Probability of nuclear excitation by electron transition in Os atoms

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An experiment on nuclear excitation by electronic transition (NEET) induced by deexcitations of the excited atomic states was performed on ^{189}Os. Osmium targets were bombarded with both a white beam and with a monochromatic 115 keV x ray beam to produce *K* vacancies in Os atoms. Online *K* x-ray spectra were measured with a Ge detector and the *L* x rays emitted from the 30.814 keV isomer state in the ^{189}Os nucleus were measured offline. The NEET probability obtained from the monochromatic beam experiment was deduced to be less than 4.1×10^{-10} per created *K* hole. This value is much lower than the values measured so far. The white beam experiment shows that these high probabilities of the NEET process in ^{189}Os, which were obtained by irradiation with white x-ray and bremsstrahlung x-ray beams, results mainly in a contribution from direct nuclear photoabsorption into the 69.537 keV state in ^{189}Os.

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I. INTRODUCTION

The deexcitation of excited states of atoms proceeds usually by x-ray and Auger electron emission. The third decay mode of deexcitation of atomic states was proposed theoretically by Morita [1]. This process is called nuclear excitation by electron transition (NEET). In the NEET process, the energy of the atomic excited states is transferred to the nucleus via the exchange of a virtual photon. The NEET process can be regarded as an inverse process to internal conversion. The most widely known example for this process is radiationless transition in muonic atoms [2]. The process can occur only when the atomic and nuclear excited states have close transition energies and involve the same spin and parity changes, although its probability may be very small. The probability for the NEET process is inversely proportional to the square of the difference between the electron transition energy and the nuclear transition energy.

The NEET process was investigated experimentally [3–8] and theoretically [5,8–14] on the ^{189}Os nucleus, because it can offer experimentally a signature for excitation of the 69.537 keV nuclear state in ^{189}Os by this process. The 69.537 keV state decays with a partial branch to a 6 h isomer state of 30.81 keV in ^{189}Os and the isomer state decays by internal conversion. The detection of conversion electrons or *L* x rays associated with *L* conversion electrons is a signature of the NEET process. The atomic and nuclear levels involved in the NEET process in ^{189}Os are shown in Fig. 1.

The first result of the experiment investigating NEET process was reported by Otozai *et al.* [3,4]. They used electrons below 100 keV to ionize the *K* shells of Os atom and detected the conversion electrons from the 6 h isomer state activated through the 69.537 keV level of ^{189}Os. Saito *et al.* [5] observed the radioactivity of the 6 h isomer in ^{189}Os after irradiation with bremsstrahlung x rays produced by 200 keV electrons and they concluded that the radioactivity was ascribed to the excitation of the 69.537 keV state in ^{189}Os induced by the NEET process. Shinohara *et al.* [6] observed *L* x rays from the isomer in ^{189}Os, after irradiation with a white beam of synchrotron radiation and they concluded that production of the activity resulted mainly from the excitation of the 69.537 keV state in ^{189}Os by the NEET process. Lakosi *et al.* [7] used bremsstrahlung x rays of 200 to 300 keV from a x ray generator. They claimed from their results that the contribution from nuclear resonance absorption could not be separated. The use of electron beam, bremsstrahlung x rays or synchrotron white beam may result in a contribution from direct nuclear resonance scattering into a range of levels which feed the 30.81 keV isomer state. Therefore, it is necessary to study the NEET process using a monochromatic x-ray beam which does not overlap any of the nuclear level energies of ^{189}Os. Ahmad *et al.* [8] employed a monochromatic 98.74 keV x-ray beam from a synchrotron radiation

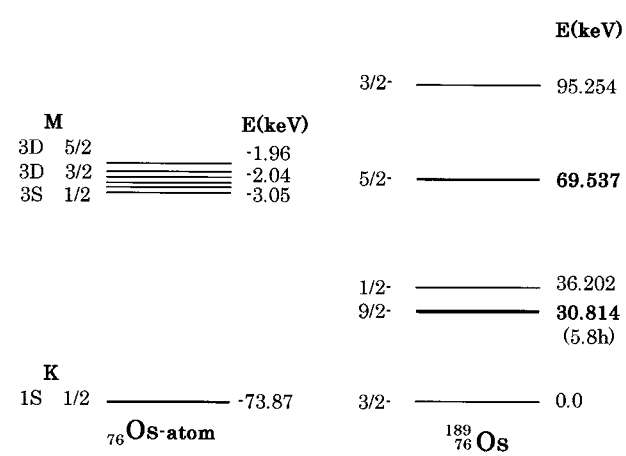

FIG. 1. Atomic and nuclear levels involved in the NEET process in ^{189}Os.

TABLE I. Summary of published NEET probabilities on ^{189}Os.

Experiment	P_{NEET}	Theory	P_{NEET} $(M1)$	P_{NEET} $(E2)$
Otozai *et al.* [3][a]	1×10^{-6}	Okamoto [9]		$\geqslant 1.6\times10^{-7}$
Otozai *et al.* [4]	$(1.7\pm0.2)\times10^{-7}$	Saito *et al.* [5]	1.58×10^{-8}	1.43×10^{-7}
Saito *et al.* [5]	4.3×10^{-8}	Pisk *et al.* [10]	2.3×10^{-7}	1.8×10^{-8}
Shinohara *et al.* [6]	$(5.7\pm1.7)\times10^{-9}$	Bondarkov *et al.* [11]	1.1×10^{-7}	2.5×10^{-9}
Lakosi *et al.* [7]	$(2.0\pm1.4)\times10^{-8}$	Ljubicic *et al.* [12]	1.06×10^{-7}	1.25×10^{-7}
Ahmad *et al.* [8]	$<9\times10^{-10}$	Tkalya [13]	1.1×10^{-10}	0.7×10^{-12}
Present work	$<4.1\times10^{-10}$	Ho *et al.* [14]	2.1×10^{-9}	
		Ahmad *et al.* [8]	1.3×10^{-10}	3.8×10^{-13}

[a]Branching ratio $=7\times10^{-5}$. All the experimental data except Ref. [3] were determined on the basis of an adopted value of 1.2×10^{-3} for the branching ratio for feeding the isomer state from the 69.537 keV state.

source to explore the NEET process. The upper limit of the NEET probability $P_{\mathrm{NEET}}<9\times10^{-10}$ obtained in their experiment was several orders of magnitude lower than the values predicted in previous measurements.

Theoretical calculations [5,8–14] were also performed applying various theoretical models which resulted in largely varying NEET probabilities, too. The existing experimental and theoretical results reported for NEET probability in ^{189}Os are summarized in Table I. It can be seen from Table I that there is a great discrepancy among the predicted and measured data.

We notice also a large difference among the measured NEET probabilities in ^{189}Os. In order to examin this difference, we performed an experiment for ^{189}Os using both white beam and monochromatic x-ray beam as Ahmad *et al.* [8] did. The main goal of this work is to evaluate the order of magnitude of the probability for NEET process using a monochromatic x-ray beam and also to make comparisons with previous experimental data and theoretical results. This is to identify the origin of the large discrepancy.

II. EXPERIMENT

Two enriched (81%) metallic ^{189}Os targets of 15 mm diameter with 4.2 mg/cm^2 (1.91 μm) and 3.5 mg/cm^2 (1.56 μm) thickness, respectively, were made by vacuum evaporation on a 15 μm Al foil.

A. Irradiation with a white x-ray beam

The irradiation with white x rays were carried out to examine the contribution from direct nuclear photoabsorption to the nuclear states which can feed the isomer state. The irradiation of ^{189}Os with white synchrotron radiation beam from a bending magnet was performed in air at beamline BL04B1 in SPring-8. The energy width of the incident photons was about 30 to 150 keV and the maximum intensity was at an energy of about 40 keV. The beam size (2 mm $\times$3 mm) was defined by slits and collimators on the beamline and measured with a fluorescent screen. The current of the electron storage ring gradually decreased from 100 mA to about 90 mA for a day.

To determine the number of ^{189}Os K vacancies produced in the target atoms, the on-line x-ray spectrum was measured with a high-purity Ge detector. The diameter and the thickness of the detector were 10 and 5 mm, respectively. The detector covered by both aluminum and copper plates was placed at 32.3 cm from the target at 90° with respect to the beam axis. A lead collimator with a 2 mm$\times$3 mm window and a 6.0 mm thick copper-plate attenuator were inserted in front of the detector to reduce the excessive counting rate. Figure 2 shows an Os K x-ray spectrum obtained during irradiation for 6.17 h. The Os K_α and K_β x rays resulting from K-hole creation can be clearly seen. The Pb K x rays are background fluorescent x-rays from the shield and collimator materials.

After irradiation, the sample was brought to a low background room and a spectrum of the Os L x ray emitted during the decay of the 30.814 keV isomer state in ^{189}Os nucleus was measured offline with a Ge low-energy photon spectrometer (LEPS). The diameter of the Ge LEPS surface

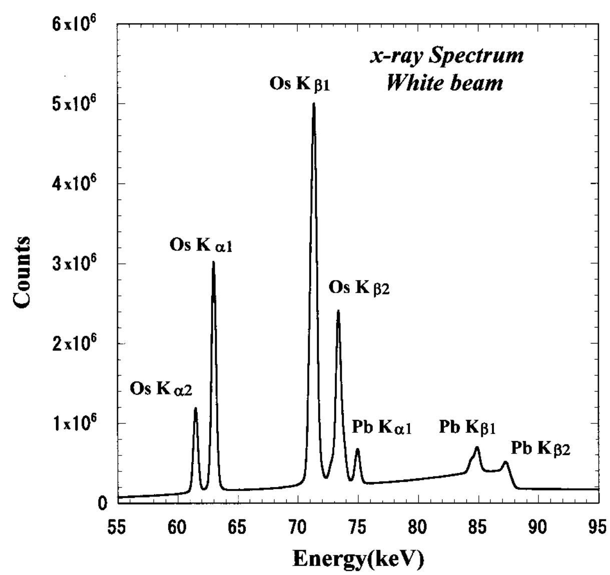

FIG. 2. Online x-ray spectrum of an Os target measured with a pure Ge detector with white x-ray beam. The irradiation time was 6.17 h.

TABLE II. Summary of experimental condition and the results.

Run No.	Target (mg/cm^2), (deg.)	Irradiation time (h)	Produced *K* vacancies	Counting time[a] (h)	Detected *L* x rays
	White beam				
Run 1	4.2, 30	6.17	1.12×10^{16}	6.67	292
Run 2	3.5, 15	7.39	1.65×10^{16}	6.67	367
Run 3	4.2, 40	6.63	1.07×10^{16}	6.67	328
	115 keV beam				
Run 1	4.2, 20	15.5	1.65×10^{15}	14.47	<10
Run 2	3.5, 12	13.8	2.08×10^{15}	12.00	<10

[a]Time of off-line measurement.

was 16 mm. The irradiated sample was placed at 5 mm in front of the detector. The experiment was performed three times in the same way. The experimental conditions are summarized in Table II.

The sum of the spectra obtained during the three offline runs is shown in Fig. 3. At the same time, the half-life of decay of this radiation was measured. Figure 4 indicates the result of the half-life measurement with a fitted curve of the exponential form. Each experimental point in this figure shows the results of summation of Os *L* x-ray intensity measured for the three runs. The obtained half-life time was 5.6 ± 0.8 h. Both the x-ray energies and the measured half-life time indicate that the measured x rays can be assigned to be *L* x rays emitted from the 30.814 keV isomer state of ^{189}Os. They also show that we can measure *L* x rays emitted from the isomer with the Ge LEPS system used in this experiment.

B. Irradiation with a monochromatic beam of 115 keV

The experimental station BL08W-B in SPring-8 was used for the measurements. A monochromatic 115 keV x-ray beam of $10^{12}-10^{13}$ photons/sec was obtained by Bragg diffraction from a single (400) Si crystal placed in the wiggler beam line. The energy width of the beam was about 100 eV. The beam energy does not overlap any of the nuclear level energies of the ^{189}Os nucleus. The targets used were the ones used in the white beam irradiation experiments. The beam sizes which were measured with a fluorescence screen at target position were 10 mm by 2 mm. The thick target was placed at 20° with respect to the beam axis and the thin target was placed at 12°. The irradiation was performed in air at the experimental station. The irradiation times were 15.5 h for the thick target and 13.8 h for the thin target as listed in Table II.

The number of *K* vacancies produced in the ^{189}Os target atoms was measured by online *K* x-ray detection using a pure Ge detector during irradiation. The detector was placed at 28.3 cm from the target center at 90° with respect to the beam axis and it did not have any collimator. A copper plate of 10.226 mm thickness was inserted in front of the Ge detector to reduce the excessive counting rate of x rays. The mean attenuation rates in the energy region of *K* x rays and

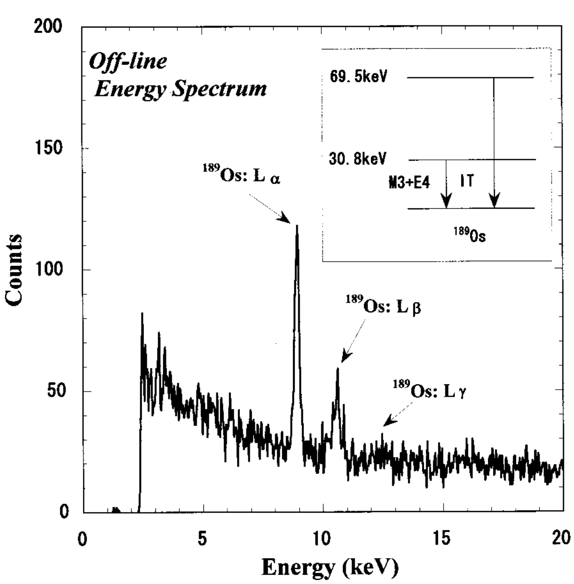

FIG. 3. Offline Os *L* x-ray spectrum summed for three runs after irradiation with white x-ray beam.

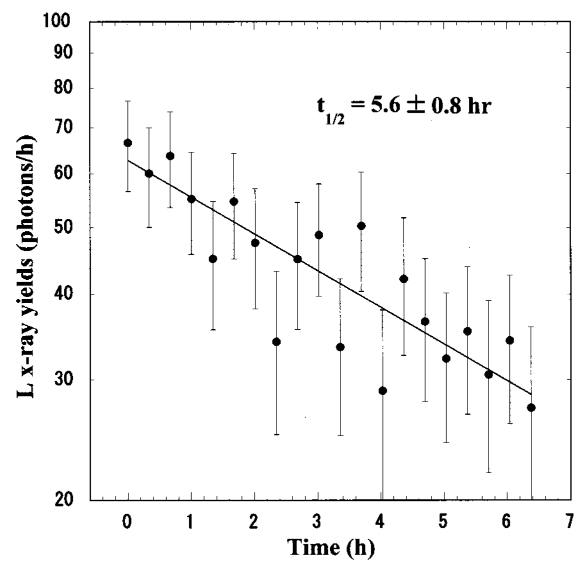

FIG. 4. Observed time variation of Os *L* x-ray intensity summed for three runs at each time. The solid line is a fitted curve of the exponential form. The half-life time was 5.6 ± 0.8 h.

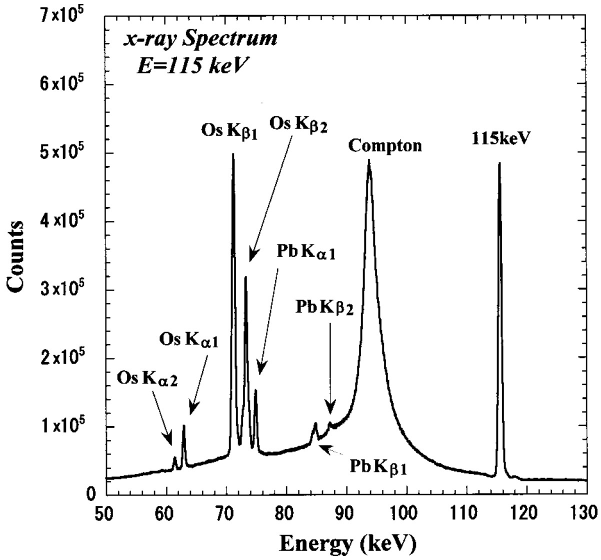

FIG. 5. Online x-ray spectrum measured of an Os target with 115 keV monochromatic x-ray beam. The irradiation time was 15.5 h.

of Compton scattering were about 10^{-4} and about 10^{-2}, respectively. Figure 5 shows an online spectrum observed during irradiation for 13.8 h with 115 keV x rays. The K_α and K_β x rays of Os can be seen clearly.

After irradiation, the target was brought to a low background room and the spectrum of the Os L x ray emitted in the decay of the 30.814 keV isomer state in the ^{189}Os nucleus was measured with the Ge LEPS. The irradiated target was placed 5 mm in front of the detector. The experiment was performed in the same way for both thick and thin targets. The experimental conditions are summarized in Table II.

The off-line spectrum for the thick target is shown in Fig. 6. The counting time was 14.47 h for the thick target and 12 h for the thin target, respectively. Backgrounds in the room used for off-line measurement were also measured. The room background spectrum is shown in Fig. 7. The counting time was 14.75 h. A comparison of Figs. 6 and 7 shows that there are few L x rays that accompany the decay of the isomer state in ^{189}Os.

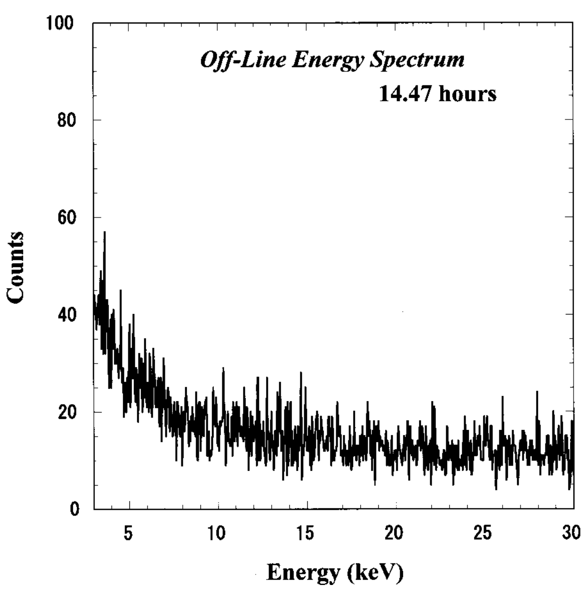

FIG. 6. Offline L x-ray spectrum measured with the Ge LEPS after irradiation. The counting time was 14.47 h.

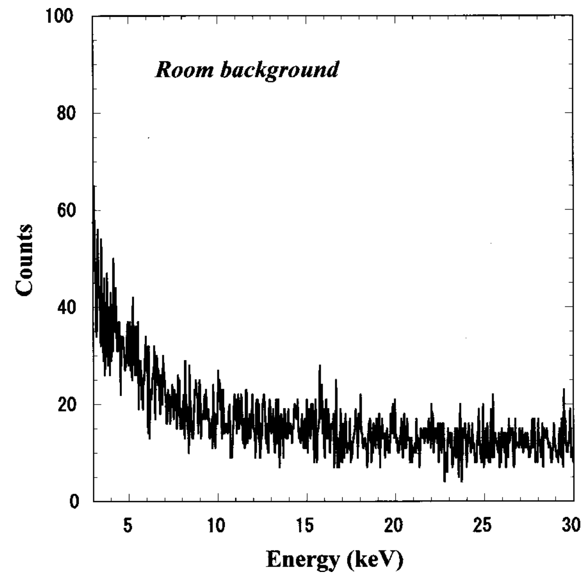

FIG. 7. Room background spectrum in the room used for offline measurement. The counting time was 14.75 h.

III. RESULTS AND DISCUSSION

In the NEET process in ^{189}Os, we consider an atomic transition occurring between an initial K vacancy and a final M-vacancy state. The decay of the initial atomic state will result in the excitation of the corresponding nuclear state. An initial K-vacancy state decays from the $70.82\,\mathrm{keV}(M1)$, $71.83\,\mathrm{keV}(E2)$, and $71.91\,\mathrm{keV}(E2)$ states of the M shell, and the 69.537 keV nuclear state $(\frac{5}{2}^-)$ as shown in Fig. 1 is excited. The state of the 69.537 keV decays promptly with a partial branch to the 30.814 keV isomer state. The isomer decays primarily by internal conversion process. L x rays associated with L conversion electrons is a signature of the NEET process.

The NEET probability per a created K vacancy can be related to the yield of the produced isomer. After irradiation, the isomer production yield (N_{isomer}) is obtained as

$$N_{\mathrm{isomcr}}=\frac{P_{\mathrm{NEET}}\cdot B\cdot N_K}{\lambda}(1-e^{-\lambda t_{\mathrm{irr}}}), \quad (1)$$

where P_{NEET} is the NEET probability, N_K the production rate of the K vacancies in ^{189}Os by irradiation, B the branching ratio for population of the isomer state, λ the disintegration constant of the isomer, and t_{irr} the irradiation time. The emitted L x rays (N_L) from the isomer is written as

$$N_L=N_{\mathrm{isomcr}}(e^{-\lambda t_0}-e^{-\lambda(t_0+t)}), \quad (2)$$

where t_0 and t are the start time and the end time of offline measurement, respectively.

In the monochromatic experiment, the total K vacancies produced in ^{189}Os were deduced from the online K x-ray measurement taking into account the self-absorption in the Os target, attenuation of K x rays in the Cu plate placed in front of the Ge detector, geometrical factors and the response function of the Ge detector. The detector efficiency was estimated from the intrinsic efficiency obtained with calibrated radioactive sources. The correction factors of attenuation and geometrical efficiency were calculated with the EGS4 code [15]. After corrections, the produced K vacancies are 1.65 $\times 10^{15}$ for thick target and 2.08×10^{15} for thin target, respectively. After similar corrections such as geometric factors for L x-ray measurement and self-absorption in the target, we obtain the number of emitted L x rays (N_L), and the NEET probability (P_{NEET}) for individual irradiation can be derived from Eqs. (1) and (2). The detection efficiency of the LEPS for L x-ray measurement was determined experimentally by using γ rays of several radioactive sources. The detected number of L x rays is at most 10. After corrections, the resulting NEET probability could not exceed an upper limit of $P_{\mathrm{NEET}}=4.1\times 10^{-10}$ for thick target and an upper limit of 2.8×10^{-10} for thin target. Otozai *et al.* [4] estimated the branching ratio B for feeding the isomer state from the 69.537 keV state to be 1.2×10^{-3}. All the experimental P_{NEET} values in Table I were obtained assuming the B value to be 1.2×10^{-3}. The values of the probabilities obtained in this experiment agreed with each other for individual irradiation and are consistent with the result of Ahmad *et al.* [8]. These are significantly lower than the values obtained from previous experiments. Most of the calculated values of the NEET probabilities in ^{189}Os are relatively high as shown in Table I, but the calculations by Tkalya [13] and Ahmad *et al.* [8] give values that are much closer to our experimental results.

On the other hand, the corrected numbers of K vacancies produced during the white beam irradiation are listed in Table II. The detected number of L x-rays from the decay of 30.81 keV isomer state are also listed. In the experiments used electrons or bremsstrahlung x rays and SR white beams for K-shell ionization, the NEET probability were very high. If the obtained L x rays after the corrections in this white beam experiment were fully attributable to the NEET process, the resulting probability could be 1.5×10^{-9} on average for the three runs. But the monochromatic beam experiment shows that it is much lower. The result from the white beam experiment is in agreement with that obtained by Shinohara *et al.* [6].

Recently, Kishimoto *et al.* [16] observed the NEET process in ^{197}Au using monochromatic x rays. The internal conversion electrons emitted from excited nuclei were detected with a silicon avalanche photodiode. The NEET probability was determined to be $(5.0\pm 0.6)\times 10^{-8}$. The common multipolarity in ^{197}Au is $M1$ which is same as that in ^{189}Os. The probability of the NEET process is very sensitive to the energy difference between the electron transition energy and the energy of nuclear transition. The energy differences are at least 1265 eV in ^{189}Os and only 51 eV in ^{197}Au, respectively. The large energy difference in ^{189}Os shows that the NEET probability in ^{189}Os should be extremely smaller than the one in ^{197}Au since the probability is inversely proportional to the square of the energy difference.

In summary, both monochromatic and white x-ray beam were used to study the NEET process in ^{189}Os. The present study gives an upper limit of $P_{\mathrm{NEET}}<4.1\times 10^{-10}$ from a measurement using 115 keV monochromatic x rays. The value is much lower than the previously obtained ones and is in agreement with calculations by Tkalya [13] and Ahmad *et al.* [8]. From white beam irradiation experiment, we demonstrated that the isomer in ^{189}Os was mainly populated via nuclear resonance absorption and that the result obtained by using white beam can not be considered to provide probabilities of the NEET process in ^{189}Os.

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