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Probing attosecond pulse trains using "phase-control" techniques

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We propose a method for the measurement of attosecond beating structures that result from the superposition of harmonics of laser radiation. It utilizes the phase control of the excitation probability of atoms through different interfering channels. The channels consist of multiphoton excitation by the fundamental frequency and single-photon excitation by the superposition of the harmonic fields, or alternatively, by each harmonic or frequency interval within the bandwidth of each harmonic. The method is sensitive to the relative phase of the harmonics, including phases introduced by chirp, and thus allows evaluation of the temporal structure of the superimposed harmonic fields. An experimental setup for the implementation of the method based on free-standing transmission grating is suggested as well.

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Among other techniques [1], the coherent superposition of high-order harmonics of a laser fundamental frequency produced in a nonlinear medium has been proposed as a most promising candidate for the generation of attosecond (as) pulse trains or even single attosecond pulses [2–14]. This would lead to the breaking of the record of a few femtosecond-pulse duration established in the last years [15–16]. In a recent work, the first experimental indication for the existence of as pulse trains has been reported [17–19]. The results of this experiment have been only qualitatively interpreted based either on the single atom response or on propagation [17,19], but since in this work the generation and measurement of the pulse trains, as well as the atomic response and propagation effects are entangled [18], a rigorous modeling of the experiment is a formidable task. Thus it is generally accepted that further and less entangled experiments are needed in order to produce quantitatively interpretable results. However, there are several problems inherent to this type of measurement, such as the dispersion introduced by beam splitters and the insufficient harmonic intensity in order to induce a nonlinear effect for a second- or higher-order autocorrelation. Thus recent efforts for the measurement of ultrashort pulses focus on cross correlations between the harmonics and a correlated infrared (IR) field. These methods rely either on the dynamics of the ionization caused by the harmonics in the presence of an IR field [20] or on phase-sensitive interference effects [21,22]. Reference [21] presents the measurement of relative phases of harmonics and a reconstruction of attosecond pulse trains. However, this is done by treating the harmonics as monochromatic waves, since the method is not mode selective, and thus cannot account for chirp within the bandwidth of each harmonic. The reconstructed pulses reflect the real ones only if this chirp is small. In the present work, we propose an alternative method to map out the relative phases *of the modes* of superimposed or individual harmonics of the laser radiation. We further suggest an experimental arrangement appropriate for the implementation of the method based on a double-pass through a freestanding transmission grating, thus overcoming the problem of the dispersion introduced by the beam splitter.

The approach we propose is based on what is known as phase control of excitation processes [23]. Excitation of an atomic system occurs through different coherent pathways, e.g., single-photon excitation through the nth harmonic of the laser field and n-photon excitation through the fundamental (Fig. 1). By varying the relative phase between the fundamental and the harmonic, assuming both as being monochromatic waves, the excitation probability varies as $\cos(\varphi_n - n\varphi_1)$, where φ_n, φ_1 are the initial phase of the harmonic and the fundamental. This variation can be probed through ionization [24] (in case that the final state is in the continuum) or more general through harmonic generation [25,26]. Probing with harmonic generation allows the study of interference below the ionization threshold and thus of lower harmonics as well. Working with low harmonics may be preferable, since they require lower phase shifting resolu-

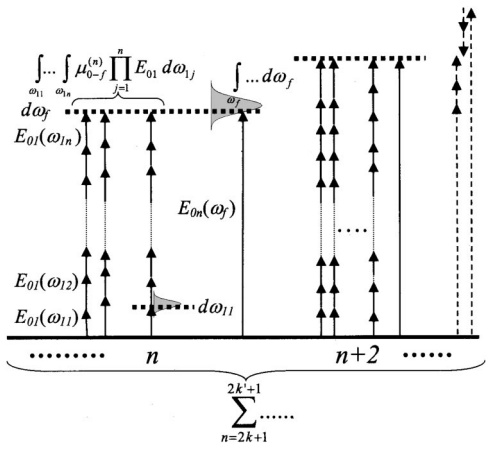

FIG. 1. Interfering channels leading to an excitation probability that depends on the relative phase between the corresponding fundamental and harmonic modes.

tion. Conventionally, phase control experiments aim at controlling the final products of the interaction, but they also probe the relative phase between the fundamental and the harmonics. If now instead of one harmonic, a coherent superposition of harmonics or several individual harmonics is used, the method can be employed to determine the relative harmonic phases and measure temporal profiles of the coherent superposition of the harmonic fields. It will be shown that since the measured quantity depends on the relative phases between each harmonic and corresponding fundamental modes, and since the phase of the fundamental modes are the same for a transform limited pulse, the measured quantity will finally depend on the relative phase between harmonic modes, including phase variations introduced through chirp. Hence it implicitly reveals this relative phase relation. Thus, variations in the measured quantity, as a function of the delay between the fundamental and the harmonic superposition, reflect variations in the electric field of the superposition of the harmonic modes. Attosecond beating eventually present in such a superposition can be evaluated through the proposed method discussed later on.

Let the superposition of the electric fields of the laser fundamental frequencies and that of its harmonics of interest be

$$\vec{E}_{\mathrm{TOT}}=\hat{x}\int_0^{\infty}\Bigg(E_{01}(\omega)e^{-i[\omega t-\varphi_1(\omega)]}+\sum_{n=2k+1}^{2k'+1}E_{0n}(\omega)e^{-i[\omega t-\varphi_n(\omega)]}\Bigg)d\omega. \tag{1}$$

$\hat{x}$ is the polarization unit vector, n denotes each harmonic, k and k' are integer numbers, and $n=1$ stands for the fundamental. E_{01} and E_{0n} are the spectral amplitudes. $\varphi_1(\omega)$ and $\varphi_n(\omega)$ correspond to the phases of the different spectral components and we assume transform limited fundamental pulses, i.e., the same initial phase $\varphi_1(\omega)$ within the spectral bandwidth. Thus the bandwidth and the chirp of each harmonic are accounted for. When this superposition interacts with an atomic system, excitation takes place from a common initial state $|0\rangle$ to a continuum of (virtual) final states $|f\rangle$. Using expressions of lowest-order perturbation theory, the excitation probability, which can be probed through harmonic emission (or ionization), is proportional to the expression

$$\sum_{n=2k+1}^{2k'+1}\int_{\omega_f}\Bigg|\int_{\omega_{11}}\cdots\int_{\omega_{1n}}\mu^{(n)}_{0-f(\omega_{11},\ldots,\omega_{1n})}\times\prod_{j=1}^{n}E_{01}(\omega_{1j})e^{i\varphi_1(\omega_{1j})}d\omega_{1j}+\mu^{(1)}_{0-f}E_{0n}(\omega_f)e^{i\varphi_n(\omega_f)}\Bigg|^2 d\omega_f \tag{2}$$

and thus to

$$C+\sum_{n=2k+1}^{2k'+1}\int_{\omega_f}\Bigg(\int_{\omega_{11}}\cdots\int_{\omega_{1n}}\mu^{(n)}_{0-f(\omega_{11},\ldots,\omega_{1n})}\times\prod_{j=1}^{n}E_{01}(\omega_{1j})d\omega_{1j}\mu^{(1)}_{0-f}E_{0n}(\omega_f)\Bigg)\times\cos\Bigg(\varphi_n(\omega_f)-\sum_{j=1}^{n}\varphi_1(\omega_{1j})\Bigg)d\omega_f, \tag{3}$$

with C a constant. The products $E_{01}(\omega_{11})E_{01}(\omega_{12})\ldots E_{01}(\omega_{1n})$ refer to the n photons of the fundamental that couple the same initial and final state as the harmonic photon does, i.e., $\Sigma_{j=1}^{n}\omega_{1j}=\omega_f$ with $\hbar\omega_f$ the energy of the transition $|0\rangle-|f\rangle$ [27] (Fig. 1). Considering a fundamental pulse Fourier-transform limited, the phase difference for one given final state ω_f is $\varphi_n(\omega_f)-\Sigma_{j=1}^{n}\varphi_1(\omega_{1j})=\varphi_n(\omega_f)-\omega_f\tau$ with τ the variable delay between the fundamental and harmonics. $\mu^{(n)}_{0-f(\omega_{11},\ldots,\omega_{1n})}$ and $\mu^{(1)}_{0-f}$ are the corresponding n-photon and single-photon dipole moments induced by each combination of modes of the fundamental and the harmonic radiation, respectively, for each pair of interfering excitation channels ($|0\rangle+\hbar\omega_{11}+\hbar\omega_{12}+\ldots+\hbar\omega_{1n}\rightarrow|f\rangle$, $|0\rangle+\hbar\omega_f\rightarrow|f\rangle$). Since interference occurs only in channels coupling the same initial and final state, the result is the coherent summation of the transition amplitudes for the single-photon ω_f (through the nth harmonic) and the corresponding n-photon (through the appropriate combination of n modes of the fundamental) excitation channels. The resulting total excitation probability is then the incoherent sum of the contributions of all interfering pairs. For a transform limited fundamental, the oscillations of the probability probe the relative phase between the fundamental and the harmonic modes (and hence between the corresponding modes of the harmonics), and thus the temporal characteristics of their superposition. Indeed, each component is oscillating as $\cos(\varphi_n(\omega_f)-\omega_f\tau)$. Summation over all components yields the temporal behavior of the coherent superposition of all harmonic modes involved. Because of the different amplitude factors in the relations given by Eqs. (1) and (3), the variations of the measured quantity will not reflect the total field as far as amplitudes are concerned, but only its temporal characteristics, i.e., the relative harmonic mode phases. Once the harmonic mode phases are extracted, e.g., through a Fourier transformation, amplitudes may be separately measured through conventional frequency domain spectroscopy. Alternatively, determination of E_{TOT} would be in principle possible by repeating the measurement selecting for each run a small frequency interval of each harmonic and measuring at the same time the field amplitude of this interval. Application of this approach for a few intervals within the bandwidth of one harmonic allows for an evaluation of its chirp. It should be noted that even if part of the interactions are not perturbative, the phase effect is not expected to be different than this resulting from the perturbative expressions used here because of the intuitive picture they provide. Thus, e.g., two-color phase control of harmonic generation is not different than what the lowest-order perturbation theory

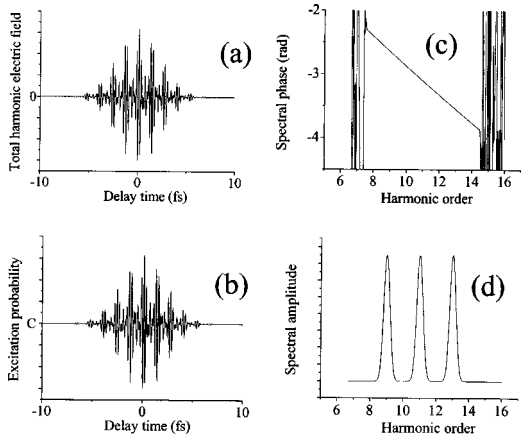

FIG. 2. Comparison of the electric field of a superposition of harmonics and the excitation probability as a function of the delay between the fundamental. Harmonic *E*-field (a); excitation probability as given in Eq. (3) (b); Fourier-transform phase of excitation probability (c); Fourier-transform spectral amplitude of excitation probability (d).

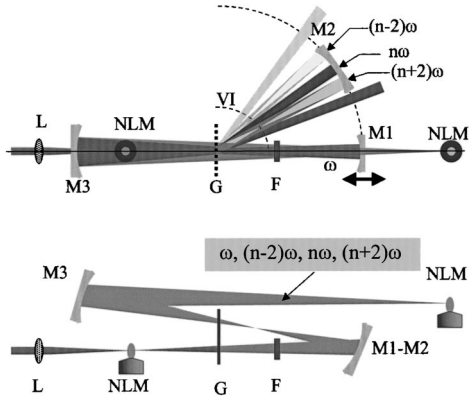

FIG. 3. Schematic of the proposed experimental setup. Upper part: top view. Lower part: side view. L: lens; NLM: nonlinear medium; G: transmission grating; M1, M2, M3: spherical mirrors; F: glass filter; VI virtual image; In this example three harmonics around the *n*th harmonic are selected and become copropagating with the fundamental. The harmonic selection along with the delayed fundamental is focused into a secondary nonlinear medium.

predicts as far as phases are concerned [26]. Considering higher-order perturbation terms, which is equivalent to solving the full problem, would hardly affect the phase relations. Some higher-order terms are discussed below. Moreover, perturbative expressions are straightforward applicable for the study of lower harmonics of harmonics of intermediate order by properly reducing the intensity of the fundamental.

In order to demonstrate the temporal relation between the measured quantity and the total field of the superposition of the 9–13 harmonics of a fundamental laser wavelength of 800 nm, the expression $\int_0^\infty \Sigma_{n=9}^{13} E_{0n}(\omega) e^{-i(\omega t-\varphi_n(\omega))} d\omega$ and the corresponding expression in Eq. (3) after numerical integration are depicted in Figs. 2(a) and 2(b). A Gaussian mode amplitude distribution and phase dependence proportional to $\omega^{0.8}$ for the spectral components (modes) of each harmonic (to account for the chirp within each harmonic) and between the harmonics (to account for the chirp between harmonics) have been used. The choice of the chirp is arbitrary as the method applies for any chirp. For demonstration purposes, the ideal case of $\int_{\omega_{11}} .. \int_{\omega_{1n}} \mu_{0-f(\omega_{11},\ldots,\omega_{1n})}^{(n)} \Pi_{j=1}^{n} E_{01}(\omega_{1j}) d\omega_{1j}$ being equal to unity has been considered, which leads to identical traces in Figs. 2(a) and 2(b). Figures 2(c) and 2(d) show the Fourier Transform of the trace depicted in Fig. 2(b). It reproduces the amplitude and phase distribution of the total harmonic field.

It is understood here that probing can be for the superposition that is avoiding energy dispersion of the interaction products (photons or electrons) in order to allow simultaneous probing of all interference terms. This requires all interfering amplitudes to be of the same order of magnitude. Alternatively, one could also select one harmonic or even frequency intervals within one harmonic either before the interaction with the atomic system or after the interaction by energy analyzing the interaction products and subsequently sum up the contributions from the different harmonics or frequency intervals. In this case, interfering excitation amplitudes can be optimized for each selected frequency interval.

An arrangement for the experimental implementation of the above method, based on a transmission grating is shown in Fig. 3. Similar arrangements have already been studied in connection with dispersionless autocorrelator setups and will be discussed in more detail elsewhere [28]. They have been used for the measurement of femtosecond-duration pulses [29]. Copropagating fundamental and harmonic beams produced in a nonlinear medium are impinging on a freestanding transmission grating (Fig. 3). The zeroth order passes straight through, while the beam diffracted in one of the first orders disperses resulting in well-separated harmonic beams. This way, a frequency range corresponding to a set of harmonics can readily be selected using an iris or appropriate obstacles. Two spherical mirrors (M1 and M2) are used to redirect the selected spectral range as well as the zeroth order back through the grating. These two mirrors are positioned so that they image the grating into itself. Moreover, they are slightly tilted so that the back reflected beams are somewhat elevated in the vertical plane and thus spatially separated from the incoming beam. The virtual image of the interaction region is different for each harmonic, but it is located on a concentric circle between the grating and the mirrors. Upon exiting the grating, all the -1st orders of the selected harmonics become copropagating with the back reflected zeroth order of the fundamental. Finally, a third mirror further directs and focuses the harmonic mixture and the fundamental to a second nonlinear medium. This last mirror relays the virtual image of the interaction region in the primary nonlinear medium into the secondary nonlinear medium. A filter positioned in the pathway of the zeroth order eliminates all the harmonic radiation, while it attenuates to a desirable degree the fundamental. An appropriate micropositioning device, on which mirror M1 is mounted, allows for temporal delay variation between the fundamental and the harmonics. Subsequently, harmonic generation (or ionization) from the interaction of the combined beams with the secondary nonlinear medium is monitored as a function of the delay of the fundamental.

A simple inspection of the geometrical arrangement immediately reveals that the setup is strictly dispersionless. All harmonics and the fundamental travel in exactly the same optical path. Furthermore, the harmonic source in the primary nonlinear medium is imaged relayed to the secondary one after a spectral selection has taken place. Consequently, both requirements of low dispersion and focusing are satisfied simultaneously in this layout. This has been verified using a three-dimensional (3D) ray-tracing code [28] in which the effect of the mirror tilt has been taken into account. The main advantage of the "*grating beam splitter*" is its flat response characteristics over the entire VUV-XUV spectral region and its high efficiency (~20% in the zeroth order and ~10% in the first order) [30]. In connection with the proposed method of detecting as trains, the suitability of this setup lies in the fact that in its part where the different wavelengths are separated, knife edges or attenuators may be used. Thus, desired spectral regions and intensity ratios may be selected.

In the preceding discussion, additional interfering channels such as $|0\rangle+\hbar\omega_{n-n'}+n'\hbar\omega_1\rightarrow|f\rangle$ or $|0\rangle+\hbar\omega_{n+n'}-n'\hbar\omega_1\rightarrow|f\rangle$, where n' is an even integer, depicted for $n'=2$ by dashed lines in Fig. 1, have not been taken into account in order to keep the picture as clear as possible. These channels, if not negligible, are not mode selective, since each of them includes photons from both the fundamental and one harmonic. Hence their interference does not give any information about chirp within the bandwidth of each harmonic. Such channels are in general expected (or can be made) to have negligible contribution because they involve nonlinear interactions, in contrast to the linear one-harmonic-photon channels. If their contribution would be not negligible, they would add low, even harmonic frequencies $n'\omega$, $2n'\omega$ to the measured interference pattern, and thus their contribution could be checked and eventually minimized. Furthermore, for the described approach that selects one harmonic in the region between the mirror and the grating, those channels do not exist.

Finally, it should be pointed out that one of the given interpretations [22] of the observed as beating in Ref. [17] has been based to the above-mentioned principle, which in the two-pulse experiment of Ref. [17], occurs due to propagation effects. In this interpretation, depending on the dispersion conditions, propagation in the medium may bring one of the laser-pulse maximum in partial overlap with the harmonics produced by the other maximum and produce new harmonics in their presence. This is equivalent with what has been described above. The delay between the one fundamental maximum and the harmonics produced by the other fundamental maximum in Ref. [17] was varied through the delay between the two laser pulses.

In conclusion, an alternative method based on phase control of atomic excitation and an experimental apparatus has been suggested that allows the mapping of relative phases of harmonic modes. The method is mode selective and thus allows the evaluation of temporal characteristics of harmonics, harmonic superposition, and the chirp of harmonics. The method is suitable for the measurement of as pulse trains or individual attosecond pulses, and hence for distinguishing a single pulse from a train of pulses.

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