High-order harmonics of 248.6-nm KrF laser from helium and neon ions

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We report high harmonic generation from a 248.6-nm KrF laser giving harmonic orders up to the 37th (67 Å) in a helium gas jet and the 35th (71 Å) in neon, for laser intensities up to 4×10^{17} W/cm^2 in 380-fs pulses. These observations are interpreted using theoretical modeling that identifies the ion species He^+, Ne^+, and Ne^{2+} as the sources of the highest harmonics.

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Recent advances in short-pulse laser technology have stimulated research into the production of coherent radiation in the extreme ultraviolet (XUV) by the generation of high odd-integer harmonic orders of a fundamental laser frequency [1–9]. The majority of this work has been performed using relatively long wavelength lasers; in particular, 1.053-μm neodymium-doped yttrium aluminum garnet (Nd:YAG) or 800-nm Ti:sapphire systems [2–4]. It has been reasoned that harmonics with the highest photon energy will be produced with such long wavelength drivers, as the furthest extent of the plateau in the atomic harmonic response for a given atom has been found both experimentally and theoretically to be given by $E_{max}\simeq I_p+3U_p$ [7], where E_{max} is the highest harmonic photon energy, I_p is the ionization potential, and U_p the ponderomotive energy ($U_p=e^2E^2/4m_e\omega^2$). These results are supported by simple semiclassical models [10,11], as well as more rigorous time-dependent solutions of Schrödinger's equation, using the single active electron (SAE) approximation [7,12]. The shortest harmonic wavelengths generated previously are the 109th harmonic (74 Å) of a Ti:sapphire laser (800 nm) using neon [4], and the 141st harmonic (75 Å) of a Nd:glass laser (1.053 μm) using helium [13].

This approach of using the quadratic scaling of ponderomotive energy with fundamental laser wavelength to produce the highest harmonic photon energy neglects generation from ions relative to that from neutral atoms. In principle, ions could produce higher-energy harmonics due to both their higher ionization potential and the correspondingly high ponderomotive energy at their saturation intensity. At the longer laser wavelengths, the harmonic signal from ions is, at best, very weak in comparison to the neutral response [14]. In contrast, Sarukura *et al.* [5] recently produced high-order harmonics from a short wavelength (248.6 nm) KrF laser, generating up to the 23rd harmonic in helium and up to the 27th harmonic in neon. An analysis of their results by Krause *et al.* [7] showed that harmonics from helium above the 13th were due to He^+, and suggested that the higher harmonics in neon were due to Ne^+. Kondo *et al.* [6] attributed neon harmonics above the 21st in the same experiment to emission from Ne^{2+}.

The generation of high harmonics from ions using short wavelength lasers can be attributed to two effects. First, the phase mismatch between fundamental and harmonic is less for short wavelengths when the refractive index is dominated by free electrons, as is the case when the gas is ionized [8]. Second, the single-atom response has been shown to be greater for short wavelength lasers [15]. Using the semiclassical model this can be attributed to the shorter available time for diffusion of the electron wave function between tunneling through the Coulomb barrier and the first recollision with the core: this time being one-half of the laser cycle. In this simple model, with significant harmonic generation occurring only on the first recollision, the reduced diffusion increases the cross section for harmonic generation [10,11]. Thus, with harmonic generation from ions already demonstrated, it is timely to investigate whether higher harmonic photon energies can be produced from ions using short wavelength lasers, than from neutral atoms with longer wavelength lasers.

We report here quantitative studies of harmonic generation from a KrF laser, with observation from the 7th (355 Å) to 35th harmonic (71 Å) in neon, and up to the 37th harmonic (67 Å) in helium. To our knowledge these results constitute the shortest harmonic wavelengths recorded to date—the 37th harmonic of 248.6 nm being approximately equivalent in photon energy to the 157th of 1.053 μm. We show that these results are in agreement with simulations of the single-atom and single-ion responses of helium and neon at the intensities present in our laser focus, and we attribute the highest harmonics in the neon targets to the response of doubly-ionized neon. These results conclusively demonstrate the potential of short wavelength lasers to produce high-energy harmonics.

The experiments were performed using the Sprite KrF laser at the Central Laser Facility of the Rutherford Appleton Laboratory. The laser, used in chirped pulse amplification mode [16], delivered up to 250 mJ on target in a 380-fs full width at half maximum (FWHM) pulse at 248.6 nm. The beam was focused using two different off-axis parabolas (OAPs). First, a 33-cm OAP was used to focus the 10-cm-diam beam to an average focal spot in vacuum of approximately 10 μm FWHM (ten times the diffraction limit, but

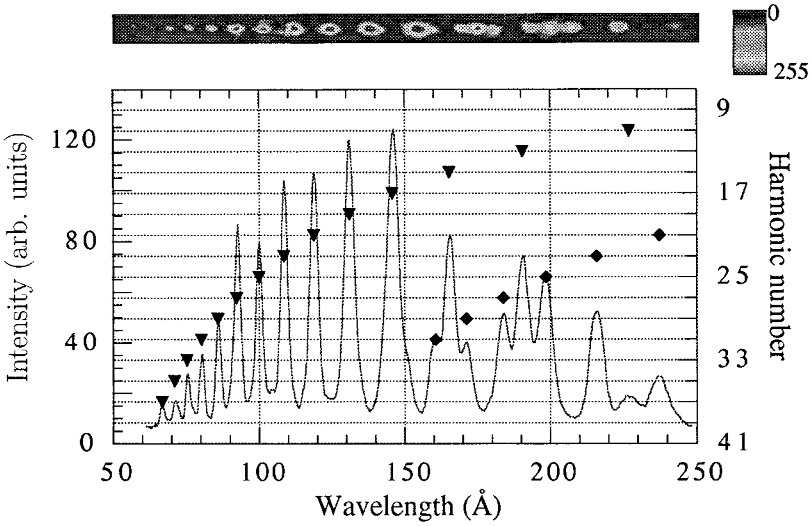

FIG. 1. Image and line-out from a typical harmonic spectrum from helium using the 100-cm OAP. The gas density is about 5×10^{17} atoms/cm^3. The first-order (▽) and second-order (◇) grating dispersion curves are shown (right axis).

with variable smaller scale structure down to the diffraction limit). This gave a mean intensity of 4×10^{17} W/cm^2 over a focal depth of about 100 μm. Second, a 100-cm OAP produced, on average, a six times diffraction-limited focal spot of 18 μm FWHM with a mean focused intensity of just over 10^{17} W/cm^2 over a length of approximately 700 μm. A solenoid-valve gas jet provided a gas target of helium or neon at the laser focus, with atomic densities ranging from 10^{16} to 10^{19} cm^{-3} and gas lengths between 0.5 and 2 mm. Densities produced at the focus were determined by Thomson scattering [17].

The harmonic emission was detected with a slitless flat-field grazing incidence XUV spectrometer (1200 line/mm Hitachi grating at 3.77°) used with an additional gold-coated grazing incidence cylindrical mirror. The grating and mirror were oriented so that their grazing incidence reflections gave perpendicular astigmatic line images of the focus of the laser beam at the detector plane. Their combined effect therefore produced a series of dispersed monochromatic images of the focus at each harmonic wavelength as shown in Fig. 1. The XUV images were detected with a Galileo double microchannel plate (MCP) coupled to a Photonic Science DarkStar intensified charge-coupled-device (CCD) camera via a 4:1 reducing fiber-optic coupler. The response of the MCP and CCD camera was calibrated with respect to Ilford Q-plate film in a separate experiment. The total spectral range on the MCP was 190 Å, with a resolution of ~1 Å. The MCP could be moved in the spectrometer dispersion direction to give a wavelength coverage from 0 to 400 Å. The acceptance solid angle of the grating and gold mirror was 1.5×10^{-5} sr. Filters of 20–200 μg/cm^2 of carbon or 27–108 μg/cm^2 of aluminum were used to eliminate the fundamental laser light and to attenuate the bright lower order harmonics. In addition, a beam block was inserted in the zero-order position to eliminate scattered XUV radiation.

A typical single-shot harmonic spectrum for helium is shown in Fig. 1. This was obtained with the 100-cm OAP and an atomic density of $\sim5\times10^{17}$ cm^{-3}. This plot is uncorrected for instrument or filter response. High harmonics up to the 37th can clearly be seen in the first-order diffracted signal. There is a relatively high level of the spectrum in second order, which is due to the blaze angle of the grating becoming optimized for second-order diffraction below about 100 Å. The response of the detector was proportional to the harmonic energy per unit solid angle.

Harmonic energy conversion efficiencies were calculated assuming that the total solid angle of emission was equal to that of the laser beam. Further calibration factors came from the absolute response of the MCP, the grating reflectivity [18], the filter transmission, the collection solid angle of the spectrometer, the focusing mirror reflectivity, and its collection solid angle. The angular distribution of the harmonics was measured in a separate series of experiments using the 33-cm OAP, without the focusing mirror [19]. The absence of the mirror reduced the overall detection sensitivity, and thus in these experiments only harmonics up to the 19th were observed. However, all the harmonics observed covered the full solid angle of the laser beam. For this reason the energy conversion measurements assume emission in the full beam angle. The assumption is validated for harmonics up to the 19th, but conversion into higher harmonics could be overestimated if the angular distribution of the beam were to narrow significantly. An upper bound of a factor of 40 on the possible error for data recorded with the 100-cm OAP is given by the fact that the acceptance solid angle of the spectrometer was 1/40th that of the laser beam. Figure 2 shows the conversion efficiencies with the 100-cm OAP for the harmonics above the 7th in both (a) helium and (b) neon. It can be seen that 30 nJ was generated at 355 Å (7th harmonic), i.e., a conversion efficiency of 10^{-7}. It is interesting to compare these efficiencies with the highest reported elsewhere [8], which were obtained using 526-nm light. Our efficiency at 355 Å is approximately the same as that at 251 Å from 526 nm, and at approximately the same XUV wavelength these efficiencies are about a factor of 2 lower. It remains to be explored whether further increase in efficiency with the KrF laser would be possible with a larger focal spot and longer confocal parameter, as was used in the 526-nm work [8]. The present results certainly suggest that the trend towards better conversion efficiency at a given harmonic wavelength using a shorter primary laser wavelength may continue beyond 526 nm.

For both helium and neon the production of the highest harmonics was largely independent of gas density until the density was lowered below a critical value, which was found to be approximately 10^{17} atoms/cm^3 for helium and neon.

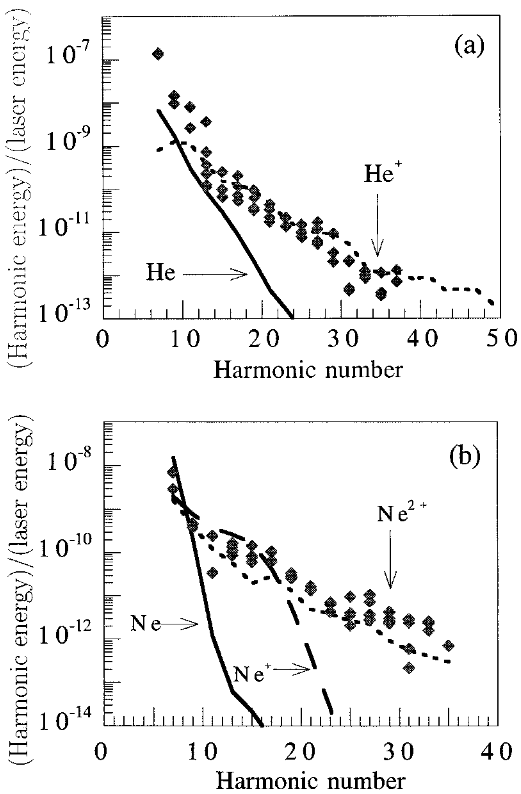

FIG. 2. Absolute harmonic conversion efficiencies (harmonic energy)/(laser energy) for (a) helium and (b) neon. Experimental data (◇) are shown alongside numerical simulations for the weighted contributions of individual neutral-atom–ion stages, as described in the text. (The whole theoretical response has been multiplied by an arbitrary factor.)

These densities are consistent with this being the point where the depth of focus (which is of the same order as the gas width) becomes shorter than the coherence length, which is in turn determined by the free-electron density because, as will be shown below, the higher harmonics are due to ions rather than neutral atoms.

Furthermore, for helium, the production of high harmonics was also almost independent of the focusing of the laser beam: the 37th harmonic being observed with both the 33- and 100-cm OAPs. In contrast, the neon response appeared to be much more sensitive to the focusing. The 33-cm OAP produced up to the 23rd harmonic and the 100-cm OAP produced up to the 35th in the same atomic densities as for helium. The difference between the 33- and 100-cm OAPs for neon is attributed to the much higher ionization rate of Ne^{2+} compared to He^{+} at the higher intensities produced by the short focal length parabola, the species generating the harmonics being depleted more rapidly in neon than in helium.

The relationship between single-atom calculations and the data recorded in an experiment is not straightforward because the net generation depends on the time- and phase-dependent integration over the whole focal region. Although single-atom calculations can take into account the temporal variations in laser intensity, they ignore spatial variations of intensity, variations of phase, and propagation effects.

Three dimensional numerical simulations for helium are performed in the SAE approximation [7,12,20]. We calculate harmonic generation from the neutral atom and the ion by solving the corresponding time-dependent Schrödinger equation. The ion response is weighted by the time-dependent ion-population obtained from the ionization rate of the neutral atom. In this way, the problem of solving the response of an N-electron atom during sequential ionization is reduced to solving N one-electron correlated wave functions. Full details of the model can be found elsewhere [20]. This model is generally too computationally expensive to use with neon, due to the greater number of ionization stages and bound electrons involved; we have therefore used a one dimensional soft core model of the form $V(x) = -Z_{eff}/\sqrt{a^2+x^2}$ to calculate the response of neon and its ions, where a is chosen for each ionization stage to give the correct ionization potential.

It is generally the case that the harmonics observed will have been generated from atoms and ions within approximately the last coherence length before the beam exits the gas jet, and this coherence length is inversely proportional to the harmonic order when the refractive index is dominated by free electrons. As a first approximation to the real experimental situation, we have linearly combined several single-atom and single-ion calculations, each performed at a different peak intensity ranging from 10^{15} to 10^{17} W/cm^2, with each individual calculation weighted for the relative volume in the focus at which its peak intensity occurred (assuming a Gaussian, TEM_{00}, beam profile). We have also multiplied the data by a factor of q^{-2}, where q is the harmonic number, to allow for the scaling with q of the coherence length (which is shorter than the confocal parameter). The overall result of this process is to enhance the generation efficiency for the lower harmonics with respect to the higher harmonics, as they will be produced over a much larger volume and will have a longer coherence length. Even this simple weighting significantly alters the predicted harmonic response. The higher harmonic efficiencies are reduced by up to two orders of magnitude with respect to the low harmonics when compared to the uniform intensity response. This method is only a rough approximation and to model the temporal and spatial dependence, the single-atom response should ideally be incorporated into a beam propagation code.

The simulated single-atom and single-ion responses for a Gaussian focus for helium and neon are shown in the Figs. 2(a) and 2(b). It can be seen that there is good qualitative agreement between the simulated and experimental results. The simulation appears to significantly underestimate the generation of the lower harmonics from neutral helium, which can be attributed to propagation effects and confirms the need for a full propagation code. The simulation does predict the change in response at the 13th-15th harmonic where He^{+} takes over from the neutral atom as the dominant response. For the neon data, the experimentally observed change in response at the 9th and 19th-21st harmonic is well reproduced by the simulations. This is due to the transition from neutral neon to Ne^{+} and from Ne^{+} to Ne^{2+}, respectively, as the dominant harmonic generator. It can be seen, in agreement with Kondo, that Ne^{2+} is responsible for all the harmonics observed above the 21st.

The observation of harmonics from Ne^{2+} raises the question of the response of higher ionization stages. Indeed, the saturation intensities of Ne, Ne^{+}, and Ne^{2+} are below

10^{17} W/cm^2 and therefore they are mostly ionized before the experimental peak laser intensity. Simulations of Ne^{3+} indicate that its response should become comparable to that of Ne^{2+} around the 35th harmonic, and certainly any harmonics beyond this point could have been attributed to Ne^{3+} as the dominant response. Although higher ionization states of the atom, i.e., Ne^{4+} or beyond, have higher saturation intensities and could therefore experience larger intensities before being completely ionized, calculations of their contribution to the harmonic generation show it to be negligible. An explanation for this in terms of the simple semiclassical model is that the higher ions require a much higher intensity to ionize, so that the oscillatory velocity of the freed electron at recollision with the core is so high that the cross section is too low to give significant conversion [21].

Also of note is the difference in conversion efficiencies between helium and neon. The 7th, 9th, and 11th harmonic efficiencies for helium are over an order of magnitude greater than those for neon. After this the difference is less, but with the helium efficiency remaining higher up until the 31st harmonic. This is contrary to the work of Kondo *et al.* [6], which showed helium and neon with approximately the same response from the 9th to the 23rd harmonic, but with helium marginally lower after the 13th.

In conclusion, we have studied the generation of high harmonics of an ultra-short-pulse KrF laser in helium and neon gas jets. We demonstrate the generation of shorter harmonic wavelengths than in previous work, up to the 37th harmonic (67 Å) in helium and the 35th harmonic (71 Å) in neon. The source of the harmonics is shown to be the ion species He^{+}, Ne^{+}, and Ne^{2+}. We also demonstrate that helium can produce very high conversion efficiencies for the lower harmonics of 248.6-nm laser radiation, giving energies up to 31 nJ and powers up to 80 kW, for the 7th harmonic (355 Å). These results prove the potential of short wavelength driver lasers for the efficient production of high-energy XUV radiation by harmonic generation.

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