High-Order Harmonic Generation Using Intense Femtosecond Pulses

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Neon gas excited by 800-nm laser pulses (15 mJ, 125 fsec) at an intensity near 10^{15} W/cm^2 generates harmonics up to the 109th order. The appearance of successively higher harmonics as the laser intensity is increased is compared to recent calculations of the strong-field atomic response. Blueshifting of the laser and harmonic wavelengths indicates a small degree of ionization until the threshold for the highest harmonics (>91st) is reached.

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Atoms driven by a strong laser field can produce coherent harmonic radiation at photon energies far exceeding the atomic binding energy [1,2]. This nonperturbative atomic response has been examined extensively [3], and the role of phase matching considered [4,5], for moderate laser intensities up to about 10^{14} W/cm^2. Recent experiments have now demonstrated generation below 10 nm using He or Ne gas, but have required vastly different intensities. Sarukura *et al.* [6] obtained the 25th harmonic of 248 nm using greater than 10^{17} W/cm^2, whereas L'Huillier and Balcou [7] observed up to the 135th harmonic of 1053 nm using about 10^{15} W/cm^2. A description of harmonic generation at these high intensities is of great interest in order to determine how to develop the harmonics as a useful source of coherent soft x rays.

Recently Krause, Schafer, and Kulander [8] reported calculations of the harmonic response for both atoms and ions at intensities that produce ionization rates up to a few times 10^{12} s^{-1}. They found a general scaling of the height and breadth of the single-atom harmonic spectrum: The emission rate of the spectrum tracks the ionization rate, while the breadth of the harmonic spectrum extends to a cutoff harmonic at a photon energy equal to $\approx I_p + 3U_p$, where I_p is the atom (or ion) ionization potential, and U_p is the ponderomotive energy, $e^2E^2/4m\omega^2$, for the laser field E at frequency ω. They predict that very high harmonics can be obtained from atoms at modest ionization rates for a long-wavelength driving laser, and their scaling can explain the maximum harmonic observed in recent experiments [6,7,9–11].

As a result of the work of Krause and collaborators, various analytical models for strong-field harmonic generation may be tested against the numerical calculations. One promising model of strong-field harmonic generation has been proposed by Becker, Long, and McIver [12]. These authors considered harmonic generation from a model atom with a short-range potential that supports one bound state and a continuum. Although they reported that the spectrum cuts off at a photon energy given by $2(I_p + U_p)$, their solution actually predicts the harmonic plateau extends out to an energy equal to $I_p + 3.2U_p$. The agreement with the Krause result suggests that this model may provide insight into the strong-field atomic response, and allow calculations of the macroscopic polarization of the atomic medium to be performed with relative ease.

In this Letter we describe very-high-order harmonic generation from neutral Ne atoms. We use femtosecond, 806-nm laser pulses (15 mJ, 125 fsec), loosely focused in Ne gas, to produce harmonics up to the 109th order. The use of femtosecond pulses reduces the deleterious effects of ionization for most of the harmonics, allowing us to examine the intensity dependence of the emission over a large range of harmonic photon energy. We report the intensity dependence for a sequence of harmonics over a range of $(0.1-1)\times10^{15}$ W/cm^2, and show that the dipole moment obtained from the model of Becker, Long, and McIver [12], when integrated over the interaction volume, can qualitatively describe our results. Our measurements also support the scaling predicted by Krause, Schafer, and Kulander [8], that several times the ponderomotive energy roughly determines the extent of the harmonic response. In addition, we find the degree of ionization of the gas by examining the blueshift of the laser spectrum, and estimate about 60% of the atoms survive the measured intensity ($\sim10^{15}$ W/cm^2) that produces the 101st harmonic.

The harmonics are generated by focusing linearly polarized, 806.5-nm laser pulses into a thin slab of Ne gas. A 5-Hz, Ti:sapphire laser [13] provides up to 35 mJ in a near-Gaussian beam (12.5 nm e^{-2} radius). The laser pulse width is 125 fsec, assuming a sech^2 pulse shape. A 60-cm lens focuses the beam into a vacuum cell and through a Ne gas tube. Laser pulses propagate perpendicular to the tube axis, through two small holes (350 μm diameter) drilled in the tube. When the tube is pressurized with Ne, the small conductance of the holes confines the gas approximately to the tube i.d., which is 1 mm thick. The surrounding cell is evacuated to 20 mTorr for a tube pressure of 25 Torr. A grazing-incidence monochromator views the harmonic radiation on the laser axis. The monochromator employs a 1200-line/mm grating at an incidence angle of 88°, with a blaze wavelength of 7.6 nm. The entrance slit is located 40 cm from the laser beam waist to prevent damage to the monochromator

grating. The harmonic radiation is detected by either an electron multiplier or a microchannel plate detector.

Phase matching of higher harmonics improves when the confocal parameter is much larger than the gas interaction length [5]. A large confocal length is obtained here at the expense of laser energy by placing an iris before the focusing lens. Beam truncation with an 8.5–12-mm variable iris yields a confocal parameter for the focused beam of 25–13 mm. Relative changes in focused intensity are determined in the absence of the gas by imaging the focal spot at 10× magnification onto a thin KDP crystal and monitoring the frequency-doubled light. The absolute intensity is found by replacing the KDP crystal with six different pinholes and measuring encircled energy. This provides a course measure of the beam profile, with the smallest pinhole measuring the peak fluence. The peak fluence is about 2 times smaller, and the beam spot size about 1.2 times larger, than that calculated for an ideal truncated-Gaussian beam. More energy appears in the spatial wings of the focused beam, resulting in a 50% reduction in peak intensity from that possible with our laser power. We estimate less than a factor of 2 uncertainty in the absolute measurements.

Figure 1 shows the intensity dependence of the harmonic radiation for harmonic orders 31 through 101, obtained with 16 Torr of Ne, using the electron multiplier detector. Each data point plotted represents the mean value of 30 laser shots. The iris is set to obtain a large confocal length with an intensity range necessary to produce the various harmonics (an 8.5-mm iris for harmonics 31 through 51; 10-mm for 61 through 81; and 12-mm for 91 and 101). The most conspicuous feature of the data, particularly as the harmonic order increases, is the dramatic change in the slope of the growth of the emission. Each harmonic initially follows a near power-law dependence, with exponents that are smaller than the harmonic order, so this is not a perturbative response. Above some critical intensity, the emission deviates from an initially steep dependence to a much slower or flat growth. This critical intensity, which due to the steepness of the growth may be somewhat arbitrarily defined for each harmonic, depends linearly on the harmonic order, and can quantify when a particular order becomes part of the slowly varying plateau portion of the harmonic spectrum.

To attempt to determine the macroscopic response, we have calculated the number of harmonic photons generated by the gas as a function of laser intensity, using the method described by L'Huillier, Schafer, and Kulander [4]. This is shown as the solid curves in Fig. 1. The dipole moment, obtained from the model proposed by Becker, Long, and McIver [12], is calculated versus intensity, an example of which is shown in Fig. 2. The spatial dependence of the microscopic polarization in the gas then follows from the spatial dependence of the laser intensity, which we take to be Gaussian with confocal parameter set by the iris and focal length used for the different harmonics. We find the harmonic radiation in the far field by integrating the induced polarization over the interaction volume. This far-field intensity is integrated over time and area to yield the total number of harmonic photons. Since neon at 16 Torr is nearly dispersionless, we take Δk_q equal to zero. We also neglect the effect of ionization, and the small effect of loss at the harmonic frequency due to single-photon photoionization. We assume the Ne gas varies only in the z direction (the propagation direction), with a profile $1/[1+(2|z|/L)^3]$, where L is taken to be 1 mm. In order to match the data, we shift the calculated curves along the x axis by 1.4 times for harmonics 31–51, and by 1.3 times for harmonics 61–81, suggesting our measured intensities are too high by 40% and 30%, respectively. The numbers at the top of each curve are the amount by which the calculated curve has been multiplied along the y axis. This arises in part because we do not know the relative wavelength sensitivity of our detection system. Finally, for the

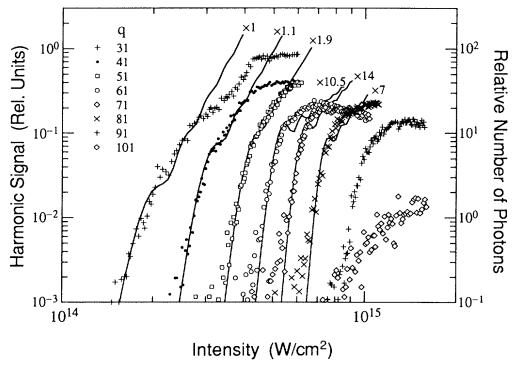

FIG. 1. Harmonic emission vs incident intensity for the 31st through 101st harmonics, at 16 Torr Ne. Solid lines are the relative number of harmonic photons predicted by integrating a model dipole response over the interaction volume.

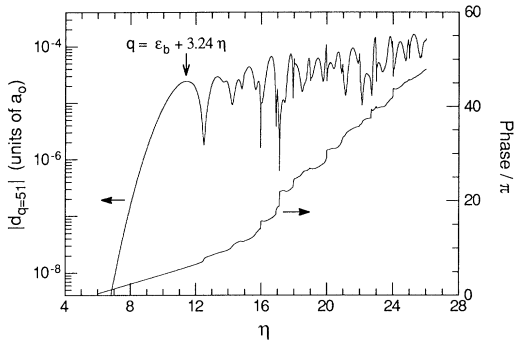

FIG. 2. Calculated magnitude and phase of the single-atom dipole response at the 51st harmonic, vs η ($\eta = U_p/\hbar\omega$), from the model of Becker, Long, and McIver [12]. The first maximum of the response occurs when $q = \varepsilon_b + 3.24\eta$, where ε_b is the Ne ionization potential I_p divided by $\hbar\omega$.

calculated curves shown, we have located the beam waist 2 mm before the center of the gas profile. The predicted emission is increased, particularly for the 61st harmonic, over that found when the beam waist coincides with the center of the gas profile. The choice of $L=1$ mm is very close to twice the coherence length for the 61st harmonic, producing a strong suppression of this harmonic in the calculation which is not observed in the experiment. A shift of the beam waist by 2 mm (10% of the confocal parameter) significantly enhances the 61st harmonic, and yields a more reasonable comparison to the data.

The predicted curves provide very good agreement with the shape of the measured emission, although the predicted curves never go flat as do the data. We have calculated the emission for a variety of gas profiles and confocal parameters, and find the initially steep slope that is followed by a reduced growth is preserved. It is the relative amplitude which is a strong function of the focusing geometry. In particular, we find a very strong enhancement with increasing confocal parameter. We believe this is a key parameter, since increases in gas density or gas length will be limited by loss at the harmonic frequencies due to photoionization. This loss limits the density-length product to about $2/\sigma_{\rm PI}$ ($\sim 4\times 10^{17}$ cm^{-2} in Ne), where $\sigma_{\rm PI}$ is the photoionization cross section.

To show how the single-atom response depends on intensity, we plot in Fig. 2 the intensity dependence of the 51st harmonic, predicted by the model of Becker *et al.* [12,14]. Both magnitude $|d_q|$ and phase are plotted versus η, which is the ratio of ponderomotive energy U_p to laser photon energy $\hbar\omega$. Comparing Fig. 2 to the curves of Fig. 1, it is apparent that the initially steep dependence on intensity for the macroscopic emission derives from the single-atom response. As indicated in Fig. 2, this response first levels off at a value of η such that $q=\varepsilon_b+3.24\eta$, where $\varepsilon_b=I_p/\hbar\omega$. This is the scaling identified by Krause, Schafer, and Kulander [8]. The phase of the dipole initially grows as 3.24η, and appears to sweep through resonances at some integer values of η. The amplitude structure that appears with increasing intensity is similar to that found by Krause, Schafer, and Kulander [3], who interpret this as interference between the different paths which contribute to the qth-order harmonic response. We have not yet attempted to employ this model to describe the highest harmonics observed.

Generation of harmonics above about the 91st order requires an intensity that results in a large ionization rate, and hence a significant fraction of ions. We cannot unambiguously determine the relative contribution of the atom and the ion to the macroscopic emission, since both will contribute an amount determined by their polarizability and density. In our experiment, we monitor electron density (and hence ion density) by a blueshift of the laser and harmonic wavelengths that results from gas ionization. A decreasing refractive index as electrons ionize during the pulse is well known to blueshift the laser spectrum [15]. The harmonic blueshift, not previously seen, can result from up-conversion of the laser shift [5].

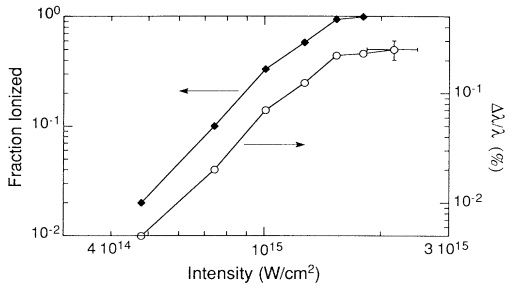

FIG. 3. Fractional blueshift $\Delta\lambda/\lambda$ (open circles) of the transmitted laser spectrum for 24 Torr of Ne, and fraction of atoms ionized (solid diamonds), estimated from the blueshift.

Figure 3 shows the fractional blueshift of the laser spectrum after passing through 24 Torr of Ne, at several values of incident intensity. This is obtained by imaging the pulse onto a 0.25-m spectrometer, and viewing the central portion of the beam. Although the shift appears to level off above 2×10^{15} W/cm^2 (not shown), the shift again increases to a level of about 0.5%. We believe this second increase in shift is associated with the ionization of Ne^+. Also shown in Fig. 3 is the fraction of atoms ionized by the end of the pulse, based on a simple model of ionization-induced phase modulation. The on-axis laser blueshift is found by taking the Fourier transform of a phase-modulated laser field, with a time-dependent phase proportional to the electron density $n_e(t)$,

$$n_e(t)=n_a^0\left\{1-\exp\left[-\int_{-\infty}^{t}W(t')dt'\right]\right\}, \qquad (1)$$

where n_a^0 is the initial atom density. For the ionization rate, we use $W(t)=W_0[\mathrm{sech}^2(t/\tau)]^5$, where W_0 is the rate at the peak intensity I_0, τ is 0.57 times the laser pulse width, and $\mathrm{sech}^2(t/\tau)$ is the laser pulse shape $I(t)/I_0$. In this form $W(t)$ is proportional to $[I(t)]^5$, where a 5th power approximates the slope of a Keldysh ionization rate over the range 10^{12}–10^{14} s^{-1}.

Blueshifts are calculated using $W(t)$ above for a range of values of peak rate W_0. By comparing each measured shift to the shifts calculated versus W_0, we identify W_0 versus measured intensity. For example, the 0.07% shift observed at 10^{15} W/cm^2 requires $W_0\approx 7\times 10^{12}$ s^{-1}. From Eq. (1), the electron density at the end of the pulse indicates 60% of the atoms survive ionization. Since harmonics up to the 101st order appear below 10^{15} W/cm^2, these harmonics are consistent with generation from the neutral atom. However, for the 0.22% shift observed at 1.5×10^{15} W/cm^2, we find $W_0\approx 5\times 10^{13}$ s^{-1}, and over 90% of the atoms ionize, mainly during the first half of the pulse. Under these conditions, harmonic generation from neutral atoms can likely occur only in the leading

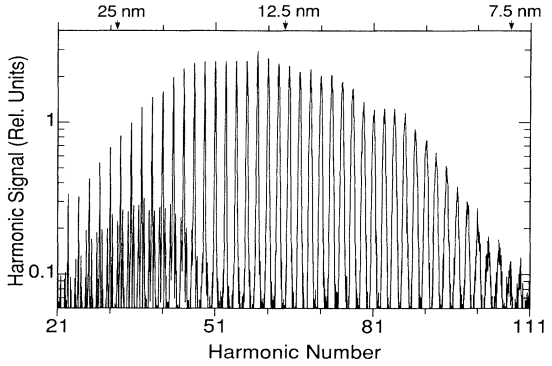

FIG. 4. Harmonic spectrum at 1.3×10^{15} W/cm^2, for 13 Torr of Ne in a 2.5-mm tube. The features running through the low harmonics are high harmonics appearing in second order of the monochromator grating. The spectral width of individual harmonics is instrument limited.

edge of the pulse, or from a spatially extended volume that excludes the high-intensity center portion of the beam.

The relatively small effect of ionization for all but the highest harmonics is seen in Fig. 4, obtained at approximately 1.3×10^{15} W/cm^2 with a 2.5-mm gas tube at 13 Torr, although it is typical of that obtained with the 1-mm tube. For this spectrum, we use the microchannel plate detector. The spectral resolution of the harmonics is instrument limited by the use of 70-μm slit widths. Calibration lines at 16 nm indicate that the resolution at least doubles the true spectral width. The roll-off at low frequencies is expected from the monochromator grating response. A key point is that the spectrum varies very slowly out to the 87th harmonic, and then slowly decreases into the noise at around the 109th. This roll-off is consistent with depletion of neutral atoms by ionization, and reduced phase matching due to free electrons. Both effects blur any sharp demarcation in the maximum harmonic order produced by the atom. We have looked extensively for still higher harmonics but have not found any, so that at our wavelength, approximately 109 is the maximum order that can practically be generated in Ne.

This work demonstrates high-order nonlinear optics in Ne gas using intense femtosecond laser pulses at 800 nm. Our measurements of the generated harmonics appear to support the scaling proposed by Krause, Schafer, and Kulander [8], and the simple model of Becker, Long, and McIver [12]. Phase matching may be determined for the high harmonics using the model atom dipole moment. As a practical matter, very high harmonics can be obtained at a low degree of ionization. Shorter optical pulses may further reduce the degree of ionization to achieve a high peak intensity. A pulse width reduction of 3 requires $\sim$5 mJ to obtain the power used in this experiment. This power may soon be available at higher pulse-repetition rates, and allow kilohertz generation of coherent soft x rays.

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