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Generation of high-order harmonics from solid surfaces by intense femtosecond laser pulses

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The interaction of intense ultrashort laser pulses from a chirped pulse amplification titanium sapphire laser system with dielectric and metallic surfaces was studied. With a p-polarized incident beam and a large angle of incidence we have observed odd and even harmonics of high order with relatively smooth roll-off in the harmonic spectra. The shortest wavelength was 55.3 nm, corresponding to the 15th harmonic order.

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When an intense femtosecond laser pulse interacts with the surface of a solid target, material in a thin surface layer is ionized very rapidly. A plasma is formed in such a short time that only limited hydrodynamic expansion is possible. The maximum free-electron density reached during the interaction with the laser pulse is of the order of the total electron density of the solid and drops to the vacuum level in a distance that can be shorter than the skin depth of the laser light. The extremely high density and the sharp plasma-vacuum interface are unique features of plasmas produced in the femtosecond laser-solid interaction.

One of the interesting aspects of very dense plasmas having an almost steplike density profile concerns the dynamics of the electronic motion in the vicinity of the plasma-vacuum interface [1,2]. When the electric-field vector of the laser light possesses a sufficiently strong normal component, electrons can be dragged back and forth across the plasma-vacuum boundary with excursions greater than the scale length of the interface. Because the restoring forces acting upon the laser-induced charge perturbations are dependent on the local density, the periodic motion across the interface can be regarded as a driven oscillation in some very anharmonic potential. The anharmonic response of the interface electrons should give rise to radiation at harmonics of the driving fundamental laser frequency.

At least two reasons can be given why the study of optical harmonic generation at the plasma-vacuum interface should be of great interest. First, the spectra of the harmonics are expected to provide useful information on the structure of the plasma-vacuum boundary and the dynamics of the electrons as the normal component of the motion samples the interface. Second, the pronounced electronic nonlinearity of the plasma-vacuum interface bears some potential for the generation of harmonics of high order and could thus represent a useful mechanism for the generation of coherent ultrashort light pulses at very short wavelengths. Other schemes to generate high-order optical harmonics have recently been proposed, which involve the interaction of intense femtosecond laser pulses with solids [3] or solid density plasmas [4] having a steep density profile. These schemes could possibly provide an interesting alternative to high harmonic production in gases [5].

Generation of harmonics of very high order (≈ 50) has been observed previously in the interaction of *nanosecond* pulses from a CO_2 laser with solid targets [6,7]. In this case, however, harmonic generation was associated with a steep density profile produced as a result of a balance between the radiation pressure of the laser pulses and the hydrodynamic expansion pressure of the plasma. In this situation the "upper shelf" density was determined by the intensity of the laser pulse and the electron temperature. The maximum plasma density attained in the CO_2-laser experiments ($\approx 2\times 10^{22}\,\mathrm{cm}^{-3}$) corresponded to a few hundred times the critical density. However, in femtosecond experiments the maximum density would be independent of the laser and plasma parameters, being limited only by the electron density of the solid material.

Second-harmonic generation during the interaction of intense ultrashort laser pulses with solid surfaces has been studied in great detail [8,9]. However, experimental work on higher-order harmonic generation has been rather scarce to date. The fifth harmonic from gold surfaces has been measured using picosecond pulses of relatively low power [10]. Recently, observation of the seventh harmonic of near infrared femtosecond pulses from bulk aluminum targets has been reported [11].

Here we wish to describe an experiment in which optical harmonics of high order have been observed during the interaction of intense femtosecond laser pulses with planar solid targets. A schematic of the experimental arrangement is shown in Fig. 1. Laser pulses polarized in the plane of incidence were focused on the target using a fused silica lens with $f = 15$ cm at an angle of incidence of 68°. Optical flats coated with a 200 nm layer of aluminum and bare glass flats served as targets. The samples were rapidly raster-scanned to provide a fresh surface for each pulse.

Specularly reflected light from the target was picked up by a toroidal holographic grating with 550 lines/mm, which was aligned to produce an image of the target plane with a magnification close to unity. For the detection of UV and XUV light a rare-earth phosphor (Gd_2O_2S:Tb) deposited on an optical flat was positioned in the image plane of the grating. The yellow-green phosphorescent light produced by

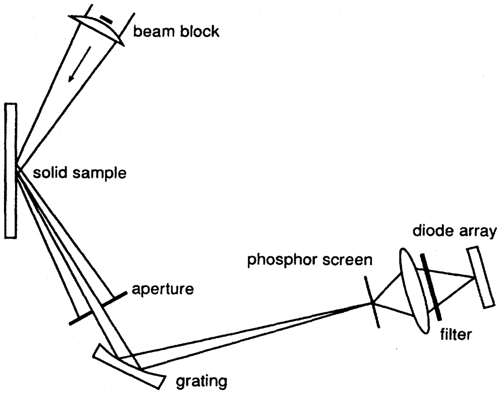

FIG. 1. Schematic of the experimental setup.

short-wavelength radiation from the target was collected by a pair of commercial fast photographic lenses aligned front to front. With this arrangement high-resolution imaging of the phosphor on a photodiode array with $f/1.1$ could be achieved. The photodiode array was coupled to a multichannel analyzer. The experiment was carried out under vacuum at a base pressure of 10^{-3} mbar. The laser pulses used in our experiment were obtained from a titanium sapphire chirped pulse amplification laser system operating at 800 nm. The pulse duration was 130 fs, and up to 30 mJ of energy were available. The peak intensity on target was estimated to be approximately 10^{17} W/cm^2 or less.

To measure the relatively weak harmonic content of the specularly reflected beam the fundamental frequency must be suppressed. This was done as follows. A small disk was positioned near the front of the focusing lens to block the center of the incident laser beam. It can be readily shown that in the target plane the obstacle produces essentially some changes in the *wings* of the intensity profile, whereas the distribution in the *center* is hardly affected. Since the induced nonlinear surface current responsible for reflected harmonics is expected to be some rapidly increasing function of the fundamental intensity, these minor deformations of the fundamental beam caused by the beam block are negligible. However, an image of the blocking disk will be reproduced in the far field of the reflected beam at the fundamental frequency. An aperture can now be used to very effectively block the strong reflected laser beam, while passing the harmonic light in the center of the beam. Provided that there are no severe phase distortions of the nonlinear source current, the beam diameter of the harmonics should scale as $n^{-1/2}$, where n is the order of the harmonic. Thus it should be possible to choose an aperture blocking the fundamental beam, while transmitting high-order harmonics practically without losses. We note that the same scheme has previously been used in high-order harmonic generation in gases [12].

In the experiment we have observed a clean image at the fundamental frequency of the blocking disk, when the beam reflected from the plasma was intercepted in the far field by some screen. This image permitted us to precisely align an aperture to block the reflected fundamental laser light. With the bare eye conspicuous second harmonic light could be seen in the specular direction confined to a cone that was somewhat narrower than the fundamental laser beam. In fact, second-harmonic emission scattered from obstacles in the vacuum chamber and the walls was responsible for most of the background light seen by our detection system. While scattered near-infrared laser light could be effectively cut off with a suitable color filter, the blue second-harmonic background could be reduced with a low-pass color filter only at the expense of the signal light from the phosphor.

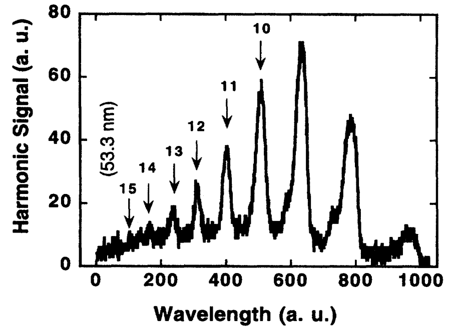

FIG. 2. Harmonic spectrum from an aluminum film. The vertical arrows indicate the order of the harmonics.

Figure 2 depicts an example of a measured harmonic spectrum in the far-UV region between 40 nm (left) and 100 nm (right) for aluminum. A series of harmonic lines can clearly be recognized, superimposed on some background signal. In this case a filter had been used to reduce the second-harmonic background, and signal accumulation over 300 laser pulses was necessary to record the spectrum (integration time of 30 s). The vertical arrows in Fig. 2 indicate the order of the harmonics. Both odd and even harmonics were observed. The highest order was the 15th harmonic at a wavelength of 53.3 nm. Similar harmonic spectra were measured for bare glass samples, with the highest order distinguishable from the background around 13 to 14.

Figure 3 compiles data of the measured strength of the harmonics as a function of harmonic order from a number of different experimental runs. It appears that the roll-off at high orders is relatively smooth. It should be pointed out, however, that these data are subject to considerable uncertainties, mainly because the properties of the phosphor were not very well known. In particular, we have noticed a pronounced spatial inhomogeneity of the phosphor layer. This effect can be attributed to our rather simple deposition method. The layers were prepared by letting the phosphor powder settle down on the substrate from a liquid suspension. The composition and thickness of the layer could not be very well controlled.

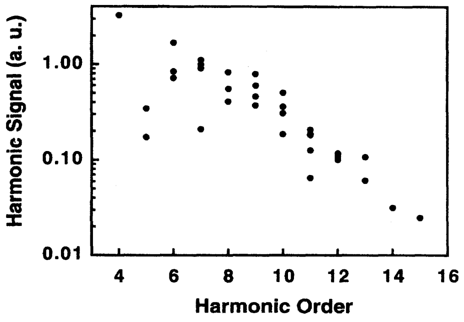

FIG. 3. Strength of the harmonics as a function of order.

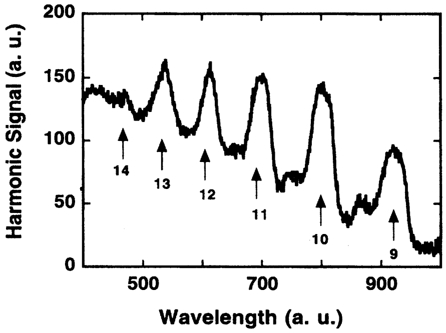

FIG. 4. Harmonic spectrum from glass showing spurious half-integral harmonics.

Keeping these reservations in mind, we can nevertheless give an estimate of the observed efficiency of harmonic production. In fact, being able to observe far-UV radiation from harmonic generation with some rather crude detection scheme suggests that the number of UV photons per pulse is relatively large. An order of magnitude estimate gives $\approx 2\times 10^9$ and $\approx 10^8$ photons per pulse at the 10th and the 15th harmonic, respectively, which corresponds to a photon conversion efficiency of $\approx 2\times 10^{-8}$ and $\approx 10^{-9}$.

Occasionally we have observed spurious lines at intermediate wavelengths between the integral harmonics, which could be interpreted as half-integral harmonic emission. An example is shown in Fig. 4, where a harmonic spectrum for glass is depicted on an expanded wavelength scale. It should be mentioned that in this case the background signal was much stronger. We have noticed that a degradation of the laser performance, e.g., a decrease of the pulse energy and concomitant distortions of the spatial and temporal pulse profile, typically resulted in a significant increase of the background up to the point where no harmonic signals could be distinguished. Visually we observed that under these conditions the laser beam reflected from the target was deformed and unstable so that the reflected light was incompletely blocked by the aperture.

We now turn to a brief discussion of our results. The experiments have been performed in a configuration in which there was a strong normal component of the electric field. Thus it is likely that the experiment predominately probes the response of the electrons in the direction perpendicular to the plasma-vacuum interface. The observation of odd and even harmonic orders of equal strength indicates that harmonic generation takes place in some environment with broken inversion symmetry, as expected for an interface process. It rules out harmonic generation caused by the nonlinear response of the ions or atoms of the bulk plasma, which would be the same mechanism responsible for high-order harmonic generation in gases. Observation of harmonic generation in the specular direction is also consistent with a coherent surface source current induced by the fundamental light. Under these conditions the direction of the harmonics is determined by the conservation of the parallel component of the wave vectors in the nonlinear process.

Finally we wish to discuss the shortest harmonic wavelength observed in the present experiments. Earlier theoretical models [1,2] have predicted that the frequency of the maximum harmonic order is limited by the plasma frequency corresponding to the upper-shelf electron density. However, a more recent computer simulation suggested that there is no sharp cutoff related to the maximum electron density and that the number of harmonics continues to increase with increasing laser intensity [13]. As regards our experimental situation, the density of conduction electrons in solid aluminum is $\approx 2\times 10^{23}$ cm^{-3}. This figure is expected to double for a solid density plasma with an electron temperature of about 100 eV [14], and the photon energy of the corresponding value of the plasma frequency is 23.5 eV. Following the existing theoretical models this value would suggest a maximum order of 15, which agrees with the experimentally observed maximum harmonic order. However, we believe that this agreement is fortuitous. It is clear that further experiments are required to explore the limit in high-order surface harmonic generation.

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[1] B. Bezzerides, R. D. Jones, and D. W. Forslund, Phys. Rev. Lett. **49**, 202 (1982).
[2] C. Grebogi, V. K. Teripathi, and H.-H. Chen, Phys. Fluids **26**, 1904 (1983).
[3] S. Hüller and J. Meyer-ter-Vehn, Phys. Rev. A **48**, 3906 (1993).
[4] S. C. Wilks, W. L. Kruer, and W. B. Mori, IEEE Trans. Plasma Sci. **21**, 120 (1993).
[5] See, e. g., *Atoms in Intense Fields*, edited by M. Gavrila (Academic Press, Boston, 1992).
[6] R. L. Carman, D. W. Forslund, and J. M. Kindel, Phys. Rev. Lett. **46**, 29 (1981).
[7] R. L. Carman, C. K. Rhodes, and R. F. Benjamin, Phys. Rev. A **24**, 2649 (1981).
[8] T. Engers, W. Fendel, H. Schüler, H. Schulz, and D. von der Linde, Phys. Rev. A **43**, 4564 (1991).
[9] D. von der Linde, H. Schulz, T. Engers, and H. Schüler, IEEE J. Quantum Electron. **28**, 2388 (1992).
[10] G. Farkas, C. Tóth, S. D. Moustaizis, N. A. Papadogiannis, and C. Fotakis, Phys. Rev. A **46**, R3605 (1992).
[11] S. Kohlweyer, G. D. Tsakiris, C.-G. Wahlström, C. Tillman, and I. Mercer, Opt. Commun. (to be published).
[12] J. Peatross, J. L. Chaloupka, and D. D. Meyerhofer, Opt. Lett. **19**, 942 (1994).
[13] P. Gibbon (private communication).
[14] H. M. Milchberg, R. R. Freeman, S. C. Davey, and R. M. More, Phys. Rev. Lett. **61**, 2364 (1988).