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On the role of harmonics superimposed to the driving field in a harmonic generation process

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Abstract. The spatially integrated emission of harmonics from two along the propagation axis spatially separated sources has been investigated as a function of the relative phase and relative emission power of the two sources. Temporal interference fringes have been observed with frequency distributions including multiples of the driving Ti:sapphire laser frequency. The interference fringes provide evidence of harmonic emission that is sensitive to the relative emission phases and intensities of the two sources. Thus they demonstrate control of the emission at the following source by the harmonics emitted at the preceding one. The observed interference frequencies are discussed qualitatively and some of them are attributed to resonant processes.

PACS. 42.65.Ky Frequency conversion; harmonic generation, including higher-order harmonic generation – 42.65.Re Ultrafast processes; optical pulse generation and pulse compression – 32.80.Qk Coherent control of atomic interactions with photons

1 Introduction

Interference of different mutually coherent excitation channels coupling the same initial and final state may lead to enhancement or cancellation of the excitation process. Thus variation of the relative phase of the excitation paths through the phases of the fields involved may result to an oscillatory modulation of the excitation probability. This effect is established for several years now as phase sensitive coherent control. One way of probing the excitation probability is harmonic generation from the excited state. Such a coherent control of ns 3rd harmonic generation has been experimentally demonstrated previously [1]. Besides the control of the harmonic amplitude, the phase sensitivity of such processes may also serve as a tool for the measurement of phases of harmonics. Such an approach based on perturbative considerations has been suggested recently [2] in relation to the measurement of ultra-short XUV pulses. This work has been followed-up by a non-perturbative investigation of the approach [3]. Nearly dispersionless experimental set-ups for the implementation of this approach have been as well recently studied [4], designed, tested [4] and are currently in use.

In the effort of evaluating less demanding experimental approaches for this type of phase sensitive measurements, we present in this work an alternative set up with which feasibility experiments have been implemented and have resulted to some interesting observations. In this set up two foci separated along the propagation axis of the laser beam, act as the two harmonic sources. In contrast to the two laterally spatially separated harmonic sources of an experiment with similar setup [5], in the present scheme the harmonics produced at the first focus participate in the processes induced at the second focus downstream of the beam and thus may affect them amongst others through phase sensitive interferences. In this case interfering nonlinear (multiphoton) channels may affect the overall harmonic generation. By introducing a variable delay between the laser fields at the two foci, fringing may be observed without special selection in the observed signal, as is the case in the experiment of reference [5]. From the phase control of the conversion efficiency, harmonic phase information could eventually be extracted.

The aim of this lead experiments were to investigate the feasibility of phase control of low harmonic generation (3rd, 5th and 7th). At the same time the measurements presented are an initial effort aiming at better understanding whether and how harmonic generation at a given beam segment may affect the generation process of harmonics (phase and amplitude) at another segment downstream of the beam and thus the overall harmonic generation. Although the coherent control of all these three harmonics is

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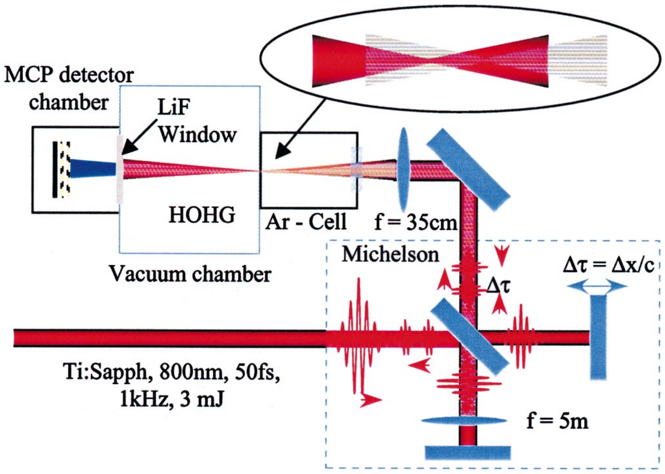

Fig. 1. Experimental set-up.

not firm in these first experiments, interesting, *a priori* not expected fringing patterns have been observed and are reported in the following together with an attempt towards their interpretation.

2 Experimental set-up

The experimental set-up is depicted in Figure 1. The laser used is a titanium sapphire oscillator followed up by a two-stage amplification producing pulses with maximum energy of 3 mJ at ∼800 nm and 1 kHz repetition rate, with a measured pulse duration of ∼50 fs. The laser beam is passed through an autocorrelator creating two colinear time-delayed pulses. The two pulses were focused *via* a 35 cm focal length lens into an Ar gas laminar flow cell, kept at constant pressure [6,7]. In one of the two autocorrelator branches a 5 m focal length lens was introduced, thus producing two spatially separated along the propagation axis focal regions in the quasi static cell. One of the two foci was placed at the ∼100 μm diameter exit pin-hole of the cell and the other one ∼3 cm before it within the same cell. Different combinations of focused intensities between 10^{13} and 10^{14} W/cm^2 were used at each of the foci. Typical Ar gas pressures in the cell were between 30 and 70 mbar. The harmonics generated in the cell entered co-linearly with the fundamental in a pumped vacuum chamber and passed through a LiF window, separating this chamber from the micro-channel-plate (MCP) detector chamber, where the superposition of the window transmitted 3rd, 5th and 7th harmonics were detected. The fundamental, that is transmitted by the LiF window, is not detected by the MCP detector, which for an unfocused beam is blind at 800 nm. This has been confirmed experimentally through the disappearance of the MCP signal upon evacuation of the generating cell.

The MCP signal was monitored through a lock-in amplifier locked at the laser repetition rate as a function of the delay introduced by the autocorrelator. The delay was generated by the translation of the mirrors of one of the branches, through a piezoelectric stage. The minimal step used was 10 nm. The zero delay was not determined in these measurements and thus all delays shown in the following are given with some unknown offset τ, which varies from spectrum to spectrum. At each position of the piezoelectric stage the detected signal was measured for two times the 100 ms time constant of the lock-in amplifier and stored in a PC.

It should be stressed here that the dual foci arrangement is not equivalent to a common auto- or cross-correlator arrangement. The arrangement does not have two geometrical branches and as such it cannot be considered as a conventional interferometer. For instance a first order autocorrelation of one harmonic performed with this arrangement would, because of energy conservation, result into a flat line. No interference fringes can be observed unless special measures are taken. Of course due to the divergence of the beams after the two foci a first order autocorrelation could eventually show fringing if the detection were spatially restricted to a small area of the overlapping cross-sections of the two beams, as has been done elsewhere [5]. This is though not the case in our set up for this and previous [6] experiments. The MCP detector is detecting practically the total beam cross-section thus performing a spatial integration of the total harmonic intensity. Diffracted intensity outside the detector cross-section is negligible. Cross-correlation measurements between the fundamental and harmonics though can be made with such an arrangement. The two branches play now the role of the radiation fields and energy can be transferred from one field to the other and *vice versa*, through up and down conversion, thus taking care for energy conservation.

3 Results and discussion

Interferometric traces have been recorded for different intensity ratios at the two foci. The traces and observed frequencies are found to be sensitive to this ratio. The trace shown in Figure 2a for example is recorded when the laser intensities delivered by the two branches of the Michelson interferometer were approximately equal, showing only the optical interference in a higher order autocorrelation of the laser beam. In the Fourier transform spectrum (Fig. 2b) the dominant frequency is that of the laser, while few lower odd and even harmonics with rapidly monotonically decreasing amplitudes are also present as expected in a higher order autocorrelation [8]. Although the two laser beams do not have the same intensity at each of the foci, the optical interference is observable due to the high contrast of a higher order autocorrelation.

However, adjusting with neutral density filters the intensities of the two interferometer branches to be such that the measured VUV signals produced by each of the two branches are comparable, higher frequencies have been observed. As discussed above the expectation was to observe, due to phase control of the harmonics produced at the second focus, a signal oscillating with delay at the frequencies of one or two or all three transmitted harmonics. Indeed temporal interference fringes were observed, but surprisingly in several spectra higher frequencies in the

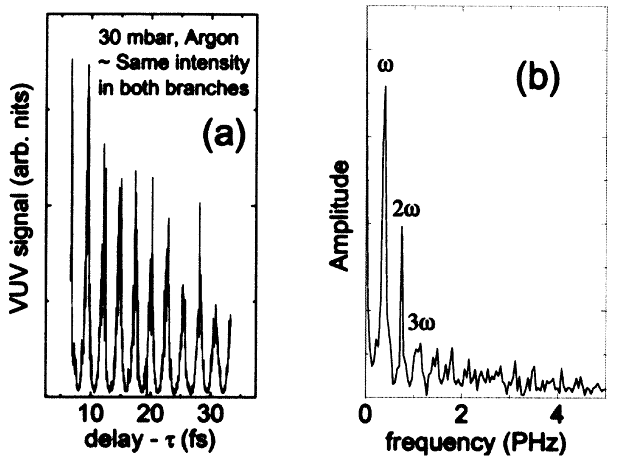

Fig. 2. Interferometric trace recorded with approximately equal laser intensities in the two interferometer branches, depicting only the optical interference (a). Fourier transform of the trace (b).

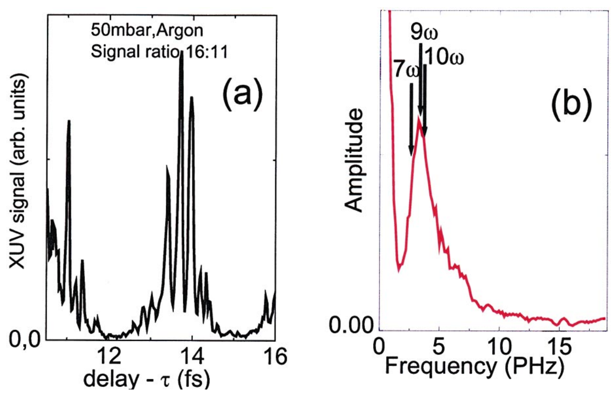

Fig. 3. Expanded region of an interferometric trace recorded with approximately equal VUV signals from the two interferometer branches, depicting high contrast fringing at high frequencies (a). Fourier transform of the trace showing a pronounced peak around the frequency of the 9th harmonic (b).

range of 9–11 times the laser frequency were the dominant features in the frequency distributions of the Fourier transform spectra, while lower frequencies appear with small amplitudes in only few spectra, depending on the signal ratio. This is at first glance surprising because the LiF window does not transmit harmonics higher than the 7th. Two examples of such spectra are shown in Figures 3 and 4.

The highest contrast and most regular interference fringing observed is depicted in Figure 3a. It has been recorded at 50 mbar argon pressure and a close to unity ratio of the signal observed by the two sources independently. This signal ratio was adjusted by the relative intensity of the two interferometer branches. The Fourier transform of the trace is shown in Figure 3b in which a pronounced peak around the 9th harmonic (3.34 PHz) frequency is dominating the spectrum apart of the frequencies originating from the optical interference. The large width of the distribution around this frequency may

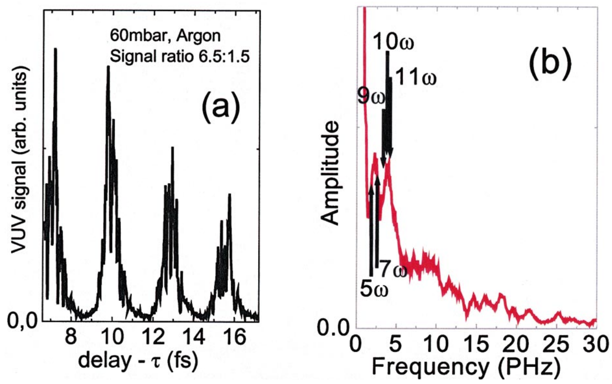

Fig. 4. Interferometric trace recorded with a 6.5:1.5 ratio of the VUV signals from the two interferometer branches, depicting fringing at the optical and higher frequencies (a). Fourier transform of the trace showing two peaks, one including the frequencies of the 5th and the 7th harmonic and the other with a maximum at ten times the laser frequency (b).

be attributed to the stability limits of the interferometric set-up, laser beam intensity fluctuations, the shifting and broadening of the harmonic frequency distributions at higher laser intensities [7] and mainly to the ac Stark shifting of near resonant atomic levels discussed bellow. This width in combination with the high amplitude at the frequency of the 9th harmonic does not allow conclusions about the presence of the 7th (and the 5th) harmonic frequency.

An interferometric trace recorded at a first to second source signal ratio of 6.5:1.5 and 60 mbar of argon is shown in Figure 4a. We show here a delay of 4 optical cycles that makes the high frequency fringing visibly less distinguishable. In the Fourier transform spectrum of Figure 4b again a clear peak in the range 9–11 times the laser frequency is present but now a second peak including the frequencies of the 5th and 7th harmonic is resolved.

Several other recorded spectra showed the same behavior to the three representative spectra of Figures 2–4.

While frequencies close to the 5th and 7th harmonic frequency can be understood by means of the phase control scenario mentioned above (5th or 7th harmonic *vs.* five or seven laser photons), which was the initial target of the study, in contrast the frequencies of the 9th harmonic or ten times the laser frequency are not transmitted by the LiF window and thus are not expected to appear in the trace. Looking at the atomic structure of Ar though it becomes immediately apparent that the 9th harmonic frequency and ten times the laser frequency are near resonant with excited atomic states of Ar. Thus the 9th harmonic is near resonant with the $5s[3/2]_1$ and $3d[1/2]$ and $3d[3/2]$ states and transitions to these states from the ground state are allowed through absorption of odd number of photons. Detunings are of the order of 1000 cm^{-1}. Already the bandwidth of the fundamental is about 450 cm^{-1} and the ponderomotive shift at the intensities used is of the order of few eV and thus more than one order of magnitude larger than the detuning. Ten times the frequency of the laser (0.375 PHz) is fully resonant with the manifold

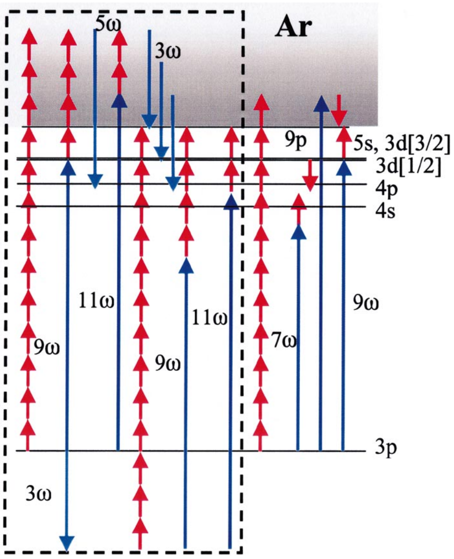

Fig. 5. Near resonant states and interfering coupling channels possibly responsible for the observed high frequency fringing (see text).

of the $9p$ Rydberg states, which are accessible from the ground state *via* absorption of an even number of photons. Furthermore in possible excitation paths of the $4s$ and $4p$ states, detuning is larger ($\sim$4000 cm^{-1}) but not large enough in order to exclude their contribution to oscillations at eight times the laser frequency. The observed dominant interference fringes are obviously due to the presence of such resonances.

It is worth noting that slight lateral displacement of the two sources that could give rise to first order autocorrelation of harmonics, as is the case of reference [5], would not be compatible with the observed higher frequencies, even if detection would be spatially restricted, because of the absorption by the LiF.

There are several scenarios that could qualitatively explain the observed higher frequency peak. Those include: (i) schemes in which the excited resonant state is coupled to the ground state through different coherent channels while the generation of the transmitted and observed harmonics is part of these channels, (ii) generation of the transmitted and observed harmonics from a phase controlled superposition of the ground with an excited state and (iii) harmonic generation from an excited state, while excitation is through different coherent channels.

An example of interfering channels of the scenario (i) is depicted in Figure 5. The diagram is not complete and one can easily add some more channels relevant to the discussions below. Since it is already dense enough we will not include more channels and those shown should be considered as representative examples. If the channels in the dashed line box are induced by photons delivered by one of the interferometer branches (one of the two sources) and the channels outside the box by photons of the other branch, the excitation probability of the resonant states will oscillate at the frequency of the 9th harmonic or ten or eight times the laser frequency as a function of the delay. Thus the third harmonic generation included in the excitation scheme will depict an efficiency that also oscillates at these frequencies. Including higher order processes would lead to the same result for the 5th or 7th harmonic as well, which have lower conversion efficiency than the third harmonic but are detected by the MCPs with higher efficiency. Unfortunately no spectrally analyzed data are available from this experiment, in order to distinguish whether one of the three harmonics as modified through transmission and detection is dominant.

In the scenario of case (ii) the 9th harmonic produced in the first focus by the IR radiation of the first branch of the interferometer propagates to the second focus, where in excites a coherent superposition

$$\Psi_{\mathrm{ge}} = a|g\rangle \mathrm{e}^{\frac{\mathrm{i}E_{\mathrm{g}}t}{\hbar}} + b|e\rangle \mathrm{e}^{\frac{\mathrm{i}E_{\mathrm{e}}t}{\hbar}}$$

of the ground $|g\rangle$ and the $5s$ or $3d$ excited $|e\rangle$ state of Ar with corresponding energies E_{g} and E_{e}. The resulting electronic wavepacket of the non-stationary state oscillates at frequency $\omega_{\mathrm{ge}} = E_{\mathrm{ge}}/\hbar = (E_{\mathrm{e}} - E_{\mathrm{g}})/\hbar$, *i.e.* at the frequency of the 9th harmonic and so does the susceptibility of the medium. A phase shifted interfering replica of the wave packet

$$\Psi'_{\mathrm{ge}} = a|g\rangle \mathrm{e}^{\frac{\mathrm{i}E_{\mathrm{g}}(t-\Delta\tau)}{\hbar}} + b|e\rangle \mathrm{e}^{\frac{\mathrm{i}E_{\mathrm{e}}(t-\Delta\tau)}{\hbar}}$$

may be produced by the $\Delta\tau$ delayed 9th harmonic (or any combination of lower harmonic photons and photons of the fundamental that leads to the same excitation) produced at the second focus by the IR radiation from the second branch of the interferometer. The interfering wave packets of the superposition $\Psi = \Psi_{\mathrm{ge}} + \Psi'_{\mathrm{ge}}$ create a delay dependent susceptibility, oscillating with a period equal to the period of the 9th harmonic. The generation efficiency of the harmonics produced at the second focus and transmitted by the LiF window hence may vary with delay at the same frequency. It should be noted again that fringing due to "optical" interference of the two 9th harmonic fields is not possible in this set up as long as their absorption is linear and spatially integrated. Similar interferences in wave packet interferometry have been studied through ionization [9]. In the present work the probe would be harmonic generation instead of ionization. The same scenario can be considered for the excitation of a superposition of the ground state with the manifold of the $9p$ states by adding one laser photon in the channels discussed above or of the $4s$ and $3p$ states.

Under scenario (iii) the resonant states become populated through two interfering channels (*e.g.* the $5s$ and $3d$ states are excited trough one 9th harmonic photon of one of the source and nine laser photons from the other source (branch) or an other appropriate combination). Population transfer to the excited state thus becomes delay dependent. The harmonic generation efficiency from the excited state is in general different than that from the ground state and thus the measured VUV intensity will depend on the oscillating population ratio between ground and excited state.

All mechanisms described above are compatible with a measured transmitted harmonic intensity that oscillates with delay at higher than the transmitted frequencies as observed in the experiment. A fingerprint of the mechanisms (ii) and (iii) could be the long lifetime of the excited states. Provided that only spontaneous emission causes decay of the induced atomic coherence (superposition) in case (ii) or of the population (excited state) in case (iii) the oscillations in the spectrum should be observable for delays much longer than those warranting temporal overlapping of the to IR pulses. In contrast the mechanism of case (i) would give rise to oscillations only as long as the fields participating in the interference overlap in time. However, fast dephasing or depopulation in cases (ii) and (iii) due to ionization of the excited states cannot be excluded. If ionization is by the preceding IR pulse no oscillations will be present at delays that bring the two IR pulses apart. At the intensities employed in the experiment ionization is not saturated and thus the dephasing or depopulation time is expected to be longer than the pulse duration. However no conclusive interpretation can be made at this stage because of lack of reliable data at longer delays.

4 Conclusions

We have reported experimental results of phase control of strong field short pulse harmonic generation in a set up consisting of two generating sources that are spatially separated along the propagation axis of the laser beam. The delay between the emission of the two pulsed sources is variable, as each source is produced by the laser radiation of each of the two branches of a Michelson interferometer. The total harmonic generation signal could be phase controlled through the delay of the two laser pulses delivered by the interferometer and possible origins of the observed oscillation frequencies have been qualitatively discussed. The study is not completed and the results should be considered as an initial indicative demonstration.

The phase sensitivity of the presented experiments may in principle provide information about the phases of the harmonics involved in the interference schemes. Since the detected harmonics are low order harmonics several resonant states complicate the analysis of the results, introducing a large amount of interfering channels. Whether few of those are dominant could possibly be concluded through quantitative theory. But still the presence of the resonances is known to complicate the determination of harmonic phases [3] in "phase control" type experiments. Thus a reasonable extension of this work would be to higher harmonics with which atomic excitation is far from resonances [3].

The main result of this work is that harmonic generation at a given position along the driving field propagation affects the generation at a position downstream and thus the overall generation efficiency. A further interesting extension of this work would thus be to investigate this type of interferences for a gradual decrease of the distance between the two sources until they eventually essentially merge.

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References

1. N.E. Karapanagioti, D. Xenakis, D. Charalambidis, C. Fotakis, J. Phys. B **29**, 3599 (1996); D. Xenakis, N.E. Karapanagioti, C. Fotakis, D. Charalambidis, Opt. Commun. **152**, 83 (1998)
2. E. Hertz, N.A. Papadogiannis, G. Nersisyan, C. Kalpouzos, D. Charalambidis, T. Halfmann, G.D. Tsakiris, Phys. Rev. A **64**, 051801(R) (2001)
3. M.G. Makris, L.A.A. Nikolopoulos, P. Lambropoulos, Phys. Rev. A **66**, 053414 (2002)
4. E. Goulielmakis, G Nersisyan, N.A. Papadogiannis, D. Charalambidis, G.D. Tsakiris, K. Witte, Appl. Phys. B **74**, 197 (2002); N.A. Papadogiannis, G. Nersisyan, E. Goulielmakis, T.P. Rakitzis, E. Hertz, D. Charalambidis, G.D. Tsakiris, K. Witte, Opt. Lett. **27**, 1561 (2002)
5. M. Kovacev *et al.*, oral and poster presentation at ICOMP IX, Elounda, Greece (2002)
6. N.A. Papadogiannis, B. Witzel, C. Kalpouzos, D. Charalambidis, Phys. Rev. Lett. **83**, 4289 (1999)
7. N.A. Papadogiannis, C. Kalpouzos, E. Goulielmakis, G. Nersisyan, D. Charalambidis, F. Augé, F. Weihe, Ph. Balcou, Appl. Phys. B **73**, 687 (2001)
8. N.A. Papadogiannis, E. Hertz, C. Kalpouzos, D. Charalambidis, Phys. Rev. A **66**, 025803 (2002)
9. M.A. Bouchene, C. Nicole, B. Girard, J. Phys. B: At. Mol. Opt. Phys. **32**, 5167 (1999)