Enhancement of harmonic emission by a sum of multiple unrelated frequency fields

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Introduction

High-order harmonic generation (HHG) is a highly nonlinear process arising from the interaction of strong infrared laser fields with atoms or molecules. HHG has a pivotal role in modern atomic, molecular and optical physics as it offers a route for the generation of coherent XUV radiation and attosecond pulses [1,2]. Moreover, HHG can provide information about the medium in which it is generated with ultrafast time resolution [3].

Since the discovery of HHG, it has been a constant concern of researchers to enhance the harmonic yield and control the properties of the harmonic radiation. One of the most versatile ways to achieve this is by using two driving fields of different frequencies. Much work has been done investigating HHG with two-colour fields, mainly in a $\omega + 2\omega$ (800 nm + 400 nm) configuration [4-13]. The general conclusion is that the intensity, polarisation, and time structure of the harmonics can be manipulated by controlling the relative intensity, polarisation and phase of the two fields.

The present work was motivated by the need to use relatively long wavelength fields (1300 nm) to obtain high order harmonics from systems with a low ionization potential, where the laser intensity must be kept at relatively low values ($<10^{14}$ W/cm^2). The maximum photon energy in the harmonic spectrum scales as λ^2 but the conversion efficiency decreases as λ^{-5} or faster [14,15]. The multiple frequency beamlines available in Artemis allowed us to investigate different combinations of two-colour fields to enhance the yield of the high harmonics generated with the long wavelength pulses while maintaining the high efficiency of the shorter wavelengths.

Experimental setup

The experiment was implemented using a combination of two laser beams centred at 1300 nm and 780 nm with 45 fs and 80 fs duration respectively. The relative intensities were controlled using variable apertures in the beams; the time delay between the two pulses was adjusted using a Newport translation stage, and their relative polarisation could be varied with a half wave plate inserted in the 780 nm beam. The harmonics were generated by focussing the beams with a 30 cm focal length lens into argon diffused into the chamber through a 100 micron diameter nozzle with 2 bar backing pressure. The harmonics were spectrally resolved by a 1200 lines/mm curved grating which focussed them onto a MCP detector. Each spectrum was averaged over 10^4 laser shots at 1 kHz.

We also took data for a combination of 1200 nm and 780 nm pulses, producing results qualitatively similar to the ones obtained with 1300 nm and 780 nm, and so we will not present them here.

Results

We investigated the effect of the multiple frequency field on HHG by recording the harmonic spectra as a function of the time delay between the two pulses for different relative polarisations. We performed the measurements in three different regimes: (1) Generating harmonics with the 1300 nm pulse and using the 780 nm as the control field ($I_{1300} >> I_{780}$), (2) Vice versa ($I_{780} >> I_{1300}$) and (3) Generating harmonics with both beams at the same intensity.

Common to all three cases was the appearance of non-integer order harmonics when the two pulses where overlapped. These intermediate harmonics are seen as two peaks between the harmonics of 1300 nm or four peaks between those of 780 nm, and are the result of frequency mixing. Their spacing corresponds approximately to odd harmonics of the minimum common multiple of the two wavelengths, 3900 nm.

Only when the intensity of the 1300 nm pulse was equal or greater than the intensity of the 780 nm pulse and the polarisation of the two beams was parallel did we observe any enhancement of the harmonics corresponding to the long wavelength field.

Figure 1(a) shows the recorded harmonic spectra as a function of the time delay between the 1300 nm and 780 nm pulses with parallel polarisations when $I_{1300} >> I_{780}$ ($I_{1300} = 1.5\times10^{14}$ W/cm^2; $I_{780} = 0.2\times10^{14}$ W/cm^2). The appearance of intermediate harmonics can be seen around zero delay accompanied by an increase in the overall signal up to the cut-off of the 1300 nm spectrum. The signal of the 1300 nm harmonics increased by a factor of four, and the enhancement was preceded and followed by a suppression of the signal at –0.1 and +0.1 ps.

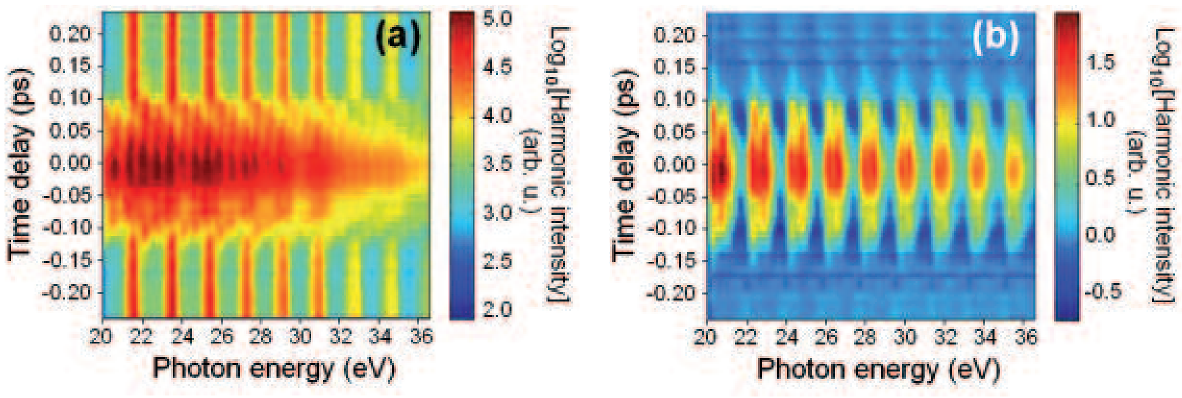

Figure 1. (a) Harmonic spectra of argon as a function of the delay between the 1300 nm and 780 nm pulses (I_{1300} = 1.5×10^{14} W/cm^2; I_{780} = 0.2×10^{14} W/cm^2, parallel polarisation). (b) Same as (a), but normalised to the spectrum with 1300 nm only.

The net effect of the control field can be better appreciated in Figure 1(b), where the harmonic intensity normalised to the spectrum of 1300 nm alone has been plotted. The most prominent effect is the appearance of the non-integer harmonics.

Figure 2 shows a similar scan acquired when the intensities of the 1300 nm and 780 nm beams were of the same order (I_{1300} = I_{780} = 0.5×10^{14} W/cm^2). The intensities were matched adjusting the cut-offs of the harmonic spectra obtained with each field alone. This configuration produced an enhancement factor in the 1300 nm harmonics of more than two orders of magnitude, as can be seen in Figure 2(b).

We also performed measurements with different relative polarisations but the degree of signal enhancement was always greater for parallel polarisations. In fact, the combination of the 1300 nm beam with a perpendicularly polarised 780 nm beam always produced a reduction of the 1300 nm harmonic signal.

We did not observe an extension of the cut-off in any case but an increase in harmonic signal up to the cut-off of the 1300 nm spectrum. Previous investigations with two-colour fields (ω,2ω) have found a modulation of the harmonic signal dependent on the relative phase between the two fields; however, the frequency ratio employed in the present work is incommensurate. This means that their relative phase cannot be well defined as it is intrinsically averaged within the pulse.

There are two possible explanations for the enhancement of harmonic signal in the two-colour field. One is that the control field enhances the ionisation yield by adding amplitude to the total field. The other is that the short wavelength pulse modifies the electric field in such a way that shortens the path of the electrons in the continuum thus reducing the wave packet spreading and increasing their recollision probability. Theoretical modelling is under way to determine the extent of each effect in the present data.

Conclusions

Using a combination of long and short unrelated wavelengths (1300 nm and 780 nm) we have been able to increase the intensity of the harmonic spectrum generated by the longer wavelength by two orders of magnitude. The maximum enhancement was observed when the two fields had similar intensities and their polarisations were parallel. This result demonstrates that the enhancement of harmonic signal in a two-colour field is not restricted to commensurate frequency ratios; moreover, the use of unrelated frequencies allows the generation of non-integer order harmonics.

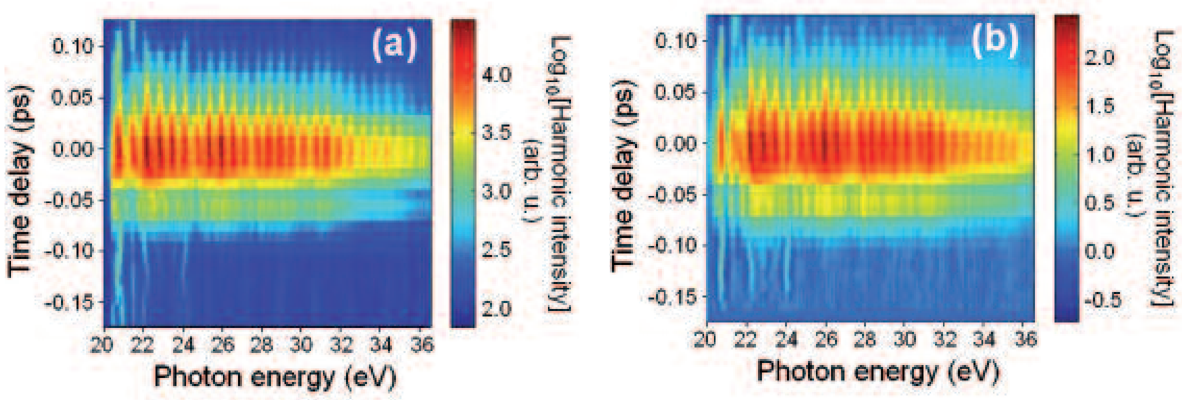

Figure 2. (a) Harmonic spectra of argon as a function of the delay between the 1300 nm and 780 nm pulses (I_{1300} = I_{780} = 0.5×10^{14} W/cm^2, parallel polarisation). (b) Same as (a), but normalised to the spectrum with 1300 nm only.

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