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NONLINEAR OPTICS

S-Matrix Approach to the Problem of High-Harmonic Generation in the Field of Intense Laser Wave

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Abstract—A method using expansion of the wave function in the basis of photonic and free atomic eigenstates is proposed for calculating the emission spectrum of an atom in a laser field. The wave function is constructed using the Kramers–Henneberger transformation so that the expression for the transition S matrix explicitly includes the nonlinear interaction with the laser field. The expansion coefficients are determined by the residual interaction, which depends on the coordinates of the classical free electron motion in the laser field. Resonances at the atomic transition frequencies explicitly arise in the emission spectrum when the residual interaction is considered in the first order. The numerical solution of the time-dependent Schrödinger equation for the hydrogen atom within the semiclassical approach is used to obtain emission spectra for laser pulses of different intensities and durations.

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1. INTRODUCTION

An interaction of high-intensity (10^{13} to $10^{15}\,\mathrm{W\,cm^{-2}}$) laser radiation with gaseous media gives rise to the high-order harmonic generation (HHG): up to the UV and soft X-ray ranges. This phenomenon has been analyzed in most of theoretical studies within the semiclassical approach, i.e., based on the time dependence of the mean atomic dipole moment, found by solving the time-dependent Schrödinger equation (TDSE) in the classical field of laser wave. Significant progress in the theoretical studies is related to the use of the strong-field approximation, which is based on the analytical method developed by Keldysh [1]. Various versions of the HHG theory have been constructed within this method, which adequately reproduced many experimental results [2, 3]. Results of the numerical solution of the TDSE for an atom in a strong laser field are used to verify the analytical methods [4–6] (see review [7]).

Recently researchers have considered problems where the semiclassical approach cannot be applied, in particular, because of the effect of the atomic resonances on the harmonic generation [8–13]. Radiation at atomic transition frequencies was observed along with harmonics in recent experiments [14]. Another practically important case is the HHG gain under conditions of multiphoton resonance with autoionization state [9–12], whose decay can be taken into account in the TDSE [12]. The limitations of the semiclassical approach to the description of HHG were discussed in detail in [15].

In this study, the spectral and polarization characteristics of HHG are formulated in terms of the S-matrix approach. The TDSE is solved using the Kramers–Henneberger transformation [16, 17], which is widely used in the theory of multiphoton processes [18–20]. The wave function (WF) of the transformed TDSE with the potential of residual interaction is expanded in the eigenstates of the free atom. As a result, the radiative transition S matrix explicitly includes nonlinear factors, which makes it possible to use approximate solutions for expansion coefficients. For comparison with the results of semiclassical approach, the fifth-harmonic emission spectra near the $2p$–$1s$ transition in the hydrogen atom are obtained by solving numerically the three-dimensional Schrödinger equation for laser pulses of different intensities and durations.

Atomic units ($e = m = \hbar = 1$ and speed of light $c \approx 137$) are used unless otherwise specified.

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2. THEORY

The evolution of the initial atomic state in a laser field is determined by the TDSE

$$i\frac{\partial\Psi}{\partial t} = \left[H_{\rm A} + V(t)\right]\Psi(t),$$
$$\Psi(t\to-\infty)\to\exp(-iE_it)\,\varphi_i. \qquad (1)$$

Here, E_i is the eigenvalue of the free-atom Hamiltonian $H_{\rm A}$ in the initial state φ_i and $V(t)$ is the operator of the atom–laser pulse interaction in the long-wavelength approximation:

$$V(t) = -\frac{1}{c}\mathbf{PA}(t) + \frac{1}{2c^2}\left|A(t)\right|^2, \qquad (2)$$

where $\mathbf{P}=\Sigma_j\mathbf{p}_j$ is the total electron momentum operator; $\mathbf{p}_j=-i\nabla_j$ is the momentum operator of the jth electron; and $\mathbf{A}(t)$ is the laser field vector potential, related to the electric field $\mathbf{E}(t)$ by the expression $\mathbf{E}(t)=-c^{-1}\partial\mathbf{A}/\partial t$.

Within the semiclassical approach the atomic emission spectrum is calculated as follows. First, the WF $\Psi(t)$ of an atom in an external laser field with equivalent interaction in the form of scalar potential $V(t)=-\mathbf{DE}(t)$ ($\mathbf{D}$ is the dipole moment operator of the atom) is found by solving Eq. (1). Then the mean atomic dipole moment $\mathbf{d}(t)=\langle\Psi(t)|\mathbf{D}|\Psi(t)\rangle$ is calculated. The spectrum of radiation with polarization $\mathbf{e}_\gamma$ is proportional [21] to $|\ddot{\mathbf{d}}_\omega\mathbf{e}_\gamma|^2$, where $\ddot{\mathbf{d}}_\omega$ is the Fourier transform for the second derivative of the dipole moment with respect to time, $\ddot{\mathbf{d}}(t)$.

Within the quantum-mechanical approach we consider the HHG as a process based on the spontaneous emission of an atom in a laser field. Following [22], we expand the space of quantum system states by including the quantized field states, which describe the spontaneously emitted photon. The total WF can be written as

$$\Psi(t) = \Psi_0(t)\,|0\rangle + \sum_\gamma \Psi_\gamma(t)\exp(-i\omega_\gamma t)\,|1_\gamma\rangle, \quad (3)$$

where only the vacuum field state $|0\rangle$ and the single-photon Fock states $|1_\gamma\rangle=a_\gamma^+|0\rangle$ are taken into account; a_γ^+ and a_γ are, respectively, the photon creation and annihilation operators in the state $\gamma\equiv\mathbf{k}\lambda$ with the wave vector $\mathbf{k}$ ($k=\omega_\gamma/c$) and helicity $\lambda=\pm1$ (summation over γ implies the integral over $\mathbf{k}$ and the sum over λ). The atomic component $\Psi_0(t)$ takes into account the nonradiative processes in the laser field (excitation, ionization), while the components $\Psi_\gamma(t)$ correspond to the spontaneous emission processes (elastic and Raman scattering of laser radiation and harmonic emission).

The Schrödinger equation for the WF of extended system (3) is constructed by replacing $H_{\rm A}$ in Eq. (1) with the unperturbed Hamiltonian of the system, $H_0=H_{\rm A}+\Sigma_\gamma\omega_\gamma a_\gamma^+a_\gamma$, and the interaction operator $V(t)$ with $V(t)+v$, where the interaction operator of the atom with the quantized field v in the Schrödinger representation (in the long-wavelength approximation) has the form

$$v = \sum_\gamma(v_\gamma a_\gamma + v_\gamma^+ a_\gamma^+), \quad v_\gamma = -c_\gamma\mathbf{Pe}_\gamma. \quad (4)$$

Here, $\mathbf{e}_\gamma$ is the unit polarization vector of the photon and $c_\gamma=1/(2\pi\omega_\gamma^{1/2})$.

Then, substituting (3) into (1) and projecting onto the photonic states, we arrive at a system of coupled equations for the atomic components of the total WF:

$$i\frac{\partial\Psi_0}{\partial t} = \left[H_{\rm A}+V(t)\right]\Psi_0 + \sum_\gamma\exp(-i\omega_\gamma t)\,v_\gamma\Psi_\gamma, \quad \Psi_0(t\to-\infty)=\exp(-iE_it)\,\varphi_i,$$
$$i\frac{\partial\Psi_\gamma}{\partial t} = \left[H_{\rm A}+V(t)\right]\Psi_\gamma + \exp(i\omega_\gamma t)\,v_\gamma^+\Psi_0, \quad \Psi_\gamma(t\to-\infty)=0. \qquad (5)$$

Within the quantum-mechanical approach, the probability amplitude for emitting a photon γ in the $i\to f$ atomic transition under the action of the laser field is determined by the general expression for the S matrix after substituting the WF (3):

$$S_{fi}(\gamma,L) = -i\int_{-\infty}^{\infty}\exp(iE_ft)\left[\langle\varphi_f|\,V(t)\,|\Psi_\gamma(t)\rangle + \exp(i\omega_\gamma t)\langle\varphi_f|\,v_\gamma^+\,|\Psi_0(t)\rangle\right]dt, \qquad (6)$$

where L is a set of laser field parameters (frequency ω, electric field strength E, unit polarization vector $\mathbf{e}$, and relative phase φ).

The components of the atomic WFs $\Psi_0(t)$ and $\Psi_\gamma(t)$ in (5) can be sought for using the unitary transformation [16, 17]

$$\Phi_{0(\gamma)}(t) = U(t)\,\Psi_{0(\gamma)}(t), \quad U(t) = \exp\bigl[-i\mathbf{P}\mathbf{r}_{\rm c}(t)\bigr]\exp\bigl[i\chi(t)\bigr], \tag{7}$$

where $r_{\rm c}(t) = \dfrac{1}{c}\int\limits_{-\infty}^{t}\mathbf{A}(t')\,dt'$ is the change in the classical coordinate of free electron in the laser field, $\chi(t) = \dfrac{1}{2}\int\limits_{-\infty}^{t}v_{\rm c}^2(t')\,dt'$, and $\mathbf{v}_{\rm c}(t) = \dot{\mathbf{r}}_{\rm c} = c^{-1}\mathbf{A}(t)$. The first (from the right) transformation on the right-hand side of (7) shifts the WF phase, and the second transformation is the shift of electron coordinates. As a result of these transformations of the free-atom Hamiltonian (with electrostatic interaction), we obtain the following relations from Eq. (5):

$$\begin{aligned} i\frac{\partial\Phi_0}{\partial t} &= \bigl[H_{\rm A} + W(t)\bigr]\Phi_0 + \sum_{\gamma}\exp(-i\omega_\gamma t)\,v_\gamma\Phi_\gamma, \quad \Phi_0(t\to-\infty) = \exp(-iE_it)\,\varphi_i, \\ i\frac{\partial\Phi_\gamma}{\partial t} &= \bigl[H_{\rm A} + W(t)\bigr]\Phi_\gamma + \exp(i\omega_\gamma t)\,v_\gamma^{+}\Phi_0, \qquad\qquad \Phi_\gamma(t\to-\infty) = 0, \end{aligned} \tag{8}$$

where the residual interaction for an atom with a nuclear charge Z has the form

$$W(t) = \sum_{j=1}^{Z}\left[\frac{Z}{r_j} - \frac{Z}{|\mathbf{r}_j - \mathbf{r}_{\rm c}(t)|}\right]. \tag{9}$$

If $\int\limits_{-\infty}^{\infty}\mathbf{A}(t)\,dt = 0$, $W(t)$ vanishes at $t\to\pm\infty$. On the other hand, W rapidly decreases with an increase in the distance between the electron and nucleus: $W\propto r_j^{-\kappa-1}$ ($\kappa\geqslant 1$) at $r_j > r_{\rm c}(t)$.

Using the expansion of $\Phi_{0(\gamma)}$ in the basis of eigenfunctions of $H_{\rm A}$,

$$\begin{aligned} \Phi_0(t) &= \textstyle\sum_n a_n(t)\exp(-iE_nt)\,\varphi_n, \;\; a_n(t\to-\infty) = \delta_{in}, \\ \Phi_\gamma(t) &= \textstyle\sum_n b_n(t)\exp(-iE_nt)\,\varphi_n, \;\; b_n(t\to-\infty) = 0, \end{aligned} \tag{10}$$

we obtain the following system of equations for the expansion coefficients from (8):

$$\begin{aligned} i\frac{da_n}{dt} &= \sum_m\exp(i\omega_{nm}t)\left[W_{nm}(t)\,a_m(t) + \sum_\gamma\exp(-i\omega_\gamma t)(v_\gamma)_{nm}\,b_m(t)\right], \\ i\frac{db_n}{dt} &= \sum_m\exp(i\omega_{nm}t)\bigl[W_{nm}(t)\,b_m(t) + \exp(i\omega_\gamma t)(v_\gamma^{+})_{nm}\,a_m(t)\bigr], \end{aligned} \tag{11}$$

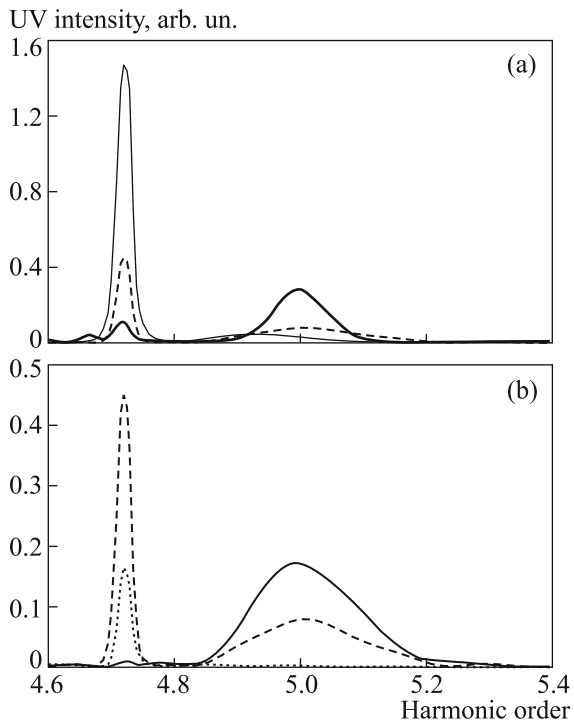
where $W_{nm} = \langle\varphi_n|W(t)|\varphi_m\rangle$ is the matrix element of the residual interaction (9) and $\omega_{nm} = E_n - E_m$ is the transition frequency.

Using the above-defined quantities, one can present expression (6) in the form

$$S_{fi}(\gamma, L) = -i\sum_n\int\limits_{-\infty}^{\infty}\exp(i\omega_{fn}t)\Bigl[V'_{fn}(t)\,b_n(t) + \exp(i\omega_\gamma t)\,V''_{fn}(t)\,a_n(t)\Bigr]dt, \tag{12}$$

where

$$V'_{nm}(t) = \Bigl\langle\varphi_n\Bigl|V(t)\,U^{+}(t)\Bigr|\varphi_m\Bigr\rangle, \qquad V''_{nm}(t) = \Bigl\langle\varphi_n\Bigl|v_\gamma^{+}\,U^{+}(t)\Bigr|\varphi_m\Bigr\rangle. \tag{13}$$



Spectra of the hydrogen atom in the vicinity of the fifth harmonic at a laser wavelength of 580 nm, calculated by solving numerically the Schrödinger equation: (a) laser-pulse peak intensity 3×10^{13} W cm^{-2} at full pulse durations $\tau = 6T_c$ (thin solid curve), $12T_c$ (dashed curve), and $24T_c$ (solid curve) and (b) laser pulse duration $\tau = 12T_c$ at intensities of 10^{13} (dotted curve), 3×10^{13} (dashed curve), and 5×10^{13} W cm^{-2} (solid curve).

3. RESULTS AND DISCUSSION

The above approaches are used below to calculate the emission spectrum of the hydrogen atom in an external laser field.

The method of numerical solution of the TDSE was described in detail in [23]. The electric field strength in the interval $0 \leqslant t \leqslant \tau$ is set as

$$E(t) = E_0 \sin^2 \frac{\pi t}{\tau} \cos \frac{2\pi t}{T_c},$$

where τ is the pulse duration, E_0 is the peak field strength, and T_c is the optical period.

To determine the contribution of the residual population to the emission spectrum, the calculations were performed on the time interval up to 48 fs, which is twice as large as the maximum of the pulse durations in use. To solve the TDSE on a spatial grid in our calculations, the Coulomb potential was smoothed at the origin of coordinates: $V_c = -1/\sqrt{r^2 + a^2}$, where $a = 0.4$. This smoothing slightly changes the energies of excited states.

The figure shows the results of numerical calculations for laser pulses of different durations and intensities in the spectral regions of the fifth harmonic and the $2p \to 1s$ transition. It can be seen that the ratio of the intensities of these lines depends strongly on the laser pulse parameters.

The emission intensity at the atomic transition frequency decreases with an increase in the pulse duration, which is in agreement with the result of the analytical theory [8], and disappears when the pulse duration greatly exceeds the optical period. In addition, the emission intensity decreases with an increase in the intensity in the pulse and disappears when passing to the tunnel ionization mode (i.e., when the Keldysh parameter [1] is smaller than unity). Thus, for a sufficiently long pulse and tunnel ionization, the residual population of the excited states is low and does not contribute to the emission spectrum. Under these conditions, the semiclassical approach does not lead to contradictions. For less intense pulses, containing several field periods, the residual population makes a significant contribution to the intensity of resonance lines. The semiclassical approach becomes inconsistent in this case.

Then we consider expression (12) for the S matrix in the case of the transition to the final ground state of the hydrogen atom ($f = i = 1\,\mathrm{s}$) with approximate solutions to (11).

In the first order of the residual interaction, the initial values of the expansion coefficients (10) are substituted into the right-hand side of (11):

$$\begin{aligned} a_n(t) &= \delta_{in} - i\int\limits_{-\infty}^{t} \exp(i\omega_{ni}t')\, W_{ni}(t')\, dt', \\ b_n(t) &= \frac{(v_\gamma^+)_{ni}}{\omega_{in} - \omega_\gamma} \exp\bigl[i(\omega_{ni} + \omega_\gamma)\, t\bigr]. \end{aligned} \tag{14}$$

Substituting expressions (14) into (12) and carrying out simple transformations, we arrive at

$$S_{ii}^{(1)}(\gamma, L) = -i\int\limits_{-\infty}^{\infty} \exp(i\omega_\gamma t)\left\{ V_{ii}''(t) + \sum_n \left[\frac{V_{in}'(t)(v_\gamma^+)_{ni}}{\omega_{in} - \omega_\gamma} + \frac{V_{in}''(t)\, W_{ni}(t)}{\omega_{ni} - \omega_\gamma} \right] \right\} dt. \tag{15}$$

Note that the singularity in the resonant term at $\omega_\gamma \to \omega_{ni}$ is eliminated by taking into account the radiative or ionization widths of the excited state.

Taking into account the dominant contribution of the resonance amplitude (15) in the vicinity of the $np{-}i$ transition frequency and using the momentum representation to calculate the matrix elements $V''_{nm}(t)$ in (13) and the multipole expansion for the residual interaction $W_{npm,1s}$ in (9), we obtain

$$S^{(1)}_{ii}(\gamma, L) = S^{(0)}_{ii}(\gamma, L) - i\frac{\gamma^n_{ii}(\gamma, L)}{\omega_{ni} - \omega_\gamma - (i/2)\Gamma_n}, \tag{16}$$

where

$$S^{(0)}_{ii}(\gamma, L) = -c_\gamma \int\limits_{-\infty}^{\infty} \exp\big[i\omega_\gamma t - i\chi(t)\big]\,(\mathbf{n}_\mathrm{c}(t)\,\mathbf{e}_\gamma)\,J^1_{ii}(r_\mathrm{c}(t))\,dt, \tag{17}$$

$$\gamma^{np}_{ii}(\gamma, L) = -\frac{c_\gamma}{3} \int\limits_{-\infty}^{\infty} \exp\big[i\omega_\gamma t - i\chi(t)\big](\mathbf{n}_\mathrm{c}(t)\,\mathbf{e}_\gamma)\Big[J^0_{i,np}(r_\mathrm{c}(t)) - 2J^2_{i,np}(r_\mathrm{c}(t))\Big]\,y^1_{i,np}(r_\mathrm{c}(t))\,dt. \tag{18}$$

Here, $\mathbf{n}_\mathrm{c} = \mathbf{r}_\mathrm{c}/r_\mathrm{c}$ and $J^\kappa_{nm} = \int\limits_0^\infty j_\kappa(pr_\mathrm{c})\,\varphi^*_n(p)\,\varphi_m(p)\,p^3 dp$, $j_\kappa(x)$ is the spherical Bessel function of order κ. The radial integral of the residual interaction $y^1_{1s2p}(r_\mathrm{c})$ in (18),

$$y^\kappa_{nl,n'l'}(r_\mathrm{c}) = \frac{1}{r_\mathrm{c}^{\kappa+1}} \int\limits_0^{r_\mathrm{c}} R_{nl}(r)\,r^\kappa R_{n'l'}(r)\,r^2\,dr + r_\mathrm{c}^\kappa \int\limits_{r_\mathrm{c}}^\infty R_{nl}(r)\,r^{-\kappa-1} R_{n'l'}(r)\,r^2\,dr,$$

is explicitly calculated for the hydrogen atom with apparent asymptotic behavior at $r_\mathrm{c} \to 0$ and $r_\mathrm{c} \to \infty$.

Thus, the emission probability amplitude in the first order of the residual interaction (15) is expressed in terms of the characteristics of classical free electron motion in an electromagnetic wave. Dependences on these characteristics (16) and (17) are determined by the quantum distributions of the coordinate and momentum in atomic states.

4. CONCLUSIONS

A method based on the S-matrix approach was proposed to calculate the emission spectrum of an atom in the field of an intense laser pulse. When the Kramers–Henneberger transformation is used for the WF, the S matrix of the photon explicitly includes a contribution of nonlinear processes and is determined by the expansion coefficients of the WF in the basis of free-atom stationary states in the potential of the residual atom–field interaction. In zero order of the residual interaction, the emission spectrum is completely determined by the electron momentum distribution in the initial state. In the first order, peaks at the frequencies of atomic transitions through excited states, including the autoionizing states of multielectron atoms, arise in the spectrum.

The emission spectra of the hydrogen atom for laser pulses of different intensities and durations were obtained in the semiclassical approach by solving numerically the Schrödinger equation. The semiclassical approach is applicable for long pulses in the range of resonant frequencies. For less intense or shorter pulses (several field periods), one should use more accurate methods, for example, the approach formulated in this study.

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