High-Order Harmonic Generation from Atoms and Ions in the High Intensity Regime

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We present calculated optical harmonic spectra for atoms and ions in the high intensity regime relevant to current short-pulse experiments. We find that ions can produce harmonics comparable in strength to those obtained from neutrals, and that the emission extends to much higher order. Simple scaling laws for the strength of the harmonic emission and the maximum observable harmonic are suggested. These results imply that the photoemission observed in recent experiments in helium and neon contains contributions from ions as well as neutrals.

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When an atom is exposed to an intense laser field, it develops a time-dependent dipole moment and radiates at odd multiples (harmonics) of the incident laser frequency. This process, known as optical harmonic generation (OHG), has been thoroughly studied at low to moderate laser intensities (e.g., 10^{13} to 10^{14} W/cm^2 at 1 μm) with pulse widths of 10–40 ps [1]. However, current laser systems can produce intensities in excess of 10^{17} W/cm^2, with pulse widths of 0.1 to 1.0 ps. Recent short-pulse experiments by several groups have observed high-order harmonic generation in the rare gases at intensities well beyond the saturation intensity for the atom and several of its ionization stages. Since ionized electrons cannot contribute to harmonic production unless their motion becomes relativistic, these experiments are probing the limits of OHG, certainly for neutrals, and possibly for ions as well. L'Huillier *et al.* [2] have observed the 53rd harmonic of 1053 nm in neon, Miyazaki and Sakai [3] have detected the 41st harmonic of 616 nm in He, and Crane *et al.* [4] have observed the 45th harmonic of 527 nm in He, as well as recombination lines indicating the presence of He^+ and He^{++}. The shortest harmonic wavelengths reported to date are 7.2 nm (the 109th harmonic of 806 nm) in Ne [5] and 9.9 nm (the 25th harmonic of 248 nm) in He [6].

These experiments raise several fundamental questions. One concerns the wavelength dependence of OHG. While several different wavelengths have been used to generate harmonics, no systematic study exists to show whether one wavelength is better than another at producing high-energy photons. Another question concerns the role of ions. The intensities developed by short-pulse lasers are sufficient to multiply ionize an atom, so it is natural to ask whether the resultant ions contribute to harmonic production. The expectations for harmonic emission from ions are unclear. Theory and experiment at lower intensity have demonstrated that an atom with a higher ionization potential (I_p) generates higher-order harmonics than an atom with a lower I_p [7]. Because the I_p of an ion is much higher than that of the neutral, ions are potentially a source for very short-wavelength harmonics. However, the ion has a lower polarizability and a correspondingly lower conversion efficiency. But the ion can experience much higher intensities before ionizing further, so the harmonic emission rates might be significant. At high intensities ionization occurs via tunneling, which means that the bound electrons make a transition directly into the continuum by tunneling through or flowing over the suppressed Coulomb barrier. Atomic resonances play little or no role in electronic excitation. Therefore, one might expect that these conditions would be unfavorable for OHG, because photoemission requires a transition from the continuum back to the ground state, which can occur only near the nucleus [8].

To answer these questions we have calculated optical harmonic spectra from several atoms and ions at intensities up to the saturation intensities, I_{sat}, attainable with 0.1–1.0 ps lasers. I_{sat} is defined for each charge state, and corresponds to the maximum intensity that a reasonable number of atoms or ions (say, 20%) experience before ionizing. This results in a peak ionization rate of a few times the inverse pulse width. Our goal is to examine the limits of harmonic emission that can be attained from a single atom or ion interacting with a short-pulse laser. The harmonic intensities that can be observed in experiments are obviously limited by the single-atom response. One of the main results of this work is the development of a simple formula that depends only on the wavelength of the driving field, the saturation intensity, and the I_p of the emitting species. This formula enables us to estimate the maximum observable harmonic photon for a specified set of experimental conditions. It also allows us to predict the harmonic order at which the contribution from ions becomes stronger than that from neutrals. We confirm for the first time that ions can emit very high-order harmonics with a magnitude comparable to that obtainable from neutrals. In addition, we show that the emission from ions will extend to much higher order than that from neutrals. Based on these results we conclude that most of the available experimental data can be explained by assuming harmonic emission exclusively from neutrals. However, we believe that recent experiments in helium and neon have in fact observed harmonic emission from ions.

The results presented in this work were obtained using methods described previously [8]. For one-electron sys-

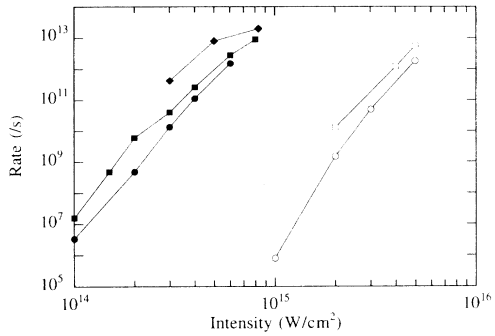

FIG. 1. Calculated ionization rates for He at 248 nm (solid diamonds), 527 nm (solid squares), and 1053 nm (solid circles), and He^+ at 248 nm (open squares) and 527 nm (open circles). The rates for helium are total (two-electron) rates.

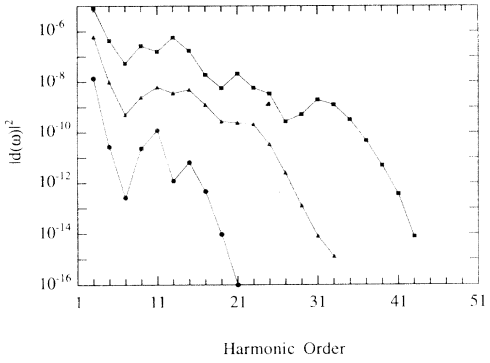

FIG. 2. Single-electron harmonic spectra $|d(\omega)|^2$ for He at 1×10^{14} (circles), 3×10^{14} (triangles), and 6×10^{14} W/cm^2 (squares).

tems such as He^+ we integrate the time-dependent Schrödinger equation directly on a numerical grid. For He and the other rare gases, we solve the time-dependent Hartree-Fock equations using a single-active-electron approximation in which we assume that only one electron interacts with the field, while the others remain fixed in their ground-state orbitals. This approximation has been tested extensively and found to produce results that can be reliably compared with experiment [9]. The output of both methods is the time-dependent dipole $d(t)$ induced by the field in the atom or ion. The square of the Fourier transform of this quantity, $|d(\omega)|^2$, is proportional to the single-atom photoemission spectrum. More importantly, $d(\omega)$ is the driving term in Maxwell's equations that must be solved to propagate the harmonic field in the nonlinear medium. The emitted harmonics are coherent, and as they propagate through the medium are subject to phase-matching conditions that determine the eventual strength of the macroscopic emission. L'Huillier, Schafer, and Kulander [1,10] have shown that at intensities up to I_{sat} in the nonperturbative regime, phase matching is relatively independent of harmonic order, and so the experimental spectra strongly resemble the single-atom spectra. Laser pulses that produce intensities which greatly exceed the saturation intensity, however, will result in a high density of free electrons which can significantly alter the phase matching.

In this paper we focus on results for He and He^+. He has been studied by several experimental groups because it has the highest neutral I_p (24.6 eV), and so can withstand higher laser intensities than the other rare gases. Figure 1 shows calculated ionization rates for He and He^+ at various wavelengths as functions of laser intensity. As expected, at a given intensity the ionization rate for He is much higher than the rate for He^+, since the I_p for He^+ (54.4 eV) is more than twice that of He. This justifies treating the two species separately in our calculations.

Figure 2 shows calculated harmonic intensities for He at 527 nm and intensities of $(1-6)\times10^{14}$ W/cm^2. These spectra all display the three typical features of experimental and theoretical harmonic spectra [1], a rapid decline over the first few harmonics followed by a plateau of relatively constant harmonic intensities and then an abrupt cutoff. As the laser intensity increases, the harmonic intensities and the extent of the plateau increase as well. At 6×10^{14} W/cm^2, the cutoff in the spectrum begins at about the 33rd harmonic.

In Fig. 3 we plot harmonic intensities for He at 527 and 1053 nm and He^+ at 527 nm at laser intensities that produce ionization rates of $\sim2\times10^{12}$/s. At low harmonic order, He at 527 nm produces the most intense harmonics. The emission from the ion becomes competitive with that from the atom near the cutoff, and extends to much higher order. At these intensities, both He at 1053 nm and He^+ at 527 nm are entering the tunneling regime (as determined by the Keldysh tunneling parameter, $\gamma\approx0.4$) but still produce abundant harmonics.

The results in Figs. 2 and 3 illustrate two simple and striking scaling relations with regard to the breadth and intensity of the plateaus. First, the maximum harmonic in the plateau (i.e., the harmonic at which the cutoff *begins*) is given quite accurately by

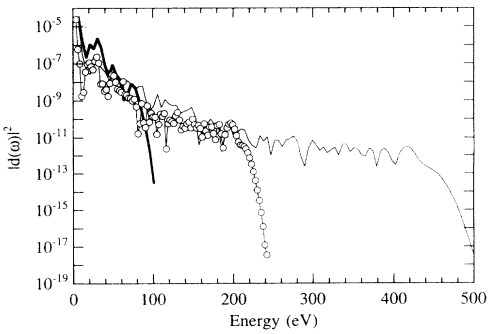

FIG. 3. Harmonic spectra for He at 527 nm (thick line) and 1053 nm (open circles), and He^+ at 527 nm (thin line) at intensities producing ionization rates of $\sim2\times10^{12}$/s (6×10^{14} W/cm^2 for He and 5×10^{15} W/cm^2 for He^+).

$$E_{max} \approx I_p + 3U_p , \quad (1)$$

where $U_p = I/4\omega^2$ is the ponderomotive shift of the ionization potential, I is the laser intensity, and ω is the frequency and the factor of 3 was determined empirically. This formula shows that the cutoff scales linearly with both the ionization potential of the atom and the laser intensity, and depends quite strongly on the wavelength when U_p is large. We have found that this simple relation predicts the calculated cutoffs in a large number of systems including the rare gases, hydrogen, and a variety of one- and three-dimensional model potentials, both Coulombic and non-Coulombic, including potentials with no (field-free) bound excited states. The only similarity in these potentials was the presence of a deep bound state separated by several photons from the continuum. The calculations considered many combinations of wavelength and intensity. Equation (1) differs significantly from previously proposed scaling laws based on either a mean Rabi frequency [11] ($I^{1/2}$ scaling) or twice the ac Stark shifted ionization potential [12].

At first glance, it may be difficult to understand the origin of photons as energetic as those predicted by Eq. (1). However, some insight can be obtained by appealing to a simple classical argument that has been used previously to explain some aspects of above threshold ionization. In this model the atom serves as a source of free electrons that are "born" in the continuum at random times during the optical cycle. The maximum cycle-averaged kinetic energy such an electron gains from the field is $3U_p$ [13]. However, in the true tunneling limit, the electrons would be produced only at the points in the cycle when the electric field is at its peak. Under such conditions, the energy of the electron would be U_p. We have clearly not reached this limit in the present calculations.

The second regularity revealed by our results is a simple dependence of the height of the plateau on the laser intensity. In the high intensity regime we find that the intensity of the plateau is proportional to the ionization rate, with a system-dependent proportionality constant. This scaling can be clearly seen in Fig. 2, where the plateau increases by 5 orders of magnitude as the ionization rate (see Fig. 1) increases by about the same amount. This scaling reflects the fact that ionization and harmonic emission both require excitation out of the ground state, and indicates that the processes responsible for harmonic generation and ionization are coupled, even in the tunneling regime.

These observations suggest that a system in which the lowest excited state is as far as possible above the ground state will produce the maximum number of harmonics, because such a system will be the most resistant to ionization. We illustrate this point in Fig. 4 by comparing calculated spectra for hydrogen at 7.1×10^{13} W/cm^2 and a 3D Yukawa potential at 2.0×10^{14} W/cm^2. The parameters of the Yukawa potential were chosen such that it has the same I_p as hydrogen, but no bound excited states. The ionization rates for the two cases are equal (corresponding to saturation for a 2 ps pulse). With no bound states, and therefore a higher saturation intensity, the Yukawa potential produces *many* more harmonics.

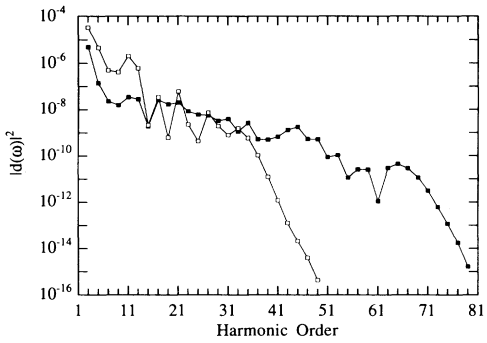

FIG. 4. Harmonic spectra for hydrogen at 7.1×10^{13} W/cm^2 (open squares) and a 3D Yukawa potential at 2.0×10^{14} W/cm^2. Both calculations were at 1064 nm. The Yukawa potential was chosen to have the same I_p as H (13.6 eV), and no (field-free) bound excited states.

We can use our simple formula to interpret several recent experiments which used laser intensities up to and above the saturation intensities for neutral atoms. Macklin *et al.* [5] have reported the first experimental evidence that the cutoffs in the harmonic spectra in Ne at 806 nm scale linearly with the laser intensity. By using Eq. (1) and calculating ionization rates for neon, we find that all of the observed harmonics (up to the 109th) in these experiments are consistent with emission from neutral neon. Macklin *et al.* reach the same conclusion by measuring blueshifts of the harmonics as a function of pressure and laser intensity. These shifts can be simply related to the free electron density produced during the pulse [5,14]. Crane *et al.* [4] have detected the 45th harmonic of 527 nm in He using a 600 fs pulse. Our calculations predict that the saturation intensity for this pulse is approximately 8×10^{14} W/cm^2. At this intensity, the calculated spectrum for neutral helium begins to cut off at near the 39th harmonic. Crane *et al.* have also observed harmonics of 1053 nm. Although they have sufficient intensity to generate high-order emission from ions, the observed spectra are not inconsistent with emission from neutral helium. However, it is possible that the two highest harmonics they observe are from ions. In contrast, one recent experiment shows clear evidence for emission from ions. Sarukura *et al.* [6] have reported the 23rd harmonic of 248 nm in He using a 285 fs pulse. We have calculated harmonic spectra for neutral He at this wavelength and find no harmonics beyond the 13th at the saturation intensity, which occurs at about 5×10^{14} W/cm^2 (see Fig. 1). He^+, however, has a saturation intensity of about 4×10^{15} W/cm^2 under these conditions, and is capable of producing the reported harmonics and cutoff. Similarly, emission from Ne^+ must be invoked to explain the high-

order harmonics detected in neon by Sarukura *et al.*

In conclusion, we presented in this paper the first calculations comparing harmonic emission from atoms and ions. We showed that in the intensity regime of current experiments the single-atom response depends strongly on the wavelength, and that ions can contribute significantly to the harmonic spectra. We emphasize that the contributions from ions can become competitive with that from neutrals only when the peak intensity in the focal volume experienced by the ions equals or exceeds their saturation intensity. At such intensities it is difficult to determine experimentally the source of the photoemission because the observed signal is phase matched over a macroscopic distance, and both species are present in the focal volume. We presented a simple formula to estimate the maximum harmonic that can be expected from a given set of experimental parameters and showed that photons with energies of several hundred eV can be generated with existing short pulse laser systems.

The *actual* emission observed in an experiment depends on the phase matching of the single-atom spectra to produce a macroscopic field. Our experience [1] indicates that, under the conditions of low pressure and weak focus typically encountered in experiments, the phase-matched results do not differ dramatically from the single-atom results. However, when comparing photon emission from ions and neutrals, two factors may tend to modify our predictions. First, the volume of the laser focus in which the intensity is high enough to produce ions may be considerably smaller than the volume in which the neutrals radiate efficiently. Second, since neutrals saturate at much lower intensities than ions, many free electrons will be present in the laser focus. These electrons may severely affect the propagation of the pump and harmonic fields, and hence reduce the strength of the coherent emission from the ions. The trend of the available experimental data is that, as expected, the effects of free electrons on high-order harmonics are more deleterious at longer incident wavelength than at shorter wavelength.

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