Photoexcitation of $^{189}Os^m$ and $^{193}Ir^m$. II. Excitation by ^{137}Cs and ^{60}Co γ rays

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Photoexcitation of 5.84 h $^{189}Os^m$ and 10.6 d $^{193}Ir^m$ was observed using high activity ^{137}Cs and ^{60}Co sources for irradiation. The highly converted weak isomeric transitions were detected by large area 2π multiwire proportional chambers. Determining resonance photon flux densities with Monte Carlo simulation, isomer excitation cross sections integrated over identified activation levels were evaluated and level half-lives were derived.

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I. INTRODUCTION

If a nuclear isomer is excited via (γ,γ') reaction, a higher lying (activation) level (AL) is populated in the first step, which subsequently feeds the metastable state. Isomers can be photoexcited using bremsstrahlung from electron beams or radioisotopes for irradiation, among which ^{60}Co has been used most extensively [1]. The AL's are populated due to resonance photoabsorption of a very narrow band in the degraded spectrum of a high activity source. Many stable nuclides have been investigated [1,2], the first principal AL's of which are lying in the energy range of 1–2.75 MeV, corresponding to the primary energies of the exciting sources. However, the role of the excitations of even lower energies is stressed in some applications, e.g., in nuclear astrophysics [3] or gamma-laser research [4]. Although isomer excitation by (γ,γ') reaction using low-energy bremsstrahlung has long been observed [5], the first photoexcitation experiments on ^{189}Os and ^{193}Ir by ^{137}Cs, a source of 661.66 keV primary energy, were reported just lately [6], which was followed by the excitation of $^{77}Se^m$, $^{79}Br^m$, and $^{191}Ir^m$ by the same source [7]. As a result, AL's were identified and it became possible to determine unknown level lifetimes.

Photoactivation studies on ^{189}Os started with the search for the effect of nuclear excitation by electron transition (NEET) [8]. Bombarding with x rays [9] and synchrotron radiation [10], the excitation of the $^{189}Os^m$ isomer was observed and attributed to the population of the 69.54 keV level via the NEET effect. By irradiating with a 200 kV x-ray generator, we also found very weak $^{189}Os^m$ radioactivity in a preliminary study [11], using a planar Ge detector to count L x rays accompanying the strongly converted $M3$ isomeric transition. Using an x-ray generator with accelerating potentials up to 300 keV and observing internal conversion electrons, we obtained more conclusive results in Ref. [6] and in paper I of this work (Ref. [16]). However, contributions from competing resonance scattering and NEET could not be separated, and the existence of NEET remained unconfirmed. In those experiments we excited also higher lying AL's via resonance scattering, and a level half-life was determined, while the excitation of $^{189}Os^m$ via AL's lying above 300 keV was left to be studied with higher energy sources in experiments accounted for in the present paper.

In what follows, we are reporting on photoexcitation of $^{189}Os^m$ and $^{193}Ir^m$, having used ^{137}Cs and ^{60}Co irradiating sources. In the case of ^{193}Ir, the photoexcitation has been assumed to proceed only via resonance scattering, populating the metastable state at 80.28 keV. $^{193}Ir^m$ had only been photoexcited previously by irradiation with a 2.75 MeV energy ^{24}Na source [2], to our knowledge.

II. EXPERIMENTS

The ^{137}Cs source used was of 67.39 TBq (1820 Ci) activity. Its main dimensions were described in an earlier paper [7]. Metallic powder Os samples of 4.38 g mass pressed into pellet form of 20 mm diameter by 3.5 mm length, and a 1.75 g Ir metal plate of $25\times25\times0.125$ mm size were irradiated in the arrangement reported previously [7], but without the pneumatic rabbit system used only there for short-lived isomers.

Os and Ir powder samples of 1–1.5 g mass were irradiated in the field of a cage type, 2.78 PBq (75 kCi) activity ^{60}Co facility at the Institute of Isotopes. Irradiation and measurement periods were 10–67 h for the Os and 3–12 d for the Ir sample, respectively.

Because of the weak isomeric activities, sensitive counting techniques were developed. Due to the low transition energies and the high multipole orders, both isomeric transitions are practically fully converted. A 90 cm^2 sensitive area flat 2π multiwire proportional counter [6] was used for measuring about 20 keV electrons from the 5.8 h half-life, 30.8 keV energy isomeric transition of $^{189}Os^m$, and a smaller, 6.2 cm^2 sensitive area flat 2π proportional counter [6] was used for measuring electrons mostly of about 70 keV energy from the 10.6 d half-life, 80.28 keV energy isomeric transition of $^{193}Ir^m$. After irradiations, the Os samples were dismounted and their content was evenly dispersed over the brass cathode plate serving as sample holder at the same time, while the Ir plate was inserted in the small counter. After the extinction of isomeric activities, the samples were left in the counters for background measurements because of the sensitivity of the counters to Os and Ir K x rays excited by environmental background radiation.

When using the large 2π counter, a sensitivity increase of about 140 was attained for $^{189}Os^m$ measurement against a

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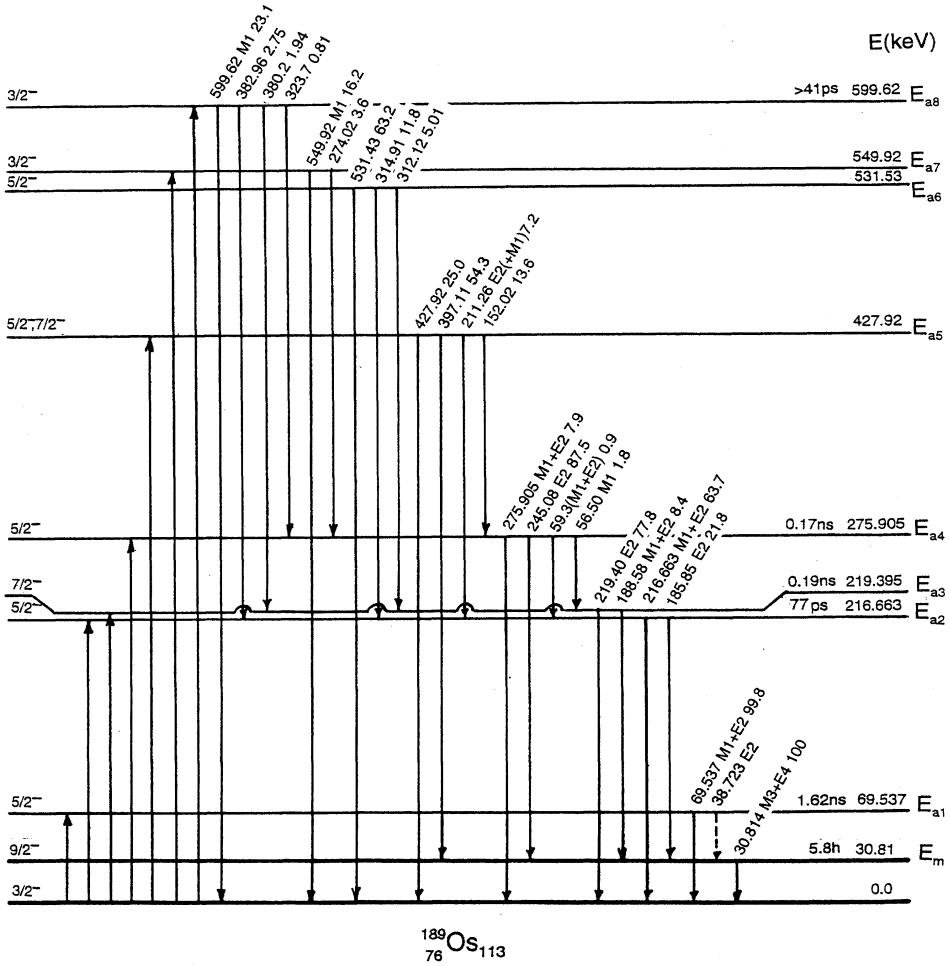

FIG. 1. Part of the ^{189}Os level structure, relevant for photoexcitation by ^{137}Cs γ rays. Activation levels E_{ai} and transitions feeding the metastable state E_m are only indicated. Levels and level parameters are taken from Refs. [12,13] and paper I (Ref. [16]). Branching ratios are in percent.

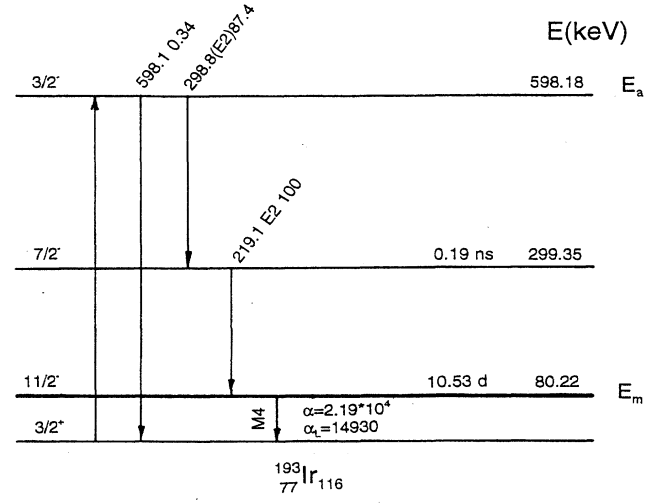

FIG. 2. Partial level diagram of ^{193}Ir [14]. A single activation level can be identified up to 662 keV.

low-energy planar Ge detector. The figure was about 25 for $^{193}Ir^{m}$ measurements in the small counter.

III. DATA ANALYSIS

The isomer activity induced by resonance scattering can be written as

$$Y = f_r(E_a)\sigma_m , \tag{1}$$

where $f_r(E_a)$ denotes the spectral density of the irradiating photon flux taken at a single AL energy E_a. The isomer excitation cross section integrated over this level, σ_m, is provided by integrating the Breit-Wigner single level formula as given in Eq. (2) of Ref. [16].

The relevant detail of the ^{189}Os level diagram up to 662 keV, the primary energy of ^{137}Cs γ rays, is shown in Fig. 1 [12]. Eight levels have been identified to be AL's, from where there are direct transitions to the ground state, and direct or cascade transitions to the isomer. Transition and level parameters (spin parity I^{π}, B_0, B_m, $t_{1/2}$) according to Ref. [12] are indicated in the figure. From Ref. [13], $I^{\pi}=5/2^{-}$ is adopted for the 216.66 keV level instead of $7/2^{-}$, [12] and the AL itself at 531.5 keV and its parameters are also taken from there [13].

TABLE I. Results of ^{137}Cs irradiations. Integrated isomer production cross sections, σ_m were determined either from adopted nuclear data or from measured isomer (partial) yields Y and calculated resonance flux densities f_r. The uncertainties (in parentheses) do not contain those of the literature data. Level half-lives were taken from the literature and/or determined from the results for σ_m. The partial yields at 216.7, 219.4, 275.9, and 599.6 keV were determined multiplying σ_m by f_r. Subtracting their sum from the total yield, the remainder was attributed to the unseparated effect of AL's at 427.9, 531.5, and 549.9 keV, one by one. For this reason, only upper limits for σ_m and lower limits for $t_{1/2}$ values of unresolved ^{189}Os levels can be given. $t_{1/2}$=half-life of the activation levels (10^{-10} s), $g=(2I_a+1)/(2I_g+1)$; I_a, I_g spin of the activation and ground level, respectively.

Nuclide	Activation level (keV)	Yield (Bq/mol)	f_r (10^5 cm^{-2} s^{-1} eV^{-1})	σ_m (mb eV) From level scheme	Ref.	This work	Partial yield (Bq/mol)	Level half-life (10^{-12} s) Literature	Ref.	This work
^{189}Os	216.7	1.12(0.04)×10^5	$2.51^{+1.23}_{-0.22}$	81.0(9.0)	[16],[13]		12 243(5998)	77(10)	[16]	
	219.4		$2.64^{+1.25}_{-0.25}$	20.1	[12]		3148(1491)	190	[12]	
				19.5	[13]					
	275.9		$3.82^{+1.84}_{-0.35}$	12.0	[12]		2772(1335)	170	[12]	
				12.1	[13]					
	427.9		$3.28^{+1.58}_{-0.33}$	$15.9g/t_{1/2}$	[12]	<536	93 782(7482)			>4.4(I_a=5/2)
										>5.9(I_a=7/2)
	531.5		$2.57^{+0.92}_{-0.25}$	$1.76/t_{1/2}$	[13]	<682				>0.26
	549.9		$2.43^{+0.90}_{-0.24}$	$2.285/t_{1/2}$	[12]	<722				>0.039
	599.6		1.88(0.25)	< 0.39	[12]		< 55	> 41	[12]	
				< 0.49	[13]					
^{193}Ir	598.2	673± 291	2.17(0.30)	$0.145/t_{1/2}$	[14]	5.15(2.5)				$2.8^{+2.8}_{-0.9}$

Below 662 keV, a single AL at 598.18 keV can be established for ^{193}Ir, as seen in the level scheme [14] in Fig. 2, for which no half-life data are available in the literature.

The population of ^{189}Os^m due to the first AL at 69.54 keV is very low. It should proceed via the 38.7 keV $E2$ transition never observed directly (indicated by dashed line in Fig. 1), postulated only owing to successful isomer activation experiments [8–10]. This level, no matter whether resonance scattering or NEET predominates in its population (see Ref. [16]), has been left out from further considerations, because of its low cross section (about two orders of magnitude less than the cross section for the next AL at 216.7 keV). Moreover, the intensity of 69.54 keV energy photons in the Compton continuum is also low, since a primary energy photon cannot be scattered to this energy in a single step.

For determination of the number of photons scattered to AL energies in the source, its cladding, the environment, and the target itself, a Monte Carlo code was developed [15]. The number of simulated particles were selected so (typically about 10 000) that the statistical error of the average fluence was less than 3%. In calculating electron densities, the actual values of the porous and plate target materials were correspondingly considered. Resonance flux densities were determined (Table I) assuming single scattering. Allowance for double scattering [7] was included in the error estimates of the values given in the table. It is worth noting that the resonance flux densities calculated on the basis of an approximate analytical formula [7] agree with the present results within 20%. The integrated isomer excitation cross sections for the AL's have also been calculated, using the available level parameters from the literature.

Since higher lying AL's and depopulating cascades cannot be identified, from ^{60}Co irradiations it is possible to derive only effective cross sections $\langle\sigma\rangle$, i.e., isomer yields related to the primary flux.

IV. RESULTS AND DISCUSSION

The results of the measurements are summarized in Tables I and II. Isomer excitation cross sections integrated over single activation levels, $\sigma_m - s$, were derived from measurements using ^{137}Cs source for irradiation, and from known level parameters. From ^{189}Os level half-lives available in the literature, we obtained σ_m values given in Table I. For the level at 216.7 keV, we determined σ_m from the x-ray experiment (Ref. [16]). Multiplying these by the correspond-

TABLE II. Yields and effective cross sections $\langle\sigma\rangle$ for isomer production by irradiation with ^{137}Cs and ^{60}Co sources. ^{115}In^m data are also given for comparison.

Isomer	^{137}Cs irradiation Yield (Bq/mol)	$\langle\sigma\rangle$(μb)	^{60}Co irradiation Yield (Bq/mol)	$\langle\sigma\rangle$(μb)
^{189}Os^m	(1.12±0.04)×10^5	1.00±0.20	(6.64±0.67)×10^5	2.69±0.54
^{193}Ir^m	673±291	(4.9±2.3)×10^{-3}	(2.34±0.12)×10^4	0.107±0.009
^{115}In^m			(1.73±0.10)×10^4	0.079±0.006

ing resonance flux densities, individual contributions resulting from the population of these levels were separated. The sum of the partial yields thus obtained was subtracted from the total isomer production, and the rest was attributed to the excitation of the remaining three levels, for which there were no literature half-life data available. Since it was not possible to resolve the individual effects of these levels, the residual yield as a whole was attributed alternately to them, as if each were alone responsible for it. Dividing then this figure by the respective resonance flux values, one by one, upper limits of σ_m were obtained for the AL's at 427.9, 531.5, and 549.9 keV. Corresponding lower limits for the respective half-lives are also indicated in the table. In order to resolve the unseparated effect of these levels, a variable energy electron accelerator could be used.

In the case of ^{193}Ir, the total isomer yield was ascribed to the excitation of the single AL at 598.2 keV, the half-life of which was determined from the measurement as about 3 ps.

In Table II, we summarized the results of the measurements using the ^{60}Co source. Above 662 keV up to 1.33 MeV, the higher primary energy of ^{60}Co, there are 26 known levels in the case of ^{189}Os, all of them with unknown half-lives, and other parameters are also unknown for most of them. The case is similar for ^{193}Ir, where the number of levels between 0.662 and 1.33 MeV is 35. For this reason, effective cross sections were only determined for excitation by ^{60}Co. The respective quantities for ^{137}Cs irradiation were also evaluated and given in the table, together with results for $^{115}In^m$ activation for comparison. It is worth noting that — in contrast to ^{189}Os and ^{193}Ir — there is only one principal AL for photoactivation of ^{115}In up to 1.33 MeV, corresponding to its much less total number (12) of levels.

Among the nuclides in Table II, ^{189}Os exhibits the largest cross sections. This fact seems to be in accordance with its high level density and relatively low spin difference $\Delta I=3$ between the ground and isomeric states, as compared to $\Delta I=4$ for ^{193}Ir and ^{115}In.

Summing up, we have measured the photoexcitation of $^{189}Os^m$ and $^{193}Ir^m$ isomers by multicurie ^{137}Cs and ^{60}Co sources for the first time, and have shown that this method may represent a tool for providing important spectroscopic information. Due to sensitive measurement techniques for counting low energy, weak isomeric activities, determination of unknown parameters of low-lying, narrow levels has proven to be possible, which can hardly be determined by other means. Photoexcitation of $^{189}Os^m$ displays the largest cross sections ever obtained with ^{137}Cs and ^{60}Co irradiations. For further studies, the use of low energy, high current accelerators or energetic synchrotron radiation is recommendable.

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