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STUDY OF THE GAMMA EMISSION FROM THE 31-YEAR ISOMER OF ^{178}Hf INDUCED BY *X*-RAY IRRADIATION

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C.B.Collins, F.Davanloo, M.C.Iosif, R.Dussart[1], J.M.Hicks
Center for Quantum Electronics, University of Texas at Dallas, Richardson, Texas, USA

S.A.Karamian[2]
Joint Institute for Nuclear Research, Dubna, 141980, Russia

C.A.Ur, I.I.Popescu
H.Hulubei National Institute of Physics and Nuclear Engineering and IGE Foundation, Bucharest, Romania

V.I.Kirischuk
Scientific Center «Institute for Nuclear Research», Kiev, Ukraine

J.M.Pouvesle
GREMI, CNRS — Universite d'Orleans, Orleans, France

P.McDaniel
Air Force Research Laboratory, DEPA, Kirtland Air Force Base, Albuquerque, New Mexico, USA

C.E.Crist
Sandia National Laboratory, Albuquerque, New Mexico, USA

[1]Present address: GREMI, CNRS — Universite d'Orleans, Orleans, France
[2]Corresponding author E-mail: karamian@cv.jinr.dubna.su

I. INTRODUCTION

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

Proposals to trigger the release of the energy of a nuclear isomer by exciting it to some higher level associated with freely radiating states have been known for over a decade [1]. To be efficient such schemes require the existence at an energy near that of the isomer of a state of mixed–K. It was proposed [1] to use the resonant absorption of x–rays from a bremsstrahlung source to excite some fraction of a high-K isomeric population to the K–mixing level. Then, the decay to the ground state via one or more γ-cascades could subsequently release the total energy of the isomer plus that of the absorbed trigger photon. The types of K–mixing states needed in such schemes to induce the decay of nuclear isomers have been reported [2] in ^{180}Ta and described in ^{174}Hf and other isomers [3].

In 1999 the use of soft x-ray irradiation to accelerate the decay of the ^{178}Hf isomer was reported [4,5]. In that work the continuous x-ray spectrum was most intense in the range of 20 to 50 keV. While the energy of the particular component causing the transition which initiated the process was not determined, the data was analyzed for a hypothesis that the energy lay at the peak of the spectral distribution, 40 keV. For a two step process of the type reported for ^{180}Ta the corresponding value of integrated cross section ($\sigma\Gamma$) in the case of the ^{178}Hf would be about 1 x 10^{-21} cm^2 keV. At such low energies, the perceptions of Mossbauer spectroscopy may be followed to yield an expected photon-absorption cross section and resonance shape given by the Breit-Wigner expression. At 20 keV a $\sigma\Gamma \approx 10^{-21}$ cm^2 keV would suggest a width of about 0.5 eV. This is superficially similar to the width deduced for the deexcitation of ^{180}Ta induced by photons.

The systematics for the nuclear transition strengths do not predict such large widths corresponding to short lifetimes ($\sim 10^{-15}$ s) as being typical for nuclear levels at $E^* =$ 2 – 3 MeV. Recommended calculations of the reduced transition probabilities, B(E1) and B(E2), give $\sigma\Gamma \approx 10^{-27}$ cm^2 keV for ^{180}Ta, a prediction two orders of magnitude lower than the experimental value [2]. The corresponding disagreement is even more impressive in the ^{178}Hf case, mainly because of the paucity of levels with appropriate quantum numbers for the decay of the high-spin intermediate state excited after the absorption of a photon by the $^{178}Hf^{m2}$ isomer. As a consequence, the properties of intermediate K-mixing states appear as an extraordinary issue which naturally excites more focused studies. In particular, more details are necessary on the acceleration of the decay of the $^{178}Hf^{m2}$ isomer in order to clarify the mechanism of the x-ray induced de-excitation of the isomer. By focusing upon a confirmation of the work previously reported [4,5], while extending it to include a study of a fragment of a cascade not present in the spontaneous decay of the isomer; the present work is aimed at meeting these requirements.

II. EXPERIMENTAL DETAILS

The experimental work was performed at the Center for Quantum Electronics, University of Texas at Dallas. The irradiating beam consisted of the bremsstrahlung radiation provided by an x–ray unit operated at 15 mA and an end point energy up to 90 keV. The device was operated in a way that ensured a duty cycle for the irradiation of about 0.6%.

The irradiated sample consisted of a sealed plastic target containing 6.3×10^{14} $^{178}Hf^{m2}$ isomeric nuclei placed in a 1 cm diameter well. The main radiative contaminants in the sample were the ^{172}Hf nucleus and its daughters at a level comparable to the intensity of the

31-year spontaneous decay of the ^{178}Hf isomer. The sample was placed at 5.5 cm distance from the emission point of the x-rays and the only absorption was due to the glass of the x-ray tube and the 2 mm plastic sealing of the x-ray device. The absorption in the sealing of the sample was negligible. .

In Fig. 1 we show the experimental arrangement. The 10% coaxial Ge detector was placed at 41.6 cm distance from the irradiated sample. A shielding built of 3 mm of Cu and 2 mm of Pb was used in order to prevent detection of scattered x-rays in the Ge detector. Thus, the rate of scattered x-rays allowed to get into the detector was measured to be about 1.5 kHz. The counting rate produced by the radiative target in the absence of the x-rays was about 4 kHz. Such an experimental arrangement resulted in a low value of the total dead-time of the acquisition chain of about 9% during the irradiation. The energy and efficiency calibration of the Ge detector was done using standard calibration sources, ^{60}Co, ^{133}Ba and ^{137}Cs. We estimated the maximum absolute detection efficiency of about 1.5 x 10^{-4} to correspond to a gamma-ray energy of 300 keV.

Data acquisition was enabled only when a signal provided by a *p-i-n* diode which monitored the x-ray beam was present. The signal from the diode was processed in order to produce a 4 ms gate centered on the intensity distribution of the x-ray signal. We recorded γ-ray energies up to 2 MeV with amplification set to give 0.25 keV/channel, allowing for a good analysis of the possible contamination of the lines of interest. Spectra have been stored each three hours and gain matched using internal γ-lines before adding them.

Special attention was devoted to the measurement of the x-ray flux emitted by the x-ray device. For this purpose we have placed the Ge detector at 8.2 m distance from the x-ray device. The direct x-ray flux could reach the active area of the detector only through a 1 mm diameter hole placed at 0.5 cm from the detector. High counting rates still required the application of absorbers. From a few measurements with different absorbers the final spectra could be reconstructed. They are shown in Fig. 2 for two conditions, with and without a 2.7 mm equivalent thickness of Al available for covering the output window. In the insert the spectrum taken with 1.5 mm Cu absorber is given. It was one of the component measurements from which the composite spectrum was assembled. In previous work [4,5] the Al was present, while in the experiment reported here it had been removed. One can see in curve a) of Fig. 2 that there is a high intensity, above 4 x 10^{11} photons cm^{-2} keV^{-1} s^{-1} at low energies near 20 keV; drastic decrease of the intensity to higher E_x values; and a clear manifestation of the characteristic K-X lines of W. All these peculiarities of the incident radiation are important for the conclusive definition of the cross section integral, as discussed below.

III. RESULTS

Acquired with the 10% coaxial Ge detector, spectra of the induced emission of γ-radiation generally resembled those obtained in the earlier work [4,5]. However, in the present experiment normalization of the spectra taken with and without x-ray irradiation was facilitated by the deliberate inclusion of lines from the ^{133}Ba source proximate to the Hf target, but not irradiated. Those fiducial lines were within about 30 keV of the 325.5 keV ($6^+ \rightarrow 4^+$) component of the ground state band, GSB, and so reduced any effects of drift or nonlinear dependence of efficiency upon energy. In practice the data were acquired so that the total numbers of photons collected in the 356.0 keV line of ^{133}Ba were as nearly equal as could be arranged. An empty target holder, the "blank," of similar mass and construction to the one carrying the isomeric nuclei was available for use. Comparative measurements showed that over 95% of the elastic and inelastic scattering of the irradiating beam arose from the mass of the holder and not its contents.

Three geometric arangements were important during the collection of data: 1) "Inbeam" in which the isomeric target was centered in the cone of irradiation as shown in Fig. 1, 2) "Outbeam" in which the target was placed out of the beam of x-rays at the position denoted as "Proximate" in Fig. 1 and the "blank" target holder replaced it in the cone of irradiation, and 3) Baseline in which the isomeric target was placed in the position of the cone of irradiation, but the x-ray source was turned off. During analysis both inbeam and outbeam spectra were scaled to the baseline spectrum which was collected during a subsequent 1 h period uninterrupted by the switching of the x-rays, so that the areas of the fiducial line in each spectra were the same.

Fig. 3 shows the results of 16 h of acquisition time, during which there were 340 sec of actual counting time enabled by the gate coincident with the detection of x-rays. From top to bottom are shown the inbeam, outbeam, and baseline spectra. The counts in the areas under the relevant peaks are summarized in Table I and Table II.

Because of the difficulties in establishing precise fiducial lines in other regions of the spectra, differences in areas under peaks could not be attained with the same high level of confidence as accomplished for the 325.5 keV line. Impressions generally followed previous reports [4,5] suggesting that not all components of the spontaneous decay cascade were enhanced by the x-rays. If several of the transitions feeding the GSB in spontaneous decay are not enhanced, the question naturally arises as to the channels followed by the induced decay of the isomer. A search through the spectra suggested several "new" components tending to confirm observations made upon data from our 1998 experiment. Fig. 4 shows the best of such lines observed in the experiment reported here. The line at 210.6 keV is close to a member of the $K^\pi = 6^+$ band known to ^{178}Hf but not previously seen in spontaneous decay of the isomer. In a baseline spectrum this line is manifested with some shifting and with relative intensity at least 4 times smaller [6]. In that case it corresponds to a known ^{172}Lu line both in energy and relative intensity. In agreement with the previous suggestions, the lines corresponding to other transitions, feeding and fed, by the 210.6 keV line may be present in our data, but statistics are not sufficient to confirm identity.

IV. DISCUSSIONS AND CONCLUSION

The first reports [4,5] of the great faculty of low energy x-rays for inducing the decay of the $^{178}\mathrm{Hf}^{m2}$ isomer, stressed all models which might have been able to predict such an effect. The problem has not been resolved by the results of the experiment reported here.

However, some illumination of the unexpected nature of the phenomenon has been realized. Because it tends to bring a convergence with systematics of the nuclear transition strengths, it is instructive to examine the details.

Of first importance is that the general phenomenology has been reproduced in accordance with Ref. [4,5] . Table I shows that with the same type of small x-ray generator traditionally used in dental medicine enhancements of the order of 2 % can be induced in the rate of spontaneous decay of the Hf isomer. From Table II it is clear that there is no comparable value of spurious enhancement found when irradiating the empty target holder with the $^{178}Hf^{m2}$ isomer in the outbeam geometry. The excess counts in the 325.5 keV line are essentially zero, as can be seen as well in Fig. 3.

The yield of triggering events would equal the product of the number of isomeric atoms in a target, the spectral flux density from Fig. 2 at the appropriate energy, and the unknown integrated cross section, $\sigma\Gamma$, for the branch of the excitation of a K–mixing level that ends in a state other than that of the initial isomer. Since each quantity is known except for the integrated cross section for the "triggering branch," that cross section can be obtained if the transition energy is estimated. Possible values of $\sigma\Gamma$ are summarized in Table III. The specific value of $\sigma\Gamma$ is strongly dependent on the position of an intermediate level because of the strong variation of the x-ray flux with energy. Assuming the resonance band lies near the maxima of flux, one can deduce $\sigma\Gamma \approx (2 - 3) \times 10^{-23}$ cm^2 keV. The emission lines of W were detected in the x-ray spectrum with resolution of 0.9 keV. The natural width is much smaller, and, in reality, flux is respectively higher in the characteristic X-ray peaks.

For the case of isomeric ^{178}Hf we have confirmed that the irradiation by x–ray photons with the energy of the order of 20-60 keV can induce the prompt release of the 2.446 MeV stored by the isomer into freely radiating states. This is an energy gain of about 60.

Further research is needed to provide greater precision to the measurements of the transition energy to the K–mixing level and to clarify properties of the cascade feeding the GSB. Such data will then facilitate a better understanding of these first evidences of the triggering of induced gamma emission from the 31–year isomer of ^{178}Hf with very low energy x–ray photons through large cross sections, $\sigma\Gamma \geq 2 \times 10^{-23}$ cm^2 keV.

TABLES

TABLE I. Comparison between the gamma emission from the target irradiated in the inbeam position with the baseline obtained without irradiation. The two sets of data were normalized according to the ratios of the areas of the 356.0 keV ^{133}Ba line in the two cases as detailed in the text.

Eγ	Nucleus	With irradiation	Baseline		Excess
(keV)		Area	Area	Norm.[a] Area	(counts)
296.8	^{178}Hf	1692 (68)	14925 (201)	1685 (27)	7 (73)
302.9	^{133}Ba	6503 (103)	58052 (304)	6554 (64)	-51 (121)
323.9	^{172}Lu	1403 (71)	12891 (222)	1455 (28)	-52 (76)
325.5	^{178}Hf	17035 (146)	147952 (451)	16703 (148)	332 (208)[b]
356.0	^{133}Ba	22617 (180)	200333 (482)	Normalizing line	
372+373	^{172}Lu	2439 (84)	21860 (250)	2468 (90)	-29 (123)
377.5	^{172}Lu	3098 (75)	27445 (225)	3098 (36)	0 (83)
383.8	^{133}Ba	3225 (76)	28160 (261)	3179 (40)	46 (86)

[a]Normalized to the 356 keV line of ^{133}Ba

[b]Estimation of the effect: 2.0 ± 1.2%

TABLE II. Comparison between the gamma emission acquired from the target in the outbeam geometry with the baseline obtained without irradiation. The two sets of data were normalized according to the ratios of the areas of the 356.0 keV ^{133}Ba line in the two cases as detailed in the text.

Eγ (keV)	Nucleus	Outbeam Area	Baseline Area	Baseline Norm.[a] Area	Excess (counts)
296.8	^{178}Hf	785 (52)	20826 (262)	832 (14)	-47 (54)
302.9	^{133}Ba	3021 (70)	75754 (352)	3028 (36)	-7 (79)
323.9	^{172}Lu	722 (49)	17737 (250)	709 (13)	13 (51)
325.5	^{178}Hf	7961 (103)	199062 (517)	7957 (88)	4 (135)
356.0	^{133}Ba	10408 (110)	260372 (551)	Normalizing line	
372+373	^{172}Lu	1258 (57)	29652 (284)	1185 (55)	73 (79)
377.5	^{172}Lu	1428 (51)	36429 (260)	1456 (19)	-28 (54)
383.8	^{133}Ba	1509 (52)	36928 (261)	1476 (19)	33 (55)

[a]Normalized to the 356 keV line of ^{133}Ba

TABLE III. Calculations of the integrated cross section, $\sigma\Gamma$ from 2% enhancement of the ^{178}Hf^{m2} line of 325.5 keV

Ex(keV)	20	30	40	50	$K\alpha_1$(W)[a]	60	$K\beta_1$(W)[a]	70
$\sigma\Gamma$(cm^{2}keV) Flux with no filter	3.2x10^{-23}	5.4x10^{-23}	1.6x10^{-22}	3.4x10^{-22}	1.6x10^{-23}	3.9x10^{-22}	3.3x10^{-23}	1.1x10^{-21}
$\sigma\Gamma$(cm^{2}keV) Flux with 2.7 mm Al filter	3.5x10^{-22}	1.2x10^{-22}	2.4x10^{-22} [b]	4.5x10^{-22}	2.0x10^{-23}	4.7x10^{-22}	4.0x10^{-23}	1.3x10^{-21}

[a]For width 50 eV.

[b]For comparison with Ref. [5]

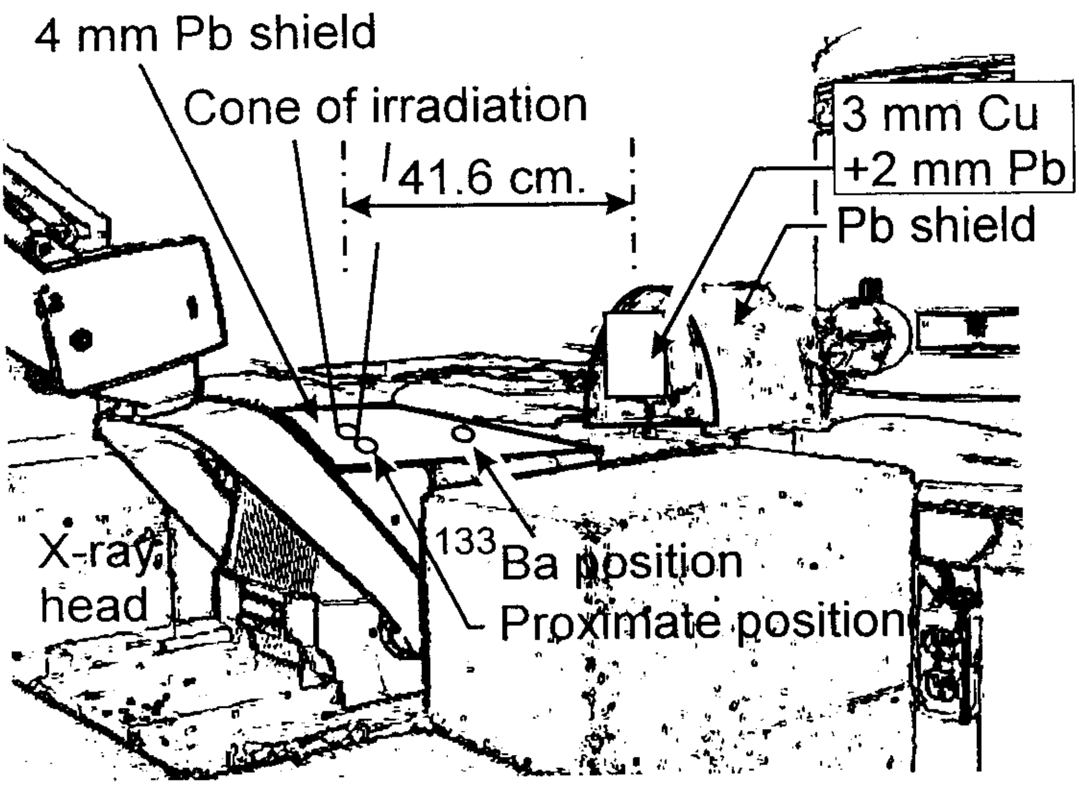

Fig. 1- Schematic drawing of the experimental arrangement showing the geometric placements and dimensions of the components.

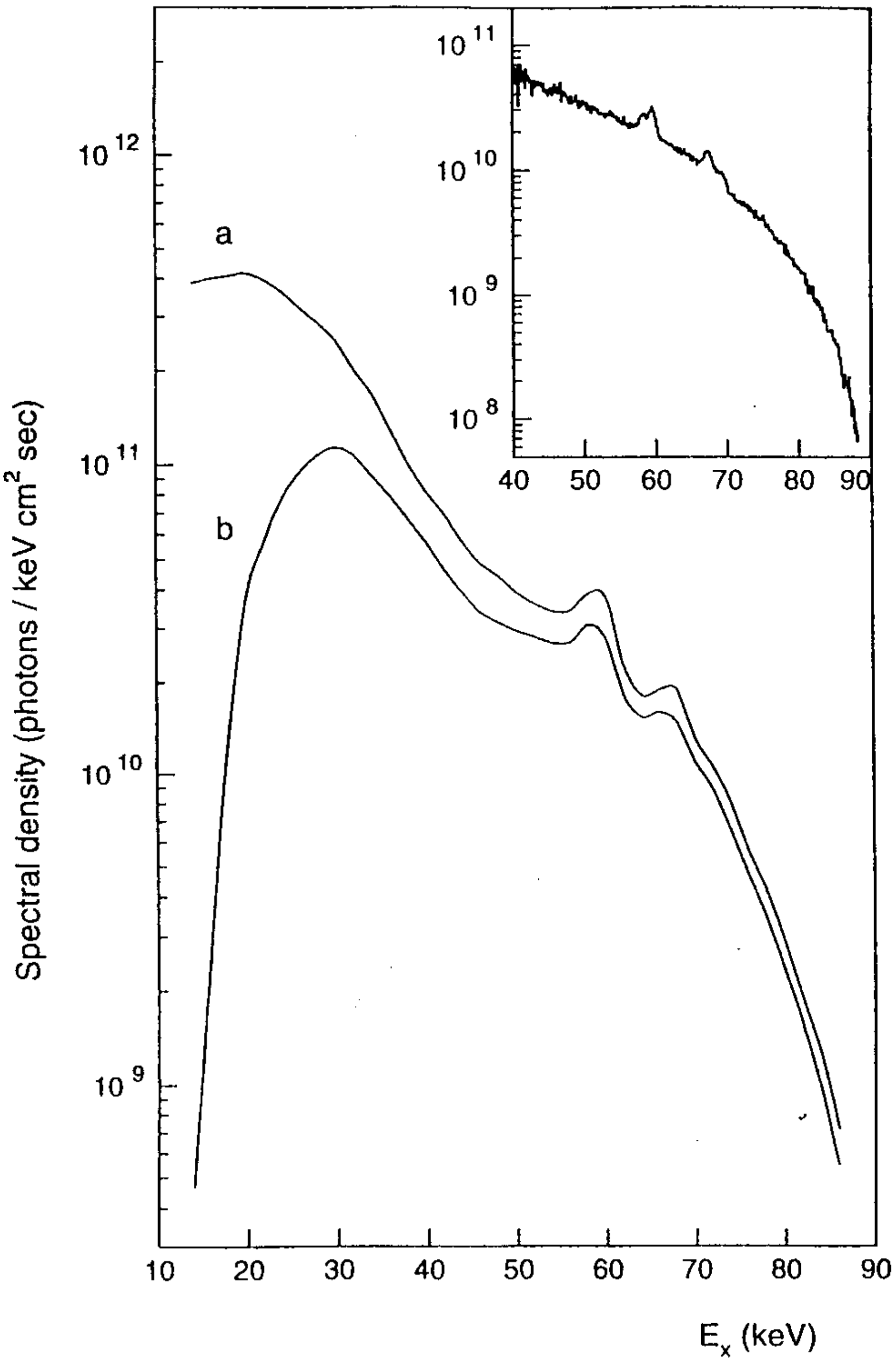

Fig. 2- Graph of the x–ray spectral flux expected at the inbeam position of the isomeric target. These experimental data for a 90 keV endpoint were measured using a Ge detector from input attenuated with a pin hole and placed at 8.2 m distance from the x-ray tube. Spectra of irradiation correspond to: a) this experiment, and b) the measurements of Ref. [4,5] which used the Al absorber on the output. The insert shows the raw data taken with a 1.5 mm Cu absorber.

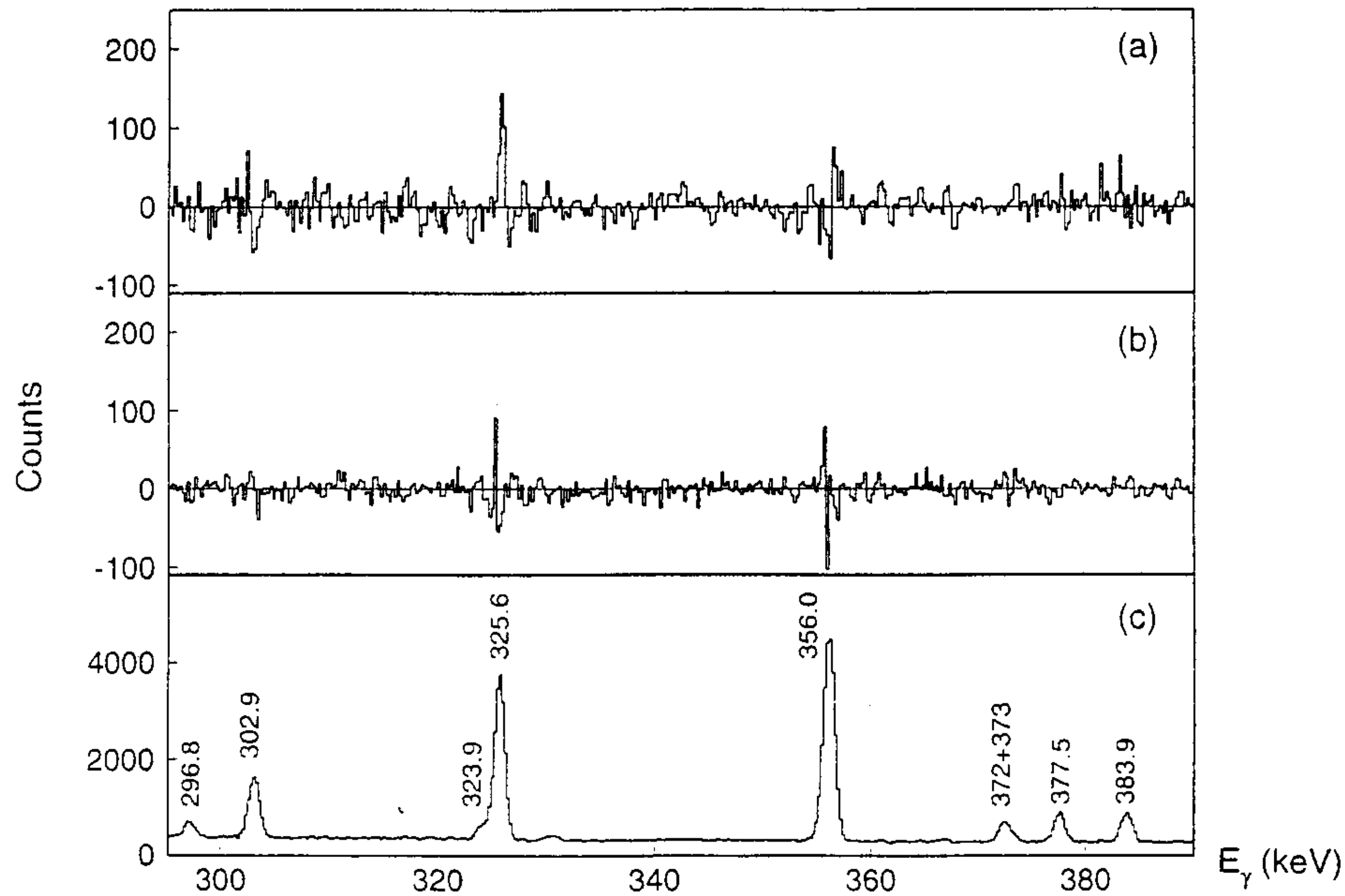

Fig. 3- Plot of the differences in counts a) and b) obtained in the spectrum of the ^{178}Hf from the isomeric target in comparison to baseline data shown as c). The region shown could be dependably normalized by comparing the areas under the 356.0 keV line of ^{133}Ba and includes the 325.5 keV ($6^+ \rightarrow 4^+$) component of the GSB of ^{178}Hf. The spectra are: a) target inbeam minus scaled baseline and b) target outbeam minus scaled baseline.

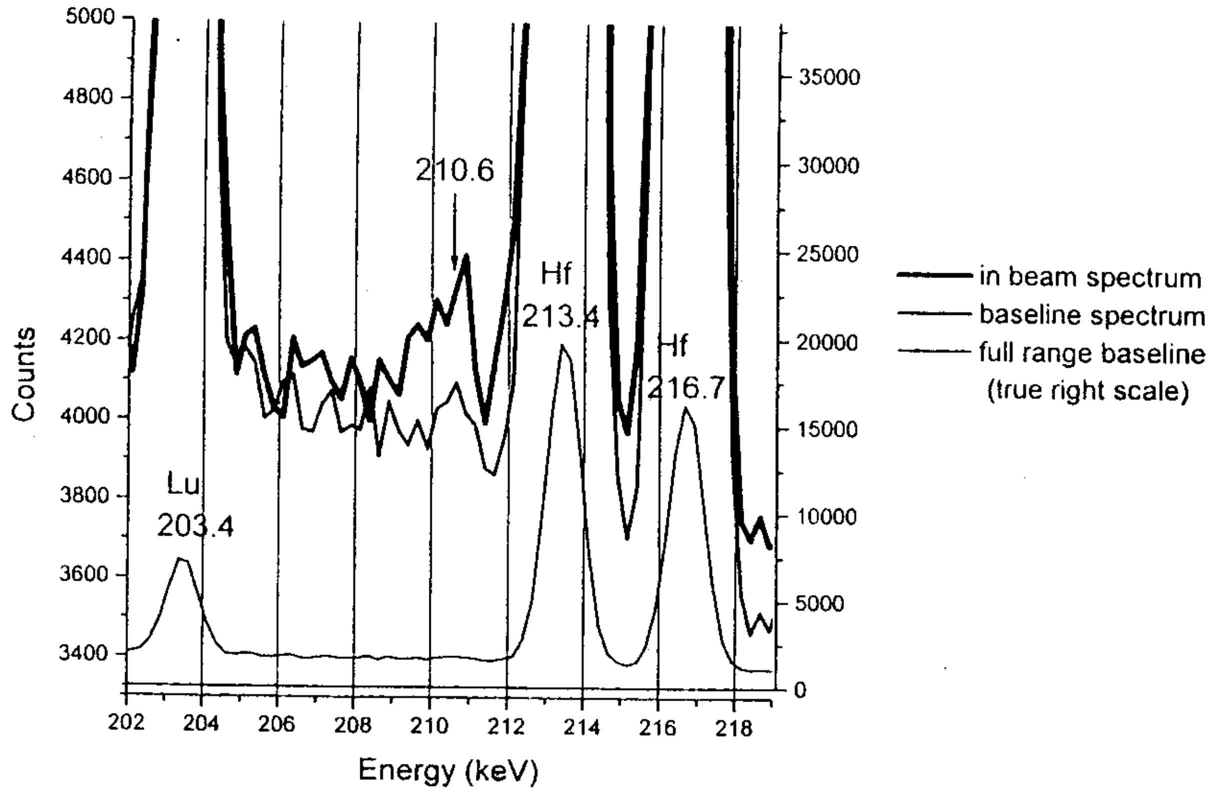

Fig. 4- Graph showing a comparison near 210 keV of the spectra obtained at the different geometries. The FWHM of the marked peaks are from left to right: 1.10, 0.91, 1.06, and 1.04 keV, respectively.

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Коллинз К.Б. и др. E15-99-224

Изучение гамма-эмиссии изомера ^{178}Hf ($T_{1/2}=31$ год), индуцированной рентгеновским излучением

Образец, содержащий $6{,}3\times10^{14}$ ядер изомера ^{178}Hf (имеющего спин 16^{+}, период полураспада 31 год и энергию возбуждения 2,446 МэВ), облучали рентгеновскими импульсами. Для генерации излучения с граничной энергией 90 кэВ использовалась установка, работающая при токе 15 mA. С помощью Ge-детектора измеряли спектры гамма-активности изомерной мишени. Было обнаружено, что интенсивность перехода $6^{+}\rightarrow4^{+}$ (325,5 кэВ) полосы основного состояния ^{178}Hf увеличивается на 2 %. Такое ускорение распада изомера ^{178}Hf соответствует интегральному сечению около 3×10^{-23} см$^{2}\cdot$кэВ, если предположить, что резонансное поглощение происходит при энергии вблизи максимума потока рентгеновского излучения, т.е. около 20 кэВ, или при энергиях, совпадающих с эмиссионными K-линиями W.

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Study of the Gamma Emission from the 31-Year Isomer of ^{178}Hf Induced by X-Ray Irradiation

A sample containing 6.3×10^{14} nuclei of the 16^{+} isomer of ^{178}Hf having a half-life of 31 years and excitation energy of 2.446 MeV was irradiated with x-ray pulses derived from a device operated at 15 mA to produce bremsstrahlung radiation with an end point energy set to be 90 keV. Gamma-spectra of the isomeric target were taken with a Ge-detector. Intensity of the 325.5 keV ($6^{+}\rightarrow4^{+}$) transition in the ground state band of ^{178}Hf was found to increase by about 2 %. Such an accelerated decay of the ^{178}Hf isomer is consistent with an integrated cross section of 3×10^{-23} cm$^{2}\cdot$keV if the resonant absorption takes place within the energy ranges corresponding to the maxima of the x-ray flux, either near 20 keV, or at the K emission lines of W.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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