradiation. (This photograph is not enhanced.)

TABLE I. The gamma ray energies and branching ratios of main nuclides of the sample.

Nuclides	Energy (keV)	Branching ratio
¹⁷⁵ Hf	343.4	0.869
⁷² Lu	181.42	0.192
	900.69	0.277
	1093.64	0.752

will be stronger than that of the latter, when the energy 20.825-keV x ray irradiates ¹⁷⁸Hf^{m2} isomers.

Figure 3 shows the experiment arrangement. Here, the Ge detector 2-1 (dashed line) is settled on the ground. From the top view, it is vertical to the page. This GC12021 HP Ge detector, made by Canberra Corporation, has a relative efficiency of 30% and the energy resolution of 60 Co in its peak value 1332.5 keV is 1.8 keV. The other Ge detector is portable and made by the Ortec Company with a 30% relative efficiency. In order to prevent the two detectors from receiving x rays and their scattered photons, their top sides are covered by the shielding layer. The Ge detectors are 2 cm away from the sample center. Through the two DSP200 multichannel digital systems, the gamma photon signals are transformed into digital signals and then recorded. Two ion chambers are used to monitor the intensity change of the x ray in front of and behind the sample. Meanwhile, the position of the sample is adjusted repeatedly to make sure the x-ray flux goes through the sample effectively. Figure 4 is a scan image of an x ray irradiating the sample, in which the rectangular light region indicates the x-ray flux and cracked black zone sample containing ¹⁷⁸Hf^{m2}. By adjusting the position of the sample repeatedly, the crack black zone is contained in a rectangular region. It means that the whole sample can receive the x-ray irradiation.

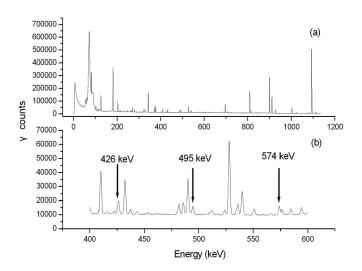


FIG. 5. The gamma spectrum of 426-, 495-, and 574-keV lines of the isomer measured. Plane (b) is for a clearer gamma count description between 400 and 600 keV.

TABLE II. Raw data of gamma rays of ¹⁷²Lu, ¹⁷⁵Hf, and ¹⁷⁸Hf^{m2} isomer without x-ray irradiation.

Nuclide	Energy (keV)	Counts	Uncertainty (1 σ)	FWHM	Count rate
¹⁷⁵ Hf	343.154	921162.75	0.18%	1.507	71.999329
¹⁷² Lu	181.038	1963286.3	0.11%	1.407	153.45311
	900.636	2017172.3	0.09%	1.823	157.66492
	1093.498	3800768.5	0.06%	1.913	297.07322
$^{178}\mathrm{Hf}^{m2}$	426.27	55516.57	3.41%	1.603	4.3019247
	494.757	38624.73	1.04%	1.731	2.9929925
	574.179	41274.93	0.98%	1.637	3.198354

D. Experiment plan

During this triggering experiment, the energy of the x ray is chosen to be 20.825 keV which is recommended by the Collins team [14] as the value to trigger 178 Hf^{m2} to accelerate the gamma emission. To obtain highly credible results, the following steps shall be noticed:

When compared with the results of Collins *et al.*, the number of the isomers used here is smaller than three magnitudes, so the gamma measurement continues until the time lasts for more than 3 h when the x ray is turned on or off.

The background should be monitored to investigate the influence caused by the structure material of BL13W1 hatch.

Because the time of using the x ray provided by SSRF is limited, the gamma ray detection with x-ray irradiation can be repeated only twice. No matter whether the triggered decay is prompt or delayed, so long as the induced gamma emission exists, the enhanced gamma ray should be accumulated and recorded by the detectors during the measurement time.

For other research results, the data used to compare are the net gamma count. Here, the net gamma count rate is proposed to take part in the comparison when the x ray is turned on and off.

III. DATA PROCESSING AND DISCUSSION

A. Gamma spectrum analysis

Two different high-purity Ge detectors receive simultaneously the gamma rays from $^{178}\text{Hf}^{m2}$ so as to ensure the effectiveness of the data. Because the measurement efficiency of the stationary detector is higher than that of the portable

TABLE III. Raw data of gamma rays of ¹⁷²Lu, ¹⁷⁵Hf, and ¹⁷⁸Hf^{m2} isomer with first x-ray irradiation.

Nuclide	Energy (keV)	Counts	Uncertainty (1σ)	FWHM	Count rate
¹⁷⁵ Hf	343.154	712801.63	0.21%	1.555	69.293986
¹⁷² Lu	181.038	1513455.1	0.13%	1.449	147.12837
	900.636	1554239	0.11%	1.862	151.09311
	1093.498	2935071.5	0.06%	1.962	285.32876
$^{178}\mathrm{Hf}^{m2}$	426.27	43531.89	2.86%	1.655	4.2021119
	494.757	30040.75	1.27%	1.801	2.8998188
	574.179	31659.11	1.16%	1.692	3.0560383

Nuclide	Energy (keV)	Counts	Uncertainty (1σ)	FWHM	Count rate
¹⁷⁵ Hf	343.154	1568101.8	0.14%	1.565	68.785851
¹⁷² Lu	181.038	3327727	0.09%	1.467	145.97301
	900.636	3419598.8	0.07%	1.873	150.00303
	1093.498	6461133	0.04%	1.974	283.422
$^{178}{ m Hf}^{m2}$	426.27	96312.74	1.93%	1.677	4.1580191
	494.757	67576.23	0.88%	1.785	2.9174049
	574.179	69369.07	0.79%	1.685	2.9948055

TABLE IV. Raw data of gamma rays of ¹⁷²Lu, ¹⁷⁵Hf, and ¹⁷⁸Hf^{m2} isomer with second x-ray irradiation.

detector, it is more suitable to acquire the information about the induced decay by analyzing the gamma spectrum from a stationary detector and applying its data. The gamma spectrum of the sample is shown in Fig. 5.

In this work, although a shielding layer is used to decrease the effect of low-energy rays and their scattered photons, the scattered high-intensity x rays still affect the measurement efficiency of the gamma ray. Therefore, ¹⁷⁵Hf and ¹⁷²Lu are used to calibrate the dead time of gamma measurement. Table I lists the radioactive nuclides of the sample and their energy of characteristic gamma rays with a large branching ratio.

The gamma spectrum analysis method is the key to obtaining accurate data. Because this sample contains ¹⁷⁵Hf, ¹⁷²Lu, and ¹⁷²Hf, among which the gamma ray from ¹⁷²Lu is especially sufficient and the energy range varies from a few keV to many MeV, the gamma ray counts of energy 426-, 495-, and 574-keV characteristic peaks of ¹⁷⁸Hf^{m2} are affected by those of ¹⁷²Lu. Here, an interactive peak fit is employed to get gamma counts of characteristic peaks with energy 426, 495, and 574 keV of 178 Hf^{m2}. For analyzing the 426-keV line, the region of interest (ROI) selected is from 419.3 to 440.6 keV. The total gamma photon count also contains the contribution from the 427.3-keV line of ¹⁷²Lu besides the gamma photons of ${}^{178}\text{Hf}^{m2}$. In the course of fitting, the good peak shape of the 426-keV line of 178 Hf^{m2} makes it easy to ignore the fact that the contribution from the 427.3-keV line of ¹⁷²Lu should be deducted. Comparing the net peak area of 426 keV of isomer after and before the deduction of gamma counts of the 427.3-keV line of ¹⁷²Lu, the former (the value obtained after such deduction) is 3200 counts, smaller than the latter (the value before such deduction), and the difference is about 0.73% of the former. The residual of the former ranges from -2.2% to 1.7% and that of the latter varies from -3.1% to 4.2%. Similar to the 426-keV gamma peak, if deducting the counts of the 493-keV line of ¹⁷²Lu, in terms of the net count of energy 495-keV line of ¹⁷⁸Hf^{m2} isomer, the former is 1014 counts less than that of the latter. The residual of the former ranges from -1.8% to 2.1% and the latter varies from -2.2% to 5.2%. Tables II, III, and IV present the raw data obtained through the interactive peak fit code. Uncertainty is composed of the statistic error and the fitting method error.

B. Data processing

As mentioned by McDaniel *et al.* [19], it is very difficult to process the data in such an experiment. In Ref. [13], the information shows that the gamma counts of ¹⁷²Lu characteristic peaks do not change with and without x-ray irradiation. Therefore, we also suppose that the gamma count rates of ¹⁷²Lu characteristic peaks do not change whether the x-ray beam is turned on or off. Two methods are used to deal with the raw data mentioned above. One is to inspect the change of the gamma count rate ratio between ¹⁷⁸Hf^{m2} and ¹⁷²Lu to identify induced γ emission; the other is to use gamma count rates of characteristic peaks of ¹⁷²Lu and ¹⁷⁵Hf

TABLE V. The ratio of gamma count rate among peaks of ¹⁷²Lu, ¹⁷⁵Hf, and ¹⁷⁸Hf^{m2} isomer.

	Without x-ray irradiation		First x-ray irradiation		Second x-ray irradiation	
	Ratio of count rate	Uncertainty (1σ)	Ratio of count rate	Uncertainty (1σ)	Ratio of count rate	Uncertainty (1σ)
$P_{426.27}/P_{181.04}$	0.0280	3.41%	0.0285	2.86%	0.0285	1.93%
$P_{495.75}/P_{181.04}$	0.0195	1.05%	0.0197	1.28%	0.0199	0.88%
$P_{574.18}/P_{181.04}$	0.0208	0.98%	0.0208	1.17%	0.0205	0.80%
$P_{426.27}/P_{343.15}$	0.0597	3.41%	0.0606	2.87%	0.0604	1.94
$P_{495.75}/P_{343.15}$	0.0416	1.06%	0.0418	1.29%	0.0424	0.89
$P_{574.18}/P_{343.15}$	0.0444	0.99%	0.0441	1.18%	0.0435	0.80
$P_{426.27}/P_{900.64}$	0.0273	3.41%	0.0278	2.86%	0.0277	1.93%
$P_{495,75}/P_{900,64}$	0.0190	1.04%	0.0192	1.27%	0.0194	0.88%
$P_{574.18}/P_{900.64}$	0.0203	3.55%	0.0202	3.09%	0.0200	0.79%
$P_{426.27}/P_{1093.5}$	0.0145	3.41%	0.0147	2.86%	0.0147	1.93%
$P_{495.75}/P_{1093.5}$	0.0101	3.57%	0.0102	3.13%	0.0103	0.88%
$P_{574,18}/P_{1093,5}$	0.0108	1.43%	0.0107	1.72%	0.0106	0.79%

	First x-ra	ay irradiation	Second x-	Second x-ray irradiation		
	Enhancement	Uncertainty (2σ)	Enhancement	Uncertainty (2σ)		
$P_{426,27}/P_{181,04}$	1.88%	8.91%	1.61%	7.84%		
$P_{495.75}/P_{181.04}$	1.0%	3.30%	2.7%	2.74%		
$P_{574.18}/P_{181.04}$	-0.34%	3.06%	-1.57%	2.53%		
$P_{426,27}/P_{343,15}$	1.49%	2.18%	1.17%	7.85%		
$P_{495.75}/P_{343.15}$	0.67%	3.28%	2.03%	2.76%		
$P_{574.18}/P_{343.15}$	-0.72%	3.17%	-1.99%	2.56%		
$P_{426.27}/P_{900.64}$	1.93%	8.91%	1.59%	7.84%		
$P_{495.75}/P_{900.64}$	1.10%	3.28%	2.45%	2.73%		
$P_{574.18}/P_{900.64}$	-0.29%	9.41%	-1.58%	7.27%		
$P_{426,27}/P_{1093,5}$	1.70%	8.90%	1.31%	7.84%		
$P_{495.75}/P_{1093.5}$	0.87%	9.49%	2.17%	7.34%		
$P_{574.18}/P_{1093.5}$	-0.52%	4.47%	-1.85%	3.27%		

TABLE VI. Comparison of the data with and without the x-ray irradiation.

to normalize and calibrate the dead time of measurement and then to determine whether the gain exists or not. Properly speaking, the results of the two methods should be consistent.

1. First processing method-internal standard method

In this method, the peaks with energy 1093.5 and 900.69 keV of 172 Lu and 343.4 keV of 175 Hf are used as the standards. The ratio of gamma count rates between the peaks of standards and the peaks of 178 Hf^{m2} isomer are presented in Table V. Here, the uncertainty is calculated with the following formula (1):

$$s = \sqrt{s_1^2 + s_2^2}.$$
 (1)

 s_1^2 is the uncertainty of gamma count rate of ${}^{178}\mathrm{Hf}^{m2}$ characteristic peak;

 s_2^2 is the uncertainty of gamma count rate of ¹⁷²Lu or ¹⁷⁵Hf characteristic peak.

Based on the data in Table V, the ratios of count rates with and without the x-ray irradiation are compared. The results of comparison are listed in Table VI. These data indicate that there is no significant enhancement at the energy 495-keV line in the confidence interval 95% (2σ) and there is no increase at energy 574-keV peak in the confidence interval of 95% (2σ).

2. Second processing method—dead time calibration method

After balancing the shielding of x rays and detecting of gamma rays, a decision is made that the count rates with the x-ray irradiation can be useful after calibrating the dead time of the measurement. The calibration factor is presented by the formula (2)

$$F = \frac{R_1}{R_2}.$$
 (2)

F is the calibration factor;

 R_1 is the count rate with the x-ray irradiation;

 R_2 is the count rate without the x-ray irradiation.

Select the gamma count rates of gamma peaks with a large branching ratio; take the average as the calibration factor F; then multiply the raw count rates of gamma peaks with energy 426, 495, and 574 keV by F to calculate the new values as the count rates during the x-ray irradiation. Table VII shows the calibration factors of the gamma peaks and the count rates calibrated.

On the basis of the data in Table VII, the count rates with and without the x-ray irradiation for $^{178}\text{Hf}^{m2}$ are compared in Table VIII. The enhancement Δ presents the difference of the count rates of $^{178}\text{Hf}^{m2}$ gamma peaks with and without the x-ray irradiation. As Δ is smaller than zero, there is surely no induced emission. If Δ is larger than zero, but the magnitude of Δ is not beyond the scope of uncertainty with 2σ , then the null

TABLE VII. Calibration factor F and count rates calibrated of 178 Hf^{m2} gamma peaks.

Energy (keV)	F	F Faverage		Energy (keV)	Count rate	
		Value	Uncertainty $(1\sigma, \%)$		Value	Uncertainty $(1\sigma,\%)$
181.038	0.9588	0.9592	0.18	426.27	4.3777	2.86
900.636	0.9583			494.757	3.0220	1.285
1093.498	0.9605			574.179	3.1866	1.17
			Second x-ray irradia	tion		
181.038	0.9513	0.9522	0.12	426.27	4.3635	1.93
900.636	0.9514			494.757	3.0604	0.89
1093.498	0.9541			574.179	3.1476	0.80

Energy (keV)	Fi	rst irradiation	Sec	nd irradiation		
	Δ (%)	Uncertainty (2σ)	Δ (%)	Uncertainty (2σ)		
426.27	1.76	5.89%	1.43	5.33%		
494.757	0.97	3.25%	2.25	2.68%		
574.179	-0.37	3.08%	-1.59	1.77%		

TABLE VIII. The comparison of the count rates with and without the x-ray irradiation for ¹⁷⁸Hf^{m2}.

increment is obtained. Only when Δ is larger than zero and the magnitude of Δ is beyond the scope of uncertainty with 2σ , a conclusion can be drawn that there is indeed emission induced by the low-energy x ray. The following data indicate the results of this experiment.

C. Discussion

From the above two sets of data obtained by applying the two processing methods in two irradiations, it is obvious that the results are consistent with each other: The 574-keV gamma line is not enhanced with the low-energy x-ray irradiation and the 495-keV line shows an increase of 1%–2.3%, but it is still in the range of the statistical fluctuation. Therefore, it should be taken as a null increase.

Since Collins et al. reported the first result of accelerated gamma ray emission from ${}^{178}\text{Hf}^{m2}$ [4], the argument about whether the ¹⁷⁸Hf^{m2} isomer could be induced to emit the gamma ray by the low-energy x ray or not has never stopped. Though some theoretical mechanisms have been put forward, such as nuclear excitation by electron transition and mixing-klevel, it is worth noticing that no nuclear energy level of ¹⁷⁸Hf has been found to support the induced decay phenomenon from the experimental point of view. This group has also calculated some nuclear energy levels with the energy beyond 2.446 MeV. Two discrete levels were simulated and a theoretical perspective on the low-energy induced decay was predicted [19]. The reports of Collins et al. on SR experiments mentioned the enhancements, when 20.825- and 9.567-keV x rays were irradiating [12,15]. What a curious coincidence! It was this coincidence that promoted us to confirm the induced emission by the x ray having energy of 20.826 keV. The result with the null enhancement of this experiment is not surprising. There are two possible reasons. One is that this isomer has not been excited to an upper level (i.e., the intermediate energy level), because there is no level found to satisfy the excited transition

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of ¹⁷⁸Hf^{m2} at all. The other is that ¹⁷⁸Hf^{m2} has been excited to levels, having energy larger than 2.446 keV, with such a small possibility that the increased gamma counts have been very weak and have been buried by a large number of Compton scattered photons. Therefore, no matter which case may be, the zero significant induced gamma ray was detected under the experimental conditions such as magnitude of isomer, x-ray intensity, and measurement efficiency.

IV. CONCLUSION

After the preparation of the sample containing 1.2×10^{11178} Hf^{m2} isomeric nuclei, the induced gamma decay experiment was conducted at Shanghai Synchrotron Radiation Facility. We irradiated the sample with the x-ray energy 20.825 keV, measured the gamma ray, and used an interactive fitting program to obtain the raw data of the gamma energy spectrum of ¹⁷⁸ Hf^{m2}, ¹⁷²Lu, and ¹⁷⁵ Hf. Due to the application of the two sorts of data processing methods, the results are consistent with each other. These results indicated clearly that there was zero enhancement for 495-, 574-, and 426-keV gamma lines of ¹⁷⁸ Hf^{m2} during a 20.825-keV x ray with 10¹² cm⁻² s⁻¹ photon flux irradiation.

ACKNOWLEDGMENTS

We are deeply grateful to the collaborators of BL13W1 Hatch at Shanghai Synchrotron Radiation Facility (SSRF), who ensured the supply of the normal beam by SSRF and helped us conduct our experiment successfully. We express our thanks to Dr. He You for some valuable information about the x-ray beam. This project is supported by the National Natural Science Foundation of China (NSFC) program, Grant No. 11075142.

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