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Spectroscopy of cooperative electron-gamma-nuclear processes in heavy atoms: NEET effect

O Yu Khetselius

Odessa State University – OSENU, P.O. Box 24a, Odessa-9, SE-65009, Ukraine

E-mail: nuckhet@mail.ru

Abstract. A consistent relativistic energy approach, based on the S-matrix Gell-Mann and Low formalism (energy approach) combined with the relativistic many-body perturbation (PT) theory, to studying NEET (nuclear excitation by electron transition) effect is presented. The calculation results of the NEET probabilities for the $^{189}_{76}Os$, $^{193}_{77}Ir$, $^{197}_{79}Au$ atoms are presented and compared with available experimental and other theoretical data.

1. Introduction

This paper goes on our work on studying the cooperative electron-gamma-nuclear processes [1-4]. The important example of the cooperative electron-gamma-nuclear process is so called NEET (nuclear excitation by electron transition) effect [5-7]. Naturally, the similar NEEC (nuclear excitation by electron capture) process should be reminded too. In both NEEC and NEET, which are at the borderline between atomic and nuclear physics, electronic orbital energy is converted directly into nuclear energy. These effects offer therefore the possibility to explore the spectral properties of heavy nuclei through the typical atomic physics experiments. In fact, the NEET is a fundamental but rare mode of decay of an excited atomic state in which the energy of atomic excitation is transferred to the nucleus via a virtual photon. This process is naturally possible if within the electron shell there exists an electronic transition close in energy and coinciding in type with nuclear one. In fact the resonance condition between the energy of nuclear transition w_N and the energy of the atomic transition w_A should be fulfilled. Obviously, the NEET process corresponds to time-reversed bound-state internal conversion. Let us remind that firstly the NEET and NEEC effects have been postulated in 1973 by Morita and Goldanskii–Letokhov-Namiot [5-7] (see also review [1] and Refs.[8-10]). Unlike the NEEC effect, the NEET process has been observed experimentally in $^{197}_{79}Au$ by Kishimoto et al (Institute of Material Structure Science, KEK, and Japan Synchrotron Radiation Research Centre, Japan) and in $^{189}_{76}Os$ by Ahmad et al (Argonne National Laboratory, USA) [11-13]. Below in table 1 we present a summary of the experimental works on NEET in $^{189}_{76}Os$. It should be noted that each of the experimental techniques has certain inherent difficulties. Analysis of this problem has been presented in Refs. [1,13]. It explains quite large difference between the results of different experiments. Saying briefly, the cited difficulties are reduced to the problem of revealing a NEET signal among the surrounding other effects. Really, use of an electron beam can cause direct Coulomb excitation of a nucleus. In this case it is hardly possible to distinguish this component from that due to the NEET process. Use of a broad continuous spectral distribution of synchrotron or bremsstrahlung X-rays results in contribution from a direct nuclear photoabsorption into the nuclear state or into a range of nuclear levels that can feed that state or the lower-lying metastable state.

Table 1. Experimental data on the NEET probabilities P_{NEET} for the isotope of $^{189}_{76}Os$.

	year	Experimental techniques	P_{NEET}
Otozai et al	1973	e^- bombardment 75-85 keV	$1\cdot10^{-6}$
Otozai et al	1978	e^- bombardment 72-100 keV	$(1.7\pm0.2)\cdot10^{-7}$
Saito et al	1981	200 keV bremsstrahlung	$(4.3\pm0.2)\cdot10^{-8}$
Shinohara etal	1987	„white“ synchrotron radiation	$(5.7\pm1.7)\cdot10^{-9}$
Lakosi et al	1995	300 keV bremsstrahlung	$(2.0\pm1.4)\cdot10^{-8}$
Ahmad et al	2000	monochromatic 100-keV X rays	$<9\cdot10^{-10}$

The theoretical models for the NEET effect were developed in Refs. [5-17] (see details in [1]). Many of the early estimates involved the using simplifying approximations that led to results at considerable variance. More lately Tkalya [16,17] has proposed a model for description of the NEET process near the K-shell ionization threshold of an atom. The QED PT with empirical estimates of the nuclear and electron matrix elements and the Dirac-Fock code by Band and Fomichev (taking into account the finite nuclear size) were used. New theoretical approach by Ahmad et al [13] is based on using the time-dependent amplitude coupled equations. These authors calculated electron wave functions using the GRASP code and tabulated values of the nuclear transition matrix elements. Therefore, the theoretical models involved the use of different consistency level approximations led to results at quite considerable variance. It is obvious that more sophisticated relativistic many-body methods should be used for correct treating the NEET effect. Really, the nuclear wave functions have the many-body character. The correct treating the electron subsystem processes requires an account of the relativistic, exchange-correlation and nuclear effects. Really, the nuclear excitation occurs by electron transition from the M shell to the K shell. So, there is the electron-hole interaction and it is of a great importance a correct account for the many-body correlation effects, including inter-shell correlations, the post-act interaction of the removing electron and hole, possibly the continuum pressure etc [1,17,18]. In any case the theoretical calculations for NEET occurring in scattering measurements are particularly useful, especially in finding candidate isotopes and transitions suitable for experimental observation. In our paper a new, consistent relativistic energy approach to calculation of probabilities of the NEET is presented. In our approach the NEET process probability and cross-section are determined within the S-matrix Gell-Mann and Low formalism (energy approach) combined with the relativistic many-body PT [18-23]. The calculation results of the NEET probabilities for the $^{189}_{76}Os$, $^{193}_{77}Ir$, $^{197}_{79}Au$ atoms within different theoretical models are presented and compared with available experimental data.

2. Relativistic energy approach to process of nuclear excitation by electron transition

The relativistic energy approach is based on the S-matrix Gell-Mann and Low formalism combined with the relativistic many-body PT [1,19-23]. Let us remind that in atomic theory, a convenient field procedure is known for calculating the energy shifts ΔE of the degenerate states [20]. Secular matrix M diagonalization is used. In constructing M, the Gell-Mann and Low adiabatic formula for ΔE is used. A similar approach, using this formula with the electrodynamical scattering matrix, is applicable in the relativistic theory [19-21]. In contrast to the non-relativistic case, the secular matrix elements are already complex in the PT second order (1storder of the inter-electron interaction). Their imaginary parts relate to radiation decay (transition) probability. The total energy shift of the state is as:

$$\Delta E = \mathrm{Re}\Delta E + \mathrm{i}\ \mathrm{Im}\Delta E, \quad (1)$$

$$\mathrm{Im}\ \Delta E = -\Gamma/2, \quad (2)$$

where Γ is interpreted as the level width, and the decay possibility P=Γ. The whole calculation of energies and decay probabilities of a non-degenerate excited state is reduced to calculation and diagonalization of the complex matrix *M*. To start with the Gell-Mann and Low formula it is necessary to choose the PT zero-order approximation. Usually, the one-electron Hamiltonian is used, with a central potential that can be treated as a bare potential in the formally exact PT [21]. The total probability of radiative decay (excitation, de-excitation) is connected with imaginary part of ΔE of the system "atom plus field" [18-21]. The corresponding corrections of the PT for ImΔE can be represented as a sum on the virtual states. In the lowest PT the separated terms of these sums correspond to the additive contributions of different physical channels into the total decay probability. Naturally the channels interference effects will appear in the next PT orders. The fundamental parameter of the cooperative NEET process is a probability P_{NEET} (cross-section) of the nuclear excitation by electron transition. In fact it can be defined as the probability that the decay of the initial excited atomic state will result to the excitation of and subsequent decay from the nuclear state. Within the energy approach a decay probability is connected with an imaginary part of energy shift for the system (nuclear subsystem plus electron subsystem) excited state. An imaginary part of the excited state energy shift in the lowest PT order can be written as [1]:

$$\begin{aligned} &\mathrm{Im}\,\Delta E = e^2\,\mathrm{Im}\,i\cdot\lim_{\gamma\to 0}\iint d^4x_1 d^4x_2 e^{\gamma(t_1+t_2)}\cdot \\ &\cdot\{D(r_{N1t1},r_{N2t2})<\Psi_I\,|\,(\hat{J}_N(x_1)\hat{J}_N(x_2))\,|\,\Psi_I>+ \\ &+D(r_{e1t1},r_{e2t2})<\Psi_I\,|\,(\hat{j}_e(x_1)\hat{j}_e(x_2))\,|\,\Psi_I>\} \end{aligned} \tag{3}$$

Here $D(r_1t_1,r_2t_2)$ is the photon propagator (for example in the Lorenz gauge); $\hat{j}_N$, $\hat{j}_e$ — are the 4-dimensional components of a current operator for the nuclear and electron (hole) subsystems; $x=(r_n,r_e,t)$ is the four-dimensional space-time coordinate of the particles, respectively; γ is an adiabatic parameter. The nuclear current can be written as follows:

$$J^P(R,t)=\psi^+_{N^*}\hat{J}^P\psi_N, \tag{4}$$

where $\hat{J}^P$ is operator of an nuclear electromagnetic transition, ψ_N is a nuclear wave function. The current operator for electron is

$$\overline{j}^{\mu}_e=\overline{\widehat{\psi}}_e\gamma^{\mu}\widehat{\psi}_e, \tag{5}$$

where γ^{μ} are the Dirac matrices. The Hamiltonian of the interaction of the electronic hole current j^{μ}_{fi} and the nuclear current $J^{\nu}_{fi}(R)$ is written as :

$$H_{int}=\int d^3r\,d^3R\;j^{\mu}_{fi}\;D_{\mu\nu}(x_N,\,r\text{-}R)\;J^{\nu}_{fi}(R) \tag{6}$$

The energy shift can be further represented as the PT set. After integration transformations the final expression for the imaginary part of energy shift can be represented as a sum of the corresponding nuclear-electron (hole) contributions:

$$\mathrm{Im}\,\Delta E=\mathrm{Im}\,E_e+\mathrm{Im}\,E_N,$$

$$\mathrm{Im}\,E_a=-\frac{z_a^2}{4\pi}\sum_F\iint dr_{e1}dr_{e2}\iint dr_{N1}dr_{N2}\cdot\Psi^*_I(1)\Psi^*_F(2)\hat{T}_a(1,2)\Psi_F(1)\Psi_I, \tag{10}$$

$$\hat{T}_a(1,2)=\frac{\sin(\omega_{IF}r_{a12})}{r_{a12}}$$

Here, as usually, $r_{a12}=|r_{a1}-r_{a2}|$, ω_{IF} is the energy of transition between the initial I and final F states; the sum on F means the summation on the final states of a system. Naturally, the form of operator in (10) is determined by a gauge of the photon propagator (look discussion in Ref. [21]). In the zeroth approximation the dependence Ψ_F, Ψ_I on the nuclear and electron coordinates (R_N , $R_{e(h)}$) is factorized ($\sim\Phi_e\Phi_N$). Therefore, the combined electron (hole)- nuclear one-photon transitions occur as each of the operators T_N, T_e in (10) contains the combination of the nuclear and electron variables. After factorization and some transformations the expression (10) can be presented in the following form:

$$\begin{aligned}\mathrm{Im}\,E_a = -\frac{z_a^2}{4\pi}\sum_{F_eF_N}\int\int dR_{N1}dR_{N2}\int\int dR_{e1}dR_{e2}\cdot\Phi^*_{Ie}(R_{e1})\Phi^*_{IN}(R_{N1})\Phi^*_{Fe}(R_{e2})\cdot\\ \cdot\Phi^*_{FN}(R_{N2})\frac{\sin\omega_{IF}R_{a\,12}}{R_{a12}}\Phi_{Fe}(R_{e1})\Phi_{FN}(R_{N1})\Phi_{Ie}(R_{e2})\Phi_{IN}(R_{N2}).\end{aligned} \tag{11}$$

The expansion of the operator $\frac{\sin(\omega_{IF}R_{a12})}{R_{a12}}$ on the spherical harmonics generates the decay probability multipole expansion. It can be written in the following known form:

$$\frac{\sin|\omega|R_{12}}{R_{a12}}=\frac{\pi}{2\sqrt{R_1R_2}}\sum_{\lambda=0}^{\infty}(\lambda)J_{\lambda+1/2}(|\omega|R_{a1})J_{\lambda+1/2}(|\omega|R_{a2})P_\lambda(\cos\widehat{R_{a1}R_{a2}}), \tag{12}$$

where J is the Bessel function of the first kind and $(\lambda)=2\lambda+1$. In fact this expansion coincides with the known power expansion; naturally the strict decreasing contribution on multipolarity corresponds to them. In our problem the power expansion parameters are the combinations $\omega^a_{IF}R_e$, $\omega^N_{IF}R_N$. Further the effects of purely nuclear transition, purely electron-(hole) transition and combined electron – nuclear transition in (11) can be distinguished. The corresponding technique of work with these expansions is well developed [8,19-21] and often used in our precious papers (look [1-4]). Finally the NEET probability P_{NEET} is connected with the imaginary part of energy of the excited nuclear-electron state. It can be shown that P_{NEET} can be presented in the following form [17]:

$$\mathrm{P_{NEET}}=\left(1+\frac{\Gamma_\mathrm{i}}{\Gamma_\mathrm{f}}\right)\frac{M^2_{\mathrm{int}}}{(\omega_N-\omega_A)^2+(\Gamma_i+\Gamma_f)^2/4}\ . \tag{13}$$

Here, as usually, $\Gamma_{i,f}$ are the widths of the initial and final electron states; M^2 is averaged over initial states and summed over the final states the square modulus of the Hamiltonian of the electron hole current- nuclear current interaction. It can be written (M_I –K transition) as follows (see Ref. [17]):

$$M^2_{\mathrm{int}}=4\pi e^2\omega_N^{2(\lambda+1)}\frac{(j_i\,\frac{1}{2}\,\lambda O\,|\,j_f\,\frac{1}{2})^2}{[(2\lambda+1)!!]^2}\left|R^{E/M}_\lambda(\omega_N)\right|^2 B(E/M\lambda;J_i\to J_f). \tag{14}$$

Here $B[E/(M)\lambda;\ J_i\text{-}J_f]$ is the reduced nuclear probability, $\left|R^{E/M}_\lambda(\omega_N)\right|$ are the atomic radial matrix elements of electric (magnetic) [E/M] multipolarity λ; $j_{i,f}$ and $J_{i,f}$ are the angular momenta of the electronic and nuclear states correspondingly. The atomic radial matrix elements $\left|R^{M}_\lambda(\omega_N)\right|$ of electric (magnetic) [E/M] multipolarity λ are expressed by means the integral:

$$\int_0^\infty dr r^2 Z_\lambda^{(1)}(\omega r)[g_i(r)f_f(r)+f_i(r)g_f(r)], \quad (15)$$

where *f(r)* and *g(r)* are the large and small components of the Dirac electronic wave functions and

$$Z_\lambda^{(1)}=\left[\frac{2}{|\omega|\alpha Z}\right]^{\lambda+1/2}\frac{J_{\lambda+1/2}(\alpha|\omega|r)}{r^\lambda\Gamma(\lambda+3/2)} \quad (16)$$

Other details can be found in Refs. [1-4,18-21,23].

3. Results and conclusions

In concrete calculation of the NEET probabilities for different atomic/nuclear systems one should calculate the corresponding matrix elements. As we will consider below M1 (E2) transition from the ground state to the first excited state in the nuclei $^{189}_{76}Os$, $^{193}_{77}Ir$, $^{197}_{79}Au$, it should be noted that the values of $B[E/(M)\lambda; J_i\text{-}J_f]$ are usually taken from the Nuclear Data tables [33] or can be estimated according the known formula (look [24,25]). In order to calculate the electronic wave functions and matrix elements we have used the relativistic many-body PT formalism [1-4,18,23]. It allows take into account accurately the relativistic, exchange-correlation, nuclear, radiative corrections (the PC code "Superatom-ISAN"). The corresponding code contains the atomic and nuclear blocks. The zeroth approximation electronic wave functions are found from the Dirac (or Dirac-Kohn-Sham) equation with potential, which includes the SCF potential, the electric and polarization potentials of a nucleus. As an account of the finite nuclear size has a sensitive effect on the energy levels of the bound electron, we usually use the smooth Gaussian (or Fermi) function of the charge distribution in a nucleus. The correlation corrections of the second and high orders are taken into account within the Green functions method (with the use of the Feynman diagram's technique). There have taken into account all correlation corrections of the second order and dominated classes of the higher orders diagrams [18]. The magnetic inter-electron interaction is accounted in the lowest (on α^2 parameter; α is the hyperfine structure constant). The radiative corrections are taken into account effectively, namely, the Lamb shift self-energy part is accounted within the generalized Ivanov-Ivanova non-perturbative procedure and the polarization part - in the generalized Uehling-Serber approximation [18]. The important feature of the whole method is using the optimized one-quasiparticle representation in the zeroth approximation, which is constructed within the method [21]. The nuclear part of the general method includes a set of the nuclear shells models, including the relativistic mean-field approach and the Dirac-Bloumkvist-Wahlborn and Dirac-Woods-Saxon models [2,26-28]. The calculation results on the NEET probability for the $^{189}_{76}Os$, $^{193}_{77}Ir$, $^{197}_{79}Au$ atoms together with the alternative theoretical (by Tkalya and Ahmed et al) [13,16,17] and experimental data (see [11-13 and Refs. therein] are listed in the table 2. Let us note that in $^{189}_{76}Os$ during the NEET process the 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 (71.911keV, E2) atomic transitions can give the contribution.

Table 2. Theoretical and experimental probabilities P_{NEET} (M1) for $^{189}_{76}Os$, $^{193}_{77}Ir$, $^{197}_{79}Au$.

Nucleus	Energy of nuclear excitation (keV)	Experimental values	Theory: Tkalya and Ahmed etal	Present work
$^{189}_{76}Os$	69.535	$<9.5\cdot10^{-10}$	$1.2\cdot10^{-10}$ $1.3\cdot10^{-10}$	$1.9\cdot10^{-10}$
$^{193}_{77}Ir$	73.04	$(2.8\pm0.4)\cdot10^{-9}$	$2.0\cdot10^{-9}$	$2.7\cdot10^{-9}$
$^{197}_{79}Au$	77.351	$(5.7\pm1.2)\cdot10^{-8}$ $(4.5\pm0.6)\cdot10^{-8}$	$3.4\cdot10^{-8}$ $4.5\cdot10^{-8}$	$4.6\cdot10^{-8}$

The corresponding nuclear state at 69.535 keV can be excited via M1 or E2 transitions from the $3/2^-$ nuclear ground state. The following energy parameters ω_N=69.535 keV, $\omega_A=E_{MI}-E_K$=70.822 keV, Γ_K=42.6 eV, Γ_M=12.8 eV are used for the $^{189}_{76}Os$ atom. Correspondingly, the energy parameters for $^{197}_{79}Au$ are as follows: ω_N=77.351 keV, ω_A=77.325 keV, Γ_K=52 eV, Γ_M =14.3 eV and for $^{193}_{77}Ir$: ω_N=73.04 keV, ω_A=72.937 keV, Γ_K =45 eV, Γ_M =12.8 eV. Analysis of all presented theoretical data shows that these results are consistent with each other and are in the physically reasonable agreement with the experimental results.

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