PHYSICAL REVIEW C **88**, 054616 (2013)

General theory of nuclear excitation by electron transitions

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(Received 16 August 2013; published 25 November 2013)

We described the nuclear excitation by electron transition (NEET) by using strict collision theory combined with quantum electrodynamics. All stages of the process are considered, which include ionization of the atom by an x-ray photon with the formation of the hole in an inner electronic shell, its decay accompanied by the excitation of the nucleus, and the subsequent deexcitation of the nucleus. The cross sections for the NEET and photoabsorption of x rays are calculated near the threshold. We also analyzed the fine structure of the NEET curve analogous to the extended x-ray-absorption fine structure, well known in x-ray optics. The results agree with the findings of Kishimoto *et al*. [Phys. Rev. C **74**, 031301(R) (2006)].

DOI: 10.1103/PhysRevC.88.054616 PACS number(s): 32.30.Rj, 23.20.Nx, 27.70.+q, 27.80.+w

I. INTRODUCTION

X rays or electrons incident on the targets can ionize their inner atomic shells. The vacancies formed are filled by electrons from upper levels mainly with the emission of x-ray photons or Auger electrons. But there is one more, although weak, decay channel of vacancies—nuclear excitation by electron transition (NEET), predicted first by Morita [1]. It arises in those cases when atomic and nuclear transitions have near energies and the same multipolarities. This effect already has been observed for the nuclei ^{197}Au [2], ^{189}Os [3–6], and ^{237}Np [7]. An attempt to observe the NEET effect at the 76-eV level of ^{235}U in a plasma produced by lasers has also been undertaken in Ref. [8].

Most precise measurements were performed by Kishimoto *et al*. [2], who irradiated the golden foil, which contained ^{197}Au, by synchrotron radiation with a very narrow bandwidth of $\Gamma_s = 3.5$ eV. Incident x-ray photons, which ionized the K orbit of the Au atom, further the electron transition between the M_1 and K levels, and led to the excitation of the isotope ^{197}Au due to the exchange by virtual photons between the electron and the nucleus. The number of excited nuclei was counted by detecting the emitted L-conversion electrons. By varying the energy of the x-ray photons, they found that the NEET edge was shifted higher by 40 ± 2 eV than the K-photoabsorption edge and was steeper.

A general formula for the NEET probability P_{NEET} has been derived by Tkalya [9] and has been corrected slightly by Harston [10]. Alternative attempts to derive the P_{NEET} have been undertaken in Refs. [6,11]. In all these papers, only the restricted scheme of the NEET has been studied, when in the initial state, the atom had a hole in an inner shell and the nucleus was in the ground state, whereas, the final state consisted of the hole in a final upper-lying shell and the excited nucleus. The NEET itself was treated as a two-step exchange by virtual photons between the atomic electrons and the nucleus. Respectively, Tkalya [9] described this process in the second-order approximation of the quantum electrodynamics (QED). Unfortunately, such an approach has some shortcomings. First, for the description of the resonances, the real energies in the propagators were replaced *ad hoc* by their complex values. But such a replacement leads to an infinite value of the wave functions at the initial moment of the scattering process when the time $t \to -\infty$. Another unpleasant procedure was a "spreading" of the δ functions, which corresponds to the energy-conservation law. More definitely, the δ function with complex energies, which expresses the energy conservation law, has been replaced by the Lorentzian function.

More correctly, the NEET problem has been solved by Morel *et al*. [12] on the basis of the decay theory [13]. They treated the NEET as an exponential decay of the vacancy, which is formed in an inner shell at the initial moment $t = 0$.

All theories discussed above failed to explain behavior of the NEET curve at the threshold since they did not regard the formation of the vacancy by incident radiation as well as the final stages of the process, i.e., the relaxation of the final atomic hole followed by the decay of the excited nucleus. Partly, such a program has been realized by Tkalya [14], who took the initial stage of the NEET process into consideration, i.e., the K-hole formation by the incident x-ray photon. It has been performed again in the modified second-order QED approach, which has the same faults. Of course, the QED, which deals with the four-dimensional space where the spatial coordinates and time are treated quite symmetrically, is the best in such second-order calculations. But for the description of multistep transitions, such as NEET, the collision theory [13] turns out to be much simpler. It has been used in our previous paper [15] where all phases of the NEET were considered, which include the formation of the hole in an inner atomic shell by incident x-ray photon, the relaxation of the electronic subsystem accompanied by energy transfer to the nucleus, and its subsequent decay. Both the NEET and the absorption of x rays were analyzed on an equal footing in the framework of a nonrelativistic theory. The main features of the NEET and x-ray absorption curves as a function of x-ray photons were explained. But we, like all previous theorists, did not touch the fine structure of the NEET curve observed in Ref. [2]. As correctly noticed by Kishimoto [2], the observed oscillations of the NEET curve near the edge have the same nature as the analogous oscillations of the absorption cross section of x-ray photons in the vicinity of their absorption threshold. Such oscillations are well known in x-ray optics as the extended x-ray absorption fine structure (EXAFS) (see, e.g., Refs. [16,17]). They arise since photoelectrons, ejected from any atom of the crystal, suffer scattering by neighboring atoms. The electron waves, coherently scattered in the backward direction, interfere with the original electron wave, which gives rise to EXAFS oscillations.

Here, we give a complete analysis of the NEET process, induced by x-ray photons. To this aim, a strict collision theory is used with inclusion of the QED methods for the relativistic second-order calculation of the NEET strength.

At the initial NEET stage, the ionization of the atom by incident x-ray photon occurs. We first regard the ionization of an isolated atom by completely ignoring its crystal environment. This simplification enables us to describe the general behavior of both the NEET and the photoabsorption curves near their thresholds. And then we take into account rescattering of the photoelectrons by neighboring atoms of the crystal, that allows us to reproduce the fine structure of the NEET curve.

II. BASIC EQUATIONS

The unperturbed Hamiltonian of the system (nucleus plus atomic electrons plus quantized electromagnetic field) is

$$\hat{H}_0 = \hat{H}_n + \hat{H}_a + \hat{H}_{\text{rad}} + \hat{H}_c, \tag{1}$$

where $\hat{H}_n$, $\hat{H}_a$, $\hat{H}_{\text{rad}}$, and $\hat{H}_c$ are the Hamiltonians of the nucleus, atomic electrons, quantized electromagnetic fields, and the crystal, respectively.

The total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}_r$ contains the perturbation operator,

$$\hat{V}_r = \hat{V}_r^{(n)} + \hat{V}_r^{(a)} + \hat{V}_r^{(c)}, \tag{2}$$

where $\hat{V}_r^{(n)}$, $\hat{V}_r^{(a)}$, and $\hat{V}_r^{(c)}$ are the interaction operators of the nucleus, the electrons of the atom absorbing x-ray photon, and the electrons of the neighboring atoms in the crystal with the quantized electromagnetic field, respectively [18],

$$\hat{V}_r^{(n)} = \frac{1}{c}\int d\mathbf{R}\, \hat{J}(\mathbf{R})\hat{A}(\mathbf{R}), \quad \hat{V}_r^{(a)} = \frac{1}{c}\int d\mathbf{r}\, \hat{j}(\mathbf{r})\hat{A}(\mathbf{r}), \tag{3}$$

and $\hat{J}(\mathbf{r})$, $\hat{j}(\mathbf{r})$ are the four-dimensional electric current density operators of the nucleus and atomic electrons, respectively, and $\hat{A}(\mathbf{r})$ is the four-vector potential operator of the electromagnetic field.

The eigenfunctions and eigenvalues of the unperturbed Hamiltonian $\hat{H}_0$ obey the equation,

$$\hat{H}_0 \chi_b = E_b \chi_b. \tag{4}$$

Let, at the initial moment $t \to -\infty$, the system be described by the wave function,

$$\chi_a = |I_g M_g\rangle \Phi_0 |1_{\mathbf{ke}}\rangle, \tag{5}$$

where the wave function $|I_g M_g\rangle$ describes the initial state of the nucleus in the ground state with spin I_g and its projection M_g on the quantization axis, Φ_0 is the initial state of the atom, $|1_{\mathbf{ke}}\rangle$ is the quantized field, which contains one x-ray photon with the wave vector $\mathbf{k}$, the polarization $\mathbf{e}$, and the energy E. The corresponding initial energy of the system equals

$$E_a = E, \tag{6}$$

where the initial energies of the atom and nucleus are taken to be zero. From now on, for brevity, we omit the wave function and the energy of the crystal, which only enter the formula Eq. (68) for the ionization cross section. At first, an electron of any initial i shell ($i = K, L, \ldots$), which has absorbed the x-ray photon, flies away with the wave vector $\vec{\kappa}$ and energy ε. Note that this energy in metals is counted from the Fermi level. Such a first intermediate state of the system is described by the wave function,

$$|c_1\rangle = |I_i M_i\rangle \Phi_{j_i m_i} |\vec{\kappa}\rangle |0\rangle, \tag{7}$$

where $|0\rangle$ stands for the wave function of the vacuum of the electromagnetic field, $|\vec{\kappa}\rangle$ is the function of the ejected electron, $\Phi_{j_i m_i}$ is the wave function of the atom that contains a hole in the i shell, specified by the quantum numbers $j_i m_i$. The corresponding eigenvalue of the unperturbed Hamiltonian $\hat{H}_0$ will be

$$E_1 = B_i + \varepsilon. \tag{8}$$

Hereafter, B_i and B_f are the binding energies of the electron in the initial and final (f) states.

Afterwards, the hole passes to level $|j_f m_f\rangle$ with the excitation of the nucleus so that the whole system undergoes a transition into the second intermediate state,[1]

$$|c_2\rangle = |I_e M_e\rangle \Phi_{j_f m_f} |\vec{\kappa}\rangle |0\rangle, \tag{9}$$

which has the energy,

$$E_2 = W_e^{(n)} + B_f + \varepsilon, \tag{10}$$

where $|I_e M_e\rangle$ and $W_e^{(n)}$ are the wave function and the energy of the excited nucleus. The nuclear and electronic transition energies involved in NEET are $E_0^{(n)} = W_e^{(n)}$ and $E_0^{(a)} = B_i - B_f$ with their mismatch,

$$\delta = E_0^{(n)} - E_0^{(a)}. \tag{11}$$

In principle, there exist two decay branches of state $|c_2\rangle$: deexcitation of the nucleus and decay of the f vacancy. But the hole widths $\Gamma_{i(f)}$ are, by many orders, larger than the nuclear width Γ_n. In particular, the width of the first excited level of ^{197}Au is $\Gamma_n = 2.38 \times 10^{-7}$ eV, whereas, the K- and M_1-hole widths are $\Gamma_K = 52$ and $\Gamma_{M_1} = 14.3$ eV [2]. Thus, at first, fast decay of the f hole occurs and only then slow nuclear decay. We will talk about single electron transition from an upper level with the binding energy B' into the f hole, which is accompanied by the emission of the photon that has the energy $E' = \hbar\omega'$.

In such a case, the system passes into another intermediate state $|c_3\rangle$ with the energy,

$$E_3 = W_e^{(n)} + B' + \varepsilon + E'. \tag{12}$$

After that, the nucleus decays into a final state $|I_f M_f\rangle$ by emitting a γ quantum with the energy $E_\gamma = \hbar\omega_\gamma$ or by means of internal conversion. For definiteness, we will only talk about

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[1] By following tradition, we call this hole state final, although its subsequent decay should also be traced.

the radiative decay. The energy of the whole system in final state $|b\rangle$ then becomes

$$E_b = W_f^{(n)} + B' + \varepsilon + E' + E_\gamma. \tag{13}$$

Such a multistep scattering process is determined by the resonant part of the transition operator,

$$\hat{T} = \hat{V}_r^{(n)} \hat{G}^+(E_a) \hat{V}_r^{(a)}, \tag{14}$$

where

$$\hat{G}^+(E_a) = (E_a + i\eta - \hat{H})^{-1} \tag{15}$$

is the Green's operator $\eta \to +0$.

In (14), we would like to separate the parts which describe the sequence of events discussed above. It can be performed in full analogy with the description of the inelastic scattering of electrons by atoms with the subsequent emission of photons [13]. To this aim, we introduce the operators,

$$\hat{\mathcal{A}} = E_a + i\eta - \hat{H}, \quad \hat{\mathcal{B}} = E_a + i\eta - \hat{H}_0 - \hat{V}_r^{(n)}, \tag{16}$$

With the aid of the operator identity (see also Ref. [13]),

$$\frac{1}{\hat{\mathcal{A}}} - \frac{1}{\hat{\mathcal{B}}} = \frac{1}{\hat{\mathcal{B}}}(\hat{\mathcal{B}} - \hat{\mathcal{A}})\frac{1}{\hat{\mathcal{A}}}, \tag{17}$$

we rewrite the transition operator (14) as

$$\hat{T} = \hat{V}_r^{(n)} \frac{1}{E_a + i\eta - \hat{H}_0 - \hat{V}_r^{(n)}} \hat{\mathcal{T}}, \tag{18}$$

where the new transition operator is

$$\hat{\mathcal{T}} = \hat{V}_r^{(a)} + \hat{V}_r^{(a)} \hat{G}^+(E_a) \hat{V}_r^{(a)}. \tag{19}$$

By performing this in the same manner, one can separate the initial transition operator $\hat{T}_{\mathrm{i}}$, responsible for the ionization of the atom by x rays and rescattering of the emitted electron by the adjacent atoms of the crystal,

$$\hat{T}_{\mathrm{i}} = \hat{V}_r^{(a)} + \hat{V}_i \hat{G}^+(E) \hat{V}_r^{(a)}, \tag{20}$$

where $\hat{V}_i = \hat{V}_r^{(1)} + \hat{V}_r^{(c)}$ and the operator $\hat{V}_r^{(1)} = c^{-1} \int j_1(\mathbf{r}) A(\mathbf{r}) d\mathbf{r}$ determine interaction with the field of an electron, which absorbs the x-ray photon and flies away from the atom. The interaction operator $\hat{V}_r^{(c)}$ of the neighboring atoms with the field is inserted into $\hat{V}_i$ to describe the rescattering of the photoelectron by these atoms.

Finally, one has

$$\hat{\mathcal{T}} = \hat{V}_r^{(a)} \hat{G}'^+(E) \hat{T}_{\mathrm{i}}, \tag{21}$$

where $\hat{V}_r'^{(a)} = \hat{V}_r^{(a)} - \hat{V}_r^{(1)}$ and $\hat{G}'^+(E)$ denotes the resolvent of the operator $\hat{H} - \hat{V}_i$ (below, we will omit the prime).

Then, the matrix elements for the transition operator (18) take the form

$$\begin{aligned} T_{ba} = \sum_{c_1,c_2,c_3} &\langle b|\hat{V}_r^{(n)}|c_3\rangle \frac{1}{E_a - E_3 + i\Gamma_n/2} \langle c_3|\hat{V}_r^{(a)}|c_2\rangle \\ &\times G_{21}^+(E) \langle c_1|\hat{T}_{\mathrm{i}}|a\rangle, \end{aligned} \tag{22}$$

where the off-diagonal elements of the Green's matrix $G_{21}^+ \equiv \langle c_2|\hat{G}^+(E)|c_1\rangle$ couple two overlapping resonant levels c_1 and c_2. The sums over c_i mean the summation over the magnetic quantum numbers M and m, respectively, for the nucleus and the atom.

The Green's matrix $G_{c'c}^+(E_a)$ is determined by a system of algebraic equations [19],

$$\begin{aligned} (E_a - E_1 - R_{11}^+) G_{11}^+ - \sum_{M_e, m_f} R_{12}^+ G_{21}^+ &= 1, \\ -R_{21}^+ G_{11}^+ - (E_a - E_2 - R_{22}^+) G_{21}^+ &= 0, \end{aligned} \tag{23}$$

where the R matrix is given by the expansion,

$$R_{cc'}^+(E_a) = V_{c,c'} + \sum_{d \neq c,c'} \frac{V_{cd} V_{dc'}}{E_a + i\eta - E_d} + \cdots. \tag{24}$$

The imaginary parts of its diagonal elements are $R_{11} = -i\Gamma_i/2$, $R_{22} = -i\Gamma_f/2 - i\Gamma_n/2$, whereas, the real parts already are included in the energies of the nuclear and electronic levels.

The solution of Eq. (23) has the form

$$G_{11}^+ = \frac{\Delta E - \varepsilon - \delta - R_{22}}{[\Delta E - \varepsilon - R_{11}][\Delta E - \varepsilon - \delta - R_{22}] - E_{\mathrm{int}}^2}, \tag{25}$$

and

$$G_{21}^+ = \frac{R_{21}}{[\Delta E - \varepsilon - R_{11}][\Delta E - \varepsilon - \delta - R_{22}] - E_{\mathrm{int}}^2}. \tag{26}$$

Here, $\Delta E = E - B_i$ specifies the energy excess of the x-ray photon over the x-ray absorption threshold B_i, and

$$E_{\mathrm{int}}^2 = \sum_{M_e, m_f} |R_{21}^+(E_a)|^2 \tag{27}$$

determines the NEET strength.

It is convenient to split the Green's functions into a sum of two terms associated with two overlapping resonant levels of the system.[2] In particular, one has

$$G_{21}^+(E_a) = \frac{R_{21}}{\mu_+ - \mu_-} \left(\frac{1}{\varepsilon - \mu_+} - \frac{1}{\varepsilon - \mu_-} \right), \tag{28}$$

where

$$\begin{aligned} \mu_+ &= \Delta E + \Delta_{\mathrm{BIC}} + i(\Gamma_i - \Gamma_{\mathrm{BIC}})/2, \\ \mu_- &= \Delta E - \delta - \Delta_{\mathrm{BIC}} + i(\Gamma_f + \Gamma_n + \Gamma_{\mathrm{BIC}})/2. \end{aligned} \tag{29}$$

Here, we introduced the following notations:

$$\Delta_{\mathrm{BIC}} = \frac{\delta}{\delta^2 + (\Gamma_i - \Gamma_f)^2/4} E_{\mathrm{int}}^2, \tag{30}$$

and

$$\Gamma_{\mathrm{BIC}} = \frac{\Gamma_i - \Gamma_f}{\delta^2 + (\Gamma_i - \Gamma_f)^2/4} E_{\mathrm{int}}^2. \tag{31}$$

The parameters Δ_{BIC} and Γ_{BIC} mean the shift and broadening of the resonant levels caused by the bound internal conversion (BIC), which is the reverse with respect to the NEET process. In this case, the excited nucleus transfers its energy back to an electron of the i shell, which returns to the vacant f level. Our expression (31) for the BIC width

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[2] Such a procedure for the overlapping resonances has been proposed in Ref. [21].

coincides with the standard formula (see, e.g., Ref. [20]) if we omit Γ_f. The width Γ_f enters our formulas since the f vacancy decays with the rate $\Gamma_f/\hbar$, which blocks the BIC channel in accordance with the Pauli principle. The estimations [15] show that

$$\Delta_{\mathrm{BIC}}, \Gamma_{\mathrm{BIC}}, \Gamma_n \ll \Gamma_{i(f)}. \tag{32}$$

Therefore, below, we will neglect both the BIC parameters and the nuclear width.

III. NEET COUPLING PARAMETER

Now, we will calculate the coupling parameter E^2_{int}, defined by Eq. (27). The corresponding matrix element R_{21} is determined by the second-order term of the expansion (24),

$$R_{21} = \sum_{d \neq c_1, c_2} \frac{\langle c_2|\hat{V}_r|d\rangle\langle d|\hat{V}_r|c_1\rangle}{\hbar\omega_{ad} + i\eta}, \tag{33}$$

where $\hbar\omega_{ad} = E_a - E_d$. The summation in (24) includes integration over wave vectors $\mathbf{q}$ of the virtual photons along with the summation over their polarizations and corresponding magnetic quantum numbers.

The transition from state $|c_1\rangle$ to $|c_2\rangle$ with the emission of the virtual photon, which has the frequency ω, wave vector $\mathbf{q}$, and polarization $\mathbf{e}_\alpha$, may proceed via two intermediate states,

$$|d_1\rangle = |I_g M_g\rangle|j_f m_f\rangle|\vec{\kappa}\rangle|1_{\mathbf{q},\alpha}\rangle, \tag{34}$$

and

$$|d_2\rangle = |I_e M_e\rangle|j_i m_i\rangle|\vec{\kappa}\rangle|1_{\mathbf{q},\alpha}\rangle. \tag{35}$$

The R matrix splits into two terms,

$$\begin{aligned} R_{21} = &\sum_{d_1} \frac{\langle c_2|\hat{V}^{(n)}_r|d_1\rangle\langle d_1|\hat{V}^{(a)}_r|c_1\rangle}{-\hbar(\omega - \omega_1) + i\eta} \\ &+ \sum_{d_2} \frac{\langle c_2|\hat{V}^{(a)}_r|d_2\rangle\langle d_2|\hat{V}^{(n)}_r|c_1\rangle}{-\hbar(\omega + \omega_2) + i\eta}, \end{aligned} \tag{36}$$

respectively, where frequencies ω_1 and ω_2 are

$$\begin{aligned} \omega_1 &= \left(E^{(a)}_0 + \Delta E - \varepsilon\right)/\hbar, \\ \omega_2 &= \left(E^{(a)}_0 - \Delta E + \delta + \varepsilon\right)/\hbar. \end{aligned} \tag{37}$$

Near the photoabsorption threshold, $\omega_2 \approx \omega_1 \approx \omega_0 = E^{(a)}_0/\hbar$.

To calculate E^2_{int}, it is useful to transform (36) to the form, which includes the photon propagator [18],

$$D_{\mu\nu}(x - x') = \frac{i}{\hbar c}\langle 0|\hat{T}\hat{A}_\mu(x)\hat{A}_\nu(x')|0\rangle, \tag{38}$$

where $\hat{T}$ is the chronological operator. This propagator depends on the difference $x - x'$ of the four-dimensional vectors $x = \{ct, \mathbf{r}\}$ and $x' = \{ct', \mathbf{R}\}$ with the time difference $\tau = t - t'$. Its introduction, which makes the bridge between collision theory and QED, is realized easily with the aid of identity,

$$\frac{1}{\hbar\omega_{ad} + i\eta} = -\frac{i}{\hbar}\int_0^\infty e^{i\omega_{ad}\tau - \eta\tau}d\tau. \tag{39}$$

By substituting the relation (39) into (36), one has

$$\begin{aligned} R_{21} = &\int d\mathbf{r}\int d\mathbf{R}\, j^\mu(\mathbf{r})_{fi} J^\nu(\mathbf{R})_{eg} \\ &\times \left(-\frac{i}{\hbar}\right)\int_{-\infty}^\infty d\tau\, e^{-i\omega|\tau| + i\omega_0\tau}\langle 0|A_\mu(\mathbf{r})A_\nu(\mathbf{R})|0\rangle. \end{aligned} \tag{40}$$

Here, the integral over time is nothing but the Fourier transform of the photon propagator, that is,

$$R_{21} = -\int d\mathbf{r}\int d\mathbf{R}\, \hat{j}^\mu(\mathbf{r})_{fi}\hat{J}^\nu(\mathbf{R})_{eg}D_{\mu\nu}(\omega_0, \mathbf{r} - \mathbf{R}), \tag{41}$$

where $D_{\mu\nu}(\omega_0, \mathbf{r} - \mathbf{R})$ is the photon propagator in the mixed frequency-coordinate representation [18],

$$D_{\mu\nu}(\omega_0, \mathbf{r} - \mathbf{R}) = -g_{\mu\nu}\frac{e^{ik_0|\mathbf{r} - \mathbf{R}|}}{|\mathbf{r} - \mathbf{R}|}, \tag{42}$$

with the wave vector $k_0 = \omega_0/c$ and the metric matrix $g_{\mu\nu}$.

Substituting (41) and (42) into (27) for pure transition of the multipolarity L and type $\pi = E$ or M, one gets the result, derived previously in [10]

$$\begin{aligned} E^2_{\mathrm{int}} = 4\pi e^2 k_0^{2L+2}&\frac{\left(j_i\tfrac{1}{2}L0|j_f\tfrac{1}{2}\right)^2}{L^2\,[(2L+1)!!]^2} \\ &\times |M_e(\pi L)|^2 B(\pi L; I_g \to I_e), \end{aligned} \tag{43}$$

where $B(\pi L; I_g \to I_e)$ is the reduced probability of the nuclear transition, $M_e(\pi L)$ stands for the matrix element for the electronic transition (for its relativistic expression, see Refs. [9,10]). It is interesting to note, however, that simple nonrelativistic calculations [22] of the NEET probability for the 31-y isomer $^{178}\mathrm{Hf}^{m2}$ practically give the same numbers as the cumbersome relativistic Hartree-Fock calculations [23,24].

IV. NEET CROSS SECTION

The cross section for the transition from $|a\rangle$ to $|b\rangle$ is determined by the formula,

$$\sigma_{a\to b} = \frac{2\pi}{\hbar c}|T_{ba}|^2\delta(E_a - E_b), \tag{44}$$

with the transition matrix T_{ba} defined in Eq. (22).

After substitution of (22) into (44), we yet have to average $\sigma_{a\to b}$ over the initial states and sum over all possible final $|b\rangle$'s. Such a summation includes integration over the states of an emitted γ quantum with frequency ω_γ, x-ray photon ω', and a photoelectron. We rewrite the volume element in the integral for the electron $d\vec{\kappa}/(2\pi)^3$ as $d\Omega_{\vec{\kappa}}\rho_e(\varepsilon)d\varepsilon$ with the solid angle $d\Omega_{\vec{\kappa}}$ and the density of the electronic states in the continuous spectrum $\rho_e(\varepsilon) = m\kappa/(2\pi)^3\hbar^2$, where m is the electron mass. By performing first integration over ω' with the aid of the δ function, we find that

$$\begin{aligned} \sigma_{\mathrm{NEET}}(E) = &\int_0^\infty \frac{\Gamma_n(e \to f)/2\pi}{\left(E_\gamma - E^{(0)}_\gamma\right)^2 + (\Gamma_n/2)^2}dE_\gamma \\ &\times \frac{\Gamma_f}{2\pi}\int_0^\infty d\varepsilon|G^+_{21}(E)|^2\sigma_{\mathrm{ion}}(\varepsilon), \end{aligned} \tag{45}$$

where $E_\gamma^{(0)} = W_e^{(n)} - W_f^{(n)}$ is the transition energy to the final state of the nucleus, $\Gamma_n(e \to f)$ is the partial nuclear width for the transition from $|I_e M_e\rangle$ to $|I_f M_f\rangle$, Γ_n is the total width of the excited state, and $\sigma_{\text{ion}}(\varepsilon)$ represents the ionization cross section,

$$\sigma_{\text{ion}}(\varepsilon) = \frac{2\pi}{\hbar c} \sum_{m_i} \int d\Omega_{\vec{\kappa}} |\langle c_1|\hat{T}_{\text{i}}|a\rangle|^2 \rho_e(\varepsilon). \tag{46}$$

Trivial integration over E_γ immediately gives the branching ratio,

$$R(e \to f) = \Gamma_n(e \to f)/\Gamma_n. \tag{47}$$

Then, the reaction cross section will be equal to the product of the nuclear excitation cross section $\sigma_{\text{NEET}}(E)$ and the branching ratio,

$$\sigma_r(E) = \sigma_{\text{NEET}}(E) R(e \to f). \tag{48}$$

As to the NEET cross section, it is determined by the integral over the electron energy (see also Refs. [14,15]),

$$\sigma_{\text{NEET}}(E) = E_{\text{int}}^2 \left(\frac{\Gamma_f}{2\pi}\right) \int_0^\infty \frac{\sigma_{\text{ion}}(\varepsilon) d\varepsilon}{[(\varepsilon - \Delta E)^2 + (\Gamma_i/2)^2][(\varepsilon - \Delta E + \delta)^2 + (\Gamma_f/2)^2]}. \tag{49}$$

Previously [14,15], the cross section $\sigma_{\text{ion}}(\varepsilon)$ was replaced by a constant $\sigma_{\text{ion}}^{(at)}$, which is possible if one ignores the environment of the atom, which absorbs the x-ray photon and then emits a photoelectron. In other words, $\sigma_{\text{ion}}^{(at)}$ means the ionization cross section of a single atom when the rescattering of the emitted electron wave by the neighboring atoms of the crystal is disregarded. In the Born approximation, the transition operator $\hat{T}_{\text{i}}$ in Eq. (46) should be replaced by $\hat{V}_r^{(a)}$.

Calculations of the ionization cross section $\sigma_{\text{ion}}^{(at)}$ at the K shell, presented in Ref. [25], show that it can really be treated as a constant. When

$$\Delta E, \Delta E - \delta \gg \Gamma_{i,} \Gamma_f, \tag{50}$$

the lower limit of integration in (49) may be replaced by $-\infty$. Then, the integral is calculated easily by the contour integration, which gives

$$\sigma_{\text{NEET}}(\infty) = P_{\text{NEET}} \sigma_{\text{ion}}^{(at)}, \tag{51}$$

where the designation $\sigma_{\text{NEET}}(\infty)$ is associated with energies which satisfy the condition (50) and P_{NEET} is the decay probability of the i hole through the NEET channel, calculated earlier in [9,10,12]

$$P_{\text{NEET}} = \left(1 + \frac{\Gamma_f}{\Gamma_i}\right) \frac{E_{\text{int}}^2}{\delta^2 + (\Gamma_i + \Gamma_f)^2/4}. \tag{52}$$

In the vicinity of the threshold, the NEET cross section can be written as

$$\sigma_{\text{NEET}}(E) = \sigma_{\text{NEET}}(\infty) F_{\text{NEET}}(E), \tag{53}$$

where $F_{\text{NEET}}(E)$ describes the energy dependence of the NEET cross section at the threshold. With the aid of the partition (28), we find this edge factor,

$$F_{\text{NEET}}(E) = \frac{f_+(E) + f_-(E) + f_\pm(E)}{(1 + \Gamma_f/\Gamma_i)[\delta^2 + (\Gamma_i - \Gamma_f)^2/4]}, \tag{54}$$

with

$$f_+(E) = \frac{\Gamma_f}{\Gamma_i}\left[\delta^2 - \left(\frac{\Gamma_i}{2}\right)^2 + \left(\frac{\Gamma_f}{2}\right)^2\right] \times \left[\frac{1}{2} + \frac{1}{\pi}\arctan\left(\frac{2\,\Delta E}{\Gamma_i}\right)\right],$$

$$f_\pm(E) = \frac{\delta \Gamma_f}{2\pi} \ln\left[\frac{(\Delta E)^2 + (\Gamma_i/2)^2}{(\Delta E - \delta)^2 + (\Gamma_f/2)^2}\right],$$

$$f_-(E) = \left[\delta^2 + \left(\frac{\Gamma_i}{2}\right)^2 - \left(\frac{\Gamma_f}{2}\right)^2\right] \times \left[\frac{1}{2} + \frac{1}{\pi}\arctan\left(\frac{2(\Delta E - \delta)}{\Gamma_f}\right)\right], \tag{55}$$

where the terms $f_+(E)$ and $f_-(E)$ represent the contribution into the integral of the squared terms of G_{21}^+ with the poles μ_+ and μ_-, respectively, whereas, $f_\pm(E)$ is associated with their interference. Our formulas (53)–(55) agree with those of Ref. [14], except for the factor $1/\pi$, missed in Ref. [14] in the expressions for F_1, F_2. Once $\Delta E \to \infty$, the factor $F_{\text{NEET}}(E) \to 1$. Note that, in (55), the term $f_-(E)$ for ^{197}Au is larger than $f_+(E)$ and $f_\pm(E)$ since $\Gamma_f \ll \Gamma_i$. Specifically, for the Au atom, $\Gamma_{M_1}/\Gamma_K \approx 0.3$. Therefore, the significant rise in the NEET curve, described by the edge factor $F_{\text{NEET}}(E)$, occurs only in the vicinity of the point $\Delta E = \delta$ (see Fig. 1).

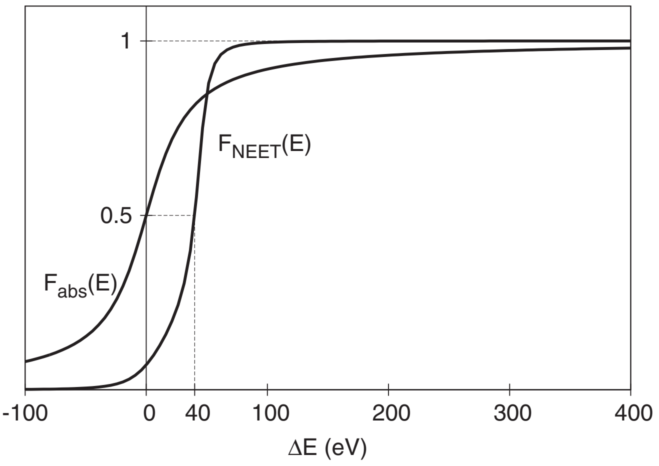

FIG. 1. The calculated NEET edge function $F_{\text{NEET}}(E)$ and the K-absorption one $F_{\text{abs}}(E)$ for ^{197}Au versus the energy $\Delta E = E - B_K$ of the x-ray photons.

V. ABSORPTION OF X RAYS

The absorption cross section of x rays by electrons according to the optical theorem (see, e.g., Ref. [13]) is determined by the imaginary part of the transition matrix element T_{aa}, which describes the elastic scattering of the x-ray photons to the zero angle,

$$\sigma_a(E) = \frac{2}{\hbar c}\,\mathrm{Im}\,T_{aa}. \tag{56}$$

From estimations (32), it follows that the NEET matrix element is $|R_{21}| \ll \Gamma_f$. As a result, one has the inequality $|G_{21}^{+}(E_a)| \ll |G_{11}^{+}(E_a)|$, which means that the absorption of x rays by the electrons of the i shell proceeds mainly without an energy transfer to the nucleus. In the same approximation, the expression (25) for $G_{11}(E_a)$ simplifies

$$G_{11}^{+}(E_a) \approx \frac{1}{\Delta E - \varepsilon + i\Gamma_i/2}. \tag{57}$$

As a result, the transition matrix becomes

$$T_{aa} = \sum_{m_i}\int \frac{d\vec{\kappa}}{(2\pi)^3}\langle a|\hat{T}_{\rm i}|c_1\rangle G_{11}^{+}(E_a)\langle c_1|\hat{T}_{\rm i}|a\rangle. \tag{58}$$

And for the absorption cross section, we have the following expression:

$$\sigma_a(E) = \frac{\Gamma_i}{2\pi}\int_0^\infty \frac{\sigma_{\rm ion}(\varepsilon)d\varepsilon}{(\varepsilon - \Delta E)^2 + (\Gamma_i/2)^2}. \tag{59}$$

Again, by replacing $\sigma_{\rm ion}(\varepsilon)$ with a constant $\sigma_{\rm ion}^{(at)}$, one gets

$$\sigma_a(E) = F_{\rm abs}(E)\sigma_{\rm ion}^{(at)}, \tag{60}$$

where the photoabsorption edge factor,

$$F_{\rm abs}(E) = \frac{1}{2} + \frac{1}{\pi}\arctan\left[\frac{2\,\Delta E}{\Gamma_f}\right]. \tag{61}$$

VI. AVERAGING

The cross sections $\sigma_a(E)$ and $\sigma_{\rm NEET}(E)$ should yet be averaged over the energy distribution of the incident photons. It is convenient to approximate it by the Lorentzian function,

$$w_s(E) = \frac{\Gamma_s/2\pi}{(E-\bar{E})^2 + (\Gamma_s/2)^2}, \tag{62}$$

where $\bar{E}$ and Γ_s are the average energy and the width of the distribution of photons emitted by a source. It may be rewritten in the integral form

$$w_s(E) = \frac{1}{\pi}\mathrm{Re}\int_0^\infty e^{i(E-\bar{E})\mu - \Gamma_s\mu/2}d\mu. \tag{63}$$

Then, the edge factor $F_{\rm abs}(E)$ convoluted with the distribution $w_s(E)$ takes the form

$$\begin{aligned}\mathcal{F}_{\rm abs}(\bar{E}) &\equiv \langle F_a(E)\rangle\\ &= \frac{\Gamma_i}{2\pi^2}\times\int_0^\infty d\varepsilon\,\mathrm{Re}\int_0^\infty d\mu\int_{-\infty}^{\infty}dE\\ &\quad\times\frac{e^{i(E-\bar{E})\mu - \Gamma_s\mu/2}}{(E-B_i-\varepsilon)^2 + (\Gamma_i/2)^2}.\end{aligned} \tag{64}$$

By the contour integration, we first calculate the integral over E, then over μ and ε. Finally, one arrives at the following formula for the averaged absorption edge factor:

$$\mathcal{F}_{\rm abs}(\bar{E}) = \frac{1}{2} + \frac{1}{\pi}\arctan\left[\frac{2\,\Delta\bar{E}}{\Gamma_{\rm a}}\right], \tag{65}$$

where $\Delta\bar{E} = \bar{E} - B_K$ and the width $\Gamma_{\rm a} = \Gamma_s + \Gamma_i$.

In analogy, only by keeping the leading term $f_{-}(E)$ in (54) do we see that the averaged NEET cross section $\bar{\sigma}_{\rm NEET}(\bar{E})$ in units of $\sigma_{\rm ion}^{(at)}$ is equal to the factor,

$$\mathcal{F}_{\rm NEET}(\bar{E}) = \frac{1}{2} + \frac{1}{\pi}\arctan\left[\frac{2(\Delta\bar{E} - \delta)}{\Gamma_{\rm NEET}}\right], \tag{66}$$

where the width is $\Gamma_{\rm NEET} = \Gamma_s + \Gamma_f$.

Both the absorption and the NEET near their edges are well described by their derivatives of (65) and (66) over the mean energy $\bar{E}$,

$$\begin{aligned}\mathcal{F}'_{\rm abs}(\bar{E}) &= \frac{(\Gamma_{\rm abs}/2)^2}{(\Delta\bar{E})^2 + (\Gamma_{\rm abs}/2)^2},\\ \mathcal{F}'_{\rm NEET}(\bar{E}) &= \frac{(\Gamma_{\rm NEET}/2)^2}{(\Delta\bar{E} - \delta)^2 + (\Gamma_{\rm NEET}/2)^2}.\end{aligned} \tag{67}$$

These functions are represented by the Lorentzians with the edge widths $\Gamma_{\rm abs}$ and $\Gamma_{\rm NEET}$.

VII. NEET FINE STRUCTURE

The general expression for the ionization cross section of the K shell, which takes the interference of the photoelectron waves into account, coherently scattered in a crystal, has the form (see, e.g., Ref. [16])

$$\begin{aligned}\sigma_{\rm ion}(\varepsilon) = \sigma_{\rm ion}^{(at)}\Bigg[1 + \sum_j \frac{1}{\kappa R_j^2}|f_j(\pi)|e^{-2\kappa^2\overline{u^2}}e^{-2R_j/\lambda(\kappa)}\\ \times\sin[2\kappa R_j + 2\delta_1(\kappa) + \theta_j(\kappa)]\Bigg],\end{aligned} \tag{68}$$

where $\sigma_{\rm ion}^{(at)}$ is the ionization cross section of an isolated atom, R_j is the distance from the atom, which emits a photoelectron to the neighboring jth atom, $\delta_1(\kappa)$ is the phase shift of the partial electron wave with the orbital angular momentum $l = 1$ scattered by the atom, $f_j(\pi) = |f_j(\pi)|e^{i\theta_j}$ is the coherent backscattering amplitude of the electrons by the jth atom, which is fixed in the crystal site, and its vibrations give rise to the factor $e^{-2\kappa^2\overline{u^2}}$, where $\overline{u^2}$ denotes a mean-squared amplitude of the vibrations of the atom about its equilibrium position. Attenuation of the wave inside the crystal is characterized by the factor $e^{-2R_j/\lambda(k)}$, where $\lambda(\kappa)$ stands for the mean-free path of the electrons in the medium. Equation (68) describes the oscillating cross section, whose beats attenuate with an increase in electron energy ε. Therefore, far away from the absorption threshold, the cross section $\sigma_{\rm ion}(\varepsilon)$ takes the constant value $\sigma_{\rm ion}^{(at)}$. In Eq. (68), a short lifetime of the vacancy in the atomic shell is neglected, but it manifests itself in the more accurate formula (59) to give a complete description of the x-ray absorption.

To estimate the amplitude $\overline{u^2}$, we use the Debye model where

$$\overline{u^2} = \frac{3\hbar^2}{M}\frac{1}{k_B\theta_D}\int_0^{\theta_D/T}\left[\frac{1}{e^x-1}+\frac{1}{2}\right]x\,dx, \tag{69}$$

θ_D is the Debye temperature, and M is the mass of the atom. For the Au crystal with $\theta_D = 165$ K, we obtained $(\overline{u^2})^{1/2} \approx 0.050$ Å.

We approximate the interaction of electrons with atoms of the crystal by a screened Coulomb potential,

$$V_c(r) = -\frac{Ze^2}{r}e^{-r/r_0}, \tag{70}$$

where r_0 is the screening length and Z is the charge number of the atom. Following Ref. [26], for r_0, we take the characteristic atomic size $r_a = a_0 Z^{-1/3}$, predicted by the Thomas-Fermi model, where a_0 is the Bohr radius of the atom. In particular, for gold, $r_0 \approx 0.13$ Å. For the elastic backscattering amplitude of electrons in the potential (70), we employ the expression,

$$f_B(\pi) = \frac{2mZe^2}{\hbar^2}\frac{1}{4\kappa^2+1/r_0^2}, \tag{71}$$

calculated in the plane-wave Born approximation, where m is the electron mass. We see that $f_B(\pi)$ is a real number, i.e., its phase is $\theta_j(\kappa) = 0$. Besides, in analogy with the EXAFS analysis of Emura *et al.* [27], we set the phase $\delta_1(\kappa)$ to be zero as well.

At last, for the mean-free path of the electrons, we apply the empirical formula of Teo [17],

$$\lambda(k) = \left(\frac{1}{0.53}\right)\left[\left(\frac{3.9}{\tilde{k}}\right)^4 + \tilde{k}\right], \tag{72}$$

where the dimensionless number is $\tilde{k} = k \times 1$ Å.

Equation (49) combined with (68) completely determines the NEET cross section. It has been calculated numerically for ^{197}Au in the golden crystal with a face-centered-cubic lattice where the edge length is $a = 4.0782$ Å. In the sum over j of Eq. (68), we took the contribution of 12 nearest neighbors separated by $R_1 = a/\sqrt{2}$ and 6 nearest neighbors separated by $R_2 = a$ into account. The calculated NEET cross section $\sigma_{\mathrm{NEET}}(\varepsilon)$, in units of $\sigma_{\mathrm{NEET}}(\infty)$, is presented in Fig. 2.

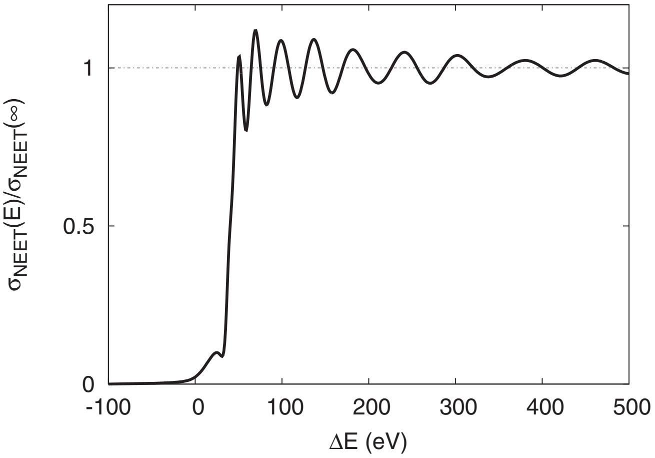

FIG. 2. The NEET cross section $\sigma_{\mathrm{NEET}}(E)$ in units of $\sigma_{\mathrm{NEET}}\infty$.

VIII. DISCUSSION

The NEET process is caused by a resonant coupling of the nuclear level and atomic hole levels via the electromagnetic field. We studied both the photoabsorption of x rays and the photoinduced NEET by making use of the collision theory. It allowed us to consider all phases of the NEET, which start with the hole formation along with the rescattering of the photoelectrons and end with the deexcitation of the nucleus. The system nucleus plus atomic electrons has two resonant levels, which are associated with two poles of the Green's matrix (54), which lie in the points $\mu_+ \approx \Delta E + i\Gamma_i/2$ and $\mu_- \approx \Delta E - \delta + i(\Gamma_f + \Gamma_n)/2$ on the complex plane of the photoelectron energy ε. A small perturbation of these complex energies is shown to be caused by the reverse BIC process.

We found the NEET cross section in the form of the integral (49), which depends on the ionization cross section $\sigma_{\mathrm{ion}}(\varepsilon)$. To understand the main features of the NEET, we first neglect the dependence of the ionization cross section $\sigma_{\mathrm{ion}}(\varepsilon)$ on the energy of the photoelectrons by replacing it with the constant value $\sigma_{\mathrm{ion}}^{(at)}$. Such an approximation immediately allowed us to get the analytical solutions (51)–(54) for the cross section $\sigma_{\mathrm{NEET}}(\varepsilon)$ in the form of the product of the NEET probability P_{NEET} far from the edge, $\sigma_{\mathrm{ion}}^{(at)}$ and the edge factor $F_{\mathrm{NEET}}(E)$, which describes the behavior of the NEET in the vicinity of the edge. The function $F_{\mathrm{NEET}}(E)$ consists of three terms, born by two resonant terms of the Green's matrix (54). The $f_+(E)$ and $f_-(E)$ represent separate contributions of the resonant levels, which correspond to the poles of the Green's matrix in the points μ_+ and μ_-, whereas, $f_\pm(E)$ results from their interference. The functions $f_+(E)$ and $f_-(E)$ describe two NEET edges at the energies $\Delta E \approx 0$ and $\Delta E \approx \delta$, respectively, with the widths Γ_i and Γ_f. The interference term becomes significant for the overlapping resonances when the mismatch of the nuclear and atomic transitions δ is smaller than the atomic widths Γ_i, Γ_f or is of the same order of magnitude. Note also that the term f_- is higher than f_+ by the factor $\sim\Gamma_f/\Gamma_i$. In the case of ^{197}Au with $\delta \sim \Gamma_i$ and $\Gamma_f/\Gamma_i \sim 0.3$, the first weak upswing at $\Delta E \approx 0$ is smashed out, whereas, in Fig. 1, the second at $\Delta E \approx \delta$ is seen absolutely clearly in correspondence with the observations [2]. In addition, it is worth noting that the main rise in the NEET curve for ^{189}Os takes place far below the photoabsorption edge as $\Delta E \approx \delta = -1287$ eV (see also Ref. [14]).

Kishimoto *et al.* [2] registered the NEET edge at the energy of x-ray photons $\Delta E_{\mathrm{NEET}} = 40 \pm 2$ eV. This point corresponds to $F_{\mathrm{NEET}} = 0.5$. It was believed [2] that there is an exact equality $\Delta E_{\mathrm{NEET}} = \delta$, which follows from the energy conservation law $\Delta E = \delta + \varepsilon$ if $\varepsilon = 0$. It is so if we do not consider the final phases of the NEET process. But the real energy conservation law $E_b = E_a$ for the whole NEET process, if the energy of the emitted γ quantum E_γ is replaced by its resonant value $E_\gamma^{(0)}$, is written as

$$\varepsilon = \Delta E - \delta - (E' - E'_0). \tag{73}$$

Since the distribution of the emitted x-ray photons over the energies E' is on the order of Γ_f, from the condition $\varepsilon = 0$, it only follows the approximate equality $\Delta E_{\text{NEET}} \approx \delta$. In addition, we see that the NEET channel in ^{178}Au becomes open at $\Delta E \approx 0$, which is owed to the equality (73). Here, the process is weak because it proceeds at the wing of the emission line of x-ray photons ω', emitted during the filling of the f hole. Note also that, if we only retained the leading term $f_-(E)$ of the edge factor, we would get the NEET edge exactly in the point $\Delta E = \delta$. But small terms $f_+(E)$ and $f_\pm(E)$ lead to any shift in the edge from this position. Really, our fitting of the NEET edge for ^{197}Au, presented in Fig. 1, shows that $F_{\text{NEET}} = 0.5$ in the point $\Delta E = 40 \pm 2$ eV when $\delta = 45 \pm 2$ eV.

By taking the table [28] energy $W_0^{(n)} = 77.351$ keV of the first excited level of ^{197}Au and by comparing it with the energy of the atomic transition in gold $W_0^{(a)} = 77.300$ keV [29], one has their mismatch of $\delta = 51$ eV. Recently, for ^{197}Au, a new value of $W_0^{(n)} = 77.339 \pm 0.003$ keV was reported [30], which lowers the mismatch up to $\delta = 39 \pm 3$ eV. So, we see that the calculated result for the mismatch δ lies between these experimental data. However, we get additional lowering of δ when an account is taken of the photoelectron rescattering inside the crystal, determined by the operator $\hat{T}_{\text{i}}$. This operator provides well-known EXAFS oscillations of the ionization cross section $\sigma_{\text{ion}}(\varepsilon)$, given in Eq. (68). Our numerical calculations of the NEET cross section (49) with (68) reveal that $\sigma_{\text{NEET}}(E)/\sigma_{\text{NEET}}(\infty) = 0.5$ if $\delta = 36$ eV, which agrees well with the data of Kirischuk *et al.* [30]. The fine structure of the NEET curve, drawn in Fig. 2, qualitatively reproduces the results of Kishimoto *et al.* [2].

The width of the NEET edge Γ_{NEET} appears to be much less than that of the K-absorption edge Γ_{abs} because $\Gamma_K \gg \Gamma_{M_1}$. For ^{197}Au by means of Eqs. (67), we found $\Gamma_{\text{NEET}} = 17.8$ and $\Gamma_{\text{abs}} = 55.5$ eV, which correlates well with the experimental data $\Gamma_{\text{NEET}} = 14 \pm 9$, $\Gamma_{\text{abs}} = 58 \pm 3$ eV [2].

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