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High-Harmonics Chirp and Optimization of Attosecond Pulse Trains

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Abstract—Plateau high harmonics are shown to be chirped. The experimental values of the chirp, in good agreement with purely classical kinematics or strong field approximation quantum theory are positive and inversely proportional to intensity. The various steps for the optimization of the attosecond pulse train are discussed.

1. INTRODUCTION

Odd harmonics of an infrared fundamental light have been the object of constant interest ever since their first observation in the late 1980s [1], first as a fascinating nonpertubative process resulting from the interaction of an atom and an intense laser pulse and second as a coherent, intense source of radiation, with a spectrum extending to the XUV region and whose remarkable properties have led to a large number of applications. The study of the harmonic spectrum, long the object of most of the experimental effort, revealed this nonperturbative character through the observation of a long plateau, a spectral region over which the successive orders, at first glance resulting from successive perturbation orders, instead of decreasing rapidly remain, on the contrary, constant. The plateau ends at the high-energy side, in a cutoff where the spectrum turns off over a few orders. At the beginning of the 1990s [2], plateau high harmonics were proposed as an ideal source for the Fourier synthesis of attosecond (as) pulses due to their periodic spectrum, which covers a wide bandwidth. In the past few years, experiments have indeed confirmed that subfemtosecond pulses could be generated from high harmonics. In one approach [3], a few-cycle pump pulse is used and two or three cutoff harmonics are spectrally selected. This method was shown to produce a single pulse of subfemtosecond duration, which has been further reduced to 250 as by the last developments of carrier-to-envelope phase control [4]. In the second approach [5], a larger group of plateau harmonics are superposed to generate a train of attosecond pulses, as was proposed in [2], with a period equal to half the fundamental cycle. In this method, longer (usually 40-fs) pump pulses are used and the envelope of the harmonic attosecond pulse train is typically of the same order. The duration of the attosecond bursts occurring twice per optical cycle of the infrared field depend on the phases of the spectral components and reach the Fourier limit only in the case of a linear spectral phase. In this paper, we shall be concerned with the synchronism of the harmonic spectral components or, expressed otherwise, with the linearity of the spectral phase. It will be shown first that plateau harmonics are not synchronous but are rather emitted at different instants depending on their order. Theory and experiments agree that this dependence is quasi-linear, which is equivalent to saying that the spectral phase has a quadratic dependence on frequency. The attosecond chirp has consequences for the duration of the harmonics bursts and, particularly, implies an optimum total bandwidth. The optimization of the attosecond pulse train additionally requires the selection of the so-called short quantum path in the single-atom response. The concept of "short" and "long" paths (or trajectories) derives from the three-step model of harmonic generation [6], which predicts that two classes of quantum paths dominate the dipole amplitude of a given harmonic. The necessity of filtering out one of them in order to produce a regular attosecond pulse train has long been known [7]. Recent observations indeed confirm this prediction. Intensity is also an important parameter controlling the chirp and, hence, the duration of the pulses. Finally, the linearity of the group delay

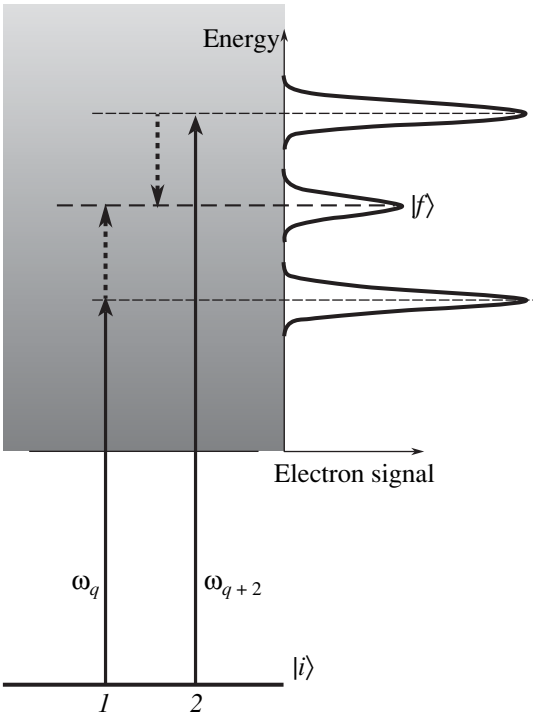

Fig. 1. RABITT principle. Energy-level diagram.

suggests that the chirped pulses can be recompressed by propagation in a suitably dispersive medium. The content of the paper is as follows: we first briefly recall the principle of the harmonic characterization in the frequency domain (RABITT) [5, 8] and summarize the phase measurements, in particular, that of the quadratic component. We then discuss the optimization of the bandwidth and the control of the short quantum path. We finally examine the possibility of generating shorter attosecond pulses by using a longer fundamental wavelength, e.g., in the mid-infrared region. The discussion hereinafter is based on previously published experimental results [5, 9, 10].

2. RABITT

Several techniques have been used to determine the attosecond pulse durations. Some work directly in the time domain [11], others, in the frequency domain by measuring both the spectral amplitudes and phases of each harmonic. It is the latter that is described now. In the case of plateau harmonics generated by a relatively long pulse of a few tens of femtoseconds, the phases of the harmonics are determined pairwise by the reconstruction of attosesecond beating by two-photon transitions (RABITT) method [12, 8] based on two-color, two-photon photoionization of a target gas. The photoelectron energy spectra, under these conditions, are comprised of narrow peaks, which are, in the case of long (typically, 40-fs) pump pulses, separated by 2ω and shifted by ω from the single-photon ionization comb due to the harmonics alone (Fig. 1). The setup (Fig. 2), which includes an attosecond optical delay line and a time-of-flight electron spectrometer, is described in detail elsewhere [5, 9]. The phase information is extracted from the modulation of these sideband peaks as a function of the optical delay, provided that one has knowledge of some atomic parameters [8, 12]. From Fig. 1 it is clear that there are two quantum paths leading from the initial state $|i\rangle$ to the final one $|f\rangle$. Second-order perturbation theory shows that the corresponding probability is proportional to an interference term that can be written as

$$\cos(2\omega\Delta t - (\phi_q - \phi_{q+2}) - (\theta_q - \theta_{q+2})), \qquad (1)$$

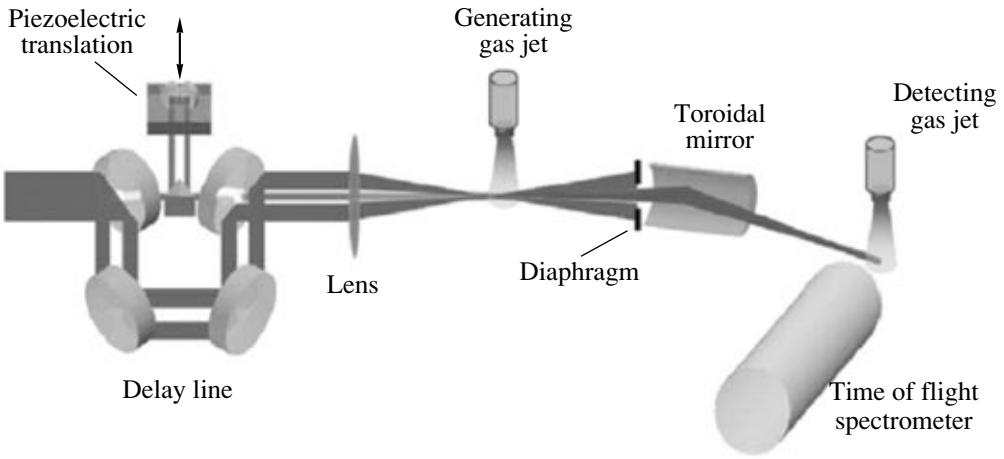

Fig. 2. RABITT setup.

where ω is the fundamental frequency, Δt is the delay between the pump pulse and the harmonic pulse train, $\phi_q - \phi_{q+2}$ is the phase difference to be measured, and $\theta_q - \theta_{q+2}$ is a calculated atomic phase difference. This atomic phase is actually a consequence of the fact that the second-order matrix element is complex, as can be shown in the framework of perturbation theory for any two-photon ATI process. The experiment amounts to recording function (1) by varying Δt and retrieving $\phi_q - \theta_{q+2}$ from the fitted cosine function. Knowledge of the phase differences for all the pairs of harmonics within the bandwidth used, added to that of the corresponding harmonic amplitudes obtained from the photoelectron energy spectra, is sufficient for a reconstruction of the temporal profile of the train [5, 8]. RABITT, like SPIDER (the standard method for complete characterization of femtosecond pulses), works in the energy domain. RABITT is not a single-shot technique; rather, it records a signal averaged over (typically) a thousand laser shots. The reconstructed time profile is therefore an average one and assumes that the harmonic phases remain reasonably constant over the measurement. Moreover, RABITT yields information only about the harmonics whose energy is above the target-gas ionization threshold or just below it. An early measurement of the harmonics 11 to 19 generated in argon is shown in Fig. 4. The time profile of the pulse train resulting from the superposition of harmonics 11–19 is represented in Fig. 4: each burst is about 250 attoseconds long. The corresponding bandwidth of 12 eV implies that the shortest Gaussian pulse that could be supported is about 150 as ($\Delta t \Delta \nu = 0.44$). The profile in Fig. 4 is significantly broader than the Fourier limit, reflecting the effect of the quadratic phase.

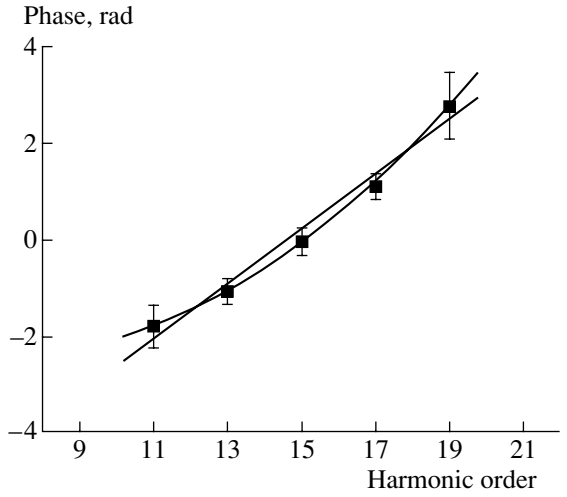

Fig. 3. Harmonics phases vs. order in argon. The quadratic fit is clearly superior to the linear one.

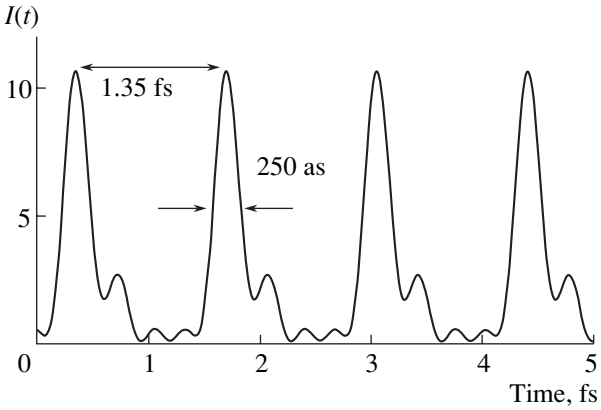

Fig. 4. Attosecond pulse train reconstructed from the measurement of Fig. 3.

3. FIRST EVIDENCE AND QUASI-CLASSICAL INTERPRETATION OF THE CHIRP

Though it is approximately linear, the spectral phase indeed shows a quadratic component, which was first noticed and analyzed in [12]. The interpretation is straightforward in the framework of the three-step recollision model of high-harmonic generation. Harmonic generation in the strong-field regime can be described as a recollision process between the electron released in the continuum and accelerated back to its parent core by the laser field after gaining kinetic energy. This kinetic energy is then transformed into photon energy when the electron wavepacket recombines with the core [6]. This process, occurring twice per optical cycle, gives rise to a sequence of flashes of (XUV) light whose spectrum is the characteristic harmonic spectrum. The usual theoretical approach used to describe high harmonics is the quasi-classical strong-field approximation [13]. This is a quantum-mechanical theory that determines the amplitude A_q of a given Fourier component of the atomic dipole as the sum of complex numbers whose phases depend on the corresponding complex trajectories of the electron wavepacket under the combined action of the electromagnetic field and the Coulomb force:

$$A_q = \sum_{\text{all trajectories}} a_q^{(i)} e^{i\Phi(i)}. \qquad (2)$$

In Eq. (5), the index i labels a given trajectory. Therefore, just from the form of Eq. (1), groups of trajectories for which the phase is *stationary* contribute most. The quasi-classical theory shows in fact that the phases are proportional to the quasi-classical action [14], and, hence, the *classical* trajectories, for which the action is stationary according to Fermat's least-action principle, dominate the sum. A simple argument can be based directly on the one-dimensional classical trajectories of

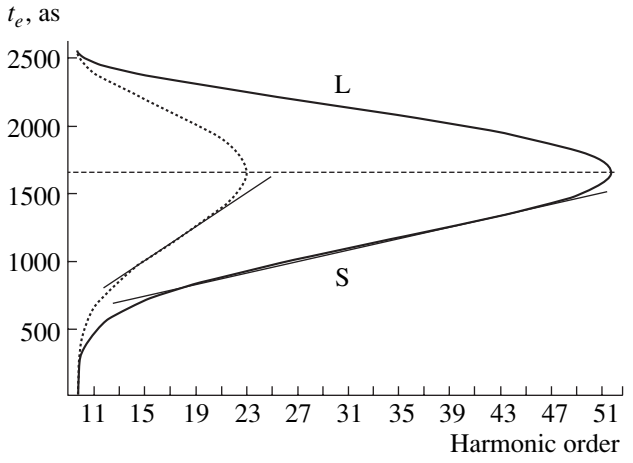

Fig. 5. Recombination time versus energy from classical kinematics.

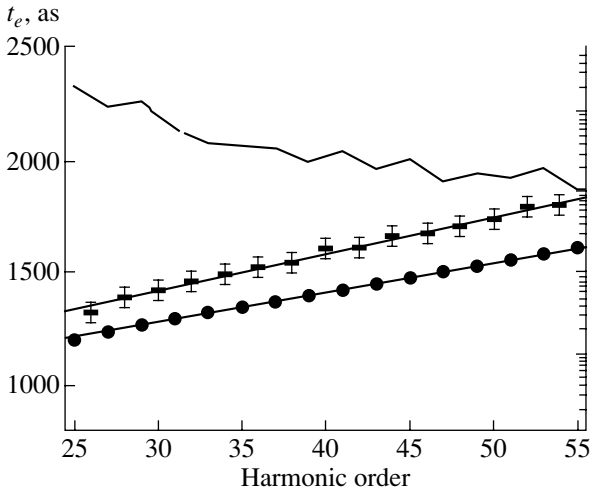

Fig. 6. Accurate measurements of the attosecond chirp in neon.

a free electron in the electromagnetic field. It can be shown that two classical trajectories correspond to any given return energy (or every harmonic order q) up to the cutoff energy: a short (S) one, for which the duration of the trajectory is less than half a cycle, and a long (L) one, whose duration is between a half and a full cycle. Equation (5) can therefore be rewritten approximately as:

$$A_q \approx a_s e^{i\Phi_S} + a_L e^{i\Phi_L}, \tag{3}$$

where Φ_S and Φ_L are given by the quasi-classical action along the short or long trajectories, respectively. Let us assume for the moment that only one trajectory is selected. (This point is actually crucial, and we shall return to it later on). The recollision (or emission) time t_e in units of $2\pi/\omega$ is obtained by solving the Newtonian equation of motion. The initial and recollision times are found to be linked by

$$-\sin x + \sin x_i + \cos x_i (x - x_i) = 0, \tag{4}$$

with $x = \omega(t_i + \tau)$ and $x_i = \omega t_i$, $t_e = t_i + \tau$, which can be solved numerically for x, given x_i between $\pi/2$ and π. Initial phases between 1.57 and 189 correspond to the "long" trajectories, while those with phases between 1.89 and π correspond to the "short" ones. We are interested, from an attosecond perspective, in the dependence of the recollision time on frequency (or harmonic order), i.e., on the harmonic photon energy, which is directly related to the kinetic energy of the recolliding electron. The classical kinetics directly provide the recombination time as a function of energy, as shown in Fig. 5. The plot of Fig. 5 can be done numerically as a parametric plot, which amounts to eliminating the initial phase x_i between the two equations energy $= f(x_i)$ and $x = f(x_i)$. This can be carried out analytically as well, as was shown by Kazamias and Balcou [12], who derived an analytical expression for the chirp. Applied to the case of harmonics 11–19 of Fig. 4, one obtains a total chirp of 230 as/12.4 eV, which can account for the observed 250-as duration of the bursts in Fig. 4. Note in Fig. 5 that the classical mechanics predicts that the chirp has the opposite sign for the long trajectory branch and that its absolute value decreases with intensity.

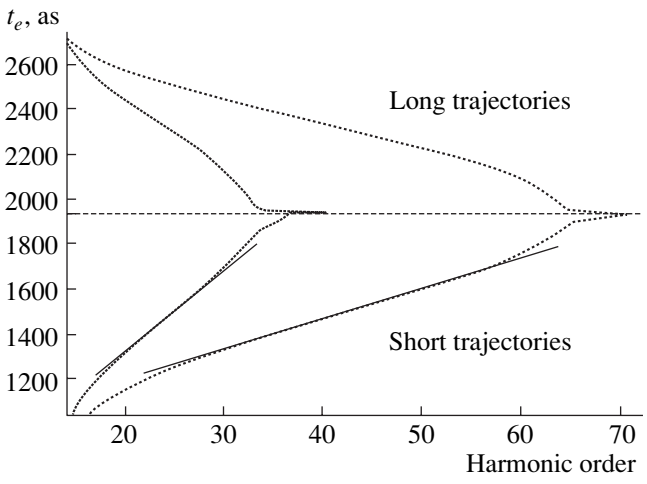

Fig. 7. Quantum calculation of the emission time versus harmonic order, to be compared with Fig. 5 (from [9]).

4. ACCURATE MEASUREMENTS AND QUANTUM THEORY

More accurate measurements in argon (over orders 11–25) (Fig. 6) and neon (over orders 25–55) confirmed the quadratic trend and yielded accurate values of the time difference between consecutive harmonics of 106 ± 8 as for argon at 1.2×10^{14} W cm^{-2} and 33 ± 3 as in neon at 3.8×10^{14} W cm^{-2}, in good agreement with the theoretical values deduced from the quantum calculation [8]. The value of 106 as per order would yield a minimum duration of 424 as for four harmonics if they were of equal amplitude. However, their amplitudes decrease rapidly after $q = 17$, and only three orders significantly contribute to the train. In neon, about three times more intensity can be used before saturation; the time delay per order is found to be roughly three times smaller. The pulses obtained by selecting harmonics in the plateau between $q = 25$ and $q = 69$ (i.e., 23 orders) could be about five times shorter than in argon, just from the bandwidth. However, the chirp contributes to a large broadening and the effective pulse duration amounts to three times the Fourier limit or 150 as in this case.

The quantum calculation uses the saddle-point method [9] and yields emission times versus photon energy (or harmonic order) in excellent agreement with the purely classical calculation above (Fig. 7) in the plateau region. Moreover, it allows one to examine the classically forbidden cutoff region, in which the emission times from both the long and short trajectory branches become equal and independent of the harmonic order. The theory also predicts that the absolute value of the chirp is inversely proportional to the ponderomotive energy, i.e., proportional to ω^2/I. This has interesting consequences, which will be somewhat developed at the end of this paper. Experimentally, the $1/I$ scaling of the chirp was well observed over a large range of intensities using xenon, argon, and neon generating gases to overcome the saturation limit, as shown in [9]. The quadratic phase is unavoidable. It is rooted in the high-harmonic generation process: to produce more energetic photons at recombination, the electron just needs more time to acquire more kinetic energy along the short trajectories. All that can be done is to apply the highest possible intensity compatible with saturation. In this respect, helium is better than neon, which is better than argon. Moreover, a recompression device can be exploited, as will be discussed below.

5. OPTIMAL BANDWIDTH

One obvious consequence of the chirp is that the pulse width does not decrease monotonously when the bandwidth increases, since the total delay between the extreme orders increases with bandwidth. It follows that the duration goes through a minimum corresponding to an optimal bandwidth (Fig. 8). For example, in

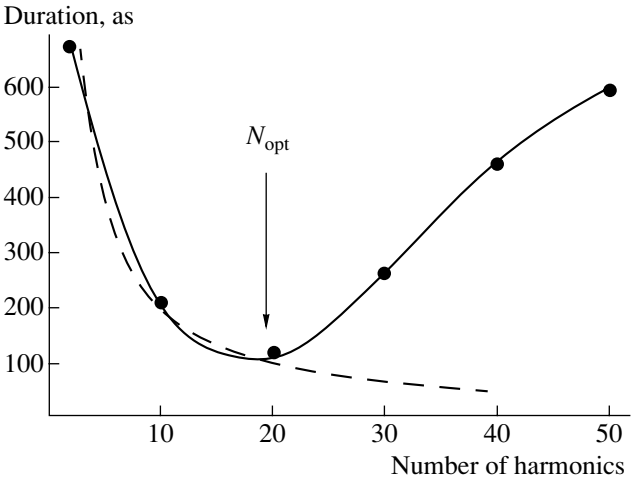

Fig. 8. Optimum bandwidth in the case of neon.

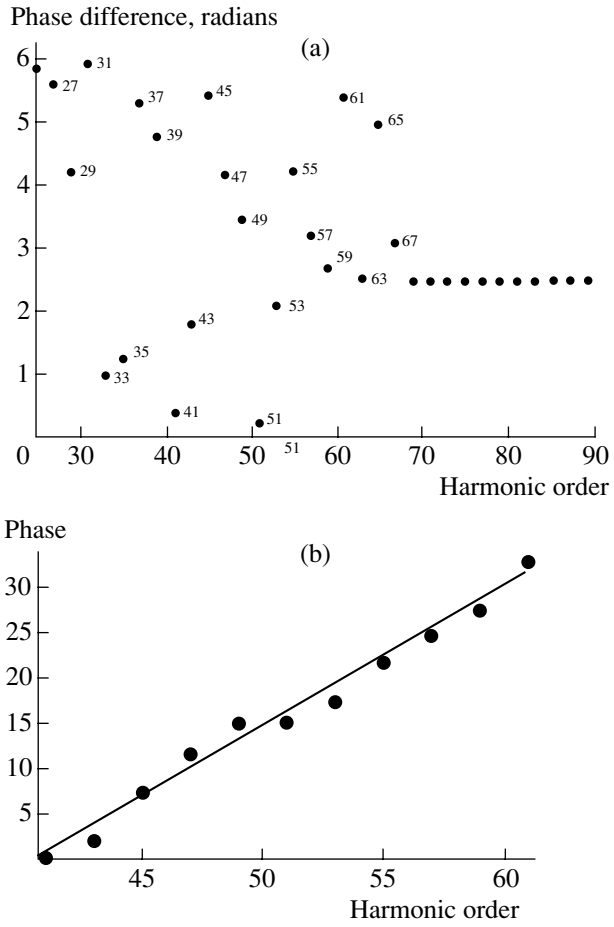

Fig. 9. (a) Harmonic phase differences in neon after Fig. 1 in [7]. The labels are the harmonic orders. (b) Phases vs. order concatenated from the differences of Fig. 9a.

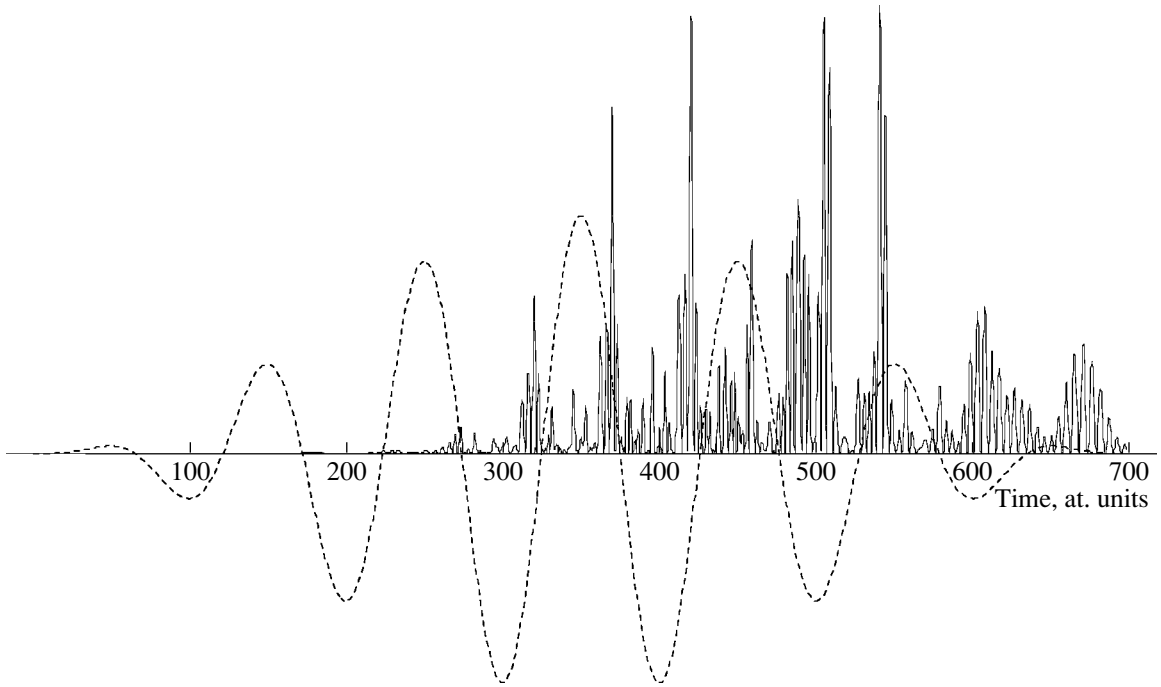

Fig. 10. TDSE square of the dipole acceleration (solid line) and pump field (dotted line) versus time. See text for details.

neon, the pulses obtained by selecting harmonics in the plateau between $q = 25$ and $q = 69$ (i.e., 23 orders) could be about five times shorter than in argon, just judging from the bandwidth. However, the chirp contributes to a large broadening and the effective pulse duration amounts to three times the Fourier limit, or 150 as in this case. Limiting the total bandwidth to only 11 harmonics yields a minimum pulse duration close to the Fourier limit of 130 as. The filtering of such a range of wavelengths is, in practice, not an easy task and is still an unsolved problem. The above statement must therefore be taken as a theoretical prediction.

6. SELECTION OF THE SHORT QUANTUM PATH

The theory, as summarized by Eq. (3), stresses that the amplitude of a given harmonic results from the interference of essentially two quantum paths. Moreover, the calculation clearly indicates that the emission times are different for a given photon energy depending on which quantum path is followed (Figs. 5 and 7). Actually, this has been known since the seminal paper of Antoine *et al.* [7]. Incidentally, one more comment about this paper may be appropriate: one of the results included there assesses the phase differences for harmonics (q = 21–61) in neon to be fairly random. This may appear to contradict the results of Figs. 5 and 7, which show a recombination time (or derivative of the phase with respect to frequency) that is almost exactly linear for a given branch. The contradiction is only apparent: if one plots the phase itself instead (obtained by concatenation from the differences), one gets an almost linear dependence (with, naturally, the small quadratic terms corresponding to the linear scaling of the emission time (Fig. 9)). The scattered aspect of the figure in [7] results from the expanded scale. Another way to more completely realize the effect of interference of all the quantum paths is to perform a numerical integration of the Schrödinger equation on a grid and to calculate the dipole acceleration as a function of time. An example of such a calculation using a code described elsewhere [15] is shown in Fig. 10. This result is rather far from the regular series of attosecond peaks that occurs twice per pump cycle. In this calculation, the target atom is argon and the 800-nm pump pulse comprises seven cycles with a sine square profile. For the first cycles, a growing harmonic burst appears close to the zero-crossings of the pump field, but it is only at the sixth crossing that the amplitude becomes noticeable. The three successive bursts resemble the naive image with a width of about 270 as, but a second series of peaks already appear around the peaks of the pump field, which is probably due the long trajectory. This is another hint that some clean-up is necessary in order to obtain an optimum pulse train. In [7] it was predicted that propagation (phase matching) would do the job. In [10] this was confirmed by comparing the quality of the mode locking as measured by RABITT to the predictions of TDSE on the one hand

and to that of the quasi-classical theory, in which one can filter out the contribution of one trajectory, on the other hand (see Fig. 1b in [10]). It thus appears that the experiment, at least using a setup like the one described in [5, 9, 10], manages to favor one quantum path (the short trajectory) over the other one. How this is actually done is not entirely clarified. One very likely reason is that the two trajectories are associated with different angular distributions of the far-field harmonic intensity and that the experiment "naturally" picks the one with the highest on-axis emission by means of the pinhole, which eliminates the infrared pump beam in the annular-pump beam geometry [5]. The connection between the quantum path and the angular and coherence properties of the harmonic emission was discussed in [1] and [16]. In particular, the experiment in [16] confirmed that the harmonic emission resulting from the long quantum path has a much higher divergence due to a faster intensity dependence of the phase. In the experiment in [9], it is not really possible to select the long trajectory; only a substantial degradation of the mode-locking quality is observed when the phase-matching conditions are changed.

7. PULSE RECOMPRESSION

The harmonic chirp has a different sign for the two dominant quantum paths, as is clear from Fig. 5 or Fig. 7. The positive chirp along the short trajectory branch suggests that propagation in a negatively dispersive medium could compensate the chirp and compress the resulting attosecond pulses. This is confirmed by a calculation [9] and a recent experiment [17]. It would be interesting to select the long trajectory branch with a negative chirp to be able to recompress the pulses in a normally dispersive medium. The off-axis angular distribution of the harmonics seems, however, to be a serious practical obstacle.

8. MIR HARMONICS

To close this discussion on the optimization of an attosecond pulse train, we will briefly consider the influence of the pump frequency on the duration of the attosecond pulses by reducing the chirp. As was already mentioned, the quasi-classical theory indicates that the absolute value of the chirp is proportional to $1/U_P$ and, therefore, also proportional to ω^2/I. It is also possible to show that the optimum pulse duration (that corresponding to the optimum bandwidth) is proportional to $\sqrt{\Delta t_e}$ with Δt_e being the time delay between two consecutive harmonics, which are, hence, separated by 2ω, i.e., twice the fundamental frequency [9]. The optimum pulse duration is therefore proportional to $\omega^{3/2}$. It is known [18] that strong-field physics may be scaled to lower intensities by using smaller ionization potential (IP) atoms (like alkalis) and longer wavelengths. The Keldysh parameter ($\sqrt{2IP}/U_P$) can thus be kept constant. An interesting source of mid-infrared (three to four or even five microns) radiation can be obtained by difference frequency generation in a nonlinear crystal [18]. Indeed, high harmonics can be generated by such a source, and one may speculate that, because of the lower frequency, the chirp will be considerably reduced and, hence, shorter pulses produced, provided the same total bandwidth can be afforded (which, of course, means a much larger number of harmonic orders) at the same or higher intensity. This will be an object of future research.

9. CONCLUSION

An attosecond pulse train generated by plateau high harmonics and a long pump pulse has a number of obvious drawbacks with respect to a single attosecond pulse (generated by cutoff harmonics) and a few-cycle pump pulse, especially since the latter allows one to control the carrier-to-envelope phase [4]. It has, however, a few advantages. One of them is that, in principle, the acceptable bandwidth can be much larger in the plateau than in the cutoff, where it is limited on the high-energy side by the rapid drop of the harmonics amplitude and on the low-energy side by the chirp. Therefore, much shorter pulses could be produced especially after recompression, as was already demonstrated.

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