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Nonperturbative theory of harmonic generation in helium under a high-intensity laser field: The role of intermediate resonance and of the ion

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The time-dependent Schrödinger equation for the helium atom with a model potential is solved on a numerical grid. The effects of intermediate-state resonance and of the ionization of the atom upon the harmonic generation are investigated. We also evaluate the contribution of the helium ion when the laser intensity is sufficiently high for most of the atoms to be ionized during the laser pulse. We find that the harmonics with order higher than 13 are due to the ion when the photon energy is 5.0 eV, while for photon energy 2.0 eV the atom produces up to about the 49th harmonic.

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Recent experiments [1,2] have demonstrated the generation of high-order harmonics in helium; up to the 23rd with pump photons [1] of 5.0 eV and up to the 41st with pump photons [2] of 2.0 eV. The peak intensities were given as 2×10^{17} and 1.4×10^{14} W/cm^2, respectively. Since, as we show below, even with a very short pulse the helium atom is totally ionized at the intensity of 10^{16} W/cm^2, we need to assess the contribution of the helium ion; otherwise the origin of the high harmonics would remain unclear. In earlier experiments, Perry *et al.* [3] studied the enhancement of multiphoton ionization due to intermediate-state resonances, while recent studies by L'Huillier, Balcou, and Lompré [4] have suggested the influence of resonance effects in the generation of high harmonics.

The theory of these processes has been led by Kulander [5] and collaborators who have shown how to employ time-dependent Hartree-Fock (TDHF) techniques for the calculation of the wave function of the atom under the influence of the field in the single-active-electron (SAE) approximation. Similar integration schemes have also been employed by others [6,7], while alternative techniques employing basis sets, and in particular B splines have been developed by Tang, Rudolph, and Lambropoulos [8]. Although their application to the nonperturbative calculation of ionization has proven quite successful, it remains to be seen how far they can go in the calculation of harmonic generation beyond perturbation theory [9].

Through the use of such basis sets, however, Tang, Rudolph, and Lambropoulos [10] have been able to demonstrate recently that for a range of photon energies up to at least 10 eV or higher, the ionization and excitation of helium is described very accurately in the SAE approximation. In this paper we implement a SAE description through a model potential on the basis of which we solve the time-dependent Schrödinger equation (TDSE) directly by converting the differential equation to a difference equation on a grid [5]. Using the time-dependent wave function, we can then calculate the time-dependent dipole moment from the Fourier transform of which we obtain the photon-emission spectrum exhibiting the harmonics. Our purpose here is to produce spectra for the frequencies and the range of intensities presently employed in experiments, to obtain the limits of useful intensities as determined by the ionization of the atom and to evaluate the role of He^+ in the generation of harmonics at intensities beyond which the neutral atom no longer exists.

The essential steps of the technique for the solution of the TDSE will be presented elsewhere [11]. As a brief reminder and summary of the steps necessary for the problem at hand, we recall that we solve the equation

$$\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = -iH(t)\Psi(\mathbf{r},t) \tag{1}$$

subject to the initial condition $\Psi(\mathbf{r},0)=|g(\mathbf{r})\rangle$, with $|g\rangle$ being the ground state of helium. The TD Hamiltonian is

$$H(t)=p^2/2+V_m(r)+h\ , \tag{2}$$

where, within the dipole approximation, the interaction h is written as $h=\epsilon\cdot\mathbf{r}E(t)\sin\omega t$, with ϵ being the unit polarization vector of the radiation, ω its frequency, and $\mathbf{r}$ the position operator of the electron. $V_m(r)$ is the model potential of the helium atom that we adopt to simplify the two-electron system. An imaginary potential [5] located at the boundary of the grid (300 a.u.) is used to absorb the wave function which reaches the boundary. The amplitude of the field ramps from zero to its maximum value linearly in the first 10 optical cycles, and then holds constant. The total propagation time of the wave packet is chosen to be 32 optical cycles. The wave function is expanded in an angular momentum basis as

$$\Psi(\mathbf{r},t)=\sum_{l,m} R_{lm}(r,t)Y_{lm}(\theta,\phi)\ . \tag{3}$$

The number of the angular momenta is determined so that further increase of the basis does not change the results significantly. Since the laser light is taken linearly polarized, only $m=0$ angular states are needed in the calculation. The spatial step size is fixed at $dr=0.1$ a.u. The time step size of $\frac{1}{1024}$ and $\frac{1}{4096}$ of an optical period are used depending on the intensity and frequency of the light involved in the specific problem. The time-dependent induced dipole is calculated along the propagation of the wave packet as

$$d(t)=\int \Psi(\mathbf{r},t)^{\dagger} z \Psi(\mathbf{r},t) dr . \quad (4)$$

The basic idea of the model potential method is to freeze one electron of the helium atom at its ionic ground state, and to treat the motion of the second electron in the ion core by a model potential with several free parameters. Then we adjust the free parameters in such a way as to fit the data from experiments or other well-established theoretical calculations. Tang, Rudolph, and Lambropoulos [10] have shown the basic physical justification for the success of the frozen-core (FC) model, provided that the photon energies employed in the calculations are considerably smaller than the energy of the first doubly excited state. This condition is satisfied in our present calculations.

The first concern in constructing a model potential is to reproduce with sufficient accuracy the known energy spectrum of the atom. The values of the energy levels from our model potential are reasonably good compared with the values of energy levels from experiments. Good values for energies of the model potential are not sufficient to guarantee equally good values for the transition moments, which reflect the dynamic properties of the atom in the laser field. Their accuracy must be tested separately. The matrix elements of some transitions calculated by the model potential and the extended-configuration-interaction (ECI) method [10] are compared in Table I. It is quite clear from the table that the values from the model potential are compatible with the more sophisticated method.

In the case of interaction between atom and strong laser field, the excited states of an atom which are nonresonant in weak field can shift into resonance due to the ac Stark shift of the atomic energy level in the laser field. The experiments by Freeman *et al.* [12] and Perry *et al.* indicate that the intermediate resonance effect plays an important role in photoionization and above-threshold ionization processes. Such resonance effects are also manifested in the harmonic generation process [4]. We present now a summary of some of our main results.

Figure 1 gives the intensities of the fifth, seventh, and ninth harmonics, defined as the square of the Fourier transform of the last eight cycles of the TD dipole oscillation, as a function of pump photon energy. The laser intensity is 4.4×10^{14} W/cm^2. The results show that the harmonics are enhanced around the photon energies of 4.35 and 4.42 eV. At 4.35 eV the seventh harmonic is enhanced by almost 2 orders of magnitude, and the ninth harmonic by about a factor of 5. The seventh harmonic becomes larger than the fifth in the range of photon energy around 4.35 eV. An examination of the energy levels of He, and in view of the results of Ref. [13], suggests that the enhancement in the harmonic generation is due to the six-photon resonance with the $1s3s$ state. The shoulder on the seventh and ninth harmonics at the photon energy of 4.42 eV is due to six-photon resonance with the state $1s3d$. We have also calculated the population left in the bound states of the atom at the end of the laser pulse, and found that the resonance in photoionization, which is mostly due to the state $1s3d$, occurs at the photon energy of 4.4 eV, which is somewhat different from the photon energy at the peak of the harmonics. Thus the shift of the $2s3d$ state, according to our result, is $6\times4.4-23.07=3.33$ eV under the laser intensity of 4.4×10^{14} W/cm^2. This shift is very close to the ponderomotive shift and to the result in Ref. [3], which solves the TDSE with HF potential.

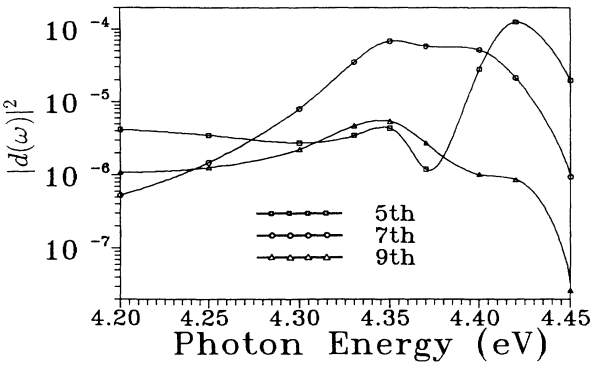

FIG. 1. The intensities of the fifth, seventh, and ninth harmonics as a function of photon energy at the intensity $I=4.4\times10^{14}$ W/cm^2 for helium. The field is ramped linearly to its maximum (over ten optical cycles) followed by a steady amplitude. The total propagation time is 32 optical cycles.

TABLE I. Matrix elements between representative states of He in atomic units (MP is the model potential; ECI, the extended configuration interaction [12]).

States	MP	ECI
$1s^2$ to $1s2p$	0.4078	0.4192
$1s^2$ to $1s3p$	−0.1978	−0.2073
$1s^2$ to $1s4p$	−0.1236	−0.1304
$1s2s$ to $1s2p$	−2.9368	−2.9242
$1s2s$ to $1s3p$	−0.9144	−0.9072
$1s2s$ to $1s4p$	−0.4643	−0.4601
$1s2p$ to $1s3d$	−2.507	−2.511
$1s2p$ to $1s4d$	−0.888	−0.884
$1s3p$ to $1s3d$	−5.170	−5.168
$1s3p$ to $1s4d$	−4.068	−4.064

It is interesting that the fifth harmonic is also enhanced by the six-photon resonance. This could not happen in the low-intensity regime where perturbation theory is applicable. The experiment by Allendorf *et al.* [14] has shown a similar phenomenon in Kr. We suspect that the origin of the enhancement of the fifth harmonic is a Raman process in which an atom absorbs six photons, and emits one pump photon and the fifth harmonics, which should become important in the high-intensity regime. More careful studies are needed to verify this interpretation.

A recent experiment [1] has produced high-order harmonic generation in He at a laser intensity of about 10^{17} W/cm^2. This raises several questions. Can the helium atom or even the helium ion survive at such high intensity? If not, what is the optimum intensity for harmonic generation by helium under the given photon energy? What is the relative weight of atom and ion in the process of harmonic generation? Because the answers to these

questions are sensitive to the energy of the pump photon, we have carried out calculations with photon energies of 2.0 and 5.0 eV, respectively. The photon energies of 5.0 and 2.0 eV were chosen for the calculation because of the recent experiments of Sarukura *et al.* [1] and Miyazaki and Sakai [2], where the highest reported harmonics were 23rd and 41st, respectively.

In order to generate high-order and high-intensity harmonics, the laser intensity must be high, but not higher than the saturation intensity for the specific photon energy and pulse duration, due to the ionization of the atom. The saturation intensities of the helium atom and helium ion are about 6×10^{14} and 5.5×10^{15} W/cm^2 at the photon energy of 5.0 eV for the pulses used in our calculations, which are shorter than those in the experiments. When the laser intensity is about 2×10^{15} W/cm^2, there is practically no population left at the ground state of the atom and at the end of the laser pulse. The helium atom cannot experience the peak intensity if it is higher than 2×10^{15} W/cm^2. Everything is ionized during the rising part of the pulse. We must then also consider the role of the helium ion, when the laser intensity is higher than 10^{15} W/cm^2. Figure 2 shows the results of the Fourier transforms of the time-dependent dipole oscillations of the helium atom and the ion at their saturation intensities. We notice that the atom produces up to the 13th harmonic. On the other hand, the ion produces up to the 41st harmonic. We have also calculated the Fourier transform of the dipole oscillations of the helium atom at other laser intensities, from below to above saturation. The number of harmonics produced is about the same when we vary the laser intensities around the value 6×10^{14} W/cm^2. From the above observation, we can reach the following conclusion: In an experiment of harmonic generation by the helium atom, with photon energy around 5.0 eV, if the peak laser intensity is higher than the saturation intensity of the helium ion, as in the experiment of Sarukura *et al.*, both the atom and the ion contribute to the process. The harmonics with order higher than 13 must be due to the ion's contribution only. For comparison, we also plot in Fig. 2 the experimental data of Sarukura *et al.*, normalized to the value of the 13th harmonic of the He ion.

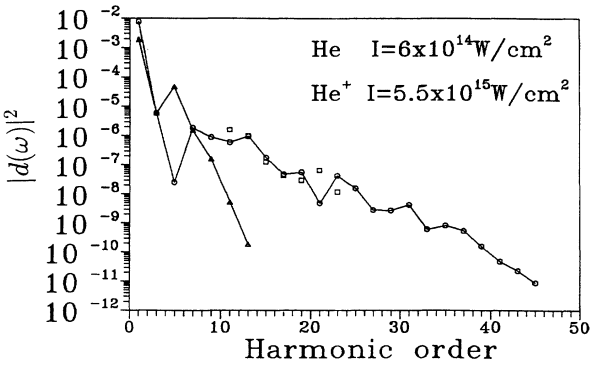

FIG. 2. Harmonic distribution with photon energy of 5.0 eV: helium atom (triangles) at the intensity $I=6\times10^{14}$ W/cm^2, helium ion (circles) at the intensity $I=5.5\times10^{15}$ W/cm^2. The pulse shape and duration in optical cycles are the same as those in Fig. 1. The squares represent the experimental data from Ref. [1] normalized to the value of the 13th harmonic of the ion of He. This normalization is based on the assumption that very little of the 13th or higher harmonics are produced by the atom.

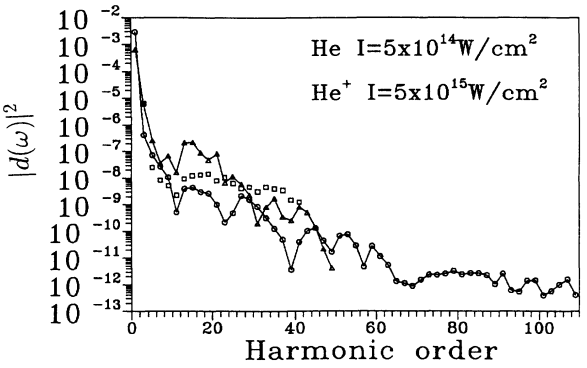

FIG. 3. Harmonic distribution with photon energy of 2 eV: helium atom (triangles) at the intensity $I=5\times10^{14}$ W/cm^2, helium ion (circles) at the intensity $I=5\times10^{15}$ W/cm^2. The pulse shape and duration in optical cycles are the same as those in Fig. 1. The squares represent the experimental data from Ref. [2] normalized to the value of the third harmonic of He. Although the peak intensity of the experiment was 1.4×10^{15} W/cm^2, optimum comparison is obtained with our 5×10^{14} W/cm^2 calculation.

Of course, the calculations only show the one atom or ion response in the laser field. As we mentioned before, one purpose of this paper is to investigate the basic role of the ion in the process of the harmonic generation of He. It is not our intention to reproduce exactly the experimental results because of the difficulties in simulating the real conditions in the experiments such as phase matching and pulse shape. The phase-matching problem of the high-order harmonics, however, has been found to be insensitive in the strong-field regime as compared to the weak-field limit [15]. Thus the single-atom results are basically conserved when considering the response of the whole medium.

We find similar results in the case of 2.0-eV photon energy, though the phenomenon is not as pronounced as that of the 5.0-eV photon. Figure 3 gives the Fourier transforms of dipole oscillations under the saturation intensities, which are about 5×10^{14} and 5×10^{15} W/cm^2 for the helium atom and ion, respectively. Our result for helium shows that the intensities of the harmonics from 13 to 21 are larger than that of the 11th harmonic, which is consistent with the outcome of the experiment [2].

In conclusion, by solving the TDSE with a model potential, we have demonstrated the effects of intermediate resonant states on harmonic generation and confirmed the role of the helium ion in the high-order harmonic generation.

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