X-ray-photon scattering by an excited atom

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The existence of resonances in a double differential cross section of anomalous inelastic scattering of an x-ray photon by an excited atom at the attosecond time scale of the photon-electron contact interaction is theoretically predicted. These resonances realize unusual types of the excited atom vacancy decay channels. The subject of investigation is the atom of Be, excited to a discrete spectrum state via the $1s \to np, n \geqslant 2$ channel. Experimental realization of the theoretically predicted quantum effect may provide a new method for increasing the energy of photons generated by an x-ray free electron laser.

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I. INTRODUCTION

The x-ray free electron laser (XFEL) [1] and the high-order-harmonic-generation technique [2] open up new possibilities for the study of fundamental processes of the microcosm. In recent years there has been significant progress in addressing one of the underlying problems: obtaining a photon pulse of high energy ($\hbar\omega$, where $\hbar$ is Planck's constant and ω is the angular frequency of the photon) and of short duration (τ). In the context of this paper, we note the results in [3] ($\hbar\omega = 10$ keV, $\tau = 50$ as; 1 as $= 10^{-3}$ fs $= 10^{-18}$ s) and [4] ($\hbar\omega = 10$ keV, $\tau = 0.7$ as). These developments stimulate studies of both the structure and the evolution of many-electron systems at atomic spatial resolution and new physical aspects of the photon-atom interaction in the femtosecond [5] (photon absorption by neon) and attosecond [6] (photon absorption by helium, argon, krypton, and xenon) regimes of photon pulse duration.

In this work, we investigate the scattering of a photon by an excited atom at the attosecond time scale [7] of the photon-electron interaction. In this case, the lifetime (τ_{1s}) of even the $1s$ deep vacancy of the excited atom is significantly longer than the duration of the photon pulse in [2–4]. For example, in neon, the measured [8] lifetime of the $1\ s^{-1}\ 3p$ state is $\tau_{1s} = 2.74$ fs $\gg 50$ as. As a result, channels of photon scattering by the excited atom appear without going through the known processes of radiative and Auger decay of the vacancy. Such scattering channels can also be physically interpreted as (not previously studied) vacancy decay channels through the absorption of a photon incident on the excited atom [see Figs. 2(b) and 2(c)]. Thus, at the attosecond time scale one finds quantum effects that are not observed when $\tau_{1s} \leqslant \tau$. We predict such a quantum effect with an example study of the process of contact (nonlinear operator with respect to the electromagnetic field) inelastic [$\omega_2 \neq \omega_1$; ω_1 (ω_2), angular frequency of the incident (scattered) photon] and elastic ($\omega_2 = \omega_1$) Thomson scattering of an x-ray photon by an excited ($1s \to np$, $n \geqslant 2$; $\tau_{1s} = 41.16$ fs) atom of beryllium (Be: nuclear charge is $Z = 4$, the configurations of the ground state and the term are $[0] = 1s^2 2s^2[^1S_0]$). We show that in the double-differential cross section, along with the profile of Compton scattering and the Thomson line, resonances of anomalous inelastic scattering ($\omega_2 > \omega_1$) occur. We note that the investigated effect has a different [transition over operator $\widehat{V}$ from (1); see Figs. 2(b) and 2(c)] quantum nature, in contrast to the experimentally observed (by Kanter *et al.* [5]) resonant radiative transition effect in the core of a Ne atom excited by XFEL radiation.

II. THEORY

The mathematical formalism of this work is based on the first order of Dirac quantum-mechanical time-dependent perturbation theory for photon-electron contact interaction of duration τ. The "contactness" of the interaction corresponds to that representation of nonrelativistic quantum theory where, at one space-time point, there are four interacting states: two photons and, for example [see Fig. 2(b)], an electron and a vacancy. The operator of contact interaction, as a periodic perturbation, can be written as

$$\widehat{V} = \frac{1}{2m_e}\left(\frac{e}{c}\right)^2 \sum_{i=1}^{N} (\widehat{A}_i \cdot \widehat{A}_i), \tag{1}$$

where $\widehat{A}_i \equiv \widehat{A}(t;\vec{r}_i)$ is the electromagnetic field operator in the second-quantization representation, c is the speed of light in vacuum, e is the electron charge, m_e is the electron mass, N is the number of atomic electrons, t is the time of spreading of the field, and $\vec{r}_i$ is the position vector of the ith atomic electron. It is the nonlinearity of operator (1) over the electromagnetic field that leads to the existence of the quantum effect of anomalous inelastic scattering of a photon by an excited atom. The condition of validity of the corresponding formulas for differential cross sections of the studied processes has the form of [9] $\hbar E_{1s}^{-1} \ll \tau \ll \tau_{1s}$, where E_{1s} is the absolute value of the total energy of the atomic state with a $1s$ vacancy in the core. For Be, we obtain 2 as $\ll \tau \ll 41$ fs. The fulfillment of this double inequality gives the physical basis to interpret the investigated effect of anomalous inelastic scattering as an effect on the "attosecond time scale" over the duration of contact interaction (1) (see also the example of the Zn atom in Sec. IV).

Let us consider the process of contact inelastic scattering of a linearly polarized (perpendicular to the scattering plane; $\perp$) x-ray photon by a Be atom in a single excited state $C_n = 1\ s^{-1}\ np[^1P_1]$, $n \geqslant 2$. Here and below, the closed shells are not specified. The scattering plane passes through the wave

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vectors of the incident and scattered photons. We limit ourselves in the single-configuration Hartree-Fock approximation to considering the main channels of the anomalous ($\omega_2 > \omega_1$) inelastic scattering

$$\omega_1 + C_n \to [0] + \omega_2, \tag{2}$$

$$\omega_1 + C_n \to 2s^{-1}np + \omega_2 \tag{3}$$

and Compton ($\omega_2 < \omega_1$) scattering

$$\omega_1 + C_n \to n_1 l_1^{N_1 - 1} \varepsilon l + \omega_2, \tag{4}$$

where $n_1 l_1 = 1s$ ($N_1 = 1$), $2s$ ($N_1 = 2$), np ($N_1 = 1$), and ε is the energy of the continuum electron of l symmetry ($l = 0,1,\ldots,\infty$). Then, using the methods of nonrelativistic quantum theory of [10] for the double-differential cross sections of processes (2), (3), and (4), we obtain, respectively,

$$\sigma_\perp^{(1)} = 6 r_0^2 \beta R_1^2(1s,np) G_{sp}, \tag{5}$$

$$\sigma_\perp^{(2)} = 2 r_0^2 \beta R_0^2(1s,2s) G_{ss}, \tag{6}$$

$$\sigma_\perp^{(3)} = r_0^2 \beta \sum_{n_1 l_1 \leqslant np} \sum_{l=0}^{\infty} \int_0^\infty H_{l_1 l} G_{l_1 l} d\varepsilon. \tag{7}$$

Here the radial parts (in Dirac notation) of the probability amplitudes of the anomalous inelastic scattering (transitions in the discrete spectrum) are

$$R_1(1s,np) = \langle 1s | j_1(qr) | np \rangle, \tag{8}$$

$$R_0(1s,2s) = \langle 1s | j_0(qr) | 2s \rangle, \tag{9}$$

$$q = \frac{\omega_1}{c}(1 + \beta^2 - 2\beta\cos\theta)^{1/2}, \tag{10}$$

the Gauss-Laplace instrumental functions are

$$G_{sp(s)} = \alpha \exp\left[-\left(\frac{\Delta + I_{sp(s)}}{\gamma_b} \right)^2 \right], \tag{11}$$

$$G_{l_1 l} = \alpha \exp\left[-\left(\frac{\Delta - I_{l_1 l} - \varepsilon}{\gamma_b} \right)^2 \right], \tag{12}$$

and the multipole structures of the squared probability amplitudes of Compton scattering (transitions into continuum) are

$$H_{sl} = N_1 (2l+1) R_l^2(n_1 s, \varepsilon l), \tag{13}$$

$$H_{pl} = (l+1)\left[R_l^2(np, \varepsilon(l+1)) + R_{l+1}^2(np, \varepsilon l) \right]. \tag{14}$$

In (2)–(14) and below the atomic system of units is used ($e = \hbar = m_e = 1$), and the variables are designated as follows: $\sigma_\perp \equiv d^2\sigma_\perp/(d\omega_2 d\Omega)$, with Ω being the exit solid angle of the scattered photon; r_0 is the classical electron radius; $j_k(qr)$ is the kth-order spherical Bessel function of the first kind; $\beta = \omega_2/\omega_1$; $\Delta = \omega_1 - \omega_2$; θ is the scattering angle (the angle between the wave vectors of the incident and scattered photons); $\alpha = 1/(\gamma_b \sqrt{\pi})$; $\gamma_b = \Gamma_{\text{beam}}/(2\sqrt{\ln 2})$; Γ_{beam} is the full width at half-maximum of the G function; and I is the difference between the total Hartree-Fock energies of the initial (C_n) and final states of the atomic system in processes (2), (3), and (4).

The appearance of a vacancy in the atomic core is accompanied by effects of radial relaxation of one-electron wave functions. The time scale for electron-electron electromagnetic interaction is characterized by the quantity $l/c \approx 0.2$ as for the Bohr radius $l \approx 5.29 \times 10^{-11}$ m. Accounting for these effects leads to modifications of integrals in (8), (9), (13), and (14). For example, for the integral in (8) we have

$$R_1(1s,np) \to \varsigma\, [R_1(1s_0,np_+) - \eta\, R_1(2s_0,np_+)], \tag{15}$$

$$\varsigma = \langle 1s_0 | 1s_+ \rangle \langle 2s_0 | 2s_+ \rangle^2, \tag{16}$$

$$\eta = \langle 1s_0 | 2s_+ \rangle / \langle 2s_0 | 2s_+ \rangle. \tag{17}$$

Here, radial parts of the wave functions of the l_0 electrons are obtained by solving the nonlinear integral-differential self-consistent-field Hartree-Fock equations for the [0] configuration. Radial parts of the wave functions of the l_+ electrons are obtained by solving the Hartree-Fock equations for the C_n configuration (in the $1s$-vacancy field).

The calculation of the double-differential cross sections for elastic ($\omega_2 = \omega_1$) Thomson (T) x-ray photon scattering via the channel

$$\omega_1 + C_n \to C_n + \omega_1 \tag{18}$$

has been performed using

$$\sigma_\perp^T \equiv (d\sigma_\perp^T / d\Omega) G, \tag{19}$$

$$G = \alpha \exp\left[-\left(\frac{\Delta}{\gamma_b} \right)^2 \right]. \tag{20}$$

In (19), the single-differential scattering cross section is calculated in the form factor approximation (the scattering probability amplitude calculation does not take into account the Kramers-Heisenberg-Waller anomalous dispersion terms) [11]:

$$d\sigma_\perp^T / d\Omega = r_0^2 \left| \sum_{n_1 l_1 \leqslant np} N_1 \langle n_1 l_1 | j_0(\chi r) | n_1 l_1 \rangle \right|^2, \tag{21}$$

$$\chi = (2\omega_1/c)\sin(\theta/2). \tag{22}$$

III. RESULTS AND DISCUSSION

During the calculation of the absolute values and forms of the scattering cross sections (5), (6), (7), and (19), the following values of ω_1, Γ_{beam}, and θ are used. The energy of the incident photon is $\omega_1 = 1$ keV. This value is considerably higher than the energy of the ionization threshold of the $1s$ shell of the Be atom, $I_{1s} = 123.09$ eV (calculated in this paper), which makes it possible to use the form factor approximation for calculating the cross section (21). For the spectral resolution of the proposed experiment, $\Gamma_{\text{beam}} = 1$ eV is taken. This value is larger than the natural linewidth of the $1s$-vacancy decay of Be, $\Gamma_{1s} = 0.016$ eV ($\tau_{1s} = \hbar\Gamma_{1s}^{-1} = 41.16$ fs) (calculated in this work), and corresponds to the spectral resolution of modern experiments on inelastic x-ray photon scattering by an atom (Γ_{beam} from 1.00 [12] to 10^{-3} [13] eV). The scattering angle $\theta = 90^\circ$ is taken. Calculations of (13) and (14) take into account the multipolarity l from 0 to 25. Inclusion of higher

TABLE I. Energy (ω_2) and intensity ($\sigma_\perp^{(1,2)}$) [see (5) and (6)] of the resonances of the anomalous inelastic scattering of linearly polarized (perpendicular to the scattering plane; $\perp$) x-ray photons by excited atomic states C_n of Be. $\omega_1 = 1$ keV, $\theta = 90^\circ$, $\Gamma_{\text{beam}} = 1$ eV.

	ω_2 (eV)[a]		$\sigma_\perp^{(i)}$ ($10^4\ r_0^2\ \text{eV}^{-1}\ \text{sr}^{-1}$)	
n	$1s \to 2s$	$1s \to np$	$i = 2$	$i = 1$
2	1111.75	1114.37	5.38	45.00
3	1114.74	1120.91	7.86	5.91

[a]See (11), where $\omega_2 = \omega_1 + I_{sp(s)}$, $I_{sp} = E(C_n) - E(0)$, $I_{ss} = E(C_n) - E(2s^{-1}np)$, and $E(Q)$ is the full Hartree-Fock energy of state Q.

harmonics ($l > 25$) changes the value of the scattering cross section, (7), by no more than 0.1%. For $\omega_1 = 1$ keV and $\theta = 90^\circ$, the primary channels of Compton scattering, (4), were transitions to states of the continuous spectrum of $p(\sim 85\%)$, $d(\sim 10\%)$, and $f(\sim 5\%)$ symmetry. Energy thresholds for the breaking of the profiles of partial Compton scattering cross sections ($\omega_2^{\max} = \omega_1 - I_{n_1 l_1}$; $I_{n_1 l_1}$ is the $n_1 l_1$-shell ionization threshold energy of the configuration of the excited state C_n) are obtained: $\omega_2^{\max}(1s \to \varepsilon l) = 836.15$ eV ($n = 2$), 825.20 eV ($n = 3$), $\omega_2^{\max}(2s \to \varepsilon l) = 989.03$ eV ($n = 2$), 983.54 eV ($n = 3$), and $\omega_2^{\max}(np \to \varepsilon l) = 991.28$ eV ($n = 2$), 997.82 eV ($n = 3$).

The results of calculations are presented in Table I and Fig. 1. In Fig. 1, the total double-differential scattering cross section is shown:

$$\sigma_\perp \equiv \sum_{i=1}^{3} \sigma_\perp^{(i)} + \sigma_\perp^{T}. \tag{23}$$

Figure 1 demonstrates the main theoretical result: in the processes of scattering of an incident photon with energy ω_1 by an excited (e.g., by a laser additional to the XFEL, with photon energy $\omega_0 \sim I_{sp}$ or in the scheme of self-amplification through the absorption of one and scattering of another XFEL photon) atom of Be, scattered photons of energy $\omega_2 \sim \omega_1 + \omega_0$ [$\sigma_\perp^{(1)} + \sigma_\perp^{(2)}$ in (23)] are produced. At the same time, the increase in the principal quantum number of the np excited electron is accompanied by a decrease in the intensity of the scattering resonance due to channel (2) and a stabilization of the scattering resonance via channel (3). In fact, the transition from state C_2 to state C_3 is accompanied by an increase in the average np-shell radius ($r_{2p} = 2.14 \to r_{3p} = 8.59$ a.u.) and, as a result, a reduction in the value of overlap of wave functions of the $1s$ and np electrons in the integral $R_1(1s,np)$. Simultaneously, the average radius of $1s$ and $2s$ shells is stabilized [$r_{1s} = 0.377$ ($n = 2$) $\to$ 0.376 ($n = 3$) a.u.; $r_{2s} = 2.19$ ($n = 2$) $\to$ 1.98 ($n = 3$) a.u.], which leads to the stabilization of the value of the integral $R_0(1s,2s)$. Increasing r_{np} and stabilizing r_{1s}, r_{2s} with $C_2 \to C_3$ also leads to a reduction of the maximum of the Compton scattering cross section profile [$\sigma_\perp^{(3)}$ in (23)].

The dependence of the Bessel functions on ω_1, ω_2, and θ should lead to a corresponding dependence of the scattering cross sections (5), (6), (7), and (19). An increased complexity of structure in the region of the anomalous inelastic scattering resonances in Fig. 1 should also be expected when the effects

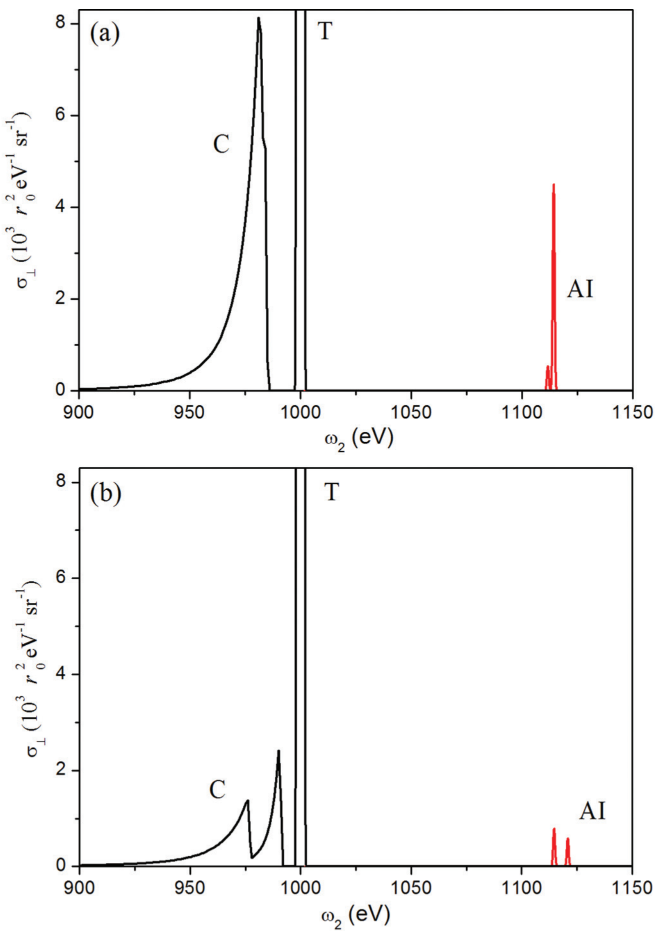

FIG. 1. (Color online) Double-differential scattering cross section of a linearly polarized (perpendicular to the scattering plane; $\perp$) x-ray photon by the excited $1s^{-1}\ np[^1P_1]$ state of Be at the attosecond time scale of the photon-electron contact interaction: $n = 2$ (a) and $n = 3$ (b). C, Compton ($\omega_2 < \omega_1$) scattering; T, Thomson ($\omega_2 = \omega_1$) scattering [$\max(10^{-3}\sigma_\perp^T) = 2.11$ (a), 2.04 (b) $r_0^2\ \text{eV}^{-1}\ \text{sr}^{-1}$]; AI, anomalous inelastic ($\omega_2 > \omega_1$) scattering. Contribution to the Compton scattering cross section of a deep $1s$ shell [$\sigma_\perp^{(3)} \sim 10^{-5}\ r_0^2\ \text{eV}^{-1}\ \text{sr}^{-1}$ for $n_1 l_1 = 1s$] is defined outside of the energy scale in Fig. 1 ($\omega_2 \leqslant 837$ eV). Spectral characteristics of the anomalous inelastic scattering resonances are listed in Table I. $\omega_1 = 1$ keV, $\theta = 90^\circ$, $\Gamma_{\text{beam}} = 1$ eV.

of multiple excitation and ionization, configuration mixing, and multiplet splitting in scattering channels (2) and (3) have been taken into account. With this, for example, expression (5) is converted to the triply differential anomalous inelastic scattering cross section,

$$d\sigma_\perp^{(1)}/d\omega_0 = \zeta_n \cdot \bar{\sigma}_\perp^{(1)}; \tag{24}$$

for $\bar{\sigma}_\perp^{(1)}$, see (5) with the substitution $I_{sp} \to \omega_0$. In the Dirac δ function approximation we have $\zeta_n \to \delta(\omega_0 - I_{sp})$, and after the integration over ω_0, one obtains (5). Here, the function ζ_n is equal to the spectral density of the excited states C_n that are created when a neutral atom absorbs a photon of energy ω_0, and the following requirement must be satisfied:

$$\int_{-\infty}^{\infty} \zeta_n d\omega_0 = 1. \tag{25}$$

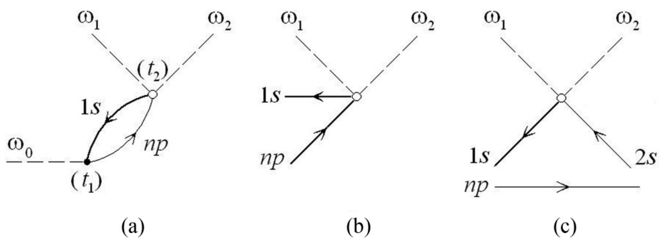

FIG. 2. Feynman diagrams for the probability amplitudes of the anomalous processes of inelastic scattering [$\omega_2 > \omega_1$; (b) see (2) and (c) see (3)] of an x-ray photon by an excited ($1s \to np$, $n \geqslant 2$) atom of Be at the attosecond time scale of the photon-electron contact interaction. The process of two-photon absorption by a neutral atom, (26), described in Fig. 2(a), is forbidden due to the Landau-Yang theorem. Left arrow, vacancy; right arrow, electron; filled circle, vertex of the interaction with the operator of radiative transition; open circle, vertex of the contact interaction with operator $j_k(qr)$. Time direction of the processes is from left to right ($t_1 < t_2$). In Fig. 2(c), the np electron is a "spectator" of the process.

Expression (24) contains the information about the dependence of the spectral characteristics of the anomalous inelastic scattering effect on the photon energy ω_0 that prepares state C_n. Study of these issues is the subject of future work.

Finally, we note the following. The anomalous inelastic scattering process, (2) [Fig. 2(b)] cannot be considered as a part of the process of absorption of two photons by a neutral atom with the formation (via a C_n virtual state) of a single photon in the final state [Fig. 2(a)]:

$$\omega_0 + W_1 \to W_{1n} \to W_2, \tag{26}$$

$$W_{1,2} \equiv \omega_{1,2} + [0], \quad W_{1n} \equiv \omega_1 + C_n. \tag{27}$$

In fact, process (26) is forbidden by the Landau-Yang theorem [14,15], since the spin of the final state is equal to 1 (but not $s = 0$ or 2). In this case, the Landau-Yang theorem in the representation of Feynman diagrams for the probability amplitude of multiphoton scattering reproduces Furry theorem [16], restated for an atom with a $^1S_0$ term of the ground state: the loop of virtual electrons and vacancies cannot have an odd number of external (real) photon lines. Thus, the process of scattering ($1s$-vacancy decay) via channel (2) is defined as an independent quantum effect and is implemented only at the attosecond time scale of the photon-electron contact interaction. Exactly this fact is reflected in the factorized analytical structure of expression (24). In this case, the potentially attainable attosecond photon pulse duration for such interaction can be achieved with an appropriate design of the experiment. The process of two-photon absorption of the type

$$\omega_0 + W_1 \to W_{1n} \to 2s^{-1}np + \omega_2 \tag{28}$$

is defined, but with probability amplitude lower by ~2 orders of magnitude (see the additional factor of 1/137) than the probability amplitude of the process of scattering ($1s$-vacancy decay) via channel (3) [Fig. 2(c)].

IV. CONCLUSIONS

We state the main result of this work as follows. The contact interaction of an x-ray photon with an excited atom at the attosecond time scale leads to the quantum effect of anomalous inelastic scattering resonances occurring in the double-differential scattering cross section. Certainly, the time delay between the photon pulse that creates the C_n state (i.e., t_1) and the photon pulse that scatters the C_n state (i.e., t_2) must satisfy the condition $t_2 - t_1 \ll \tau_{1s}$. The question of experimental realization of this condition is left open. We also leave open the problem of generating a coherent beam of scattered photons of energy $\omega_2 \sim \omega_1 + \omega_0$.

It is interesting to investigate this effect for deep shells of heavy elements of the periodic table with a strong discrete structure of the photoabsorption spectrum. For example, for atomic zinc ($\tau_{1s} \cong 0.39$ fs), experiments [17] reveal intense photoexcitation resonance $1s \to 4p$ with transition energy $\omega_0 \cong 9.66$ keV, much higher than the transition energy $I_{sp} \cong 114.37$ eV for Be atom (Table I; $n = 2$). In this case (0.02 as $\ll \tau \ll 0.39$ fs) we should expect a broad series of anomalous inelastic scattering resonances up to a resonance with energy $\omega_2 \sim \omega_1 + \omega_0$. Therefore, the inclusion of photon scattering by an excited atom at the attosecond time scale of the photon-electron contact interaction from (1) in an experiment on the generation of hard x rays may provide a means to a noticeable and controlled increase in the angular frequency of the incident XFEL photon.

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