Spectral Phase Distribution Retrieval through Coherent Control of Harmonic Generation

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The temporal intensity distribution of the third harmonic of a Ti:sapphire laser generated in Xe gas is fully reconstructed from its spectral phase and amplitude distributions. The spectral phases are retrieved by cross correlating the fundamental laser frequency field with that of the third harmonic, in a three laser versus one harmonic photon coupling scheme. The third harmonic spectral amplitude distribution is extracted from its field autocorrelation. The measured pulse duration is found to be in agreement with that expected from lowest order perturbation theory both for unstretched and chirped pulses.

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The driving of a nonlinear medium through two different mutually coherent fields, e.g., through n photons of the fundamental laser field and one photon of its nth harmonic, leads to interference effects that allow the control of the interaction products. Such an effect is the well-known quantum interference process of phase sensitive coherent control [1]. By varying the relative phase between the two fields the induced atomic or molecular polarization may be continuously modified and with it excitation rates, ionization, and/or dissociation branching ratios, autoionization [2], as well as harmonic generation [3–5], can be manipulated in a controlled manner. In particular, harmonic generation may similarly be manipulated by coherently increasing or decreasing the field amplitude by controlling the nonlinear polarization of the generating medium through the phase of the driving field. Utilizing this type of control effect in reverse order, the relative phase between mutually coherent fields may be determined by measuring a rate proportional to the square of the polarization induced by the polychromatic field. Such an approach has been recently proposed for the determination of the relative spectral phase distribution of an individual harmonic or a superposition of harmonics [6], an issue highly pertinent to attosecond pulse (as) metrology [7–11]. Knowledge of the spectral phase and amplitude distributions of the harmonics is required for the reconstruction of the as pulses. The approach is related to what is known as the RABBITT method [8], one of the most widely used methods in as pulse characterization. However, RABBITT determines a mean relative phase between subsequent harmonics and not the phase distribution within the bandwidth of each harmonic. In contrast, the proposed method [6] is frequency mode selective and thus accounts for the entire spectral phase distribution, allowing complete reconstruction of the waveform. The phase distribution retrieval though is based on lowest order perturbation theory (LOPT), which is a valid assumption for many but not all coupling schemes. The limits of the validity of the method have been assessed in this respect theoretically [12]. This study indicates that phases can be retrieved when excitation is far from dynamic resonances. Thus for higher harmonics and high IR fields the validity of the approach has to be verified *through ab initio* theory. The method is in any case valid for lower harmonics, below the plateau, that are far from resonances. This is an unexplored and important spectral region of harmonic generation, as these harmonics may give rise to attosecond pulses with wavelengths suitable for single photon atomic and molecular electronic excitations.

In the present Letter we present the first experimental implementation of such an approach, in determining the spectral amplitude or phase distribution of the third harmonic (TH) of a femtosecond Ti:sapphire laser beam, generated in Xe gas.

The scheme under investigation is depicted in Fig. 1. The TH field $E_3^{(1)}$ to be characterized, generated in a nonlinear medium (NLM1), is cross correlated with the nearly

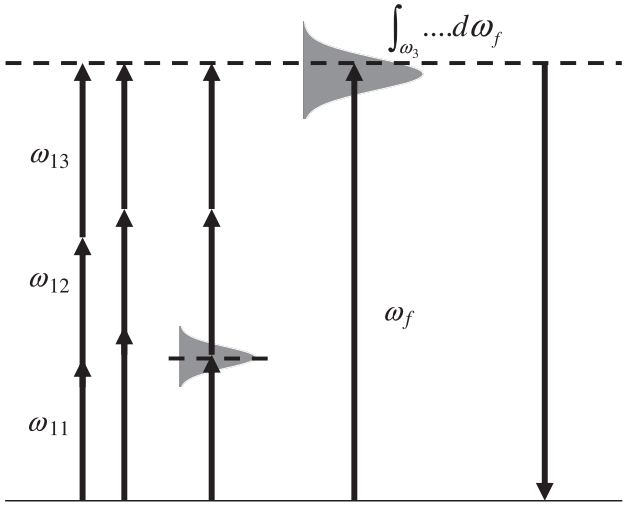

FIG. 1. Coupling scheme.

Fourier transform limited (FTL) field of the laser E_1 in a three versus one photon coupling scheme. The measured quantity is the TH intensity $I_3 \propto E_3 E_3^*$ produced in a second medium (NLM2) by E_1 in the presence of the field E_3 produced in NLM1. The variation of each spectral component $\omega_f = 3\omega$ (monochromatic wave) of the TH field in NLM2 is due to the induced polarization at 3ω: $\frac{\partial E_{3\omega}(z,t)}{\partial z} \propto P(3\omega, z, t)$, z being the propagation axis, with its origin at the entrance of NL2. $P(3\omega, z, t)$ consists of interfering nonlinear and linear terms $P(3\omega, z, t) = P^{\mathrm{NL}}(3\omega, z, t) + P^{\mathrm{L}}(3\omega, z, t) = \chi^{(3)} E_\omega^3(z, t) + \chi^{(1)} E_{3\omega}(z, t)$, where $E_{3\omega}$ can be decomposed as $E_{3\omega} = E_{3\omega}^{(1)} + E_{3\omega}^{(2)}$, $E_{3\omega}^{(i)}$ $(i = 1, 2)$ being the harmonic fields produced in NL1 and NL2, respectively. $|E_{3\omega}^{(1)}|$ is practically constant, while the average $|E_{3\omega}^{(2)}|$ is initially zero and increases with z. The TH field at the exit of the NLM2 becomes:

$$E_{3\omega}(z = L) = E_{3\omega}(z = 0) + A \int_0^L [\chi^{(3)} E_\omega^3(z, t) + \chi^{(1)} E_{3\omega}^{(1)}(z, t) + \chi^{(1)} E_{3\omega}^{(2)}(z, t)] dz$$

with A a constant and L the length of the NLM2. Since $E_{3\omega}(z = 0)$ and $\chi^{(1)} E_{3\omega}^{(1)}(z, t)$ oscillate with the phase of the TH entering NLM2, while $\chi^{(3)} E_\omega^3(z, t)$ and $\chi^{(1)} E_{3\omega}^{(2)}(z, t)$ with three times the phase of the fundamental the measured signal I_3 oscillates as $\cos(\varphi_{3\omega} - 3\varphi_\omega)$. If, instead of harmonic generation, only ionization would occur and be measured in NLM2 [6], the yield would be determined only by the square of the interfering terms $\chi^{(3)} E_\omega^3(z, t)$ and $\chi^{(1)} E_{3\omega}^{(1)}(z, t)$, thus depicting the same type of modulation. It is worth noting that the field $E_{3\omega}(z, t)$ is affected by the induced polarization at ω, contributing, through a parametric process, to the fundamental field strength: $P(\omega, z, t) = P^{\mathrm{NL}}(\omega, z, t) + P^{\mathrm{L}}(\omega, z, t) = \chi^{(3)} E_{3\omega} E_\omega^*(z, t) E_\omega^*(z, t) + \chi^{(1)} E_\omega(z, t)$, where $E_\omega = E_\omega^{(1)} + E_\omega^{(2)} \simeq E_\omega^{(1)}$. This system of coupled equations governs the energy exchange between the fundamental and the TH fields, ensuring energy conservation.

For the polychromatic TH field the measured TH intensity becomes proportional to:

$$C + \int_{\omega_f} B(\omega_f) \cos\left[\varphi_3(\omega_f) - \sum_{j=1}^{3} \varphi_1(\omega_{1j})\right] d\omega_f, \quad (1)$$

where B is a function of ω_f, C is a constant, and $\varphi_1(\omega_{1j})$ and $\varphi_3(\omega_f)$ are the spectral phase distributions of the fundamental and the TH fields, respectively. $j = 1, 2, 3$ refer to the three photons of three modes of the fundamental that couple the same initial and upper state as the TH photon does, i.e., $\sum_{j=1}^{3} \omega_{1j} = \omega_f$. For a FTL fundamental, the relative phase for each given frequency ω_f within the bandwidth (i.e., for each upper state $|f\rangle$) is given by $\varphi_3(\omega_f) - \sum_{j=1}^{3} \varphi_1(\omega_{1j}) = \varphi_3(\omega_f) - \omega_f \tau$ with τ being the variable delay of the cross correlation. For each frequency ω_f the TH emission probability oscillates as $\cos[\varphi_3(\omega_f) - \omega_f \tau]$. The total emitted TH power results then from the incoherent sum of the oscillatory contributions of all the frequency components ω_f (integration over all different frequencies ω_f) within the given bandwidth. Thus the measured cross-correlation signal yields the spectral phase difference $[\varphi_3(\omega_f) - \omega_f \tau]$ distribution and hence the relative spectral phase distribution of the TH pulse. For the reconstruction of the TH pulse, its field amplitude distribution has to be determined separately.

The cross correlation has been performed utilizing the dispersionless transmission grating based interferometer [13] shown in Fig. 2. The TH to be characterized is produced in the first Xe gas cell (NLM1). The geometry is such as to give rise to a double passage of the beam through a fused silica grating (G): in the first passage the different spectral components are split into the various diffraction orders while in the second, after retroreflection on the spherical mirrors SM1 and SM2, are recombined. The fundamental beam (IR) is selected in the zero order diffraction branch by insertion of a glass plate (F) that absorbs the TH. The TH is spatially separated from the other wavelengths in the first order diffraction branch. This allows its selection by appropriately positioning the suitably sized mirror SM2. In the second passage through the grating, the fraction of the selected fundamental that is zero order diffracted and the fraction of the selected TH that is first order diffracted recombine and copropagate towards

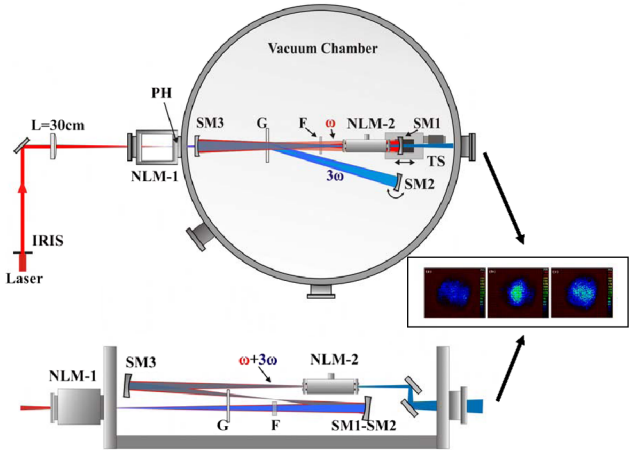

FIG. 2 (color online). Top view (upper panel) and a side view (lower panel) of the experimental setup. L: $f = 30$ cm lens. NLM1, NLM2: nonlinear medium (gas cell) 1 and 2. PH: pinhole. G: 600 lines/mm grating SM1, SM2, SM3: spherical mirrors. F: 3 mm-thick BK7 glass filter. TS: translation stage. Inset: spatial interference fringes of the TH recorded for three different delays (from left to right $\lambda/2$, 0, $\lambda/4$) between the two overlapping pulses. The images are captured by a CCD camera at the far field.

the spherical mirror SM3. In order for these outgoing beams not to counterpropagate with the incoming beams, the mirrors SM1 and SM2 are slightly tilted, thus elevating the retroreflected beams as to impinge the grating 1 cm above the incoming ones. The two copropagating beams are finally focused by the mirror SM3 into the second nonlinear medium (NLM2), a second gas cell filled with Xe as well. The TH field produced in the NLM1 is cross correlated with the IR field by translating the mirror SM1 and measuring the TH yield of the NLM2. It is worth noting that the IR pulse is precompensated in the compressor of the laser system for the chirp introduced by the optical elements (lens, entrance window of the first cell). This ensures an almost FTL 56 fs long laser pulse in the second THG cell, where the cross-correlation measurement takes place. The laser pulse is further near FTL in the first THG cell, as the exit of the first cell is windowless and the 1 mm thick LiF entrance window of the second cell does not introduce significant dispersion. Spatiotemporal overlap and copropagation of the two fields is optimized by ensuring that in the far field the final TH field exhibits a single spatial fringe pattern. Such single spatial fringe patterns are shown in the inset of Fig. 2 for three different delays. The entire grating interferometer is placed in a vacuum chamber kept at 10^{-4} mbar. This prevents (i) additional TH generation at the foci of the mirrors SM1 and SM2 formed between the mirrors and the grating and (ii) dispersion of the TH through propagation in air. The TH signal at the exit of the second gas cell is spatially integrated and recorded with a linear photodiode. Cross-correlation traces have been measured for an unstretched TH field and a chirped one. Stretching of the TH is achieved by propagation through 6 mm of fused silica, which also introduced chirp in the fundamental. In this case, in order to fulfill the requirement for a FTL fundamental in the second interaction cell, stronger precompensation had to be applied. As criterion for the presence of a FTL fundamental has been used the maximization of the THG in the second cell adjusting the precompensation of the fundamental (THG in the first cell was switched off pumping out the Xe gas). At constant pulse energy, the THG maximizes when the driving laser intensity is maximized, i.e., when the laser pulse duration is minimized (FTL pulse).

The measured traces for the unstretched and the chirped TH are shown in Figs. 3(a) and 3(b), respectively. The zero of the cross-correlation signal axis is arbitrarily chosen at the middle of the trace. The larger width of the trace in the Fig. 3(b) indicates already the chirp of the pulse. In expanded (inserted portions), symmetric with respect to the zero delay, areas of the trace of Fig. 3(b) from -60 to -50 fs and from 50 to 60 fs, the chirp of the pulse is clearly visualized. The number of cycles in the interval -60 to -50 fs is $\sim$10.5 while in the interval 50 to 60 fs it is increased to $\sim$12. At negative delays the carrier frequency is markedly lower. Figure 3(c) partially shows results from

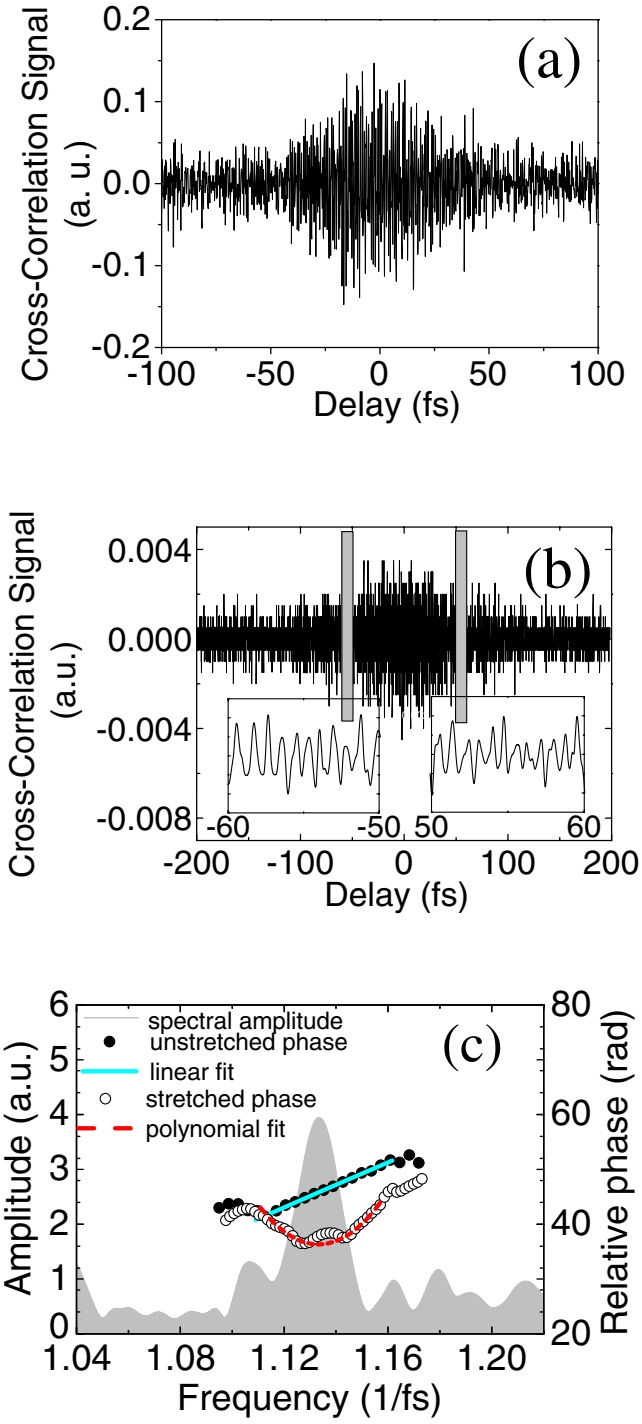

FIG. 3 (color online). IR-TH cross-correlation trace of: an unstretched pulse (a), of a chirped pulse. The inset portions are expanded areas of the trace from -60 to -50 fs and 50 to 60 fs depicting the chirp of the stretched pulse (b), Fourier transform spectra (c). In (c): phase distribution of the unstretcehd pulse (filled circles), phase distribution of the chirped pulse (open circles), TH field amplitude determined from the FT of the TH field autocorrelation (filled area), linear fit (straight line), polynomial fit (dashed line). As zero of the y axis in the cross-correlation traces is taken the THG signal produced at long delays for which the pulses are not overlapping and thus no interference occurs. (a.u. stands for arbitrary units.)

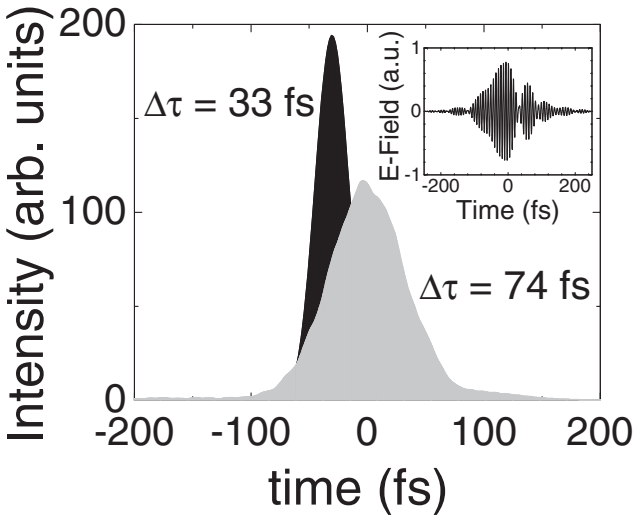

FIG. 4. Reconstructed unstretched (filled black area) and chirped (filled gray area) pulse. The inset trace is the field distribution of the stretched pulse reconstructed using the row measured phases (prior polynomial fit).

the Fourier transform (FT) of the traces. The filled dots represent the spectral phase distribution of the unstretched TH field, showing fairly linear frequency dependence, while for the stretched TH field an almost quadratic distribution is retrieved (open circles).

For the determination of the TH field amplitude spectral distribution a first order autocorrelation (AC) has been performed. According to the Wiener-Khinchin Theorem the FT of it is equivalent to the power spectrum. For the AC, the interference of the recombined zeroth-first-order diffracted (the absorbing glass plate has been removed) and the first-zeroth-order diffracted TH fields is recorded by properly positioning the mirror SM3. Translating the mirror SM1 the field AC trace has been measured. The TH field amplitude distribution is determined from the FT of the trace and is shown in Fig. 3(c) (filled area).

From the retrieved spectral phase and amplitude distributions the third harmonic pulses have been reconstructed and are shown in Fig. 4. The resulted duration of the unstretched pulse (Fig. 4 filled black area) is found to be 33 ± 5 fs. This result is in a very good agreement with the expected duration from the valid for the THG lowest order perturbation theory, $\tau_{\mathrm{THG}} = \frac{\tau_L}{\sqrt{3}} = 32$ fs, τ_L being the 56 fs duration of the IR laser pulse. The reconstructed chirped TH field is shown in the inserted portion of Fig. 4. The modulated envelope is due to the noisy phase distribution. In order to reconstruct a smooth pulse a polynomial is fitted in the smooth parts of the phase distribution. The fit is shown in Fig. 3(c) with the dashed line. Using the phases of this polynomial distribution the pulse of Fig. 4 (filled gray area) has been reconstructed. Its duration is (74 ± 15) fs. Taking into account all dispersive elements of the apparatus and the chirp they introduce, the expected duration of the chirped TH (after propagation in the fused silica plate) has been estimated to 85 fs. This measured value is also in fairly good agreement with the estimated one. It is worth noting that the method applied does not account for the spatial phase distribution of the TH, like all other approaches measuring "average" phases of harmonics [14].

As long as the deviation from the perturbative regime does not affect the retrieved phases, the demonstrated full reconstruction of the gas TH pulses can be extended to higher harmonics, including superpositions of harmonics and thus attosecond pulses. This is feasible by replacing the fused silica grating through a free standing transmission grating [13], using a gas with high ionization potential in NLM2, measuring ionization, and using high precision stabilized interferometer arrangements [15].

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[1] M. Shapiro *et al.*, Chem. Phys. Lett. **149**, 451 (1988).
[2] Ce Chen *et al.*, Phys. Rev. Lett. **64**, 507 (1990); A. D. Bandrauk *et al.*, Chem. Phys. Lett. **200**, 399 (1992); T. Nakajima *et al.*, Phys. Rev. Lett. **70**, 1081 (1993); Phys. Rev. A **50**, 595 (1994); E. Dupont *et al.*, Phys. Rev. Lett. **74**, 3596 (1995); B. Sheehy *et al.*, Phys. Rev. Lett. **74**, 4799 (1995); Langchi Zhu *et al.*, Science **270**, 77 (1995); Phys. Rev. Lett. **79**, 4108 (1997).
[3] S. Cavalieri *et al.*, Phys. Rev. A **55**, 2941 (1997).
[4] D. Xenakis *et al.*, Optics Communications **152**, 83 (1998); N. E. Karapanagioti *et al.*, J. Phys. B **29**, 3599 (1996).
[5] E. Charron *et al.*, Phys. Rev. Lett. **71**, 692 (1993); E. Cormier *et al.*, J. Phys. B **30**, 3095 (1997); U. Andiel *et al.*, Europhys. Lett. **47**, 42 (1999).
[6] E. Hertz *et al.*, Phys. Rev. A **64**, 051801 (2001).
[7] M. Drescher *et al.*, Science **291**, 1923 (2001).
[8] P. M. Paul *et al.*, Science **292**, 1689 (2001); Y. Mairesse *et al.*, Science **302**, 1540 (2003).
[9] P. Tzallas *et al.*, Nature (London) **426**, 267 (2003).
[10] Y. Kobayashi *et al.*, Opt. Lett. **23**, 64 (1998); T. Sekikawa *et al.*, Phys. Rev. Lett. **88**, 193902 (2002); Y. Nabekawa *et al.*, Phys. Rev. Lett. **94**, 043001 (2005).
[11] H. Niikura *et al.*, Nature (London) **417**, 917 (2002); **421**, 826 (2003).
[12] M. Makris *et al.*, Phys. Rev. A **66**, 053414 (2002).
[13] E. Goulielmakis *et al.*, Appl. Phys. B **74**, 197 (2002).
[14] M. B. Gaarde *et al.*, Phys. Rev. Lett. **89**, 213901 (2002); S. Kazamias *et al.*, Phys. Rev. A **69**, 063416 (2004); L. A. A. Nikolopoulos *et al.*, Phys. Rev. Lett. **94**, 113905 (2005).
[15] M. Kovačev *et al.*, Phys. Rev. Lett. **95**, 223903 (2005).