Basis of displaced Coulomb states in the problem of the generation of high-order harmonics

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This paper proposes a method for quantum-mechanically analyzing the dynamics of a hydrogenlike system, based on the representation of the electron wave function in a basis of displaced Coulomb states. This method can be used to construct an approximate model for the generation of the high harmonics of a strong light field by a hydrogenlike system. A plateau is present in the calculated spectra of the harmonics, with a high-frequency limit (when the width of the laser pulse is greater than 15–20 optical periods) in the region where the photon energy equals $I+3U$, where I is the ionization energy and U is the ponderomotive energy. If the laser pulse is shorter, a significant lengthening of the plateau occurs; the dependence of the high-frequency limit of the plateau on the pulsewidth is investigated. The generation of high-order harmonics in the field of a pulse with phase modulation (chirp) is also studied; these harmonics broaden somewhat and shift toward higher frequencies if the pulse is negatively chirped, and the harmonics shift toward lower frequencies if the pulse is positively chirped. Several possible mechanisms leading to this effect are discussed.
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1. INTRODUCTION

The generation of high-order harmonics, observed when intense laser radiation interacts with gases[1–4] and plasma,[5,6] is one of the most interesting nonlinear optical phenomena and is being actively studied in modern laser physics. An atom or an ion in the field of a high-intensity (greater than 10^{13} W/cm^2) electromagnetic wave interacts with it in an essentially nonlinear manner. In particular, it emits electromagnetic waves at frequencies that are multiples of the frequency of the incident radiation.

A plateau is observed in the spectrum, containing tens of odd harmonics and terminating in a rather sharp high-frequency limit. It was shown in the numerical experiments of Ref. 7 that this limit lies in the region of photon energies close to $I+3U$, where I is the ionization potential and U is the ponderomotive energy of an electron; consequently, the order of a generated harmonic cannot appreciably exceed

$$N_{\max}=\frac{I+3U}{\hbar\omega}, \tag{1.1}$$

where ω is the frequency of the external radiation.

On the whole, the experimental results agree with this conclusion, although it is difficult to check Eq. (1.1) exactly.

It is difficult to analytically study the process of generating high-order harmonics, mainly because perturbation theory cannot be used for calculating processes in an atom in a strong field. In this connection, several simplified models have been proposed for theoretically analyzing the generation of high-order harmonics. These include the semiclassical model,[8] the two-level system of Ref. 9 (which cannot be described by any potential in space), and quantum-mechanical models using various model potentials: a one-dimensional potential of the type $1/\sqrt{1+x^2}$,[10,11] a three-dimensional δ-function potential,[12] and several others. The plateau in the spectrum of the harmonics is, to all appearances, a fundamental property of a nonlinear system and is present in all the models enumerated above. Even when a one-dimensional classical anharmonic oscillator oscillates, there is a plateau in the Fourier components of its displacement.[13]

The case of a hydrogenlike system is important in practice and is considered in the analytical quantum-mechanical theories for the generation of high-order harmonics of Refs. 14 and 15. A serious disadvantage of these theories is that they cannot take into account the nonmonochromaticity of the external field (if it is not small), i.e., the finite width of the laser pulse, its chirp, or the presence in it of radiation of different wavelengths of comparable intensity. It is crucial to create theories that are free from these drawbacks; the practical importance of this is associated, in particular, with the fact that, in recent experiments[4] involving the use a short laser pulse, harmonics were observed with orders up to one and a half times that given by Eq. (1.1).

This paper proposes a method for quantum-mechanically analyzing the processes that occur in an atom, based on the representation of the electron wave function in a special basis—a basis of displaced Coulomb states. We should point out that the basis of displaced harmonic-oscillator states is used in quantum mechanics.[16] A basis of displaced Coulomb states can be used in calculating the dynamics of a hydrogenlike system in various problems. In this paper, it is used to study the generation of high-order harmonics. The proposed model of the generation of high-order harmonics is not absolutely rigorous but gives results close to those of Refs. 14 and 15 for the case of a monochromatic field. At the same time, it can be used to calculate the generation of high-order harmonics in the field of a short chirped pulse. The results agree with the experimental data of Ref. 4.

2. BASIS OF DISPLACED COULOMB STATES

This section describes a method for approximately solving Schrödinger's equation, using the basis of displaced Coulomb states.

Schrödinger's equation for the electron wave function $\Psi(\mathbf{r},t)$ in the one-electron approximation, neglecting relativistic and spin effects, is written as

$$i\dot{\Psi}(\mathbf{r},t)=\left(\left(\frac{\hat{\mathbf{p}}+c^{-1}\mathbf{A}(\mathbf{r},t)}{2}\right)^2+\hat{V}(\mathbf{r})\right)\Psi(\mathbf{r},t), \qquad (2.1)$$

where $\mathbf{A}(\mathbf{r},t)$ is the vector potential of the external electromagnetic field of the wave, $\hat{V}(\mathbf{r})$ is the potential of the nucleus (or of the atomic cone), and $\hat{\mathbf{p}}$ is the momentum operator. Here and below, we use the atomic system of units. The solution of the wave equation can be written as

$$\Psi(\mathbf{r},t)=\sum_{j=1}^{\infty} b_j(t)\varphi_j(\mathbf{r})+\int c(\mathbf{k},t)\chi(\mathbf{k},\mathbf{r})d\mathbf{k}, \qquad (2.2)$$

where $\varphi_j(\mathbf{r})$ and $\chi(\mathbf{k},\mathbf{r})$ are the eigenfunctions of some Hamiltonian. How rapidly the series in Eq. (2.2) converges for a specific problem depends on the choice of the basis $\{\varphi_j(\mathbf{r})\}$. The eigenfunctions of the unperturbed energy operator (the so-called Coulomb states) are usually chosen as basis functions.

We shall use the basis consisting of the Coulomb states centered at the position of a free classical electron in the field of the electromagnetic wave,

$$\varphi_j=\varphi_j(\mathbf{r}-\mathbf{R}),$$

where φ_j are the Coulomb states (subscript j is equivalent to the set of subscripts n, l, and m—the principal, orbital, and magnetic quantum numbers), and $\mathbf{R}$ is the position of the free electron:

$$\ddot{\mathbf{R}}(t)=-\mathbf{E}(t)=\frac{1}{c}\dot{\mathbf{A}}(t), \quad \mathbf{R}(\omega)=\frac{\mathbf{E}(\omega)}{\omega^2}; \qquad (2.3)$$

here $\mathbf{E}(t)$ and ω are the electric field and frequency of the external wave. The displaced Coulomb states are the eigenfunctions of the energy operator $p^2/2+\hat{V}(\mathbf{r}-\mathbf{R})$.

Let us substitute the expansion given by Eq. (2.2) into Eq. (2.1). After replacing $\rho=\mathbf{r}-\mathbf{R}$ and doing some manipulations, we get in the long-wavelength approximation

$$i\dot{b}_n(t)=\varepsilon_n b_n(t)+\sum_{j=1}^{\infty}\langle\varphi_n(\boldsymbol{\rho})|\hat{V}(\boldsymbol{\rho}+\mathbf{R}(t))$$
$$-\hat{V}(\boldsymbol{\rho})|\varphi_j(\boldsymbol{\rho})\rangle b_j(t)+\delta W_n(t), \qquad (2.4)$$

where ε_n is the energy of the (undisplaced) Coulomb state φ_n, and $\delta W_n(t)$ denotes a term that depends on $c(\mathbf{k},t)$. This system of (an infinite number of) integrodifferential equations, combined with a similar equation for $c(\mathbf{k},t)$, is equivalent to Schrödinger's equation, Eq. (2.1) and consequently is exact (in the long-wavelength approximation).

However, it is not possible to solve such a system, while retaining $\delta W_n(t)$. The main approximation used in this paper is that $\delta W_n(t)$ is neglected in Eq. (2.4). Below (for brevity) we call the system of displaced states of the discrete spectrum the basis. If the series in Eq. (2.2) converges rapidly enough, it can be cut off at a finite number of terms. After this, Eq. (2.4) reduces to a system of a finite number of ordinary differential equations,

$$i\dot{b}_n(t)=\varepsilon_n b_n(t)+\sum_{j=1}^{N}\langle\varphi_n(\boldsymbol{\rho})|\hat{V}(\boldsymbol{\rho}+\mathbf{R}(t))$$
$$-\hat{V}(\boldsymbol{\rho})|\varphi_j(\boldsymbol{\rho})\rangle b_j(t), \qquad (2.5)$$

which can be solved numerically. The conditions under which the approximations used here are justified are discussed at the end of this section.

Note that the action of the external field on the atom has been reduced to a time-dependent shift in the argument of the potential. The atom–field interaction is described in the same way in the Kramers coordinate system (see, for instance, Ref. 17); however, our approach has the advantage that the explicit form of the function $\varphi_j(\rho)$ is known from the very beginning, whereas it is difficult to obtain an analytical expression for the eigenfunctions of the Kramers–Henneberger potential for a hydrogenlike system.

To solve the system of Eqs. 2.5, it is necessary to compute the matrix elements

$$f_{n,l,m;n',l',m'}=\langle\varphi_{nlm}(\boldsymbol{\rho})|\hat{V}(\boldsymbol{\rho}+\mathbf{R})|\varphi_{n'l'm'}(\boldsymbol{\rho})\rangle. \qquad (2.6)$$

Below, the entire treatment is carried out for a hydrogenlike system in the field of a plane-polarized wave.

It is well known from the general theory[18] that quantities such as that given by Eq. (2.6) differ from zero only for $m=m'$. Below, it is assumed everywhere that $m=0$ holds, since this is the only value of the magnetic quantum number that is possible in the initial $1s$ state of the atom, from which the transitions to the excited states occur. There are no other selection rules for the matrix elements given by Eq. (2.6).

It is natural to carry out the integration in Eq. (2.6) in spherical coordinates $\{\rho,\theta,\varphi\}$, choosing the direction of $\mathbf{R}$ to be along the axis. We represent $V(\boldsymbol{\rho}+\mathbf{R})$ by the well-known expansion in Legendre polynomials $P_k(-\cos\theta)$:

$$-V(\boldsymbol{\rho}+\mathbf{R})=\frac{1}{|\boldsymbol{\rho}+\mathbf{R}|}$$
$$=\begin{cases}\dfrac{1}{R}\displaystyle\sum_{k=0}^{\infty}P_k(-\cos\theta)\left(\frac{\rho}{R}\right)^k, & \rho\leq R\\ \dfrac{1}{\rho}\displaystyle\sum_{k=0}^{\infty}P_k(-\cos\theta)\left(\frac{R}{\rho}\right)^k, & \rho>R.\end{cases}$$

The dependence of φ_{nl0} on θ is also expressed in terms of Legendre polynomials. Integration over θ in Eq. (2.6) results in the integrals

$$\int_{-\pi}^{\pi}P_l(\cos\theta)P_k(\cos\theta)P_{l'}(\cos\theta)\sin\theta\,d\theta.$$

This is a particular case of the so-called $3j$ symbols, whose values are given in Ref. 18. They differ from zero only when k satisfies the condition $|l-l'|\leq k\leq l+l'$ (the angular momentum-addition rule). Thus, only a finite number of terms remain in the sum over k in Eq. (2.6).

The radial part of φ_{nl0} is a generalized Laguerre polynomial multiplied by a power of ρ and an exponential of

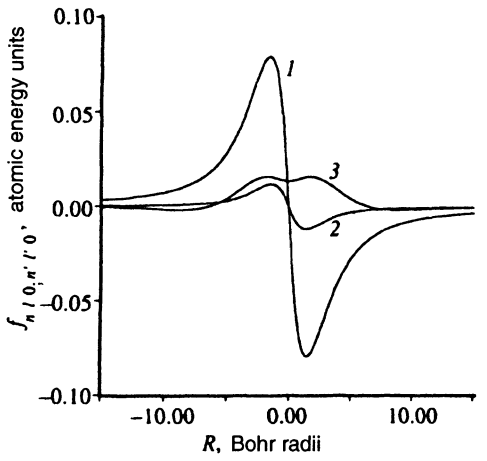

FIG. 1. Matrix elements given by Eq. (2.6); curve *1* corresponds to $n=1$, $l=0$; $n'=2$, $l'=1$. Curve *2* corresponds to $n=1$, $l=0$; $n'=7$, $l'=1$. Curve *3* corresponds to $n=2$, $l=1$; $n'=7$, $l'=1$.

ρ, and therefore the integral over the radial variable is found by integrating by parts. The final expression for the matrix elements of Eq. (2.6) is extremely cumbersome; the important thing is that it contains only finite sums, and therefore the calculation of the matrix elements by computer presents no fundamental difficulties.

Figure 1 presents some results of this calculation. It can be seen that the absolute values of the matrix elements corresponding to transitions to the upper levels are small. This means that the occupations $b_i b_i^*$ of these levels will increase slowly with time, and the series in Eq. (2.2) converges rather rapidly.

The matrix elements for small R, i.e., for high frequency (or) weak fields, are especially small [see Eq. (2.3)].

From a physical viewpoint, the expediency of using a basis of displaced Coulomb states in the case of high frequencies can be explained by the following considerations: It can be concluded from an analysis of the results of exact numerical experiments (for example, Ref. 19) that, in a high-frequency field, even when the mean electron energy exceeds the ionization energy, the electron continues to oscillate near the nucleus for many light periods. These oscillations are taken into account explicitly in the basis of displaced Coulomb states, and it is therefore obvious that it gives a better description of such an electron state than does the system of undisplaced states of the discrete spectrum.

If the external field is weak, one can use perturbation theory, the results of which are described in detail in the literature. This makes it possible to use the case of a weak field in testing the numerical calculations in models that use a basis of displaced Coulomb states.

If the initial state of the atom is strongly excited, our approximations are justified for fairly strong and low-frequency fields. Actually, the matrix elements of transitions from the upper states are small for any R; the continuous spectrum of the basis of displaced Coulomb states can be neglected because an electron in a strongly excited state is far from the nucleus and is almost free; consequently, the probability that it will absorb a photon and be ionized is very small.

If the electron was in the ground state before the field was switched on, and the field is strong and has a low frequency, calculations in which the states of the continuous spectrum of the basis of displaced Coulomb states are neglected can be regarded only as model calculations. However, this case is important in practice in the problem of the generation of high-order harmonics; therefore, the next section chiefly relates to intense, low-frequency external fields.

3. GENERATING THE HIGH-ORDER HARMONICS OF A HYDROGENLIKE SYSTEM

This section is devoted to a description of one possible application of the basis of displaced Coulomb states—calculation of the radiation of a hydrogenlike system in the field of an electromagnetic wave.

As is well known, the radiation intensity (in the dipole approximation) is proportional to $\ddot{d}(t)^2$, where $d(t)$ is the dipole moment of the atom:

$$d(t)=-\langle\Psi(\mathbf{r},t)|r\cos\vartheta|\Psi(\mathbf{r},t)\rangle,$$

here ϑ is the angle between $\mathbf{r}$ and the z axis, directed along the external field. After we expand $\Psi(\mathbf{r},t)$ in the basis of displaced Coulomb states, we get

$$d(t)=-R(t)-\sum_{k,j} b_k^*(t)b_j(t)\langle\varphi_k(\boldsymbol{\rho})|\rho\cos\theta|\varphi_j(\boldsymbol{\rho})\rangle. \tag{3.1}$$

The matrix elements appearing in this expression,

$$z_{kj}=\langle\varphi_k(\boldsymbol{\rho})|\rho\cos\theta|\varphi_j(\boldsymbol{\rho})\rangle,$$

are calculated analogously to Eq. (2.6).

In order to compute $\ddot{d}_\omega$, it is sufficient to find the spectrum d_ω of the quantity $d(t)$, since $\ddot{d}_\omega=-\omega^2 d_\omega$. However, by using Ehrenfest's theorems, it is immediately possible to find the spectrum of the acceleration $\ddot{d}_\omega$ or the velocity $\dot{d}_\omega$ of the electron. From Ehrenfest's second theorem, $\langle\ddot{z}\rangle=-\langle E\rangle$, where $\mathbf{E}$ is the total field acting on the electron: $\mathbf{E}=\mathbf{E}_{\text{wave}}+\mathbf{E}_{\text{atom}}$, we get

$$\ddot{d}(t)=E_{\text{wave}}+\left\langle\Psi(\mathbf{r},t)\left|\nabla_{\mathbf{r}}\left(\frac{1}{r}\right)\right|\Psi(\mathbf{r},t)\right\rangle=-\ddot{R}(t)$$

$$-\sum_{k,j} b_k^*(t)b_j(t)\nabla_R f_{nl0;n'l'0}(R). \tag{3.2}$$

The derivative of the matrix elements given by Eq. (2.6) with respect to R in this equation can be calculated numerically.

The first derivative of the dipole moment can be computed without the help of numerical differentiation. In fact, using Ehrenfest's first theorem, $\langle\dot{z}\rangle=\langle p_z\rangle+A_z/c$, we get

$$\dot{d}(t)=-\dot{R}(t)-i\sum_{k,j} b_k^*(t)b_j(t)z_{kj}(\varepsilon_k-\varepsilon_j), \tag{3.3}$$

where ε_k and ε_j are the energies of the kth and jth states.

Since the motion of the electron is calculated approximately, the values of $\omega^2 d_\omega$, $\omega\dot{d}_\omega$, and $\ddot{d}_\omega$ may not coincide.

A comparison of these values can be used to evaluate the correctness of the approximations that have been made.

The general procedure of a numerical experiment is as follows: The system of the first N differential equations from Eq. (2.5) was solved numerically by a fourth-order Runge–Kutta method over several tens of optical periods. The number N was chosen so that including additional equations no longer changed the results substantially, and ranged from 21 to 34. Before the beginning of the pulse, the electron is in the $1s$ state. The external field amplitude either attained its maximum value in about the first 20–30 optical periods and then remained constant (a "long" pulse) or smoothly increased and then decreased (a "short" pulse). The spectrum of the harmonics was calculated in the long-pulse case over several optical periods in the region where the field amplitude was established; for a short pulse, the spectrum of the harmonics was calculated over the entire duration of the pulse.

The correctness of the numerical calculations was checked by means of several test problems. If the external field is weak and its frequency is close to that of the transition between some two levels, the atom can be regarded as a two-level system, the dynamics of which are well known; the results of numerical calculations were extremely close to the latter. Another test problem is the generation of harmonics in a weak field, which can be studied by means of perturbation theory. In this case, the amplitude of the nth harmonic is proportional to E^n, where E is the amplitude of the external field. Our numerical calculations agree with this conclusion.

The closeness of the spectra of the harmonics calculated in terms of the various derivatives of the dipole moment is not only a criterion of the applicability of the approximations that are used but is also an effective test of the numerical calculations, since the population of each state makes different contributions to the right-hand sides of Eqs. (3.1), (3.2), and (3.3). In our computations, all three spectra were close. The $\omega\dot{d}_\omega$ spectrum was the least noisy; below, all the results are given for this spectrum.

The specification of a smooth shape for the turning-on of the external field was important for obtaining low-noise spectra of the harmonics. In the case of a long pulse, the field amplitude increased according to the law

$$E(t)=E_0\left(1-\frac{1}{\cosh(t^2/\tau^2)}\right),$$

where τ is the duration of the leading edge, and t varies from 0 to several times τ. In a short pulse, the field amplitude varied according to the law

$$E(t)=E_0\exp(-t^2/\tau^2),$$

where t varies from minus several times τ to plus several times τ.

Even with this smooth shape of the turning-on of the external field and with the maximum accuracy of the numerical calculation, some noise is present in the spectrum. Its level can be reduced by averaging over several (complex) spectra. In the case of a long pulse, these spectra are taken in different time intervals. In the case of a short pulse, the spectra were calculated for peak amplitudes of the field that differed slightly (by several percent) and were then averaged.

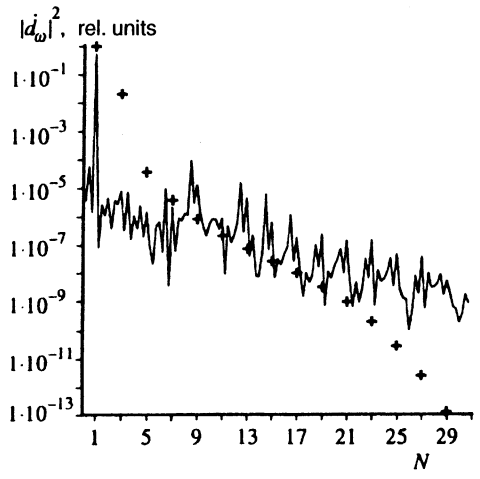

FIG. 2. Spectrum of the harmonics generated in a field with an intensity of 8×10^{13} W/cm^2 and a quantum energy of $0.1I$. The solid curve shows the calculation in a model that uses a basis of displaced Coulomb states. The crosses show the calculation in the ionization model.

The resulting spectra of the harmonics possess a plateau with a high-frequency limit whose position in the case of a long pulse is well described by Eq. (1.1). Figure 2 shows the spectrum calculated for $\hbar\omega=I/10$ and $U=0.45I$ (I is the ionization potential of the atom). The harmonics with $N<10$ are hard to distinguish, since the spectrum is affected by the resonance frequencies of the atom; the harmonics with higher numbers are easy to distinguish, and their intensities decrease appreciably beginning with $N=25$ (the high-frequency limit of the plateau). The same figure shows the spectrum of the harmonics obtained in the "ionization" theory of Ref. 15. The closeness of the results is evidence that the described model is applicable for fields with a ponderomotive energy at least up to $I/2$ and a quantum energy greater than $I/10$.

In the spectrum of the harmonics generated by a short pulse, the high-frequency limit of the plateau shifts toward higher frequencies and becomes less sharp. If the position of this limit is described by the equation

$$N_{\max}=\frac{I+\alpha(\tau)U}{\hbar\omega},$$

α varies from 3 for large τ to 5–7 for small τ. These results qualitatively agree with the experimental data of Ref. 4. It can be seen from Fig. 3 that the minimum pulsewidth at which a theory of the type of Refs. 14 and 15 is still applicable (i.e., the external field amplitude can still be considered slowly varying) is about 20 optical periods.

Reference 4 proposed a possible explanation for the displacement of the high-frequency limit of the plateau: An atom in a laser field with a rapidly increasing intensity (the leading edge of a short pulse) cannot become ionized at those intensities at which it is ionized when the field increases slowly; when it is ionized at higher intensities, the atom emits harmonics with higher numbers. In our model, the escape of an electron to the upper states can be regarded

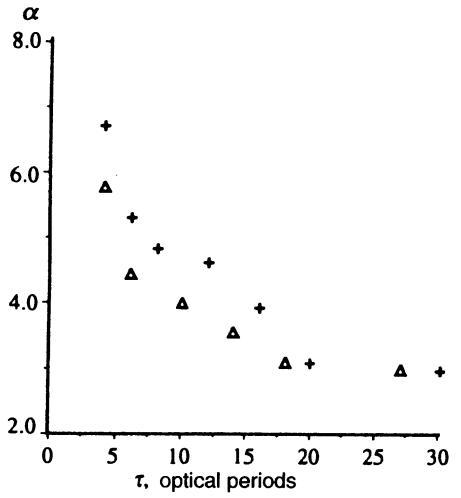

FIG. 3. Length of the plateau vs the pulse duration. The triangles are for an external field with $U=0.11I$ and $\hbar\omega=I/20$.. The crosses are for an external field with $U=0.45I$ and $\hbar\omega=I/10$.

as an analog of ionization. Figure 4 shows the dependence of the occupation $|b_1(t)|^2$ of the first state on the field amplitude $E(t)$ for three different pulse rise times τ. The points on the figure represent the occupations at the instant at which $R=0$, i.e., when the displaced states coincide with the usual states; for clarity, the points on the figure are connected with line segments. The results shown in Fig. 4 support the assumption made above concerning the character of the ionization of the atom when the laser field increases rapidly.

In the model described here, it is possible to study the generation of high-order harmonics in the field of a short, chirped pulse. Figure 5 shows the results of a calculation in a field with $U=0.25I$ and $\hbar\omega=I/5$ (the high-frequency limit of the plateau is $N=9$) for a chirp given by

$$T\frac{d\omega}{dt}=0.003\omega$$

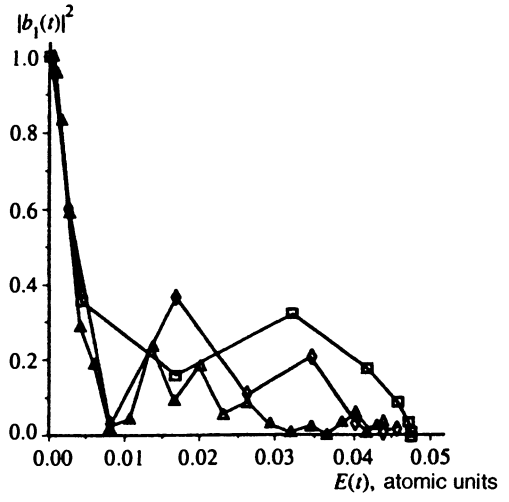

FIG. 4. Occupation of the $1s$ state vs the external field for three different pulse rise times. The squares correspond to $\tau=6$ optical periods. The rhombuses correspond to $\tau=10$ optical periods. The triangles correspond to $\tau=20$ optical periods.

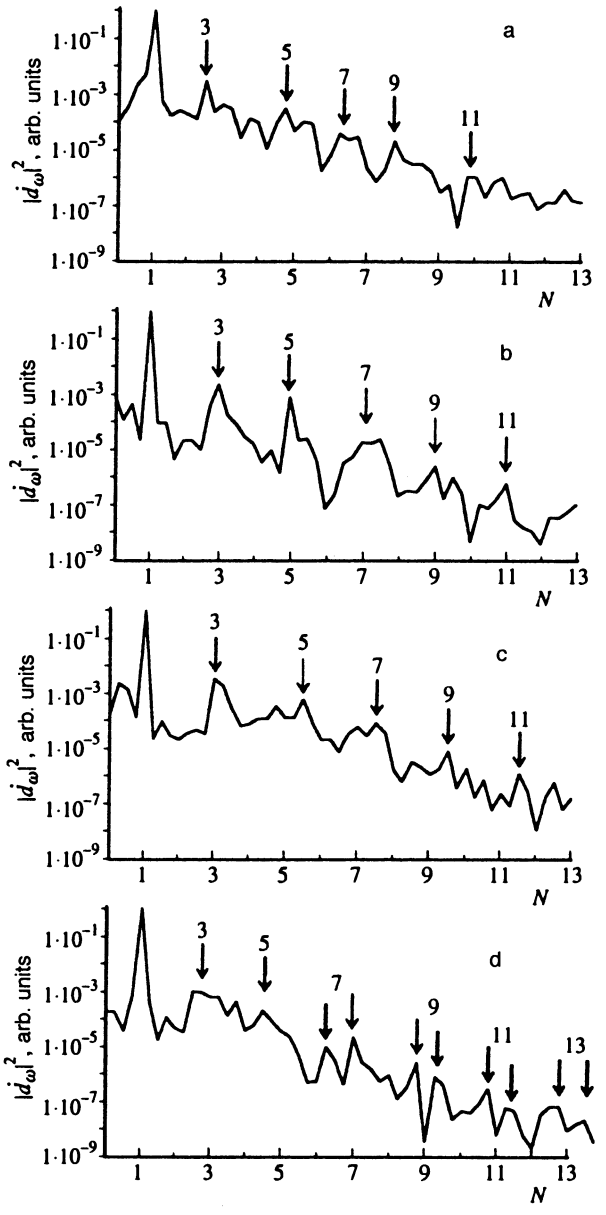

FIG. 5. Spectrum of the harmonics for various chirps of the laser pulse: (a) positive chirp, (b) pulse without chirp. (c) negative chirp, (d) frequency-modulated pulse.

(T is the optical period) and a pulsewidth of $\tau=32T$. Although the spectra are very noisy, the qualitative features noted in Ref. 4 can be seen in them: when the chirp is positive, the harmonics shift toward lower frequencies; when the chirp is negative, the harmonics shift toward higher frequencies, and their half-widths are greater than in the absence of chirp. The dependence of the frequency shift of the harmonics on the chirp of the laser pulse can be used to tune a source of coherent short-wavelength radiation.

Several mechanisms are possible by which pulse chirp affects the frequency and halfwidth of the emitted harmonics. Reference 4 makes the assumption that, in the field of a negatively chirped laser pulse, an atom is ionized more slowly than in the absence of chirp or with a positive chirp and, consequently, a wider range of frequencies of the external field is delayed before the atom is ionized, which also

explains the broadening of the harmonics. In our opinion, the retardation of the ionization in the case of negative chirp can be associated with the fact that the presence of chirp introduces asymmetry into the pulse of ponderomotive energy [and into the pulse of R, see Eq. (2.3)]; its leading edge becomes more sloped for negative chirp and becomes steeper for positive chirp; therefore ionization occurs later in the case of negative chirp.

Another possible mechanism is associated with the fact that the phase of a harmonic depends on the intensity of the external field.[20] Therefore, a harmonic emitted even by an unchirped pulse is chirped. Depending on whether this chirp is compensated by the chirp of the external field or is added to it, these features can be present in the spectrum of the harmonics emitted by a chirped pulse.

The behavior of high-order harmonics generated in the field of a chirped pulse is possibly associated chiefly with the frequency variation of the field within one optical period. This mechanism can be deduced by means of a semiclassical model of the generation of high-order harmonics: in the presence of chirp, the durations of the half-period during which the field tears the electron away from the nucleus and of the half-period when the field returns it to the nucleus and recombination occurs are different. This (very small) difference causes the recombination to occur differently (in particular, earlier or later) than it does when these two half-periods are equal. This can cause a certain displacement and broadening of the harmonics emitted by an electron during multiple ionization and recombination.

To test this assumption, we calculated the spectrum of the harmonics generated in the field of a frequency-modulated pulse with a modulation period of eight optical periods (Fig. 5d). The mean value of $|d\omega/dt|$ was chosen to be the same as in the pulses for which the spectra of the harmonics in Figs. 5a and 5c were recorded. Since the frequency-modulation depth is very small, the first two mechanisms discussed above for displacing and broadening the harmonics are not important in this case. At the same time, it can be seen that the spectra of the harmonics generated in a pulse without a chirp (Fig. 5b) and in a frequency-modulated pulse (Fig. 5d) differ strongly. In particular, the characteristic splitting of harmonics 7, 9, 11, and 13 is traced in the spectrum of the harmonics in Fig. 5d. It is apparently associated with the fact that, in the time interval in which the frequency increases, harmonics are generated that are displaced toward lower frequencies, whereas, when the frequency decreases, the harmonics are displaced towards higher frequencies. This displacement is close to the frequency displacement in the field of a chirped pulse (Figs. 5a and 5c). The displacement of the harmonics can thus be determined by the rate of frequency variation and not by the frequency variation during the entire pulse.

To all appearances, different mechanisms for the displacement and broadening of the harmonics by the chirp of a pulse can make different contributions, depending on the frequency and intensity of the external field, the pulse duration, and the number of the harmonic.

We should point out that these results do not exhaust all the possibilities of the model. In particular, the model makes it possible to study the generation of high-order harmonics in a bichromatic field. Experiments with such fields are described in Ref. 21.

4. DISCUSSION OF THE RESULTS; CONCLUSION

As mentioned above, the neglect of the continuous spectrum of the basis of displaced Coulomb states when the low-frequency external field is strong cannot be justified rigorously. This can be partially deduced from the fact that the functions of the discrete spectrum of the basis of displaced Coulomb states describe states (of the undisplaced basis) with an energy that is both larger and smaller than the ionization energy; i.e., they (partially) describe the electron states in the continuous spectrum of the undisplaced basis. From a mathematical viewpoint, this means that the subspace of functions $\{\varphi_j(\mathbf{r}-\mathbf{R})\}$ overlaps the subspace $\chi(\mathbf{k},\mathbf{r})$. Thus, electron ionization is partially taken into account in our model.

Comparison of the results with the results of the theory of Ref. 15, which explicitly takes ionization into account, has essential significance for the foundation of this model. A representation of the electron wave function in a complete basis of plane waves is used in a the ionization model. However, this can only be solved in a first approximation. The model described in this paper, on the other hand, uses an incomplete basis of discrete, displaced Coulomb states, but the solution of the system of Eqs. 2.5 is obtained exactly. In this connection, the ionization model and the model that uses a basis of the displaced Coulomb states in a certain sense complement each other.

Thus, this paper has introduced a basis of displaced Coulomb states that can be used in various problems involving the dynamics of a hydrogenlike system in an external field. Using this basis, an approximate model has been constructed for generating the high-order harmonics of a hydrogenlike system. The model has been used to study the generation of harmonics in the field of a short pulse, and it has been shown that, for a pulse that lasts less than 15–20 optical periods, harmonics are efficiently generated with orders that significantly exceed the high-order limit of the plateau in the spectrum of harmonics generated in the field of a pulse of longer duration. This effect is associated with nature of the ionization of an atom when the external field amplitude grows rapidly. The generation of high-order harmonics in the field of a chirped pulse has been studied; the harmonics emitted in the field of a negatively chirped pulse are somewhat broadened and are shifted toward higher frequencies, whereas the harmonics are shifted to lower frequencies in a field of a positively chirped pulse. Different mechanisms resulting in this effect have been considered.

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