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Harmonic generation beyond the saturation intensity in helium

A. Sanpera,[1] P. Jönsson,[2] J.B. Watson,[1] and K. Burnett[1]
[1]*Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, United Kingdom*
[2]*Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden*
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We present results from numerical simulations of the interaction of an intense ultrashort (100 fs) laser pulse with a neutral helium (He) atom and its ion He^+. The simulations are done for a number of laser intensities in the range of 10^{14}–10^{16} W/cm^2 at the KrF-laser wavelength. These intensities range from close to well above the saturation intensity of the neutral atoms and it is therefore necessary to study the contribution to harmonics from neutral atoms as well as ions. Our approach is based on a two-step procedure where we first calculated the neutral atom response in the single active electron approximation. We observe that harmonics are still produced by the neutral atoms above the saturation intensity, but the simple classical law for the cutoff is not obeyed. In the second step we calculated the response of ions. We show that the contribution from ions extends through and beyond the region where the contribution from the neutral atoms disappears. We find good agreement between our numerical results and the experimental ones. We also present some ideas about the importance of the laser defocusing and phase matching at such high intensities.

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I. INTRODUCTION

In the past few years there have been several experimental studies [1–5] of harmonic generation at laser intensities between 10^{14} and a few times 10^{17} W/cm^2. These intensities range from close to well above the saturation intensity of any rare gas. The saturation intensity (I_{sat}) is conventionally understood as the intensity at which a sample of atoms is mostly ionized. In principle, going past the saturation intensity means that we should now have clear experimental evidence of the role of ions in harmonic generation. Due to the larger ionization potential of rare gas ions compared to the neutral atoms, the harmonics generated by ions are expected to extend to a higher order than the ones generated by neutral atoms. Interpreting the results of present experiments is not, however, as simple as one might wish; going above the saturation intensity does not mean we completely deplete the neutral population. Moreover, the neutral atoms that are left will experience very high intensities indeed. This point may not have been fully taken into account in the interpretation of harmonic generation based on steady state response. Our calculation, which models the full response for a pulse, brings this point out clearly.

Calculations using one-dimensional model atoms and again considering the full pulse response have shown that the number of harmonic peaks increases with the peak laser intensity, even when the intensity goes above the point of saturation [6]. This increase, however, is not linear with the peak laser intensity as it is for intensities below the saturation intensity. This does not imply a complete breakdown of the cutoff rule, as one has to think in detail about the response of an ever diminishing number of atoms in a rapidly increasing field. This means we will get higher and higher harmonics coming from a smaller and smaller number of atoms. The efficiency in producing these high harmonics will be greatly reduced due to the small amount of atoms left. As a consequence, a complete and definitive answer about the role of ions cannot be deduced from the previously mentioned experiments since it is not easy to discriminate completely between the contributions from atoms and ions.

A qualitative explanation of the experiments performed to date can be obtained by combining the single neutral-atom response with that of ions, for a full pulse. This is what we present in this paper, so that experimentalists may better assess the relative role played by the single-atom or ion response. We shall address the case of helium and compare the responses of neutral He for a full pulse with that of He^+, the latter produced along the pulse as the neutral atoms are ionized. Helium has the largest ionization potential of all rare gases (U_i = 24.6 eV) and can therefore survive to very high intensities in the ionization process. The intensities we have used in the simulations range from 6×10^{14} to 2×10^{16} W/cm^2 for a fixed KrF-laser wavelength. The choice of this wavelength is related to the efficiency in producing harmonics. Larger wavelengths will produce higher-order harmonics, but with an associated lower efficiency [7]. This lower efficiency may well be an important issue in the attempts to observe the contribution from ions using longer wavelength lasers.

The harmonic spectra are obtained using the acceleration of the electric dipole moment $\ddot{d}(t)$. Closely related calculations using a steady state response have been previously presented in the literature [8–10]. In these calculations the harmonic spectra were determined from the dipole moment itself. Both quantities are easily related, but at high intensities the simple relation $\ddot{d}(t) = -\omega^2 d(t)$

cannot be employed because the dipole moment and its velocity do not vanish at large times [11]. It has been shown that when significant amounts of ionization are produced during the laser pulse, large final dipole moments are also produced. The presence of a final dipole moment can give rise to a background in the harmonic spectrum that increases with increasing intensity; this background is spurious and may hide harmonics that are produced during the process. For intensities around or beyond the saturation intensity the ionization is obviously large and the dipole acceleration has shown to be the most reliable method to calculate the harmonic emission.

Another interesting feature arises in our calculations since the overall response from ions is related to the amount of ionization as the pulse proceeds. By computing the ionization of neutral atoms during the pulse, we can calculate the ions' contribution more precisely. This is done by solving first the time-dependent Schrödinger equation for helium, obtaining a time-dependent ionization function $P_{\rm ion}(t)$. This function $P_{\rm ion}(t)$ is then included in the calculation of the ions' response. By doing that, we achieve a more realistic response of the ions for the whole pulse.

Finally, our results are compared with the available experimental data [3,5]. Quantitative comparisons should, of course, involve a calculation of the macroscopic issues of propagation with all its associated complexity. The single-atom response is, nevertheless, useful in indicating at what intensities neutral atoms and ions are competitive. We obtain, however, excellent agreement with the published results [3,5], stressing the fact that phase-matching effects could, in fact, be less important in the saturation regime [1]. Before describing our method and the results we obtain, we shall briefly review the experimental results in the next section.

II. EXPERIMENTAL RESULTS

To our knowledge there are at least five different experiments [1–5] in helium, at intensities beyond or around the saturation intensity. In an experiment with a gas target the saturation intensity is determined by the breakdown of the power law that relates the number of created ions with intensity I^q, where q is the order of the process. Krause *et al.* [8] define the saturation intensity as the intensity reached when approximately 20% of the atoms are left unionized. That results in an ionization rate that is a few times the inverse of the pulse width; in other words, the ionization rate times the pulse duration is close to one.

The experiments mentioned above correspond to different wavelengths ($248 \leq \lambda \leq 1053$ nm), peak intensities ($10^{14} \leq I \leq 10^{17}$ W/cm^2), pulse durations, and saturation intensities. Therefore the quantitative results vary greatly. In almost all the cases, nevertheless, the qualitative features are the same. A plateau in the harmonic generation is encountered. The cutoff to this plateau is given approximately by $U_i + 3U_p$ [8]. It has become conventional to infer the species, ion or neutral atom, responsible for generating a particular range of harmonics by using the cutoff rule (via the dependence of the cutoff on ionization potential). As an example, in the experiments performed by Sarukura *et al.* [5] with KrF, the cutoff rule has been used to infer generation of harmonics by He^+. Those inferences are based on the single-atom response at constant intensity close to saturation [8,9,12] and none of them take into account pulse shape or phase matching effects. The point we want to emphasize here is that even though only a small fraction of the neutral atoms survives to the saturation intensities, it radiates in the presence of a very high field. As a consequence, the harmonics generated from this small fraction can be comparable to those produced by ions over a fair range of the transition region.

III. METHOD

Our approach is based on a two-step procedure where, as a first step, the ionization and harmonic generation for the neutral helium atom is calculated in the single active electron (SAE) approximation with an absorbing boundary condition [8]. The validity of this approximation relies on the assumption that ionization is a stepwise process. It is well known that the SAE approximation leads to reasonable results for photons energies below 10 eV [9]. (At extrahigh intensities and ludicrously short pulses, direct multiple electron production may be possible [13].) For more energetic photons there is the possibility of a simultaneous double ionization and then the SAE will not be valid. The effective potential $V_{\rm eff}(r)$ of helium is constructed from Hartree calculations on its ground state, using a modified version of the Hartree-Fock program of Froese Fischer [14,15]. Our results for the effective potential and the helium ground state are displayed in Fig. 1. The ionization potential we obtain from the effective potential is $U_{\rm He} = 0.901$ a.u. (24.50 eV), very close to its experimental value (24.58 eV). From the decrease of the norm of the neutral atoms' wave function, a time-dependent ionization function $P_{\rm ion}(t)$ is obtained as the pulse proceeds. In the second step, we solve the time-

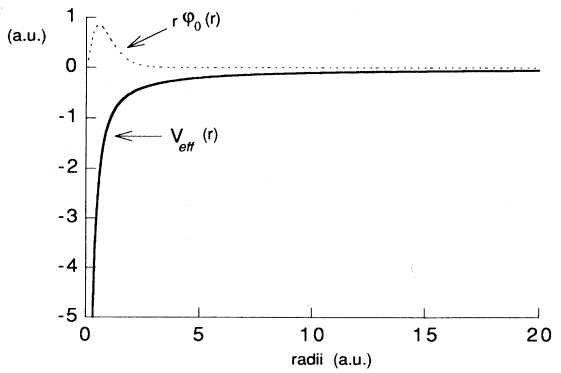

FIG. 1. Effective potential $V_{\rm eff}(r)$ and ground state wave function $\varphi_0(r)$ for helium. The calculated ionization potential is $U_{\rm He} = 0.90$ a.u. (24.5 eV).

dependent Schrödinger equation for the ion and then the ion wave function is weighted by the time-dependent $P_{\rm ion}(t)$ ion-population function. [At each temporal step, the wave function corresponding to He^+, $\Psi_{He^+}(\vec{r},t)$, is multiplied by $P_{\rm ion}(t)$. We assume an adiabatic behavior in the ions' response.] In this way the problem is reduced from the time propagation of a two-electron wave function to the propagation of two one-electron wave functions. The numerical time evolution of the one-electron wave function is performed using standard techniques [16,17]. We begin with the time-dependent Schrödinger equation

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t} = H(t)\Psi(\vec{r},t), \tag{1}$$

where $H(t)$ is the total Hamiltonian of the system

$$H = \{-\tfrac{1}{2}\vec{\nabla}^2 + V_{\rm eff}(r) + \vec{E}(t)\cdot\vec{r}\sin\omega t\}. \tag{2}$$

We use the length gauge and consider the laser to be linearly polarized along the z axis. Here $V_{\rm eff}(r)$ either refers to the effective potential if we deal with He or is simply equal to $-2/r$ if we deal with He^+. We expand the wave function $\Psi(\vec{r},t)$ in an angular basis

$$\Psi(\vec{r},t) = \sum_{l=0}^{l_{\max}} \frac{1}{r}\chi_l(r,t)Y_l^0(\theta), \tag{3}$$

where for simplicity only spherical harmonics with $m=0$ are introduced. This kind of simplified expansion is correct provided that the atom is initially in an s state. By using this angular basis we obtain a set of $l_{\max}$ coupled one-dimensional differential partial equations. The interaction term $\vec{E}\cdot\vec{r}$ couples each χ_l component with $\chi_{l\pm1}$ and the Schrödinger equation reduces to

$$\begin{aligned} i\frac{\partial}{\partial t}\chi_l(r,t) = &\left\{-\frac{1}{2}\frac{\partial^2}{\partial r^2} + V_{\rm eff}(r) + \frac{l(l+1)}{2r^2}\right\}\chi_l(r,t) \\ &+rE(t)\sin\omega t\,[c_l^+\chi_{l+1}(r,t) \\ &+c_l^-\chi_{l-1}(r,t)]\,. \end{aligned} \tag{4}$$

Here $c_l^{\pm}$ are the Clebsch-Gordan coupling coefficients. To solve Eq. (4) the Hamiltonian is split into the atomic and the interaction term with a symmetric decomposition. Each term is propagated in time with a semi-implicit Crank-Nicholson scheme. The box size is 300 a.u. with a spatial step $\Delta r = 0.10$ and we use an absorbing mask function boundary. Typically, the time step varies between 1024 and 4096 points per optical cycle. Because of the absorbing boundary the decrease of the norm of $\Psi(\vec{r},t)$ can be used as a measure of ionization. The time-dependent ion population is then given by

$$P_{\rm ion}(t) = 1 - \langle\Psi(\vec{r},t)\,|\Psi(\vec{r},t)\rangle. \tag{5}$$

As we pointed out before, this function will be used to weight the ions response at each temporal step, as the pulse proceeds. The harmonic emission is computed using the acceleration form as

$$P(\omega) \propto |\,a(\omega)\,|^2 = \left|\int_0^T \ddot{d}(t)\,e^{-i\omega t}\,dt\right|^2, \tag{6}$$

where T is the duration of the laser pulse and $\ddot{d}(t)$ the acceleration of the dipole moment

$$\ddot{d}(t) = \frac{d^2}{dt^2}\langle\Psi(\vec{r},t)\,|z|\,\Psi(\vec{r},t)\rangle. \tag{7}$$

The acceleration is calculated using Ehrenfest's theorem

$$\ddot{d}(t) = -\left\langle\Psi(\vec{r},t)\right|\frac{\partial}{\partial z}[V_{\rm eff}(r) + H_{\rm int}]\left|\Psi(\vec{r},t)\right\rangle. \tag{8}$$

In many studies the harmonic spectra is computed as

$$P(\omega) \propto \omega^4\left|\int_0^T d(t)e^{-i\omega t}dt\right|^2. \tag{9}$$

A partial integration

$$\begin{aligned} \int_0^T \ddot{d}(t)e^{-i\omega t}dt = &\,e^{-i\omega t}\dot{d}(T) + i\omega e^{-i\omega t}d(T) \\ &-\omega^2\int_0^T d(t)e^{-i\omega t}dt \end{aligned} \tag{10}$$

shows that the two methods are equivalent only if $d(T)$ and $\dot{d}(T)$ are negligible. If there is a significant amount of ionization during the pulse, the value of the final dipole moment can be quite large and, as a consequence, the two expressions (6) and (9) lead to different results [11,18]. Finally, the number of partial waves $l_{\max}$ used in the expansion of $\Psi(\vec{r},t)$ has to be carefully checked for each intensity. We consider that convergence is achieved when the harmonic spectrum obtained does not change significantly when increasing the number of partial waves included in the calculation. Since the different partial waves are coupled through the interaction term, it is clear that the number of partial waves has to be increased with the laser intensity. However, the harmonic generation emission is relatively insensitive to the high angular momentum components [20] and convergent results are obtained with few partial waves, i.e., $l_{\max} < 30$.

IV. NUMERICAL RESULTS

For the results presented here we use two kinds of pulses: short pulses with a linear turn on of 10 cycles followed by a flat part of 22 optical cycles, and more realistic pulses, with a $\sin^2$ envelope function, where the interaction lasts 128 cycles (105 fs). We first investigate harmonic emission at intensities around $I_{\rm sat}$ for neutral atoms and ions separately. We start by using the shortest pulses with a linear turn on of 10 cycles. To calculate the saturation intensity corresponding to each charge state (He or He^+) we compute the ionization rates through the decay of the normalization. As an example, in Fig. 2 we plot the ground state population and nor-

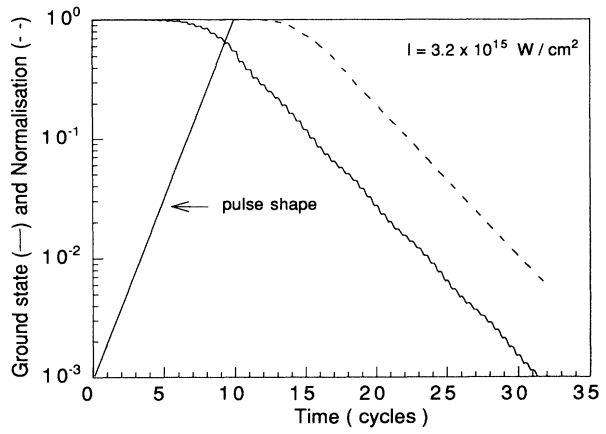

FIG. 2. Normalization and ground state population of He as a function of time (logarithmic scale) for a short pulse of 32 cycles with a linear turn on of ten cycles and intensity $I = 3.2 \times 10^{15}$ W/cm^2.

malization together, with the pulse profile for an intensity $I = 3.2 \times 10^{15}$ W/cm^2. We can see that the decay is purely exponential once the atom has adjusted its behavior to the external field, i.e., when it reaches the constant intensity part of the pulse and then the ionization rate can be computed either using the normalization function or the ground state population. Our predictions for the saturation intensities of He and He^+ at this wavelength (and pulse shape) are about 5×10^{14} and 4×10^{15} W/cm^2, respectively, in very good agreement with Ref. [8].

In Fig. 3 we plot the harmonic spectra for the neutral atoms at the intensities $I = (6,\ 20,\ \text{and}\ 32) \times 10^{14}$ W/cm^2. For clarity we plot only the peak intensities for each harmonic (joined by a line to guide the eye). The first point we should make is that in spite of going above the saturation intensity ($I_{sat} \simeq 5 \times 10^{14}$ W/cm^2), the number of harmonics does indeed increase with intensity. Second, the efficiency in harmonic generation decreases after exceeding I_{sat} as we should expect. Increasing the laser intensity further results in a gradual saturation of the higher harmonic orders. When this occurs the position of the cutoff will no longer increase linearly with intensity and then will not follow the $U_i + 3U_p$ rule. The complete saturation of the harmonic order occurs when there is no population left in the initial state and everything has been ionized. It can be seen in Fig. 2 that for $I = 3.2 \times 10^{15}$ W/cm^2 only 50% of the ground state population survives the linear turn on of the pulse. (The ground state population is calculated by projecting the time-dependent wave function onto the initial ground state.) It is this part left in the ground state that will experience the highest intensity and therefore generates the most energetic harmonics but with a lower efficiency. With greatly increasing intensity the ground state becomes depleted faster resulting in a poorer harmonic efficiency [7,20]. A similar kind of harmonic degradation has been seen experimentally [3].

To better understand these features we plot now the whole harmonic spectra (including the background) for the previous cases: He at $I = 2 \times 10^{15}$ W/cm^2 and $I = 3.2 \times 10^{15}$ W/cm^2 [Figs. 4(a) and 4(b), respectively]. We find that the edge of the plateau region gradually becomes broader [as can be seen by comparing Fig. 4(a) to Fig. 4(b)] as the intensity increases. For the

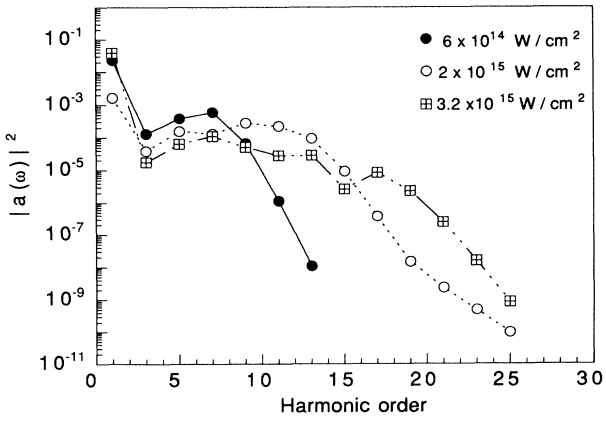

FIG. 3. Harmonic spectra of helium at intensities (a) $I = 6 \times 10^{14}$ W/cm^2 (solid dots), (b) $I = 2 \times 10^{15}$ W/cm^2 (dots), and (c) $I = 3.2 \times 10^{16}$ W/cm^2 (squares) for a short pulse with a linear turn on. For clarity we plot only the peak intensities of each harmonics joined by a line to guide the eye.

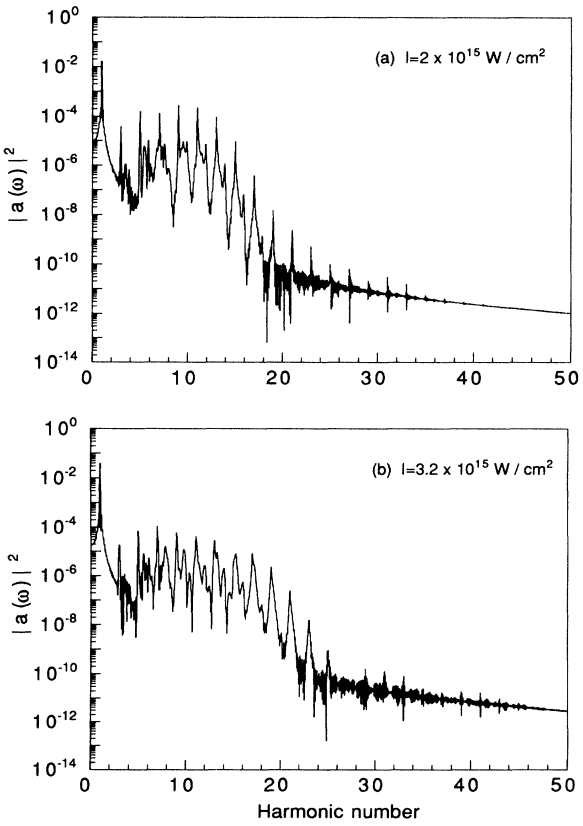

FIG. 4. (a) Harmonic spectrum of helium for a linear turn on pulse at intensity $I = 2 \times 10^{15}$ W/cm^2. (b) Harmonic spectrum of helium for a linear turn on pulse at intensity $I = 3.2 \times 10^{15}$ W/cm^2.

higher intensity [Fig. 4(b)] case we can clearly see that the high-order harmonics are broader than the low-order ones. The higher-order harmonics are produced at the flat part of the pulse, when there are just a few neutral atoms left. It is clear that they should be broadened in frequency since they are produced in very short time, just before the ground state becomes completely depleted.

We have seen that for short pulses the saturation intensity does not imply a strict limit on obtaining higher harmonics beyond the $U_i + 3I_{\rm sat}/4\omega^2$ order. We therefore repeat the calculations for a more realistic $\sin^2$ pulse of 128 cycles at different laser peak intensities, including now the ions' response during the pulse. The contribution from ions is weighted at each temporal step by the function $P_{\rm ion}(t)$ as it is computed from the decay of neutral-atom normalization. In Figs. 5(a) and 5(b) we plot the neutral atoms and ions response together with the pulse profile at two different intensities $I = 3.2 \times 10^{15}$ and $I = 1.2 \times 10^{16}$ W/cm^2. The first case [Fig. 5(a)] corresponds to an intensity below the $I_{\rm sat}$ of He^+, whereas in the second case [Fig. 5(b)] the intensity exceeds the He^+ saturation intensity. The overall response is therefore very different. For the lower intensity case, the neutral atoms ionize quickly to He^+, but very little He^{2+} is produced at the end of the pulse. For the latter case, however, the ions rapidly ionize to He^{2+} and very few He^+ ions in fact experience the peak intensity. We can see in Fig. 5(b) that the peak ion population is reached before the peak laser intensity and it decays very fast to He^{2+}.

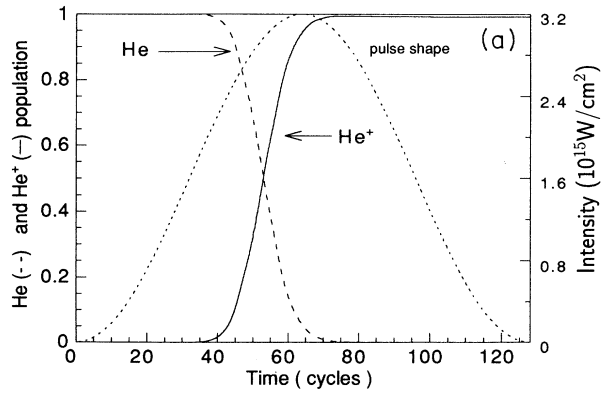

FIG. 5. (a) Neutral (He) and ion (He^+) populations for a $\sin^2$ pulse profile of 128 cycles (105 fs) and peak intensity $I = 3.2 \times 10^{15}$ W/cm^2. (b) Neutral (He) and ion (He^+) populations for a $\sin^2$ pulse profile of 128 cycles (105 fs) and peak intensity $I = 1.2 \times 10^{16}$ W/cm^2.

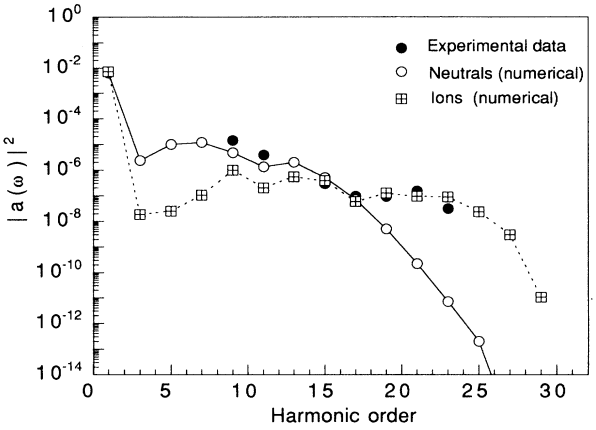

FIG. 6. Neutral [He (dots)] and ion [He^+ (squares)] response at $I = 3.2 \times 10^{15}$ W/cm^2. Experimental data (solid dots) from Ref. [5], normalized to the 13th harmonic, have been included for comparison.

We compare our harmonic generation results with the experimental ones from Refs. [3,5]. The best fit between experimental and numerical results occurs at intensities around $I \simeq 3.5 \times 10^{15}$ W/cm^2. This is shown in Fig. 6, where the experimental data are plotted together with the neutral atom and ion response at $I = 3.2 \times 10^{15}$ W/cm^2. (The experimental data are normalized to the 13th harmonic.) The agreement between the experimental and the numerical spectra is remarkably good. We can see that both neutral atoms and ions show the characteristic structure of a plateau followed by a cutoff. The plateau of ions extends to higher orders than the neutral atoms and it is crossed by the neutral-atom cutoff around the 15th–17th harmonic. So, in fact, only the higher peaks (19–23 orders) are produced by ions exclusively.

The fact that such good agreement between numerical simulations and experiments is reproduced by using a peak intensity almost two orders of magnitude lower than the one claimed in the experiments suggests that the effect of the defocusing of the laser beam at such intensity, gas density, and experimental geometry is important. A full treatment of the defocusing inside a gas will require a complex calculation on the refractive index gradient produced by ionization. However, a rough approximation following the work done by Rae [19] for a one-dimensional model atom indicates that the effective intensity could easily be more than one order of magnitude weaker than the peak laser intensity in vacuum. We could not assert that defocusing is solely to blame for the apparent decrease in intensity. In fact, other transverse properties of the beam—intensity variations, filamentation, poor beam quality—as well as longitudinal propa-

gation effects should be taken into account to have a full description of the experiments. It is quite impossible, at the present time, to combine a full treatment of single-ion atom evolution within a pulse with all the issues of beam propagation. The present study does, however, provide exceptionally strong motivation for pursuing studies of this kind.

V. CONCLUSIONS

We have calculated single-atom harmonic emission from neutral atoms and ions at intensities beyond the corresponding saturation intensity using realistic pulses. We have shown that the concept of saturation intensity does not imply a total restriction in obtaining higher-order harmonics. As a consequence we have seen that the contribution of neutral atoms in experiments running at very high intensities extends to higher orders than the ones previously mentioned in the literature. Nevertheless, we have found some differences in the behavior of the harmonic spectrum when the intensity goes beyond the corresponding saturation intensity. First of all, the efficiency of the harmonics clearly diminishes; the plateau intensity becomes lower for intensities above the saturation intensity than for the saturation intensity itself. We have also found that the cutoff of the plateau does not scale linearly with the intensity anymore and shows an obvious tendency to saturate. We have compared our results with the experimental ones and found excellent agreement for an intensity almost two orders of magnitude below the one claimed experimentally. This agreement suggests that phase matching plays a less important role in the saturation regime. Moreover it emphasizes that beam propagation effects at such intensities and gas densities result in significant lower effective intensities. Finally, we want to point that we have not been able to reproduce these results by using the dipole moment form of the harmonic calculation. This is what we would expect in this rapidly ionizing regime.

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