PHYSICAL REVIEW C, VOLUME 61, 051304(R)

Nuclear excitation by electronic transition in ^{189}Os

I. Ahmad, R. W. Dunford, H. Esbensen, D. S. Gemmell, E. P. Kanter, U. Rütt, and S. H. Southworth
Argonne National Laboratory, Argonne, Illinois 60439
(Received 28 January 2000; published 21 April 2000)

Monochromatic x rays have been used to explore the phenomenon of nuclear excitation by electronic transition (NEET) in the ^{189}Os atomic/nuclear system. A new theoretical approach to calculating this process has also been developed and predicts a value for the ''NEET probability,'' P_{NEET}, of 1.3×10^{-10}. P_{NEET} is the probability that a given atomic excitation (in this case a K vacancy), will result in the excitation of a specific nuclear state (in this case the 69.5-keV level in ^{189}Os). This value is much lower than most of the calculated values given in the literature for this system. Our measurement gives the result $P_{NEET}<9\times10^{-10}$, an upper limit which is several orders of magnitude lower than the values found in previous measurements, but which is consistent with the new calculation.

PACS number(s): 23.20.Nx, 21.10.Tg, 27.70.+q, 32.80.Hd

Nuclear excitation by electronic transition (NEET) is a rare but fundamental mode of decay of an excited atomic state. It is a process by which the energy of the atomic state is transferred via the exchange of a virtual photon into excitation of the atom's nucleus. It can only occur when the atomic and nuclear states have closely matching transition energies and also involve the same changes in spin and parity. The NEET process, which competes with the ''normal'' decay modes involving x-ray and/or Auger-electron emission, was first postulated in 1973 by Morita [1] and is similar to related processes seen in the decay of muonic atoms [2].

Consider an atomic system, such as the ^{189}Os that we discuss here, in which an atomic transition can occur between an initially excited K-vacancy state and a final M-vacancy state. Let us assume that the nucleus of that atom can undergo an excitation to a level involving the same changes in angular momentum and parity as are involved in the atomic transition. This situation is illustrated in Fig. 1. NEET can occur when the two product states are nearly degenerate. They are coupled by a residual interaction V_{em}, the electromagnetic interaction of the electron hole with the protons in the nucleus. Let φ_i denote the atomic wave function and ψ_j the nuclear wave function. Following the creation of a K hole, the initial state has a product wave function $|\alpha\rangle=|\varphi_K\psi_0\rangle$. The residual interaction generates an amplitude for the state $|\beta\rangle=|\varphi_M\psi_1\rangle$ and one can detect this component by measuring the nuclear decay. We can write the time evolution of the total wave function as

$$|\Phi(t)=a_\alpha(t)|\alpha\rangle+a_\beta(t)|\beta\rangle, \tag{1}$$

where the amplitudes a_α and a_β have initial ($t=0$) values of 1 and 0, respectively. We determine the two time-dependent amplitudes from the following coupled equations, which include the off-diagonal matrix element, $\kappa=\langle\alpha|V_{em}|\beta\rangle$, and the decay rates of both states explicitly:

$$i\hbar\frac{da_\alpha}{dt}=(E_\alpha-i\Gamma_\alpha/2)a_\alpha+\kappa a_\beta,$$
$$i\hbar\frac{da_\beta}{dt}=\kappa a_\alpha+(E_\beta-i\Gamma_\beta/2)a_\beta, \tag{2}$$

where (E_α,Γ_α) and (E_β,Γ_β) are the energies and decay widths of the two product states, $|\alpha\rangle$ and $|\beta\rangle$, respectively. The associated decay probabilities are

$$P_\alpha=\frac{\Gamma_\alpha}{\hbar}\int_0^\infty|a_\alpha(t)|^2dt,\quad P_\beta=\frac{\Gamma_\beta}{\hbar}\int_0^\infty|a_\beta(t)|^2dt. \tag{3}$$

The state $|\beta\rangle$ can decay either by a nuclear or by an electronic transition from the M vacancy. An electronic transition will, however, still result in a nuclear decay at a later time. Thus P_β is equal to the ''NEET probability'', P_{NEET}, defined as the probability that the decay of the initial excited atomic state will result in the excitation of and subsequent decay from the corresponding nuclear state.

The coupled equations (2) for the coefficients, a_α and a_β, can be solved analytically and this leads to an exact, if somewhat complex, expression for P_{NEET} ($=P_\beta$). For small κ, this expression reduces to

$$P_{NEET}^{\kappa\to0}=\frac{\Gamma_\alpha\Gamma_\beta}{\Gamma_\alpha}\frac{\kappa^2}{(E_\alpha-E_\beta)^2+\left(\frac{\Gamma_\alpha+\Gamma_\beta}{2}\right)^2}. \tag{4}$$

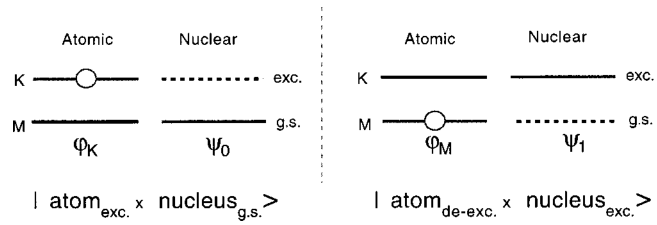

FIG. 1. Initial and final atomic and nuclear states involved in a NEET transition.

0556-2813/2000/61(5)/051304(5)/$15.00

TABLE I. Summary of published theoretical and experimental work on NEET in ^{189}Os.

THEORY			
		P_{NEET} $(M1)$	P_{NEET} $(E2)$
Morita [1]	1973	–	1×10^{-6}
Okamoto [3]	1977	–	1.5×10^{-7}
Pisk *et al.* [4]	1989	2.3×10^{-7}	1.8×10^{-8}
Bondar'kov *et al.* [5]	1991	1.1×10^{-7}	2.5×10^{-9}
Ljubicic *et al.* [6]	1991	1.06×10^{-7}	1.25×10^{-7}
Tkalya [7]	1992	1.1×10^{-10}	7×10^{-13}
Ho *et al.* [8]	1993	2.1×10^{-9}	–
Present work	2000	1.3×10^{-10}	3.8×10^{-13}
EXPERIMENT			
		Method	P_{NEET}
Otozai *et al.* [9]	1973	e^- bombardment 75–85 keV	1×10^{-6}
Otozai *et al.* [10]	1978	e^- bombardment 72–100 keV	$(1.7\pm0.2)10^{-7}$
Saito *et al.* [11]	1981	200-keV bremsstrahlung	$(4.3\pm0.2)10^{-8}$
Shinohara *et al.* [12]	1987	''White'' synchrotron radiation	$(5.7\pm1.7)10^{-9}$
Lakosi *et al.* [13]	1995	300-keV bremsstrahlung	$(2.0\pm1.4)10^{-8}$
Present work	2000	Monochromatic 100-keV x rays	$<9\times10^{-10}$

Estimates of P_{NEET} for various atomic/nuclear systems have been given by several authors [1,3–8], beginning with Morita [1]. Many of the early estimates involved the use of simplifying approximations that led to results at considerable variance with Eq. (4). Also, it was not recognized at first that P_{NEET} tends to be significantly higher for $M1$ transitions than for $E2$ transitions, mainly because of the involvement of atomic s states, which leads to a stronger coupling. The more recent calculations of Tkalya [7] give values that are close to those obtained from Eq. (4).

The system that has received the most experimental attention in NEET studies is ^{189}Os [9–13]. This is because it is a heavy system (more compact atomic wave functions) and because it offers a convenient signature for excitation of the $5/2^-$ nuclear state at 69.537 keV in that this state decays promptly (1.6 ns) with a partial branch ($\sim1.2\times10^{-3}$, see Ref. [10]) to a lower-lying metastable (6-h half-life) $9/2^-$ state at 30.814 keV, which in turn decays primarily by internal conversion and can readily be measured. (Unless otherwise specified, we use the values given in Refs. [14,15] for the nuclear properties of ^{189}Os.) For these reasons and also to make comparisons with previous experimental and theoretical work, we too have made measurements and calculations for the case of ^{189}Os. We exploit the large fluxes of tunable, monochromatic, high-energy x rays now available from a third-generation synchrotron radiation source.

In the NEET process in ^{189}Os that we consider here, an initial K-vacancy state decays via an electronic transition from the M shell. The KM_{I} (70.822 keV, $M1$), KM_{IV} (71.840 keV, $E2$), and KM_{V} (71.911 keV, $E2$) atomic transitions [16] can contribute. The corresponding nuclear state at 69.537 keV can be excited via $M1$ or $E2$ transitions from the $3/2^-$ nuclear ground state.

The theoretical expression for the electromagnetic ($M1$ or $E2$) coupling matrix element that we have used is

$$\kappa^2=4\pi e^2 B\left(\Pi L,\frac{3}{2}^-\rightarrow\frac{5}{2}^-\right)\times\left\langle j_K\frac{1}{2}L0\middle|j_M\frac{1}{2}\right\rangle^2\left(\frac{q^{L+1}|m_{\Pi L}(q)|}{(2L+1)!!}\right)^2, \qquad (5)$$

where ΠL represents $M1$ or $E2$, and q ($=35.24$ Å^{-1}) is the wave number of the nuclear transition. The atomic matrix elements $m_{\Pi L}(q)$, defined by Eq. (12) of Ref. [7], were calculated using wave functions from the GRASP2 code [18], and tabulated values [14] of $B(M1)$ and $B(E2)$ were used for the nuclear transition. The Clebsch-Gordan coefficient refers to the total angular momenta j_K of the K-vacancy and j_M of the M-vacancy states.

Inserting the calculated values of κ together with the atomic transition energies given above, and the calculated atomic level widths of Ref. [17], we obtain the following NEET probabilities:

$$P_{NEET}(M1)=1.3\times10^{-10} \quad \text{and} \quad P_{NEET}(E2)=3.8\times10^{-13}.$$

Previous experimental determinations of P_{NEET} in ^{189}Os were achieved employing a variety of technical approaches to produce the initial K-vacancy states (see Table I). Each of these techniques has certain inherent difficulties. For example, use of an electron beam can cause direct Coulomb excitation of the nuclear state and it is hard to distinguish this component from that due to the NEET process. Similarly, the use of a broad continuous spectral distribution of synchrotron or bremsstrahlung x rays (''white light'') results in a contribution from direct nuclear photoabsorption into the nuclear state or indeed into a range of nuclear levels that can feed that state or the lower-lying metastable state. In the present work, we employed x-ray beams from a wiggler operated by the Basic Energy Sciences Synchrotron Radiation

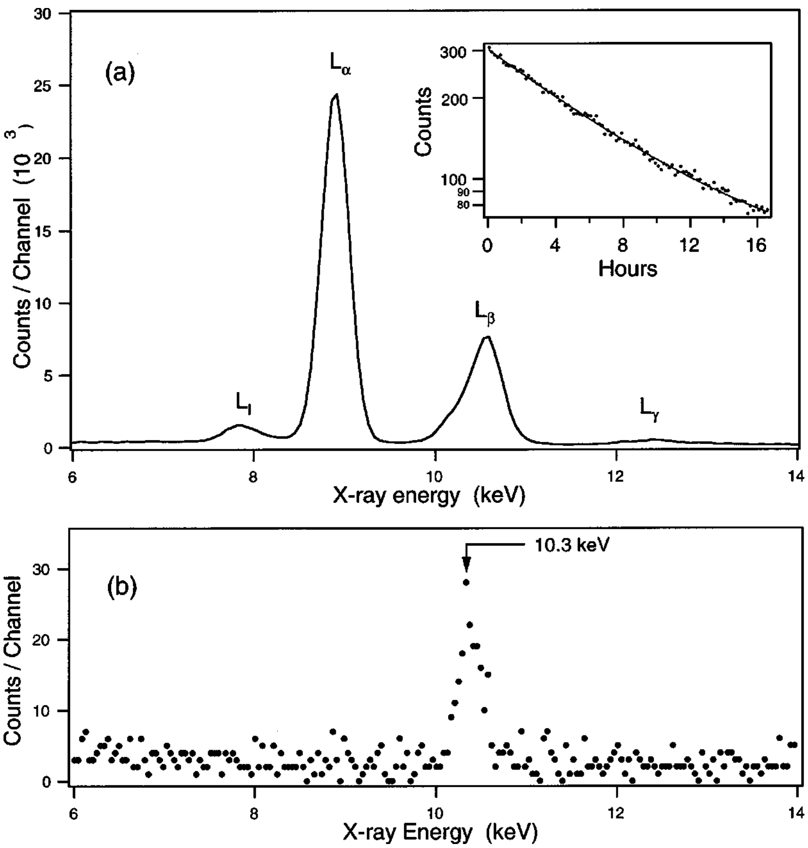

FIG. 2. (a) The osmium L x-ray spectrum obtained in the initial runs after irradiation with a white x-ray beam. The inset shows a corresponding measurement of the 6-h decay of this radiation together with a fitted curve of the form $a+be^{-t/\tau}$. (b) The x-ray spectrum summed for two targets irradiated with 98.74-keV x rays as described in the text. The total irradiation time was 43.9 h and the total counting time was 37.4 h.

Center [19] at the Advanced Photon Source at Argonne National Laboratory. After initial measurements with a white beam, in which the difficulties mentioned above became very apparent, we switched to the use of a monochromatic 98.74-keV x-ray beam to produce the K-vacancy states. This beam (5×10^{11} photons/s) was formed by Bragg diffraction from a single (440) Si crystal placed in the wiggler beam. The diffraction angle was $2\theta=7.5°$. This particular beam energy was chosen because it lies above the osmium K edge at 73.9 keV [16], does not lie near the energy of a nuclear level in ^{189}Os, and corresponds to a convenient and intense diffraction. The energy width of the beam was about 0.1% (100 eV) and the beam-spot size at the target was 0.2 mm wide and 4 mm high. The incident x-ray beam contained a comparably intense component at 49.37 keV (below the osmium K edge) and also a few-percent component at 148.1 keV. None of the beam components had energies overlapping any of the ^{189}Os nuclear level energies, thereby avoiding problems with nuclear resonant absorption.

The 30.814-keV metastable nuclear state of ^{189}Os decays by internal conversion. This decay was measured off-line using a Ge (LEPS) detector to count the L x-ray spectrum associated with the L-conversion electrons. Figure 2(a) shows the L x-ray spectrum obtained in the initial runs using a white beam. The inset shows a corresponding measurement of the decay of this radiation. The half-life of the 30.8-keV state was measured to be 5.65 ± 0.15 h, in good agreement with the tabulated value [14] of 5.8 ± 0.1 h.

The monochromated x-ray beam from the wiggler was incident upon a thin (9.3 mg/cm^2) layer of isotopically separated (95.3%) metallic ^{189}Os electroplated onto a 0.015-in.-thick Cu disk using the method of Stuchbery [20]. Individual targets were irradiated in this fashion for periods of about 20 h. Large numbers of K vacancies were produced, some of which were expected to lead via NEET to the 69.5-keV state and thence to the 30.8-keV metastable state of the nucleus. The number of OS K vacancies generated was monitored by on-line observation of the K x rays using a Ge detector.

After irradiation the targets were removed and the L x rays associated with decays of the metastable state were detected in a low-background shielded underground counting room where counting with a Ge detector proceeded also for about 20 h. Figure 2(b) shows the results summed for two targets and a total counting time of 37.4 h. From a comparison with Fig. 2(a), it is apparent that within the sensitivity of this measurement, there was no evidence of the x rays that accompany the decay of ^{189}Os metastable state. [The peak at 10.3 keV in Fig. 2(b) is due to weak natural background radiation and is only observable in extremely well shielded conditions.]

After taking into account factors such as the number of K holes created during the irradiation (and their decay), the branching ratio for feeding the metastable state from the 69.5-keV state, geometrical factors, the emission probability for L x rays in the isomeric decay [12], self-absorption in the target, etc., we obtained the result $P_{NEET}<9\times10^{-10}$. This value is significantly smaller than the various values obtained in previous measurements and predicted by previous calculations (see Table I). It is, however, consistent with our

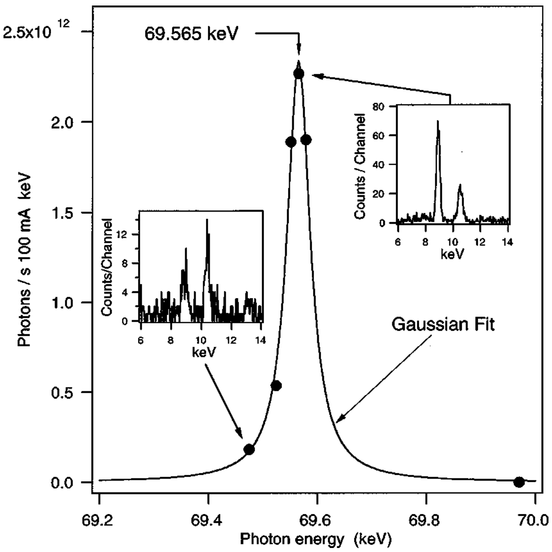

FIG. 3. The effective number of incident photons/s/keV per 100 mA ring current at the nuclear resonance energy deduced from the measured production of isomeric decays resulting from nuclear photoabsorption as the x-ray beam energy was scanned across the 69.5-keV level energy. The result is the energy profile of the incident beam shown plotted here and fitted with a Gaussian. The insets show the *L* x-ray spectra at two representative points. Note that the spectrum obtained at the peak is similar to that in Fig. 2(a), whereas the one taken at the lowest beam energy is dominated by the weak 10.3-keV background peak exhibited in Fig. 2(b).

calculated value derived from Eq. (4) namely,

$$P_{NEET}(M1) = 1.3 \times 10^{-10}.$$

To demonstrate that our irradiation and detection systems were functioning properly and that our various assumptions about the atomic and nuclear properties of ^{189}Os were correct, we changed monochromator crystals and switched to a nominal beam energy of 69.8 keV. This beam was used to measure nuclear resonant absorption into the 69.5-keV state in ^{189}Os. The beam energy was varied about the resonance energy by means of small rotations of a (400) Si monochromator crystal in Laue geometry in the wiggler beam. This resulted in small (a few mm) variations in the position of the beam at the target and these movements were accommodated by corresponding linear translations of the target.

Six x-ray beam energies were used in the scan ranging from 69.47 to 69.97 keV. At each energy, an irradiation of about 20 h was followed by a similar counting period. The number of L_α x rays from the decay of the 30.8-keV metastable state was measured and from this the number of incident photons/s/keV per 100 mA ring current at the nuclear resonance energy was deduced. The result was an energy profile of the incident beam (the energy width of the nuclear level is 4×10^{-10} keV) and is shown plotted in Fig. 3. (The insets in Fig. 3 show the *L* x-ray spectra at two representative points in the scan.) The absorption was found to be a maximum at a beam energy of 69.565 keV. We used a Ge detector on-line to determine the energy of the Rayleigh-scattered photons and, in addition, we measured absorption over the *K* edge of tungsten (69.525 keV [16]). Based on these measurements, we estimate the error in the beam-energy determination to be about ± 30 eV (primarily a systematic error—the relative errors are much smaller). The resonance energy is thus in reasonable agreement with the tabulated value [14] of 69.537 keV. Also, the energy width measured for the incident beam (51 eV) and the integrated intensity (1.3×10^{11} photons/s/100 mA) both agree with the calculated values. Thus the experimental technique and also the values taken for the atomic and nuclear properties of ^{189}Os (in particular the branching ratio for feeding the $9/2^-$ metastable state) appear to be sound.

In summary, a new measurement using monoenergetic x rays and a new calculation of P_{NEET} in ^{189}Os have been performed. Both result in values that are significantly lower than previously obtained. The measurement sets an upper limit for P_{NEET} of 9×10^{-10}, consistent with our calculated value of 1.3×10^{-10}. As part of the work, we have also determined a new value of 5.65 ± 0.15 h for the half-life of the 30.8-keV state in ^{189}Os.

We gratefully acknowledge the help of F. Markun, J. Greene, and A. Stuchbery in the preparation of the targets. P. Montano, M. Beno, and G. Jennings provided valuable assistance in beamline operations. This work was supported by the U.S. Department of Energy under Contract No. W-31-109-ENG-38.

[1] M. Morita, Prog. Theor. Phys. **49**, 1574 (1973).
[2] See, for example, S. Bernow *et al.*, Phys. Rev. Lett. **21**, 457 (1968), and references therein.
[3] K. Okamoto, in *Laser Interactions and Related Plasma Phenomena*, edited by H. J. Schwartz and H. Hora (Plenum, NY, 1977) p. 283.
[4] K. Pisk, Z. Kaliman, and B. A. Logan, Nucl. Phys. **A504**, 103 (1989).
[5] M. D. Bondar'kov and V. M. Kolomietz, Izv. Akad. Nauk SSSR, Ser. Fiz. **55**, 983 (1991).
[6] A. Ljubicic, D. Kekez, and B. A. Logan, Phys. Lett. B **272**, 1 (1991).
[7] E. Tkalya, Nucl. Phys. **A539**, 209 (1992).
[8] Y-k. Ho *et al.*, Phys. Rev. C **48**, 2277 (1993).
[9] K. Otozai, R. Arakawa, and M. Morita, Prog. Theor. Phys. **50**, 1771 (1973).
[10] K. Otozai, R. Arakawa, and T. Saito, Nucl. Phys. **A297**, 97 (1978).
[11] T. Saito *et al.*, J. Inorg. Nucl. Chem. **43**, 1963 (1981).
[12] A. Shinohara *et al.*, Nucl. Phys. **A472**, 151 (1987).
[13] L. Lakosi, N. C. Tam, and I. Pavlicsek, Phys. Rev. C **52**, 1516 (1995).
[14] R. B. Firestone, Nucl. Data Sheets **59**, 869 (1990).
[15] S. G. Malmskog, V. Berg, and A. Bäcklin, Nucl. Phys. **A153**, 316 (1970).
[16] See, for example, G. P. Williams, X-ray Data Booklet, Center

for X-ray Optics, Lawrence Berkeley Laboratory, PUB-400 Rev, 1986.

[17] See, for example, H. J. Leisi *et al.*, Helv. Phys. Acta **34**, 161 (1961); E. J. McGuire, Phys. Rev. A **5**, 1043 (1972).

[18] F. A. Parpia, C. F. Fischer, and I. P. Grant, Comput. Phys. Commun. **94**, 249 (1996), and references therein.

[19] P. A. Montano, G. S. Knapp, G. Jennings, E. Gluskin, E. Trakhtenberg, I. B. Vasserman, P. M. Ivanov, D. Frachon, E. R. Moog, L. R. Turner, G. K. Shenoy, M. J. Bedzyk, M. Ramanathan, M. A. Beno, and P. L. Cowan, Rev. Sci. Instrum. **66**, 1839 (1995); P. A. Montano, Y. Lin, U. Rütt, M. A. Beno, G. Jennings, and C. W. Kimball, Proc. Int. Soc. Opt. Eng. **3773**, 262 (1999).

[20] A. E. Stuchbery, Nucl. Instrum. Methods Phys. Res. **211**, 293 (1983).