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Nuclear Excitation by Electronic Transition (NEET)

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Abstract

We present a report on recent measurements using the Advanced Photon Source at Argonne National Laboratory to explore the phenomenon of Nuclear Excitation by Electronic Transition (NEET) in the ^{189}Os atomic/nuclear system.

Nuclear Excitation by Electronic Transition (NEET) is a fundamental (but rare) 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 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 mu-mesonic atoms [2].

A typical NEET situation is shown in Fig. 1 where we consider an atomic transition occurring between an initial K-vacancy state and a final M-vacancy state.

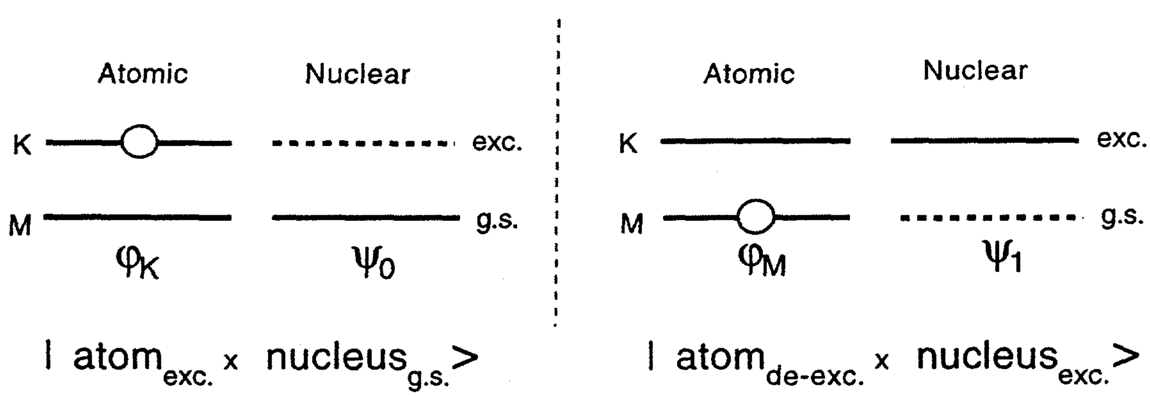


Fig. 1 Initial and final atomic and nuclear states involved in a NEET transition.

NEET can occur when these 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. Following the creation of an initial K-hole state $\varphi_K\psi_0$, the residual interaction generates an amplitude for the state $\varphi_M\psi_1$ and one can detect this component by measuring the nuclear decay. We can write the time evolution of the total wavefunction as

$$\Phi(t) = a_K(t)\varphi_K\psi_0 + a_M(t)\varphi_M\psi_1 \quad , \tag{1}$$

where the amplitudes a_K and a_M 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, $\kappa = \langle \varphi_K\psi_0 | V_{em} | \varphi_M\psi_1 \rangle$, and the decay rates of both states explicitly:

$$\begin{aligned} i\frac{da_K}{dt} &= (E_K - i\Gamma_K/2)a_K + \kappa a_M \quad , \\ i\frac{da_M}{dt} &= \kappa a_K + (E_M - i\Gamma_M/2)a_M \quad , \end{aligned} \tag{2}$$

where (E_K,Γ_K) and (E_M,Γ_M) are the energies and decay widths of the two product states, $\varphi_K\psi_0$ and $\varphi_M\psi_1$, respectively. The associated decay probabilities are

$$P_K = \Gamma_K \int_0^\infty |a_K(t)|^2 dt, \qquad P_M = \Gamma_M \int_0^\infty |a_M(t)|^2 dt \quad . \tag{3}$$

The M-hole state $\varphi_M\psi_1$ can decay either by a nuclear decay or by an electronic transition. An electronic transition will, however, still result in a nuclear decay at a later time. Thus P_M 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 (Eq. 2) for the coefficients, a_K and a_M, can be solved analytically and this leads to an exact, if somewhat complex, expression for P_{NEET} $(=P_M)$. For small κ, this expression reduces to

$$P_{NEET}^{\kappa\to 0} = \frac{\Gamma_K + \Gamma_M}{\Gamma_K} \frac{\kappa^2}{(E_K - E_M)^2 + \left(\frac{\Gamma_K + \Gamma_M}{2}\right)^2} \quad . \tag{4}$$

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 have a stronger overlap with the nucleus. 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 [9 – 13] is ^{189}Os. This is because it is a heavy system (more overlap between atomic wavefunctions and the nucleus) and because it offers a convenient signature for excitation of the nuclear state in that this state decays promptly (1.6 ns) with a partial branch (~1.2 x 10^{-3}) to a lower-lying metastable (6-hr. half-life) state, which in turn decays, primarily by internal conversion, and can readily be measured. 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 high-energy x-rays now available from a third-generation synchrotron radiation source. Figure 2 shows the atomic and nuclear levels involved.

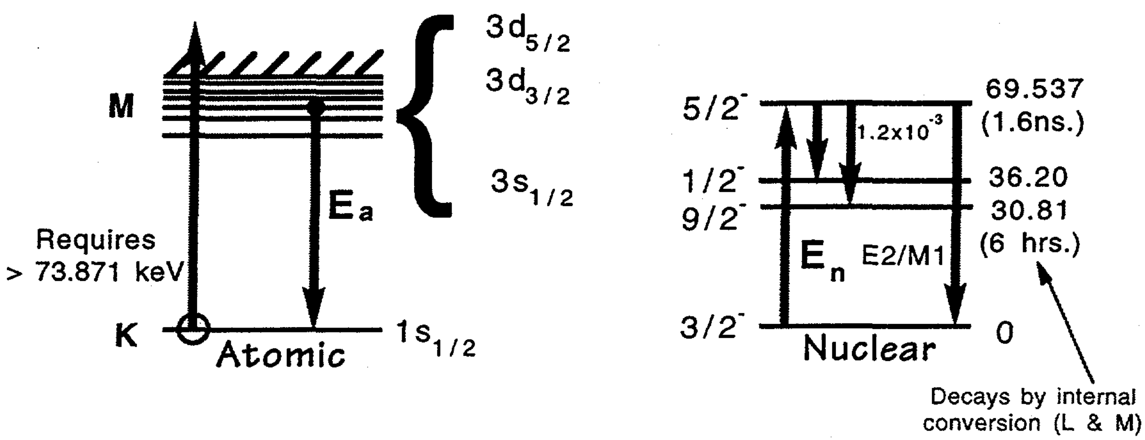


Fig. 2 Atomic and nuclear levels involved in NEET in ^{189}Os

In this NEET process in ^{189}Os, an initial K-vacancy state decays via an electronic transition from the M-shell. The corresponding nuclear state that is excited is the $5/2^-$ state at 69.537-keV. The transition from the nuclear ground state is known [14] to proceed via E2 or M1. Thus the KM_I (70.822 keV, M1) and the KM_{IV} (71.840 keV, E2) and KM_V (71.911 keV, E2) atomic transitions can contribute.

In Table I we list the values of P_{NEET} that have been both calculated and measured for ^{189}Os. In cases where both M1 and E2 transitions have been calculated, the table shows both values.

The theoretical values given in Table I for the "present work" are derived from Eq. 4 using the atomic transition energies given above [15], the calculated atomic level widths [16], the tabulated values of B(E2) and B(M1) for the nuclear transition [14] (employed to obtain the nuclear matrix elements), and atomic matrix elements calculated using wavefunctions from the "Grasp2" code [17].

One notices that the values, both calculated and measured, for P_{NEET} in ^{189}Os have exhibited a steady decrease as a function of time. We surmise that this trend reflects continuing improvements in calculational and experimental techniques.

The previous experimental determinations of P_{NEET}, listed in Table I, were achieved employing a variety of technical approaches to produce the initial K-vacancy states. Each of these techniques has certain inherent difficulties. For example, use of an electron beam causes direct Coulomb excitation of the nuclear state and it is hard to distinguish this component from 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 photo-absorption into the nuclear state or indeed into a range of nuclear levels that can feed the metastable state. In the present work, we employed x-ray beams from Argonne's Advanced Photon Source (APS). After initial measurements with a white beam, in which the difficulties mentioned above became very apparent, we switched to the use of a monochromatic 100-keV x-ray beam to produce the K-vacancy states. The nominal 100-keV x-ray beam (5 x 10^{11} photons/s) was formed by Bragg diffraction from a single (440) Si crystal placed in the BESSRC wiggler beam. The energy width of the beam was about 0.1% (100 eV). The incident beam also contained a comparably intense component at 50 keV (below the K-threshold for Os) and also a few-percent component at 150 keV. None of the beam components had energies overlapping any of the ^{189}Os nuclear level energies, thereby avoiding problems with nuclear resonant absorption.

TABLE I

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	1998	1.3×10^{-10}	3.8×10^{-13}

Experiment

			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	$(1.7\pm0.2)\times10^{-7}$
Saito et al. [11]	1981	200-keV bremsstrahlung	$(4.3\pm0.2)\times10^{-8}$
Shinohara et al. [12]	1987	"White" synchrotron radiation	$(5.7\pm1.7)\times10^{-9}$
Lakosi et al. [13]	1995	300-keV bremsstrahlung	$(2.0\pm1.4)\times10^{-8}$
Present work	1998	Monochromatic 100-keV x-rays	$< 9\times10^{-10}$

The APS is one of three recently constructed high-energy third-generation synchrotron light sources. It is a companion to the 6-GeV storage ring at Grenoble, France (ESRF) and the 8-GeV ring in Hyogo, Japan (SPring-8). The APS stores 100 mA of 7-GeV positrons or electrons. Ground-breaking for the APS occurred in June, 1990 and the first x-ray beams were produced in March, 1995. The storage ring has 40 sectors, of which 35 are dedicated to producing x-ray beams for users. Each of these sectors has one bending-magnet x-ray source and one insertion-device source. We used an elliptical multipole wiggler located at Sector 11 operated by the Basic Energy Sciences Synchrotron Radiation Center (BESSRC). This source produces intense beams of high-energy x-rays, whose polarization can be switched at a 10 Hz rate if desired. Further details on the APS and BESSRC facilities are given in Ref. 18, and also at the APS website: http://www.aps.anl.gov

The 31-keV nuclear metastable state of ^{189}Os decays by internal conversion. This decay was measured using a Ge detector to count the L x-ray spectrum associated with the L-conversion electrons. In Fig. 3 we show the L x-ray spectrum obtained in the initial runs using a white beam. Figure 4 shows the corresponding measurement of the 6-hr. decay of this radiation.

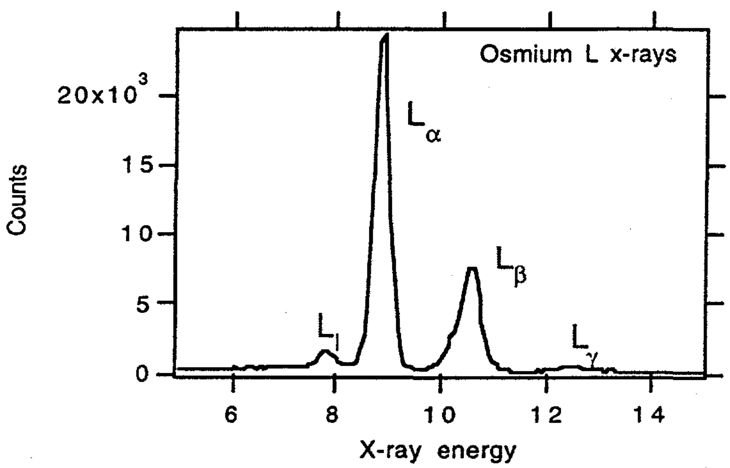


Fig. 3

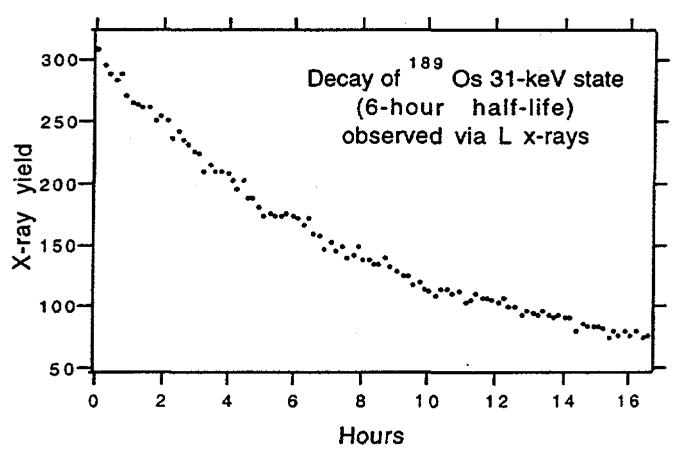
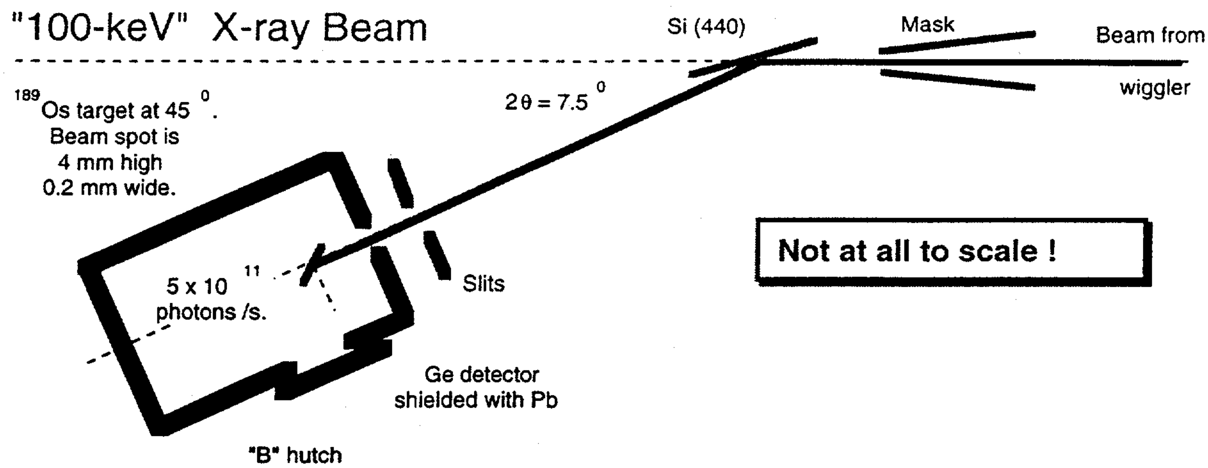


Fig. 4

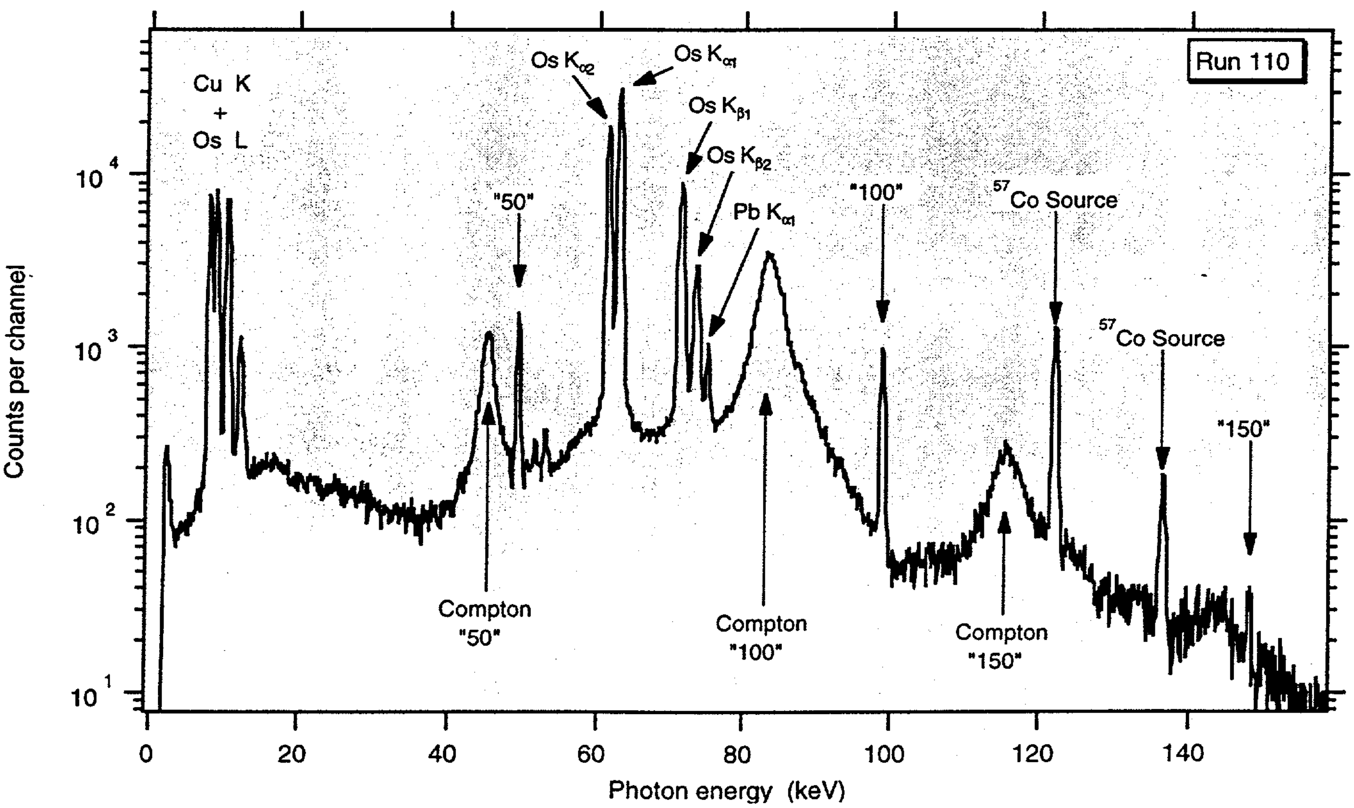
The monochromated x-ray beam from the wiggler was incident upon a thin (approx. 9 mg/cm^2) layer of isotopically separated (95.3%) metallic ^{189}Os electroplated onto a 0.015"-thick Cu disk using the method of Stuchbery [19]. Figure 5 is a schematic illustration of the arrangement used for the irradiations. Individual targets were irradiated in this fashion for periods of about 20 hours. Large numbers of K-vacancies were produced, some of which were expected to lead via NEET to the 70-keV state and then to the 31- 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 (Fig. 6 shows a spectrum).

After irradiation the targets were removed and the L x-rays associated with decays of the metastable state were detected in a shielded underground counting room where counting with a Ge detector proceeded also for about 20 hours. Figure 7 shows the results summed for two targets and a total counting time of 37.4 hours. For comparison, the insert shows the spectrum expected for the Os L x-rays



"100-keV" X-ray Beam
Si (440)
Mask
Beam from
wiggler
^{189}Os target at 45 ^{0}.
Beam spot is
4 mm high
0.2 mm wide.
2θ = 7.5 ^{0}
5 x 10 ^{11}
photons /s.
Slits
Not at all to scale !
Ge detector
shielded with Pb
"B" hutch

Fig. 5



Run 110
Cu K
+
Os L
Os $K_{\alpha 2}$
Os $K_{\alpha 1}$
Os $K_{\beta 1}$
Os $K_{\beta 2}$
Pb $K_{\alpha 1}$
"50"
"100"
^{57}Co Source
^{57}Co Source
"150"
Compton
"50"
Compton
"100"
Compton
"150"
Counts per channel
Photon energy (keV)

Fig. 6

(also shown in Fig. 3). Within the sensitivity of this measurement, there was no evidence of the x-rays that follow the nuclear decay of ^{189}Os (the peak at 10.3 keV is from weak background radiation from the molecular sieve in the detector – only observable in very well shielded conditions).

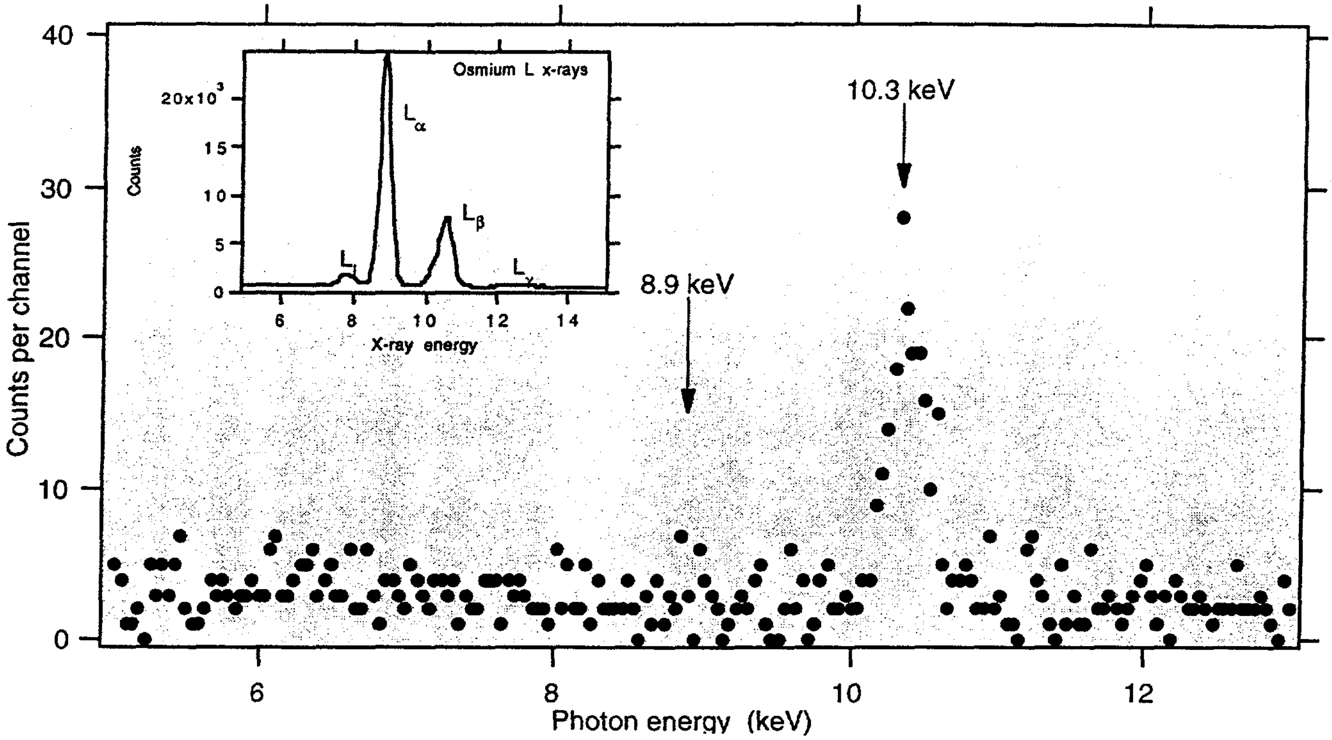


Fig. 7

After allowing for self-absorption in the target, 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, and other factors, we obtain $P_{NEET} < 9 \times 10^{-10}$. This value is significantly smaller than those obtained in previous measurements (Table I) and is consistent with our calculated value for P_{NEET} (M1) = 1.3×10^{-10}.

To demonstrate that our irradiation and detection systems were functioning correctly, we switched

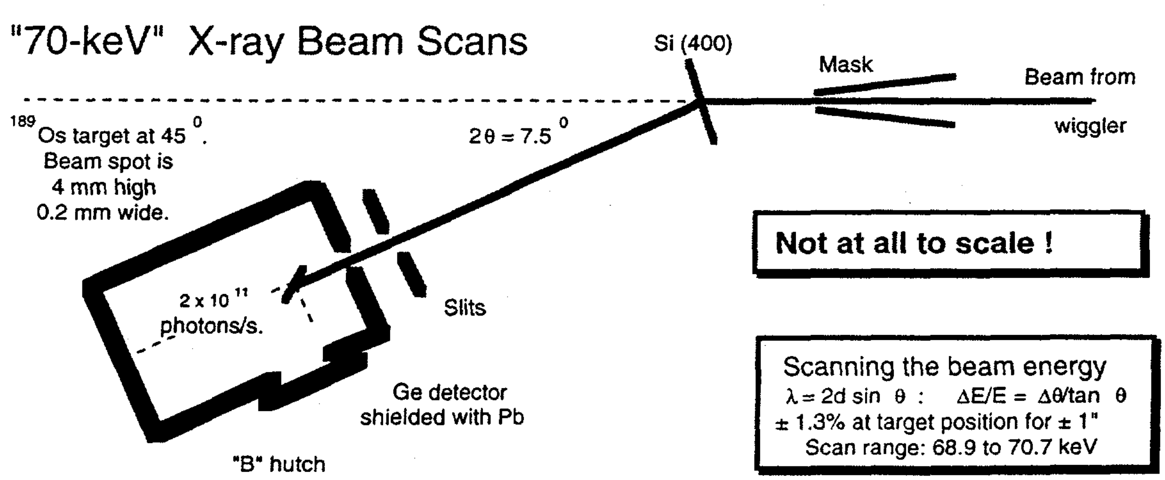


Fig. 8

to a monochromatic 70-keV beam and measured the nuclear resonant absorption into the 70-keV state in ^{189}Os. The beam energy was scanned by rotation of a (400) Si Laue monochromator crystal. Figure 8 shows the experimental arrangement and Fig. 9 is a spectrum recorded online with the Ge detector.

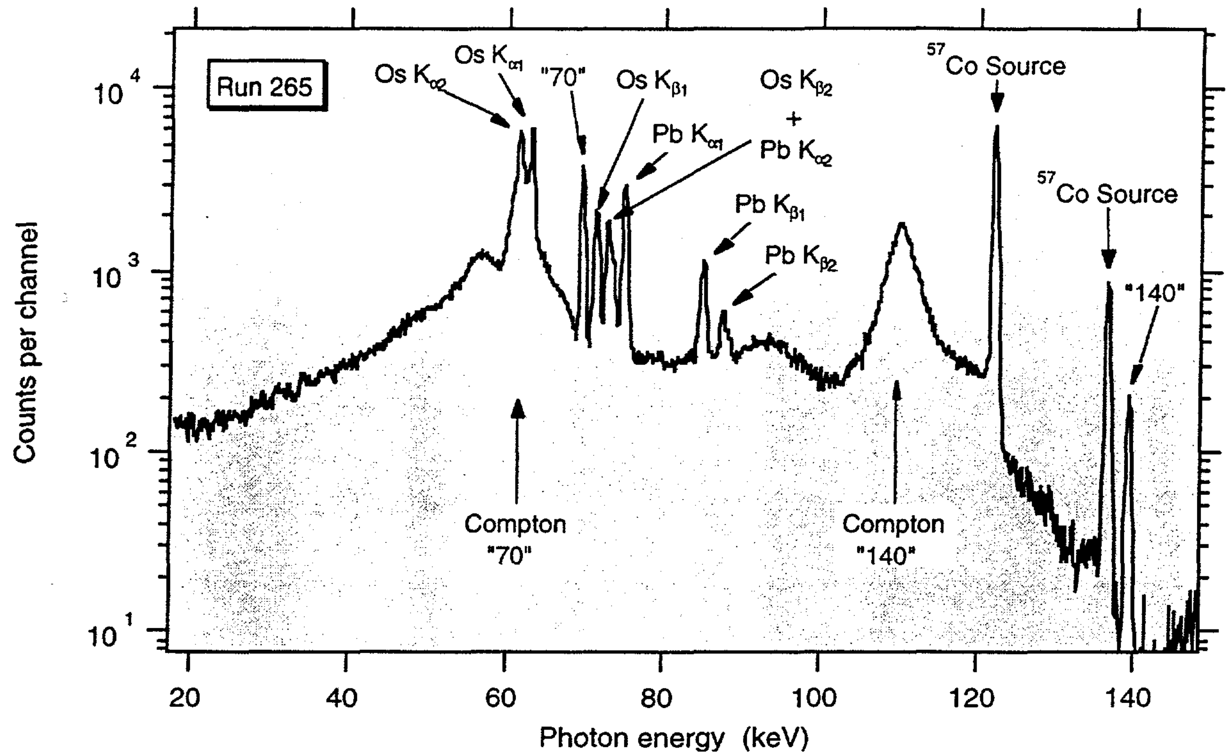


Fig. 9

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 hours was followed by a similar counting period. The number of L x-rays from the metastable decay was measured and from this the number of incident photons/s/keV per 100 mA ring

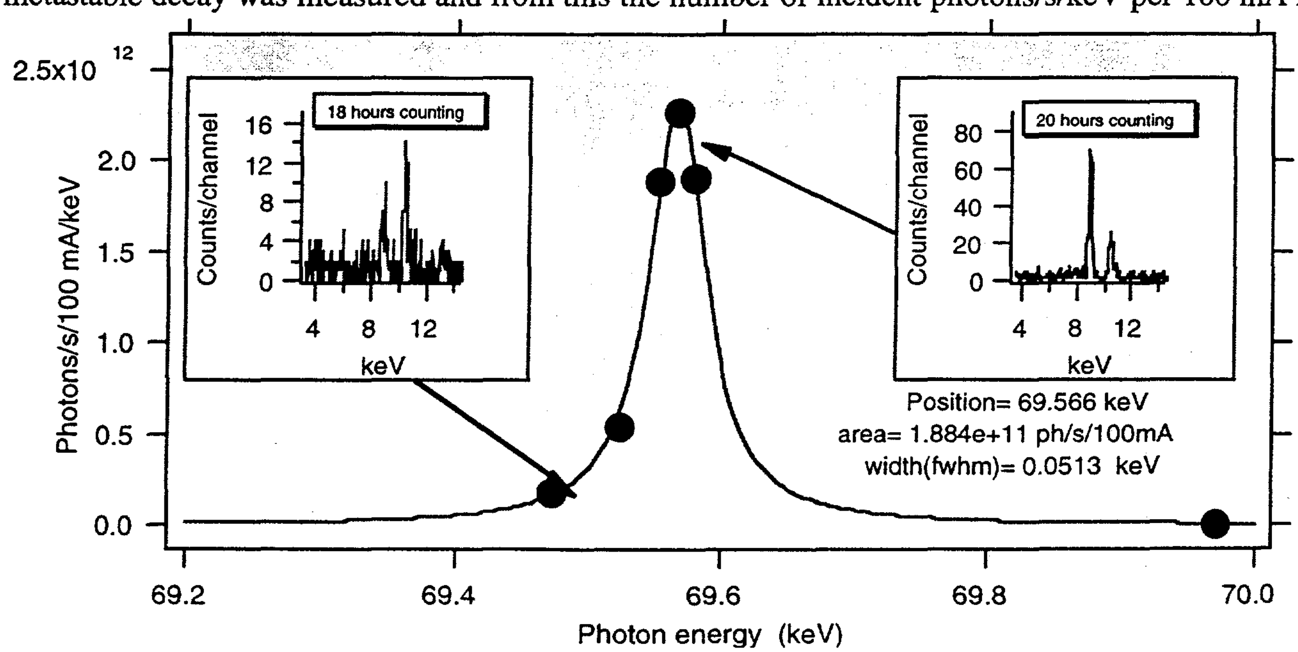


Fig. 10

current at the nuclear resonance energy was deduced (the energy width of the nuclear level is 4 x 10^{-10} keV [14]). The result was an energy profile of the incident beam and is shown plotted in Fig. 10. (The inserts show the L x-ray spectra at a couple of representative points.) One sees that the absorption is a maximum at a beam energy of 69.566 keV 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 (1.9 x 10^{11} photons/s/100 mA) both agree with the calculated values. Thus the experimental technique appears to be sound.

Conclusion

A new measurement and a new calculation of P_{NEET} in ^{189}Os have been performed. Both values are significantly lower than previously obtained. The measurement sets an upper limit for P_{NEET} of 9 x 10^{-10}, consistent with our calculated value of 1.3 x 10^{-10}.

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