Calculations of Multiple-Harmonic Conversion of 1064-nm Radiation in Xe

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The relative strengths of harmonic intensities, generated in xenon by a strong infrared laser, are calculated from the time-dependent dynamics of the electronic charge density. The (nonperturbative) solution of the time-dependent Schrödinger equation for a single-electron model of the atomic system is found to predict line strengths in reasonable agreement with the observed intensities. A single time-dependent calculation provides the intensity predictions for all orders for a given laser wavelength and intensity.

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Recent experiments by Ferray *et al.*[1] have revealed efficient generation of high harmonics (up to order 31) by pulsed 1064-nm-wavelength radiation incident on xenon, krypton, and argon with number densities in the range of 10^{17} to 10^{18} atoms/cm^3. At the intensities of the experiment (10^{13} to 10^{14} W/cm^2) the gases become highly ionized by multiphoton ionization during the course of the pulse. These experiments complement earlier results obtained at shorter wavelengths by McPherson *et al.*[2] on these same systems.

Although the lower harmonics (less than eleventh order in Xe) may be regarded as generated by dipole moments to virtual bound levels, it has been predicted that higher harmonics involve free-free dipole transitions or, equivalently, a dressed continuum.[3]

The published results of Ferray *et al.*[1] for intensities of 3×10^{13} W/cm^2 show a strong third harmonic followed by a succession of odd harmonics observable out to a maximum value (21st order in xenon). Although the decrement between third and fifth harmonic is more than an order of magnitude,[4] the relative intensities of the higher harmonics fall only by an order of magnitude. The intensity appears to fall significantly for possible harmonics higher than 21st order.

Theoretical predictions of harmonic intensities require two steps. First one must determine, using the time-dependent Schrödinger equation (or more general Bloch equation) for a given electric field, the atomic response and thence the polarization field. Next one must determine, for this polarization field, the emerging radiation field predicted by a paraxial wave equation (or by more general Maxwell equations). Although the general problem of pulsed and focused radiation traveling through a responsive medium requires elaborate numerical simulation, simplified approximations are available.[5] The experiments of Ferray *et al.* were done with a focused beam in which the confocal parameter was larger than the propagation length. Treatments of harmonic generation of focused Gaussian beams based on perturbation theory (nonlinear susceptibility)[5–7] show that, as the propagation distance increases, the harmonic power depends not only on the harmonic distribution of the polarization field but on phase-matching conditions. One can show that, to a good approximation, in the case of a loosely focused Gaussian beam (a reasonable approximation to the experiments) traveling through a uniform nonlinear medium the emerging power in the optical harmonics is directly proportional to the single-atom spectrum, except in the case of a near-resonant excitation by a particular harmonic. In what follows we shall therefore assume that our calculated single-atom results provide a reasonable description of the relative distribution of harmonic intensities emerging from a small source. Corrections to this result may increase the relative high-order harmonics by up to an order of magnitude. (A detailed description of this approximation is being prepared for publication.)

We have carried out a numerical simulation of these experiments using a model in which the atoms are assumed to be uniformly distributed within a volume that receives uniform illumination from a time-dependent pulse. We model the field as a ramped amplitude (over five optical cycles) followed by a steady amplitude equal to that of the peak experimental intensity (typically for fifteen to twenty optical cycles). The atomic dynamics is treated with the model developed by Kulander,[8–10] in which each xenon atom is idealized as a single electron moving in an effective electrostatic potential (a frozen-core Hartree-Slater potential). We calculate the ground-state orbitals for the outer 5*s* and 5*p* electrons, then freeze all but one of the electrons. Thus there are no collective excitation effects in this single-electron model. We determine the time evolution of the wave function for the unfrozen electron interacting with the time-varying uniform electric field (electric-dipole interaction). For this purpose we integrate a three-dimensional time-dependent Schrödinger equation within a finite spatial volume, using a finite difference scheme,[8] to obtain a time-dependent wave function. Our procedure allows probability to flow irreversibly out of the finite volume, thereby modeling ionization. We determine the ionization probability by evaluating the flux through an absorbing boundary. From our wave functions $\Psi(r,t)$ we determine the time-varying expectation value of the dipole moment. In atomic units this value is

$$\langle d(t)\rangle = \int d^3r\,\Psi(r,t)^* z \Psi(r,t)\,. \tag{1}$$

This time-varying atomic dipole moment, assumed to be distributed uniformly within the (macroscopic) excitation volume with a phase determined by the pump field, provides the polarization field responsible for the generation of optical harmonics. When perturbation theory provides an accurate description of the atomic response then the Fourier transform of this dipole moment, when multiplied by the atomic number density and divided by the appropriate power of the electric field amplitude, provides the susceptibility (linear or nonlinear) at the harmonic frequencies. It should be emphasized that, although we use a simplified model of the Xe atom, our solutions are not obtained through perturbation theory. Our dipole moment, and the derived polarization field and spectrum, are not obtained as a series in powers of the electric field, as is customary in traditional nonlinear optics.[5] Our results represent all orders of the nonlinearities; to extract nonlinear susceptibilities it would be necessary that we be able to fit our results to the proper power-law dependence on intensity over a range of intensities. However, at these intensities this procedure is not valid for the higher harmonics.

The square of the Fourier transform of $\langle d(t)\rangle$ gives the distribution of frequencies radiated by the atom and so a plot of this quantity displays the fluorescence spectrum. (This spectrum is often determined from the autocorrelation function, with the invocation of the Wiener-Khinchine theorem[11] and the assumption of a steady process. In our case the direct evaluation of the Fourier transform is much simpler.) Under the approximation mentioned above this quantity provides the relative distribution of optical harmonics emerging from a small sample.

We typically integrate over tens of optical cycles. We observed that, after the initial field amplitude ramp (five cycles), the spectrum became stationary. This will not be the case when the wavelength is close to an n-photon resonance. Over the time interval that we examine, the ionization is still slight (less than 1% for this intensity) and is proceeding at a steady rate. At the chosen frequency we see no evidence for Rabi oscillations; the field frequency is far from resonance with any discrete excited state.

Our results for an intensity of 3×10^{13} W/cm^2 on Xe show a succession of odd harmonics similar to those of Ferray *et al.*[1] for the 5th to 21st harmonic. Figure 1 shows relative values of our spectral intensity (the square of the Fourier transform of the dipole moment). For comparison, we have indicated the experimental results of Ferray *et al.* (who reported only relative values) shifted to coincide at the seventh harmonic. The distribution of harmonics in the range of the fifth to the thirteenth follows very closely their relative values. Our 15th to 21st harmonics are significantly lower than theirs. This may be due to our neglect of collective effects, due to contributions from 5s electrons, or simply due to the experimental intensity being slightly higher than that reported. We find indications of even higher harmonics than those reported, although our 25th harmonic is predicted to be 3 orders of magnitude smaller than their 19th harmonic. The widths of the peaks are the widths appropriate to either the ionization rate of the atom or the overall integration time (pulse bandwidth), whichever is larger.

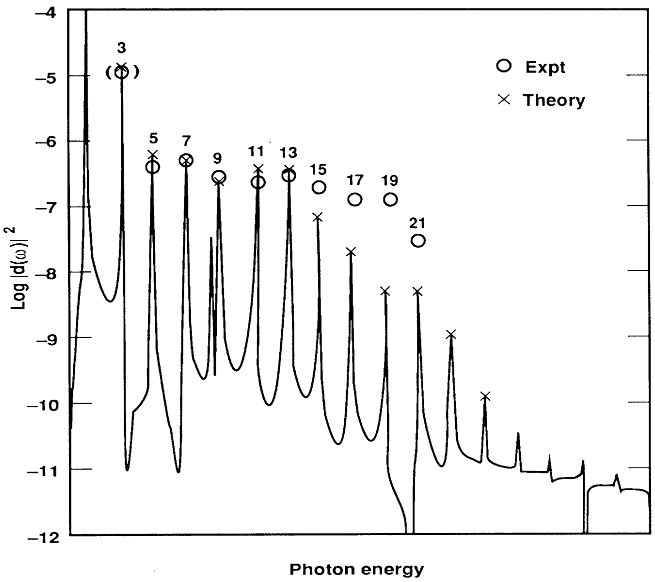

FIG. 1. Predicted and measured harmonic intensities (see Ref. 4). Results are shifted to agree for the seventh harmonic.

We observe a small side peak near the ninth harmonic. We believe that it originates with an atomic resonance transition, probably involving promotion of the 5p electron to a 4f orbital. It becomes relatively less pronounced when we consider longer pulses, and so we conclude that the excitation occurs as a result of nonadiabatic behavior during the pulse rise.

Our calculations show a continuum background underlying the harmonics. We have not elucidated the origin of this continuum, but we find that it is sensitive to pulse duration and shape. It remains at least an order of magnitude weaker than the harmonics. Such background is observed in the experiments.[1]

We have also modeled ionization by weaker (10^{13} W/cm^2) and stronger (10^{14} W/cm^2) fields. The succession of harmonics beyond the third is absent at the lower intensity and is observed out to higher values at the higher intensity. Therefore there appears to be an abrupt threshold for the onset of the production of multiple higher harmonics.

Our results suggest that a simple one-electron atomic model is adequate to model the generation of high optical harmonics by intense fields. Our model predicts a significant increase in the number of orders as the field intensity is increased from 1×10^{13} to 3×10^{13} W/cm^2. To model intense field ionization the only currently available nonperturbative alternative to direct integration of the time-dependent Shcrödinger equation, as discussed here, are methods based on Floquet theory.[12,13] It would be interesting to compare our time-dependent dipole moment with one calculated from the relevant Floquet state.

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