Excitation of $^{77}Se^m$, $^{79}Br^m$, and $^{191}Ir^m$ by ^{137}Cs γ rays

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(Received 20 January 1994; revised manuscript received 9 December 1994)

Excitation of the 17.4 s $^{77}Se^m$, 4.9 s $^{79}Br^m$, and 4.9 s $^{191}Ir^m$ isomers by γ rays of a strong ^{137}Cs source was observed for the first time. Using a 67 TBq (1800 Ci) source for irradiation, isomeric activities were measured by means of γ spectrometry. Identifying principal activation levels, photoexcitation cross sections were determined, allowing level lifetimes to be deduced.

PACS number(s): 21.10.Tg, 23.20.Lv, 27.50.+e, 27.80.+w

I. INTRODUCTION

Nuclear isomers can be produced via the (γ,γ') reaction by populating first a higher-lying (short-lived), so-called activation level, which subsequently decays to the metastable state. In the extensive studies carried out in the last 40 years on a number of stable nuclides having long-lived metastable states, photoexcitation of this type has been observed, using—not considering bremsstrahlung of electron accelerators for this time—high-activity radioisotopes, first of all ^{60}Co, but also ^{24}Na, ^{46}Sc, $^{116}In^m$, ^{140}La, ^{142}Pr, and ^{182}Ta sources for irradiation. Utilizing the low-background off-beam conditions for the measurement of the isomeric activities produced by the Compton-scattered γ rays in the source, such investigations resulted in a wealth of information for the properties of the activation levels as well as practical applications in gamma-activation analysis, activity and dose-rate measurements of multicurie sources, burnup determination of spent reactor fuel, and hot atom chemistry. A comprehensive review of these studies has been given in Ref. [1].

The investigations have been concentrated on several stable nuclides, the activation levels of which are usually lying in the energy range of 1 to 2.76 MeV, corresponding to the principal γ energies of the exciting sources. However, some motivations have emerged lately to study even lower-lying states, involved, e.g., in nuclear astrophysics [2,3] or γ-laser research [4]. To provide low-energy photons, the use of ^{137}Cs of primary energy 661.66 keV, widely used in kCi amounts, has been considered.

The first experiments using ^{137}Cs γ rays were performed on ^{189}Os and ^{193}Ir nuclides most recently [5], the evaluation of which in terms of isomer excitation cross section is in progress. Presently we are reporting the first results on photoexcitation of ^{77}Se, ^{79}Br, and ^{191}Ir isomers by a ^{137}Cs source.

II. EXPERIMENTS

The ^{137}Cs source was of 67.39 TBq (1820 Ci) activity at the time of the measurement with an active diameter 33.5 mm×42 mm length, encapsulated in a stainless steel cylinder of outer diameter 38 mm×49 mm length. The irradiation setup was according to Fig. 1. A pneumatic rabbit system was used for providing rapid transport of samples between the irradiation and measurement positions. A well type NaI(Tl) scintillation detector with dimensions of diam. 58 mm×58 mm length, well diam. 25 mm×43.5 mm depth, and a 1024 channel analyzer were used for counting isomeric activities in the convenient 100–200 keV γ-ray energy range.

The metallic powder Se and Ir and the NH_4Br samples of natural isotopic composition were inserted in aluminum capsules of internal diam. 13.5 mm×35 mm length and 1 mm wall width. The samples were pressed into shapes of diam. 10 mm uniformly, while their length varied between 8 and 20 mm. The position of the sample was not fixed inside the capsule so as to facilitate slipping to the ends when the capsule got stuck having been shot into the irradiation and measurement positions.

The samples were irradiated for 5–6 times the half-lives of the respective isomers to attain saturation, then counting and, again, irradiation followed. The number of activation-counting cycles was 100 for the Se and Br and 150 for the Ir samples. The main data for the isomers [6] and the samples, the irradiation and counting periods, and the number of counts are summarized in Table I. The self-absorption of the signature transition in the sample material was determined from tabulations for extended sources [7].

III. DATA ANALYSIS

The isomer production yield, i.e., isomeric activity, is determined from the measured count rate, the detection efficiency, the γ-ray emission probability of the signature transition, and the transparency of the sample for this γ energy. On the other hand, the yield for a nucleus to be activated can be written as

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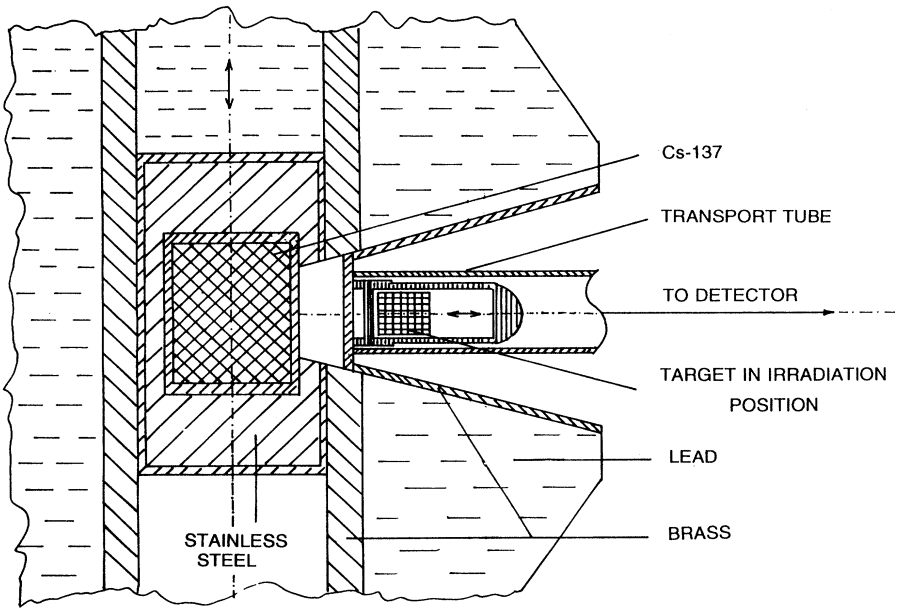

FIG. 1. Irradiation setup.

$$Y = f_r(E_a)\int \sigma(E)dE = f_r(E_a)\sigma_m \qquad (1)$$

in the case of a single activation level, where the resonance flux density $f_r(E_a)$ is taken at the level energy E_a, and the isomer excitation cross section integrated over the level is

$$\sigma_m = \frac{2I_a+1}{2I_g+1}\frac{\lambda_a^2}{4}\Gamma\frac{B_0}{\alpha+1}B_m \ . \qquad (2)$$

Here I_a and I_g are the spins of the activation and ground states, respectively, λ_a is the wavelength of the γ ray of energy E_a, Γ is the total level width, B_0 and B_m are the branching ratios for the ground and metastable states, respectively, and α is the internal conversion coefficient for the ground state transition.

We estimated the contribution of secondary electrons (produced via photoelectric absorption and Compton scattering) to the isomer production through (e,e') reaction, and found that the effect was more than two orders of magnitude smaller than the direct photon contribution, for each activation level of the isotopes in question. The cross sections for electron excitation were calculated on the basis of the Robl theory [8].

For calculating the resonance flux in the unit energy interval we used the approximation [1]

$$f_r = \omega_E\left\{\frac{n_1}{H}\int f_p\frac{e^{-\mu_i' y}(1-e^{-\mu_1 y})}{\mu_1}dz + \frac{n_2}{H}\int f_p\frac{e^{-\mu_2 y'}(1-e^{-(\mu_2'-\mu_2)y'})}{\mu_2'-\mu_2}dz + \frac{n_4 f_p}{\mu_4(1-e^{-\mu_4' L})}\left[1-\frac{\mu_4' e^{-\mu_4 L}-\mu_4 e^{-\mu_4' L}}{\mu_4'-\mu_4}\right]\right\}\frac{1-e^{-\mu_4' L}}{\mu_4' L} \ , \qquad (3)$$

where the first, second, and third members of the sum represent the contributions from the scattering in the source, its cladding, and the target, respectively. We disregarded the scattering from the third medium (stainless steel, brass, and lead shielding, see Fig. 1) in this approximation, because of the complex geometry.

The notations are as follows. ω_E is the cross section of the Compton scattering from a single electron, calculated from the Klein-Nishina formula, n_1, n_2, and n_4 are electron densities, the corresponding μ's and μ''s are total attenuation coefficients for primary and scattered photons in the source, cladding, and target, while H and L are source and target heights, respectively.

$$y = \frac{R\sqrt{D^2+(z-H)^2}}{D}, \quad y' = \frac{yd}{R} \ , \qquad (4)$$

where R is the radius of the source cylinder, d is the thick-

TABLE I. Primary data for irradiation and counting. The isomer deexcitation data are taken from Ref. [6]. Uncertainties are in parentheses.

Nuclide	^{77}Se	^{79}Br	^{191}Ir
Half-life of the isomer (s)	17.45	4.86	4.94
γ energy (keV)	161.9	207.2	129.4
γ emission yield	0.524	0.758	0.257
Photopeak detection efficiency	0.56	0.46	0.66
Irradiation time (s)	90×100	30×100	30×150
Measurement time (s)	10×100	5×100	5×150
Sample length (mm)	20	19	8
Sample mass (g)	4.30	4.128	2.85
Number of counts	1745 (262)	722 (110)	45 (43)
Extrapolated count rate (counts/s)	2.40 (0.40)	2.93 (0.66)	0.12 (0.12)
Sample transparency	0.70 (0.05)	0.75 (0.05)	0.10 (0.02)

TABLE II. Main data for calculation of the resonance flux density.

Nuclide			^{77}Se		^{79}Br	^{191}Ir (10^{23}cm^{-3})	Electron density
Primary and scattered γ energies (keV)	662	521	440	250	384	659	
Attenuation coefficient (cm^{-1})							
CsCl	0.304	0.363	0.427	0.985	0.495	0.305	10.2
Fe	0.579	0.647	0.706	1.004	0.757	0.580	22.05
Se	0.183	0.207	0.230	0.375			6.79
NH_4Br	0.176				0.238		6.58
Ir	0.452					0.454	10.95
Klein-Nishina cross section ω_E (10^{-31} cm^2 eV^{-1})		4.901	4.488	6.122	4.363	5.805	
Scattering angle	0°	37.75°	52.46°	105.91°	63.88°	4.61°	

ness of its cladding (including also the 1.75 mm thick steel closing plate at the end of the transport tube; see Fig. 1), while D is the distance between the central points of the source and the target.

In calculating electron densities, the actual values of the porous target materials were considered. The total attenuation coefficients were determined from the tables of Veigele [9]. The primary flux f_p was determined using the tables in Ref. [7].

The angular distribution of the scattered photons was not considered in this formula. Furthermore, the target was regarded as a point. For ^{191}Ir about 22% higher and 17% lower flux values were calculated at the front and back ends of the cylinder, respectively.

The contribution from double scattering was taken into account on the basis of the approximation [10]

$$\frac{f_r(\text{double scattering})}{f_r(\text{single scattering})} = \frac{n\bar{\omega}_E \Delta E}{\bar{\mu}} . \quad (5)$$

Here ΔE is the difference between the energies of the primary quanta and the activation level, and $\bar{\omega}_E$ and $\bar{\mu}$ are mean values of the scattering cross section and attenuation coefficient, respectively, in this energy interval. This correction was negligible (1.5%) for ^{191}Ir because of the small ΔE, but amounted to about 58% for ^{79}Br and 61–84 % for ^{77}Se levels.

The main parameters for the calculation of the resonance flux are given in Table II. Because of the small scattering angle, the presence of the source container can be neglected in the case of ^{191}Ir, while considerable effects may arise in the other two cases. For this reason, the resonance flux densities for ^{77}Se and ^{79}Br activation levels may be underestimated.

IV. RESULTS AND DISCUSSION

The results are summarized in Table III. The integrated isomer production cross sections have been calculated for the activation levels from adopted half-lives, spins, and branching ratios (to be read from Figs. 2–4 below) on the basis of Eq. (2) on the one hand, and from measured isomer activation yields on the other hand. Level half-lives, where not directly taken from the literature, have been derived from the measured cross sections.

A. ^{77}Se

The relevant detail of the level scheme up to 662 keV, the primary energy of ^{137}Cs, is shown in Fig. 2 [11]. The levels at 521, 439, and 250 keV from where there are direct transitions to the ground state and direct or cascade transitions to the metastable state were established to be activation levels. The cross sections σ_m for these

TABLE III. Results. 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 partial yields at 250 and 440 keV were determined by multiplying σ_m with f_r. By subtracting their sum from the total yield, that for 521 keV was obtained. The uncertainties (in parentheses) do not contain those of the literature data. Level half-lives were taken from the literature and/or determined from our results for σ_m. B_m =isomer branching ratio.

Nuclide	Activation level (keV)	Y (Bq/mol)	f_r (10^5 cm^{-2} s^{-1} eV^{-1})	σ_m (mb eV) Ref. [11]	σ_m (mb eV) This work	Partial yield (Bq/mol)	Level half-life (10^{-12} s) Ref. [11]	Level half-life (10^{-12} s) This work
^{77}Se	250	2756 (550)	2.51 (1.25)	2.0		302 (150)	9680	
	440		2.54 (0.80)	5.0		765 (240)	23	
	521		2.42 (0.75)	0.50	11.6 (5.6)	1689 (619)	90	4(2)
^{79}Br	384	423 (102)	2.34 (0.84)		3.0 (1.0)			6 (2)$\times 10^3 B_m$
^{191}Ir	659	1275 (1222)	2.83 (0.52)		7.5 (7.3)		5×10^{-5}–120	0.9–13.6

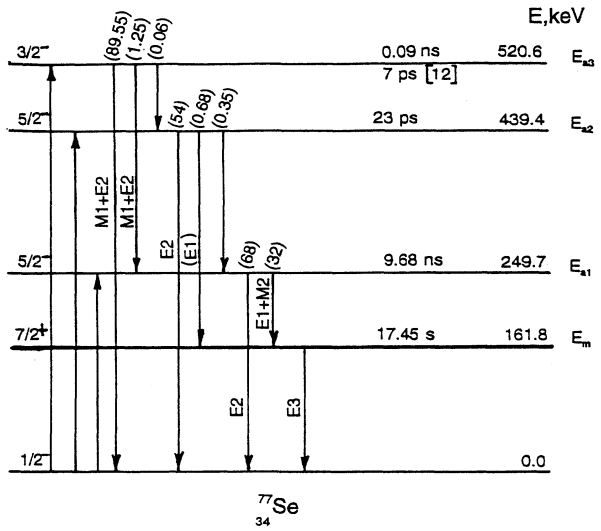

FIG. 2. Partial level scheme of ^{77}Se [11] up to 662 keV, the primary energy of ^{137}Cs. Only those (activation) levels (E_{ai}) and transitions are indicated which are involved in the population of the metastable state (E_m). Branching ratios in percent are in parentheses.

levels were determined from the parameters given in Ref. [11]. We tried to separate the measured yield into contributions from individual activation levels in the following manner. Adopting the literature half-lives 9.68 ns and 23 ps for the 250 and 440 keV levels, respectively, we obtained the values for σ_m given in Table III. Multiplying these with the corresponding resonance flux densities, we determined the isomeric activities resulting from the excitation of these two levels. The sum of the partial yields thus obtained was subtracted from the total isomer yield and the rest was attributed to the excitation of the third level at 521 keV. Dividing this by the corresponding resonance flux, an experimental cross section was obtained, also indicated in Table III.

It can be seen that this cross section is an order of magnitude larger than that determined from the half-life 90 ps adopted by Nuclear Data Sheets [11]. We derived 4±2 ps for this level which, in turn, is in reasonable agreement with a previous literature value 7 ps [12].

It is to be mentioned at this point that the value of 90 ps, not supported by our measurement, has been based on an order of magnitude mistake in the original publications of Cauchois and co-workers [13], who carried out isomer excitation experiments by bremsstrahlung irradiation, determining integrated cross sections and reduced level widths in terms of $u = g\Gamma_0\Gamma_m/\Gamma$. It is easy to recognize that the right value deduced for this quantity from the integrated cross section would be on the order of 10^{-7} eV instead of 10^{-8} eV given by the authors and used directly by the compilers of Nuclear Data Sheets [11].

Thus the half-life deduced from [13] for the 521 keV level would correctly be 9 ps. Even this value is too long, since all the cross sections given in [13] are too low, both in view of the data of Table III for the 250 and 440 keV levels (deduced from the adopted level parameters in Nuclear Data Sheets [11]), and of those for higher-lying levels provided by recent photoactivation experiments by LINAC bremsstrahlung [14]. In this way, notwithstanding the uncertainties associated with the flux estimation, our result can easily be reconciled with the earlier values, including 7 ps coming from a Coulomb excitation experiment [12].

In the case of experiments with radioactive sources, a comparison between effective cross sections, i.e., isomer yields, related to the primary flux may be useful, because this is the only quantity that can be evaluated when activation levels are not known, or many of them are involved. Considerable increase is found in these terms from the present 5.5×10^{-32} cm^2 to a 49×10^{-32} cm^2 value obtained with a ^{24}Na exciting source [1].

B. ^{79}Br

We assume the principal activation level at 384 keV (Fig. 3). From here, a transition to the $\frac{3}{2}^-$ ground state is known, but no transition to the isomer has been observed [15]. Hence, we propose a low intensity 176.5 keV transition to the $\frac{9}{2}^+$ metastable state. From the possible $\frac{3}{2}^+$ or $\frac{5}{2}^+$ spin-parity assignments given for this level [15], we select $\frac{5}{2}^+$; thus the excitation may take place by an $E1$ transition, while the decay to the metastable state proceeds by an $E2$ transition, as indicated in the figure. No data for the half-life of this level are available in the literature.

We have determined an integrated cross section as 3±1 mb eV (Table III), from which we deduce 6 ± 2 ns for the ratio of the half-life of this level to its branching to the isomer. That is, an isomer branch of, e.g., 10^{-2} corresponds to a half-life of 60 ps.

An order of magnitude higher cross section can be de-

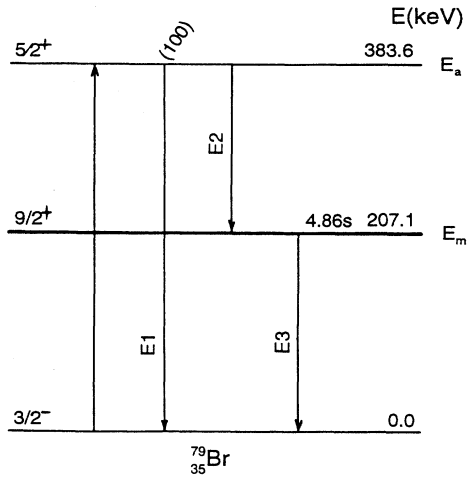

FIG. 3. Fragment of the level diagram of ^{79}Br (proposed). The principal activation level assigned $\frac{5}{2}^+$ is populated by an $E1$ transition from the ground state, while a postulated $E2$ transition of low branching feeds the metastable state.

termined from the data adopted by Nuclear Data Sheets [15] for the next activation level located at 761 keV, in accordance with the results of photoactivation experiments with ^{46}Sc and ^{60}Co sources [1,16]. Thus the lower-lying level explored presently could not result in a large surplus yield in those experiments. Further enhancement results from even higher-lying levels up to 2.75 MeV, pointed out by high-activity ^{142}Pr and ^{24}Na sources [1], which can be seen in terms of the effective cross section, upon comparing 8×10^{-33} (^{137}Cs) with 6.5×10^{-32} cm^2 (^{24}Na).

C. ^{191}Ir

Direct ground state and cascade transitions to the isomer having been observed, it is easy to identify a single activation level at 659 keV, as indicated in the level diagram [17]; see Fig. 4. For the half-life of this level, upper (0.12 ns) and lower (0.05 fs) limits only have been given in the literature [17]. A value of about 7 ps can be deduced from our measurement (Table III).

The effective cross section increases from the present 2×10^{-32} to 17.3×10^{-32} cm^2 for ^{24}Na [1].

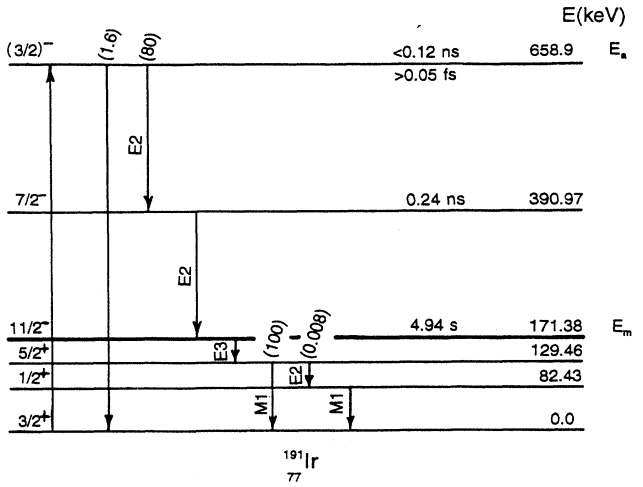

FIG. 4. Partial level scheme of ^{191}Ir [17]. A single activation level can be identified.

V. CONCLUDING REMARKS

The main results of this study can be summarized as follows.

Using adopted parameters for the lower-lying activation levels involved in ^{77}Se^m production, a new half-life was determined for the third level, contradicting recent evaluations in Nuclear Data Sheets, but in reasonable agreement with previous data.

One principal activation level was identified for photoexcitation of ^{79}Br^m, and a spin-parity assignment was made. Postulating an $E2$ transition to the metastable state, the ratio of the level half-life to the isomer branching was determined.

For ^{191}Ir, a single activation level was located, allowing the determination of its half-life for the first time.

It is to be noted that the direct observation of the activation levels suggested here for ^{79}Br and ^{191}Ir is desirable by variable end point energy bremsstrahlung.

The situation concerning ^{77}Se approaches an ideal status—similar to the most extensively studied case of ^{115}In [18], or, e.g., that of ^{83}Kr examined just recently for the first time [19]—that photoactivation can quantitatively be accounted for relying on accepted nuclear data available in recent tabulations, even if these tabulations need to be reconsidered in the present particular case. Hence, we did not make allowance for a possible nonresonant reaction mechanism claimed to have a major role in photoexcitation [20]. We rather think that the present results provide further evidence for the resonance character of the excitation, in addition to those in Ref. [21].

The issue for ^{79}Br and ^{191}Ir indicates that photoactivation by ^{137}Cs γ rays may represent a tool for determining unknown parameters of nuclear levels difficult to excite by other means. Thanks to the sensitive isomer activation techniques, very weak transitions to some low-lying, narrow states can in certain cases be explored. Even by this technique, however, the weak population of these states calls for the use of strong exciting sources, preferably energetic synchrotron radiation.

The authors are indebted to the staff of the Laboratory of the National Command for Civil Defense for providing the use of the ^{137}Cs source.

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