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Antoine, P; L'Huillier, Anne; Lewenstein, M

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Attosecond Pulse Trains Using High–Order Harmonics

Philippe Antoine,[1,3] Anne L'Huillier,[2] and Maciej Lewenstein[1]
[1]*Commissariat à l'Energie Atomique, DSM/DRECAM/SPAM, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette, France*
[2]*Department of Physics, Lund Institute of Technology, S-221 00 Lund, Sweden*
[3]*Laboratoire de Physique Atomique et Moléculaire, Chemin du Cyclotron, Université Catholique de Louvain, 2 B-1348 Louvain-la-Neuve, Belgium*
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We demonstrate that high-order harmonics generated by an atom in intense laser field form trains of ultrashort pulses corresponding to different trajectories of electrons that tunnel out of the atom and recombine. Propagation in an atomic jet allows us to select one of these trajectories, leading to a train of pulses of extremely short duration. [S0031-9007(96)00866-6]

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When an intense short-pulse laser is focused into a jet of rare gases, high-order harmonics are generated. The harmonic spectra present a decrease for the first harmonics, followed by a broad plateau of almost constant conversion efficiency, ending up by a sharp cutoff. Apart from the first and last harmonics, the spectra look like a comb of peaks with constant amplitudes, equally spaced in frequency by twice the fundamental photon energy (only odd harmonics are emitted, owing to the inversion symmetry). A question raised almost immediately, after the first spectra had been observed [1], was whether the harmonics were emitted in phase. In the time domain, the emitted signal would then consist of a train of pulses separated by half the laser period, of duration in the attosecond range. There is a clear analogy here with mode-locked lasers, where axial modes oscillating in a laser cavity are locked in phase, leading to the production of trains of short pulses [2]. This idea attracted a lot of attention, but also scepticism. Indeed, time-dependent numerical calculations for the single-atom response showed that the harmonics were, in general, not in phase. In addition, it was believed that propagation in a macroscopic medium would destroy any possible phase locking.

The understanding of physics involved in the generation of high-order harmonics made considerable progress, owing to the elaboration of the so-called two-step model [3]. In this quasiclassical description, an electron tunnels through the potential barrier formed by the combined Coulomb and electromagnetic fields. It can then be regarded as a free particle oscillating in the laser field. When it returns towards the nucleus, it can recombine to the ground state, emitting high-energy photons. Quantum-mechanical approaches [4,5] gave firm grounds to this interpretation, expressing, in particular, the time-dependent dipole moment (whose Fourier transform gives the harmonic components) as a sum of contributions from the different trajectories of the electron in the continuum. Finally, the fact that the phase of each harmonic component of the dipole moment varies rapidly with the fundamental intensity was found to be extremely important to understand propagation effects and the coherence properties of the emitted radiation [6,7].

The problem of attosecond pulse generation was recently discussed by Corkum *et al.* [8]. They proposed a clever way of selecting one pulse from a train of pulses obtained by phase-locked harmonics. Their idea is based on the high sensitivity of harmonic generation to the degree of ellipticity [9]. By creating a laser pulse whose polarization is linear only during a short time, close to a laser period, the emission could be limited to this interval. Thus, a single attosecond pulse could be produced. Note that this idea is based on the assumption that trains of attosecond pulses could be generated in the case of linearly polarized light, by appropriate filtering.

In the present Letter, we analyze the problem of the production of attosecond pulse trains, with the much deeper understanding brought about by the quasiclassical interpretation and its quantum-mechanical formulation. We show that, although the harmonics in the plateau region are not strictly speaking phase locked, the time-dependent single-atom emission consists of a train of ultrashort pulses, with two dominant pulses per half cycle, corresponding to the two main trajectories giving rise to harmonic emission. Under certain geometrical conditions, only one of these two contributions gets phase matched, leading to trains of ultrashort pulses, with one pulse per half cycle. The duration of the pulses is essentially that obtained by assuming the harmonics to have the same phase and amplitude (about 120 attoseconds for eleven harmonics of 825 nm radiation).

Consider N harmonics, all with the same amplitude and phase. The intensity $I(t)$ of the total signal emitted by these harmonics reads

$$I(t) \propto \left| \sum_{q=q_0}^{q=q_0+N-1} e^{-i(2q+1)\omega t} \right|^2 = \frac{\sin^2(N\omega t)}{\sin^2(\omega t)} . \quad (1)$$

$I(t)$ is a periodic function with periodicity $T_L/2 = \pi/\omega$, half the laser period (1.37 fs for the 825 nm wavelength considered later on). It consists of a succession of sharp peaks, with full width at half maximum $\Delta T \simeq T_L/2N$ ($\simeq$

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120 as for $N = 11$). As beautifully shown by Siegmann [2] for the case of mode-locked lasers, the regular pulsed structure of $I(t)$ is not much affected by any amplitude variation of the different frequency sidebands. It is, however, completely ruined if the frequency sidebands are not phase locked. Note that phase locking does not mean here that the harmonic components have the same phase, but rather that the phase difference (φ) between two consecutive harmonics is constant, so that a factor $\exp[-i(2q + 1)\varphi]$ enters the sum in Eq. (1).

To examine whether harmonics are phase locked, we calculated the phase difference between the harmonic components for a neon atom exposed to a 825 nm wavelength radiation, at an intensity of 4×10^{14} W/cm^2, using the model described in Refs. [5,10]. The results are shown in Fig. 1. The phase difference between two harmonics is apparently completely random in the plateau region. The cutoff [11] occurs for harmonic energies higher than $\simeq I_p + 3.2U_p \simeq 69\hbar\omega$, where I_p is the ionization potential, U_p the ponderomotive energy (proportional to laser intensity), and ω the laser frequency. This result confirms the results of the earlier calculations [12], namely that the harmonics in the plateau region are not phase locked. The phase locking exhibited by the harmonics in the cutoff is not very interesting, because the harmonic amplitude decreases very rapidly with the process order, so that only one or two harmonics really contribute to $I(t)$, leading to broad light pulses.

In Fig. 2, we plot in solid line the function

$$I(t) = \left| \sum_{q=q_0}^{q=q_0+N-1} d\big((2q + 1)\omega\big) e^{-i(2q+1)\omega t} \right|^2 \quad (2)$$

over one optical cycle. $d\big((2q + 1)\omega\big)$ denotes the $(2q + 1)$th harmonic component of the dipole moment. We choose 11 harmonics, from the 41st to the 61st, in the plateau region, and the same laser intensity and wavelength as in Fig. 1. The linearly polarized electric field is chosen proportional to $\cos\omega t$ so that its extrema occur at $t = \pm k\pi/\omega$, with k integer. Surprisingly, the train of pulses is not as irregular as one could have expected on the basis of the phase variation shown in Fig. 1. It consists of two dominant peaks (labeled by τ_1 and τ_2) and several less significant peaks (per half cycle).

In our theory, the atomic dipole moment is expressed as a sum of complex contributions corresponding to processes in which an electron tunnels to the continuum, undergoes subsequent (quasi)free evolution under the influence of the laser field, returns to the nucleus and recombines. In a recent paper [7], we analyze the relevant trajectories leading to harmonic emission, using a saddle-point method. The trajectories are determined by the stationarity condition of the quasiclassical action $S(\mathbf{p}, t, \tau) - (2q + 1)\omega t$ with respect to variations of the canonical momentum $\mathbf{p}$, the time t for harmonic emission, and the return time τ, i.e., the time spent by the electron in the continuum. Physically, it means that only those trajectories are selected for which (i) the electron returns to the nucleus after time τ, (ii) its kinetic energy at the moment of tuneling is negative and equal to $-I_p$, and (iii) its kinetic energy just before recombination is determined by energy conservation and equal to $(2q + 1)\hbar\omega - I_p$. The atomic dipole moment is essentially determined by two of these trajectories, which correspond to the shortest return times τ_1 and τ_2, within one laser cycle. The two main peaks in Fig. 2 appear at the emission times corresponding to these two trajectories. To check that they can be interpreted as the two light bursts emitted by the electron following these two trajectories, we calculated the temporal profile [Eq. (2)] by taking into account only the contribution of return times smaller than one cycle. The result is shown by the dashed curve in Fig. 2. The two main peaks are almost unchanged, indicating that they are due to processes occurring within a laser cycle, whereas the smaller peaks disappear. Note that, for lower intensities, such that the harmonics enter the cutoff region,

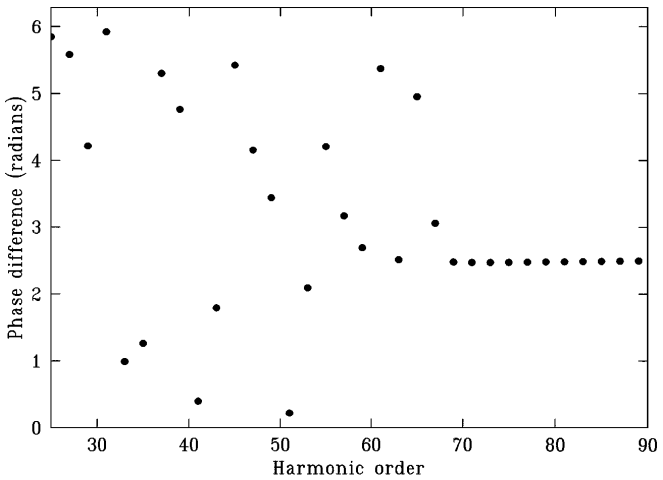

FIG. 1. Phase difference between the hamonic components for a neon atom exposed to a laser light of wavelength 825 nm, and intensity 4×10^{14} W/cm^2.

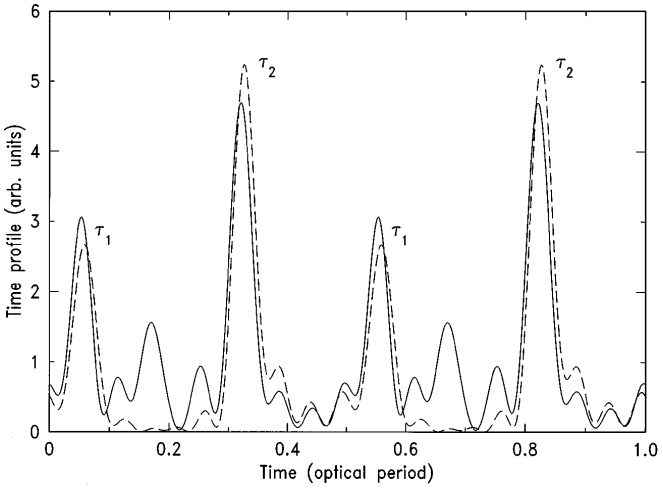

FIG. 2. Train of pulses of 11 harmonics (from 41st to 61st) emitted by a neon atom. Laser parameters same as in Fig. 1. The dashed line corresponds to return times smaller than one cycle.

the atomic dipole is dominated by a single contribution, corresponding to the (complex) return time τ_2.

The properties of the two dominant trajectories are summarized in Fig. 3. Here we show the ionization and recombination times as a function of intensity. The solid curves correspond to the 41st harmonic alone, the dashed curves, to the 61st harmonic, whereas the solid dots are the intensity-dependent positions of the dominant peaks in the train of pulses of Fig. 2. They represent the cumulative results for the filtered atomic response [Eq. (2)]. As expected, the positions of the bursts in the cumulative response fall between the emission times for the 41st and 61st harmonics. Also, for small intensities, the two dominant peaks merge together, and only one of them (τ_2) remains significant.

For the trajectory labeled by the index 2, the ionization time is $\simeq 0$, when the electric field is maximal and the tunneling process most probable. The probability amplitude of the harmonic emission process is, however, reduced significantly by quantum diffusion, since the return time τ_2 is relatively long. For the first trajectory, tunneling takes place later, when the electric field has a smaller amplitude, with a lower probability. However, quantum diffusion is not as important for this process, since the return time τ_1 is smaller. The probability amplitudes for these two emission processes are actually comparable and interfere very efficiently, leading to apparently random phases (see Fig. 1) as well as harmonic strengths. By analyzing the emission process in the *time* rather than in the *frequency* domain, the two processes are naturally separated. They give rise to regular trains of attosecond pulses, with two bursts per half laser cycle.

To determine the signal emitted by a macroscopic medium, one calculates first the harmonic fields $E((2q+1)\omega, r)$ at the exit of the medium, by solving propagation (Maxwell) equations [10]; r is here the radial coordinate. The source term that enters the propagation equation for a given harmonic field is the laser-induced polarization at the harmonic frequency, proportional to the single-atom response and to the atomic density. The laser pulse is modeled as a Gaussian beam with a confocal parameter $b = 5$ mm. The peak intensity at the focus is 6.6×10^{14} W/cm^2. We consider two cases: (a) the 1 mm long atomic gas jet is placed 2 mm after the laser focus, and the intensity at the center of the medium is therefore equal to 4.4×10^{14} W/cm^2; (b) the gas jet is centered at the laser focus. Note that since the harmonic field $E((2q+1)\omega, r)$ is obtained through the integration of a propagation equation, it accounts au-

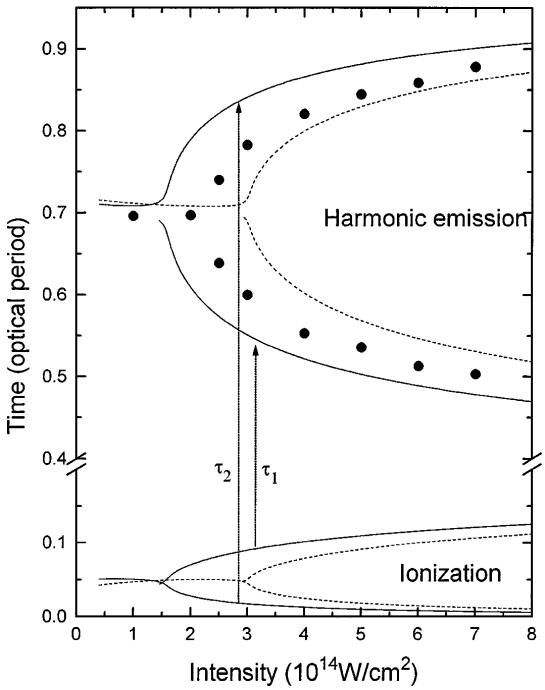

FIG. 3. Ionization times (lower branch) and recombination (harmonic emission) times (upper branch) for the two dominant trajectories $i = 1, 2$ as a function of laser intensity. Solid lines correspond to 41st harmonic, dashed lines to 61st harmonic, and solid dots represent dominant peak positions for the cumulative response of the atom [Eq. (2)].

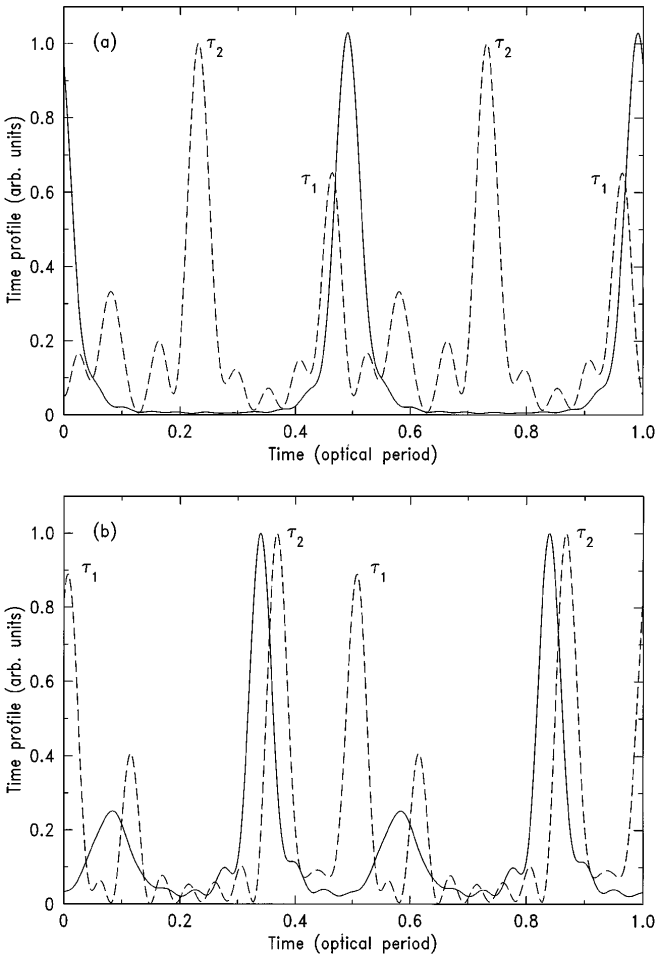

FIG. 4. Time profile of the filtered harmonic signal including 41st-61st harmonics (solid line) emitted by the macroscopic medium located at (a) before ($z = -2$ mm), or (b) at the focus ($z = 0$ mm), generated by a laser pulse of intensity 6.6×10^{14} W/cm^2. The dashed line denotes the corresponding single-atom response.

tomatically for the effect of phase matching. In Figs. 4(a) and 4(b) we plot in solid line the total signal (integrated over the radial coordinate r) for the two cases (a) and (b),

$$I(t) = \int \left| \sum_{q=q_0}^{q=q_0+N-1} E((2q+1)\omega, r) e^{-i(2q+1)\omega t} \right|^2 2\pi r \, dr \tag{3}$$

over one optical cycle. The filter applied (selecting harmonics from the 41st to the 61st) is the same as in Fig. 1. We show the single-atom response obtained at the same intensity in dashed line. In case (a), in order to compare single- and many-atom responses in a meaningful way, we have accounted for a (weak) geometrical phase shift [$\simeq 0.1$ in the units of Fig. 2(a)] of the fundamental.

The key result of this Letter is that propagation selects here 1 and only 1 attosecond pulse per half cycle *with no additional broadening*. The propagated harmonic components are locked in phase, so that the macroscopic signal contains a train of well defined 120 as pulses. In the conditions of Fig. 4(a), the selected peak is the one corresponding to the shortest trajectory, with the return time τ_1. All other contributions (with the return time τ_2 or longer than one period) practically vanish. In Fig. 4(b), another trajectory is selected. In that case, the peak selected corresponds to the contribution of the trajectory with the return time τ_2. Other contributions are considerably reduced, giving rise to small and broad background pulses in between the dominant ones.

To understand the role of propagation, we recall [6] that phase matching between the nonlinear polarization in the medium and the generated harmonic field is essentially determined by the interplay between the phase variation ($\delta\Psi_{\text{geo}}$) induced by the geometrical phase shift of the fundamental across the focus and the variation of the total dipole phase in the nonlinear medium (following the distribution of laser intensity). In the present work, we separate the contributions from the different trajectories by analyzing the temporal output of the nonlinear medium for a selection of harmonics. The effect of propagation on the two main trajectories can be understood by considering the phase variation ($\delta\Psi_i$) induced by each of them. As illustrated in Fig. 3, the return times τ_1 and τ_2 have a different intensity dependence. As the laser intensity increases, τ_2 increases, whereas τ_1 decreases. The phases corresponding to the two contributions (approximately equal to $-U_p\tau_i$; see [7]) vary differently with the laser intensity, and consequently in the nonlinear medium. This affects phase matching dramatically. Depending on the geometrical conditions, i.e., on the behavior of $\delta\Psi_{\text{geo}}$, the contribution of one trajectory can be enhanced (if $\delta\Psi_{\text{geo}} + \delta\Psi_i$ is small) and the others reduced. When the laser is focused before the gas jet, for example, as is the case of Fig. 4(a), the phase variation induced by focusing is small and the selected trajectory is the one with the slowest phase variation (τ_1).

In summary, we have shown that a single atom generates harmonics in the form of a train of attosecond pulses, even though the Fourier components of the atomic dipole moment are not locked in phase. There are, in general, several pulses per half period corresponding to various energetically allowed electronic trajectories. Propagation selects only one of these trajectories, and locks harmonics in phase. The temporal output of the medium then consists of a train of sharp pulses, with only one pulse per half cycle. This selection can be controlled by changing the position of the atomic jet relative to the laser focus.

This result is very important for two reasons. First, it opens the route towards the production and utilization of attosecond pulses. Trains of attosecond pulses can be used to probe phenomena with the same periodicity (e.g., induced by the same laser). Second, by considering ten harmonics (forming a train of attosecond pulses) instead of only one, the intensity of the radiation increases by a factor of 100. Typical numbers for a harmonic of energy $\simeq 50$ eV, are 10^9 photons [13] with a measured 100 fs pulse duration [14], and an estimated focal spot of $\simeq 1\ \mu\text{m}^2$. The mean intensity of a single harmonic pulse may thus reach 10^{13} W/cm^2. The intensity corresponding to a train of pulses combining ten such harmonics would then be $\simeq 10^{15}$ W/cm^2. There are several fascinating applications of such an intense extreme ultraviolet radiation in the area of strong field laser-atom physics.

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