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Time-resolved harmonic generation in an ionizing gas

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Abstract. We report on a time-resolved study of the fifth harmonic generated in xenon by 140 ps pulses from a Nd:YAG laser in the 10^{13} W cm^{-2} intensity regime. Absolute timing between the driving laser pulse and the harmonic pulse could be determined by means of reference harmonics generated in non-linear crystals and a single common streak camera. Above a certain laser intensity the centre of the observed harmonic pulse was shifted earlier in time relative to the laser pulse. We found the intensity dependence of this shift to be approximately linear within the intensity range used which is consistent with the results of numerical simulations taking the ionization and dispersion of the xenon gas into account.

1. Introduction

The generation of high-order harmonics of short-pulse, high-power lasers has recently attracted considerable attention. Extremely short-wavelength harmonics have been reported, for example the 135th harmonic of a 1053 nm Nd: glass laser (L'Huillier and Balcou 1993), the 109th harmonic of a 806 nm Ti: sapphire laser (Macklin *et al* 1993), the 45th harmonic of a 526 nm frequency doubled Nd: glass laser (Crane *et al* 1992), and the 33rd harmonic of a 248 nm KrF-excimer laser (Sarukura *et al* 1991). As this short-wavelength radiation may be coherent and of very short duration, harmonic generation is an interesting and useful source of intense XUV and soft x-ray radiation. Because of their short pulse duration (<1 ps) and high peak brightness these sources complement existing synchrotron sources in this spectral range. If frequency tunable high-intensity lasers are used, the tunability of the harmonic radiation also makes them complementary to existing x-ray lasers.

The use of radiation produced by harmonic generation in various applications has only recently begun to be explored. Haight and Peale (1993) used subpicosecond pulses from a dye laser as the pump, and various harmonics from the same laser as probes in a series of pump-probe experiments performed to investigate surface states in solid state physics. Balcou *et al* (1993) used high-order harmonics generated with a tunable dye laser to investigate the photoionization cross sections in several rare-gas atoms.

High-order harmonics have been obtained at focused intensities in the range 10^{14}–10^{18} W cm^{-2} where a non-negligible amount of ionization of the medium takes place during the laser pulse. This raises questions concerning the role of ions in the harmonic generation process and the effect of free electrons on phase matching of the harmonics

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in the medium. The ionization reduces the number of neutral atoms contributing to the harmonic generation, and the presence of free electrons leads to plasma dispersion. The temporal and spatial evolution of these phenomena will affect the temporal and spatial structure of the harmonic pulse and will ultimately limit the conversion efficiency to high-order harmonics.

The degree of ionization will be different at different points in space and time within the focal volume during the laser pulse and depends upon the peak intensity used. The spatial and temporal profiles of the generated radiation are thus expected to be complex, even if the driving laser pulse is smooth and uniform in both space and time. In certain applications, the usefulness of this radiation depends critically on the uniformity of its temporal and spatial profiles. It is therefore important to study these properties and determine how they depend on different experimental conditions. By comparing these properties between experiment and theory, new understanding can also be achieved of the fundamental processes involved.

Many of the reported experiments on high-order harmonic generation have been performed with ultra-short laser pulses, with pulse lengths of the order of a few picoseconds or less. Studies of the temporal behaviour of harmonics generated with such short pulses have so far not been possible because of the lack of detectors with suitable time resolution. However, the relevant physics, which determines the temporal structures can be studied in harmonics generated by longer laser pulses.

In a recent experiment Faldon *et al* (1992) investigated the temporal structure of the seventh and ninth harmonics of a 50 ps Nd:YLF laser (1053 nm) produced in xenon. The temporal profiles obtained for the harmonics were compared to the results of a numerical model which incorporated the experimental temporal profile of the laser pulse. This model took into account the time and space dependent ionization of the medium and the dispersion of the photoelectrons. One characteristic effect predicted by that model was an intensity-dependent shift in time of the peak of the harmonic pulse relative to the laser pulse. This effect was due both to the depletion of the neutral medium and to the propagation in the medium being influenced by the dispersion of the photoelectrons. However, the shift could not be observed experimentally as the laser pulse and the harmonic pulse were recorded with different streak cameras and the temporal jitter in the triggering of the streak cameras was larger than the expected shifts. No absolute timescale could be obtained in that experiment.

In this paper we report on a study where we specifically investigated, for the first time to our knowledge, the timing of the harmonic radiation relative to the driving laser pulse. The temporal profile of the laser pulse, the fifth harmonic generated in an unsaturated non-linear crystal and the same harmonic generated in an ionizing gas were recorded simultaneously using a single streak camera. In this way the influence of ionization on the temporal structure of the harmonic radiation could be investigated. In particular, the absolute shift in time of the harmonic radiation relative to the laser pulse was determined and studied for various laser intensities. The experimental shifts were compared with numerical simulations, using the same model as Faldon *et al* (1992).

The numerical model used describes the generation of an electric field at frequency $q\omega$ by a focused laser beam of frequency ω and its propagation in a non-linear medium undergoing multiphoton ionization. It is assumed that the spatial profile of the laser radiation is Gaussian and that it propagates in the positive z direction. The temporal profile of the laser field may be chosen either to be Gaussian or to match exactly the shape of the experimental pulse. The model includes both the time and space dependent

phase matching and depletion of the neutral atoms. As a first step, for each grid point in time, the density of neutral atoms, ions of different ionization stages (determined from the lowest-order perturbation theory), and free electrons are calculated at every grid point (r, z) in space. From these values, the local phase mismatch is calculated. Secondly, the amplitude and the phase of the partial waves, originating in different grid points, are calculated. Finally, the total field at the point of observation, far from the interaction region, is obtained through numerical integration over the total volume.

2. Experimental details

The experimental set-up is illustrated schematically in figure 1. The laser used in this experiment was a Q-switched and mode-locked Nd:YAG laser (Continuum PY61C) at

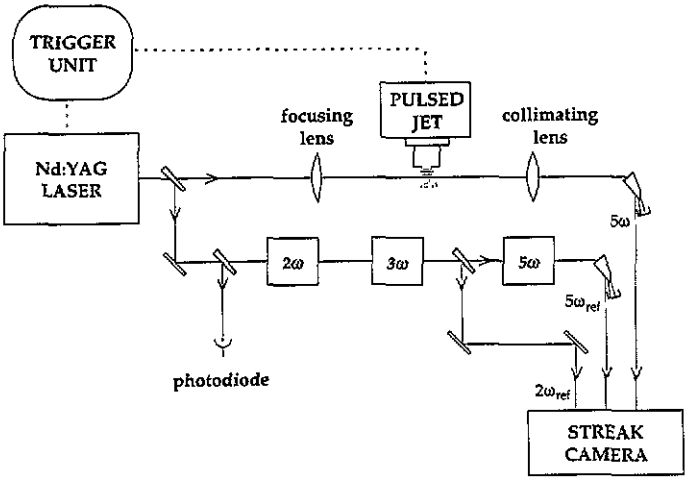

Figure 1. Experimental set-up for time-resolved harmonic generation.

the Lund High-Power Laser Facility (Svanberg *et al* 1994) operating at 1064 nm. The mode locking was accomplished by the combined use of a saturable absorber dye and an intra-cavity acousto-optic modulator. The oscillator produced pulses of approximately 2 mJ energy which were amplified in a single-stage amplifier. By keeping the voltages of the oscillator and amplifier flash lamps constant, but changing their mutual timing, the amplifier gain, and hence the output energy of the laser could be continuously varied up to 100 mJ without changing the final beam profile. The pulse length was determined by the thickness of an etalon in the oscillator cavity. In the present experiment, the pulse length was approximately 140 ps FWHM. However, there were significant fluctuations in the pulse length on a shot-to-shot basis, and we therefore registered and stored the pulse shape and pulse length of every laser shot individually. The long wavelength cut-off in the spectral sensitivity of the streak camera (Hamamatsu C1587 with S-20 cathode) is around 850 nm. Consequently, the infrared laser pulses could not be recorded directly. Instead a small fraction of the pulse energy was split off and passed through a KD*P crystal, and the second harmonic radiation, at 532 nm, was recorded. Great care was taken to ensure that no saturation effects occurred in the crystal. The temporal spread due to dispersion in the crystal was estimated to be less than 0.5 ps. The pulse energies were measured and recorded individually using a fast

photodiode detecting a portion of the laser light split by a wedged glass plate placed in the laser beam before the focusing lens. The photodiode was cross-calibrated against a calorimeter.

The laser pulses were focused by a 190 mm focal length 'best-form' lens ($f\# = 32$) into a vacuum chamber where they interacted with a jet of xenon gas from a pulsed nozzle (Lasertechnics LPV). The choice of the focusing optics was a compromise between two conflicting requirements. A larger value of the focal length of the lens would on one hand mean a larger volume of atoms emitting harmonics but would on the other hand result in lower intensities available with the laser used because of the increase in focal spot size. The density of the gas in the interaction region was not measured directly, but estimated from profile and flow measurements which gave a value of approximately 5×10^{17} atoms/cm^3 corresponding to a peak xenon pressure of 20 mbar in the centre of a Lorentzian density distribution with a full-width at half maximum of approximately 1 mm. The spot size radius w_0 was estimated to be about 30 μm with a 30% uncertainty.

The generated harmonics and the fundamental radiation left the vacuum system through a quartz window and were collimated using a quartz lens ($f = 200$ mm). The fifth harmonic, at 213 nm, was isolated from the fundamental radiation and the third harmonic by a quartz Pellin–Broca prism. By using a relatively low dispersion prism instead of a grating, unnecessary temporal smearing of the fifth harmonic was avoided (for first-order dispersion from a 1200 mm^{-1} grating, illuminated with a spot diameter of 12 mm, the temporal stretch is about 10 ps). Higher-order harmonics were all absorbed in the quartz optics and in the air outside the vacuum chamber. Finally, the harmonic radiation was focused onto the entrance slit of the streak camera which was optically triggered by a fast photodiode. The temporal resolution was estimated to be 5 ps. The output of the streak camera was recorded with a cooled CCD camera (Hamamatsu C3140-60/61) and stored on a computer hard disk for further analysis.

At the same time as the fifth harmonic was generated in the xenon gas, the same harmonic was also generated using non-linear crystals. The second harmonic radiation was generated in one KD*P crystal and the third harmonic was produced in another KD*P crystal by mixing ω and 2ω. Finally, the fifth harmonic was obtained by mixing 2ω and 3ω in a BBO crystal. This fifth harmonic radiation was recorded with the streak camera by illuminating a separate part of the entrance slit. In this way, not only an absolute time reference, but also a reference temporal profile could be obtained on a shot-to-shot basis. The total dispersion of the non-linear crystals was estimated to be less than 5 ps. To confirm that no saturation occurred in the crystals at any intensity used, the agreement between the square root of the second harmonic profile and the fifth root of the fifth harmonic profile generated in the crystals was monitored.

The repetition rate of the laser and consequently the production rate of the harmonic radiation was 10 Hz. However, the storage of each streak camera image limited the real data acquisition rate to at best 1 shot every 30 s.

3. Results

A large number of temporal profiles were recorded spanning a range of pulse intensities corresponding to pulse energies between 30 and 80 mJ. The lower limit was determined by the sensitivity of the streak camera and the upper limit by the laser system.

The peak intensity of each pulse was calculated (Perry *et al* 1988) according to

$$I_0 = \frac{E_{\text{pulse}}}{\tau_p \pi w_0^2/2} \tag{1}$$

where E_{pulse} is the total energy of the pulse and $\tau_p = \int_{-\infty}^{\infty} T(t)\,dt$ ($T(t)$ is the dimensionless, normalized time profile of the pulse). Assuming the pulse is Gaussian in time, we have $\tau_p = 1.06 T_{\text{FWHM}}$, where T_{FWHM} is the full width at half maximum of the temporal profile. E_{pulse} was corrected for reflective losses in the optics. The uncertainty in our intensity determination was ±70%.

Examples of recorded profiles at different intensities are seen in figure 2. The profiles of the laser pulses were calculated as the square-root of the second harmonic profiles generated in the crystal.

The width ratio of the fifth harmonic pulse generated in the gas and the fifth harmonic pulse generated in the crystal was found to be constant (=0.85) at low intensities. The temporal broadening of the harmonic pulse in the crystals explains part of the deviation from unity. At higher intensities the ratio begins to decrease indicating that the temporal profile of the harmonic radiation generated in the gas becomes narrower than its counterpart produced in crystals where ionization does not occur. The maximum narrowing is about 20% at the highest intensity used.

A comparison between experimental and calculated harmonic profiles is seen in figure 3.

An obvious definition of the temporal shift between the laser pulse and the fifth harmonic radiation generated in the gas would be to measure the time difference between the peak of the second or fifth harmonic generated in the crystal and the peak of the fifth harmonic generated in the gas. However, we chose a slightly different definition, namely the time difference between centres of the pulses at half maximum. This definition emphasizes the overall shift but is less sensitive to fluctuations in the shape of the temporal profile. The relative shift is defined as the absolute shift divided by the rise time of the fundamental and takes into account shot-to-shot variations of the laser pulse width.

Figure 4(*a*) shows the experimental relative shift as a function of laser intensity. Each data point is an average of 10 experimental values thus carrying the same statistical weight. There is no apparent shift below 1.7×10^{13} W cm^{-2} but above that value an approximately linear intensity dependence is observed. The maximum relative shift observed in our experiments was 0.2 (corresponding to 20 ps for a 140 ps laser pulse).

4. Discussion

The relationship between the non-linear polarization and the driving field, is in accordance with the lowest-order perturbation theory:

$$P_q^{\text{NL}}(r, t) = \frac{1}{2^{q-1}} \left(\sum_i N_i(r, t)\, \chi_i^{(q)} \right) E_1^q(r, t) \tag{2}$$

where $\chi_i^{(q)}$ is the qth-order non-linear susceptibility and $N_i((r, t)$ is the atomic (ionic, for $i > 1$) number density.

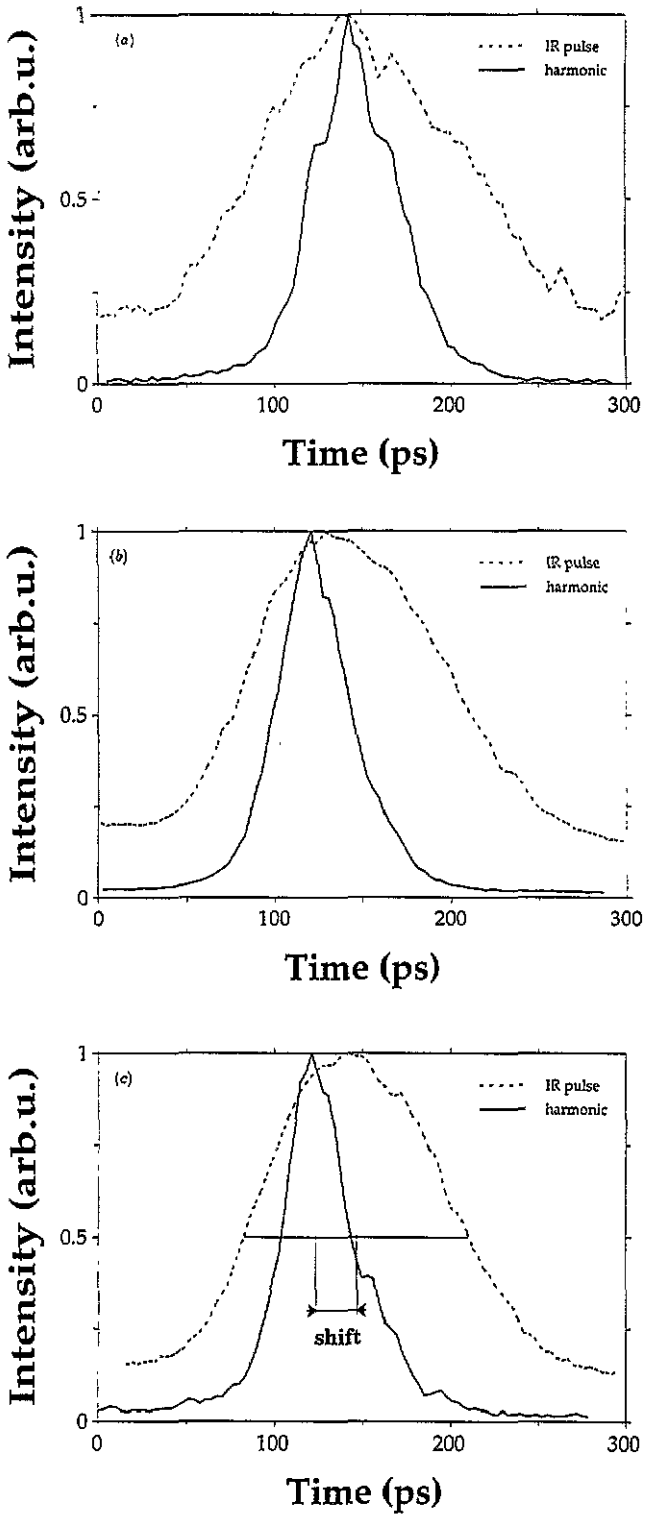

Figure 2. Experimental temporal profiles of the fifth harmonic in xenon (full curve) and laser pulse (broken curve). The laser pulse is calculated by taking the square-root of the second harmonic profile from the crystal. The intensity uncertainty is ±70%. (*a*) At 1.4×10^{13} W cm^{-2}. (*b*) at 2.3×10^{13} W cm^{-2}. (*c*) at 3.5×10^{13} W cm^{-2}.

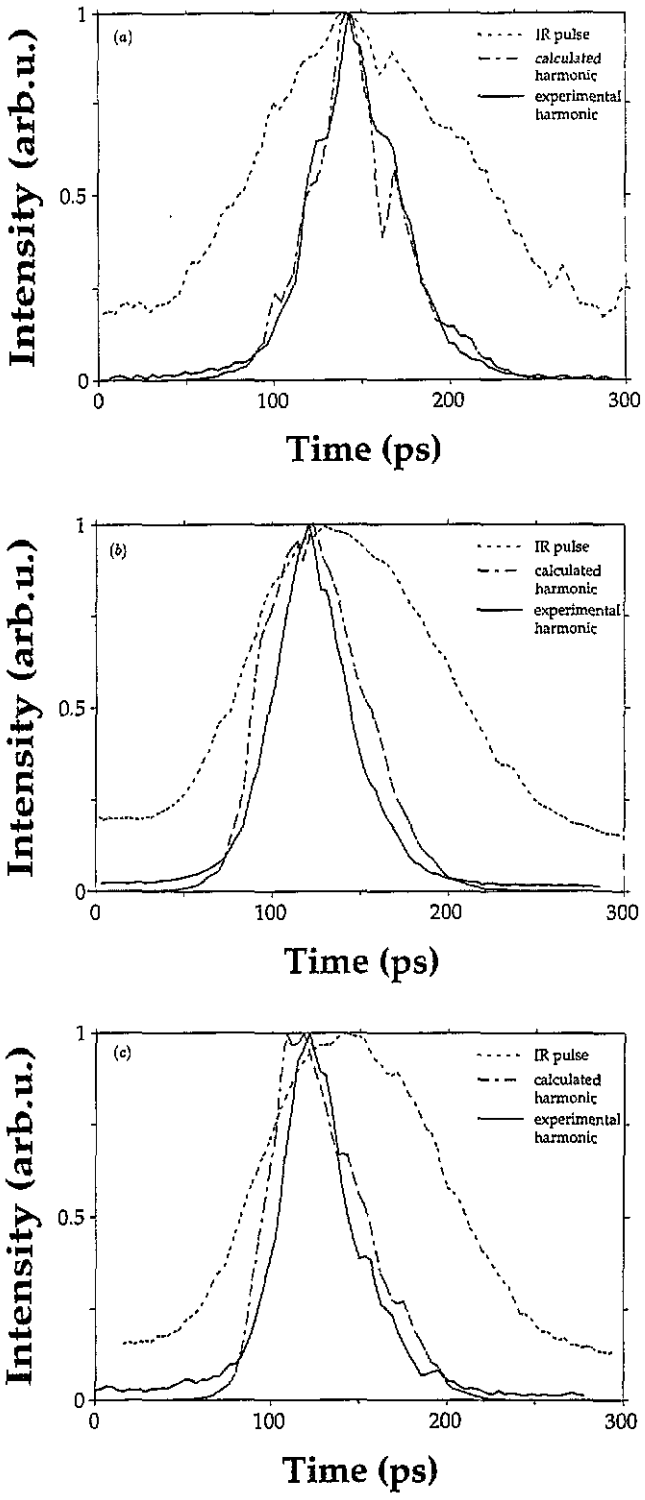

Figure 3. Theoretical fifth harmonic in xenon (chain curve) and experimental fifth harmonic (full curve) with experimental laser pulse (broken curve) as input. The laser pulse is calculated by taking the square root of the second harmonic profile from the crystal. The intensity uncertainity is ±70%. (*a*) At $1.4\times10^{13}\,\mathrm{W\,cm^{-2}}$, (*b*) at $2.3\times10^{13}\,\mathrm{W\,cm^{-2}}$, (*c*) at $3.5\times10^{13}\,\mathrm{W\,cm^{-2}}$.

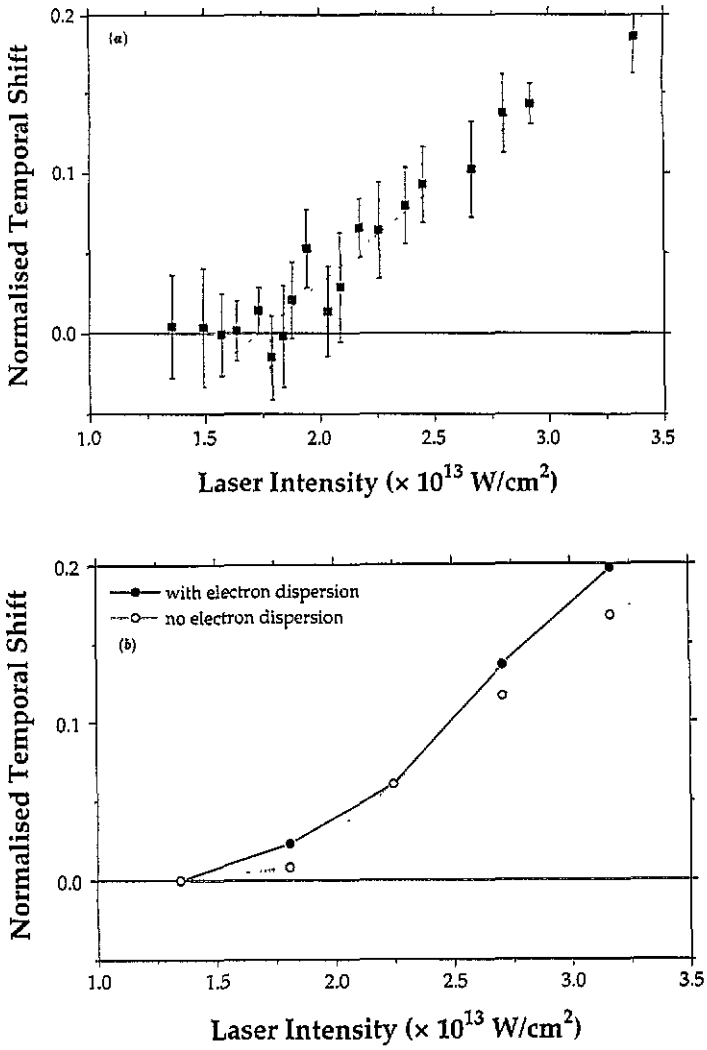

Figure 4. Relative temporal shift as a function of laser intensity. (*a*) Experimental data (intensity uncertainty is ±70%). (*b*) Calculated data with 140 ps FWHM Gaussian laser pulse as input (full line: photoelectron dispersion included, dotted line: photoelectron dispersion excluded).

The propagation equation for the qth harmonic field E_q coupled to the non-linear polarization of the medium P_q^{NL} can be written as (in CGS units):

$$\nabla_\perp^2 E_q + 2ik_q \frac{\partial}{\partial z} E_q = -\frac{4\pi}{c^2}(q\omega)^2 P_q^{\mathrm{NL}} \mathrm{e}^{-\mathrm{i}\Delta k q z} \tag{3}$$

where $\nabla_\perp$ acts on the transverse coordinate r, ω is the driving laser frequency, k_q is the wavevector of the qth harmonic field, and Δk_q is the phase mismatch between E_q and P_q^{NL} (in phase with E_1).

Perturbation theory is not applicable for high-order harmonic generation. However, experimental results (Lompré *et al* 1990) show that at the intensities used in our experiment, deviations from the lowest-order perturbation theory are small for the low-order

harmonics (such as the fifth harmonic, studied in this work) and the harmonic intensity scales with laser intensity as I^q.

The total phase mismatch between the polarization field and the generated harmonic radiation is a sum of two terms: $\Delta k_q = \Delta k_q^{\text{geom}} + \Delta k_q^{\text{disp}}$. Here Δk_q^{geom} is a geometrical phase mismatch orginating from the fact that the laser beam is focused. For a Gaussian beam the geometrical phase mismatch close to the focus is

$$\Delta k_q^{\text{geom}} = 2\frac{q-1}{b} \tag{4}$$

and can be the dominating term when the non-linear medium is a low-density neutral gas. For a given harmonic and a given focusing geometry this geometric phase mismatch is constant. The second term, Δk_q^{disp} originates from the difference in the refractive index of the non-linear medium at frequencies ω and $q\omega$. This dispersive phase mismatch can be expressed as:

$$\Delta k_q^{\text{disp}} = k_q - qk_1 = (n_q - n_1)\, q\omega/c. \tag{5}$$

The resulting harmonic field is a vector sum (i.e. both the amplitudes and phases are taken into account) of partial harmonic wavelets generated in each atom (or ion). Photoionization affects the harmonic intensity principally in two ways. Firstly, the density of neutral atoms decreases (and the density of ions of different ionization stages increases) changing the polarization. Ionic susceptibilities are not very well known but are expected to be much less than the susceptibility of neutral atoms. This leads to a decrease in harmonic generation. Secondly, the free electrons created during the ionization process affect the dispersive properties, Δk_q^{disp} of the non-linear medium. In a fully singly ionized gas with a pressure of 20 mbar, the density of free electrons will be approximately 5×10^{17} cm^{-3}. The plasma frequency of a free electron gas is given by

$$\omega_p = \sqrt{\frac{e^2 n_e}{m_e \varepsilon_0}} \tag{6}$$

where e is the electron charge, n_e the electron density, m_e the electron mass, and ε_0 the permittivity of free space. Under our experimental conditions ω_p was much less than the optical frequency ω, and the dispersion due to the free electrons could be expressed as

$$n - 1 \approx -\frac{\omega_p^2}{2\omega^2}. \tag{7}$$

The phase mismatch due to the electron gas in the medium then becomes:

$$\Delta k_q^e = \frac{q^2-1}{q}\,\frac{\omega_p^2}{2\omega c} \tag{8}$$

where c is the speed of light. The phase mismatch is positive and increases with harmonic order. For the fifth harmonic in xenon under the experimental conditions ($p_0 = 20$ mbar, $\lambda_{\text{laser}} = 1064$ nm), the phase mismatch in the neutral gas, Δk_5 is only about 0.84 cm^{-1}, but the phase mismatch due to free electrons, Δk_5^e when the gas is ionized is about 72 cm^{-1}. Apparently the phase mismatch due to the free electrons completely dominates the mismatch due to the dispersion in the neutral gas. However, during the rise time of a laser pulse, the non-linear medium is exposed to a whole range of intensities, and if the peak intensity is higher than the saturation intensity for ionization, then the phase

mismatch becomes a rapidly varying function of time. The intensity variation in r (due to the spatial mode of the laser) and in z (due to focusing) also affects the phase matching and the total time dependence of the harmonic intensity.

If the peak intensity of the laser is sufficiently high, a temporal shift of the harmonic with respect to the driving laser pulse should be expected. At the beginning of the laser pulse the intensity is low and no ionization takes place. The harmonic intensity increases according to the perturbative I^q law. As time progresses the laser intensity and hence the ionization rate of the medium increase. If the peak intensity of the pulse is sufficiently high, depletion of the neutral atoms and reduction in phase matching occur early during the rise time of the laser pulse. The harmonic generation yield is therefore reduced and the maximum harmonic intensity appears before the peak of the laser pulse and a temporal shift is observed.

This shift could be reproduced rather well numerically despite the simplicity of the model employed. The detailed temporal shape of the harmonic pulse however, could not be reproduced as well. A series of calculations simulating a 35% higher gas pressure were also done. These calculations gave approximately the same shifts but rather different shapes of the detailed structures. This illustrates the need for more accurate gas density measurements, in order to make exact profile modelling possible. Figure 4(*b*) shows the calculated relative time shift as a function of laser intensity. The full line includes the photoelectron dispersion whereas the dotted line represents the situation when the photoelectron dispersion is not included, but the depletion of the medium is still included. The harmonic radiation starts to shift at an intensity closely coinciding with the saturation intensity for ionization in xenon, $I_{sat} = 2.2 \times 10^{13}$ W cm^{-2} at $\lambda = 1$ μm and $T_{FWHM} = 140$ ps (L'Huillier *et al* 1992). The depletion of the medium contributes most to the observed temporal shift in the intensity range used.

5. Conclusions

We have performed temporal measurements of harmonic radiation generated in an ionizing gas. We have studied, in particular, the occurrence in time of harmonic radiation relative to the generating laser pulse. As expected, the harmonic radiation pulses coincide in time with the laser pulses as long as the laser intensity is sufficiently low. However, as the laser intensity increases above a certain threshold intensity, a shift in time between the laser pulse and the harmonic pulse is observed which increases approximately linearly with laser intensity. This behaviour is reproduced in numerical simulations, using a simple model which takes into account the ionization and dispersion of the xenon gas. The numerical calculations show that the dominating effects are depletion of the neutral gas and the dispersion from the liberated photoelectrons which are both due to the ionization of the gas. The depletion of the medium contributes most to the shift.

The duration of the harmonic pulse is also altered by the ionization process. However, the harmonic pulse width at the highest laser intensity used, is only reduced about 20% relative to the pulse width at the lowest intensities.

Acknowledgments

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