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Photoexcitation of ^{189}Os^m and ^{193}Ir^m. I. Excitation of ^{189}Os^m by low-energy x rays

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Production of 5.84 h ^{189}Os^m by 200 through 300 kV bremsstrahlung has been measured and attributed to population of four nuclear states between 69.54 and 275.91 keV. From the measurements and known level parameters, contributions to isomer production from individual levels have been separated, and the half-life of the 216.66 keV level has been determined as 77 ± 10 ps. Besides nuclear resonance absorption (NRA), the first level has long been assumed to be also excited by another process called nuclear excitation by electron transition (NEET), via virtual photon exchange due to recombination of vacancies produced in the K shell by incident x rays. However, contribution from competing NRA cannot be separated, and the claimed dominance or even the existence of the NEET process has remained unconfirmed.

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I. INTRODUCTION

The study of low-lying nuclear levels has been motivated by various reasons. Among several reaction mechanisms, electromagnetic modes of excitation by real and virtual photons play a distinguished role, involving direct interactions with the nucleus via inelastic photon scattering or Coulomb excitation, while there exist also rare mechanisms coupled to atomic deexcitations via higher order processes.

Particularly, excitation of long-lived metastable states has an impact on divers fields such as γ-laser research [1], nuclear astrophysics [2], and spectroscopy of nuclear (activation) levels establishing an effective coupling between the ground state and the isomer. Population of ^{176}Lu^m [3] and depopulation of ^{180}Ta^m [4] by resonance photon absorption may reduce drastically effective half-lives in stellar interiors, strongly affecting the abundances of these nuclides as a result of nucleosynthesis along the s process [2], while emphasizing the role of low-lying activation levels. Determination of the properties of such activation levels may provide unique spectroscopic information, often hardly accessible by other means [5].

The energy released during interactions in the atomic environments may also lead to excitation of the nucleus in some cases, via absorption of a virtual photon. The most widely known examples for such processes are radiationless transitions in muonic atoms [6] and radiationless positron annihilation [7]. Similar higher order electromagnetic processes may also take place upon recombination of vacancies induced in the atomic electron shells. Such phenomena were first considered by Morita [8], who treated the possibility of resonance between atomic and nuclear transitions of the same multipolarities and about the same energies. This process was called nuclear excitation by electronic transition (NEET). NEET was investigated in the cases of ^{189}Os [9–13], ^{197}Au [14], and ^{237}Np [15], and positive results were reported. In a similar mechanism called inverse internal electron conversion (IIEC, or nuclear excitation by electron capture, NEEC) [16], the electron capture from the continuum by a vacancy in the electron shell was considered. The excitation of the ^{235}U^m isomer (75 eV, 26 min) through this process in a laser produced uranium plasma was experimentally observed by Izawa and Yamanaka [17]. This phenomenon (initially attributed to NEET) was considered to be much more probable via free-bound electronic transitions than via transitions between two bound states. However, Arutyunyan *et al.* [18], after repeating this experiment with a null result, observed the population of the ^{235}U^m isomer in a plasma induced by a high intensity electron beam (500 keV, 150 kA) with a cross section of 10^{-32}–10^{-31} cm^2, 2–3 orders of magnitude less than the result of Izawa and Yamanaka.

Excitation of ^{235}U^m in intense laser fields by a third-order mechanism called inverse electron bridge (IEB; also inverse internal Compton effect or nuclear excitation by inelastic photoelectric effect) [19] was theoretically studied with a favorable conclusion recently [20]. The tuned frequency external radiation field stimulates directly, here again, an electron (even an $E1$) transition rather than a nuclear one, utilizing the much larger width of the former, and nuclear excitation occurs as a consequence of a virtual photon exchange. Recently, the excitation of the extremely low energy first level of ^{229}Th at 3.5 eV was proposed [21] by laser stimulation via this effect.

The study of the excitation of ^{189}Os^m was previously restricted to the search for the NEET effect. This effect was first postulated to take place in this nuclide by Otozai *et al.* [9,10]. In their experiments, bombarding osmium with 72–100 keV electrons, the production of the ^{189}Os^m isomer was attributed to the population of the 69.54 level via the NEET mechanism. Saito *et al.* used bremsstrahlung from a 100 kV Cockcroft-Walton generator [11], and found a lower value for the NEET probability P, although it must not depend on "prehistory," i.e., the mechanism for vacancy production. Lakosi *et al.* used bremsstrahlung from a 200 kV x-ray generator [12]. In addition to ^{189}Os^m activity measurement, Shinohara *et al.* registered even the K x rays accompanying recombination of vacancies induced by synchrotron radiation, and obtained the least value for P as yet by a direct evaluation [13].

Calculations were also performed [22–29] applying vari-

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TABLE I. Experimental and theoretical NEET probabilities in ^{189}Os. All the experimental data were determined on the basis of an adopted value of 1.2×10^{-3} for the isomer branching B_m. Calculations were performed for the two atomic transitions indicated in Fig. 2.

		Theory		
Experiment	Ref.	$M1$	$E2$	Ref.
10^{-6}	[9]		$\geq1.6\times10^{-7}$	[22]
$(1.7\pm0.2)\times10^{-7}$	[10]	1.58×10^{-8}	1.43×10^{-7}	[11]
$(4.3\pm0.2)\times10^{-8}$	[11]	2.3×10^{-7}	1.8×10^{-8}	[23]
$(4.0\pm2.7)\times10^{-8}$[a]	[12][a]	1.1×10^{-7}	2.5×10^{-9}	[24]
$(5.7\pm1.7)\times10^{-9}$	[13]	1.06×10^{-7}	1.25×10^{-7}	[25]
$(2.0\pm1.4)\times10^{-8}$	Present work	1.2×10^{-9}		[26]
		1.1×10^{-10}	0.7×10^{-12}	[27]
		3.4×10^{-10}		[28]
		2.1×10^{-9}		[29]

[a] Present reevaluation.

ous theoretical models which resulted in largely varying NEET probabilities, too. In Table I the existing experimental and theoretical results reported for the NEET effect in ^{189}Os are summarized. A general tendency of a gradual decrease in a range of three orders of magnitude is clearly visible, both for the experimental and theoretical results.

In the present work we report on the photoexcitation of ^{189}Os^{m} including a search for the NEET effect in it. Since the study of the level scheme has revealed that a number of low-lying levels are to be involved in the excitation of the isomer via nuclear resonance scattering (NRS), the energy range of low-energy x-ray irradiations was extended up to 300 keV to cover three further activation levels. Also, following our earlier experiment [12], an instrumental development was undertaken [30,31] in order to improve the detection sensitivity and obtain more conclusive results (this paper). On the basis of this development, the energy range was further extended using ^{137}Cs and ^{60}Co sources, for excitation of ^{193}Ir as well (paper II).

II. EXPERIMENTAL

Elemental Os powder samples pressed into pellet form of 2 cm diameter were irradiated by a Philips type MCN 321 x-ray generator at the National Office of Measures, Budapest. The tube was operated at 200–300 kV voltage and 7–15 mA electron current. The 1.5-mm-thick Be window was at 6.7 cm distance from the point of the incidence of the electron beam on the W anode, while the Os samples were placed at 3.1 cm from the window. Thus, an overall distance of 9.8 cm from the origin of bremsstrahlung was taken into account for the calculation of the photon flux. No filter was used. The irradiation geometry is shown in Fig. 1.

Os samples of 1–3 g were used. Irradiations lasted for 5–6 h. Because of the weak isomeric radioactivities excited by low-energy x rays, a sensitive counting technique was applied. A 90 cm^2 sensitive area flat 2π multiwire proportional counter was developed [30] for measuring low energy (about 20 keV) electrons from the highly converted

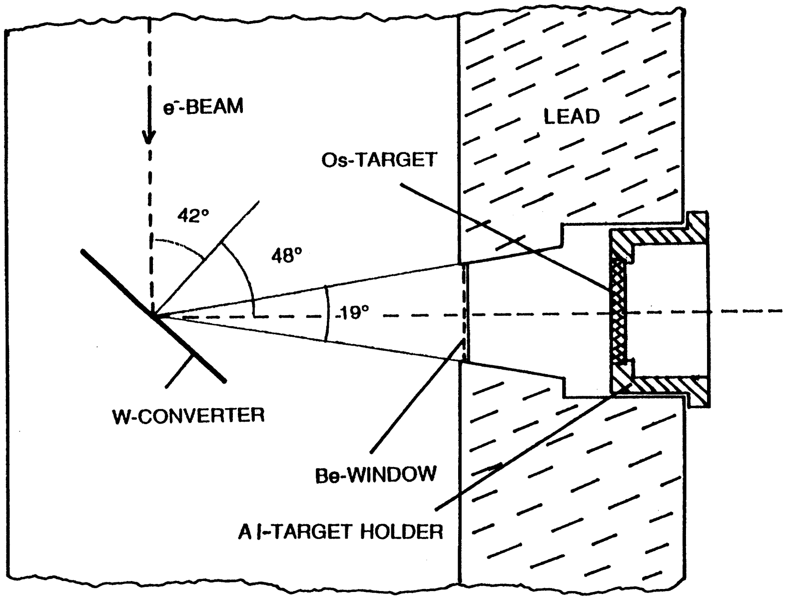

FIG. 1. Diagram of the irradiation geometry.

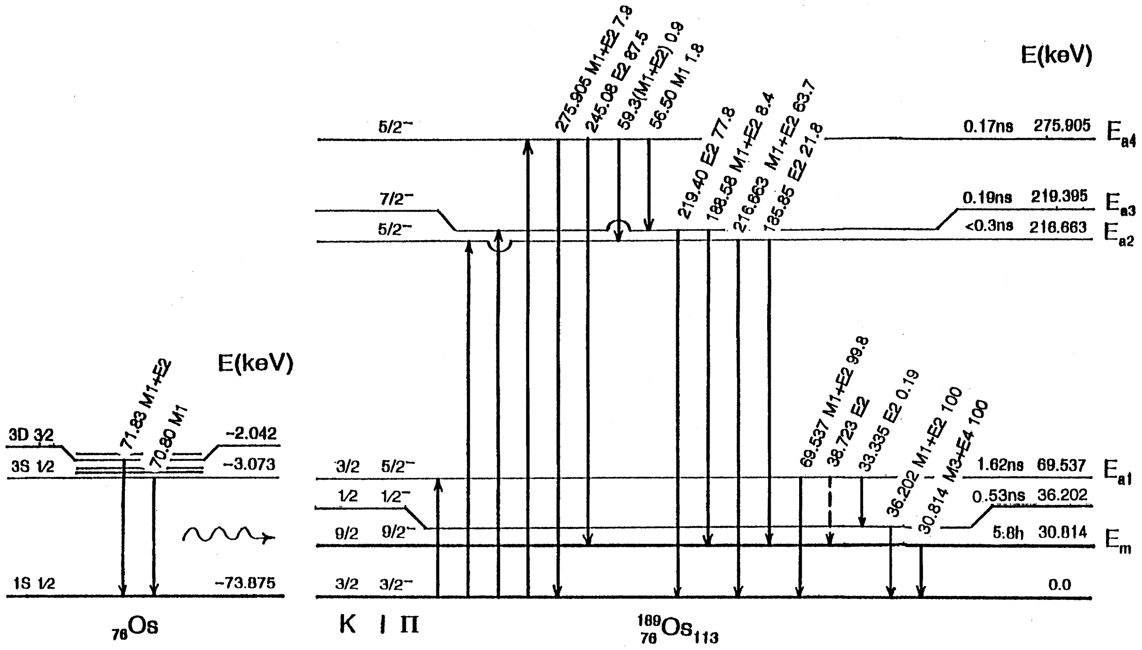

FIG. 2. Fragment of the level scheme of ^{189}Os up to 300 keV. Activation levels ($E_{\mathrm{a}i}$) and transitions feeding the metastable state (E_m) are indicated [32,33]. Branching ratios are given in percent. A partial atomic level diagram relevant to possible NEET mechanism is also shown (energies are taken from Ref. [34]).

($\alpha = 3.24 \times 10^5$), 5.8 h half-life, 30.8-keV energy isomeric transition [32]. After irradiations, the samples were dismounted and their content was evenly dispersed over the brass cathode plate serving as sample holder of the proportional counter at the same time. Measurements lasted for several hours. Afterwards, the samples were left in the counter for background measurements, because even inactive samples emitted KX rays; γ spectra always exhibited their presence, excited by the environmental background radiation. More technical details are given in Refs. [30,31].

When using the 2π counter, a sensitivity increase by a factor of about 140 was attained against a low-energy planar Ge detector for ^{189}Os^m measurement.

III. DATA ANALYSIS

A. Identification of the activation levels

The relevant fragment of the ^{189}Os level scheme up to 300 keV is shown in Fig. 2 [32]. Direct ground state and direct or cascade transitions to the isomer having been observed, it is easy to establish activation levels at 216.66, 219.40, and 275.91 keV. ($I^\pi = 5/2^-$ was adopted for the 216.66 keV level [33], instead of $7/2^-$ given by Ref. [32].) However, an activation level should also exist at lower energy; its existence was postulated by the successful isomer excitation experiments performed by 72–100 keV electron [9,10] and up to 200 keV energy photon bombardments [11–13]. The only candidate for this is the $5/2^-$ 69.54 keV level, even if the 38.7 keV $E2$ transition from it (indicated by dashed line in the figure) to the $9/2^-$ 30.81 keV metastable state was directly never observed. Through this level both NEET and resonance scattering processes may feed the isomer. A partial atomic level diagram for the NEET mechanism is also shown in the figure, indicating two transitions of $M1$ and $M1+E2$ multipolarities, the same character as that of the nuclear transition linking the ground state with the 69.54 keV level. Contributions from atomic shells other than indicated in the figure are less.

B. General formalism

For a single nucleus to be activated, the isomer yield induced by (γ,γ') NRS can be written as

$$Y_{\mathrm{NRS}} = \int f(E,E_0)\sigma(E) f_a dE = f_r(E_a,E_0) f_a \sigma_m, \qquad (1)$$

where the spectral flux density, $f(E,E_0)$ (E_0 is the end-point energy) is taken at the activation level energy E_a. The flux attenuation in the sample has been taken into account by the factor $f_a = (1 - e^{-\mu d})/\mu d$. The isomer excitation cross section integrated over a single activation level is

$$\sigma_m = \frac{2I_a+1}{2I_g+1}\pi^2\left(\frac{\hbar c}{E_a}\right)^2 \Gamma \frac{B_0}{\alpha+1} B_m. \qquad (2)$$

Here I_a and I_g are the spins of the activation and ground levels, respectively, B_0 is the ground state branching ratio, and B_m stands for the sum of all branches populating the metastable state, either directly or via cascade, α is the internal conversion coefficient for the ground state transition, and Γ is the total width of the activation level related to the level lifetime.

TABLE II. Calculated flux densities, attenuation corrections $f_a=(1-e^{-\mu d})/\mu d$, integrated cross sections, and NEET contributions $Y_{\mathrm{NEET}}/PB_m=\int_{E_{\mathrm{thr}}}^{E_0} f(E,E_0)\sigma_K f_a dE$.

	Photon flux density (10^6 cm^{-2} s^{-1} eV^{-1})			Attenuation correction, f_a		Calculated NRS isomer excitation cross section
Level energy (keV)	200 kV 15 mA	250 kV 7 mA	300 kV 7 mA	$d_1=0.318$ (g/cm^2)	$d_2=0.478$ (g/cm^2)	(10^{-26} cm^2 eV)
69.54	15.08	9.59	11.94	0.667	0.609	$3.54B_m$
216.66		0.475	1.24	0.892	0.854	$6.18/t_{1/2}(10^{-10}$ s)
219.40		0.428	1.18	0.903	0.860	2.01
275.88			0.261		0.909	1.20
Y_{NEET}/PB_m	3.24	1.83	2.58			
(10^{-10} Bq/nucleus)	(d_1)	(d_2)	(d_2)			

The isomeric activity due to the population of the 69.54 keV level is assumed to be consisting of two components; contributions of NRS and the NEET effect. The NEET contribution is

$$Y_{\mathrm{NEET}}=PB_m\int_{E_{\mathrm{thr}}}^{E_0} f(E,E_0)\sigma_k(E)f_a dE, \tag{3}$$

where P is the NEET probability, B_m is the (as yet unobserved) isomer branching of the 69.54 keV level, $E_{\mathrm{thr}}=73.87$ keV is the energy of the K absorption edge, and σ_k is the K-shell ionization cross section of osmium for the incident x rays, taken from Ref. [35].

The spins, half-lives, and branching ratios of the activation levels and internal conversion coefficients of the ground-state transitions are known, except for the half-life of the $5/2^-$ 216.66 keV level (an upper limit is given) and the isomer branching B_m of the $5/2^-$ 69.54 keV level.

Since the unknown B_m appears in both constituents of the yield [see Eqs. (1) through (3)], neither P nor even the product PB_m can be determined from a single measurement, unless further provisions are made for either of them.

C. Determination of the irradiating photon flux

The spectral distribution of bremsstrahlung as the function of the photon energy E and the energy of electron beam E_0 was calculated on the basis of a semiempirical formula developed for thick-target spectra [36]. Accordingly, the number of bremsstrahlung photons per unit energy interval per incident electron is

$$\frac{dN(E,E_0)}{dE}=\frac{kZ(E_0-E)[1-\exp(-3E/E_k)]}{E(E/E_0)^{1/3}[1-\exp(-E_0/E_k)]}\times\exp(-0.2R\mu_B/\tan\alpha), \tag{4}$$

where 1.1×10^{-6} (keV interval electron)$^{-1}$ was taken for k [36]. R is the range of an electron with initial energy E_0, μ_E is the total attenuation coefficient of a photon with energy E, and α is the angle between the incident electron beam and the normal to the converter, 42° in the present case (Fig. 1).

In this formula, the electron energy loss, electron backscatter loss, and photon attenuation in the converter were considered, and overall agreement of 20% with measurement below 300 keV was attained [36]. The characteristic radiation was not taken into account, because we are not interested in the spectrum below 69.5 keV, the ionization potential E_K of the K shell for $Z=74$ (tungsten).

From the number of bremsstrahlung photons, we determined the flux density, taking into account the electron current and the distance from the point of incidence of the beam on the anode to the sample, 9.8 cm in our case.

Representative results of calculations for 200, 250, and 300 kV irradiations are given in Table II, where calculated cross sections for isomer excitation by NRS, integrated over each respective activation level, are also indicated. Figure 3 shows corresponding bremsstrahlung spectra.

IV. RESULTS AND DISCUSSION

After irradiations carried out at 200, 230, 250, 280, and 300 kV generator voltages in altogether eight runs, isomer production yields were determined from the measured activities corrected for saturation loss due to insufficient sample thickness, using the calibration of the proportional counting [30]. The yield curve as a function of the end-point energy can be seen in Fig. 4. The partial yields for the activation levels at 219 and 276 keV were calculated using Eq. (1) and subtracted from the overall yields determined for each run. In such a way, a series of partial yields were obtained for acti-

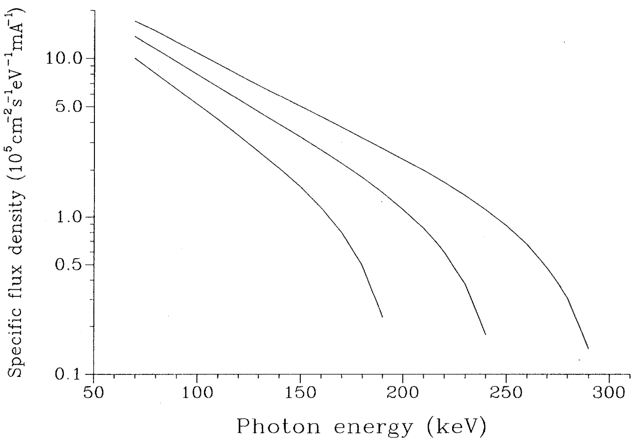

FIG. 3. Bremsstrahlung flux densities calculated for 200, 250, and 300 kV end-point energies at target distance.

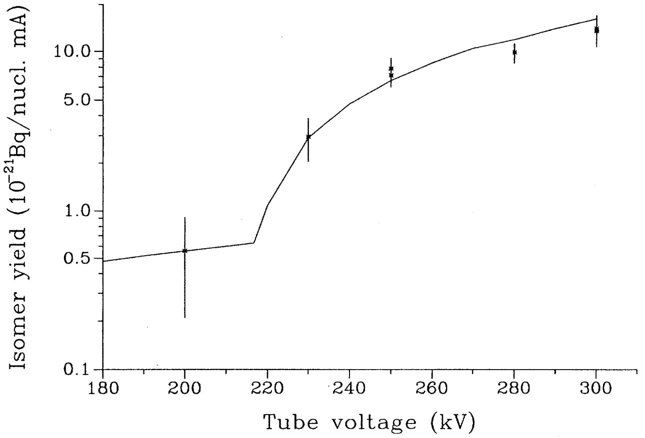

FIG. 4. Isomer excitation function measured and calculated (solid curve).

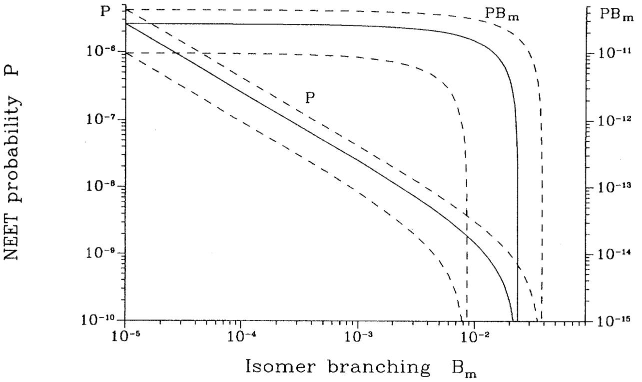

FIG. 5. Interdependence between NEET probability P and isomer branching B_m of the 69.54 keV level, according to Eq. (5), derived from the 200 kV experiment. Dashed lines indicate error limits.

vation via the 217 keV level, the half-life of which is not known. Calculating the flux data, cross sections and corresponding half-lives were derived for this level. As a weighted mean of seven runs, a cross section of $(8.1\pm0.9)\times10^{-26}$ cm^{2}eV and, from this, a half-life of $(0.77\pm0.10)\times10^{-10}$ s were obtained. (The errors contain the uncertainties of the measurements only.) The solid curve in Fig. 4 was calculated using this cross section (and those in Table II).

Inspection of the cross section values inferred from literature half-lives and branching ratios for the upper-lying two levels (Table II) shows that they do not play a crucial role. Allowing even 50% uncertainty in that value for the 219 keV level, its influence is less than 14% on the experimental cross section evaluated for the 217 keV level, while this figure caused by a similar uncertainty of the cross section for the 276 keV level alone is less than 2%.

From the run performed by 200 kV irradiation, the equation

$$3.24\times10^{10}PB_m+35.6B_m=0.84\pm0.53 \qquad (5)$$

follows for the determination of the NEET probability P and the isomer branching B_m of the 69.5 keV level, the only activation level below 200 keV. [The first (NEET) term is also present upon irradiation by higher generator voltages, but its relative importance is gradually decreasing with the energy increase, and obscured by uncertainties associated with larger yields.] From this equation, an upper limit on PB_m as $((2.6\pm1.6)\times10^{-11}$ or, on the other hand, on B_m as $2.35\pm1.5\%$ can be imposed, by neglecting NRS and NEET contributions alternately. As can be seen, the value of B_m plays a key role in the interpretation of the results. Both NRS and NEET are involved in the isomer excitation through that.

B_m was first estimated by Otozai *et al.* [9,10], based on the branching ratio of the 33.3 keV $E2$ transition, given experimentally by Harmatz *et al.* [37] as 6.0%, to the $K,I^\pi=1/2,1/2^-$ level at 36.2 keV (Fig. 2). Considering that the 33.3 and 38.7 keV transitions are both of $E2$ character and about equal transition energies, but the 38.7 keV transition from the $3/2,5/2^-$ level at 69.5 keV to the $9/2,9/2^-$ isomeric state is K forbidden in the first order, while the 33.3 keV transition is not, Otozai *et al.* assumed that the branching of the two transitions differed only by an empirical hindrance factor $\sim1/50$. In this way, they obtained $B_m=1.2\times10^{-3}$, which led to 1.7×10^{-7} for P [10]. All the experimental P values given in Table I were obtained assuming this B_m in the evaluations. However, a more recent value of 0.18% instead of 6.0% is available [32] for the branching ratio of the 33.3 keV transition. Taking this value, a recalculation leads to $B_m=3.6\times10^{-5}$, which corresponds to 5.7×10^{-6} for P. Obviously, this figure would be too large, i.e., either B_m is too small, or some other mechanism may also play a role in the excitation. Indeed, Tkalya [38] attributed the result of Otozai *et al.* [10] to (e,e') inelastic scattering, while for the contribution of the NEET process he estimated a value two orders of magnitude less.

It is to be noted that due to simultaneous ionization of the outer atomic shells during the irradiation, atomic level energies along with transition energies change. As a consequence, actual P values may differ from theoretical ones, depending on the real conditions tuned to or off resonance, but the NEET probability cannot exceed an upper limit $P_{M1}=5.7\times10^{-7}$ via an $M1$ transition [28].

Our result is $(2.0\pm1.4)\times10^{-8}$ for P as shown in Table I, assuming 1.2×10^{-3} for B_m. It is to be stressed that we do not state to have a result for the NEET probability as given in Table I; it is based on a particular B_m value and indicated only for the sake of comparison with other results. While adopting 3.6×10^{-5} for B_m, the value $(7.2\pm4.5)\times10^{-7}$ ensues for P. These data would mean that NEET is the dominant process (by about 95% in the former and more than 99% in the latter case). P and B_m values, pertaining mutually together to fit in Eq. (5), can be read from Fig. 5.

Among theoretically calculated P values indicated in Table I, the most recent results are first of all to be considered, which supersede earlier ones of the same authors. Tkalya used his value 3.4×10^{-10} [28] for the reevaluation of previous measurements. From the results of Shinohara *et al.* [13] obtained by synchrotron radiation he deduced 3.4×10^{-3} for B_m [38]. Using this branching, he concluded on the result of Otozai *et al.* [10] as mentioned above.

If we adopt $P=3.4\times10^{-10}$, [28] $(1.8\pm1.1)\times10^{-2}$ is obtained for B_m, which means that NRS is prevailing over

NEET, similarly to Tkalya's interpretation [38] on the result of Shinohara *et al.* [13]. By accepting, in turn, $B_m = 3.4 \times 10^{-3}$ [38], $P = (6.5 \pm 4.8) \times 10^{-9}$ results from Eq. (5), rendering over 80% majority to NEET, not considering a third possible excitation mechanism (inverse electron bridge may be a candidate [19]). This means that our result is in agreement within error limits both with that of Shinohara *et al.* [13] and, on the other hand, with the P value of Tkalya [28]. It is not incompatible even with Ho's calculated result, $P = 2.1 \times 10^{-9}$, [29] either.

It can be concluded that, since NRS may alone be responsible for the population of the isomer, neither the present, nor any of the earlier results can be considered to provide an experimental proof for NEET in ^{189}Os. It has also to be stressed that no similar experiment can solve the problem. Evidence of NEET could only be given if there were independent data available on B_m. Yet, the present study is relevant in giving upper limits for the product of the (possible) NEET probability and the isomer branching of the 69.5 keV level, and also for the latter alone. While the possible participation of NEET (or a third mechanism) in ^{189}Os^{m} production cannot be excluded, it has been confirmed at the same time, that the isomer is populated via NRS involving identified activation levels. Our results are consistent with level parameters obtained previously by different experimental techniques. Also, it has become possible to establish the half-life of the $5/2^{-}$ 216.7 keV level.

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