XUV-IR Multiphoton Ionization

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1.Introduction.

The production of short-pulse, coherent, XUV radiation by High Harmonic Generation (HHG) has become a routine operation in many laboratories equipped with an intense femtosecond Titanium-Sapphire laser[1]. The required intensity of 10^{14} to 10^{15} W.cm^{-2} is easily reached with 1-2 mJ, 40-100 fs pulses focused by a long focal length lens (1m). The most usual medium for HHG is a noble gas. Xenon or argon are the most efficient ones (with efficiencies of the order of 10^{-5}) while neon and helium allow for the generation of the shortest wavelengths (2-4 nm) albeit with a reduced efficiency (10^{-9}-10^{-8}). For symmetry reasons, only the odd harmonics of the fundamental frequency are generated and a typical spectrum like the one in Fig. 1 consists of narrow lines separated by twice the fundamental photon energy (1.55x2 eV in the case of Ti:Sap lasers). The harmonic pulses are naturally synchronized with the pump pulse and usually much shorter. This latter property combined to the high brightness, coherence and directivity of HH make them ideal for pump-probe experiments and particularly for multicolor-multiphoton transitions requiring a spatio-temporal overlap of the IR and XUV pulses. Such applications have been carried out in atomic and molecular and solid state Physics. The present work is about recent studies of multiphoton ionization of rare gas atoms by a superposition of HH and IR pulses and their applications to the metrology of femto and attosecond XUV pulses produced by HHG.

From a fundamental point of view, such transitions are of the Above-Threshold Ionization (ATI) type when the HH UVX photons have an energy larger than the ionization potential of the atom, which is the usual situation. The atom is photoionized by absorbing a single HH photon but it may absorb or emit one or several IR photons: the kinetic energy of the photoelectron is then changed by one or several quanta of the fundamental radiation. These ATI transitions are therefore easily monitored by electron spectrometry.

Two-color, IR/UVX ATI transitions have several interesting and unique properties: circular dichroism for instance, is not observed in photoionization of non-chiral systems like an atom in the ground state (although it may be observed in double photoionization). However circular dichroism is predicted in non-chiral systems for two-photon, two-color ATI transitions, due to interference in the transition amplitude.

The presence of the IR field induces multiphoton transitions but also produces ac-Stark shifts of the atomic levels and in particular leads to an *increase* of the ionization potential equal to the ponderomotive energy. This induces a corresponding *decrease* of the electron energy and a broadening of the kinetic energy distribution. In the case of a very short harmonic peak, the broadening is proportional to the derivative of the ionization potential change with respect to time at the center of the harmonic pulse. This property, that we called "ponderomotive streaking of the ionization potential" has been applied to measuring 10fs harmonic pulses with a 40 fs IR pulse, without the limitations of the usual cross-correlation method[3]. The lower limit of this method using Ti:sapphire harmonics is about 3fs.

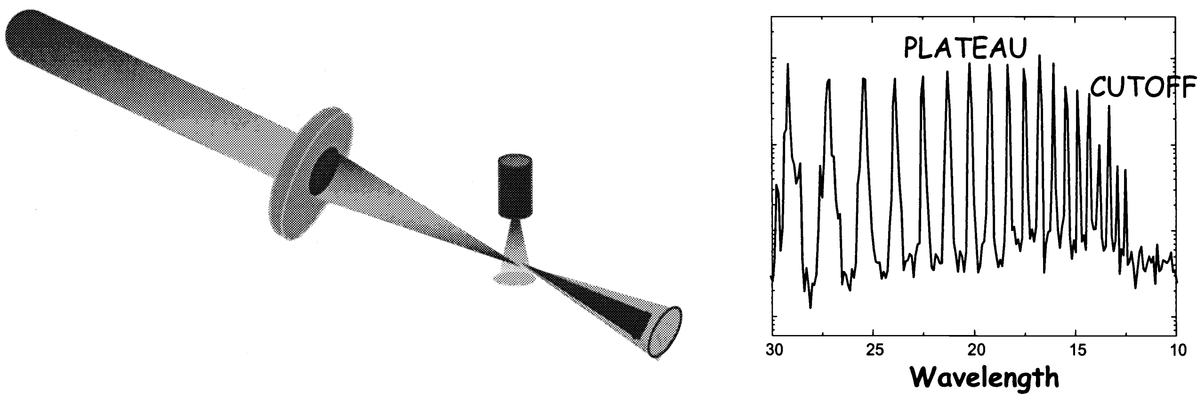

Fig. 1 Harmonic generation: an intense fs laser pulse is focused in a gas jet resulting in a spectrum of odd harmonics with constant amplitudes up to the cutoff region.

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Another property of ATI produced by superposing *several* consecutive harmonics and the fundamental light is that a given final state may in general be reached through several quantum paths: for example, a final state differing by one fundamental photon from the energy of a photoelectron peak caused by a given harmonic may be due to the absorption of on harmonic and one IR photon or the absorption of the next-order harmonic and the emission of one IR photon. The transition amplitude turns out to depend on the phase difference of the two consecutive harmonics. By scanning the relative phase of the IR and harmonic pulses, the harmonic phases can therefore be retrieved. This is a unique method to determine this phase and in particular to test the possibility of attosecond pulse train generation by HH. Again using ATI in argon we have observed that high harmonics of orders from 11 to 19 are indeed almost phase-locked and, therefore, correspond in the time domain to a train of $250x10^{-18}$ s (attosecond) pulses.

2.Two-color multiphoton Above-Threshold Ionization (ATI)

Two-color ATI transitions involving one HH, XUV photon and one IR photon from the fundamental pump laser have been investigated with the following setup:

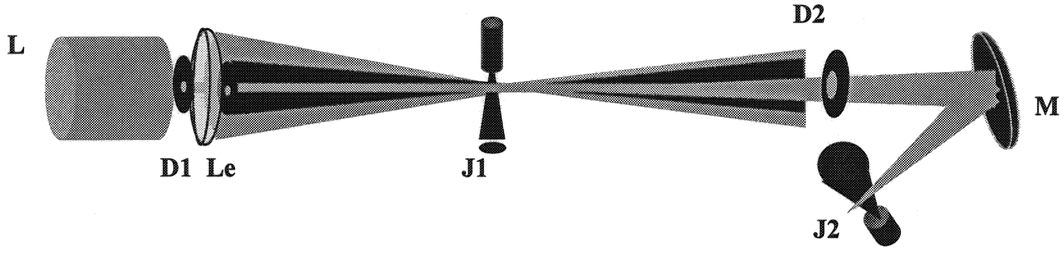

Fig.2 Schematic experimental setup

The laser beam (L) is split into two secondary collinear beams by the annular diaphragm (D1) : the most intense outer part is focused by the lens (Le) into a first atomic jet (J1) in which HH are generated and is stopped by the aperture (D2) located some 30 cm after (J1) and conjugated from (d1) by the lens. The harmonic beam as well as the central (weak) part of the laser beam which propagates through (J1) without interaction pass through the aperture (D2) and are both focused into a second atomic jet (J2) by the mirror (M). The intensities and polarization the two secondary beams as well as the delay between them may be controlled and varied to suit the needs of particular experiments as detailed hereafter. Photoelectrons produced by interaction of the IR pulse from the central beam and HH in the jet (J2) are analyzed with an electron energy spectrometer (for instance time-of-flight).

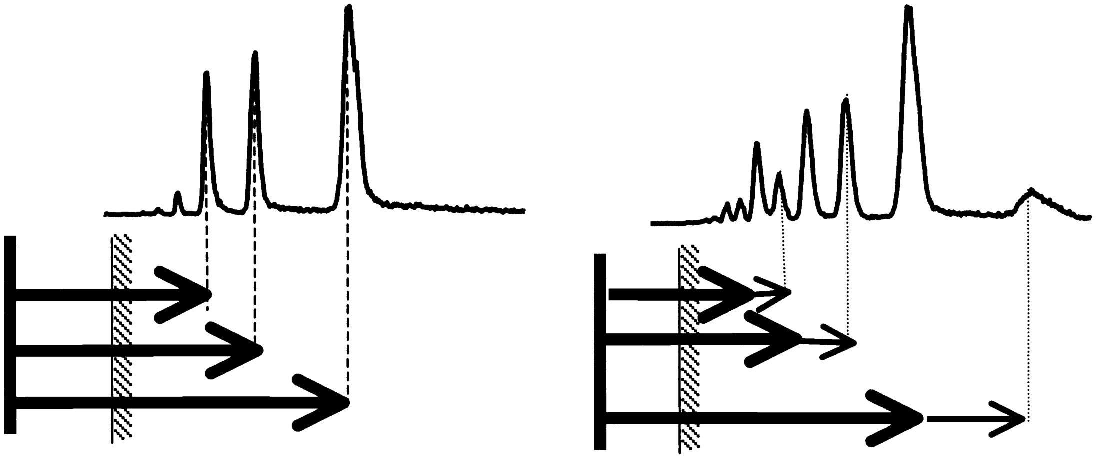

Fig. 3 XUV-IR two-photon ionization transitions and photoelectron time-of-flight spectra. Left: High Harmonics alone. Right: HH+ IR added, showing extra peaks, signature of such transitions.

Typical photoelectron energy spectra resulting from the two-color interaction give rise to discrete peaks at energies ($\hbar = 1$):

$$E_q=(2q+1)\omega \pm \omega - I_P \qquad (1)$$

Where ω is the fundamental (IR) photon energy, q, an integer number and I_P the atom ionization energy. The IR intensity required to induce a two-color ATI ionization is easily estimated from a classical argument: The photoelectron created by absorption of one HH (of order $2q+1$) photon has an energy $U_0= (2q+1)\omega - I_P$. The drift kinetic energy it gains from the IR field is given by $\sqrt{8U_P U_0}$ with U_P the ponderomotive energy[2]. It must be at least equal to the energy of one IR photon which correspond to an intensity of a few times 10^{13}W.cm^{-2}. In practice, the IR intensity necessary to observe such *sidebands* is about an order of magnitude less.

Interest in such transitions arises both from fundamental and applied viewpoints. The sections 3 and 5 investigate two fundamental properties while Sections 4 and 6 report on applications to the measurement of XUV ultrashort pulses.

3.Circular Dichroism

Non-chiral systems like atoms in the ground state display optical activity only in some cases of double-photoionization. However, second order perturbation theory predicts[3] that some circular dichroism does exist for two-color, two-photon ionization for special geometry: it can be shown that if the two beams are propagating collinearly, say along the z direction, with their polarization lying in the (xy) plane, and if the photoelectron is detected in the y direction (azimutal angular distribution at θ=90°) then maximum circular dichroism should be observed when the linear polarization of the HH photon is at 45° of the x or y axis.

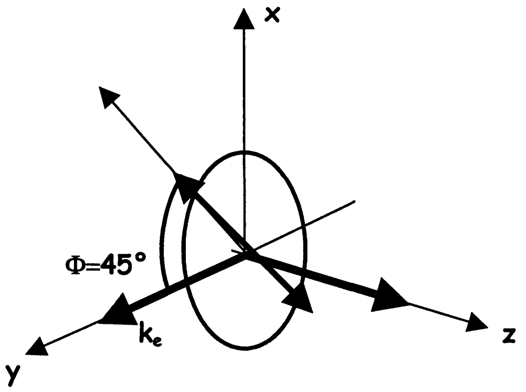

Fig. 4 Geometry for maximum dichroism in two-photon, two-color ionization

This can be understood qualitatively in the following way: the quantization axis being taken along z, the HH linear polarization is a superposition of a right and a left circular polarization and induces transitions such that Δm=±1, ΔL=±1. The possible final states, if the initial state is an s state, have angular momenta 0 and 2 and the s and d outgoing waves interfere. This interference gives rise to the circular dichroism.

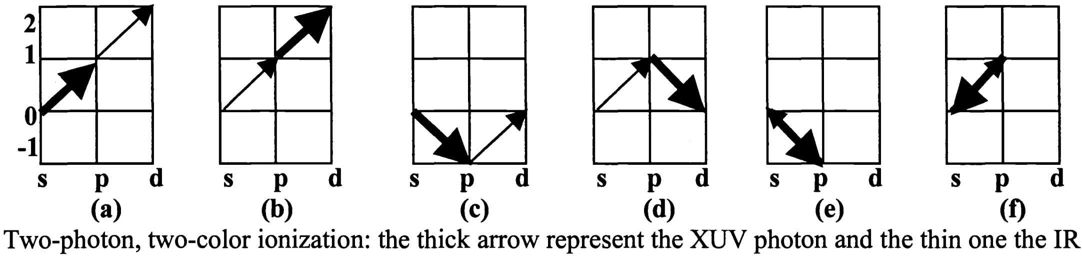

Fig. 5 Two-photon, two-color ionization: the thick arrow represent the XUV photon and the thin one the IR. Each box display a possible combination of a right circular IR photon and a linear blue photon. Numbers are m values.

Diagrams similar to those of Fig.5 may by drawn for initial states with m=-1 and m=1 (initial p state like in argon). The experiment is rather delicate since great precautions must be taken to insure real circular polarization of the IR beam. Moreover the effect is small. The setup used is that of Fig. 2 with introduction of a quarter-wave plate on the small central beam and a half-wave plate on both. We have used harmonics 13-19 in argon and detected a small dichroism on harmonics 13 and 15, as predicted by the theory.

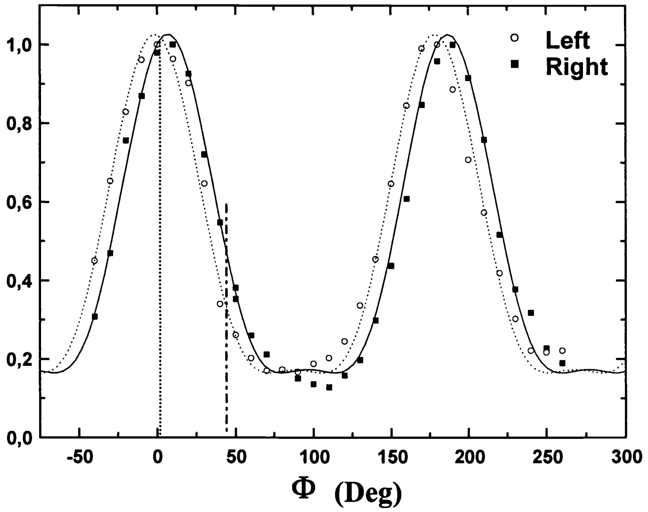

Fig. 5 azimutal angular distributions with respect to the angle Φ between the HH linear polarization and the direction of the detector

The two curves are shifted with respect to each other and the dichroism is measured by this phase-shift: it is zero close to Φ=0° where the two curves cross and maximum around 45°, in agreement with second-order perturbation theory. According to theory[3] the dichroism should rapidly decrease with the harmonic order (or the photoelectron kinetic energy)]. The experimental accuracy is insufficient to confirm this trend. Moreover, the calculation predicts a $\sin(2\Phi)$ dependence for the azimuthal angular distribution while the experiment data is fitted by sin polynomial of higher order. It is likely that this is the result of non-perturbative effects due to the IR intensity as elaborated in the next sections. Indeed, if the IR intensity is high enough, several IR photons may be exchanged for each XUV photon absorbed. For instance two IR photons may be absorbed resulting in a photoelectron with kinetic energy (q+2)ω coinciding with that of the next order harmonic photoelectron. Alternatively, one IR photon may be absorbed and another one emitted, resulting in the net absorption of one XUV photon. In the latter case the usual photoeffect is modified by the presence of the IR which changes the photoelectron angular distribution and kinetic energy.

4.Ponderomotive streaking of the ionization potential

One experimental criterion to decide of the validity of second order perturbation theory is the shift of the photoelectron peaks in the energy spectra. An IR field induces ac-Stark shifts on the atomic energy levels. These shifts affect mostly the loosely bound Rydberg states which are *up-shifted* by the ponderomotive energy U_P and much less the ground state which is *down-shifted* by about 0.1 U_P. As a consequence, the ionization potential is *increased* by about 1.1U_P and correpondingly, the photoelectron energies *reduced* by the same amount. The minimum observable shift obviously depend on the energy resolution of the spectrometer, the statistics of spectra etc. : in our time-of-flight spectrometer, a shift of 100meV is easily detected. When such shifts appear, it is clear that the second order approximation must break down. In cross-correlation measurements, one insures that the intensity is low enough to keep the dependence of the sideband peaks linear with the IR intensity to avoid cumbersome and inaccurate corrections[ref]. This is obviously a limitation since low IR intensity means low sideband amplitude and, accordingly, low dynamic range in the measurement. It occurred to us that it would be interesting to use more IR intensity and more amplitudes in the sidebands and take directly advantage of the shifts for measuring XUV ultrashort pulses. The more so as the cross-correlation time resolution is strongly limited by the IR pulse duration. In fact as the cross-correlate width is something like $\sqrt{\tau_{XUV}^2 + \tau_{IR}^2}$, the deconvolution procedure necessary to retrieve τ_{XUV} loses all accuracy when $\tau_{XUV} << \tau_{IR}$ which is unfortunately the most frequent situation. The ponderomotive shift of the ionization potential offers an attractive solution to this problem and gives access to much higher resolution[4]. Let us assume that the XUV pulse *is* much shorter than the IR one and, moreover that a delay may be introduced between the two in such a way that, in the second jet (J2 of Fig. 1), the XUV pulse occurs in the rising edge of the IR pulse. Then the ponderomotive shift is approximately a linear function of time during the XUV pulse. Roughly, the photoelectron energy spectra is broadened by an amount equal to $\frac{dU_P}{dt}\tau_{XUV}$. Measuring the peak width, and providing knowledge of the rise time of the IR pulse yields therefore directly the XUV pulse duration. The IR pulse profile is normally well established by autocorrelation. For a gaussian pulse at the inflexion point for instance, the rate of increase of the intensity is 0.8577 U_P/τ_{IR} while for an hyperbolic secant square pulse it is .78 U_P/τ_{IR}. It should be stressed that one measurement for a well adjusted delay between the two pulses is sufficient. For the sake of demonstration, we have varied this delay though and verified though numerical simulations that the same duration was obtained for all delays corresponding to non-zero rate of increase (i.e. excluding the situation where the XUV and IR pulses are peak to peak: in this case the photoelectron peak is shifted but not broadened and the measurement yields information on the peak IR intensity only).

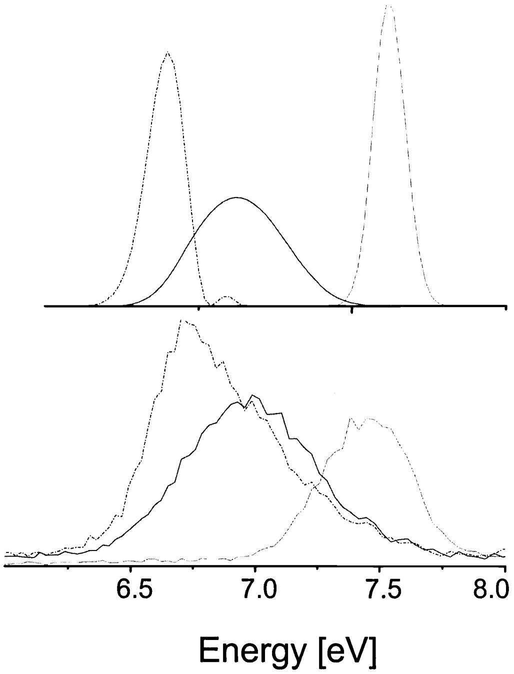

Fig.6 Experimental (lower part) and theoretical (upper part) demonstration of the ponderomotive streaking of the ionization potential as a method for measurement of ultrashort XUV pulses. The three curves in each part correspond respectively to zero IR intensity (dashed line); 2.5×10^{13}W.cm^{-2} and zero delay (dot-dashed line); same intensity and XUV pulse delayed by about 10fs (solid line).

The experimental result of Fig.6 was obtained with the harmonic 15 in argon and an IR intensity of about 2.5×10^{13}W.cm^{-2} producing a Stark shift of about 1eV. It is worth mentioning that this method is applicable to any quasi monochromatic XUV or X-ray pulse with a duration much shorter than the IR pulse but long enough for allowing the spectral resolution of the photoelectron peak from its sideband. Since the optical cycle at 800nm (Ti:Sapphire laser) is 2.7 fs, one may estimate the resolution of the method to 3-5fs. For a shorter pulse, the corresponding energy spectrum would yield peak widths larger that their separation. It follows that the ponderomotive streaking of the ionization potential provides an attractive alternative to the cross-correlation method with a number of advantages: considerably improved resolution, wide spectral range and potentially single-shot operation. This method has been used[5] to determine the duration of the shortest single harmonic pulse as yet produced (2.7 fs). It is insufficient though to characterize subfemtosecond pulses in the time domain an another approach is required as discussed hereafter.

5.IR-modified photoionization angular distributions

Still another signature of non perturbative effects of the IR field can be observed in the angular distributions of the photoelectrons resulting from the net absorption of one harmonic photon. In the absence of IR, the polar distributions of such photoelectrons is given by the well known relation:

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{4\pi}(1 + \beta \frac{\cos^2\theta - 1}{2}) \qquad (2)$$

where θ is the angle between the directions of the field polarization and that of the photoelectron momentum, β the asymmetry parameter and σ_0 the total cross-section[6]. The presence of an intense IR field deeply modifies such angular distribution as shown in Fig.7. Trying to fit such a distribution by Eq.(2) leads o unphysical values of β larger than 2. Since, in order to make a transition corresponding to the absorption of one net XUV photon, at least two IR photons must be exchanged (one absorbed and one emitted), the angular distribution corresponds to a three-photon one and would have the form[7]:

$$\beta_0 + \beta_2 \cos^2\theta + \beta_4 \cos^4\theta + \beta_6 \cos^6\theta \qquad (3)$$

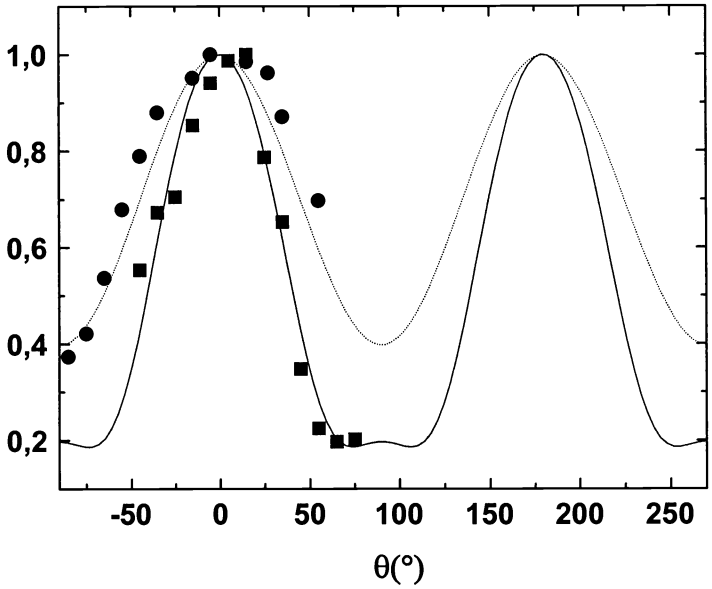

Fig.7 Angular distributions of photoelectrons produced by absorption of one photon of harmonic 13 in argon in absence (circles) and presence (squares) of IR with intensity of about 10^{13}W.cm^{-2} . The lines are fits according to Eq.(2) (dashed line) and Eq.(3) (solid line).

The angular distribution is more peaked than the one with no IR field present as higher angular momenta become accessible in the final state. Thus from an initial p state, photoionization leads to s or d waves without the IR field but to s, d and g waves by absorption and emission of two IR photons. Third-order perturbation should in principle allow to calculate the correction but, because the three-photon transition involves an intermediate state in the continuum, divergences appear in the calculation and numerical techniques to solve the time-dependent Schrodinger equation are required. This work involves difficulties as well and is currently under progress.

6.Observation of a subfemtosecond pulse train from High Harmonics

As seen above, there are methods to determine the duration of the Harmonic pulses with a good resolution. However, if one consider now, not a single harmonic but group of harmonics of consecutive orders, the temporal beating of these harmonics could lead to intensity fluctuations well below the resolution of these methods. Actually, theory does predict that a group of consecutive harmonics from the plateau should form in the time domain a train of extremely short (subfemtosecond) pulses repeating with a period equal to half the fundamental optical cycle[8]. Indeed if a group of spectral components separated by 2ω are locked in phase, their Fourier transform is a periodic succession of ultrashort spikes of period T/2 (with T=2π/ω) and a duration equal to 1/2N where N is the number of spectral components (i.e. harmonics orders) in phase. Note that it is sufficient that the spectral phases are a linear function oif the frequency (or of the harmonic order) to produce a train of attosecond pulses: if $\varphi(q) = \varphi_0 + aq$, the q^th^ harmonic phase factor writes $e^{iq(\omega t+a)}$ and they still form the geometric progression whose sum leads to the ultrashort pulses. A small amount of quadratic chirp on the other hand yields a phase factor of the form $e^{iq(\omega t+a+bq)}$ and rapidly deteriorates the contrast:

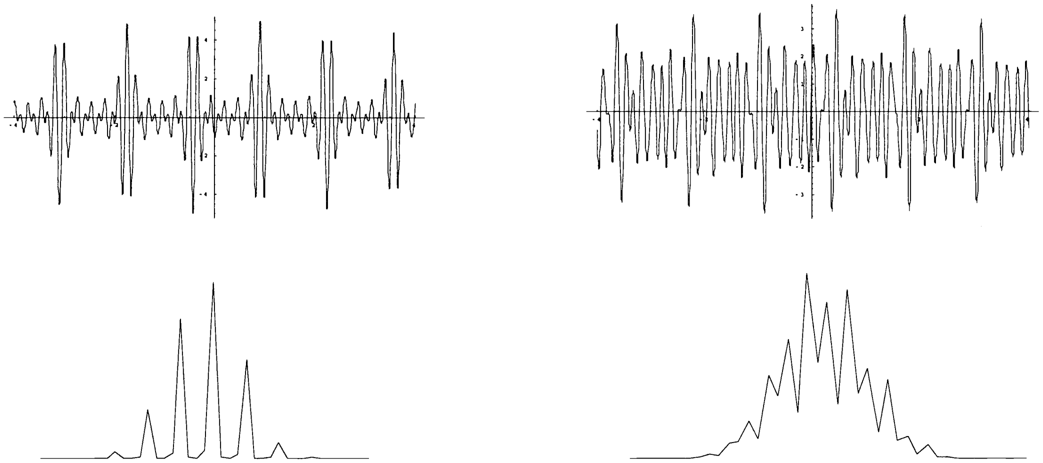

Fig. 8 Field from the superposition of 5 harmonics (q=11 to 19) with linear spectral phases (a=1.; b=0) (upper left)and quadratic (a=1.; b=0.3)(upper right); the corresponding autocorrelates are respectively on the lower left and lower right.

It would be of course desirable to be able to autocorrelate the pulses produced by such a group of harmonics. This would readily inform on their time structure as shown in Fig.8. Unfortunately, the intensity of high harmonics is insufficient to allow for the observation of two-photon processes. Cross-correlation with the IR pump field would obviously lack the resolution. If the analyzis of the pulse in the time domain appears for the time being impossible, an elegant solution exist in the frequency domain, as proposed by Véniard, Taieb, Maquet. The VTM method, as we shall call it, is based on a quantum interference which involves the phase difference between two consecutive harmonics: consider an atom irradiated simultaneously by such two harmonics of orders q and q+2 and an IR field of modest intensity such as to warrant the validity of second-order perturbation theory. The transition amplitude from the ground state to the final state $|\Psi_f\rangle$ of energy (q+2)ω −ω=q ω +ω is the sum of the two terms:

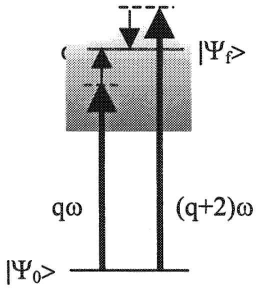

Fig.9 The quantum interference basis of the VTM method for the harmonic phase retrieval

Each term, which represent the amplitudes to reach the final state through one of the two quantum paths indicated in Fig. 9 contains the product of the IR and HH fields and writes (only the phase factors are shown):

$$e^{i(q+2)\omega t+i\varphi_{q+2}}e^{-i\omega t+i\varphi}+e^{iq\omega t+i\varphi_q}e^{i\omega t-i\varphi}=e^{i(q+1)\omega t}\left(e^{i(\varphi_{q+2}+\varphi)}+e^{i(\varphi_q-\varphi)}\right) \qquad (4)$$

where φ_q , φ_{q+2} and φ are the two harmonics phases and the fundamental phase (with respect to the harmonics) respectively. This latter phase may be written as ωτ if τ is a delay between the fundamental and harmonic fields. To the terms of Eq.(4) one should add the terms obtained by exchanging the order of the photons but they have the same phase factor and do not change the result. Another phase term arises from the second order matrix elements themselves which are complex, due to the divergence appearing in the summation over the intermediate states (which introduces a principal part and a complex term). Denoting these phases by θ_{q+2} and θ_q and taking the modulus square of (4) yields:

$$1+\cos(\varphi_{q+2}-\varphi_q+\theta_{q+2}-\theta_q+2\omega\tau) \qquad (5)$$

As a consequence, the probability of the transition leading to the final state $|\Psi_f\rangle$ of Fig.9 contains an oscillating term whose phase includes the phase difference between the two harmonics. Experimentally, this transition can be monitored by following the amplitude of the sidebands discussed here above as a function of the delay between the part of the fundamental pulse that generates the harmonics (the outer part in Fig.1) and that of the small central part which creates the sidebands. If everything else is kept constant, the oscillation should have a period equal to half the fundamental period (factor 2ω in Eq.(5)). To retrieve the harmonics phase difference, the atomic phase must be subtracted from the phase of the cosine function. This phase must be independently obtained by calculation.

The experiment has been described in details elsewhere[9] and its result has shown unambiguously that for the harmonics 11 to 19 generated in argon the phases where indeed a linear function of the order as shown by the squares and the linear fit in Fig.10.

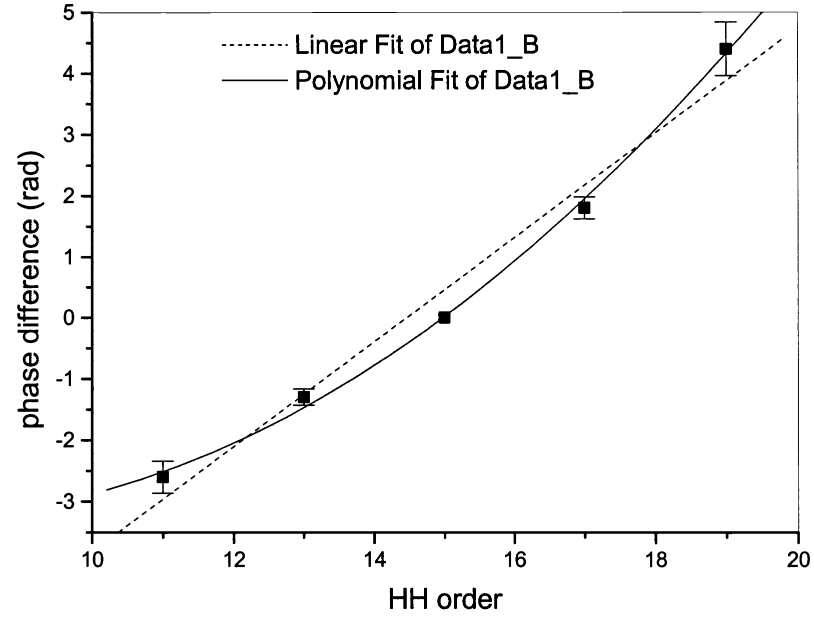

Fig.10 Experimentally determined harmonics phases vs order (q=11-19). The dotted line is a linear fit, the solid line a quadratic fit through the same data.

The pulse train which can be deducted from this measurement, the amplitudes of the corresponding harmonics and the calculation of the atomic phases was shown to have substructures of 250 as with the period of 1.3 fs as predicted by Eq(5). The phases of Fig. 10 could be fitted with a quadratic function of q (solid line in the Fig.) which may suggest that, were more harmonics added to the group of five, their contributions could damage the constructive interference, as easily seen when extrapolating the quadratic fit to the next few harmonics.

According to the theory, the locking of the harmonic phases occurs through the phase matching during the propagation, i.e. the macroscopic response of the medium[8]. The single atom response would lead, at best, to *four* attosecond pulses per fundamental optical cycle (instead of two after propagation and as in the present experimental result). It would be very useful to experimentally control the harmonics phases either at the single atom level[10], through control of the macroscopic generation or through optical elements after the generation, to be able shape the final pulse train.

7. Conclusion

We have explored experimentally multicolor, multiphoton XUV-IR ionization processes both within and beyond the perturbative limit. We have obtained original results including circular dichroism and IR-modified photoionization angular distributions. We have, in the course of these studies, designed a new method for measuring ultrashort pulses with a resolution of 3 fs and avoiding most of the drawbacks of cross-correlation. Finally, and perhaps most importantly, we have applied our experience of such transitions to implement the VTM method for harmonic phase retrieval and test the harmonics 11-19 in argon. The result leads us to assert that such harmonics form in the time domain a train of sub-pulses as short as 250 as, thus breaking the femtosecond barrier for the first time.

8. References

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