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Generation of high harmonics in laser plasmas

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Abstract

The generation of high harmonics in laser–plasma interactions on the steep density gradients is discussed, especially by using short-pulse UV laser radiation. Low intensity experiments with $5 \cdot 10^{15}$ W/cm^2 generate harmonics in the VUV range, and the efficiency can be optimized by modifying the density gradient. To obtain higher harmonics and to clear some disagreements among different results concerning polarization properties of harmonics we switched for higher intensities. Our experimental arrangement makes it possible to obtain a $5 \cdot 10^{17}$ W/cm^2 intensity with a prepulse as low as 10^7 W/cm^2 using a table-top system. Preliminary results and future trends for high-harmonics generation with this method are given.

Keywords: Harmonics generation, Laser plasma, Short-pulse lasers

1. INTRODUCTION

One of the possible methods to generate coherent radiation in the VUV and EUV spectral range is by high harmonics of a visible laser beam. Efficient high-harmonics generation of odd order has been observed in different neutral gases. Although this method is, in principle, limited by the ionization of the gas, this difficulty was overcome by applying ultrashort pulses (Chang *et al.*, 1997; Spielmann *et al.*, 1997), thus reaching very short wavelength radiation up to 0.5 keV photon energy. The synchronization of high harmonics made it possible to obtain attosecond pulses, too (Papadogiannis *et al.*, 1999; Drescher *et al.*, 2001).

On the other hand, harmonics generated in a laser–plasma interaction may offer an alternative efficient method. As early as 1980, high harmonics were obtained from laser–plasma interactions with IR laser beams. In that case, plasma profile steepening resulted in a sharp density gradient; nonlinear oscillation of the electrons crossing the critical surface as driven by the electric field of the laser (Bezzerides *et al.*, 1982) was the source of harmonics. The situation drastically changed in the recent decade with the ultrashort laser pulses. In plasmas generated on solid surfaces by ultrashort laser pulses, the scale length of the plasma may remain less than the laser wavelength; thus no profile steepening is necessary for favorable initial conditions.

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Even- and odd-order harmonics were generated using a Ti:sapphire laser by Kohlweyer *et al.* (1995), and soon afterward harmonics up to the 75th order of a Nd laser were obtained on the wavelength of 14.0 nm (Norreys *et al.*, 1996). Even a small, table-top KrF laser with nonrelativistic intensity generated harmonics to the wavelength of 82.8 nm (Földes *et al.*, 1996).

In this article, some aspects of high-harmonics generation are summarized, especially with the aim of showing the differences among results obtained with lasers of different parameters, that is, wavelength and pulse duration. The literary data is compared with our own results obtained by a KrF laser beam. The application of this ultraviolet laser beam is of interest because, for a short wavelength pumping laser, lower orders of harmonics are needed for the same given short wavelength harmonics radiation that may result in an effective conversion to short wavelength coherent radiation. Also, according to PIC simulations (Gibbon, 1996), KrF lasers are candidates for generating harmonics with wavelengths even in the water window.

2. REVIEW OF EXPERIMENTS ON HIGH-HARMONICS GENERATION

Experiments were carried out in different laboratories using different experimental parameters such as laser wavelength, intensity, and pulse duration. Parts of these experiments were carried out with lasers of durations shorter than 200 fs. The results of Kohlweyer *et al.* (1995), von der Linde *et al.*

(1995), and Gizzi *et al.* (1996) show that *p*-polarized laser radiation can generate even and odd order high harmonics. These results are in agreement with theories based on the resonant absorption of laser light at the critical surface (von der Linde *et al.*, 1992; Lichters *et al.*, 1996). In these experiments, the generated harmonics were found to propagate to the specular direction up to intensities of 10^{18} W/cm^2 (Zepf *et al.*, 1998). On the other hand, the experiments carried out with longer laser pulses, that is, with pulses of 500 fs and longer, showed a different behavior. In this case, intense harmonic generation was observed both for *p*- and *s*-polarized laser beams (Földes *et al.*, 1996; Norreys *et al.*, 1996; Ishizawa *et al.*, 1999). The experiments of Norreys *et al.* (1996) showed the until now highest, 75th order of harmonics by using the 1-ps duration Vulcan laser beam of 10^{19} W/cm^2 intensity. The experiments that were carried out by these high intensities, however, did not show specular reflection any more. Rippling of the critical surface was considered to be the reason for diffuse harmonics generation and for efficient harmonics generation with the *s*-polarized radiation as well. This was confirmed by the results of Chambers *et al.* (1998) obtained with the Titania KrF laser system in which case harmonics become diffuse above a certain threshold intensity of 10^{16} W/cm^2, and surface rippling was found to appear at this threshold, too.

The selection rules for harmonics generation were defined (Lichters *et al.*, 1996) with respect to parity (i.e., odd or even order) and polarization of the generated harmonics. The dependence on the polarization of the incident laser beam is essential. According to the theoretical models based on the critical surface processes, *p*-polarized radiation is much more efficient for harmonics generation than the *s*-polarized one. Another important selection rule is that an even harmonics number will be *p*-polarized even for an *s*-polarized laser beam (von der Linde *et al*., 1992; Lichters *et al*., 1996). This was really found by Gizzi *et al*. (1996) for the second harmonics. Therefore we investigated the polarization of the second and third harmonics generated by a 248-nm laser beam (Veres *et al*., 1999). As in this case the harmonics were already in the VUV spectral range, the polarization was determined by a polarization analyzer consisting of three Au-coated mirrors using the polarization properties of Fresnel reflections on them. A similar polarizer was recently used by Krushelnik *et al*. (2002), too. Our results showed that the *p*-polarized radiation was only four times more efficient for second harmonics generation, whereas the efficiency was approximately the same for the third harmonics for both laser polarizations. The most interesting result was that the polarization of the generated harmonics was the same as that of the laser beam, that is, harmonics kept laser polarization. In this experiment, the harmonics propagated specularly; therefore probably no critical surface rippling was present for the low, nonrelativistic intensity of the less than 10^{16} W/cm^2 UV laser beam. Another possible explanation proposed is the Faraday rotation of the laser electric field (Ishizawa *et al*., 1999). In our case, no rotation of the laser polarization was detected. Although recent experiments (Tatarakis *et al*., 2002) showed the existence of very high magnetic fields and proved their role in harmonics generation, those experiments were carried out with ultrahigh intensities of 10^{20} W/cm^2 in contrast to the $5 \cdot 10^{15}$ W/cm^2 (Veres *et al*., 1999) for which case magnetic field effects were negligible.

Thus, we can see that even and odd harmonics can be generated by nonrelativistic laser pulses keeping the polarization of the *s*- or *p*-polarized laser beam. To understand this phenomenon, we tried to improve the analytical model of von der Linde (1992), taking into account processes at the critical layer nonperturbatively. Although the nonperturbative model could explain harmonics generation by *s*-polarized radiation due to the ponderomotive force at the critical layer, it gives a *p*-polarized even harmonics for *s*-polarized radiation, too, that is, it does not explain the observed polarization of the generated harmonics. A recent model of Gál and Varró (2001) starts from another point of view. In their model, the evanescent electromagnetic field in the skin layer of the overdense plasma was calculated, too. It was shown that the magnetic field has a local maximum there; thus, even for nonrelativistic intensities, the full Lorentz force must be taken into account, that is, including the $\mathbf{v} \times \mathbf{B}$ force, and it may serve as a source of high harmonics generated in the skin layer. Harmonics generated in this way conserve the polarization of the laser beam.

The other main parameter, which may be decisive in the harmonics-generating mechanism, is the scale length of the plasma in which harmonics generation occurs efficiently. PIC simulations of Zepf *et al*. (1998) showed for high intensities that an optimal scale length exists. We carried out experiments by modifying the density gradient using a 700-fs pulse generating a preplasma 0–40 ps prior to the main KrF laser pulse (Földes *et al.*, 1999). Figure 1 shows that the intensity of the observed third harmonics has a pronounced maximum for 5-ps delay. Similar maxima were observed for the second harmonics, too. The plasma scale length corresponding to this delay was estimated to be $L/\lambda \approx 0.3$, which is roughly the same as that of the optimum for resonance absorption (Ginzburg, 1961). Our observation for an optimal scale length was confirmed by Ishizawa *et al.* (2000), extending the optimization for the fourth order. The optimization—and perhaps a selectivity for a given order—of harmonics is good news for harmonics as a usable VUV light source. On the other hand, this observation did not select among the possible mechanisms, because the conditions for an optimal resonance absorption are similar to the optimum for harmonics generation with the full Lorentz force by using nonrelativistic pulses.

3. THE PERSPECTIVES OF UV LASERS FOR HARMONICS GENERATION

PIC simulations of Gibbon(1996) suggested that—in contrast to earlier expectations—the shortest observable har-

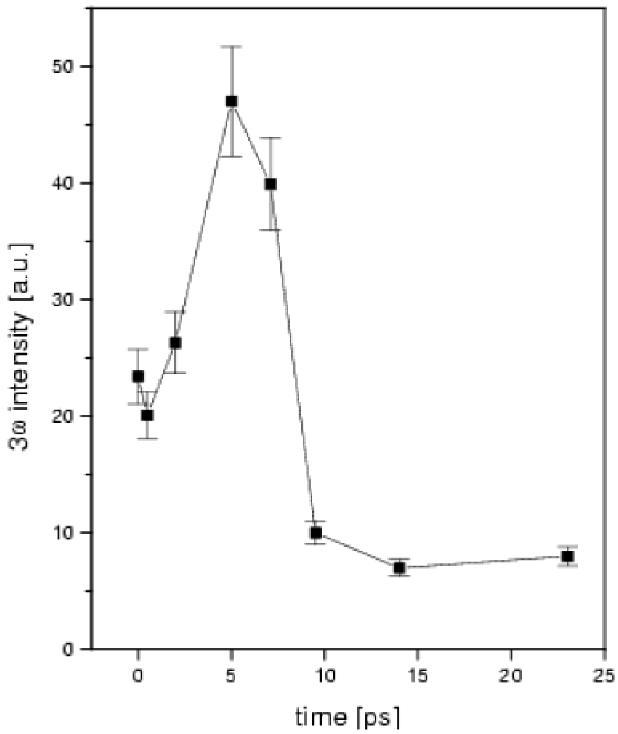

Fig. 1. Dependence of third-harmonic intensity on time delay after prepulse. The laser beam was *p*-polarized; statistical errors are shown.

monics wavelength is not limited, and the harmonic order is simply determined by $I\lambda^2$. The wavelength of a given harmonic order is shorter for short wavelength lasers; thus, they can efficiently produce harmonics of short wavelengths up to the water window. That was the reason why experiments with a KrF laser pulse may be of interest. An advantage of our KrF laser system based on a double discharge excimer laser (Szatmári, 1994) is that it is not a CPA laser, that is, the only source of prepulse is the ASE of the KrF amplifier. In the first experiments (Földes *et al.*, 1996, 1999; Veres *et al.*, 1999), the laser was focused by an *f*/10 lens, obtaining a modest, $5 \cdot 10^{15}$ W/cm^2 intensity. Due to the bad focusability and long (≈15-ns) duration of the ASE prepulse, its level could be kept below 10^7 W/cm^2. Noting that the observed 3ω radiation already had a wavelength as short as 82.8 nm, it is encouraging to go toward higher intensities. On the other hand, the experiments of Chambers *et al.* (1998) by the very high intensity Titania laser system could reach only the fourth harmonic. They observed a strong rippling of the critical surface for high intensities, that is, above 10^{16} W/cm^2. The laser pulse used therein had, however, a strong prepulse; therefore, it is not clear whether their result is a consequence of the preplasma effects or its intrinsic nature.

To obtain high focused intensity, the laser pulse of 20-mJ energy on the 248-nm wavelength with 600-fs pulse duration was focused by an *f*/2 off-axis parabola mirror from JANOS Technology, Inc. with an effective focal length of 10 cm. To measure the focal distribution, a large magnification was applied using a special CaF_2 microscope objective consisting of four lenses giving 56× magnification with $f=$ 13.21 mm. The laser beam was diffraction limited, the focal spot diameter was found to be 2 μm, in agreement with the expectations. These measurements were carried out using a full-power, but attenuated laser beam. It means that the laser was fired with a full-power amplifier, and the total intensity was attenuated later by using mirrors; thus, modifications of the beam quality by the amplifier were taken into account.

Figure 2 shows a horizontal cross section of the focal intensity distribution. Approximately 55% of the laser intensity was found inside the 2-μm focal spot, which corresponds to $\approx 5 \cdot 10^{17}$ W/cm^2 intensity. Thus the beam was approximately diffraction limited. The intensity of the ASE prepulse in the focal plane was as low as 10^7 W/cm^2, similar to the case of lens focusing. It means that it was well under the threshold of plasma generation and that the contrast was nearly 10^{11}.

To investigate high harmonics the *p*-polarized laser beam was focused on a target at an angle of 45°, which can result in $3.4 \cdot 10^{17}$ W/cm^2 intensity on the target. The targets were thin, 200-nm Al and C layers on glass plates. Each shot hit a fresh target element. The target mechanics were accurate enough that when scanning the total area of the 5-cm diameter target, the deviation from the ideal plane was ≈3 μm. This is less than the 6-μm Rayleigh range in the present case; that is, it is possible to have each shot approximately in the focal plane. The VUV spectrometer was based on a Jobin–Yvon holographic grating of 550 l/mm, which imaged the source onto the detector, which was an MCP with a phosphor screen. The visible light of the screen was then detected by a CCD. The first experiments showed intense second and third harmonic generation from a C target. Figure 3 illustrates a time-integrated VUV spectrum of a 260-nm graphite layer on a glass plate. The laser beam was *p*-polarized. Besides the carbon lines that originate at the later stage of the recombining plasma, an intense third-harmonic signal can be distinguished. Some

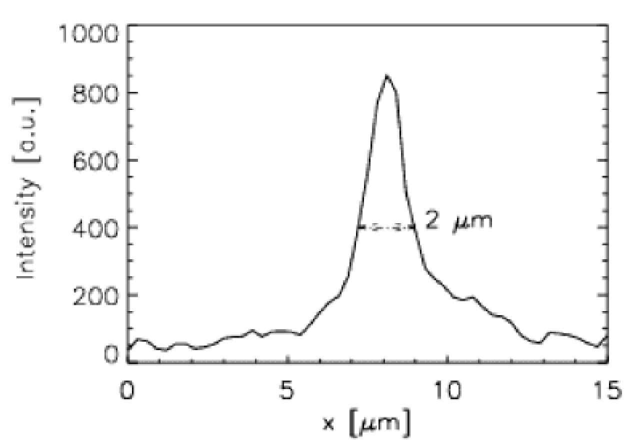

Fig. 2. Focal spot distribution. Fifty-five percent of laser energy is within the 2-μm focal diameter.

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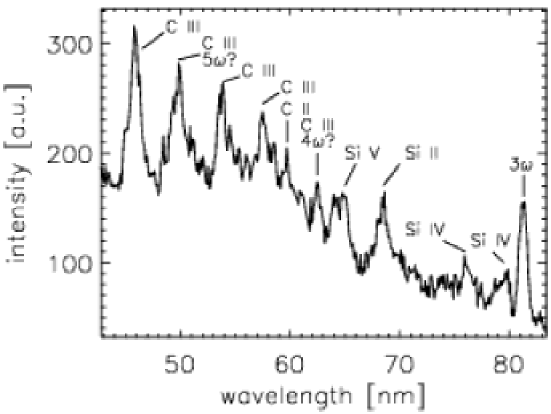

Fig. 3. VUV spectrum of 260-nm-thick C on glass with intense third-harmonic signal.

Si lines from the glass plate can be seen, too. From these preliminary data it is not clear at present whether higher harmonics occur because the position of the 4ω and 5ω radiation corresponds to intense C-lines, too. A possible problem is that although the focusing is optimized without the solid target, it is possible that the focusing onto the target was not optimal yet. Thus the on-target intensity could be significantly lower than the nominal $3.4 \cdot 10^{17}$ W/cm^2 given above. Therefore until now we could not clearly demonstrate the appearance of higher than third-order harmonics.

A question arises whether our pulse duration is appropriate for high harmonics generation. We did see that harmonics generation could be optimized by having a plasma scale length of (0.2–0.3) λ. Our 700-fs pulse duration was close to the optimum (Földes *et al*., 1999) at $5 \cdot 10^{15}$ W/cm^2 intensity. Increasing intensity increases the expansion velocity. According to the classical model of plasma expansion (Max, 1982) the expansion velocity scales as $I^{1/2}$. On the other hand, investigations were carried out to measure the spectral distribution of the specularly reflected laser light. According to these results, the expansion velocity scales as $I^{1/4}$ only. According to the $I^{1/2}$ scaling, the optimal pulse duration at $5 \cdot 10^{17}$ W/cm^2 intensity is only 70 fs and further increasing it to $5 \cdot 10^{19}$ W/cm^2 means it must be as short as 7 fs. In the other case, a 220-fs pulse may be optimal for $5 \cdot 10^{17}$ W/cm^2 in the case of $I^{1/4}$ dependence and even for $5 \cdot 10^{19}$ W/cm^2 it is sufficient to shorten it to 70 fs. There is another possibility that may help in harmonics generation with pulses longer than 100 fs. The fact that some groups observed high harmonics even for longer pulses of several picoseconds duration as did Ishizawa *et al.* (1999) and even the harmonics of the highest order with a longer than 1-ps pulse of very high intensity (Norreys *et al.*, 1996) contradicts to these expectations. Maybe even in this case one has additional help for harmonics generation in profile steepening as was found at the analysis of early experiments by Bezzerides *et al.* (1982).

4. CONCLUSIONS

The experimental works presented in this article and the ones cited here aim to clarify the mechanism of high-harmonics generation in laser plasmas generated by ultra-short pulses even for nonrelativistic laser intensities. It is fundamental to use a prepulse-free laser to obtain well-defined, clean conditions. This is fulfilled by our KrF laser in which case a contrast better than 10^{10} is obtained. Further, systematic investigations with higher than 10^{17} W/cm^2 intensity will follow, which should give more insight into these problems, among others, by separating intrinsic critical surface rippling from the one of a preformed plasma.

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