Nonrelativistic quantum theory of the contact inelastic scattering of an x-ray photon by an atom

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The nonrelativistic analytical structure of the doubly differential cross section of the *contact* inelastic scattering of an x-ray photon by a free atom is determined by means of the irreducible tensor operator theory outside the frame of the impulse approximation. For the neon atom in the vicinity of the 1s shell ionization threshold our theory predicts the existence of the distinct *fine* structure of the cross section caused by transitions of the atomic core electrons into the excited *discrete* spectrum states. The results of our calculations with inclusion of the effects of radial relaxation, inelastic scattering through the intermediate states, and elastic Rayleigh scattering, are predictions, while at the 22 keV incident photons they compare well with the synchrotron experiment by Jung *et al.* [Phys. Rev. Lett. **81**, 1596 (1998)].

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I. INTRODUCTION

Theoretical and experimental studies of the scattering of an x-ray photon by a many-electron system provide fundamental information on the scatterer [1], particularly on the nature and role of the many-particle effects, and their quantum interference [2]. Numerous experimental (see, e.g., the works by Laukkanen *et al.* [3] and Pašič and Ilakovac [4]) and theoretical (see, e.g., the works by Bergstrom *et al.* [5] and Costescu and Spanulescu [6]) studies of this process have been performed for the incident photon energies *far above* the ionization thresholds of atomic *inner* shells. In this case, together with the possible return of an atom to its ground state (elastic Rayleigh scattering), the final scattering state turns out to be "atomic residue+electron (one or several) in the *continuous* spectrum" (inelastic nonresonant Compton scattering). The transitions (with very small intensity) into the *discrete* states are detected experimentally (Agarwal [7]), but practically has not been studied theoretically. In the nonrelativistic approximation, the amplitude of *such* Compton scattering is determined by the matrix element of the *contact* part (in the Coulombic calibration for the electromagnetic field) of the operator of the photon–atomic-electron interaction,

$$\hat{Q} = \frac{1}{2m_e}(e/c)^2\sum_{i=1}^{N}(\mathbf{A}_i\cdot\mathbf{A}_i), \tag{1}$$

where c is the speed of light in the vacuum, e is electron charge, m_e is electron mass, N is the number of atomic electrons, $\mathbf{A}_i\equiv\mathbf{A}(\mathbf{r}_i,0)$ is the electromagnetic field operator (in secondary quantization representation) at the time $t=0$, and $\mathbf{r}_i$ is the position vector of the ith atomic electron.

In this work, on the example of the neon atom (Ne; ground-state configuration $[0]=1s^2 2s^2 2p^6(^1S_0)$) we present the results of a theoretical investigation of doubly differential cross section of the *contact* inelastic scattering of an x-ray photon by a *free* atom *near* the 1s ionization threshold. In our recent paper [8] the analytical structure of the operator's (1) matrix element for the atom with *any* ground-state term has been obtained by the irreducible-tensor-operator theory methods. This structure allows one to consider a wide hierarchy of the *many-particle effects*, and the transition into both continuous and discrete (inelastic resonant Landsberg-Mandelstam-Raman scattering [9,10]) spectrum states. In this work we specify the results of Ref. [8] both in the vicinity of the 1s shell ionization threshold and in the far-above-the-threshold region of nonresonant Compton scattering.

Experimental and theoretical studies of the role of the many-particle effects and the transition into both continuous and discrete spectrum states in the process of *contact* inelastic scattering of the x-ray photon in the energy regions of inner-shell ionization thresholds of *free* atoms is in great demand in modern physics. Particularly, they are important in the context of creation (and their applications) of the x-ray free electron laser (see, e.g., the works by Kornberg *et al.* [11] and Plönjes *et al.* [12]) and of a laboratory-plasma x-ray laser generating with the participation of the deep 1s shell in lighter (charge number $Z\leq 20$) atoms (see, e.g., the reviews by Daido [13] and Kapteyn *et al.* [14]), and in laser thermonuclear fusion [generation and control of directed-to-target $K\alpha$ radiation (see, e.g., the works by Lindl *et al.* [15] and McDonald *et al.* [16])].

II. THEORY

Consider the process of the contact inelastic scattering of the linearly polarized x-ray photon by the n_1l_1 shell of an atom with the $^1S_0$ ground-state term (the closed shells are omitted in configurations notations):

$$\hbar\omega_1 + [0] \rightarrow n_1 l_1^{4l_1+1} n_2 l_2 (^1L_J) + \hbar\omega_2,$$

$$n_1 l_1 \leq f, \quad n_2 l_2 > f, \tag{2}$$

where $\hbar\omega_1$ ($\hbar\omega_2$) is the energy of the incident (scattered) photon, $\omega_{1,2}$ is the photon cyclic frequency, $\hbar$ is the Planck constant, f is the Fermi level (i.e., the set of the quantum numbers of the atomic valence shell), in the LS coupling scheme, ^1L_J is the resulting term of the open n_1l_1 and n_2l_2 shells, and it is taken into account that in the process (2) the

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total spin of the atomic system is conserved ($\Delta S=0$, $J=L$).

Then the general expression for the total doubly differential cross section of the processes (2) derived in Ref. [8] reduces to the form (in atomic units, $e=\hbar=m_e=1$)

$$\frac{d^2\sigma_{\perp}}{d\omega_2 d\Omega} = r_0^2\beta \sum_{n_1l_1\leq f} \underset{n_2l_2>f}{S} A_{12}G(\omega_{12},\Delta_{12}), \tag{3}$$

$$A_{12} = \sum_{t=0}^{\infty} (4t+2)[C_{12}^{(t)}R_t(n_1l_1,n_2l_2)]^2, \tag{4}$$

$$C_{12}^{(t)} = \sqrt{(2l_1+1)(2l_2+1)}\begin{pmatrix} l_1 & t & l_2 \\ 0 & 0 & 0 \end{pmatrix}, \tag{5}$$

$$R_t(n_1l_1,n_2l_2) = \int_0^{\infty} P_{n_1l_1}(r)j_t(qr)P_{n_2l_2}(r)dr, \tag{6}$$

$$G(x,y) = \frac{1}{\gamma_b\sqrt{2\pi}}\exp\left[-\left(\frac{x-y}{\gamma_b\sqrt{2}}\right)^2\right], \tag{7}$$

$$q = |\mathbf{k}_1 - \mathbf{k}_2| = \frac{\omega_1}{c}\sqrt{1+\beta^2-2\beta\cos\theta}. \tag{8}$$

In Eqs. (3)–(8), r_0 is the classical electron radius, Ω is the solid angle of the scattered photon escape, P_{nl} is the radial part of the nl electron wave function, $\gamma_b=\Gamma_{beam}/2\sqrt{2\ln 2}$, Γ_{beam} is the full width at half maximum of the instrumental Gaussian function G, $\Delta_{12}=E(n_1l_1^{4l_1+1}n_2l_2)-E(0)$, E are the total Hartree-Fock energies of respective configurations, $\beta=\omega_2/\omega_1$, $\omega_{12}=\omega_1-\omega_2$, t is the multiplicity of the contact transition, $|l_1-l_2|\leq t\leq l_1+l_2$, j_t is the spherical first-type Bessel function of the order t (Poisson integral representation [17]),

$$j_t(x) = \frac{x^t}{2^{t+1}t!}\int_{-1}^{+1}(1-z^2)^t\cos(zx)dz,$$

q is the module of the scattering vector (i.e., the vector of the momentum transferred to the atom), $\mathbf{k}_1$ ($\mathbf{k}_2$) is the wave vector of the incident (scattered) photon, and θ is the scattering angle (i.e., the angle between the vectors $\mathbf{k}_1$ and $\mathbf{k}_2$). The $\perp$ symbol in Eq. (3) means that in the suggested experimental scheme the polarization vector of both incident ($\mathbf{e}_1$) and scattered ($\mathbf{e}_2$) photons are perpendicular to the scattering plane, $(\mathbf{e}_1\cdot\mathbf{e}_2)^2=1$. The scattering plane is the plane containing the wave vectors of incident and scattered photons. The symbol S means summation (integration) over the final excited one-particle contact scattering states of the discrete (continuous) spectra.

In this work we do not consider the multiple excitation and/or ionization of the atom ground state and the effects of the configurations mixing in the scattering states. Further on, we only include, by the methods of theory of nonorthogonal orbitals (Löwdin [18], Jucys *et al.* [19]), the many-particle effect of *radial relaxation* of the scattering final-state electron wave functions in the Hartree-Fock field of the core n_1l_1 vacancy. In this case, Eq. (3) for the Ne atom is modified as follows:

(1) $ns\to ml$ transition ($n=1,2$)

$$\frac{d^2\sigma_{\perp}^{(ns)}}{d\omega_2 d\Omega} = 2r_0^2\beta N_{ns}^2\sum_{l=0}^{\infty}\underset{ml>f}{S} M_{nm}^{(l)}, \tag{9}$$

$$M_{nm}^{(l)} = (2l+1)R_l^2(ns,ml)G(\omega_{12},\Delta_{nsml}), \tag{10}$$

where $R_l\to B_l$ for $l=0$, 1.

(2) $2p\to ml$ transition,

$$\frac{d^2\sigma_{\perp}^{(2p)}}{d\omega_2 d\Omega} = 6r_0^2\beta N_{2p}^2\sum_{l=0}^{\infty}\underset{ml>f}{S} L_m^{(l)}, \tag{11}$$

$$L_m^{(l)} = [lR_{l-1}^2(2p,ml)+(l+1)R_{l+1}^2(2p,ml)]G(\omega_{12},\Delta_{2pml}), \tag{12}$$

where $R_1(2p,ms)\to B_1(2p,ms)$ and $R_{0,2}(2p,mp)\to B_{0,2}(2p,mp)$.

Here for ml discrete spectrum states $l\leq m-1$ and, for example, the terms N_{ns} and $B_l(ns,ml)$ in Eqs. (9) and (10) for $n=1$ and $l=1$ has the form

$$N_{1s} = \langle 1s_0|1s_+\rangle\langle 2s_0|2s_+\rangle^2\langle 2p_0|2p_+\rangle^6, \tag{13}$$

$$B_1(1s,mp) = R_1(1s_0,mp_+) - R_1(1s_0,2p_+)\frac{\langle 2p_0|mp_+\rangle}{\langle 2p_0|2p_+\rangle}, \tag{14}$$

$$\langle 2p_0|mp_+\rangle = \int_0^{\infty} P_{2p_0}(r)P_{mp_+}(r)dr. \tag{15}$$

In Eqs. (13)–(15), the radial parts of the wave functions of the $1s_0$, $2s_0$, $2p_0$ electrons are obtained by the solution of nonlinear integrodifferential self-consistent field Hartree-Fock system of equations for the atom's ground-state configuration, while the radial parts of the wave functions of the $1s_+$, $2s_+$, $2p_+$, mp_+ electrons are optimized in the configuration $1smp$ of the scattering final state (in the field of the $1s$ vacancy). In the case where the radial relaxation effect is ignored, $N_{1s}=1$ in Eq. (13), and in Eq. (14) the second term is zero due to the orthogonality condition $\langle 2p_0|mp_0\rangle=0$ at $m>f$. Here the radial part of the excited mp_0 electron wave function is optimized in the Hartree-Fock field of the atomic residue $1s^{-1}$ built with the ground-state wave functions.

In contrast to the widely used literature of *impulse approximation* (see, e.g., the review by Pratt [1]), Eqs. (4), (10), and (12) include both the transitions into the *discrete* scattering states and the fact that the scattering final-state wave functions of the *continuous* spectrum of *fixed l* symmetry are *not* the plane waves but are obtained by solving the Hartree-Fock equations with the Coulomb asymptote behavior at $r\to\infty$. The structure of doubly differential cross section from Eqs. (3), (9), and (11) may be modified to include multiple excitation and/or ionization of the atom ground state *and* a wide hierarchy of the many-particle effects *via* constructing the initial and final states of scattering in the frame of con-

TABLE I. Spectral characteristics of the most distinct $1s \to ml$ resonances of doubly differential cross section of the contact inelastic scattering of the linearly polarized (perpendicularly to the scattering plane) x-ray photon ($\hbar\omega_1 = 1275$ eV) by the Ne atom [see Eq. (9) and Fig. 2] calculated (*a*) including, and (*b*) disregarding radial relaxation effects. Here $\sigma_\perp \equiv d^2\sigma_\perp / d\hbar\omega_2 d\Omega$, $r_0^2 = 7.941 \times 10^{-26}$ cm^2, $\theta = 90°$, $\Gamma_{beam} = 0.23$ eV, and $[n] \equiv 10^{-n}$.

		$\hbar\omega_2$ (eV)		$\sigma_\perp$ (r_0^2 eV^{-1} sr^{-1})	
l	*m*	*a*	*b*	*a*	*b*
s	3	411.40	412.41	1.48 [−5]	5.98 [−8]
	4	408.47	408.61	0.35 [−5]	1.17 [−8]
	5	407.59	407.65	0.14 [−5]	0.41 [−8]
p	3	409.58	409.82	2.30 [−5]	4.80 [−5]
	4	407.97	408.20	0.72 [−5]	2.09 [−5]
	5	407.38	407.48	0.31 [−5]	0.80 [−5]

figurations mixing (see, e.g., the works by Brown [20], Surič [21], and Hopersky *et al.* [2]).

III. RESULTS AND DISCUSSION

Calculated cross section from Eq. (9) for $n=1$ are presented in Table I and Figs. 1 and 2. Here we did not include the final $2sml$ and $2p^5ml$ states of scattering since their contribution to the cross section at *given* ω_1 and ω_2 turned out to be about 30 times less than that from the $1sml$ scattering states. The Gaussian function width is taken to be $\Gamma_{beam} = \Gamma_{1s} = 0.23$ eV (from Gelius [22]), where Γ_{1s} is the natural *K*-level width in the Ne atom. In other words, we assume that

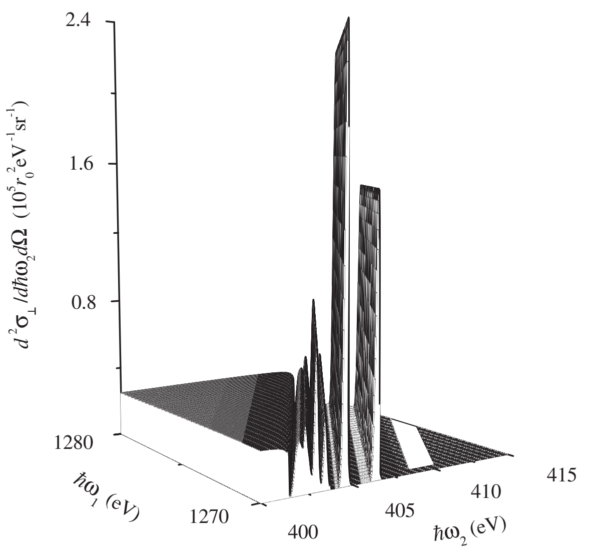

FIG. 1. Doubly differential cross section of the contact inelastic scattering of the linearly polarized (perpendicularly to the scattering plane) x-ray photon by the Ne atom (including the effects of radial relaxation) at $\hbar\omega_1 \sim I_{1s} + 0.4$ keV. Here $I_{1s} = 868.40$ eV is the $1s$ shell threshold ionization energy (our nonrelativistic calculation), $\theta = 90°$, $\Gamma_{beam} = 0.23$ eV.

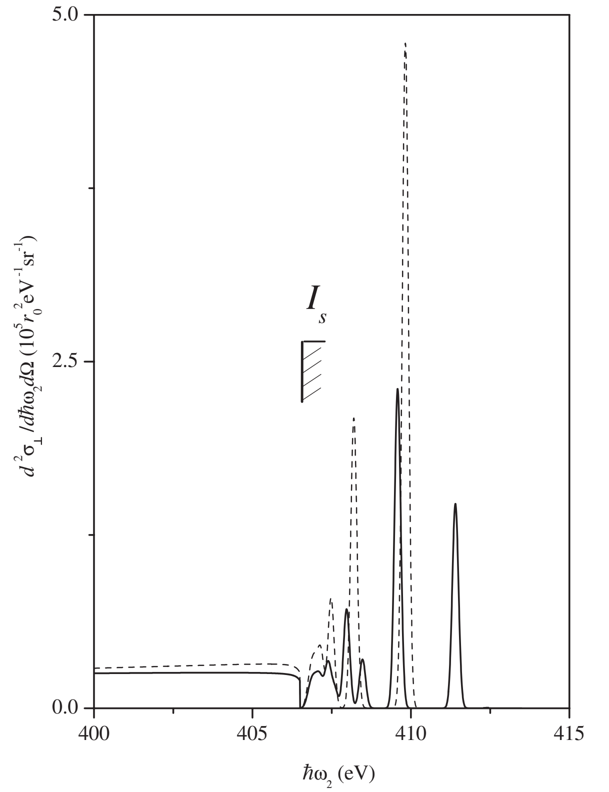

FIG. 2. See Fig. 1 at $\hbar\omega_1 = 1275$ eV. Dashed line, the calculation disregarding the effects of the radial relaxation. $I_s = \hbar\omega_1 - I_{1s}$. The resonances spectral characteristics are presented in Table I.

the experimental energy spectral resolution for the incident and scattered photons are the same. When calculating the sums over the *ml* discrete spectrum excited states, and over the multiplicities *l*, we included the $1s \to ml$ transitions with $m \leq 10$ and $l \leq 5$. The inclusion of the complete set of the *ml* discrete spectrum scattering states ($m = 3$ to ∞, and $l = 0$ to ∞), and thus eliminating the physically meaningless minimum in theoretical scattering cross section near $\omega_2 = I_s$ (see Fig. 2) is the subject of a separate study.

The calculation by the methods of Refs. [8,23,24] showed that the overall contribution from the inelastic resonant Compton $K\alpha_{1,2}$ emission structure of doubly differential scattering cross section (the channel of inelastic scattering through the formation of the intermediate $1s\varepsilon p$ state of continuous spectrum [8], $\omega_1 + [0] \to 1s\varepsilon p \to 2p^5\varepsilon p + \omega_2$), and elastic Rayleigh scattering (the channel of elastic scattering [23,24], $\omega_1 + [0] \to [0] + \omega_1$) for the Ne atom at $\omega_1 = 1275$ eV and $\omega_2 = 400$ to 415 eV was not greater than 10^{-16} (r_0^2 eV^{-1} sr^{-1}). Therefore, for the incident and scattered photon energy ranges studied, the theory of this work predicts the appearance of the distinct *fine* structure of the *total* doubly differential scattering cross section due practically exclusively by the *contact* type of inelastic scattering.

In order to verify the accuracy of the results presented in Table I and in Figs. 1 and 2 we calculated the cross sections from Eqs. (9) and (11) and the cross section of the Rayleigh scattering [23,24] in the case of the unpolarized [i.e., $(\mathbf{e}_1 \cdot \mathbf{e}_2)^2 \to (1 + \cos^2\theta)/2$] x-ray photon at $\omega_1 = 22$ keV by the Ne atom at an angle of $\theta = 90°$ for the scattered photon ener-

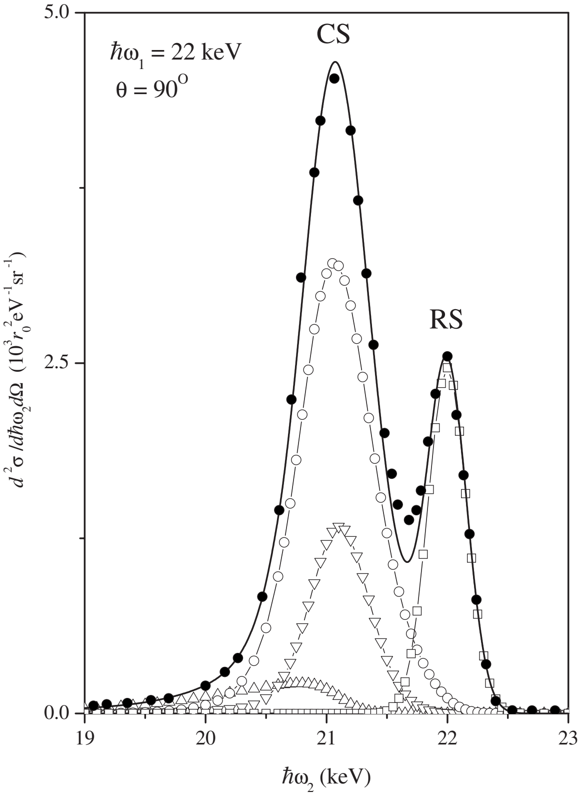

FIG. 3. Doubly differential cross section of scattering of the unpolarized x-ray photon by the Ne atom at $\hbar\omega_1 \gg I_{1s}$. Theory of this work: open circles and up and down triangles, the contributions of the $2p \to \varepsilon l$, $2s \to \varepsilon l$, and $1s \to \varepsilon l$ transitions, respectively; squares, contribution from the Rayleigh scattering (Hopersky *et al.* [23,24]); solid line, total cross section. Solid circles, synchrotron experiment (in arbitrary units) by Jung *et al.* [25]. Γ_{beam}=375 eV (from Ref. [25]). CS denotes Compton scattering and RS Rayleigh scattering.

gies ω_2=19 to 23 keV. When calculating the cross sections from Eqs. (9) and (11) we included only the transitions in the εl states of the *continuous* spectrum with l=0 to 25. The calculation showed that at the Compton's profiles' maxima of respective symmetries, the principal contributions to the $1s \to \varepsilon l$ transition are given by the p-symmetry states (72%), whereas the contributions to the $2s \to \varepsilon l$ (57%) and $2p \to \varepsilon l$ (62%) transitions are mostly from the d, f, g, and h symmetry states. Inclusion of the higher-l ($l > 25$) harmonics change the results by not less than 0.1%.

This calculation's results are compared in Fig. 3 with the *respective* synchrotron experiment by Jung *et al.* [25], and with the calculation by Biggs *et al.* [26] within the *nonrelativistic* impulse approximation (Eisenberger and Platzmann [27], Kane [28]). Experimental Compton and Rayleigh profiles are obtained in arbitrary units for intensity. Because of that, we "tied" their results to the theoretical values of the cross section from Eqs. (9) and (11) at the maximum of the doubly differential cross section of Rayleigh scattering at ω_2=22 keV. One can see good agreement of this work's theory with the experiment. The results of the impulse approximation [26] (see also the works by Eisenberger [29], Clementi and Roetti [30], Lahmam-Bennani *et al.* [31], and Jaiswal and Shukla [32] for the Ne atom) are just ~1% higher than ours at the Compton's profile maximum, and they are not shown in Fig. 3.

Note that the transition from nonrelativistic impulse approximation to Eqs. (9) and (11) leads to the *visible redistribution* of the *partial* contributions of the atomic shells to the total Compton profile. Indeed, in the nonrelativistic impulse approximation the partial contributions is equal (from Biggs *et al.* [26]) 6.59% ($1s$), 33.19% ($2s$), and 60.22% ($2p$), while in our theory we have 2.78% ($1s$), 28.24% ($2s$), and 68.98% ($2p$) (see Fig. 3). Note also that according to fundamental work by Eisenberger and Platzmann [27], the applicability criterion for the nonrelativistic impulse approximation to the theoretical description of the probability of the photon–atom contact Compton scattering is the inequality

$$\eta = qa_0/Z \gg 1, \tag{16}$$

where a_0 is the Bohr radius. However, for the neon atom at a scattering angle of θ=90° and the energies (at the Compton-profile maxima) ω_1=22 keV and ω_2=21 keV of the incident and scattering photons, respectively, η=0.815, which does not satisfy the inequality (16). Nevertheless, the calculation within the nonrelativistic impulse approximation reproduces well the results of the experiment by Jung *et al.* [25]. In this work we do not analyze the cause of this phenomenon. Note that a similar phenomenon was observed in recent experiments reported, for example, in works by Laukkanen *et al.* [3] ($\eta \cong 0.74$ for the contact Compton scattering of the 60 keV photon by the copper atom at an angle of θ=90°) and Pašič and Ilakovac [4] ($\eta \cong 1.27$ for the contact Compton scattering of the 86.54 keV photon by the germanium atom at an angle of θ=180°).

IV. CONCLUSION

To conclude, we suggest the nonrelativistic version of the many-particle quantum theory of the *contact* inelastic scattering of an x-ray photon by a *free* atom *without* any limitations (see, e.g., the works by Eisenberger and Platzmann [27], Williams [33], Cooper [34], Holm and Ribberfors [35], and Karazija [36], and the recent work by Pratt *et al.* [37]) of the impulse approximation. We predict the existence of resonant Landsberg-Mandelstam-Raman structure of a doubly differential cross section of the contact inelastic scattering near the $1s$ shell ionization threshold. QED provides the following *interpretation* of this structure. Theory developed in this work indicates that the presence of a QED *vacuum* is manifested *even* in the *x-ray* range of the photon scattered by a light atom. Indeed, as shown by Akhiezer and Berestetskii [38], the contact-scattering amplitude expressed in terms of the $\hat{Q}$ operator from Eq. (1) is a nonrelativistic limit of the relativistic scattering amplitude through intermediate single-electron states with *negative* frequencies ("Dirac sea") [39,40]).

One may suppose that the realization of the project of the creation of an x-ray free electron laser with tunable wavelengths from 60 to 1.0 Å (incident photon energy from 200 eV to 12.4 keV) will provide the opportunity for further experimental investigation predicted in this work of extended-in-space (see Fig. 1) *resonant* Landsberg-Mandelstam-Raman structure.

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