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K. J. Schafer, J. L. Krause and K. C. Kulander
University of California, Lawrence Livermore National Laboratory
Livermore, CA 94551

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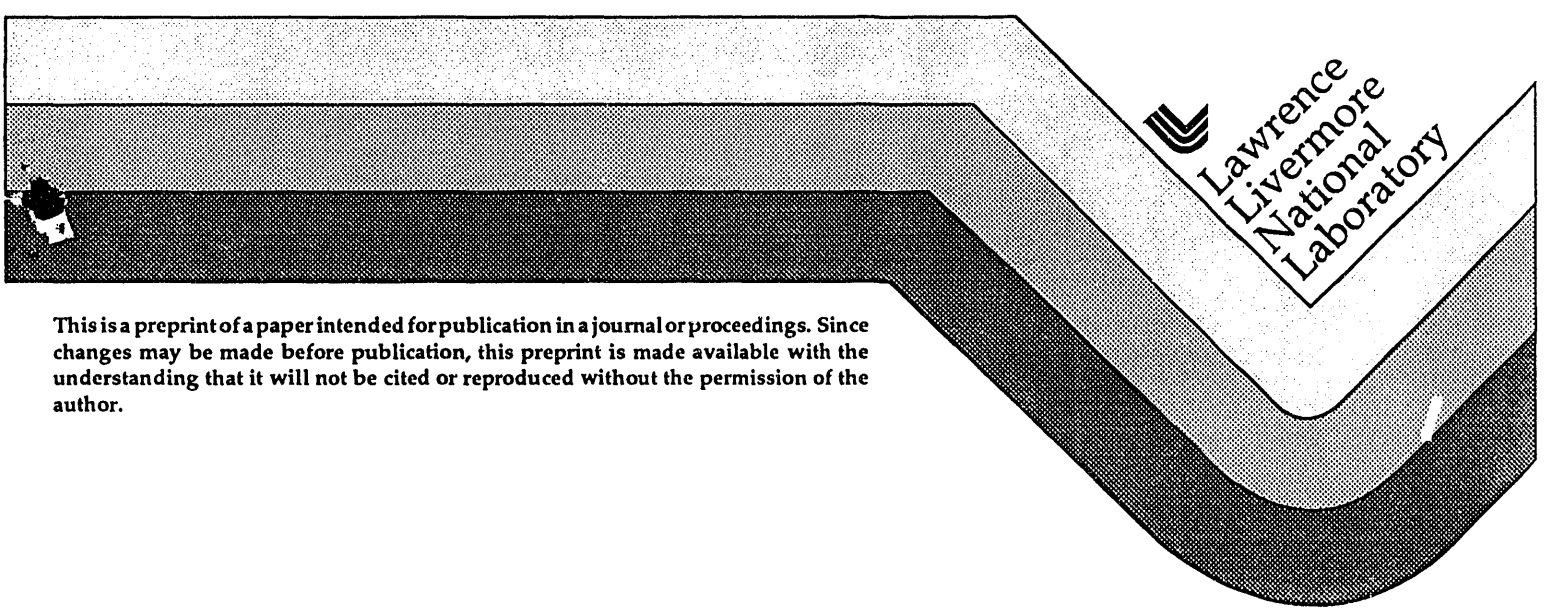

High-order harmonic generation with short-pulse lasers

Kenneth J. Schafer, Jeffrey L. Krause and Kenneth C. Kulander

Physics Department
Lawrence Livermore National Laboratory, Livermore, CA 94550

Abstract

Recent progress in the understanding of high-order harmonic conversion from atoms and ions exposed to high-intensity, short-pulse optical lasers is reviewed. We find that ions can produce harmonics comparable in strength to those obtained from neutral atoms, 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.

Introduction

Experiments employing short-pulse, high intensity lasers to excite rare gas atoms have produced surprisingly strong XUV emission due to harmonic conversion of the pump radiation.[1-6] It is expected that this phenomenon can be exploited to provide an inexpensive, flexible source of short wavelength, coherent, tunable, short pulse radiation. The development of laser systems capable of producing more than 10 TW with focused intensities above 10^{18} W/cm^2 and pulses shorter than a picosecond has enabled several groups to observed high-order harmonic generation 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 harmonic generation, certainly for neutrals, and possibly for ions as well. L'Huillier *et al.*[7] have observed the 133rd harmonic of 1053 nm in neon, Miyazaki and Sakai[4] have detected the 41st harmonic of 616 nm in He, and Crane *et al.*[5] 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)[6] in Ne, and 9.9 nm (the 25th harmonic of 248 nm)[3] in He.

These experiments raise several fundamental questions. One concerns the wavelength dependence of harmonic generation. 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. It is natural to ask whether the ions that result from such high intensities can contribute to harmonic production. 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. By examining the limits of harmonic emission that can be attained from a single atom or ion interacting with a short-pulse laser we hope to develop predictive capabilities which can establish from first principles the parameter space within which optimum harmonic conversion using existing state-of-the-art short pulse optical lasers can be achieved.

SINGLE ATOM CALCULATIONS

The results presented in this work were obtained using methods described previously.[8-11] For one electron systems 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 (SAE) 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. The development of the SAE model has made possible the study of strongly non-perturbative processes with manageable computational effort. Its

success demonstrates that the ionization and polarization dynamics of the rare gases become dominated by single excitations in the strong field regime. 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:[12]

$$\sigma(\omega) \equiv \omega^3 |d(\omega)|^2 = \omega^3 \left| \frac{1}{T_2 - T_1} \int_{T_1}^{T_2} dt\, e^{i\omega t} \int d\vec{r}\, \Psi(r,t)\, z\, \Psi(r,t) \right|^2 \qquad (1)$$

or equivalently

$$= \omega^{-1} \left| \frac{1}{T_2 - T_1} \int_{T_1}^{T_2} dt\, e^{i\omega t} \int d\vec{r}\, \Psi(r,t)\, \ddot{z}\, \Psi(r,t) \right|^2 \qquad (2)$$

where $\ddot{z} = [H,[H,z]]$ is the acceleration form of the dipole. This latter choice is found to be more tractable because it weights the value of the wave function near the origin which is more likely to be uncontaminated by numerical errors. The use of the acceleration form has allowed us to obtain converged harmonic emission spectra for much higher intensities than would have been possible with Eq. 1. Because the entire electronic wave function appears in the dipole moment expression we must sum the contributions of the individual active electrons before transforming and squaring; the separate dipoles add coherently. The interval for the transform is chosen to be well after the laser turn-on to minimize transient effects. Equation (2) provides the emission spectrum for a given intensity and wavelength. Because the atoms within the laser focus experience a range of intensities during the pulse, these calculations must be repeated for many intensities. For driving laser pulses shorter than 50 fs the transform in Eq. 2 is taken over the entire pulse because under these circumstances the transient effects are real and can be significant.

The harmonic intensities that can be observed in experiments are obviously limited by the single-atom response. More importantly, $d(\omega)$ is the driving term in the Maxwell's equations that must be solved to propagate the harmonic field in the nonlinear medium. The emitted harmonics are coherent and as such are subject to phase-matching conditions. Propagation of the generated harmonic fields through the excited medium can, in principle, significantly alter the emitted spectrum. Therefore, a complete theoretical treatment of harmonic generation requires two equally important steps: (1) calculation of the polarization fields induced in the individual atoms by the driving laser and (2) determination of the macroscopic phase matching of these harmonic fields. L'Huillier *et al.*[13-16] have shown that at intensities up to I_{sat} in the non-perturbative 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.

The same time-dependent calculations provide the ionization rates of the atom for these wavelengths and intensities. Harmonic generation from a neutral atom decreases dramatically when the atom ionizes because the polarizability of the remaining ion is substantially lower than the neutral. Therefore it is important to determine the lifetime of an atom in the field in order to evaluate the total emission during the pulse. Of course, ions subjected to much higher intensities will become polarized, emitting harmonic photons up to the point when they themselves become ionized. The ionization rates are determined by calculating the flux of probability through a surface well removed from the atom. In Fig. 1 we show the calculated ionization rates for neutral and singly ionized helium as functions of laser intensity for several wavelengths. From these results it is clear that the peak intensity helium will survive in a 1 ps pulse is approximately 6×10^{14} W/cm^2 and for the ion, 5×10^{15} W/cm^2. We note that the rates at these intensities are only weakly dependent on the wavelength even though it requires at least twenty-one 1053 nm photons as compared to eleven of the 527 nm photons to ionize helium. This is a clear manifestation of the non-perturbative nature of the dynamics under these conditions.

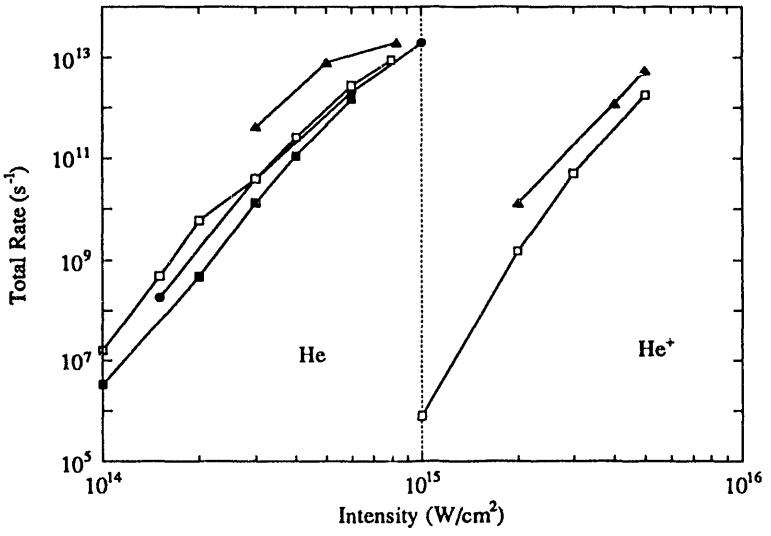

Fig. 1. Ionization rates for He and He^+ at 1053 nm (filled squares), 620 nm (filled circles), 527 nm (open squares) and 248 nm (filled triangles).

In Fig. 2 we show emission spectrum for neon for an intensity of $6x10^{14}$ W/cm^2 at 806 nm (slightly below the saturation intensity appropriate for the experiment of Macklin, *et al.*[6]). The spectrum shows the characteristic structure of the single-atom emission in the non-perturbative regime. There is a broad background on which there are narrow peaks at frequencies corresponding to odd multiples of the incident

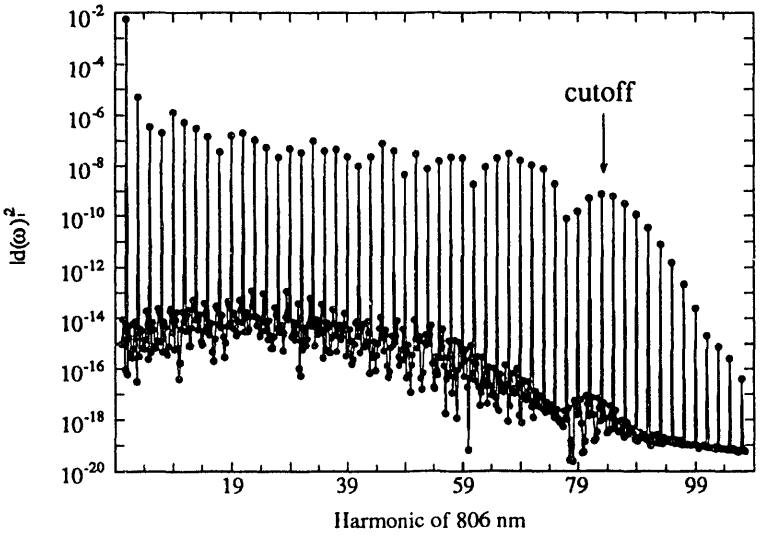

Fig. 2. Single atom spectrum for neon at 806 nm and $6x10^{14}$ W/cm^2.

photon energy. Because of the inversion symmetry of an atom in a linearly polarized field, even harmonics are forbidden. The harmonic strengths exhibit the following pattern, a rather rapid decline for the first few orders, then a broad plateau during which the intensity drops slowly, followed by an abrupt *cutoff*. The width of the harmonic peaks is determined by the shorter of the ionization lifetime or the pulse length. If we consider only the harmonic strengths as functions of intensity, we see the plateau rises and broadens with increasing intensity. This is shown in Fig. 3 where emission strengths for He at three different intensities for 527 nm (applicable to the experiments of Crane, *et al.*[5]) are displayed.

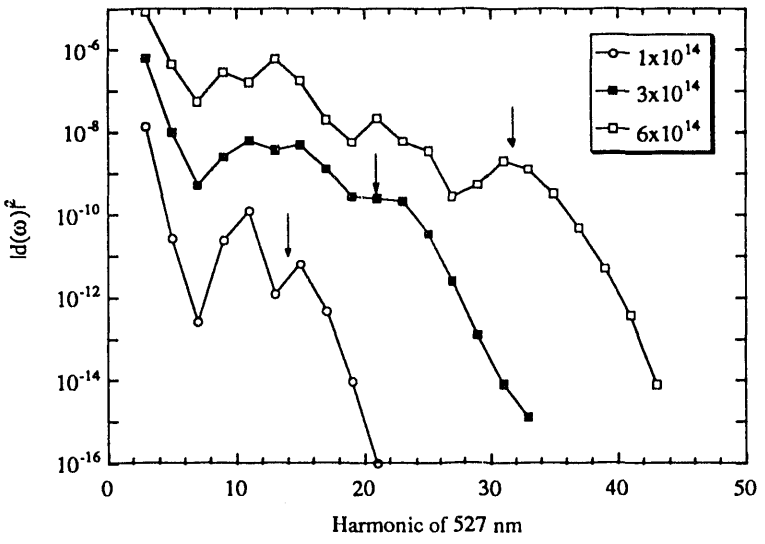

Fig.3. Single atom spectra for neutral helium at 527 nm.

SIMPLE SCALING LAWS

The results in Figs. 1 to 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:[17]

$$E_{max} \cong I_0 + 3U_P \qquad (3)$$

where $U_p=I/4\omega^2$ is the ponderomotive shift of the ionization potential, I is the laser intensity, ω is the frequency and the factor of three was determined empirically. We indicate this predicted cutoff by the arrows in figures 2 and 3. Above this energy the conversion efficiency drops rapidly with order. This formula shows that the cut-off 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 1- and 3-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.

At first glance, it may be difficult to understand the origin of photons as energetic as those predicted by Eq. 3. However, some insight can be obtained by considering a "two step" semiclassical model in which the atom is a source of free electrons that are "born" near the nucleus (the only place they can absorb enough photons to reach the continuum) at arbitrary times during an optical cycle.[18] Two types of classical orbits occur: those that return to the region near the nucleus and those that do not. Harmonic generation occurs

only for those orbits which have at least one additional collision with the nucleus. This is borne out by numerical calculations which show that high-order harmonic production is completely accounted for by considering only transitions that end in the ground state, which is localized around the nucleus. Therefore, within this simple model the maximum energy that the emitted photon can have must be the energy that the electron has *at the time it revisits the vicinity of the nucleus*. We find that for electrons that are born near the nucleus, regardless of their initial energy distribution, the maximum energy at the return time is 3.17 U_p plus the field free ionization potential. This predicts the cutoff remarkably well.

As a result of this scaling law we can identify a number of important factors in maximizing high energy harmonic production. First, for a given pulse length, one wants to work near the saturation intensity for the species of interest. This will result in the highest conversion efficiency and highest energy harmonics. For peak intensities higher than the saturation intensity, the source will be destroyed during the rise of the pulse. Second by using shorter pulses, higher peak intensities can be employed, again leading to more and stronger harmonics. Also atoms with higher ionization potentials can survive to higher intensities producing

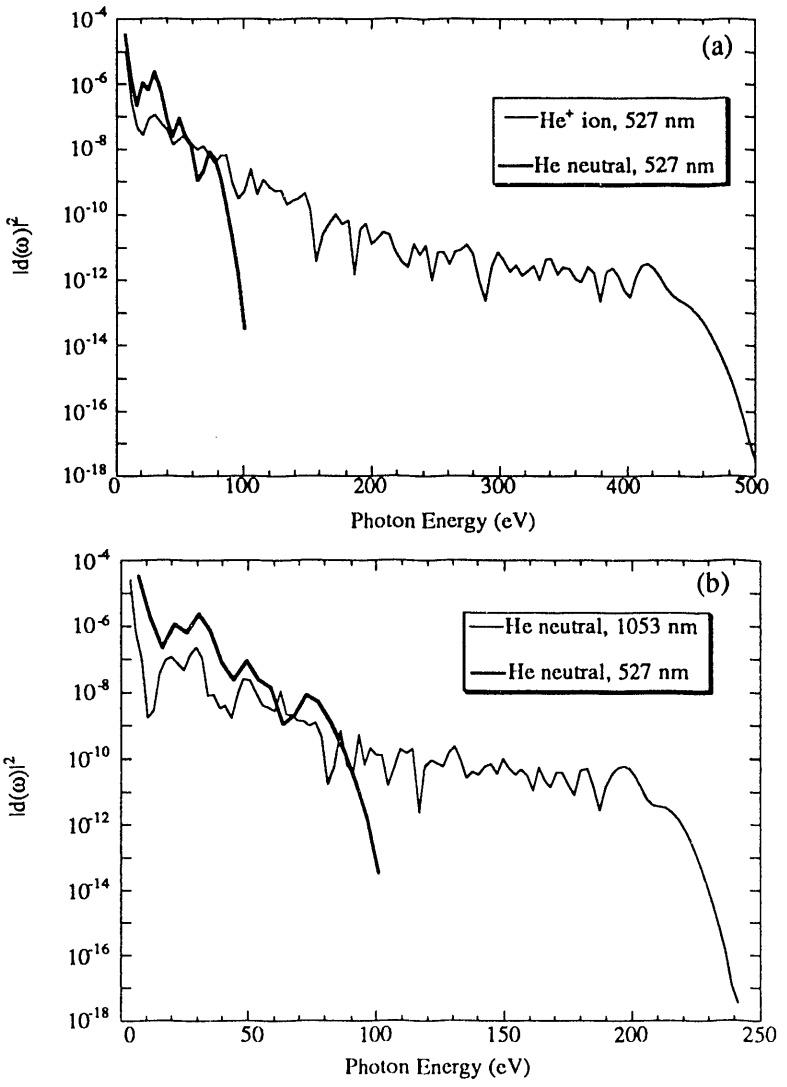

Fig. 4. Single atom spectra for helium neutral and ion at 527 and 1053 nm.

broader plateaus. This leads to the question as to whether it is more desirable to generate harmonics from ions rather than neutrals because of the higher ionization potentials for ions. This is clearly the case. In Fig 4(a) we show that the helium ion, whose saturation intensity for a 1 ps pulse is approximately $5x10^{15}$ W/cm^2, will emit harmonic photons with energies above 400 eV (less than 3 nm). Figure 4(b) also shows the advantage of using longer pump wavelengths as predicted by Eq. 3 to produce high energy harmonics by comparing results in helium neutral for 1053 and 527 nm. Although the emission strength for the shorter wavelength pump is higher for the harmonics within its plateau, the cutoff is much higher for the longer wavelength.

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. 3, where the plateau increases by five 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.

COMPARISON WITH EXPERIMENT

The experiments of Crane *et al.*[5] in helium show a plateau which extends up to the 45th harmonic when excited by a 650 fs pulse at 526 nm with a peak intensity above 10^{17} W/cm^2. From Fig. 1 we conclude the saturation intensity of neutral helium is approximately $6x10^{14}$ W/cm^2 and that for the ion is $5x10^{15}$ W/cm^2. Therefore, there is plenty of intensity to polarize either the atoms or ions in the focal volume. Our calculations predict that the harmonics from the neutral should cut off after the 35th, so that we believe the higher ones observed are due to emission from ions. In the data there is actually in a reduction in the harmonic strengths of more than an order of magnitude just above the 35th harmonic. We also predict the ion emission should extend up to 177th order. In more recent experiments harmonics up to 67th order have been seen. The difficulty in seeing these very high orders is most likely due to phase matching being poor in the presence of electrons which must accompany the production of an ionic medium.

Additional experiments have shown the reliability of the scaling law for the maximum harmonic order given be Eq. 3. Macklin *et al.*[6] have seen up to the 109th order of an 806 nm, 125 fs pulse in neon. They report that these harmonics are all from the neutrals, as predicted by our scaling law. L'Huillier and coworkers[7] have observed the 133rd harmonic from a 1 ps, 1053 nm laser, also in neon. They did not see a cutoff because of detector limitations, but we would predict the plateau of the neutral would extend above the 153rd harmonic. This result, combined with the 526 nm result of Crane *et al.*[5] in helium shows the benefit of using longer pump wavelengths to reach higher energy harmonics. Again we note the conversion efficiency is generally higher when the incident wavelength is shorter.

From these studies we have found that most harmonic photons come from neutral atoms. One recent experiment, however, shows clear evidence for emission from ions. Sarukura *et al.* [3] 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 $5x10^{14}$ W/cm^2 (see Fig. 1). He^+, however, has a saturation intensity of about $4x10^{15}$ W/cm^2 under these conditions, and is capable of producing the reported harmonics and cutoff. This is illustrated in Fig. 5 where we have scaled the experimental data of Sarukura onto single atom spectra. The cutoffs predicted by Eq. 3 are indicated by arrows as before. This simple exercise neglects phase matching which is not expected to play a role in determining the cutoff at this wavelength. Similarly, emission from Ne^+ must be invoked to explain the high-order harmonics detected in neon by the same workers.[3]

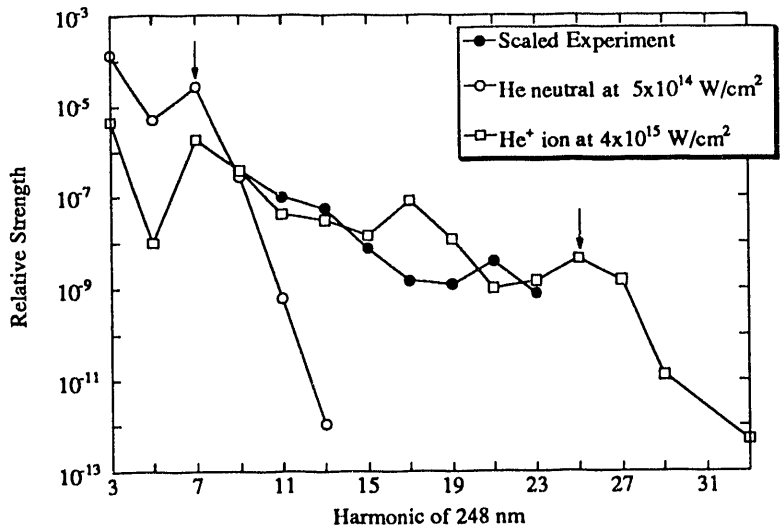

Fig. 5. Comparison of single atom spectra and the experiment of reference 3.

CONCLUSIONS

In this paper we have shown that in the intensity regime of current experiments, the single atom response depends strongly on the pump laser 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 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 considerable smaller than the volume in which the neutrals radiate efficiently. Secondly, 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. Phase matching in the presence of free electrons at long pump wavelengths is the major remaining challenge facing workers in this field.

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