Scattering of an intense laser beam by atomic systems

Mihai Dondera*

Department of Physics and Centre for Advanced Quantum Physics, University of Bucharest, MG-11, Bucharest-Măgurele 077125, Romania

(Received 29 November 2021; accepted 31 January 2022; published 15 February 2022)

A nonrelativistic quantum approach is adopted to describe the scattering of an intense laser beam from a simple atomic system. Two forms of the corresponding transition amplitude (TA) are derived, one involving the kinetic momentum operator of the electron and the other involving the force operator. The spectral and angular distribution of the scattered radiation is expressed in terms of TA by a relation recalling the classical one. The formalism is applied to the scattering of radiation by an electron (free or bound) with the absorption of two photons from the incident laser beam. Analytic and numeric results are presented for the distributions of the emitted photons in the case of the nonlinear Compton scattering from a hydrogenic atom in the ground state.

DOI: 10.1103/PhysRevA.105.023108

I. INTRODUCTION

According to classical theory an electron in *accelerated* motion emits radiation [1]. When the acceleration is caused by an external electromagnetic radiation beam, the emitted radiation is identified with the scattered radiation. The properties of the scattered light follow from the law of motion $\mathbf{r}(t)$ of the electron. Based on Eqs. (14.65) and (14.64) in Ref. [1], we express the energy of the radiation emitted in the solid angle $d\Omega$ around the observation direction $\mathbf{n}$, with the frequency in the spectral interval $d\omega$ and with polarization $\mathbf{s}_\lambda$ (orthogonal to $\mathbf{n}$), as follows:

$$d^2W_\lambda^{(\mathrm{cl})} = \frac{1}{(2\pi)^2}\,\frac{e_0^2}{c}\,\left|\mathcal{A}_{\mathrm{cl}}^{(\mathbf{n},\lambda)}(\omega)\right|^2 d\omega\, d\Omega, \tag{1}$$

with $e_0 \equiv e/\sqrt{4\pi\varepsilon_0}$, where e is the electron charge. The classical amplitude $\mathcal{A}_{\mathrm{cl}}^{(\mathbf{n},\lambda)}(\omega)$ is defined conveniently as the component along $\mathbf{s}_\lambda$ of the negative of the integral appearing in Eq. (14.64) of the quoted paper; see also Appendix A, Eq. (A1). The nonrelativistic approximation of this amplitude, taken in the first order of $\boldsymbol{\beta}_{\mathrm{cl}}(t) \equiv \mathbf{v}(t)/c = \dot{\mathbf{r}}(t)/c$, is given by

$$\mathcal{A}_{\mathrm{cl}}^{(\mathbf{n},\lambda)}(\omega) = \frac{1}{c}\int_{-\infty}^{\infty} dt\, e^{i\omega(t-\mathbf{n}\cdot\mathbf{r}(t)/c)} \times \mathbf{s}_\lambda^*\cdot[\mathbf{a}_{\mathrm{cl}} + (\mathbf{n}\cdot\boldsymbol{\beta}_{\mathrm{cl}})\,\mathbf{a}_{\mathrm{cl}} + (\mathbf{n}\cdot\mathbf{a}_{\mathrm{cl}})\,\boldsymbol{\beta}_{\mathrm{cl}}], \tag{2}$$

where $\mathbf{a}_{\mathrm{cl}} = c\dot{\boldsymbol{\beta}}_{\mathrm{cl}} = \dot{\mathbf{v}}$ is the electron acceleration. For the scattering problem one needs to know $\mathbf{r}(t)$ for the electron exposed to the laser beam (and, possibly, to another external force field), then to evaluate the quantities entering the above equation. The acceleration of the electron appears explicitly in Eq. (2) and the amplitude $\mathcal{A}_{\mathrm{cl}}$ vanishes with it.

In quantum theory, the scattering is described in terms of photons: the electron (bound or free initially) absorbs one or more photons from the laser field and *spontaneously* emits one photon of a frequency different from that of the laser photons—having much lower probabilities, the processes with the emission of more photons are disregarded in the following. At low intensity of the laser, the scattering is *linear*, it being dominated by the absorption of *one* laser photon by the electron, a process having a rate proportional to the laser intensity. At higher laser intensity the number of absorbed photons is not defined anymore and the dependence of the emitted radiation on laser intensity becomes nonlinear. Nevertheless, in the emitted radiation spectrum one can distinguish approximately the contributions corresponding to various fixed net numbers of absorbed photons. When the radiation scattering (linear or nonlinear) is accompanied by the scattering of the electron (free or bound initially), the process is of Compton type. Nonlinear processes of Raman or Rayleigh types are also possible for intense laser scattering by bound electrons.

Besides intensity, the frequency of the laser strongly influences the features of the scattering. For example, if the laser intensity overcomes the atomic intensity $I_0 \approx 3.51 \times 10^{16}\ \mathrm{W/cm^2}$ by a factor of the order 10^3, the electron dynamics is relativistic and the scattering is highly nonlinear for an incident frequency in the infrared range. For the description of the nonlinear Compton scattering (NLCS) in this regime a *nonperturbative* fully relativistic framework is required—see, for example, Refs. [2–8]; the electron is considered as being initially in a state of definite momentum with few exceptions, for example, Ref. [9] where the initial state of the electron is a wavepacket (continuous superposition of states with different momenta). We note here that the attribute "nonperturbative" is used in this paper solely with reference to the interaction of the electron with the laser modes (the laser field is usually described in classical theory, a treatment justified at high laser intensities). On the other hand, for a free electron laser in the keV range and having an intensity of the same order as above ($10^3\, I_0$), the scattering is dominantly linear, the processes with the absorption of more than one photon having relatively reduced rates. In these conditions the electron dynamics is essentially nonrelativistic and a perturbative approach is of interest in order to describe NLCS. A nonrelativistic (NR)

*mihai.dondera@fizica.unibuc.ro

treatment is also justified for the case of low (or intermediate) frequencies, for not very high intensities. The relativistic nonperturbative studies above cited make extensive use of the so-called Volkov solutions (exact solutions of the Dirac equation for an electron interacting with an intense laser pulse). An earlier NR example for the application of the Volkov solutions in the case of NLCS by free electrons is found in Ref. [10]. It is to be mentioned that NR Volkov solutions are frequently used in the description of two related strong field processes, above-threshold ionization (ATI) and high-order harmonic generation (HHG); see, for instance, Ref. [11] and references therein. Successful NR approaches of these processes, investigating the interaction of intense laser fields with bound electrons, are mainly based on the so-called strong-field approximation (SFA). In the simplest SFA one fully neglects the influence of the potential force (usually the Coulomb force electron-nucleus) on the process duration, the electron being treated as moving solely under the influence of the Lorentz force. It is remarkable that SFA, initially formulated in dipole approximation, can be improved in order to take into account magnetic and nondipole (MND) effects. This objective is attained by the construction of Volkov-type solutions appropriate for different field configurations—two significant recent examples are Refs. [12,13], aiming, respectively, to describe ATI for spatially structured laser fields (twisted fields, in particular) or to extend the description of HHG to the case of atoms exposed to a superposition of noncollinear beams (with the purpose to control the role of the magnetic field).

Apparently, the connection between the emission of radiation and the accelerated motion of the electron is lost when passing from classical to quantum theory. The primary aim of this paper is to show how the quantum description of the radiation scattering can be formulated in order to reestablish this connection. This is achieved in Sec. II, where we treat the laser field classically and make the operator associated to the force exerted on the electron appear explicitly in the formal description of the scattering. The angular and spectral distribution of the energy emitted by the electron during its interaction with the laser pulse is expressed with the help of a transition amplitude (TA), for which one first infers a formula, Eq. (10), involving the *kinetic momentum* operator of the electron in the laser field. Then, in tight analogy with the treatment of the radiation emission in classical electrodynamics, an alternative form of TA is deduced, Eq. (17), relying on the total *force* operator. We mention that for the derivation of Eqs. (10) and (17) the interaction electron laser is treated *nonperturbatively*. A part of the formalism presented in Sec. II, referring to the photon emission, is similar in particular to that used in the recent theoretical papers Refs. [14,15]. The approaches proposed in these papers for the study of the radiation scattering off bound electrons are based on the time dependent Schrödinger equation (TDSE), thus being inherently nonperturbative. In the present paper the formalism, also relying on TDSE, is further developed in order to remove its dependence on the quantization volume and to obtain closed analytic equations relating the transition amplitude to the states of the electron in the laser field.

References [14,15] are mainly motivated by the results presented in Ref. [16], an experimental paper reporting the observation of photons with energy around 18 keV, which is the double of the energy of photons of a free electron laser beam sent on a beryllium target. The results concerning the photon spectra in this energy range were interpreted in Ref. [16] as describing the "concerted" scattering of two photons absorbed from the laser beam, producing one higher-energy photon. A large redshift of the NLCS peak versus the peak for linear Compton scattering (LCS) of the second laser harmonics was observed and not understood up to now. The theoretical investigations in Refs. [14,15] seem to not confirm the experiment and lead to the conclusion that the atomic binding of the electron could not be the cause of the NLCS shift seen in Ref. [16]. Two other possible causes of the redshift are investigated in Ref. [15], mainly by two-dimensional simulations. These are the electron-electron Coulomb interaction and the linear scattering of a laser photon by a photoelectron (which absorbs another laser photon). It is found that their inclusion does not significantly alter the scattering profile for NLCS, in particular its position, and thus it is unlikely that these are causing the redshift.

Given the complexity of the analyzed process and of the implied TDSE calculations, independent approaches of it are highly desirable. In particular, an approach based on lowest-order perturbative theory (LOPT) should be a good choice for conditions like in the experiment in Refs. [16], where the measured NLCS signal has a *quadratic* dependence of the laser intensity. We remark that perturbative studies treating directly NLCS with the absorption of two photons are relatively few—two such examples are found in Refs. [17,18]. Reference [17] considers the nonlinear scattering by a He atom for a rather low incident photon energy (500 eV). The LOPT calculation of the transition amplitude is made manageable by an approximate treatment of a part of the terms corresponding to the Feynman diagrams of the process. The terms neglected in Ref. [17] were shown in Ref. [14] to be important at that photon energy and of decreasing significance at higher energies (in the keV range). Reference [18] studies the scattering by *free* electrons of defined momenta, with two photons absorbed from a low intensity incident field. The QED calculation of the angular photon distribution gives a result which, up to a factor accounting for correlations of photon pairs, agrees in the Thomson limit (where the photon momenta are negligible) with those given by the semiclassical [2] and classical theories [19].

As discussed in Ref. [14], for scattering by bound electrons in conditions similar to those of Ref. [16], there could exist good reasons to prefer the NLCS investigation by *nonperturbative* against *perturbative* calculations—the main argument resides in the increasing difficulties to obtain in these conditions *accurate* results for NLCS in perturbation theory. Nevertheless, we appreciate that in the actual stage it is also of interest to consider simplified models based on LOPT (embodying supplementary approximations) and to analyze the measure in which the NLCS features known at the present (from experiment or more accurate theoretical simulations) can be explained by these models. Going on this direction, the formalism from Sec. II is specialized in Secs. III and IV to the nonlinear Compton scattering in a perturbative regime. It is then applied in conditions close to those of Ref. [16]. Our treatment, describing NLCS perturbatively with respect to the interaction electron-laser beam, can be regarded as complementary to that used in Refs. [14,15]. In Sec. III we derive two

formulas, Eqs. (38) and (39), for the distribution fully characterizing, respectively, the scattering of a monochromatic plane wave by a free electron in a general state and by a bound electron in a stationary state—we note that supplementary approximations are adopted in order to make the calculations feasible or to simplify the form of the equations. These approximations are formulated relative to the matrix elements from the expressions of the transition amplitude, Eqs. (24) and (26).

In Sec. IV we consider NLCS from a hydrogenic atom in the ground state. For the distribution in frequency and emission direction of the scattered radiation we present two expressions, one in impulse approximation (IA) and the other based on exact descriptions of the initial and final states of the electron in the Coulomb potential. Their derivation implies the integration over the final momentum of the electron, performed as in the case of the usual Compton scattering—see, for example, Refs. [20,21]. Numerical results characterizing the spectral and angular distribution of the scattered radiation are presented for values of the parameters of the incident laser in the ranges of those used in the experiment [16], then analyzed in relation with existing experimental and theoretical results. The main features of the spectra presented in Ref. [16] are reproduced qualitatively, with the notable exception of the above-mentioned redshift. Finally, we refer to the (integrated) angular distribution of the scattered radiation. In its case, the IA results for scattering on bound electrons are compared with those corresponding to free electrons (the equations for the latter case are included in Appendix C) and with the results presented in Ref. [15].

II. THE AMPLITUDE OF THE RADIATION SCATTERING

For simplicity, as an atomic system we consider an electron moving in an attractive potential V. The Hamiltonian operator [22] of the system is $H_a \equiv \mathbf{P}^2/2m_e + V$, where $\mathbf{P}$ is the kinetic momentum operator and m_e the mass of the electron. We first allow the atomic system to interact solely with the laser beam, assumed to be of the type of a pulse introduced at a time moment t_0 and removed at t_1, thus having a duration $\tau = t_1 - t_0$. Working in the Coulomb gauge, we describe the laser field with the help of a vector potential $\mathbf{A}_L(\mathbf{r}, t)$, the corresponding electric and magnetic fields being given by the well-known relations $\mathbf{E} = -\partial\mathbf{A}_L/\partial t$ and $\mathbf{B} = \boldsymbol{\nabla} \times \mathbf{A}_L$. In these conditions the evolution of the atomic system on the time interval (t_0, t_1) is governed by the TDSE corresponding to the Hamiltonian operator $\mathcal{H}_{aL} \equiv \boldsymbol{\Pi}^2/2m_e + V$, where $\boldsymbol{\Pi} \equiv \mathbf{P} - e\mathbf{A}_\mathbf{L}$ is associated to the kinetic momentum $\mathbf{p}_{\rm kin}$ of the electron in the laser field. The state of the atomic system, evolving from an initial state $|\psi_0\rangle$ (arbitrary at this stage), is described by a state vector $|\psi_i(t)\rangle$ satisfying this TDSE and the initial condition $|\psi_i(t_0)\rangle = |\psi_0\rangle$. The range of the values for $\mathbf{p}_{\rm kin}$ which matter for the process we investigate is required to satisfy the condition $p_{\rm kin} \ll m_e c$, or $\beta \ll 1$, where $\boldsymbol{\beta} \equiv \mathbf{p}_{\rm kin}/m_e c$. The same symbol is used below to denote the operator associated to $\mathbf{p}_{\rm kin}/m_e c$:

$$\boldsymbol{\beta} = \frac{1}{m_e c}\,\boldsymbol{\Pi} = \frac{1}{m_e c}\,(\mathbf{P} - e\mathbf{A}_L). \tag{3}$$

In order to describe the radiation scattering we let now the atomic system interact with a more general electromagnetic field, the superposition $\mathbf{A}_L(\mathbf{r}, t) + \mathbf{A}_X(\mathbf{r})$ of the classical laser field $\mathbf{A}_L(\mathbf{r}, t)$, and of a quantized one. To the latter we associate in the Coulomb gauge the vector potential operator

$$\mathbf{A}_X(\mathbf{r}) = \sum_{\mathbf{k}\lambda} \sqrt{\frac{\hbar}{2\varepsilon_0 \omega \mathcal{V}}}\,(\mathbf{s}_\lambda\, e^{i\mathbf{k}\cdot\mathbf{r}}\, a_{\mathbf{k}\lambda} + \mathbf{s}_\lambda^*\, e^{-i\mathbf{k}\cdot\mathbf{r}}\, a_{\mathbf{k}\lambda}^\dagger), \tag{4}$$

where $\mathcal{V}$ is the quantization volume. The wave vector $\mathbf{k}$ and the polarization vector $\mathbf{s}_\lambda(\mathbf{k})$ are orthogonal, $\mathbf{s}_\lambda^* \cdot \mathbf{k} = 0$, and define a general plane-wave mode of the radiation which we simply call the $\mathbf{k}\lambda$ mode. The operators $a_{\mathbf{k}\lambda}$ and $a_{\mathbf{k}\lambda}^\dagger$ annihilate and create, respectively, a photon of the mode $\mathbf{k}\lambda$, with an energy $\hbar\omega$ ($\omega = ck$) and a momentum $\hbar\mathbf{k}$. The Hamiltonian operator $\mathcal{H}$ of the composed dynamical system (electron and radiation) can be written as the sum of an "unperturbed" Hamiltonian $\mathcal{H}_0$, equal to $\mathcal{H}_{aL}$ plus the energy operator of the quantized field, and of a Hamiltonian describing the interaction between the atomic system moving in the laser field and the quantized field. This interaction Hamiltonian contains a term linear in $\mathbf{A}_X$, $\mathcal{H}_1' \equiv -(e/m_e)\mathbf{A}_X \cdot \boldsymbol{\Pi}$, and a quadratic one, $e^2\mathbf{A}_X^2/2m_e$, neglected in the following. We treat the interaction electron-quantized field in the first order of perturbation theory, applied for the interaction $\mathcal{H}_1'$. This term is responsible (in the first order) for processes involving either the absorption or the emission of a single photon. If the quantized field is initially in the vacuum state $|0\rangle$ (our case), only the spontaneous emission of a photon can appear. Implicitly, adopting this treatment, processes with two or more photons of the quantized field are systematically disregarded.

The possible final states of the radiation are then the vacuum state and the photon number states of the type $|1_{\mathbf{k}\lambda}\rangle$, having one photon present in the mode $\mathbf{k}\lambda$ (and zero photons in all the other modes). It follows that the state vector $|\Psi\rangle$ of the composed system can be approximated by the sum $|\Psi^{(0)}\rangle + |\Psi^{(1)}\rangle$, where the first term describes the evolution of the unperturbed system from the initial state $|\psi_0\rangle \otimes |0\rangle$, and has the form $|\Psi^{(0)}(t)\rangle = |\psi_i(t)\rangle \otimes |0\rangle$. The second one can be represented conveniently as $|\Psi^{(1)}(t)\rangle = \sum_{\mathbf{k}\lambda} |\psi_\lambda^{(1)}(\mathbf{k}, t)\rangle \otimes |1_{\mathbf{k}\lambda}\rangle e^{-i\omega t}$, where the equations for the components $|\psi_\lambda^{(1)}(\mathbf{k}, t)\rangle$ follow from the inhomogeneous equation satisfied by $|\Psi^{(1)}\rangle$, $i\hbar\,(\partial|\Psi^{(1)}\rangle/\partial t) = \mathcal{H}_0|\Psi^{(1)}\rangle + \mathcal{H}_1'|\Psi^{(0)}\rangle$. Imposing effectively the latter equation one finds that the vectors $|\psi_\lambda^{(1)}(\mathbf{k}, t)\rangle$ can be searched in the form

$$\left|\psi_\lambda^{(1)}(\mathbf{k}, t)\right\rangle = -\frac{e}{m_e}\sqrt{\frac{\hbar}{2\varepsilon_0 \omega \mathcal{V}}}\,\sum_j s_{\lambda j}^*(\mathbf{k})|\phi_j(\mathbf{k}, t)\rangle, \tag{5}$$

where j is a Cartesian index. The ket vectors $|\phi_j(\mathbf{k}, t)\rangle$, three for each $\mathbf{k}$, satisfy the *inhomogeneous* TDSE

$$i\hbar\frac{\partial|\phi_j\rangle}{\partial t} = \mathcal{H}_{aL}|\phi_j\rangle + e^{i\omega t}e^{-i\mathbf{k}\cdot\mathbf{r}}\,\Pi_j|\psi_i\rangle, \tag{6}$$

and vanish at the initial time moment t_0. We note that an equation of the same type appears in Ref. [15], where it plays a central role; unlike there, we work here with vectors defined such the inhomogeneous term [the last one in Eq. (6)] is independent of the volume $\mathcal{V}$ and of $\mathbf{s}_\lambda$—this is very convenient in

order to take the formal limit $\mathcal{V} \to \infty$ and to perform the sum over polarizations.

Putting together the above findings we have

$$|\Psi(t)\rangle = |\psi_i(t)\rangle \otimes |0\rangle - \frac{e}{m_e} \sum_{\mathbf{k}\lambda j} \sqrt{\frac{\hbar}{2\varepsilon_0 \omega \mathcal{V}}}\, s^*_{\lambda j}(\mathbf{k}) |\phi_j(\mathbf{k}, t)\rangle \otimes |1_{\mathbf{k}\lambda}\rangle e^{-i\omega t}. \tag{7}$$

The extraction of the electron and photon distributions from the state vector $|\Psi(t_1)\rangle$ of the composed system at the end of the laser pulse—even assuming the vectors $|\phi_j(\mathbf{k}, t_1)\rangle$ are known—is not a trivial operation. This task simplifies if $|\phi_j(\mathbf{k}, t)\rangle$ are decomposed in terms of a complete system of states $\{|\psi'_m(t)\rangle\}$ satisfying *homogeneous* TDSE and taken such to evolve to a complete system of eigenvectors $|u_m\rangle$ of H_a, at the end of the laser pulse, i.e., $|\psi'_m(t_1)\rangle = |u_m\rangle$. The completeness property of the set $\{|\psi'_m(t)\rangle\}$, inherited (as also the orthonormalization property) from that of the set $\{|u_m\rangle\}$, allows one to expand $|\phi_j(\mathbf{k}, t)\rangle$ in the form

$$|\phi_j(\mathbf{k}, t)\rangle = \sum_m c_{mj}(\mathbf{k}, t) |\psi'_m(t)\rangle. \tag{8}$$

By imposing Eq. (6), it follows that the expansion coefficients at $t = t_1$, denoted simply $c_{mj}(\mathbf{k})$, are given by

$$c_{mj}(\mathbf{k}) = \frac{1}{i\hbar} \int_{t_0}^{t_1} dt\, e^{i\omega t} \langle \psi'_m(t)| e^{-i\mathbf{k}\cdot\mathbf{r}}\, \Pi_j(t) |\psi_i(t)\rangle. \tag{9}$$

The generalized matrix element under integral (9) is built with TDSE solutions of two kinds, $|\psi_i(t)\rangle$ and $|\psi'_m(t)\rangle$, evolving, respectively, *from* an initial state $|\psi_0\rangle$ and *to* a final state $|u_m\rangle$.

Replacing the vectors (8) in Eq. (7) taken at $t = t_1$, and changing the index m into f (more suggestive for indicating "final" states), one finally expresses the sum over radiation modes from $|\Psi(t_1)\rangle$ as a linear combination of the vectors $|u_f\rangle \otimes |1_{\mathbf{k}\lambda}\rangle$.

According to the rules of quantum theory, the probability to find the electron in the final state $|u_f\rangle$ *and* a photon of the mode $\mathbf{k}\lambda$ to be emitted is equal to the squared modulus of the coefficient of $|u_f\rangle \otimes |1_{\mathbf{k}\lambda}\rangle$ from the expansion of $|\Psi(t_1)\rangle$. Multiplying this probability by the photon energy $\hbar\omega$ and taking the limit of $\mathcal{V}$ covering the whole physical space, one can infer the energy $d^2W_\lambda^{(f)}$ emitted in the spectral interval $d\omega$ and in the solid angle $d\Omega$ centered on the direction $\mathbf{n} \equiv \mathbf{k}/k$. We conveniently define the amplitude of the electron transition $i \to f$, with the emission of a photon $\mathbf{k}\lambda$:

$$\mathcal{A}_{i\to f}^{(\mathbf{k}\lambda)} \equiv -i \frac{\omega}{m_e c} \int_{t_0}^{t_1} dt\, e^{i\omega t} \times \langle \psi'_f(t)| e^{-i\mathbf{k}\cdot\mathbf{r}}\, \mathbf{s}^*_\lambda(\mathbf{k}) \cdot \mathbf{\Pi}(t) |\psi_i(t)\rangle. \tag{10}$$

With the help of this dimensionless amplitude, differing from $\mathbf{s}^*_\lambda(\mathbf{k}) \cdot \mathbf{c}_f(\mathbf{k})$ by the factor $\hbar\omega/m_e c$, the emitted energy distribution can be written as

$$d^2W_\lambda^{(f)} = \frac{1}{(2\pi)^2} \frac{e_0^2}{c} \left|\mathcal{A}_{i\to f}^{(\mathbf{k}\lambda)}\right|^2 d\omega\, d\Omega. \tag{11}$$

We now notice that the distribution we have obtained has the same form as the classical one in Eq. (1), with the important difference that the electron can be finally found in various quantum states. We also remark that the form (10) is not the direct quantum counterpart of the expression (2) for the classical amplitude $\mathcal{A}_{\rm cl}^{(\mathbf{n},\lambda)}$; instead, it is related to another form of $\mathcal{A}_{\rm cl}^{(\mathbf{n},\lambda)}$; see Eq. (A2) from Appendix A.

Equations (10) and (11), as they stand, implicitly contain an approximation: one neglects the emission of radiation in the absence of the laser pulse. A better (and more general) formula for the amplitude (10) is obtained, as in the classical case [1], including under the integral a convergence factor $e^{-\varepsilon|t|}$, extending the interval (t_0, t_1) to $(-\infty, \infty)$, and finally taking the limit $\varepsilon \to 0$. This "recipe" is understood in the following as applicable to all equations expressing the transition amplitude.

We note that the above equations require some adaptations for the scattering involving final or initial states of the electron from the continuum. Also, in the case of a monochromatic beam (of infinite duration), in place of $d^2W_\lambda^{(f)}$ we have to consider the energy emitted per second (the power), $d^2W_\lambda^{(f)}/\tau$, with $\tau \to \infty$.

The expression (10) of the TA is not convenient in every circumstance. In tight analogy with the classical treatment of the radiation emission by an accelerated charge [1], an equivalent form of TA can be derived. To this end it is preferable to start from the symmetric expression

$$\mathcal{A}_{i\to f} = -i \frac{\omega}{2m_e c} \int dt\, e^{i\omega t} \times \langle \psi'_f| e^{-i\mathbf{k}\cdot\mathbf{r}}\, \mathbf{s}^* \cdot \mathbf{\Pi} + \mathbf{s}^* \cdot \mathbf{\Pi}\, e^{-i\mathbf{k}\cdot\mathbf{r}} |\psi_i\rangle, \tag{12}$$

where we use the simplified notations $\mathbf{s}$ for $\mathbf{s}_\lambda(\mathbf{k})$ and $\mathcal{A}_{i\to f}$ for TA. Equations (12) and (10) are equivalent since the operators $e^{-i\mathbf{k}\cdot\mathbf{r}}$ and $\mathbf{s}^* \cdot \mathbf{\Pi}$ commute:

$$[\mathbf{s}^* \cdot \mathbf{\Pi}, e^{-i\mathbf{k}\cdot\mathbf{r}}] = [\mathbf{s}^* \cdot \mathbf{P}, e^{-i\mathbf{k}\cdot\mathbf{r}}] = -\hbar(\mathbf{s}^* \cdot \mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{r}} = 0, \tag{13}$$

the last equality being due to the orthogonality of the vectors $\mathbf{s}_\lambda(\mathbf{k})$ and $\mathbf{k}$. For the derivation of another form of TA, detailed in Appendix A, it is convenient to define in advance an operator $\mathring{O}$ ("O ring"), associated to an arbitrary operator O such that the time derivative of the matrix element $\langle \psi'_f|O|\psi_i\rangle$ is equal to $\langle \psi'_f|\mathring{O}|\psi_i\rangle$ (for any indices i and f); see Eqs. (A4) and (A5). In particular we have

$$\mathring{\mathbf{\Pi}} = \mathbf{F}, \quad \mathring{\boldsymbol{\beta}} = \frac{1}{m_e c} \mathbf{F}, \tag{14}$$

where $\mathbf{F}$ is the self-adjoint operator corresponding to the total force, i.e., the sum of the quantum Lorentz force in the laser field and of the potential force,

$$\mathbf{F} \equiv \mathbf{F}_e + \mathbf{F}_m + \mathbf{F}_p, \tag{15}$$

with

$$\mathbf{F}_e \equiv e\mathbf{E}, \quad \mathbf{F}_m \equiv \frac{e}{2m_e}(\mathbf{\Pi} \times \mathbf{B} - \mathbf{B} \times \mathbf{\Pi}), \tag{16}$$

and $\mathbf{F}_p \equiv -\nabla V$.

In Appendix A it is shown that the transition amplitude can be expressed (up to terms of an order in β higher than 1,

negligible in the NR case) in the form

$$\widetilde{\mathcal{A}}_{i\to f} = \frac{1}{2m_e c}\int dt\, e^{i\omega t}\,\langle\psi'_f|e^{-i\mathbf{k}\cdot\mathbf{r}}\mathbf{s}^*\cdot\widetilde{\mathbf{F}} + \mathbf{s}^*\cdot\widetilde{\mathbf{F}}e^{-i\mathbf{k}\cdot\mathbf{r}}\,|\psi_i\rangle, \tag{17}$$

where $\widetilde{\mathbf{F}}$ is the self-adjoint operator

$$\widetilde{\mathbf{F}} \equiv \mathbf{F} + \tfrac{1}{2}\,[(\mathbf{n}\cdot\boldsymbol{\beta})\,\mathbf{F} + \mathbf{F}\,(\mathbf{n}\cdot\boldsymbol{\beta}) + (\mathbf{n}\cdot\mathbf{F})\,\boldsymbol{\beta} + \boldsymbol{\beta}\,(\mathbf{n}\cdot\mathbf{F})], \tag{18}$$

equal to to the sum of the total force operator (15) and of an auxiliary self-adjoint operator, linear in $\mathbf{F}$ and $\boldsymbol{\beta}$. In the latter, in order to obtain a consistent nonrelativistic expression, $\mathbf{F}$ is replaced by $\mathbf{F}_e + \mathbf{F}_p$ since the magnetic force is of the first order in β. Setting aside (temporarily) the contribution of $\mathbf{F}_p$ in Eq. (18), we get the explicit formula

$$\widetilde{\mathbf{F}} \equiv \mathbf{F}_e + \mathbf{F}_m + \frac{1}{2m_e c}\times[(\mathbf{n}\cdot\boldsymbol{\Pi})\,\mathbf{F}_e + \mathbf{F}_e\,(\mathbf{n}\cdot\boldsymbol{\Pi}) + (\mathbf{n}\cdot\mathbf{F}_e)\,\boldsymbol{\Pi} + \boldsymbol{\Pi}\,(\mathbf{n}\cdot\mathbf{F}_e)]. \tag{19}$$

To include the contribution of the potential force in the last equation one replaces $\mathbf{F}_e$ by $\mathbf{F}_e - \boldsymbol{\nabla}V$.

We note that the above expressions for TA are obtained working in the Coulomb gauge (for both fields, classical and quantized). In practice it might be of interest to solve TDSE in a different gauge for the laser field, this raising the problem to express TA in an arbitrary gauge G. One easily verifies the gauge invariance of the matrix elements that enter in Eq. (10) or (17) to a general gauge transformation, $\mathbf{A}\to\mathbf{A}_G = \mathbf{A} + \nabla g$ and $\Phi\to\Phi_G = \Phi - \partial g/\partial t$ (Φ is the scalar potential; in the above equations for TA the potentials are $\mathbf{A} = \mathbf{A}_L$ and $\Phi = 0$), generated by an arbitrary function $g(\mathbf{r}, t)$. For verification one uses (i)the transformation $|\psi\rangle\to|\psi_G\rangle = e^{i(e/\hbar)g}|\psi\rangle$ of the states under a gauge transformation and (ii) the identity $\boldsymbol{\Pi}|\psi\rangle = e^{-i(e/\hbar)g}\,\boldsymbol{\Pi}_G\,|\psi_G\rangle$, where $\boldsymbol{\Pi} = \mathbf{P} - e\mathbf{A}$ and $\boldsymbol{\Pi}_G = \mathbf{P} - e\mathbf{A}_G$ are the operators associated to the kinetic momentum in the two gauges.

What happens in the classical limit with the above equations is almost transparent at the formal level. When in Eq. (17), for example, the operators are replaced by the quantities to which they are associated, one obtains the classical amplitude (2), with $\mathbf{a}_{\rm cl} = [e(\mathbf{E} + \mathbf{v}_{\rm cl}\times\mathbf{B}) - \boldsymbol{\nabla}V]/m_e$, multiplied by the constant scalar product $\langle\psi'_f(t)|\psi_i(t)\rangle = \langle u_f|\psi_i(t_1)\rangle$. The summation over the final electron states in Eq. (11) then leads to the classical result for the emitted energy, described by Eqs. (1) and (2).

For the validity of Eqs. (10) and (17), the interaction laser-atomic system was not assumed as being small—thus they are *exact* (or nonperturbative) with respect to this interaction. Comparing the two expressions of the TA, Eqs. (10) and (17), one sees that the former is simpler, while the latter clearly shows that the radiation is emitted only on time intervals where the motion of the electron is accelerated (i.e., the force $\mathbf{F}$ is nonvanishing), this feature being attractive also for practical calculations.

We include here just a few considerations related to the evaluation of Eqs. (10) and (17), leaving the problem in its full complexity for a future analysis. In a concrete calculation, the state vectors required under integrals (10) or (17) have to be known as functions of time. The most natural way to find them is to solve numerically TDSE using appropriate methods. For simple systems the integration of TDSE is rather easy in the case of low laser frequencies, where the dipole approximation is successfully used. At higher frequencies and high intensities, where the TDSE integration is much more difficult, there were proposed alternative nonrelativistic or semirelativistic formulations [23–25], seemingly computationally more efficient than the usual approaches based on the minimal coupling for the light-matter interaction, and aiming to ensure a smooth transition between the nonrelativistic and relativistic descriptions.

In close relation with the values of the parameters relevant for scattering, the approximation methods could present a high interest (see also Sec. I). In particular, the choice of SFA can be taken in consideration for laser intensities such that the effect of the Lorentz force becomes dominant versus that of the potential force (usually the Coulomb force electron-nucleus). In the spirit of SFA (as it is used in the study of ATI or HHG) the states from the matrix element in Eq. (10) or (17) are expressed using the evolution operators $U_{aL}(t, t')$ ($t' = t_i$ or t_f) for which one inserts the SFA equations (in the latter the potential V is treated perturbatively) [26]. For low to moderate laser intensities, the methods based on perturbation theory can represent the appropriate choice (see also the discussion from Sec. I), especially in the case the laser field is close to a monochromatic one. The transition amplitude is then approximated replacing the electron states in the matrix elements from Eq. (10) or (17) by their perturbation series in the field strength [27]. As discussed in Sec. III, even if Eq. (10) is simpler to apply for PT calculations, Eq. (17) is also attractive to this end due to the more intuitive aspect of the involved relations. An advantage of Eq. (17) versus Eq. (10) manifests, for example, when the potential force $-\boldsymbol{\nabla}V$ vanishes or has a negligible effect—in this case the application of Eq. (17) for a given process requires TDSE solutions in a PT order lower (by one unit) than in the case of Eq. (10). In fact, if additional approximations are used for the PT corrections of the state vectors, both equations could be necessary for consistency reasons. We mention here that a different approach might be tried for independent checkings of the PT results, based on an extension of the TDSE treatment used in Refs. [28,29], where the studied process is the *stimulated* Compton scattering—NLCS could be partially simulated by a two-color Compton scattering in the appropriate perturbative regime, with one color corresponding to the incident laser and the other to a scattered radiation mode.

III. SCATTERING WITH ABSORPTION OF TWO LASER PHOTONS

We apply the formalism from Sec. II to the scattering of a monochromatic laser beam, in the *perturbative* regime for the interaction laser-atomic system. The application is centered on the simplest nonlinear process of this type, the scattering with the absorption of two laser photons. The laser field is modeled by a plane electromagnetic wave, monochromatic and linearly polarized, described by the vector potential

$$\mathbf{A}_L(\mathbf{r}, t) = \tfrac{1}{2}\,A_0\,\mathbf{s}_0\,e^{i(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t)} + \text{c.c.}, \quad \mathbf{s}_0\cdot\mathbf{k}_0 = 0. \tag{20}$$

The wave is polarized along $\mathbf{s}_0$ and propagates in the direction $\mathbf{n}_0 = \mathbf{k}_0/k_0$ of the wave vector $\mathbf{k}_0$. The amplitude A_0 is assumed to be such that the parameter

$$\xi \equiv \frac{|e|A_0}{m_e c} \tag{21}$$

satisfies the condition $\xi \ll 1$, for which the relativistic effects are negligible for an electron quivering in the laser field (20). This classical condition has to be complemented by a quantum one, requiring that the energy $\hbar\omega_0$ of a laser photon is much smaller than the rest energy of the electron or, equivalently, that the momentum of the laser photon in $m_e c$ units, $\varkappa_0 \equiv \hbar k_0/m_e c$, is much less than 1. Recalling that the wave intensity is given by $I = \varepsilon_0\, c\, E_0^2/2$, where $E_0 = \omega_0 A_0$, one finds that in the keV spectral range (such that the incident photon energy is much less than $m_e c^2 \approx 511$ keV) the condition $\xi \ll 1$ is satisfied even for intensities much exceeding the atomic intensity I_0, as in the experiment [16].

The scattering with the absorption of m laser photons and emission of a photon $\mathbf{k}\lambda$ is accompanied by the transfer, from the laser beam to the atomic system, of a momentum $\mathbf{Q}_m$ and of an energy Δ_m, given by

$$\mathbf{Q}_m \equiv m\hbar\mathbf{k}_0 - \hbar\mathbf{k}, \quad \Delta_m \equiv m\hbar\omega_0 - \hbar\omega. \tag{22}$$

For the scattering by a *free* electron having an initial momentum equal to zero [the system termed below as a free electron "at rest" (FER)], the spectrum of the emitted frequencies is reduced to a single line for each scattering angle θ (the angle between $\mathbf{n}$ and $\mathbf{n}_0$), the position of which is given by the relation

$$\omega_m \equiv \frac{m\omega_0}{1 + m\varkappa_0(1 - \cos\theta)}, \tag{23}$$

a consequence of the relativistic laws expressing the conservation of the momentum and energy. If the NR conservation laws are used one obtains an approximation $\widetilde{\omega}_m$ differing from ω_m by a term of the order $\varkappa_0^3$, negligible in the present conditions—below we do not make a distinction between $\widetilde{\omega}_m$ and ω_m. The line is replaced by a true distribution of the emitted frequencies in the case the initial momentum of the electron is not defined.

In the following we concentrate on the nonlinear Compton scattering with the absorption of $m = 2$ laser photons by (1) a *free* electron initially in a nonstationary state (a wave packet) and (2) an electron moving in a nonvanishing potential V, initially in a stationary or a nonstationary state. Taking into account the practical difficulties of the exact perturbative calculations in case 2 (for some of the involved terms), we adopt below some other simplifications and approximations—some of them are extended also to case 1. Details for each case are given after the identification of the dominant contributions to the transition amplitude. For the calculation of TA we apply Eqs. (10) and (17) (for consistency reasons they are both used, see below) for the monochromatic plane wave (20). We first transcribe Eq. (10) in the simplified form

$$\mathcal{A}_{i\to f} = \frac{1}{m_e c}\int dt\, e^{i\omega t}\, \mathcal{M}_{fi}, \tag{24}$$

where

$$\mathcal{M}_{fi} \equiv -i\omega\langle\psi'_f|e^{-i\mathbf{k}\cdot\mathbf{r}}\,\mathbf{s}^*\cdot\mathbf{\Pi}|\psi_i\rangle, \tag{25}$$

and $\mathbf{s}$ is the polarization vector of the emitted photon. The expression (17) of the amplitude can be put in a form similar to Eq. (24):

$$\widetilde{\mathcal{A}}_{i\to f} = \frac{1}{m_e c}\int dt\, e^{i\omega t}\, \widetilde{\mathcal{M}}_{fi}, \tag{26}$$

with

$$\widetilde{\mathcal{M}}_{fi} \equiv \tfrac{1}{2}\langle\psi'_f|e^{-i\mathbf{k}\cdot\mathbf{r}}\,\mathbf{s}^*\cdot\widetilde{\mathbf{F}} + \mathbf{s}^*\cdot\widetilde{\mathbf{F}}\,e^{-i\mathbf{k}\cdot\mathbf{r}}|\psi_i\rangle, \tag{27}$$

where $\widetilde{\mathbf{F}}$ is given by Eq. (19).

In order to treat perturbatively the considered process, we keep from the expansion of $\mathcal{M}_{fi}$ or $\widetilde{\mathcal{M}}_{fi}$ in powers of A_0 the terms quadratic (in A_0) contributing to the *absorption* of two laser photons. We first consider the matrix element $\widetilde{\mathcal{M}}_{fi}$, Eq. (27). The main simplification we adopt in case 2 is to set aside the contribution of all the terms involving the potential force, thus the operator $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}$ is the same in the two cases and contains terms of the first and second order in A_0. These are obtained replacing the expressions for $\mathbf{F}_e$, $\mathbf{F}_m$, and $\mathbf{\Pi}$ in Eq. (19). Then we may write

$$\widetilde{\mathbf{F}} = \widetilde{\mathbf{F}}^{(1)} + \widetilde{\mathbf{F}}^{(2)}, \tag{28}$$

where

$$\widetilde{\mathbf{F}}^{(1)} \equiv e\mathbf{E} + \frac{e}{2m_e}(\mathbf{P}\times\mathbf{B} - \mathbf{B}\times\mathbf{P}) + \frac{e}{2m_e c} \times[(\mathbf{n}\cdot\mathbf{P})\mathbf{E} + \mathbf{E}(\mathbf{n}\cdot\mathbf{P}) + (\mathbf{n}\cdot\mathbf{E})\mathbf{P} + \mathbf{P}(\mathbf{n}\cdot\mathbf{E})], \tag{29}$$

and

$$\widetilde{\mathbf{F}}^{(2)} = \widetilde{\mathbf{F}}^{(2)}_a + \widetilde{\mathbf{F}}^{(2)}_b, \tag{30}$$

with

$$\widetilde{\mathbf{F}}^{(2)}_a \equiv -\frac{e^2}{m_e}\mathbf{A}_L\times\mathbf{B}, \tag{31}$$

and

$$\widetilde{\mathbf{F}}^{(2)}_b \equiv -\frac{e^2}{m_e c}[(\mathbf{n}\cdot\mathbf{A}_L)\mathbf{E} + (\mathbf{n}\cdot\mathbf{E})\mathbf{A}_L]. \tag{32}$$

We note that for the wave (20) $\widetilde{\mathbf{F}}^{(2)}_a$ is along $\mathbf{n}_0$ while $\widetilde{\mathbf{F}}^{(2)}_b$ has the direction $\mathbf{s}_0$. The perturbative approximation (in the lowest order) of $\widetilde{\mathcal{M}}_{fi}$ is then obtained by combining properly the terms of $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}$ with the zeroth-order approximation or *PT* correction of the first order for the states $|\psi_i(t)\rangle$ and $|\psi'_f(t)\rangle$. The notations $|\psi^{(0)}(t)\rangle$ and $|\psi^{(1)}(t)\rangle$ are used below to indicate, respectively, the zeroth-order approximation and the first-order correction of a TDSE solution $|\psi(t)\rangle$, either $|\psi_i(t)\rangle$ or $|\psi'_f(t)\rangle$. The (unperturbed) final states are naturally taken of stationary type and we denote by E_f a possible final energy of the electron. For the initial state we consider first the stationary case, and denote by E_i the corresponding energy. Then the implications for the nonstationary case are pursued.

We proceed with the explicit enumeration of the dominant contributions to $\widetilde{\mathcal{M}}_{fi}$, using case 2 as reference. The simplest one, which we denote $\widetilde{\mathcal{M}}^{(2)}_{fi}$, originates from $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}^{(2)}$. In the expansion of $\widetilde{\mathcal{M}}_{fi}$, the quantity $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}^{(2)}$ (more precisely its positive frequency part) has to be coupled with the unperturbed states $|\psi_i^{(0)}(t)\rangle$ and $|\psi_f^{\prime(0)}(t)\rangle$. When it is explicitly

written, one finds that $\widetilde{\mathcal{M}}_{fi}^{(2)}$ is given by a factor proportional to $e^{-2i\omega_0 t}$, multiplying the matrix element $\langle\psi_f'^{(0)}|e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}|\psi_i^{(0)}\rangle$ of the exponential operator $e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}$.

Another contribution, denoted $\widetilde{\mathcal{M}}_{fi}^{(1)}$, is obtained coupling the terms of the *first order* in A_0, $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}^{(1)}$, with the first-order correction of the initial or final state (and zeroth-order approximation of the other one). From this category we only keep the terms due to the electric force, $\mathbf{s}^*\cdot(e\mathbf{E})$ (its positive frequency part), dominant versus the others, which are giving corrections of a higher order in β or $\varkappa=\hbar\omega/m_ec$. We address first the case of a *stationary* initial state, for which one obtains that $\widetilde{\mathcal{M}}_{fi}^{(1)}$ reduces, up to a factor proportional to $e^{-2i\omega_0 t}$, to the matrix element $\langle\psi_f'^{(0)}|\mathcal{O}|\psi_i^{(0)}\rangle$ of an operator $\mathcal{O}\equiv e^{i(\mathbf{k}_0-\mathbf{k})\cdot\mathbf{r}}G(E_i+\hbar\omega_0)e^{i\mathbf{k}_0\cdot\mathbf{r}}\mathbf{s}_0\cdot\mathbf{P}+\mathbf{s}_0\cdot\mathbf{P}e^{i\mathbf{k}_0\cdot\mathbf{r}}G(E_f-\hbar\omega_0)e^{i(\mathbf{k}_0-\mathbf{k})\cdot\mathbf{r}}$, where $G(z)\equiv(z-H_a)^{-1}$ is the resolvent operator of argument z, attached to H_a. The regime where the energies E_i and E_f are small (versus the energy $\hbar\omega_0$ of a laser photon) is of high interest, it allowing a simple approximate treatment of the matrix element, favored by the presence in $\mathcal{O}$ of the exponential operators involving photon momenta. The approximation consists in the replacement of the resolvent operators $(E_i+\hbar\omega_0-H_a)^{-1}$ and $(E_f-\hbar\omega_0-H_a)^{-1}$ with the constant operators $1/\hbar\omega_0$ and $-1/\hbar\omega_0$—this is equivalent to keeping only the contribution of the intermediate states of energies E_j such that $|E_j-E_{i,f}|$ is much lower than $\hbar\omega_0$ (thus one neglects the contribution of the high-energy states), and to setting $|E_j-E_{i,f}|=0$. Then it follows that $\mathcal{O}$ reduces to $[e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar},\mathbf{s}_0\cdot\mathbf{P}]/\hbar\omega_0=(1/c)(\mathbf{s}_0\cdot\mathbf{n})(\omega/\omega_0)e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}$. The result one obtains this way for $\widetilde{\mathcal{M}}_{fi}^{(1)}$ keeps the same analytic form in the case the initial state is *nonstationary*: this can be understood if one represents $|\psi_i^{(0)}(t)\rangle$ as a packet of stationary states and one approximates the resolvent operators as above (by constants independent on the involved energies) in all the terms of $\widetilde{\mathcal{M}}_{fi}^{(1)}$ corresponding to the components of the packet.

Consequently, taking the sum $\widetilde{\mathcal{M}}_{fi}^{(2)}+\widetilde{\mathcal{M}}_{fi}^{(1)}$ of the two contributions above discussed, for the matrix element (27) one derives the approximate result

$$\widetilde{\mathcal{M}}_{fi}=-\frac{i}{4}\frac{e^2A_0^2}{m_ec}\,\omega_0\,(\mathbf{s}^*\cdot\mathbf{u})\times\left\langle\psi_f'^{(0)}\middle|e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}\middle|\psi_i^{(0)}\right\rangle e^{-2i\omega_0 t},\tag{33}$$

where the real vector $\mathbf{u}$ has to be replaced by

$$\mathbf{u}_2\equiv\mathbf{n}_0+\left(2+\frac{\omega}{\omega_0}\right)(\mathbf{n}\cdot\mathbf{s}_0)\,\mathbf{s}_0.\tag{34}$$

The first term of $\mathbf{u}_2$ corresponds to $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}_a^{(2)}$, and the second corresponds to the sum of the contributions due to $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}_b^{(2)}$ and $\mathbf{s}^*\cdot(e\mathbf{E})$ from above. In particular, for the emission along the laser polarization ($\mathbf{n}=\mathbf{s}_0$) only the contribution of the magnetic term $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}_a^{(2)}$ survives since in this case $\mathbf{s}^*\cdot\mathbf{u}_2=\mathbf{s}^*\cdot\mathbf{n}_0$.

The matrix element $\mathcal{M}_{fi}$, Eq. (25), can be treated following a procedure similar to that applied to $\widetilde{\mathcal{M}}_{fi}$, with $\mathbf{s}^*\cdot\mathbf{\Pi}=\mathbf{s}^*\cdot\mathbf{P}-\mathbf{s}^*\cdot(e\mathbf{A}_L)$ in place of $\mathbf{s}^*\cdot\widetilde{\mathbf{F}}$. The calculation of the contribution to $\mathcal{M}_{fi}$, corresponding to $\mathbf{s}^*\cdot\mathbf{P}$, becomes feasible if we neglect the effect of the interaction term $-(e/m_e)\mathbf{A}_L\cdot\mathbf{P}$ from the Hamiltonian $\mathcal{H}_{aL}$ and approximate the resolvent operators $G(E_i+2\hbar\omega_0)$ and $G(E_f-2\hbar\omega_0)$ (they are involved in the *PT* corrections to the state vectors of the *first* order in the interaction term $e^2A_L^2/2m_e$) by $1/2\hbar\omega_0$ and $-1/2\hbar\omega_0$, respectively. The other contribution, coming from $\mathbf{s}^*\cdot(e\mathbf{A}_L)$, is approximated as in the case of the term $\mathbf{s}^*\cdot(e\mathbf{E})$ from above. For $\mathcal{M}_{fi}$ one finally obtains the same form as for $\widetilde{\mathcal{M}}_{fi}$, Eq. (33), where the vector $\mathbf{u}$ is now replaced by

$$\mathbf{u}_1\equiv\frac{\omega}{2\omega_0}\left[\mathbf{n}_0+2\frac{\omega}{\omega_0}(\mathbf{n}\cdot\mathbf{s}_0)\,\mathbf{s}_0\right].\tag{35}$$

Comparing the corresponding expressions for the transition amplitude, $\mathcal{A}_{i\to f}$ [Eq. (24)] and $\widetilde{\mathcal{A}}_{i\to f}$ [Eq. (26)], one sees that they differ (only) by factors of the type $\mathbf{s}^*\cdot\mathbf{u}$, where $\mathbf{u}$ is given by Eq. (34) or (35). At this stage we remark that the integrals from Eqs. (24) and (26) take significant values for frequencies around ω_2 [given by Eq. (23)], for which the ratio ω/ω_0 in Eqs. (34) and (35) is less than 2 by a term of the order $\varkappa_0\ll1$. The inconsistency of the above results for TA is then explainable since in their derivation there were neglected terms of this order. The consistency is reached in zeroth order for $\varkappa_0$, approximating $\omega/\omega_0=2$ in Eqs. (34) and (35)—in this case the vectors $\mathbf{u}_{1,2}$ reduce both to

$$\mathbf{u}_0\equiv\mathbf{n}_0+4(\mathbf{n}\cdot\mathbf{s}_0)\,\mathbf{s}_0.\tag{36}$$

In fact, a NR formula valid in the first order for $\varkappa_0$ and suggested by Eq. (35) can be indirectly uncovered by the examination of some results from the literature, for the scattering by a free electron. Equation (4) in Ref. [18] indicates that a factor of the form $\mathbf{s}^*\cdot\mathbf{u}_0$ has to appear in Eq. (33). Then a correction of the first order in $\varkappa_0$ can arise only from the first factor seen in Eq. (35), $\omega/2\omega_0$. This implication is confirmed by Eqs. (3.36) and (3.42) in Ref. [2], which are leading in the NR approximation to a correction factor for the angular distribution equal to $(\omega_2/2\omega_0)^2$. Guided by the above arguments, we arrive to the same result for the matrix elements $\mathcal{M}_{fi}$ and $\widetilde{\mathcal{M}}_{fi}$, given by Eq. (33), by taking

$$\mathbf{u}\equiv\frac{\omega}{2\omega_0}\,\mathbf{u}_0=\frac{\omega}{2\omega_0}\,[\mathbf{n}_0+4(\mathbf{n}\cdot\mathbf{s}_0)\,\mathbf{s}_0],\tag{37}$$

for which $\mathcal{M}_{fi}=\widetilde{\mathcal{M}}_{fi}$ and $\mathcal{A}_{i\to f}=\widetilde{\mathcal{A}}_{i\to f}$.

We first specialize Eq. (33) for a free electron, case 1, assumed to be initially in an arbitrary state $|\psi_0\rangle$. The state function in the momentum representation, at the initial moment, is $\varphi(\mathbf{p}_0)\equiv\langle\mathbf{p}_0|\psi_0\rangle$, where $|\mathbf{p}_0\rangle$ is a state of a defined momentum $\mathbf{p}_0$ ($\mathbf{p}_0$ is any real vector). The unperturbed state evolving freely from $|\psi_0\rangle$ is then given by the superposition $|\psi_i^{(0)}(t)\rangle=\int d\mathbf{p}_0\varphi(\mathbf{p}_0)|\mathbf{p}_0\rangle\exp[-iE(\mathbf{p}_0)t/\hbar]$—for the energy of a free electron of a momentum $\mathbf{p}$ we use the general notation $E(\mathbf{p})\equiv\mathbf{p}^2/2m_e$. For the unperturbed final state we take $|\psi_f'^{(0)}(t)\rangle=|\mathbf{p}\rangle\exp(-iE(\mathbf{p})t/\hbar)$, i.e., a state of a defined momentum $\mathbf{p}$ and an energy $E_f=E(\mathbf{p})$. Using these states, for the matrix element from the right-hand side of Eq. (33) one obtains the simple result $\varphi(\mathbf{p}-\mathbf{Q}_2)\exp\{i[E(\mathbf{p})-E(\mathbf{p}-\mathbf{Q}_2)]t/\hbar\}$. Replacing the corresponding amplitude, Eq. (24) (with the notation for polarization restored), in Eq. (11) (adapted for a continuum of final states $|\mathbf{p}\rangle$), one obtains the power $d^3\mathcal{P}_\lambda\equiv d^3W_\lambda/\tau$ (in the limit $\tau\to\infty$) emitted in the spectral range $d\omega$ and solid angle $d\Omega$, for a final electron

momentum in $d\mathbf{p}$. It can be expressed as follows:

$$d^3\mathcal{P}_\lambda = \frac{\alpha}{32\pi}\,\xi^4\,(\hbar\omega_0)^2\,|\mathbf{s}_\lambda^*\cdot\mathbf{u}|^2\,|\varphi(\mathbf{p}-\mathbf{Q}_2)|^2 \times \delta(E(\mathbf{p})-E(\mathbf{p}-\mathbf{Q}_2)-\Delta_2)\,d\omega\,d\Omega\,d\mathbf{p}, \quad (38)$$

where $\alpha = e_0^2/\hbar c \approx 1/137$ is the fine-structure constant. Like in the relativistic study [9], no interference effects can appear between the contributions to the scattering of the different components of the wave packet.

We next apply Eq. (33) in case 2, for an initial state of stationary type. In the matrix element $\widetilde{\mathcal{M}}_{fi}$ we use *exact* unperturbed states for the electron in the potential V. The unperturbed TDSE solution corresponding to the initial state of energy E_i is $|\psi_i^{(0)}(t)\rangle = |u_i\rangle\exp(-iE_it/\hbar)$, where $|u_i\rangle$ is an eigenvector of H_a for the eigenvalue E_i. For the final state, belonging to a continuum of states, the choice is $|\psi_f'^{(0)}(t)\rangle = |\mathbf{p}-\rangle\exp(-iE(\mathbf{p})t/\hbar)$, where $|\mathbf{p}-\rangle$ is a scattering state of incoming type [30], for the final energy $E_f = E(\mathbf{p})$. Proceeding as above, we are led to the relation

$$d^3\mathcal{P}_\lambda = \frac{\alpha}{32\pi}\,\xi^4\,(\hbar\omega_0)^2\,|\mathbf{s}_\lambda^*\cdot\mathbf{u}|^2\,|\langle\mathbf{p}-|e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}|u_i\rangle|^2 \times \delta(E(\mathbf{p})-E_i-\Delta_2)\,d\omega\,d\Omega\,d\mathbf{p}. \quad (39)$$

The results obtained using this equation and its integrated version are called below "exact"—the attribute "exact" is related to the use of exact states in the matrix element, otherwise the results are approximate (the treatment is in PT, some force terms were ignored, and a supplementary approximation was imposed).

We get a formula simpler than Eq. (39) working in IA, in which the role of the binding potential V during the light scattering is totally neglected—the electron being *free*, one can now apply Eq. (38), with the appropriate choice of the initial state, $|\psi_0\rangle = |u_i\rangle$, usually the ground state of the electron in the potential V; we note that for the free electron this state is of nonstationary type. For an easy comparison of the two predictions (in IA and the exact one), we write the IA formula as follows:

$$d^3\mathcal{P}_\lambda = \frac{\alpha}{32\pi}\,\xi^4\,(\hbar\omega_0)^2\,|\mathbf{s}_\lambda^*\cdot\mathbf{u}|^2\,|\langle\mathbf{p}|e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}|u_i\rangle|^2 \times \delta(E(\mathbf{p})-E(\mathbf{p}-\mathbf{Q}_2)-\Delta_2)\,d\omega\,d\Omega\,d\mathbf{p}, \quad (40)$$

where $\langle\mathbf{p}|e^{i\mathbf{Q}_2\cdot\mathbf{r}/\hbar}|u_i\rangle = \varphi(\mathbf{p}-\mathbf{Q}_2)$, with $\varphi(\mathbf{p}) \equiv \langle\mathbf{p}|u_i\rangle$. One sees that Eq. (40) differs from Eq. (39) by (a) the change of the final state in the matrix element, from $|\mathbf{p}-\rangle$ to $|\mathbf{p}\rangle$, and (b) the initial energy E_i from the argument of the δ function, which is replaced by $E(\mathbf{p}-\mathbf{Q}_2)$.

If the electron is not observed, the relevant quantities are the powers $d^2\mathcal{P}_\lambda$, obtained from Eq. (40) or (39) by integration over the final momentum $\mathbf{p}$ of the electron, and $d^2\mathcal{P} \equiv \sum_\lambda d^2\mathcal{P}_\lambda$—the latter for the case the polarization of the emitted photon is not detected.

IV. SPECTRAL AND ANGULAR DISTRIBUTIONS FOR THE SCATTERING FROM A GROUND-STATE HYDROGENIC ATOM

In the following we refer to the radiation scattering from a ground-state hydrogenic atom. Rather simple expressions can be derived for IA and exact *integrated* distributions, where $|u_i\rangle$ from the start formulas, (40) and (39), describes now the ground state of the electron in the Coulomb potential of the nucleus. In both cases, the integration in $\mathbf{p}$ can be done analytically, like in the LCS case; see, for instance, Ref. [21] and references therein.

The NLCS power distribution can be generically expressed in the form

$$\left(\frac{d^2\mathcal{P}}{d\omega\,d\Omega}\right)_{\rm NLCS} = \frac{\alpha}{32\pi}\left(\frac{\omega}{2\omega_0}\right)^2 \times \xi^4\,(\hbar\omega_0)^2\,J(Q_2,\Delta_2)\,(\mathbf{s}\cdot\mathbf{u}_0)^2, \quad (41)$$

where, for simplicity, the polarization vector is taken real and the polarization index (of $\mathbf{s}$ and $\mathcal{P}$) is omitted. Explicitly, the last factor [see Eq. (37)] is

$$(\mathbf{s}\cdot\mathbf{u}_0)^2 = [(\mathbf{s}\cdot\mathbf{n}_0)+4(\mathbf{n}\cdot\mathbf{s}_0)\,(\mathbf{s}\cdot\mathbf{s}_0)]^2. \quad (42)$$

For the scattering geometry (close to that used in Ref. [16]) where the direction $\mathbf{n}$ is in the plane $(\mathbf{s}_0,\mathbf{n}_0)$, the sum over polarizations is reduced to a single term of the type (41) if the vector $\mathbf{s}$ (orthogonal to $\mathbf{n}$) is taken in the same plane. This special geometry is characterized by the directions

$$\mathbf{s} = \cos\theta\,\mathbf{s}_0 - \sin\theta\,\mathbf{n}_0, \quad \mathbf{n} = \sin\theta\,\mathbf{s}_0 + \cos\theta\,\mathbf{n}_0, \quad (43)$$

where θ is the scattering angle. In its case

$$(\mathbf{s}\cdot\mathbf{u}_0)^2 = (1-4\cos\theta)^2\,\sin^2\theta, \quad (44)$$

and with this value for $(\mathbf{s}\cdot\mathbf{u}_0)^2$, Eq. (41) describes now the power distribution summed over polarizations (for the actual scattering geometry).

The function $J(Q,\Delta)$ is defined in IA by

$$J^{(\rm IA)}(Q,\Delta) \equiv \frac{8}{3\pi}\,\frac{(bQ/m_e)^5}{[(E(Q)-\Delta)^2+(bQ/m_e)^2]^3}, \quad (45)$$

and depends also on the atomic number Z by the parameter $b \equiv \hbar Z/a_0 = \alpha Z m_e c$. For the exact distribution, $J(Q,\Delta)$ is replaced by

$$J^{(\rm ex)}(Q,\Delta) \equiv \frac{256}{3}\,\frac{m_e b^6 Q^2(b^2+3Q^2+p^2)}{[(b^2+Q^2-p^2)^2+4b^2p^2]^3} \times \frac{e^{-2\nu\gamma}}{1-e^{-2\pi\nu}}, \quad (46)$$

with $\nu \equiv b/p$ and

$$\gamma \equiv \arccos((b^2+Q^2-p^2)/\sqrt{(b^2+Q^2-p^2)^2+4b^2p^2}).$$

The asymptotic final momentum p is determined by the energy conservation, $p = \sqrt{2m_e(E_1+\Delta)}$, where the ground-state energy E_1 is conveniently expressed as $E_1 = -b^2/2m_e$.

For comparison we also need the results for the LCS distributions. The equations corresponding to the differential LCS distributions are presented in Appendix B. The integration of Eqs. (B2) and (B3) with respect to the final electron momentum leads to the generic relation

$$\left(\frac{d^2\mathcal{P}}{d\omega\,d\Omega}\right)_{\rm LCS} = \frac{\alpha}{8\pi}\left(\frac{\omega}{\omega_0}\right)^2 \xi^2\,(\hbar\omega_0)^2\,J(Q_1,\Delta_1)\,(\mathbf{s}\cdot\mathbf{s}_0)^2, \quad (47)$$

which has to be understood like above: the function $J(Q_1,\Delta_1)$ is replaced according to Eq. (45) or (46) in order to describe

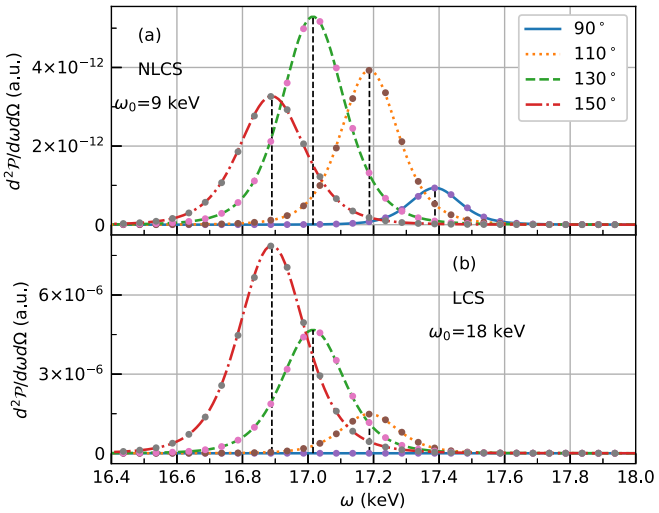

FIG. 1. Spectral profiles of the emitted power for NLCS (upper panel) and LCS (lower panel) by a hydrogenic atom with $Z = 1$. The IA results are represented with lines, and the exact ones are represented by symbols. The laser frequency ω_0 is 9 keV for NLCS and 18 keV for LCS. The values of the scattering angle θ are indicated in the legend [which is common to Figs. 1(a), 1(b), 2(a), and 2(b)]. Vertical dashed lines are drawn for the frequencies ω_m given by Eq. (23).

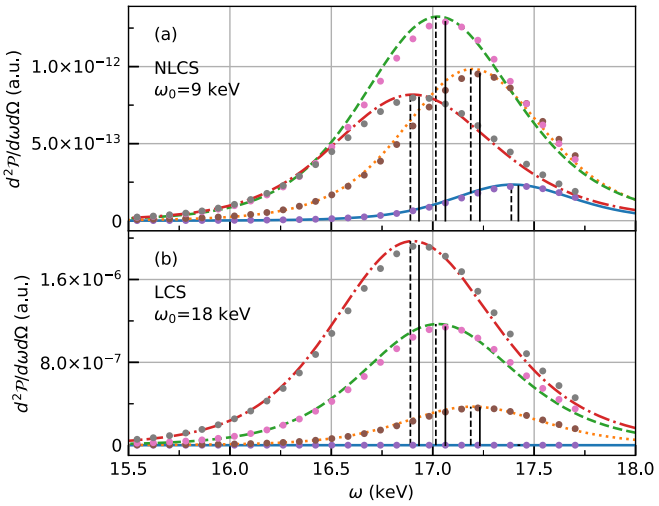

FIG. 2. Same as for Fig. 1, with $Z = 4$. The vertical solid lines correspond to the maxima for the exact profiles.

the IA or exact distributions. For the geometry (43) we have $(\mathbf{s}\cdot\mathbf{s}_0)^2 = \cos^2\theta$.

We note that for $\nu \ll 1$ the ratio of the functions (46) and (45) is close to 1 for frequencies around the Compton frequency (23), the latter being approximated in the nonrelativistic case by the solution of the equation $E(Q_m) = \Delta_m$ ($m = 1, 2$).

The distributions (47) and (41) differ by their orders in the parameter ξ, $\xi^2 \propto I$ for LCS and $\xi^4 \propto I^2$ for NLCS. The total power emitted in NLCS is then much smaller than in LCS for $\xi \ll 1$. They also differ by the angular factors they contain. In particular, the distribution (47) vanishes for emission along laser polarization ($\theta = 90°$). In contrast, Eq. (41) gives a finite result in this case, entirely due to the magnetic term $\widetilde{\mathbf{F}}_a^{(2)}$—this result is not affected in particular by the quality of the approximations leading to the term proportional to $(\mathbf{s}\cdot\mathbf{s}_0)$ in Eq. (42).

In the following we present some numerical results which, in our opinion, are of help to clarify in what measure the results known from the literature, experimental [16] or based on numerical simulations [15], can be understood using an approximate description like that behind Eqs. (41)–(47). We first describe and analyze comparatively the IA and exact spectral *and* angular distributions, for NLCS and LCS. At the same time and where possible, we discuss the relation of our findings with results presented in Refs. [15,16].

We choose to work in conditions which are similar to those used in Ref. [16]. In Figs. 1 and 2 the laser frequency ω_0 is 9 keV for NLCS, and twice this value for LCS—the frequencies are chosen for a comparison like that presented in Fig. 2(a) in Ref. [16]. The parameter ξ [Eq. (21)] is taken as 10^{-3}. In Figs. 1 and 2 we show IA results (with lines) and exact results (with symbols) for NLCS (upper panels) and LCS (lower panels) from a hydrogenic target with $Z = 1$ and 4, respectively. Four scattering directions satisfying Eq. (43) are considered—the values of the scattering angle are indicated in Fig. 1(a).

From Figs. 1 and 2 one sees that, for each scattering direction, the two spectral profiles, in IA and the exact one, are similar and almost coincide for $Z = 1$ (or $b = 1$ in a.u.)—we note, however, that the exact distribution ends at a frequency less than $2\omega_0$, the maximum frequency being imposed by the energy conservation at the threshold, $E_1 + \Delta_m = 0$.

The maxima of both profiles are shifted to red from the value $m\omega_0$ (18 keV). The frequency corresponding to the maximum of the IA profile for a given θ is well approximated by ω_m given by Eq. (23)—this is clearly seen in both figures, the vertical dashed lines being drawn at the frequencies ω_m. In Fig. 1 the peaks of the exact profiles are almost at the same frequencies, while in Fig. 2 (where they are indicated by solid vertical lines) they are shifted slightly to frequencies higher than ω_m—a blueshift of the same kind was found in Ref. [15] for NLCS simulated by TDSE calculations. The magnitude of the relative blueshift in Fig. 2 (for $Z = 4$) is greater in our case than in Ref. [15], apparently due to the potential considered (pure Coulomb here, "soft" Coulomb in Ref. [15]). Resuming the above discussion, the results indicate that the shift of a Compton peak (from $m\omega_0$, with $m = 1, 2$) is approximated in IA by $m\omega_0 - \omega_m$, and by this value plus a small blueshift depending on the parameter b (=Z in a.u.) for the exact profile. It increases in either case with the angle θ for both types of scattering, LCS and NLCS. For a given θ, the shifts for NLCS at a frequency ω_0 and LCS at $2\omega_0$ are practically the same. These findings clearly show that the impulse approximation (in the current version, above formulated) is not able to predict a supplementary redshift of the NLCS peaks of the kind observed in Ref. [16]. The same is true for the exact case since the blueshifts for NLCS and LCS are comparable (or negligible). We note, however, that the peak value of the NLCS distribution is finite at $\theta = 90°$, in accord to the persistence of the NLCS signal seen in Ref. [16] for photons emitted along the laser polarization. We also remark

that the increase with θ of the height of the NLCS peak, seen in Fig. 2(a) from Ref. [16] (on the interval 89°–131°), is qualitatively confirmed: the present calculations predict in fact that the peak height (regarded as a function of θ) has a maximum around $\theta = 130°$, where the last factor of Eq. (41) reaches its maximum value—the weak angular dependence of $J(Q_2, \Delta_2)$ does not change significantly the position of the maximum.

We might be inclined to assume that the absence of an additional redshift of the NLCS peaks in the present calculations is a consequence of the approximations made in the derivation of Eqs. (40) and (39), like for example [if one takes as starting point Eq. (24)] the neglect of the contribution of the terms involving the potential force or the approximate treatment of the intermediate states in the matrix element $\widetilde{\mathcal{M}}_{fi}^{(1)}$. However, it is rather unlikely that this is the right explanation if we take into account that the TDSE calculations of Ref. [15], not implying these approximations, are leading to the same (negative) conclusion about the redshift. We also remark that the agreement between IA and exact results for the examined distribution is similar in the two cases, NLCS and LCS; see also Refs. [20,21] for in-depth discussions of the subject in the context of the usual Compton scattering. Large deviations are expected to appear for NLCS, like in the LCS case [21], if the comparison would refer to distributions of the emitted photons *and* scattered electrons.

Besides the doubly differential distribution of the emitted radiation measured in the experiment [16], other types of distributions are also of interest. Reference [15] compares the TDSE calculated angular distributions (for LCS and NLCS) for initially *bound* electrons with predictions based on known analytical equations for *free* electrons, Klein-Nishina for LCS [32], and Brown-Kible for NLCS [2], the latter in the NR limit. The agreement obtained, remarkable for LCS and rather good for NLCS, is surprising in some measure since in the case of a FER the radiation spectrum is reduced to a Compton line, which is replaced by a Compton peak in the case of the bound electron. We extend below the comparison by the inclusion of the IA results, this allowing us to make plausible the explanation of the above-mentioned agreement as a consequence of the validity of IA in the present conditions. The comparison is done at the level of the cross sections of the processes. To obtain the differential *photon* cross section of a scattering process in the monochromatic case one divides the corresponding radiation power to the energy $\hbar\omega$ of the emitted photon and to the photon incident flux (the ratio $I/\hbar\omega_0$ of the laser intensity I to the incident photon energy $\hbar\omega_0$) [33]. In particular, the doubly differential (in frequency and emission direction) cross section (DDCS) is

$$\frac{d^2\sigma}{d\omega d\Omega} = \frac{\hbar\omega_0}{I}\frac{1}{\hbar\omega}\frac{d^2\mathcal{P}}{d\omega d\Omega} = \frac{\omega_0}{\omega}\frac{1}{I}\frac{d^2\mathcal{P}}{d\omega d\Omega}. \tag{48}$$

To express the various cross sections it is convenient to employ the parameter $\eta \equiv \xi/\sqrt{2}$ (frequently used in the literature) in place of ξ, and the classical radius of the electron $r_0 = e_0^2/m_e c^2$.

In Appendix C we consider in some detail the FER case—it is needed for comparison and is also useful to check some equations from Sec. III and from the current section.

Using the IA formulas (41) and (47), and the definition (48), we express the corresponding DDCS as follows:

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)^{(\mathrm{IA})}_{\mathrm{NLCS}} = \frac{\omega}{2\omega_0}\left(\frac{r_0\eta}{2}\right)^2 \hbar J^{(\mathrm{IA})}(Q_2, \Delta_2)\,(\mathbf{s}\cdot\mathbf{u}_0)^2 \tag{49}$$

for NLCS and

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)^{(\mathrm{IA})}_{\mathrm{LCS}} = \frac{\omega}{\omega_0}\, r_0^2\, \hbar J^{(\mathrm{IA})}(Q_1, \Delta_1)\,(\mathbf{s}\cdot\mathbf{s}_0)^2 \tag{50}$$

for LCS. One verifies that Eqs. (C1) and (C2) of the FER limit are recovered by observing in Eq. (45) the behavior

$$J^{(\mathrm{IA})}(Q_m, \Delta_m) \to \delta(E(Q_m) - \Delta_m), \quad m = 1, 2, \tag{51}$$

in the limit $b \to 0$ ($Z \to 0$).

The IA angular distributions are calculated integrating Eqs. (49) and (50) in ω on the interval corresponding to the NLCS or LCS peak (at fixed emission direction). It is useful to express them with functions defined by the relation

$$F_m(\omega_0, \theta) \equiv \int d\omega\, \frac{\omega}{m\omega_0}\, \hbar J^{(\mathrm{IA})}(Q_m, \Delta_m), \tag{52}$$

where the integral is computed over the scattering peak and $m = 1, 2$. Then

$$\left(\frac{d\sigma}{d\Omega}\right)^{(\mathrm{IA})}_{\mathrm{NLCS}} = F_2(\omega_0, \theta)\left(\frac{r_0\eta}{2}\right)^2 (\mathbf{s}\cdot\mathbf{u}_0)^2 \tag{53}$$

and

$$\left(\frac{d\sigma}{d\Omega}\right)^{(\mathrm{IA})}_{\mathrm{LCS}} = F_1(\omega_0, \theta)\, r_0^2\, (\mathbf{s}\cdot\mathbf{s}_0)^2. \tag{54}$$

The free-electron limit is easy to obtain: using Eq. (51) (valid for $b \to 0$) we have

$$F_m(\omega_0, \theta) \to \left(\frac{\omega_m}{m\omega_0}\right)^2, \tag{55}$$

yielding the first factors from Eqs. (C4) and (C5) of the FER case.

We compare numerically the angular distributions above defined, working in conditions chosen as in Ref. [15] (slightly different from those of Figs. 1 and 2), $\omega_0 = 9.25$ keV and a laser intensity I for which $\eta \approx 1.62 \times 10^{-3}$. In Fig. 3 are shown the results for $d\sigma/d\Omega$, for LCS in the upper panel and NLCS in the lower one. With lines are represented the IA results based on Eqs. (54) (for LCS) and (53) (for NLCS). The IA results are compared in Fig. 3(a) with those based on the Klein-Nishina formula, Eq. (C6) (represented by symbols), without or including the (less than 1) factor $(\omega_1/\omega_0)^2$. In Fig. 3(b) the comparison is made with the results calculated with the Brown-Kible formula, Eq. (C4), without or with the factor $(\omega_2/2\omega_0)^2$. In both panels there is a clear agreement between the results in IA and those for the free electron (both including the correction factors), this indicating that the behavior (55) extends beyond the strict limit of the free electron.

We now compare the above results with those represented in Figs. 8 and 9 of Ref. [15]. For LCS one sees that the results including the factor $(\omega/\omega_0)^2$ (line and circles) from Fig. 3(a) are practically the same (at visual level) with those in Fig. 8 from Ref. [15]. For the NLCS case one first observes

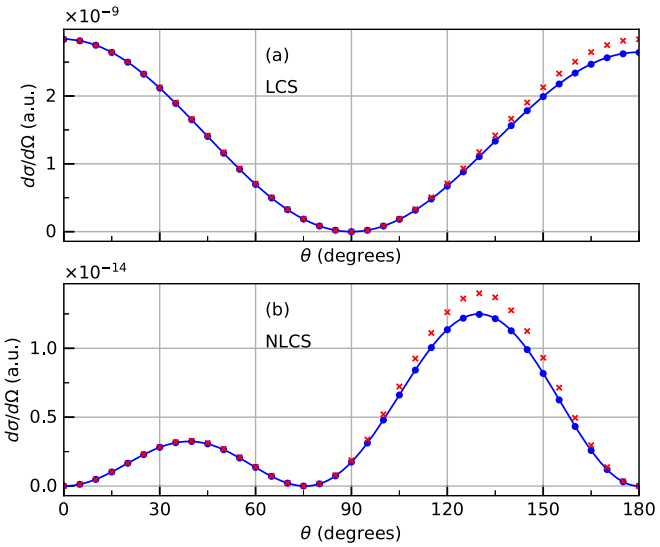

FIG. 3. Photon angular distributions for LCS (upper panel) and NLCS (lower panel). Lines represent the IA results for a ground-state hydrogenic atom with $Z = 1$, computed using Eq. (54) for LCS and Eq. (53) for NLCS. The results shown by symbols are based on Klein-Nishina (upper panel) and Brown-Kible (lower panel) formulas, Eqs. (C6) and (C4) (see other details in the text). The laser frequency ω_0 is taken to be 9.25 keV (≈340 a.u., the same value for both panels), and the field amplitude $E_0 = 107$ a.u. (the latter matters for NLCS only). The emitted photon direction is in the laser polarization-propagation plane $(\mathbf{s}_0, \mathbf{n}_0)$.

that the angular distributions from Fig. 3(b) and Fig. 9 of Ref. [15] are similar [34]. The discrepancy seen in this case in Ref. [15] between TDSE results and those for the free-electron case is likely to be caused *mainly* by the use of the Brown-Kible formula in the NR limit, without the correction factor $(\omega_2/2\omega_0)^2$ (this explanation for the discrepancy is mentioned also in Ref. [15] as possible)—around the dominant maximum of $d\sigma/d\Omega$ in Fig. 3(b) this factor is of the order of 10%, which is the same as the discrepancy level found in Ref. [15].

In light of the above findings, the agreement of the angular distributions for bound and free electrons seen in Ref. [15] gets a rather simple explanation if one assumes the validity of the impulse approximation at the level of DDCS—this explanation is strengthened by the similarity of the scattering profiles from Fig. 7 in Ref. [15] and the IA profiles from Figs. 1 and 2. However, a direct quantitative comparison of the TDSE and IA profiles makes sense solely if they are obtained in the same conditions and for the same atomic potential. It seems promising to follow this comparison versus the scattering angle θ, in particular the value $\theta = \pi/2$ could prove interesting. This case, not investigated in Refs. [14,15] but observed in Ref. [16], deserves some attention since, as discussed in the context of Eq. (34), NLCS is here mainly of magnetic origin in IA and the comparison with results based on TDSE would give an indication about the validity of the approximate treatment leading to this finding.

V. CONCLUSIONS

Two forms of the transition amplitude for the scattering of an intense laser beam from a simple atomic system are derived in the framework of the nonrelativistic quantum theory; differing by terms of the order β^2, these are equivalent in the NR approximation. The corresponding equations, (10) and (17), are nonperturbative with respect to the interaction electron-laser fields, involving exact TDSE solutions describing the evolution of the atomic system in the presence of the external electromagnetic field. The second form of TA, Eq. (17), deserves a special interest, it showing that the emission of radiation is possible, like in the classical case, for accelerated motions only. The procedure used for its derivation is suitable for relativistic generalization.

The application of the developed formalism, in the case of the nonlinear Compton scattering with the absorption of two photons from the incident laser beam, led us to nonrelativistic approximations for the distribution of the power of the emitted radiation, fully differential with respect to the attributes of the scattered photons and electrons; see Eqs. (38) and (39) for the scattering by a free-electron wave packet and, respectively, by a bound electron in a stationary state.

Formulas describing the spectral and angular distribution of the radiation (solely) scattered from a hydrogenic atom in the ground state are then presented—they are obtained either in impulse approximation or using exact descriptions of the initial and final Coulomb states of the electron. The numerical results obtained evaluating these formulas for conditions (regarding the incident laser and the scattering geometry) similar to those of the experiment [16] reproduce a good part part of the experimental observations. For the actual atomic system and the adopted approximations, we find that there is no redshift of the NLCS peak for a given laser frequency relative to the LCS peak for a double incident frequency. This finding disagrees with the main result of Ref. [16], remaining compatible with the results from Refs. [14,15]. Further investigations are required for the validation of the above considered approximations and for a better understanding of their impact on the properties of the nonlinear Compton peak. Finally we consider the angular distribution of the scattered radiation. After the derivation of the IA formulas for the corresponding NLCS and LCS cross sections, we examine their relation to the analog quantities in the case of a free electron initially at rest. The numerical results obtained for the angular distribution (including the correction factors depending of the frequencies) allow us to interpret the agreement observed in Ref. [15] between results for bound and free electrons as a consequence of the validity of the impulse approximation at the level of DDCS.

ACKNOWLEDGMENTS

The author thanks Viorica Florescu and Henri Bachau for their suggestions and comments. This work was supported by the Romanian Ministry of Education and Research, CNCS UEFISCDI, Grants No. PN-III-P4-ID-PCE-2016-0792 and No. PN-III-P4-ID-PCCF-2016-0164, within PNCDI III.

APPENDIX A: ALTERNATIVE EXPRESSIONS FOR THE TRANSITION AMPLITUDE

The exact relativistic formula for the classical amplitude [see Eqs. (14.64) and (14.65) in Ref. [1]] entering Eq. (1)

is

$$\mathcal{A}_{\rm cl}^{(\mathbf{n},\lambda)}(\omega) = -\mathbf{s}_\lambda^* \cdot \int_{-\infty}^{\infty} dt \times e^{i\omega(t-\mathbf{n}\cdot\mathbf{r}(t)/c)} \frac{\mathbf{n}\times[(\mathbf{n}-\boldsymbol{\beta}_{\rm cl})\times\dot{\boldsymbol{\beta}}_{\rm cl})]}{(1-\mathbf{n}\cdot\boldsymbol{\beta}_{\rm cl})^2}, \quad \text{(A1)}$$

its nonrelativistic approximation being given by Eq. (2). It is possible to express this amplitude [see also Eq. (14.67) from Ref. [1]] in a more compact form,

$$\mathcal{A}_{\rm cl}^{(\mathbf{n},\lambda)}(\omega) = -i\omega \int_{-\infty}^{\infty} dt\, e^{i\omega(t-\mathbf{n}\cdot\mathbf{r}(t)/c)}\, \mathbf{s}_\lambda^* \cdot \boldsymbol{\beta}_{\rm cl}, \quad \text{(A2)}$$

equivalent to Eq. (A1): one demonstrates the equivalence integrating by parts, using the identity $\frac{d}{dt}\, e^{i\omega(t-\mathbf{n}\cdot\mathbf{r}(t)/c)} = i\omega(1-\mathbf{n}\cdot\boldsymbol{\beta}_{\rm cl})\, e^{i\omega(t-\mathbf{n}\cdot\mathbf{r}(t)/c)}$ and the orthogonality relation $\mathbf{s}_\lambda^*\cdot\mathbf{n}=0$.

By analogy with the classical case, we are looking for an equivalent form of the quantum transition amplitude (10). As seen below, it is more advantageous to start from the symmetric formula (12), transcribed here in the form

$$\mathcal{A}_{i\to f} = -\frac{i\omega}{2}\int dt\, e^{i\omega t} \times \langle\psi_f'|e^{-i\mathbf{k}\cdot\mathbf{r}}\,\mathbf{s}^*\cdot\boldsymbol{\beta} + \mathbf{s}^*\cdot\boldsymbol{\beta}\, e^{-i\mathbf{k}\cdot\mathbf{r}}\,|\psi_i\rangle, \quad \text{(A3)}$$

involving the operator $\boldsymbol{\beta}$ defined by Eq. (3) and using the simplified notation $\mathbf{s}$ in place of $\mathbf{s}_\lambda(\mathbf{k})$.

In order to prepare the integration by parts of Eq. (A3) we first express the time derivative of the matrix element $\langle\psi_f'|O|\psi_i\rangle$, where O is a general operator (possibly time dependent) and the two states $|\psi_i\rangle$ and $|\psi_f'\rangle$ evolve in time according to the same TDSE, corresponding to the Hamiltonian $\mathcal{H}_{aL} = \boldsymbol{\Pi}^2/2m_e + V$. Consequently, we can write

$$\frac{d}{dt}\langle\psi_f'|O|\psi_i\rangle = \langle\psi_f'|\mathring{O}|\psi_i\rangle, \quad \text{(A4)}$$

where the symbol $\mathring{O}$ (O ring) is defined by

$$\mathring{O} = D(O) \equiv \frac{\partial O}{\partial t} + \frac{1}{i\hbar}[O,\mathcal{H}_{aL}]. \quad \text{(A5)}$$

The operator $\mathring{O}$, alternatively denoted $D(O)$ to indicate the result of an operation D done on O, has the usual properties of a derivative (for operators), in particular $D(O_1O_2) = \mathring{O}_1O_2 + O_1\mathring{O}_2$. Important examples are here the operators $\mathring{\boldsymbol{\Pi}}$ and $\mathring{\boldsymbol{\beta}}$, given by Eq. (14). In the case of the exponential from Eq. (A3) the calculation leads to the result

$$D(e^{-i\mathbf{k}\cdot\mathbf{r}}) = -\frac{i\omega}{2}(\mathbf{n}\cdot\boldsymbol{\beta}\, e^{-i\mathbf{k}\cdot\mathbf{r}} + e^{-i\mathbf{k}\cdot\mathbf{r}}\,\mathbf{n}\cdot\boldsymbol{\beta}). \quad \text{(A6)}$$

The partial integration in Eq. (A3) using the equality $i\omega e^{i\omega t} = de^{i\omega t}/dt$, the properties of $D(O)$, and Eq. (A6) gives the result

$$\mathcal{A}_{i\to f} = -\frac{i\omega}{4}\int dt\, e^{i\omega t}\langle\psi_f'|e^{-i\mathbf{k}\cdot\mathbf{r}}\,\Gamma + \Gamma\, e^{-i\mathbf{k}\cdot\mathbf{r}}\,|\psi_i\rangle + \frac{1}{2}\int dt e^{i\omega t}\langle\psi_f'|e^{-i\mathbf{k}\cdot\mathbf{r}}\,\mathbf{s}^*\cdot\mathring{\boldsymbol{\beta}} + \mathbf{s}^*\cdot\mathring{\boldsymbol{\beta}}\, e^{-i\mathbf{k}\cdot\mathbf{r}}\,|\psi_i\rangle, \quad \text{(A7)}$$

where $\Gamma \equiv (\mathbf{n}\cdot\boldsymbol{\beta})(\mathbf{s}^*\cdot\boldsymbol{\beta}) + (\mathbf{s}^*\cdot\boldsymbol{\beta})(\mathbf{n}\cdot\boldsymbol{\beta})$ is of the order β^2. Performing another partial integration, now for the first integral from this equation, and neglecting the terms originating from $D(e^{-i\mathbf{k}\cdot\mathbf{r}})$ [their contribution is the order β^2 relative to Eq. (A3)], one deduces the formula (we use a slightly different notation for TA)

$$\widetilde{\mathcal{A}}_{i\to f} = \frac{1}{2}\int dt\, e^{i\omega t}\langle\psi_f'|e^{-i\mathbf{k}\cdot\mathbf{r}}\,(\mathbf{s}^*\cdot\mathring{\boldsymbol{\beta}} + \mathring{\Gamma}/2) + (\mathbf{s}^*\cdot\mathring{\boldsymbol{\beta}} + \mathring{\Gamma}/2)\, e^{-i\mathbf{k}\cdot\mathbf{r}}\,|\psi_i\rangle. \quad \text{(A8)}$$

The operator $\mathring{\Gamma}$ can be expressed in terms of $\boldsymbol{\beta}$ and $\mathring{\boldsymbol{\beta}}$:

$$\mathring{\Gamma} = (\mathbf{n}\cdot\mathring{\boldsymbol{\beta}})(\mathbf{s}^*\cdot\boldsymbol{\beta}) + (\mathbf{s}^*\cdot\mathring{\boldsymbol{\beta}})(\mathbf{n}\cdot\boldsymbol{\beta}) + (\mathbf{n}\cdot\boldsymbol{\beta})(\mathbf{s}^*\cdot\mathring{\boldsymbol{\beta}}) + (\mathbf{s}^*\cdot\boldsymbol{\beta})(\mathbf{n}\cdot\mathring{\boldsymbol{\beta}}). \quad \text{(A9)}$$

Replacing it in Eq. (A8), then using Eq. (14) for $\mathring{\boldsymbol{\beta}}$, one finally obtains Eq. (17).

We remark here that other forms of the amplitude, equivalent to Eq. (17) in the nonrelativistic regime, can be found as above if one starts from Eq. (10) or the same equation with commuted operators inside the matrix element—these forms are more compact but less symmetric than Eq. (17).

APPENDIX B: DIFFERENTIAL DISTRIBUTIONS FOR LCS

For checking and reference purposes we collect here a part of the equations specific to LCS, analogous to those for NLCS in Sec. III. They are equivalent with formulas derived long ago, working in the so-called A^2 approximation [20]. One recovers simply these formulas if in Eq. (25) one neglects the contribution of the operator $\mathbf{s}^*\cdot\mathbf{P}$. The approximation of the matrix element describing LCS (proportional to A_0) is

$$\mathcal{M}_{fi} = \frac{i}{2}\frac{\omega}{\omega_0}\, eA_0\,\omega_0\,(\mathbf{s}^*\cdot\mathbf{s}_0) \times \left\langle\psi_f'^{(0)}\middle|e^{i\mathbf{Q}_1\cdot\mathbf{r}/\hbar}\middle|\psi_i^{(0)}\right\rangle e^{-i\omega_0 t}. \quad \text{(B1)}$$

We note that if we start from Eq. (27) and keep only the contribution of the electric force (dominant versus the other terms), $\widetilde{\mathbf{F}}^{(1)} = e\mathbf{E}$, we get the same result excepting the factor ω/ω_0—in order to recover this correction factor one has to consider the effect of the neglected terms.

Using the matrix elements (B1) and reasoning as in Sec. III we get the relations analogous to Eqs. (40) and (39), describing the power distribution of the emitted radiation in IA,

$$d^3\mathcal{P}_\lambda = \frac{\alpha}{8\pi}\left(\frac{\omega}{\omega_0}\right)^2 \xi^2\,(\hbar\omega_0)^2\,|\mathbf{s}_\lambda^*\cdot\mathbf{s}_0|^2\,|\langle\mathbf{p}|e^{i\mathbf{Q}_1\cdot\mathbf{r}/\hbar}|u_i\rangle|^2 \times \delta(E(\mathbf{p}) - E(\mathbf{p}-\mathbf{Q}_1) - \Delta_1)\, d\omega\, d\Omega\, d\mathbf{p}, \quad \text{(B2)}$$

and the exact power distribution:

$$d^3\mathcal{P}_\lambda = \frac{\alpha}{8\pi}\left(\frac{\omega}{\omega_0}\right)^2 \xi^2\,(\hbar\omega_0)^2\,|\mathbf{s}_\lambda^*\cdot\mathbf{s}_0|^2\,|\langle\mathbf{p}-|e^{i\mathbf{Q}_1\cdot\mathbf{r}/\hbar}|u_i\rangle|^2 \times \delta(E(\mathbf{p}) - E_i - \Delta_1)\, d\omega\, d\Omega\, d\mathbf{p}. \quad \text{(B3)}$$

The formula (B2) applies also in the case of a free electron, initially in an arbitrary state (a wave packet) $|u_i\rangle$—in this case the matrix element which it contains reduces to $\varphi(\mathbf{p}-\mathbf{Q}_1)$, with $\varphi(\mathbf{p}) \equiv \langle\mathbf{p}|u_i\rangle$.

APPENDIX C: CROSS SECTIONS FOR SCATTERING BY A FREE ELECTRON AT REST

In this Appendix we apply the formulas (38) and (B2) in the FER case, then we deduce the corresponding (double and single) differential cross sections. The distribution of the momentum $\mathbf{p}_0$ of a free electron having a definite momentum equal to zero is $\delta(\mathbf{p}_0)$. Then, setting $|\varphi(\mathbf{p}-\mathbf{Q}_2)|^2 = \delta(\mathbf{p}-\mathbf{Q}_2)$ in Eq. (38), integrating over the final momentum $\mathbf{p}$ of the electron, and using the definition (48), one obtains the result

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)^{(\mathrm{FER})}_{\mathrm{NLCS}} = \frac{\omega}{2\omega_0}\left(\frac{r_0\eta}{2}\right)^2 (\mathbf{s}\cdot\mathbf{u}_0)^2\, \hbar\,\delta(E(Q_2)-\Delta_2). \tag{C1}$$

Similarly, for linear Compton scattering, replacing in Eq. (B2) the squared matrix element from its right-hand side by $|\varphi(\mathbf{p}-\mathbf{Q}_1)|^2 = \delta(\mathbf{p}-\mathbf{Q}_1)$ and integrating over $\mathbf{p}$, one obtains

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)^{(\mathrm{FER})}_{\mathrm{LCS}} = \frac{\omega}{\omega_0}\, r_0^2\, (\mathbf{s}\cdot\mathbf{s}_0)^2\, \hbar\,\delta(E(Q_1)-\Delta_1). \tag{C2}$$

The argument of the function $\delta(E(Q_m)-\Delta_m)$ (with $m=1,2$) vanishes for the frequency $\widetilde{\omega}_m = \omega_m$ [see Eq. (23)], where the last equality is valid in NR approximation. In the same approximation

$$\hbar\,\delta(E(Q_m)-\Delta_m) = \frac{\omega_m}{m\omega_0}\,\delta(\omega-\omega_m). \tag{C3}$$

Then the integration in frequency is immediate and for the angular distributions we get the simple results

$$\left(\frac{d\sigma}{d\Omega}\right)^{(\mathrm{FER})}_{\mathrm{NLCS}} = \left(\frac{\omega_2}{2\omega_0}\right)^2\left(\frac{r_0\eta}{2}\right)^2 [\mathbf{s}\cdot\mathbf{n}_0 + 4(\mathbf{n}\cdot\mathbf{s}_0)(\mathbf{s}\cdot\mathbf{s}_0)]^2 \tag{C4}$$

and

$$\left(\frac{d\sigma}{d\Omega}\right)^{(\mathrm{FER})}_{\mathrm{LCS}} = \left(\frac{\omega_1}{\omega_0}\right)^2 r_0^2\,(\mathbf{s}\cdot\mathbf{s}_0)^2. \tag{C5}$$

We note that the result (C4) coincides, up to notations and if we set aside the first factor, with that given in Ref. [2], Eq. (3.44)—the latter was obtained for low laser intensities and in the NR limit of a semiclassical calculation. The more direct approach in Ref. [18] is leading in the so-called Thomson limit to a result [Eq. (5) in Ref. [18]] identical to Eq. (C4) for the actual scattering geometry (up to a factor, equal to the degree of coherence of the laser beam). It is worth mentioning that equations of the same form are obtained in classical theory: up to the factor depending on the frequencies, Eq. (C5) is the Thomson differential cross section while Eq. (C4) describes the classical angular distribution of the second harmonic (in the lowest order of η^2) [19].

Comparing now Eq. (C5) with the relativistic (Klein-Nishina) formula for LCS,

$$\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{KN}} = \frac{r_0^2}{4}\left(\frac{\omega_1}{\omega_0}\right)^2\left(\frac{\omega_1}{\omega_0}+\frac{\omega_0}{\omega_1}-2+4(\mathbf{s}\cdot\mathbf{s}_0)^2\right), \tag{C6}$$

one sees that they coincide in the NR domain—the expression from the last bracket in Eq. (C6) is equal to $4(\mathbf{s}\cdot\mathbf{s}_0)^2$ plus a negligible term of the order $O(\varkappa_0^2)$.

[1] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
[2] L. S. Brown and T. W. B. Kibble, Phys. Rev. **133**, A705 (1964).
[3] F. Ehlotzky, K. Krajewska, and J. Z. Kaminski, Rep. Prog. Phys. **72**, 046401 (2009).
[4] C. Harvey, T. Heinzl, and A. Ilderton, Phys. Rev. A **79**, 063407 (2009).
[5] M. Boca and V. Florescu, Phys. Rev. A **80**, 053403 (2009).
[6] F. Mackenroth and A. Di Piazza, Phys. Rev. A **83**, 032106 (2011).
[7] K. Krajewska and J. Z. Kaminski, Phys. Rev. A **85**, 062102 (2012).
[8] D. Seipt, S. G. Rykovanov, A. Surzhykov, and S. Fritzsche, Phys. Rev. A **91**, 033402 (2015).
[9] A. Angioi, F. Mackenroth, and A. Di Piazza, Phys. Rev. A **93**, 052102 (2016).
[10] F. Ehlotzky, J. Phys. B **20**, 2619 (1987).
[11] C. J. Joachain, N. J. Kylstra, and R. M. Potvliege, *Atoms in Intense Laser Fields* (Cambridge University, New York, 2012).
[12] B. Böning, W. Paufler, and S. Fritzsche, Phys. Rev. A **99**, 053404 (2019).
[13] E. Pisanty, D. D. Hickstein, B. R. Galloway, C. G. Durfee, H. C. Kapteyn, M. M. Murnane, and M. Ivanov, New J. Phys. **20**, 053036 (2018).
[14] D. Krebs, D. A. Reis, and R. Santra, Phys. Rev. A **99**, 022120 (2019).
[15] A. Venkatesh and F. Robicheaux, Phys. Rev. A **101**, 013409 (2020).
[16] M. Fuchs, M. Trigo, J. Chen *et al.*, Nat. Phys. **11**, 964 (2015).
[17] A. N. Hopersky, A. M. Nadolinsky, and S. A. Novikov, Phys. Rev. A **92**, 052709 (2015).
[18] E. Ugaz, Phys. Rev. A **50**, 34 (1994).
[19] E. S. Sarachik and G. T. Schappert, Phys. Rev. D **1**, 2738 (1970).
[20] P. Eisenberger and P. M. Platzman, Phys. Rev. A **2**, 415 (1970).
[21] R. H. Pratt, L. A. LaJohn, V. Florescu, T. Surić, B. K. Chatterjee, and S. C. Roy, Radiat. Phys. Chem. **79**, 124 (2010).
[22] The Schrödinger picture is used systematically throughout the paper.
[23] M. Førre and S. Selstø, Phys. Rev. A **101**, 063416 (2020).
[24] T. K. Lindblom, M. Førre, E. Lindroth, and S. Selstø, Phys. Rev. Lett. **121**, 253202 (2018).
[25] M. Førre, S. Selstø, J. P. Hansen, and L. B. Madsen, Phys. Rev. Lett. **95**, 043601 (2005).

[26] Equation (17) can be also considered for a fresh reevaluation of MND effects in HHG, a process for which the majority of existing calculations are based on the analysis of the induced dipole moment of the atomic system in the presence of the laser field [11].

[27] The short way to recover the equations which would be obtained applying directly the methods of the standard perturbation theory [see, for example, Ref. [35]] for scattering in the monochromatic case is to start from the expansion of Eq. (10) in the power series of the field amplitude.

[28] H. Bachau, M. Dondera, and V. Florescu, Phys. Rev. Lett. **112**, 073001 (2014).

[29] H. Bachau, M. Dondera, and V. Florescu, J. Mod. Opt. **63**, 402 (2016).

[30] The use of incoming states can be understood appealing to the general collision theory or, more directly, by extending the reasoning given in Ref. [31], from the case of photoionization to that of Compton ionization.

[31] M. Dondera, Phys. Rev. A **82**, 053419 (2010).

[32] O. Klein and Y. Nishina, Nature (London) **122**, 398 (1928).

[33] We notice that the cross sections so defined depend on the laser intensity I for processes implying the absorption of two or more laser photons—in order to obtain a quantity independent of I for the case of NLCS one can further divide Eq. (48) by I or by the photon incident flux; see, for instance, Ref. [35] for the definition of the generalized cross sections.

[34] They differ in particular by a scale factor originating from the different definitions adopted for the cross sections in Ref. [15] [Eqs. (20) and (21)] and in the present paper [Eq. (48)]; see also Ref. [33].

[35] F. H. M. Faisal, *Theory of Multiphoton Processes* (Springer, New York, 2013).