

# Probability of nonradiative excitation of nuclei in transitions of an electron in an atomic shell

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The probability of nonradiative excitation of nuclei in atomic transitions (the so-called NEET process) is derived on the basis of QED. The existing experimental and theoretical results for the nuclei  $^{189}\text{Os}$ ,  $^{197}\text{Au}$  and  $^{237}\text{Np}$ , are reviewed and analyzed. The theoretical estimates of the probabilities differ significantly from the experimentally observed values. It is shown that the interpretation of the experimental results on excitation of a low-lying (76.8 eV) isomeric state of  $^{235}\text{U}$  in a laser plasma with the help of the NEET mechanism is incorrect. The possibility of measuring the NEET probability in coincidence experiments is discussed.

## 1. INTRODUCTION

Nuclear excitation by electron transition (NEET) was proposed by M. Morita in 1973 as a highly efficient process for exciting atomic nuclei into isomeric states when one of the inner atomic shells is ionized.<sup>1</sup> The crux of the phenomenon consists of the following. If an atom is capable of a transition which is close in energy to and has the same multipolarity as a nuclear transition, then besides the standard channels for deexcitation of the atom—x-ray and Auger emission—the process of nonradiative excitation of the nucleus by direct transfer of energy to the nucleus from the atomic shell is also possible.

The process of nonradiative excitation of a nucleus, but for muon transitions in  $\mu$ -mesic atoms, was itself first studied by D. F. Zaretskii back in 1959.<sup>2</sup> The phenomenon was subsequently studied in many experimental and theoretical works, especially in the 1960s. The interaction of a nucleus with an electron shell was studied in detail in different orders of perturbation theory by V. A. Krutov. In particular, in Ref. 3 Krutov and Fomenko considered, among other processes, the deexcitation of a nucleus by a process inverse to NEET. The method proposed in Ref. 3 can be used (and is used) for studying different nuclear excitation mechanisms.

The theoretical calculations made by M. Morita in Ref. 1 indicate that the NEET mechanism has a high efficiency. In a subsequent series of experimental papers<sup>4–9</sup> the excitation of low-lying levels of the nuclei  $^{189}\text{Os}$ ,  $^{197}\text{Au}$ , and  $^{237}\text{Np}$  accompanying irradiation of targets consisting of thin foils of these metals by electrons and photons with energies sufficient to ionize the *K* shells of the atoms was explained by the NEET process. The results of the experiments of Refs. 10 and 11 on the formation of the isomeric nuclei  $^{235m}\text{U}$  in a hot plasma, produced on the surface of natural uranium by  $\text{CO}_2$  laser radiation, were interpreted with the help of the same mechanism.

There are a number of theoretical treatments of the NEET process. In Ref. 12 calculations, on the whole confirming the results obtained in Refs. 1 and 4–11, were performed and nuclei meeting the requirements for NEET were proposed. D. P. Grechukhin and A. A. Soldatov pointed out,<sup>13</sup> however, that the expression employed in Refs. 1 and 12 for the electron-nucleus interaction Hamiltonian is not entirely correct (essentially because the nuclear current was determined incorrectly) and consequently the results were much too high. Indeed, the Hamiltonian in Ref. 1 had the

form

$$H_{int} = - \frac{Ze^2}{|\mathbf{r} - \mathbf{R}_p|} \quad (1a)$$

instead of the usual form (see, for example, Refs. 2 and 13)

$$H_{int} = - \sum_{p=1}^z \frac{e^2}{|\mathbf{r} - \mathbf{R}_p|}, \quad (1b)$$

where  $Z$  is the charge of the nucleus and  $\mathbf{r}$  and  $\mathbf{R}_p$  are, respectively, the radius vectors of the electron and one of the protons in the nucleus. After expanding the denominator in Eq. (1a) in multipoles the following formula was obtained for the interaction energy:

$$E_{int} = - \frac{4\pi Ze^2}{2L+1} \langle R^L \rangle \langle r^{-L-1} \rangle, \quad (2)$$

where  $L$  is the multipolarity of the nuclear transition. Next, the  $L$ th power of the radius of the nucleus  $R_0 = 1.2A^{1/3}$  fm, where  $A$  is the atomic weight of the element, was substituted for the average value  $\langle R^L \rangle$ , and the quantity  $\langle r^{-L-1} \rangle$  was estimated as  $r_n^{-L-1}$ , where  $r_n$  is the radius of the Bohr orbit with  $n = (n_i + n_f)/2$  and  $n_{i(f)}$  are the principal quantum numbers of the initial and final atomic states. The presence of  $Z$  in Eq. (1a), the substitution of  $R_0$  for  $\langle R \rangle$ , and the fact that the angular parts of the matrix elements were neglected resulted, as will be shown below, in an overestimate of the probability of NEET by several orders of magnitude.

An attempt at a relativistic calculation was made in Ref. 14. The fifth-order diagram in  $e$  was studied (the Coulomb interaction between the electrons in the shell in the initial state was taken into account). The use of hydrogen-like electronic wave functions and some other unjustified approximations (see the analysis in Ref. 15), however, resulted in overestimation of the NEET probability.

The contribution of third-order diagrams in  $e$  to the probability of nonradiative excitation of the nucleus was calculated in Ref. 15. It was assumed that the smallness introduced by the extra electron-photon vertex in many cases can be compensated by a relative increase in the probability of excitation of the nucleus due to an increase in the number of accessible final states as a result of photon emission. It will be shown in Sec. 2 that taking into account the finite widths of the atomic levels in the second-order diagram does indeed

result in an expression for the probability of the process that already includes the contribution of all diagrams describing higher-order channels for de-excitation of the atomic shell.

In Ref. 16, in order to determine the efficiency of NEET, it was proposed that the atom be treated as a system of two coupled oscillators (the nucleus and the atomic shell) with close resonance energies. In this case, in the opinion of the authors, the probability of excitation of the nucleus does not depend on the magnitude of the nuclear matrix element, but rather is determined only by the properties of the corresponding atomic transition. As a consequence, the numerical estimates of the probability of NEET in the model of Ref. 16 are significantly higher than all previously obtained values. An excitation mechanism similar to that studied in Ref. 16 will be analyzed in detail in Sec. 4.

The problem of NEET was subjected to serious critical analysis in Refs. 17 and 18. Calculations of the NEET probability for the nuclei  $^{189}\text{Os}$ ,  $^{197}\text{Au}$ , and  $^{237}\text{Np}$ , performed with the help of time-dependent perturbation theory for nonrelativistic quantum mechanics, were performed in Ref. 17. In Ref. 18, in addition to the three nuclei indicated above, the nuclei  $^{193}\text{Ir}$  and  $^{161}\text{Dy}$  were also studied and the possibility of NEET for the nuclei  $^{235}\text{U}$  was likewise analyzed. The calculations were performed on the basis of quantum electrodynamics (QED). The discrepancies between the numerical results of Refs. 17 and 18 arise primarily for two reasons: first, in Ref. 18 the widths of both atomic levels participating in the process were taken into account (which, as will be shown below, is important in order to obtain the correct formula for the probability of the process), while in Ref. 17 only the width of the lower level was taken into account; second, in the numerical calculations reported in Ref. 17 employed nuclear matrix elements, which, in our opinion, were not entirely correctly reconstructed from existing experimental data.

Due to the significant discrepancies between different theoretical estimates, contradictory experimental results (for example, for the nucleus  $^{189}\text{Os}$ , and the possible interest of experimenters in the process of excitation of nuclei in atomic transitions, in the present paper the formulas for the probability of NEET are derived consistently on the basis of QED and the experimental results presented in Refs. 4–11 are discussed. The system of units  $\hbar = c = 1$  is employed.

## 2. PROBABILITY OF EXCITATION OF THE NUCLEUS IN AN ATOMIC TRANSITION

Following Ref. 18, we consider the NEET mechanism starting with the stage when excitation of the nucleus actually occurs in an electron transition between atomic shells. We neglect the previous history—the process in which a vacancy is formed in one of the lower shells. This approximation can be justified by estimating the rates of both processes. Consider, for example, the  $K$ -shell of atoms with  $Z = 70$ –80. The total width of a  $K$ -shell vacancy is equal to 30–50 eV, which corresponds to a lifetime of  $\sim 10^{-17}$  s. On the other hand, NEET is practically localized in the “upper” atomic (in our case  $L$  or  $M$ ) shell participating in the process. The residence time of 100-keV beam electrons (the energy must be high enough to achieve efficient ionization of the  $K$ -shell) in the  $K$ -shell and the time in which an electron knocked out of the  $K$ -shell leaves the region of interaction are equal to about  $10^{-19}$  s. For this reason, without significant loss of accuracy,

it can be assumed that there is enough time for the  $K$ -shell vacancy to forget by the time it decays the process by which it was formed. Similar arguments also apply in the case when NEET occurs in electron transitions between the upper shells of an atom. Although significantly lower electron energies are required to ionize these shells, the ratio obtained above for the times of the processes involved also holds here, since vacancies in these shells have significantly longer lifetimes. On the basis of what was said above, the diagrams in Fig. 1 which are of second and third order in the electromagnetic interaction constant  $e$  are most important for describing NEET.

We now proceed to the calculation of the probability. The nuclear wave functions of states with energies  $E_I$  and  $E_F$

$$\begin{aligned}\Psi_I(t, \mathbf{R}) &= \exp(-iE_I t) \Psi_I(\mathbf{R}), \\ \Psi_F(t, \mathbf{R}) &= \exp[-i(E_F - i\Gamma_N/2)t] \Psi_F(\mathbf{R})\end{aligned}\quad (3)$$

satisfy the normalization condition

$$\int d^3R |\Psi_{I(F)}(\mathbf{R})|^2 = 1.$$

In the wave functions (3)  $\Gamma_N$  is the width of the isomeric nuclear level  $|F\rangle$ . We assume that the initial state is stationary. The wave functions of the electron hole, which has energy  $\mathcal{E}$  in subshells with binding energies  $\mathcal{E}_b$  and widths  $\Gamma$  (we assume  $\mathcal{E} \equiv -\mathcal{E}_b$ , i.e., the energies of the electron holes are positive), have the form

$$\psi_{i(n, f)}(t, \mathbf{r}) = \exp[-i(\mathcal{E}_{i(n, f)} - i\Gamma_{i(n, f)}/2)t] \psi_{i(n, f)}(\mathbf{r}), \quad (4)$$

where  $|i\rangle$  is the initial state (vacancies are located in the “lower” shell),  $|n\rangle$  is the final state of the hole for a second-order diagram and an intermediate state for third-order diagrams, and  $|f\rangle$  is the final state of the electron hole for the diagrams in Figs. 1b and c. Thus we shall consider the excitation of the nucleus not in an electron transition but rather, more conveniently, in a transition of a hole from lower atomic levels to higher atomic levels.

In the calculations the wave function of an electron hole was represented as

$$\psi(\mathbf{r}) = \begin{pmatrix} g_{ij}(r) & \Omega_{im}(\mathbf{n}) \\ -if_{i'j'}(r) & \Omega_{i'm}(\mathbf{n}) \end{pmatrix} \quad (5)$$

with the normalization condition

$$\int_0^\infty dr r^2 [g^2(r) + f^2(r)] = 1.$$

Here  $g(r)$  and  $f(r)$  are, respectively, the large and small components of the radial wave functions,  $\mathbf{n} = \mathbf{r}/r$ , the

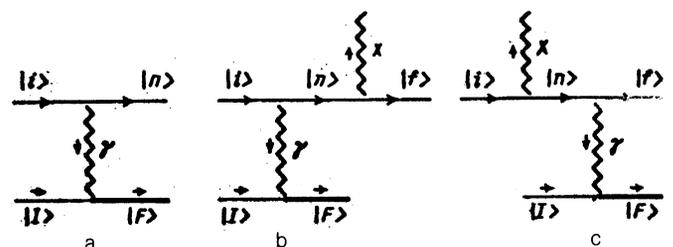


FIG. 1. Diagrams of the NEET process: a) second-order perturbation theory; b) and c) direct and exchange third-order diagrams, respectively.

spherical spinors are defined as

$$\Omega_{jm}(\mathbf{n}) = \sum_{m_1, m_2} \left( l m_1 \frac{1}{2} m_2 | j m \right) Y_{lm_1}(\mathbf{n}) \chi_{m_2},$$

$\chi_m = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$  if  $\begin{pmatrix} 0 \\ 1 \end{pmatrix}$ ,  $Y_{lm}(\mathbf{n})$  are spherical functions,  $(l m_1 \frac{1}{2} m_2 | j m)$  are Clebsch-Gordan coefficients, and  $l$  and  $j$  are the orbital and total angular momenta of the states of the electron hole.

The elements of the second-order  $S$ -matrix corresponding to the diagram in Fig. 1a have the standard form

$$S_{fi}^{(2)} = \frac{-i}{\omega_N - \omega_A - i(\Gamma_i + \Gamma_f + \Gamma_N)/2} \int d^3r d^3R e \bar{\psi}_f(\mathbf{r}) \gamma^\mu \psi_i(\mathbf{r}) \times D_{\mu\nu}(\omega_N; \mathbf{r} - \mathbf{R}) e \Psi_{F^+}(\mathbf{R}) J^\nu \Psi_I(\mathbf{R}), \quad (6)$$

where  $\omega_N = E_F - E_I$ ,  $\omega_A = \mathcal{E}_i - \mathcal{E}_f$ ,  $\gamma^\mu$  are Dirac matrices,  $\bar{\psi} = \psi^\dagger \gamma^0$ , and  $J^\nu$  is the electromagnetic-current 4-vector of the nucleus. The coordinate-frequency representation<sup>19</sup> is used for the photon propagator in Eq. 6:

$$D_{\mu\nu}(\omega_N; \mathbf{r} - \mathbf{R}) = -g_{\mu\nu} \frac{\exp(i\omega_N |\mathbf{r} - \mathbf{R}|)}{|\mathbf{r} - \mathbf{R}|}, \quad (7)$$

where  $g_{\mu\nu}$  is the metric tensor and the electron in the shell is the source of the field.

The formula (6) is obtained after integrating over the times at both vertices and over the energy of the intermediate photon. The following well-known relation is employed for the time integration:<sup>20</sup>

$$\exp[i(\mathcal{E} - i\Gamma)t] = \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\Omega \frac{e^{i\Omega t}}{(\mathcal{E} - i\Gamma) + \Omega}, \quad (8)$$

which gives

$$\int dt_1 dt_2 \exp[-i(\omega_A - \omega - i(\Gamma_i + \Gamma_f)/2)t_1 + i(\omega_N - \omega - i\Gamma_N/2)t_2] = \frac{-i}{\omega_A - \omega - i(\Gamma_i + \Gamma_f)/2} \frac{-i}{\omega_N - \omega - i\Gamma_N/2}. \quad (9)$$

Next, the integral over the energy  $\omega$  of the intermediate photon is calculated as

$$\int d\omega \rightarrow 2\pi i \operatorname{Res}(\omega = \omega_N - i\Gamma_N/2).$$

After a similar integration over times at the vertices and energies of the intermediate photon and electron hole, we obtain the following expression for the element of the third-order  $S$ -matrix corresponding to the sum of the diagrams in Figs. 1b and 1c:

$$S_{fi}^{(3)} = 2\pi e^3 \delta(\mathcal{E}_i - \mathcal{E}_f - \omega_x - \omega_N) \times \sum_n \left\{ \left[ \int d^3r \bar{\psi}_f(\mathbf{r}) \hat{\mathcal{A}}^*(\mathbf{r}, \omega_x) \psi_n(\mathbf{r}) \right] \times \left[ \int d^3r d^3R \bar{\psi}_n(\mathbf{r}) \gamma^\mu \psi_i(\mathbf{r}) D_{\mu\nu}(\omega_N, \mathbf{r} - \mathbf{R}) \Psi_{F^+}(\mathbf{R}) J^\nu \Psi_I(\mathbf{R}) \right] \times [\omega_N - (\mathcal{E}_i - \mathcal{E}_n) + i(\Gamma_i + \Gamma_n + \Gamma_N)/2]^{-1} + \left[ \int d^3r d^3R \bar{\psi}_f(\mathbf{r}) \gamma^\mu \psi_n(\mathbf{r}) D_{\mu\nu}(\omega_N, \mathbf{r} - \mathbf{R}) \Psi_{F^+}(\mathbf{R}) J^\nu \Psi_I(\mathbf{R}) \right] \times \left[ \int d^3r \bar{\psi}_n(\mathbf{r}) \hat{\mathcal{A}}^*(\mathbf{r}, \omega_x) \psi_i(\mathbf{r}) \right] \times [\omega_x - (\mathcal{E}_i - \mathcal{E}_n) - i(\Gamma_i + \Gamma_n)/2]^{-1} \right\}. \quad (10)$$

Here  $\omega_x$  is the energy of the emitted x-ray,  $\hat{\mathcal{A}} \equiv \gamma^\mu \mathcal{A}_\mu$ , and

$\mathcal{A}_\mu(\mathbf{r}, \omega_x)$  is the wave function of the photon. The derivation of Eq. (10) employed the following representation of the electron hole propagator:

$$G_{\alpha\beta}(t_f - t_i; \mathbf{r}_f, \mathbf{r}_i) = -i \sum_n \int \frac{dE}{2\pi} \frac{\exp[iE(t_f - t_i)]}{E + \mathcal{E}_n - i\Gamma_n/2} \psi_{n,\alpha}(\mathbf{r}_f) \bar{\psi}_{n,\beta}(\mathbf{r}_i) \exp(-\Gamma_n t_i), \quad (11)$$

which is constructed with the help of the integral relation (8) from the functions (4) according to the standard rules of QED. The summation in Eq. (11) extends, generally speaking, over all possible intermediate states. But, nuclei for experimental investigations of NEET are selected so that the energy  $\omega_N$  of the nuclear transition is close to the energy difference  $\mathcal{E}_i - \mathcal{E}_n$  between the initial state and one of the intermediate states of the electron hole. This makes it possible to make immediately two simplifications in the theoretical calculations. First, the single-level approximation can be used in the matrix elements (10) and (11) and, second, only the direct diagram of Fig. 1b need be considered in order to estimate the contribution of the third-order diagrams to the probability of the NEET process, since in this situation the first term in braces in Eq. (10) will be significantly greater than the second term, where there is no resonance.

For the subsequent calculations we employ the standard technique. After the phonon propagator (7) is expanded in multipoles, the longitudinal and scalar components in the amplitude  $S_{fi}^{(2)}$ , as is well known, cancel (see, for example, Ref. 21). As a result, the matrix element (6) can be written as a sum over electric ( $E$ ) and magnetic ( $M$ ) multipoles

$$S_{fi}^{(2)} = - \frac{4\pi i \omega_N}{\omega_N - \omega_A - i(\Gamma_i + \Gamma_n + \Gamma_N)/2} \times \sum_{L,m} \sum_a \int d^3r d^3R e \psi_{n^+}(\mathbf{r}) \alpha \psi_i(\mathbf{r}) \mathbf{B}_{Lm}^a(\mathbf{r}) e \Psi_{F^+}(\mathbf{R}) \hat{\mathbf{J}} \Psi_I(\mathbf{R}) \mathbf{A}_{Lm}^a(\mathbf{R}), \quad (12)$$

where  $a = E, M$ ,  $\alpha = \gamma^0 \gamma$ , and in the Coulomb gauge (see Ref. 21) the electromagnetic potentials are

$$\mathbf{A}_{Lm}^E(\mathbf{R}) = \left( \frac{L+1}{2L+1} \right)^{1/2} j_{L-1}(\omega_N R) \mathbf{Y}_{LL-1,m}(\mathbf{n}) - \left( \frac{L}{2L+1} \right)^{1/2} j_{L+1}(\omega_N R) \mathbf{Y}_{LL+1,m}(\mathbf{n}), \quad (13)$$

$$\mathbf{A}_{Lm}^M(\mathbf{R}) = j_L(\omega_N R) \mathbf{Y}_{LL,m}(\mathbf{n}),$$

$j_L(x)$  is a Bessel function,  $\mathbf{Y}_{LJ,M}(\mathbf{n})$  are vector spherical harmonics, defined as

$$\mathbf{Y}_{LJ,M}(\mathbf{n}) = \sum_{m,\mu} (L m 1 \mu | J M) Y_{Lm}(\mathbf{n}) \xi_\mu,$$

where  $\xi_\mu$  ( $\mu = \pm 1, 0$ ) are the components of the spin vector of the photon in a cyclic basis related to the components  $e_i$  ( $i = 1, 2, 3$ ) in the Cartesian basis by the relations

$$\xi_{\pm 1} = \mp (e_1 \pm i e_2) / 2^{1/2}, \quad \xi_0 = e_3.$$

The potentials  $\mathbf{B}_{Lm}^{E(M)}(\mathbf{r})$  are obtained from  $\mathbf{A}_{Lm}^{E(M)}(\mathbf{R})$  by replacing the Bessel functions in the formulas (13) by Hankel functions of the first kind  $h_L^{(1)}(\omega_N r)$ .

In order to calculate the third-order amplitude (10) we employ the following representation for the wave function for the photon with momentum  $\mathbf{k}$  (Ref. 21):

$$\begin{aligned} \mathcal{A}_\mu(\mathbf{r}, t) = & \exp(-i\omega_x t) \left( \frac{2\pi}{\omega_x} \right)^{1/2} \\ & \times (2\pi)^{1/2} \sum_{L,m} (2L+1)^{1/2} i^L D_{\mu m}^L(\varphi_{\mathbf{k}}, \vartheta_{\mathbf{k}}, 0) \\ & \times [i\mathbf{A}_{Lm}^E(\mathbf{r}) + \mu\mathbf{A}_{Lm}^M(\mathbf{r})], \end{aligned} \quad (14)$$

where  $D_{\mu m}^L(\varphi_{\mathbf{k}}, \vartheta_{\mathbf{k}}, 0)$  are Wigner  $D$ -functions.

In order to calculate the probability of the NEET process it is necessary, in particular, to integrate over the angular variables of the momentum  $\mathbf{k}$  of the emitted photon. For real photons the multipolarity  $L$  is always  $\geq 1$ .  $D$ -functions of the form  $D_{\varphi m}^L(\omega, \vartheta, 0)$  are orthogonal on the sphere. As a result, the contribution of the term corresponding to interference of the second- and third-order diagrams in the expression  $|S_f^{(2)} + S_f^{(3)}|^2$  to the probability of the process will be zero. For this reason, in what follows, the probability  $W_{\text{NEET}}^{(2)}$  of the second-order process (diagram Fig. 1a) can be calculated separately from the contribution  $W^{(3)}$  of the third-order diagram (Fig. 1b).

Consider the expression (12). We use the standard definitions of the matrix element (ME) of the nuclear current<sup>21</sup>

$$\begin{aligned} \int d^3R e\Psi_{F^+}(\mathbf{R}) \hat{\mathbf{J}} \Psi_I(\mathbf{R}) \mathbf{A}_{LM}^{E(M)}(\mathbf{R}) = & \frac{i\omega^L}{(2L+1)!!} \left( \frac{L+1}{L} \right)^{1/2} \\ & \times \langle J_F M_F | \mathcal{M}_{LM}^{E(M)}(\mathbf{R}) | J_I M_I \rangle \cdot \begin{cases} -1 & \text{for } E \\ 1 & \text{for } M \end{cases}. \end{aligned} \quad (15)$$

Here  $\mathcal{M}_{LM}^{E(M)}(\mathbf{R})$  is the electromagnetic transition operator and  $J_{I(F)}$  and  $M_{I(F)}$  are the spin of the nucleus and its projection on the quantization axis in the initial (final) state. The reduced probability of the nuclear transition is expressed in terms of the matrix element (15) as follows:

$$B(E(M)L; I \rightarrow F) = \frac{1}{2J_I+1} \sum_{M_I, M_F} |\langle J_F M_F | \mathcal{M}_{LM}^{E(M)} | J_I M_I \rangle|^2. \quad (16)$$

As far as the electronic part is concerned, by taking into account the selection rules in the matrix element of the nuclear current the electronic part can be reduced to the following form with the help of the appropriate formulas from Refs. 22 and 23:

$$\begin{aligned} \int d^3r \mathbf{B}_{LM}^{E(M)}(\mathbf{r}) \psi_n^+(\mathbf{r}) \alpha \psi_i(\mathbf{r}) = & i \left[ \frac{L(2L+1)(2j_i+1)}{4\pi(L+1)(2j_n+1)} \right]^{1/2} \mathfrak{M}_L^{E(M)}(\omega_N) \\ & \times \left( j_i - \frac{1}{2} L 0 | j_n - \frac{1}{2} \right) (j_i m_i L M | j_n m_n) \cdot \begin{cases} 1 & \text{for } E \\ -1 & \text{for } M \end{cases}. \end{aligned} \quad (17)$$

The radial matrix elements in Eq. (17) were calculated from the formulas

$$\begin{aligned} \mathfrak{M}_L^E(\omega) = & \int dr r^2 \left\{ h_L^{(1)}(\omega r) [g_i(r)g_n(r) + f_i(r)f_n(r)] \right. \\ & \left. - \frac{h_{L-1}^{(1)}(\omega r)}{L} [(\kappa_i - \kappa_n - L)g_i(r)f_n(r) + (\kappa_i - \kappa_n + L)f_i(r)g_n(r)] \right\}, \\ \mathfrak{M}_L^M(\omega) = & \frac{\kappa_i + \kappa_n}{L} \int dr r^2 h_L^{(1)}(\omega r) [g_i(r)f_n(r) + f_i(r)g_n(r)], \end{aligned} \quad (18)$$

where  $\kappa = (l-j)(2j+1)$ . The matrix elements (18) were calculated numerically. The mean field and the electronic wave functions were determined in the relativistic form of the Hartree-Fock-Slater method using A. A. Soldatov's program (a description and the necessary references can be found in Ref. 24).

In order to obtain the probability  $W_{\text{NEET}}^{(2)}$  for excitation of the nucleus in an atomic transition, we square the absolute value of the expression (12), divide by the time, and sum over the final and average over the initial states of the nucleus and the electron. Using the relations (15)–(17) it is easy to show that

$$W_{\text{NEET}}^{(2)} = (\Gamma_i + \Gamma_n + \Gamma_N) \frac{E_{\text{int}}^2(L; \omega_N; i \rightarrow n, I \rightarrow F)}{(\omega_N - \omega_A)^2 + (\Gamma_i + \Gamma_n + \Gamma_N)^2/4}, \quad (19)$$

where the squared interaction energy  $E_{\text{int}}$  denotes the quantity

$$\begin{aligned} E_{\text{int}}^2(L; \omega_N; i \rightarrow n, I \rightarrow F) = & 4\pi e^2 \frac{\omega_N^{2(L+1)}}{[(2L+1)!!]^2} (2L+1) \left( j_i \frac{1}{2} L 0 | j_n \frac{1}{2} \right)^2 \\ & \times |\mathfrak{M}_L^{E(M)}(\omega_N)|^2 B(E(M)L; I \rightarrow F). \end{aligned} \quad (20)$$

If, now, the width of the nuclear state is neglected compared with the widths of the atomic levels, the final expression for  $W_{\text{NEET}}^{(2)}$  assumes the form

$$W_{\text{NEET}}^{(2)} = \Gamma_i P, \quad (21)$$

where

$$P = \left( 1 + \frac{\Gamma_n}{\Gamma_i} \right) \frac{E_{\text{int}}^2(L; \omega_N; i \rightarrow n, I \rightarrow F)}{(\omega_N - \omega_A)^2 + (\Gamma_i + \Gamma_n)^2/4} \quad (22)$$

is the probability, introduced by M. Morita in Ref. 1, of a transition of the nucleus to an excited state accompanying the decay of the electron "hole" in the atomic level  $|i\rangle$ . In what follows, we denote by  $\Delta$  the difference  $\omega_N - \omega_A$ , appearing in the denominator, between the energies of the atomic and nuclear transitions.

The key part of the formula (22) is the expression (20) for the interaction energy of the nuclear and electronic currents. In order to verify its correctness, we employ the relations for the widths of radiative transitions with energy  $\omega_N$  between nuclear levels<sup>21</sup>

$$\begin{aligned} \Gamma_{N^*}(E(M)L; \omega_N; I \rightarrow \bar{F}) = & 8\pi \frac{\omega_N^{2L+1}}{[(2L+1)!!]^2} \frac{L+1}{L} B(E(M)L; I \rightarrow F) \end{aligned} \quad (23)$$

and between the lower atomic shells<sup>25</sup>

$$\begin{aligned} \Gamma_{A^*}(E(M)L; \omega_N; i \rightarrow n) = & 2e^2 \omega_N \frac{L(2L+1)}{L+1} \left( j_i \frac{1}{2} L 0 | j_n \frac{1}{2} \right)^2 (\text{Re} [\mathfrak{M}_L^{E(M)}(\omega_N)])^2. \end{aligned} \quad (24)$$

It is easy to see that Eq. (20) can be rewritten in terms of Eqs. (23) and (24) as

$$\begin{aligned} E_{\text{int}}^2 = & \frac{1}{4} \Gamma_{A^*}(E(M)L; \omega_N; i \rightarrow n) \\ & \times \Gamma_{N^*}(E(M)L; \omega_N; I \rightarrow F) (1+1/\delta^2), \end{aligned} \quad (25)$$

where  $\delta = \text{Re} [\mathfrak{M}_L^{E(M)}(\omega_N)] / \text{Im} [\mathfrak{M}_L^{E(M)}(\omega_N)]$  is the ana-

log of a function well-known in the theory of internal electron conversion, and Re and Im denote, respectively, the real and imaginary parts. Now, cutting the photon line in the diagram of Fig. 1a (i.e., switching to two independent radiation processes) we obtain, instead of Eq. (25), as we should, a product of two radiation widths. (Indeed,  $1/\delta^2$  evidently vanishes and the factor of  $1/4$  in Eq. (25) is a consequence of the summation over the final states of the emitted photon performed in Eqs. (23)–(24). This once again confirms the correctness of the relations (20) and (25) obtained above.

We now return to the expressions for the probabilities of nonradiative excitation of the nucleus  $W_{\text{NEET}}^{(2)}$  (19) and  $P$  (22). Taking into account the finite widths of the atomic levels participating in the NEET process results in the appearance of additional terms of order  $\Gamma_n/\Gamma_i$  in these formulas. We are not interested in further decay channels for a vacancy in the upper subshell  $|n\rangle$ . They could also include emission of an x-ray, as in Ref. 15, and the Auger process. It is the total probability of these processes that gives the width  $\Gamma_n$ . For this reason, introducing into the wave functions (4) the finite widths of the atomic states, we took into account, by means of the diagram in Fig. 1a, in particular, the contributions of the diagrams in Figs. 1b and c. We shall demonstrate this in greater detail.

We now calculate the probability of the third-order process. Squaring the absolute value of the expression (10) (without the second term in braces), summing over the final and averaging over the initial quantum numbers, and integrating over the momentum of the emitted x-ray, as the intermediate state, we obtain the following relation for the probability:

$$W_{fi}^{(3)} = 8\pi e^4 \omega_X \omega_N^{2(L'+1)} \frac{L'(2L'+1)}{L'+1} \frac{2L+1}{[(2L+1)!!]^2} B(L; I \rightarrow F) \times \sum_{j_n} \left( j_i - \frac{1}{2} L0 | j_n - \frac{1}{2} \right)^2 \left( j_n - \frac{1}{2} L0 | j_i - \frac{1}{2} \right)^2 \times \frac{|\mathfrak{M}_L(\omega_N; i \rightarrow n)|^2 |\text{Re}[\mathfrak{M}_{L'}(\omega_X; n \rightarrow f)]|^2}{(\mathcal{E}_n - \mathcal{E}_i + \omega_N)^2 + (\Gamma_n + \Gamma_i + \Gamma_N)^2/4}, \quad (26)$$

where  $L'$  is the multipolarity of the emitted photon.

The final expression is obtained by using the formulas (20) and (24) for the interaction energy and the width of the atomic transition. Using these formulas and limiting the calculation to the single-level approximation, we can put the expression (26) into the form

$$W^{(3)} = \sum_{i, L'} \Gamma_{A'}(\omega_X; L'; n \rightarrow f) \times \frac{E_{ini}^2(E(M)L; \omega_N; i \rightarrow n, I \rightarrow F)}{(\mathcal{E}_n - \mathcal{E}_i + \omega_N)^2 + (\Gamma_n + \Gamma_i + \Gamma_N)^2/4}, \quad (27)$$

where all possible final states of the hole and multiplicities of the emitted x-ray are summed over in order to calculate the total probability. It is obvious that the summation of the widths of the atomic transitions over  $|f\rangle$  and  $L'$  in the formula (27) will give a quantity that is approximately equal to the total width of the vacancy in the intermediate state  $\Gamma_n$  (minus the probabilities of Auger processes), i.e.,

$$W^{(3)} \approx \Gamma_n \frac{E_{ini}^2(E(M)L; \omega_N; i \rightarrow n, I \rightarrow F)}{(\mathcal{E}_n - \mathcal{E}_i + \omega_N)^2 + (\Gamma_n + \Gamma_i)^2/4}. \quad (28)$$

This is the additional term that appears in the formula (19) when the width of the level  $|n\rangle$  is taken into account.

Now the ratio of the probabilities of the processes shown in Figs. 1a and 1b, which we would have obtained if we have neglected the finite width of the state  $|n\rangle$  in the wave function (4), becomes obvious:

$$W^{(3)}/W^{(2)} \approx \Gamma_n/\Gamma_i, \quad (29)$$

and since in most cases the state  $|i\rangle$  corresponds to the  $K$ -shell and  $|n\rangle$  corresponds to the  $L$  and higher shells, even in the indicated case  $W^{(3)}$  is only a correction (sometimes, it is true, a quite significant correction) to  $W^{(2)}$ .

### 3. NEET IN THE NUCLEI $^{189}\text{Os}$ , $^{197}\text{Au}$ , $^{237}\text{Np}$ , $^{193}\text{Ir}$ , $^{161}\text{Dy}$ , AND $^{235}\text{U}$

Before proceeding to specific examples, we stipulate at the outset that the data on the total widths of the atomic levels were taken from Ref. 26 for the  $M$ -shell and from Ref. 27 for the  $K$ - and  $L$ -shells. The values employed below for the binding energies  $\mathcal{E}_b$  of the atomic levels and the energies  $\omega_A$  of the atomic transitions were taken from Refs. 28–30; in addition, in the case of the experimentally observed transitions preference was given to the measured values of  $\omega_A$ .

A portion of the diagrams of the atomic and nuclear levels of  $^{189}\text{Os}$  is presented in Fig. 2. The data on the energies, spins, and lifetimes of the nuclear levels were taken from Ref. 31. The rotational quantum number  $K$  and the quantum numbers in Nilsson's model  $[Nn_z\Lambda]$  were taken from Ref. 32.

According to Ref. 31, the reduced probabilities of

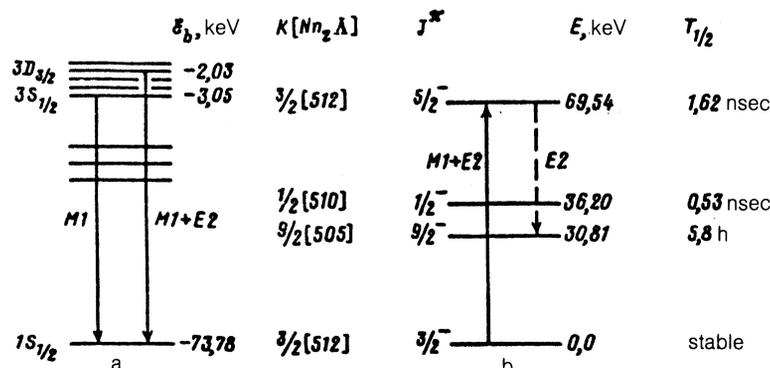


FIG. 2. Portions of the energy level diagrams of (a) the osmium atom and (b) the  $^{189}\text{Os}$  nucleus.

the transitions of interest to us are as follows:

$$B(M1; 3/2^- \rightarrow 5/2^-) = 5.45 \times 10^{-7} \text{ fm}^2,$$

$$B(E2; 3/2^- \rightarrow 5/2^-) = 69.84 \text{ fm}^4.$$

The widths of the atomic states in the  $M_1$ - and  $K$ -shells are equal to  $\Gamma(M_1) = 20.4 \text{ eV}$  and  $\Gamma(K) = 42.6 \text{ eV}$ . A numerical calculation of the atomic matrix element of the  $M$  1-transition  $3S_{1/2} \rightarrow 1S_{1/2}$  gave the value  $|\mathfrak{M}_1^M(\omega_N)|^2 = 113.4$ . Using the data presented, the interaction energy and the relative probability of excitation of the nucleus on one  $K$ -shell vacancy are, according to the formulas (20) and (22),

$$E_{int}^2(M1; M_1 \rightarrow K; 3/2^- \rightarrow 5/2^-) = 3.8 \times 10^{-4} \text{ eV}^2,$$

$$P_{M1} = 3.4 \times 10^{-10}, \quad P_{M1}(\Delta = 0) = 5.7 \times 10^{-7}.$$

The second of these values for  $P_{M1}$  is the maximum efficiency at resonance of  $\omega_A$  with  $\omega_N$ .

An analogous calculation for the transition  $3D_{3/2} \rightarrow 1S_{1/2}$  gives for  $M$  1- and  $E$  2-transitions results which are approximately four and two orders of magnitude smaller, respectively. The contribution of other atomic shells is even smaller. Thus the value  $P$  for the levels studied with  $\Delta \leq 1.3 \text{ keV}$  should fall into the range  $3.4 \times 10^{-10} \leq P \leq 5.7 \times 10^{-7}$ . In this sense, the evolution of the experimental results given in Refs. 4–7 as the measurement methods improved is interesting:  $P \approx 10^{-6}$  (Ref. 4),  $P = (1.7 \pm 0.2) \times 10^{-7}$  (Ref. 5),  $P = (4.3 \pm 0.2) \times 10^{-8}$  (Ref. 6),  $P = (5.7 \pm 1.7) \times 10^{-9}$  (Ref. 7). The last three values are consistent with the range obtained here. There is, however, a problem. In many cases we actually do not know the exact energy of the atomic transition. When the samples are irradiated with  $\gamma$ -rays in experiments with bremsstrahlung<sup>6</sup> and synchrotron radiation<sup>7</sup> (wide spectrum), ionization of the outer shells of the atoms can occur at the same time. Then the binding energies of the inner shells and, as a consequence, the energies of the transitions between them can change, i.e., conditions which are closer to resonance and the opposite conditions can be realized.

We now calculate the cross section  $\sigma_N$  for the excitation of the nucleus  $^{189}\text{Os}$  into the state  $5/2^-$  (69.537 keV) by the NEET mechanism by 100 keV electrons and the cross section  $\sigma_I$  for the formation of the isomer  $9/2^-$  (30.814 keV). The cross section for the ionization of the  $K$ -shell (we denote it as  $\sigma_A$ ) by 100-keV electrons is equal to about 6.2 barns (this estimate is based on the well-known empirical formula given in Ref. 33). From the relation

$$\sigma_N = \sigma_A P \quad (30)$$

we have  $\sigma_N \approx 2.1 \times 10^{-33} \text{ cm}^2$ . In order to find  $\sigma_I$  it is necessary to know the probability for the state  $9/2^-$  (30.814 keV) to be occupied when the level  $5/2^-$  (69.537 keV) decays; there are no experimental data on the  $E$  2-transition between the indicated levels (in Fig. 2 this transition is denoted by the dashed line). This transition is  $K$ -forbidden in first order. We introduce the intensity reduction factor  $F_{E2} = B(E2; 5/2^- \rightarrow 9/2^-) / B(E2; W)$ , where the quantity in the denominator is the reduced probability in the Weisskopf model. Then the total width is  $\Gamma(E2; 5/2^- \rightarrow 9/2^-) = (1 + \alpha) \Gamma'(E2; 5/2^- \rightarrow 9/2^-) = 1.71 \times 10^{-9} F_{E2} \text{ eV}$ , where the internal electron conversion coefficient  $\alpha = 357.6$  was calculated here using the program of Ref. 22. Correspondingly, the fraction of nuclei which occupy a long-

lived state is equal to

$$\xi = \Gamma(E2; 5/2^- \rightarrow 9/2^-) / \Gamma(5/2^-) \approx 5.7 \cdot 10^{-3} F_{E2}.$$

The decrease in the intensities of  $K$ -forbidden transitions is usually less than  $10^2$  for each unit of  $K$ -forbiddance. For this reason, the quantity  $F_{E2}$  probably falls in the range  $10^{-2} \leq F_{E2} \leq 10^{-1}$ . As a result, we obtain the following estimate for the cross section for the excitation of the long-lived isomer  $^{189m}\text{Os}$  in the NEET process when the sample is irradiated with 100 keV electrons:

$$1.2 \times 10^{-37} \text{ cm}^2 \leq \sigma_I \leq 1.2 \times 10^{-36} \text{ cm}^2. \quad (31)$$

When the sample is irradiated with 100 keV photons we obtain the following range for  $\sigma_I$  with the cross section for photoionization from the  $K$ -shell  $\sigma_A \approx 10^{-21} \text{ m}^2$ ,<sup>34</sup>

$$2 \times 10^{-35} \text{ cm}^2 \leq \sigma_I \leq 2 \times 10^{-34} \text{ cm}^2. \quad (32)$$

We underscore the fact that Eqs. (31)–(32) were obtained for monochromatic beams of electrons and photons under the assumption that ionization of the outer atomic levels does not occur at the same time.

The results presented above show that the NEET mechanism could not give the experimentally observed yield of isomeric osmium nuclei. Of all the possible excitation processes, in the present case inelastic scattering of beam electrons by  $^{189}\text{Os}$  has the highest cross section. In Ref. 35 the cross section for the excitation of the  $5/2^-$  level (69.5 keV) was calculated with relativistic electron wave functions in the initial and final states, and the effect of the atomic shell was taken into account; the atomic shell, in turn, was calculated using the code already mentioned above (these calculations are described in detail in Ref. 36).<sup>13</sup> The cross sections obtained for 100 keV beam electrons were as follows:  $\sigma_{ee}^{E2} = 3.0 \times 10^{-31} \text{ cm}^2$  and  $\sigma_{ee}^{M1} = 5.6 \times 10^{-33} \text{ cm}^2$ , whose sum is approximately 3.1 times smaller than the excitation cross sections of the  $5/2^-$  level (69.5 keV) which were reconstructed from the experimental data given in Ref. 5. Our result<sup>35</sup> is in good agreement with a similar calculation performed by M. D. Bondar'kov and I. M. Kolomiits in Refs. 15 and 37. The question of how other mechanisms for filling a  $K$ -shell vacancy in the experiments in Refs. 5 and 7 [for example, inverse internal electron conversion (IIEC)] might affect the probability of excitation of osmium nuclei was specially investigated in Ref. 38. It was found that the significant difference between the results obtained in Refs. 5 and 7 (we recall that in Refs. 5 and 7 the  $K$ -shell was ionized by an electron beam and by synchrotron radiation, respectively) cannot be explained by IIEC, and as pointed out in Ref. 16 it is a consequence of systematic errors in the experiments performed in Refs. 5 and 7. For this reason, in the experiments of Refs. 4 and 5 the data on the ratio  $\sigma_N(E) / \sigma_N(E_0)$  as a function of the energy  $E$  of the incident electron ( $E_0$  is the minimum energy for which excited nuclei could be observed) are more reliable than the data on the cross sections  $\sigma_N(E)$  for the excitation of the  $5/2^-$  (69.5 keV). These data are in good agreement with the theory.<sup>37</sup>

One experiment was performed on the nucleus  $^{197}\text{Au}$ .<sup>8</sup> Thin gold foil was bombarded with 100 keV electrons. After irradiation ceased the conversion electrons from the  $L$ -shell, which arise when the  $1/2^+$  nuclear state (77.345 keV) decays, were recorded. The measured value is  $P = (2.2 \pm 1.8) \times 10^{-4}$ .

The energy of the  $3S_{1/2} \rightarrow 1S_{1/2}$  transition in the gold atom is very close to the energy of the nuclear transition  $\omega_N = 77.35$  keV:  $\Delta \approx 50$  eV. The widths are  $\Gamma(M_1) = 20.9$  eV and  $\Gamma(K) = 49.6$  eV. The reduced probability of the nuclear transition is  $B(M_1; 3/2^+ \rightarrow 1/2^+) = 3.05 \times 10^{-7}$  fm<sup>2</sup>.<sup>39</sup> The atomic matrix element is  $|\mathfrak{M}_1^M(\omega_N)|^2 = 106.8$ . The calculation gave the following results:

$$E_{int}^2(M_1; M_1 \rightarrow K; 3/2^+ \rightarrow 1/2^+) = 3.07 \times 10^{-4} \text{ eV}^2, \\ P_{M_1} = 1.3 \cdot 10^{-7}, \quad P_{M_1}(\Delta=0) = 3.5 \times 10^{-7}.$$

NEET is significantly weaker in the case of atomic transitions from other levels. Using the experimental values for the cross section for ionization of the  $K$ -shell of gold by 100 keV electrons  $\sigma_A \approx 4.4$  barns,<sup>40</sup> we now obtain  $\sigma_N \approx 6 \times 10^{-31}$  cm<sup>2</sup> for the cross section for the excitation of the  $1/2^+$  nuclear state (69.545 keV) in NEET. It is important to note that the total cross section  $\sigma_{ee}^{E_2+M_1}$  for the excitation of the  $1/2^+$  level (77.35 keV) with inelastic scattering of 100 keV electrons by <sup>197</sup>Au nuclei, as calculation shows, is equal to approximately  $4.1 \times 10^{-32}$  cm<sup>2</sup>. Thus here we have one of those rare cases when, because of the very small energy difference  $\Delta$ , the NEET process predominates over the  $(e, e')$  process.

In Ref. 9 neptunium atoms were irradiated with 122 keV and 136 keV  $\gamma$ -rays from <sup>57</sup>Co. The intensities of the x-ray lines from electron transitions into the  $L$ -shell, which arise when the latter is ionized as a result of conversion decay of the  $5/2^-$  state (59.537 keV), were measured. The measured value is  $P = (2.1 \pm 0.6) \times 10^{-4}$ .

According to Ref. 41, the reduced probability of a nuclear transition with energy  $\omega_N = 102.96$  keV is equal to  $B(E_1; 5/2^+ \rightarrow 7/2^-) = 0.769 \times 10^{-7}$  fm<sup>2</sup>. The widths of the atomic levels are  $\Gamma(L_2) \approx 4$  eV,  $\Gamma(L_3) \approx 6$  eV, and  $\Gamma(K) = 94.2$  eV, and the energies of the corresponding transitions are  $\omega_A(L_2 \rightarrow K) = 97.067$  keV and  $\omega_A(L_3 \rightarrow K) = 101.068$  keV. The computed values of the atomic matrix

elements are as follows:

$$|\mathfrak{M}_1^E(\omega_N; L_2 \rightarrow K)|^2 = 3.4, \quad |\mathfrak{M}_1^E(\omega_N; L_3 \rightarrow K)|^2 = 4.2.$$

Thus for the atomic transition  $2P_{3/2} \rightarrow 1S_{1/2}$

$$E_{int}^2(E_1; L_3 \rightarrow K; 5/2^+ \rightarrow 7/2^-) = 0.96 \times 10^{-5} \text{ eV}^2, \quad P_{E_1} = 2.9 \cdot 10^{-12},$$

and for the  $2P_{1/2} \rightarrow 1S_{1/2}$  transition

$$E_{int}^2(E_1; L_2 \rightarrow K; 5/2^+ \rightarrow 7/2^-) = 0.76 \cdot 10^{-5} \text{ eV}^2, \quad P_{E_1} = 2.3 \cdot 10^{-13}.$$

Since the cross section for photoionization from the  $K$ -shell by 122 keV  $\gamma$ -rays is equal to  $\approx 10^{-21}$  cm<sup>2</sup> (Ref. 34) and the fraction of nuclei populating the isomeric  $5/2^-$  state (59.537 keV) with the decay of the  $7/2^-$  level (102.96 keV) is practically equal to unity,<sup>41</sup> we obtain  $\sigma_I \approx 3 \times 10^{-33}$  cm<sup>2</sup> for the cross section for the excitation of an isomer in NEET.

Experiments with <sup>193</sup>Ir and <sup>161</sup>Dy nuclei have not yet been performed. But these nuclei have stable ground states (which is very convenient for performing experiments) and transitions whose energies are close to the energies of atomic transitions (see Ref. 42). We present the data on them in Table I, which briefly summarizes the current situation with NEET.

Experimental and theoretical information, no less contradictory than for osmium-189, exists for uranium-235. On the other hand, the low-lying (76.8 eV) isomeric state in <sup>235</sup>U was very efficiently excited in a laser plasma in the experiment described in Ref. 10. In Ref. 10, Izawa and Yamataka considered, on the basis of the incorrect calculations of Ref. 1, NEET as a mechanism for the excitation of the nuclei. According to Refs. 10, 11, and 43,  $P$  must be equal to approximately  $10^{-11}$  in order to explain the yield of uranium isomers observed in Ref. 10. On the other hand, a similar experiment described in Ref. 44, performed at the I. V. Kurchatov Atomic Energy Institute, was unsuccessful. The unsuccessful attempt to observe the excitation of <sup>235m</sup>U in a plasma produced by CO<sub>2</sub> laser radiation on the surface of

TABLE I. Experimental and theoretical values of the probability of NEET.

Nucleus	Probability $P$	
	Experiment	Theory
<sup>189</sup> Os	$\approx 10^{-6}$ [4] $(1.7 \pm 0.2) \cdot 10^{-7}$ [5] $(4.3 \pm 0.2) \cdot 10^{-8}$ [6] $(5.7 \pm 1.7) \cdot 10^{-9}$ [7]	$1.5 \cdot 10^{-7}$ [12] $2.5 \cdot 10^{-7}$ [14] $1.1 \cdot 10^{-7}$ [15] $2.31 \cdot 10^{-7}$ [16] $1.2 \cdot 10^{-9}$ [17] $3.4 \cdot 10^{-10}$ *
<sup>197</sup> Au	$(2.2 \pm 1.8) \cdot 10^{-4}$ [8]	$3.5 \cdot 10^{-5}$ [14] $2.2 \cdot 10^{-5}$ [16] $4.2 \cdot 10^{-7}$ [17] $1.3 \cdot 10^{-7}$ *
<sup>237</sup> Np	$(2.1 \pm 0.6) \cdot 10^{-4}$ [9]	$1.5 \cdot 10^{-7}$ [14] $2.6 \cdot 10^{-4}$ [16] $8.5 \cdot 10^{-9}$ [17] $3.1 \cdot 10^{-12}$ *
<sup>183</sup> Ir	—	$4.6 \cdot 10^{-6}$ [14] $6.1 \cdot 10^{-9}$ *
<sup>161</sup> Dy	—	$6.6 \cdot 10^{-11}$ *

\*—present work

uranium dioxide ceramic, even with 6%  $^{235}\text{U}$  content (natural uranium was employed in Ref. 10), is very sobering. In Ref. 44 the parameters of the plasma obtained in the experiment of Ref. 10 were first reproduced and then "exceeded." The second attempt was more successful. In 1989 a series of experiments on excitation of the low-lying isomeric  $1/2^+$  state (76.8 eV) of  $^{235m}\text{U}$  in a plasma, produced with an electron beam on the surface of highly enriched (99.99%) metallic uranium, was performed in the Triton facility.<sup>45</sup> The parameters of the plasma in Ref. 45 were close to those in Ref. 10, but, first of all, the measured excitation efficiency was significantly lower than in Ref. 10 and, second, the isomers were formed as a result of inelastic scattering of beam electrons (whose energy reached 500 keV) by uranium nuclei, during which high-lying levels were populated first and uranium isomers were only formed in transitions from them.<sup>46</sup> Neither the NEET mechanism nor other mechanisms, including also "plasma" mechanisms (IIEC, etc.; see Ref. 46), for nuclear excitation had any connection with this.

As far as the efficiency of excitation of uranium by the NEET mechanism in plasma experiments<sup>10,44,45</sup> is concerned, here only a qualitative assessment can be made. The following reduced probability was obtained in Ref. 47 for the isomeric  $E3$ -transition between the ground and low-lying nuclear states:  $B(E3; 7/2^- \rightarrow 1/2^+) = 0.28 \text{ fm}^6$ . NEET is more likely to occur in the atomic transitions  $6P_{3/2} \rightarrow 5D_{3/2,5/2}$ ,  $6P_{1/2} \rightarrow 5D_{5/2}$ . The corresponding interaction energies are equal to

$$E_{int}^2(E3; P_2 \rightarrow O_3; 7/2^- \rightarrow 1/2^+) = 5.1 \times 10^{-18} \text{ eV}^2,$$

$$E_{int}^2(E3; P_3 \rightarrow O_4; 7/2^- \rightarrow 1/2^+) = 1.6 \times 10^{-18} \text{ eV}^2,$$

$$E_{int}^2(E3; P_3 \rightarrow O_5; 7/2^- \rightarrow 1/2^+) = 0.9 \times 10^{-18} \text{ eV}^2.$$

Since we are talking about a plasma whose electron temperature is equal to several tens of electron volts, where multiple ionization of the levels occurred, it is difficult to make any judgments about the energy of the atomic transitions indicated above. Since, however, the squared interaction energy of the nuclear and atomic currents is equal to only  $10^{-17}$ – $10^{-18} \text{ eV}^2$ , we can see that even at resonance the probability  $P$  will be of the order of  $10^{-11}$  only if the widths of the levels participating in the transition are less than  $10^{-3} \text{ eV}$ . This, however, is unrealistic in practice, because of the collisional broadening of atomic levels in plasma.<sup>48</sup>

#### 4. EXCITATION OF NUCLEI BY THE INVERSE ELECTRONIC BRIDGE MECHANISM

The relatively recent work Ref. 16 was one of the latest in a series of theoretical investigations of NEET. This paper contains a number of assertions which require careful analysis, and some numerical results which differ radically from other theoretical estimates. The model employed in Ref. 16 agrees with the experimental data only for  $^{237}\text{Np}$  (see Table I).

The nuclear excitation mechanism proposed in Ref. 16 reduces to the process shown in Fig. 3a. This process was first studied in Ref. 49 by I. S. Batkin and was termed Compton excitation of the nucleus. Later it was studied in Ref. 50, where it was termed inverse electronic bridge (IEB) (since it is actually the reverse of the process of deexcitation of the

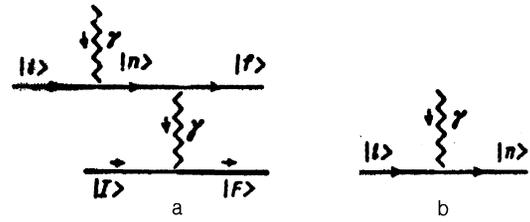


FIG. 3. a) Direct diagram of the IEB process; b) diagram describing the excitation of an atom by a photon.

nucleus through an electronic bridge). Following Ref. 16, we shall analyze the efficiency of this mechanism in the simplest case—the transition of a hole between discrete states of the atomic shell, assuming at the outset that the vacancy already exists in one of the upper subshells of the state  $|i\rangle$ . Then the process studied in Ref. 16 corresponds to the diagram shown in Fig. 3a: The external radiation is absorbed by the atomic shell and as a result the vacancy is transferred from the shell  $|i\rangle$  into the inner shell  $|n\rangle$ , after which re-emission of the  $\gamma$ -ray by the shell and excitation of the nucleus occur. It is proposed that the efficiency  $P_\gamma$  of the process be defined as the ratio of the excitation cross section of the nucleus to the photoabsorption cross section of the atom (diagram Fig. 3b).

The amplitude corresponding to the direct diagram of IEB (Fig. 3a), written with the help of the functions and relations of Sec. 2, has the form

$$S_{fi}^{(3)} = e^3 \sum_n \{ (\mathcal{E}_i + \omega_\gamma - \mathcal{E}_i - \omega_n - i[\Gamma_n + (\Gamma_i + \Gamma_f + \Gamma_n)/2])^{-1} \times \iint d^3r d^3R \bar{\Psi}_f(\mathbf{r}) \gamma^\mu \Psi_n(\mathbf{r}) D_{\mu\nu}(\omega_n; \mathbf{r} - \mathbf{R}) \Psi_{F^+}(\mathbf{R}) \hat{J}^\nu \Psi_I(\mathbf{R}) \times \int d^3r \bar{\Psi}_n(\mathbf{r}) \hat{\mathcal{A}}(\mathbf{r}, \omega_\gamma) \Psi_i(\mathbf{r}) [\mathcal{E}_n - \mathcal{E}_i - \omega_\gamma + i(\Gamma_i + \Gamma_n)/2]^{-1} \}, \quad (33)$$

where  $\omega_\gamma$  is the energy of the incident photons. The following expression is obtained for the nuclear excitation cross section from this formula:

$$\sigma^{(3)} = \frac{\lambda_\gamma^2}{4\pi} \frac{\Gamma_n \Gamma_{A^+}(\omega_\gamma; i \rightarrow n) [1 + (\Gamma_i + \Gamma_f)/2\Gamma_n]^2}{(\mathcal{E}_n - \mathcal{E}_i - \omega_\gamma)^2 + (\Gamma_i + \Gamma_n)^2/4} \times \frac{E_{int}^2(L; \omega_n; n \rightarrow f, I \rightarrow F)}{(\mathcal{E}_i + \omega_\gamma - \mathcal{E}_i - \omega_n)^2 + \Gamma_n^2 [1 + (\Gamma_i + \Gamma_f)/2\Gamma_n]^2}. \quad (34)$$

Here  $\lambda_\gamma = 2\pi/\omega_\gamma$ , and the width of the nuclear level was neglected compared with  $\Gamma_n$ .

The cross section for the excitation of an atom by a photon is calculated similarly, and in our notation it has the form

$$\sigma^{(1)} = \frac{\lambda_\gamma^2}{8\pi} \frac{\Gamma_n \Gamma_{A^+}(\omega_\gamma; i \rightarrow n)}{(\mathcal{E}_n - \mathcal{E}_i - \omega_\gamma)^2 + \Gamma_n^2/4}. \quad (35)$$

The formula for the desired efficiency or the relative probability  $P_\gamma = \sigma^{(3)}/\sigma^{(1)}$  of excitation of the nucleus when the atom is irradiated with resonance photons (resonant, since in the derivation of the expression for  $\sigma^{(3)}$  we considered only the contribution of the direct diagram in the single-level approximation, which, generally speaking, is correct only near resonance  $\omega_\gamma \approx \mathcal{E}_n - \mathcal{E}_i$ )

$$P_\gamma \approx \frac{E_{int}^2(L; \omega_n; n \rightarrow f, I \rightarrow F)}{(\mathcal{E}_n - \mathcal{E}_i - \omega_n)^2 + \Gamma_n^2}, \quad (36)$$

evidently has the same structure as the formula (22), and it includes, of course, the nuclear matrix element through the expression for the electron-nuclear interaction energy (20) and the reduced probability of the nuclear transition (16). The differences, which are not fundamental, between  $P_\gamma$  and  $P$  from Eq. (22) are related to the fact that the IEB, in contrast to the processes studied in Sec. 2, includes the previous history—the formation of a vacancy in an inner atomic shell.

## 5. CONCLUSIONS

The NEET probability computed in this paper prevents this mechanism from playing a leading role in the excitation of isomers in the experiments of Refs. 4–10 and 45. In order to separate NEET from the background formed by other processes, such as, for example, inelastic scattering of electrons by the nucleus or Compton excitation of nuclear levels,<sup>51</sup> in most cases a special coincidence experiment must be performed, in which the  $K$ -shell electron leaving the atom is correlated with the conversion electron formed as a result of the decay of the isomeric level. The only exception is <sup>197</sup>Au, where the cross section for the excitation of the isomeric nuclear level in the NEET process accompanying irradiation of gold-197 atoms with 100 keV electrons is an order of magnitude larger than the cross section for inelastic scattering of electrons by nuclei. This is what makes the nucleus <sup>197</sup>Au most promising for experimental investigations of NEET. The <sup>193</sup>Ir nucleus is also interesting, because the difference between the energies of the atomic and nuclear transitions (about 100 eV) is comparatively small for it too. Calculations show that in a similar experiment with an electron beam the cross section for NEET on <sup>193</sup>Ir will be approximately equal to the cross section for the excitation of the isomer accompanying inelastic scattering of electrons:  $\sigma_{\text{NEET}} = 3.5 \times 10^{-32} \text{ cm}^2$ ,  $\sigma_{\text{ee}^{M^1+E^2}} = 2.4 \times 10^{-32} \text{ cm}^2$ .

The computed values of the matrix elements of the atomic transitions were presented in the examples studied in the text. When necessary, this makes it possible to compare the results with calculations performed using other programs. The energy  $E_{\text{int}}$  of interaction of the nuclear and atomic currents is very sensitive to the magnitude of the atomic matrix element, i.e., to the accuracy with which the atomic wave functions of the  $K$ -,  $L$ -, and  $M$ -states are calculated, in particular, near zero, where the Neumann function has a singularity. The real part of the matrix element can be tested according to the known widths of the corresponding atomic transitions and the imaginary part can be tested according to the probability of internal electron conversion of low-energy nuclear transitions, where it makes the dominant contribution.

Apparently, the correctness of the calculation of  $E_{\text{int}}$  can now be checked in a different type of experiment—laser excitation of a low-lying isomeric state (with energy less than 5 eV) of <sup>229</sup>Th by the inverse electronic bridge mechanism.<sup>52</sup> As we saw in Sec. 4, the IEB cross section depends directly on the energy  $E_{\text{int}}$  of the interaction of the nuclear and electronic currents—the basic parameter, concerning whose value the greatest disagreements exist in the theoretical estimates of the NEET probability.

As far as the experimentally obtained absolute yield of isomeric nuclei is concerned,<sup>4–9</sup> no currently known mechanism gives the required excitation cross section. As we have

already mentioned, this is, in all probability, attributable to inadequate accuracy of the measurements. On the basis of what has been said above, it seems reasonable, on the one hand, to increase the accuracy of measurements of the nuclear excitation cross sections and, on the other, to perform coincidence experiments in order to determine the NEET cross sections themselves.

In conclusion it should be noted that in spite of the apparent simplicity of the NEET mechanism, from the viewpoint of theoretical models, reality may turn out to be much more complicated. The point is that in a different version of the process of nonradiative excitation of nuclei, namely, annihilation of positrons in an atomic shell, there is also a significant disagreement between the theoretical calculations and the experimental results (see, for example, the review Ref. 53). The reason for this state of affairs could be the same in both processes.

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