Enhancement of high harmonic signal generated by a two-colour orthogonally polarized laser field

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Introduction

Over the last decade High Harmonic Generation (HHG) has become one of the most intensively studied strong field process in nonlinear optics. The interest in high harmonic radiation is rooted in its spatial [1] and temporal coherence [2,3] and in the information on the atomic and molecular structure that the harmonic signal carries [4-8]. The process during which high harmonics are produced is well understood in terms of the three step model [9]. The electron is set free by the intense laser field through tunnel ionisation (1). Once in the continuum the electron is accelerated by the electric field (2) and eventually driven back to the parent ion (3). In the re-collision step, emission of high energy photons may occur, resulting in generation of a high harmonics spectrum of the fundamental laser that ends with a sharp cut-off.

The ionisation and re-collision steps take place within an optical cycle and are therefore inherently ultra-fast and the harmonic emission is the result of the recombination of the electron with its ion. Thus the harmonic signal can be effectively used as an ultra-fast probe of atomic and molecular systems [4]. High harmonics have also attracted a lot of interest in virtue of their potential in producing bursts of XUV and soft X-ray radiation pulses with duration of less than 100 attoseconds [10,11] (1 as = 10^{-18} s), which can have important applications in attosecond science (they can be used in pump-probe experiment) or in the developing of a new generation of laser systems (HHG pulses can be used to seed free electron lasers [17]). However, to fully exploit the potential of HHG, greater control over the HHG process and a significant improved conversion efficiency are required.

The addition of a weak perturbing field has been demonstrated as an efficient way to obtain this twofold intent. Enhancement of the HHG has been observed when using orthogonally polarized two-colour laser fields [12,13]. However, the interpretation of the result is still a matter of debate as it is not clear whether the effect of the second field is merely an enhancement of the ionisation or is also related to the motion of the electron in the continuum.

To extend the cut-off region to higher photon energy a longer wavelength can be used. The maximum photon energy that can be produced follows the cut-off law [18] $E_{max} = I_{ip} + 3.17\,U_p$, where I_p is the ionisation potential of the medium and $U_p = eE_0^2/4m_e\omega^2$ is the ponderomotive energy acquired by the electron driven by the oscillating electric field. Here E_0 is the amplitude of the electric field, e and m_e the electron charge and mass respectively and ω the laser frequency.

As the ponderomotive energy is proportional to the square of the wavelength the use of a frequency in the mid-infrared can extend the cut-off by tens of harmonic orders. Unfortunately the efficiency of the process decrease approximately as λ^{-5} and this is a limitation for the use of longer wavelengths for practical uses [14].

Recently the problem of reduced efficiency in high harmonics generated at longer wavelength has been theoretically investigated by L. Chipperfield *et al.* who showed how the mixing of a small number of harmonics of a 800 nm laser field with the same polarization can be used to synthesize a waveform that maximizes the re-collision energy (as for long wavelengths) while keeping the recombination probability high (as in the case of short wavelengths) [15].

To experimentally address the question of whether it is possible to combine the higher HHG efficiency of shorter wavelengths with the higher photon energies generated at longer wavelengths we investigated high harmonic generation in argon by a 1300 nm (fundamental) beam and its second harmonic (650 nm) that is polarised orthogonally to the fundamental. The objective of the experiment was to verify if the harmonic yield can be enhanced and if the cut-off can be further extended by the weak second harmonic added to the fundamental as well as to determine how the enhancement depends on the relative phase between the two fields.

Experimental methods

The experimental layout of the two-colour experiment is shown in Fig 1. The main beam from the KLM Red Dragon laser system was delivering 9 mJ pulses of 80 fs (measured with a Swamp optics GRENOUILLE) at 780 nm with 1 KHz repetition rate and was used to pump a Light Conversion HE TOPAS producing a 0.950 mJ beam with centre wavelength at 1300 nm. The pulse duration of the IR was 40 fs (measured by a second order autocorrelator).

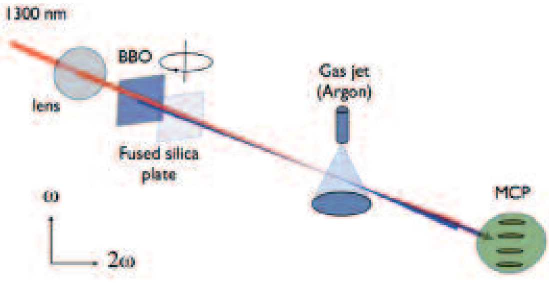

Figure 1. Experimental setup for the two-colours experiment in Artemis.

The 1300 nm beam was sent into the interaction chamber and focused by a 30 cm focal lens just below a continuous flow gas-jet of argon, formed by supersonic expansion of 2 bar of gas through a 100 μm nozzle diameter. A Type I BBO crystal with thickness of 300 μm doubled the frequency of the 1300 nm beam with 20% conversion efficiency. The BBO crystal was placed after the lens to avoid a phase shift due to propagation through the lens and mounted on a kinematic mount to control its tilt. A fused silica glass plate of thickness 300 μm was mounted on a rotation stage and set just after the BBO crystal. This setup allowed us to vary continuously the phase between the IR and its second harmonic as the phase the two beams acquire during the propagation in a dispersive medium is proportional to the wavelength-dependent refractive index. The position of the foci (the same for the two colours) with respect to the gas-jet were set to maximize the harmonic signal.

The laser intensity in the interaction region was calculated to be 10^{14} W/cm^2 and this value was in agreement with the position of the cut-off recorded when only the 1300 was present in the interaction chamber. This was done by changing the tilt of the crystal away from the phase matching angle. The high harmonics produced were spectrally dispersed by a flat-field (1200 lines/mm) and collected by a microchannel plate (MCP) coupled with a phosphor screen. The back of the phosphor screen was imaged by a CCD camera and the images recorded by an acquisition program. Each image was the result of an average of 200 images with 20 ms exposure time.

Results

High harmonics spectra from argon with the two-colour laser fields with orthogonal polarization are shown in figure 2.

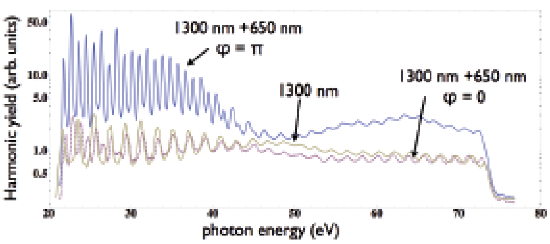

Figure 2. High harmonics generated with fundamental and with the two-colour (ω + 2ω) laser field for two values of the relative phase between the two fields. The first peak is the 23th harmonic of the 1300 nm.

The harmonics from the fundamental field alone were taken tilting the angle of the doubling crystal so that the phase matching condition was not met and no appreciable second harmonic was generated. When the angle of the BBO was at the phase matching angle the harmonics were enhanced by a factor of 2. Further enhancement was obtained by changing the phase between the two fields. Figure 3 shows the periodic oscillation of the harmonic yield as a function of the phase difference between the fundamental and second harmonic (calculated in terms of the different optical path).

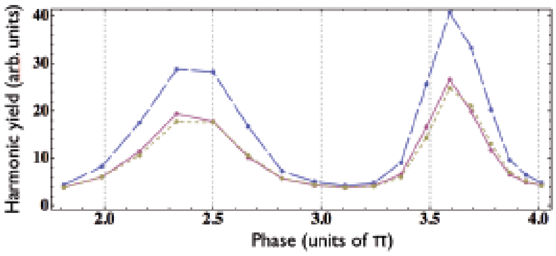

Figure 3. Modulation of the harmonic yield as a function of the relative phase for different harmonics: 24th (dashed), 26th (dotted) and 28th (uniform line).

The π periodicity proves that the enhancement is due to the combination of the two fields as a time shift of full cycle of the second harmonic corresponds to a shift of only half a cycle of the fundamental. As the reference spectrum was taken with minima second harmonic emission from the BBO crystal the laser intensity was higher than in the two colour spectrum, when part of the beam was used to efficiently produce the second harmonic, with a resulting electric field of smaller amplitude. Figure 4 shows the ionisation rate for different values of the phase calculated using the ADK formula [16]. The amplitude of the electric field is normalized to the amplitude of the fundamental field, to take into account the redistribution of laser power between the fundamental and the second harmonic when the crystal is at the phase matching angle. When the phase φ between the two fields is 0 (or π) the ionisation rate becomes higher close to T/4, where T is the fundamental laser period, but classical trajectories calculations show that the re-collision probability is lower in this conditions and the HHG are weaker.

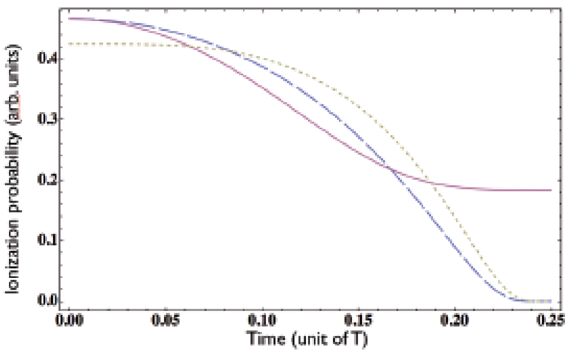

Figure 4. Tunnel ionisation probability in the fundamental field (dashed line), for φ = 0 (uniform line) and for φ = π/2 (dotted line)

When $\varphi = \pi/2$ the ionisation probability is close to the single colour case as the amplitudes of the fields are similar. This suggests that the harmonic enhancement is rather related to the trajectories of the electrons in the continuum rather than to favorable ionisation conditions. Whereas the even harmonics generated by the combined effect of the two colours were clearly present in the low energy part of the spectrum, we did not observed even harmonics at photon energies above 45 eV. The reason is not clear and further analysis is required.

Conclusions

We generated harmonics in a two-colour orthogonally polarized laser field and observed an enhancement of about one order of magnitude respect to the single colour case, where only the fundamental beam was interacting with the gas-jet. The results are quite significant as they shows that the low efficiency in high harmonic generation at long wavelength can be compensated by adding a weak perturbing field, leading to the production of harmonic photons of higher energy than could be obtained from more drive traditional sources (800 nm) and with comparable efficiency. More analysis is required to fully understand the mechanism that is at the origin of the harmonic signal enhancement.

Acknowledgements

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