High-Order Harmonic Generation in Atom Clusters

T. D. Donnelly,[1,*] T. Ditmire,[2] K. Neuman,[1] M. D. Perry,[2] and R. W. Falcone[1]
[1]*Department of Physics, University of California at Berkeley, Berkeley, California 94720*
[2]*Lawrence Livermore National Laboratory, P.O. Box 808, L-443, Livermore, California 94550*
(Received 24 August 1995)

We report the generation of short-wavelength, high-order harmonics of intense laser radiation from atom clusters. Clusters containing about 10^3 atoms are produced in a high-pressure gas jet. We show them to be a unique nonlinear medium. Compared with monomer gases they yield a higher appearance intensity for a given harmonic order, stronger nonlinear dependence of harmonic signal on laser intensity, higher-order harmonics, and reduced saturation of the harmonic signal at high laser intensity.

PACS numbers: 42.65.Ky, 32.80.Wr, 36.40.Gk

The interaction of high-intensity laser light with matter can result in the production of high-order harmonics of the laser frequency. The harmonic radiation is coherent and can be generated at photon energies well above the atomic binding energy. Models for such processes inherently involve nonperturbative interaction of the intense laser field with matter. High-order harmonics have been produced in monomer gases [1,2], molecules [3], and at solid surfaces [4,5]. Here we report the generation of high-order harmonics of intense laser radiation from atom clusters containing approximately 10^3 atoms, and show that clusters are a unique nonlinear medium with potentially useful properties.

Monomer gas targets are typically used to produce high-order harmonic radiation in the vacuum ultraviolet from intense, ultrashort-pulse lasers operating in the near visible spectral region. The efficiency of conversion of laser energy to a single harmonic is typically $\leq 10^{-7}$ at wavelengths below 30 nm [2]. A model of these harmonics involves an electron, driven to extreme distances from an atom by the oscillating laser field, radiating harmonics upon collision with the core on its return trajectory [6,7]. Harmonics have also been generated from thin plasma layers at the surface of a laser-irradiated solid; efficiencies on the order of 10^{-4} at somewhat longer wavelengths (200 nm) are observed, along with limited spatial coherence [4,8]. Here the model for harmonic generation involves laser-driven electrons oscillating through the sharp density gradient present at the surface [5].

Clusters of atoms suggest a new type of nonlinear medium for the generation of short-wavelength light. *Recent studies [9,10] have shown that clusters are a highly efficient source of incoherent short-wavelength radiation when irradiated by a high-intensity laser pulse. We show in this Letter that clusters are also a source of coherent short-wavelength radiation.* An intense, ultrashort-pulse laser irradiating a cluster will tend to ionize electrons, producing a microplasma which is held together inertially for time scales on order of the laser pulse (<100 fs), even in the presence of electron heating [10,11]. Compared with the situation of a high field interacting with the valence electron of a single atom described above, electrons in a cluster will be subject to (1) partial shielding of the laser field by *both bound and free* charges, (2) an initial binding potential that is modified from the single atom case by nearest atomic neighbors in the cluster, (3) the Coulomb field of all ions in the cluster (space charge) as electrons are driven away from the cluster, and (4) scattering in the cluster during their oscillatory motion. For the work described in this Letter, the free electron oscillation amplitude (determined by the peak laser intensity) is calculated to be on order of the cluster radius (typically 2 nm).

In our experiments, we focus a short-pulse, high-intensity laser [12] (140 fs, 825 nm) onto argon gas targets including both a pulsed-gas jet and a static-gas cell. Harmonic radiation is collected using an on-axis spectrograph which incorporates a variable-line-spaced grazing-incidence grating with nominally 1200 lines/mm. The radiation is imaged onto a single-stage, multichannel plate electron multiplier that is fiber-optic coupled to a charge-coupled-device (CCD) camera to allow digital recording of the data. The laser beam is clipped using an aperture at the focusing lens which produces a flat-top spatial beam profile and an $f/50$ focusing geometry. We are the in the weak focusing limit [13], with a laser spot size of 50 μm at focus. Although the uncertainty in the determination of the absolute laser intensity is a factor of 2, relative intensities in experimental runs comparing monomer and cluster targets are accurate to several percent.

The 1.5 mm long static-gas cell is at room temperature and contains only monomers. The laser enters and exits (along with harmonics) through small holes drilled in the cell walls by the laser itself using pulses preceding experiments which record harmonics. This short cell is contained in a larger chamber which is under vacuum. Pressure measurements are made in the gas line leading to the cell and monitor the actual pressure in the cell.

The sonic, pulsed-gas jet has an aperture diameter of 600 μm and is operated at room temperature. It produces

 0031-9007/96/76(14)/2472(4)$10.00

clusters containing approximately 500 atoms for a backing pressure of 115 psi [14–16], scaling (nonlinearly) to larger clusters at higher backing pressures [14]. Because the average interparticle spacing in solid Ar is 0.37 nm [17], this implies a cluster radius of approximately 2 nm. We operate in a regime where nearly all of the gas released from the jet will participate in cluster formation [16]. The laser-cluster interaction length near the exit of the jet is approximately 2 mm.

Figure 1 shows the yield of the 23rd harmonic as a function of laser intensity, I (W/cm^2), from (a) monomers (in the static-gas cell) and (b) clusters. Harmonic yield from the Ar monomers (at 40 Torr) is fit initially to the indicated power law, I^9, with saturation near an intensity of 1.5×10^{14} W/cm^2. Harmonic yield from the Ar clusters appears at higher intensities, and is fit (at a backing pressure of 115 psi) initially to the indicated power law, I^{17}, and then to a reduced power law, I^4, at intensities above 2×10^{14} W/cm^2, without an observation of saturation similar to that seen using the monomers. Errors in the power law fits I^n are approximately $n \pm 1$. Increased appearance intensity, increased power law dependence on laser intensity, and reduced saturation are also observed at other high-harmonic orders, although the exact appearance intensity and power law vary slightly with order number. Reproducibility of the appearance intensity for the harmonics was within the absolute intensity calibration (a factor of 2), but monomer and cluster harmonic yields always behaved relatively as shown.

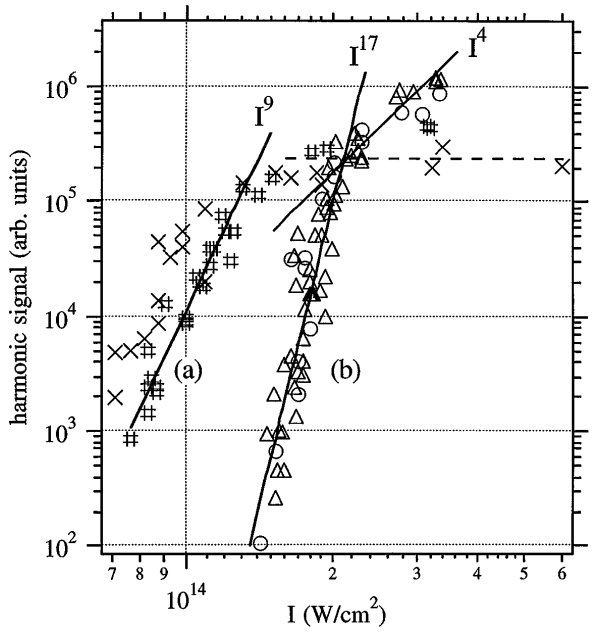

FIG. 1. Intensity of the 23rd harmonic as a function of peak laser intensity from (a) Ar monomers and (b) Ar clusters. Data are shown from: (Δ) sonic jet clusters (115 psi backing pressure), (○) sonic jet clusters (135 psi), (×) static-gas cell (70 Torr), and (#) static-gas cell (40 Torr). - - - indicates saturation of the monomer harmonics. Best power law fits to the data are given.

Figure 2 shows the harmonic spectrum for various laser intensities and targets; spectra were chosen from data generated in the high-laser-intensity regime for each target. Figure 2(a) shows harmonics for a supersonic gas jet, with a backing pressure of 255 psi, and a laser intensity of 9×10^{14} W/cm^2; Fig. 2(b) shows harmonics for a sonic gas jet, with a backing pressure of 115 psi, and a laser intensity of 3×10^{14} W/cm^2; and Fig. 2(c) shows harmonics for a static-gas cell, with a pressure of 50 Torr, and a laser intensity of 6×10^{14} W/cm^2. The backing pressure of the supersonic jet yields a cluster size of $\geq$3000 atoms [16], corresponding to a cluster radius $\geq$3 nm. We indicate specific harmonic orders to draw attention to the increasing value of the harmonic order at which the cutoff appears in the three sets of data (monomer = 29th, sonic jet cluster = 31st, supersonic jet cluster = 33rd). We define the cutoff to be the harmonic at which the signal drops to less than $\frac{1}{4}$ of the signal of the previous harmonic. The monomer data [Fig. 2(c)] are obtained at a peak laser intensity which is more than twice the monomer ionization intensity, demonstrating that the 29th harmonic is the cutoff harmonic. Additionally, the cutoff is observed to be fairly independent of static-gas pressure; Ar clusters have a consistently higher cutoff than monomers and the cluster cutoff is seen to increase with increasing cluster size.

We note that the static-gas cell and the gas jets produce similar average atom densities of about 10^{18} cm^{-3} [18]. This minimizes the contribution of potentially dissimilar phase matching conditions in the monomer and cluster experiments, assuming that the refractive index is linear in average atom density. However, the frequency dispersion

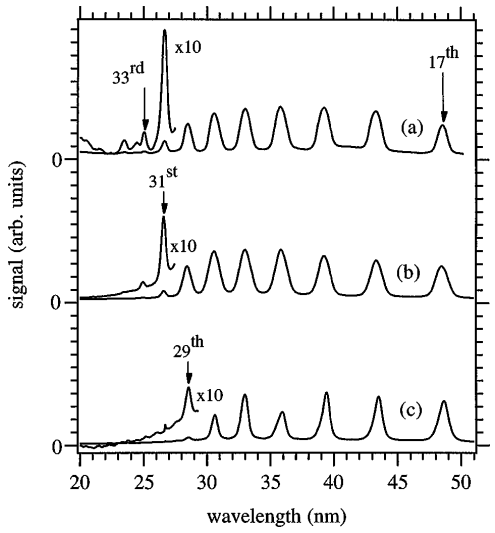

FIG. 2. Intensities of the harmonic spectra from Ar monomers and Ar clusters. The harmonic cutoff wavelength is indicated. All spectra are scaled to unit height: Data are shown from (a) supersonic jet, (b) sonic jet, and (c) static-gas cell.

of a gas of clusters remains a relatively unexplored issue; it is possible that reduced dispersion in the cluster medium results in higher harmonic yields. Finally, we note that since the detection system we employed allows spatial resolution in the plane orthogonal to the spectral dispersion plane of the grating, we could observe that the harmonics from the monomers and the clusters had identical spatial (divergence) properties.

We believe that our observations are consistent with a simple model of harmonic generation from atoms embedded in a solid-density environment. First, we consider the increased appearance intensity in the case of the clusters. We verified that the increase is not due to defocusing of the laser beam by checking the appearance intensity of particular harmonics as a function of jet backing pressure. Increased appearance intensity can be explained by the shielding of the laser field within the cluster. Consider that the field inside a dielectric sphere, relative to an external field, is shielded by a factor of $3(\varepsilon + 2)^{-1}$, where ε is the dielectric constant of the sphere. In our case, ε will reflect contributions from both bound and unbound electrons in the cluster. The index of refraction of solid-density Ar is 1.2; this leads to a reduction in the laser intensity by a factor of 0.7 before the cluster is ionized. Once ionization of the cluster begins, *and harmonic production maximizes*, the contribution to the dielectric constant from the plasma electrons (determined from the Drude model) must be included. We quantify this by tracking the ionization [19] of the cluster. Figure 3 shows the electron density as a function of time and the resulting effect of shielding on the laser intensity within the cluster. The model assumes that electrons have a temperature of 10 eV (approximately the ponderomotive energy at the peak laser intensity) over the course of the laser pulse; however, the results are not sensitive to electron temperature. (See Refs. [9] and [10] for discussions of electron heating in cluster targets at these and higher intensities.) The narrow peak in the intensity curve and the corresponding sharp rise of the electron density are due to a resonance which occurs near an electron density close to the critical density for the laser wavelength. As seen in Fig. 3, shielding results in a time-averaged decrease in the intensity of the laser within the cluster, and may explain the increase in the appearance intensity of the cluster harmonics relative to the monomer harmonics.

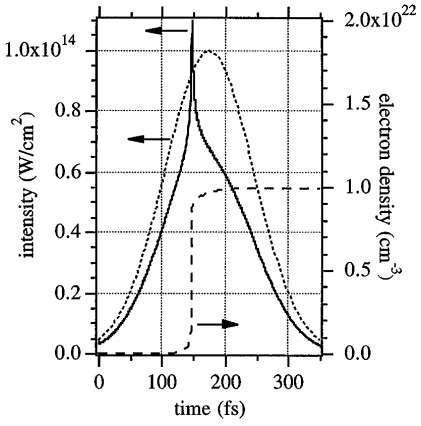

FIG. 3. The effect of shielding on the laser intensity seen by atoms in the cluster: electron density in the cluster (- - -), laser intensity in vacuum (···), and laser intensity in the cluster (——).

Next, we consider the power law dependence of cluster harmonic yield on laser intensity. We attribute the increased slope in the case of the clusters to the difference between the atomic potential which an electron experiences when bound by a single atom vs when bound in a solid. Ignoring surface effects, we model this difference using a one-dimensional, soft Coulomb potential [20]: the monomer target as a single well and the cluster as the single well plus two nearest neighbors, each offset by 0.37 nm. Using this semiclassical model to describe electron motion when driven by a laser pulse, we can determine the spectral density of the electron's emission [21]. From a series of such spectra, generated for various laser intensities, we can compare the nonlinearity of the harmonic yield vs laser intensity for the two different potentials. The power law dependencies for individual harmonics vary, and our calculations are subject to numerical noise. However, our result is that the three-well potential gives a more nonlinear dependence (average power law of I^{20}) compared to the single-well potential (average power law of I^{15}) for the 19th through 23rd harmonics modeled. We note that experimental power law fits of harmonics from various gases in the cutoff region are typically I^{12} [13,22].

Finally, we consider both the higher harmonic cutoff and reduced saturation in harmonic yield at high laser intensity for the clusters. The harmonic cutoff in monomer gases is conventionally considered to occur at a phonon energy of $I_p + 3U_p$ (where I_p is the binding energy of the electron in the atom and U_p is the ponderomotive energy of the free electron evaluated at the laser field that ionizes the atom) [6,7]. This results from the requirement that the electron be born near the peak of the laser field and then return to its parent ion in order to scatter and radiate harmonics. We expect that Coulomb (space-charge) forces will result in an increase in the effective binding energy of electrons in the cluster compared with isolated atoms, resulting in a higher photon energy (shorter wavelength) cutoff in clusters than monomer harmonics. Because of space-charge restraints on particle motion, the laser intensity may be allowed to rise to arbitrarily high limits without resulting in electron removal from the cluster, yielding harmonics at even shorter wavelengths. We also note that electrons may scatter from other atoms in the cluster.

In conclusion, we have shown that atom clusters have a strong nonlinearity which can be used to generate coherent, short-wavelength radiation in the form of high-order harmonics. These harmonics display an increased appearance intensity, higher energy cutoff, lack of saturation at

high laser intensity, and identical spatial properties when compared to monomer harmonics. Future studies will determine the actual efficiency of the cluster harmonic yields at much higher intensities and the phase matching properties of a dense medium of clusters. Using elliptically polarized laser light we should be able to determine an electron's ability to scatter from other than its parent ion in a cluster and the resulting contribution to the increase in the cutoff energy of the cluster harmonics.

The authors acknowledge helpful conversions with Ken Kulander. This work was supported by the U.S. Air Force Office of Scientific Research and through a collaboration with Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

*Current address: Department of Physics, Swarthmore College, Swarthmore, PA 19081.

[1] A. L'Huillier and P. Balcou, Phys. Rev. Lett. **70**, 774 (1993); J. J. Macklin, J. D. Kmetec, and C. L. Gordon, Phys. Rev. Lett. **70**, 766 (1993).
[2] T. Ditmire, J. K. Crane, H. Nguyen, L. B. DaSilva, and M. D. Perry, Phys. Rev. A **51**, R902 (1995).
[3] Y. Liang, S. August, S. L. Chin, Y. Beaudoin, and M. Chaker, J. Phys. B **27**, 5119 (1994).
[4] C. L. Carman, C. K. Rhodes, and R. F. Benjamin, Phys. Rev. A **24**, 2649 (1981).
[5] B. Bezzerides, R. D. Jones, and D. W. Forslund, Phys. Rev. Lett. **49**, 202 (1982).
[6] J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).
[7] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[8] R. Carman and C. Aldrich, in *Laser Techniques for Extreme Ultraviolet Spectroscopy*, edited by T. J. McIlrath and R. R. Freeman (AIP, New York, 1982), p. 224.
[9] A. McPherson *et al.*, Nature (London) **370**, 631 (1994).
[10] T. Ditmire, T. D. Donnelly, R. W. Falcone, and M. D. Perry, Phys. Rev. Lett. **75**, 3122 (1995).
[11] S. P. Gordon, T. D. Donnelly, A. Sullivan, H. Hamster, and R. W. Falcone, Opt. Lett. **19**, 484 (1994).
[12] T. Ditmire and M. D. Perry, Opt. Lett. **18**, 426 (1993).
[13] A. L'Huillier, L. Lompre, G. Mainfray, and C. Manus, in *Atoms in Intense Laser Fields*, edited by M. Gavrila (Harcourt Brace Jovanovich, Boston, 1992), p. 139.
[14] O. F. Hagena and W. Obert, J. Chem. Phys. **56**, 1793 (1972).
[15] O. F. Hagena, Surf. Sci. **106**, 101 (1981).
[16] J. Wormer, V. Guzielski, J. Stapelfeldt, and T. Moller, Chem. Phys. Lett. **159**, 321 (1989).
[17] C. L. Briant and J. J. Burton, J. Chem. Phys. **63**, 2045 (1975).
[18] M. D. Perry, C. Darrow, C. Coverdale, and J. K. Crane, Opt. Lett. **17**, 523 (1992); J. K. Crane (private communication).
[19] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).
[20] J. Javanainen and J. H. Eberly, Phys. Rev. A **39**, 458 (1989).
[21] K. Kulander and B. Shore, Phys. Rev. Lett. **62**, 524 (1989).
[22] C. G. Wahlstrom, J. Larsson, A. Persson, T. Starczewski, and S. Svanberg, Phys. Rev. A **48**, 4709 (1993).