# Temporal and Spectral Tailoring of High-Order Harmonics

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(Received 10 November 1997)

We demonstrate that the main features of high harmonic generation by ultrashort laser pulses can be explained in terms of the intensity dependent phase of the atomic polarization. Focusing conditions and chirped driving fields may be used to control the harmonic spectrum, while temporal compression provides a unique way to tailor the time profile. [S0031-9007(98)07968-X]

PACS numbers: 42.65.Ky, 32.80.Rm

Recently, a great deal of attention has been devoted to high-order harmonic generation (HHG) by ultrashort laser pulses (cf. [1,2]). Indeed, this is a promising way to generate subfemtosecond, i.e., attosecond, pulses of radiation of very high frequency. New phenomena arising in harmonic spectra at these short laser pulse durations have been reported both in theoretical calculations and in experiments. First, single atom simulations revealed that, for 27 fs pulses, harmonics in the plateau region are not any more resolved and disappear in a structured continuum [3]. Cutoff harmonics, however, are found broad but discrete with regular quadratic phases. On the other hand, recent experiments showed that the spectrum could evolve from an irregular continuum to well-resolved harmonic peaks by simply playing on the focusing conditions or on the chirp of the fundamental beam [4,5].

These strange phenomena are of the utmost interest for two reasons: for attosecond pulse generation, the largest compressible spectral bandwidth is desirable; for applications that require a high degree of coherence, such as extreme ultraviolet (XUV) interferometry, well-resolved harmonics are necessary. Thus controlling and optimizing harmonics widths is a challenging task. Unfortunately, the above-mentioned effects have not thus far been understood in the framework of the usual adiabatic theory of HHG, in which a slowly varying envelope approximation is used to describe the laser pulse. In effect, nonadiabatic effects due to the short pulse duration have been invoked to interpret the results [3,6], and, in particular, the behavior of the harmonic phase. Indeed, a change in intensity on the time scale of the laser period could alter the harmonic generation process [7]. In the quasiclassical description of HHG [8], an electron first tunnels through the potential barrier and then oscillates quasifreely in the laser field, gaining kinetic energy. When it returns back to the nucleus, it can recombine and emit harmonic photons. If the laser intensity varies sufficiently rapidly, electrons entering the continuum while the laser intensity is increasing can experience an additional acceleration before returning. This could produce a blueshift on the rising edge of the laser envelope. Conversely, electrons ionized after the peak of the pulse would be decelerated, returning later, which would lead to a redshift of the spectrum. The observed spectral characteristics would thus be the signature of the distorted atomic response to the short laser pulse. This raises many fundamental questions: What is the pulse duration necessary to induce a nonadiabatic response of the atom? Can the conflicting observed phenomena and theoretical results be reconciled and explained within the adiabatic theory? What is their physical origin and can they be controlled?

In this Letter, we show that the above-mentioned phenomena are the manifestation of the same effect, namely, the intensity variation of the adiabatic phase accumulated by the electron on its trajectory [9,10]. This is a very general process, whose magnitude is amplified by the short pulse duration. For the first time, we perform full nonadiabatic calculations, from the single atom response to the three dimensional propagation of the generated harmonic fields, and compare them with the adiabatic theory. In contrast to Ref. [11], where the influence of ionization on phase matching was studied in the one dimensional limit, we focus here on the incidence of the "pure" 3D propagation on the spectrum, below the saturation intensity. The results indicate that the adiabatic approach is still valid for 27 fs pulses, and that the harmonic phase characteristics can be used to control the temporal and the spectral properties of the harmonic beam. In particular, we show that the temporal compression provides a unique opportunity for tailoring the harmonic emission in the form of a train of ultrashort pulses with variable periodicity and number of peaks. Finally, the adiabatic approximation breaks below 27 fs, and the influence of the phase of the driving field is dramatic already for 10 fs pulses.

First, we calculated the nonadiabatic single atom response to the fast driving field in the strong field approximation (SFA) [12] and compared it with the adiabatic result. The time dependent dipole moment of an atom can be written in the SFA as

$$\vec{x}(t) = i \int_{-\infty}^{t} dt' \int d^3\vec{p}\,\vec{d}^{*}[\vec{p} - \vec{A}(t)]a^{*}(t) \times \exp[-iS(\vec{p}, t, t')]\vec{E}(t') \cdot \vec{d}[\vec{p} - \vec{A}(t')]a(t'), \quad (1)$$

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where $a(t)$ is the ground state amplitude, $\vec{A}(t)$ is the vector potential, $\vec{d}(\vec{p})$ is the dipole moment of the ground-continuum transition, and $\vec{E}(t)$ is the electric field. In the above expression, $S(\vec{p}, t, t')$ is the *quasiclassical action* for an electron born in the laser field at $t'$ with a canonical momentum $\vec{p}$, and returning to the origin at $t$ (return time $\tau = t - t'$),

$$S(\vec{p}, t, t') = \int_{t'}^{t} dt'' \left( \frac{[\vec{p} - \vec{A}(t'')]^2}{2} + I_p \right).$$

The expression (1) can be clearly interpreted in the Feynman's spirit as an integral over all possible electronic trajectories characterized by $t'$, $t$, and $\vec{p}$. Nonadiabatic theory employs directly Eq. (1) for a given temporal form of the laser pulse $\vec{E}(t)$. In the adiabatic theory, the expression (1) is used to calculate the harmonic components of $\vec{x}(t)$ for a field of given *constant* amplitude $\mathcal{E}$ and phase $\phi$. The time dependence of the Fourier components (and their spectral broadening) is then introduced by replacing $\mathcal{E}$ and $\phi$ by a slowly varying pulse envelope $\mathcal{E}(t)$, and a modulated phase $\phi(t)$.

We applied both methods to the conditions of Ref. [3], i.e., an argon atom interacting with a 27 fs, 810 nm pulse with a peak intensity of $3 \times 10^{14}$ W/cm$^2$. In these calculations, we neglect ionization since the considered intensity is (slightly) below saturation for this small pulse duration. The spectrum obtained using the nonadiabatic approach is presented in Fig. 1(a). In the plateau region, the spectrum is "white" with overimposed irregular fluctuations, whereas harmonics in the cutoff are broad but well-resolved, and exhibit regular quadratic phases. Amazingly, the behavior of such a spectrum can be well-described by the adiabatic approach. The underlying phenomenon is the phase modulation induced by the intensity dependence of the dipole phase. It has been shown that this phase is determined by the value of the action acquired by the electron along the most relevant trajectory [9,10]. Since the action $S$ is primarily determined by the ponderomotive energy $U_p \propto I\lambda^2$, the dipole phase should be $\Phi = -S \simeq -U_p\tau$, where $\tau$ is the return time, i.e., the time between tunneling and recombination. Let us first consider the cutoff region. There, $\tau$ is close to half of a period, which gives a good estimate for the slope of the linear intensity dependence of the dipole phase. Since the harmonic emission is efficient close to the top of the Gaussian laser pulse, the phase changes quadratically with time, and thus corresponds to a linear frequency chirp. The rising edge of the harmonic pulse is shifted to the blue, and the falling edge, to the red. The nonadiabatic results, profile and phase, are strikingly similar to the adiabatic results. The main difference appears in the temporal profiles: The nonadiabatic is delayed by roughly 1.3 fs compared to the adiabatic. This shift in time can be related to the physics of the process. Since the amplitude of the emission is mainly determined by the ionization probability, the emission, that corresponds to the recombination, is delayed by the value of the return time (close to half of a period in the cutoff region). In the plateau region, there are typically two electron trajectories that give dominant contributions interfering with each other [10]. One of these trajectories corresponds to a short return time, whereas the other, to a return time close to one period, and thus to a strong intensity dependence of the phase. This contribution undergoes particularly strong phase modulation in the time dependent field, and consequently strong spectral broadening, leading to an overlapping of neighboring peaks and to a white spectrum [see Fig. 1(a)].

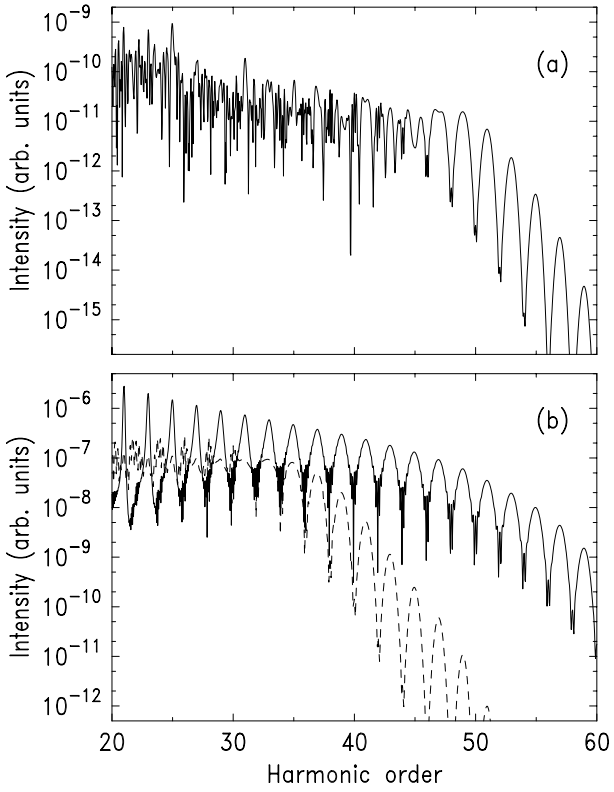


FIG. 1. (a) Single atom spectrum generated in argon by a 27 fs laser pulse at $3 \times 10^{14}$ W/cm$^2$. (b) Propagated harmonic spectrum calculated in the same condition as (a) with the confocal parameter $b = 5$ mm, and the atomic jet located 2 mm after the focus (solid line), and at the focus (dashed line).

The accuracy of the adiabatic theory can be higher for the macroscopic harmonic signals. In order to calculate them, we propagate the harmonics generated by individual atoms using Maxwell equations in the paraxial approximation [13]. The atomic polarization which is the source of the harmonic fields is proportional to the dipole moment $x(t)$. In the nonadiabatic theory, we calculate the full time dependence of $x(t)$ from Eq. (1) at each point of the fine spatial grid, Fourier transform it, and propagate *all* components of the continuous Fourier spectrum independently using Maxwell equations. In the adiabatic theory, only *harmonic* Fourier components are propagated for a given

value of the driving field amplitude $\mathcal{E}$ and phase $\phi$. They are then made time dependent, and phase modulated by replacing $\mathcal{E} \rightarrow \mathcal{E}(t)$ and $\phi \rightarrow \phi(t)$.

Propagation and phase matching bring new effects into account, i.e., the interplay between the dynamically induced phase of the atomic polarization and the phase of the focused fundamental (say, Gaussian) beam [9]. We consider here the case of a relatively tight focusing with a confocal parameter $b = 5$ mm, assuming a 1 mm wide argon jet with a Lorentzian density profile. The 27 fs laser pulse has the intensity $3 \times 10^{14}$ W/cm$^2$ at *the center of the jet*. In such a case, the macroscopic signal in the cutoff region resembles that of the single atom, but in the plateau it depends critically on the position $z$ of the jet with respect to the focus. The reason is that propagation may favor contribution of some, and diminish contributions of other, electronic trajectories [14]. If the jet is located after the focus [cf. $z = 2$ mm, solid line in Fig. 1(b)], the trajectory with the short return time $\tau$, and small action, is selected. Its contribution is characterized by a very weak intensity dependence (that becomes weaker for lower harmonic orders). As a result, phase modulation is small, and the spectrum contains very well-resolved harmonics with decreasing widths for decreasing harmonic orders. Conversely, at $z = 0$ [jet at the focus, dashed line in Fig. 1(b)], the contribution of electronic trajectories with longer $\tau$, and thus stronger intensity dependence of the phase, is dominant. In this case, the phase modulation is very strong, and the spectrum remains white with overimposed irregular structures, similar to the case of the single atom in Fig. 1(a). Note that, even though the intensity at the center of the jet is the same in both cases, the cutoff position for $z = 0$ is shifted toward lower harmonic orders due to phase matching effects [15].

The nonadiabatic results of Fig. 1(b) and the results of adiabatic theory are practically indistinguishable for $z = 2$, and very similar for $z = 0$. The discussed mechanism explains a long standing mystery, why well-resolved macroscopic harmonic spectra are observed in experiments with pulses as short as 27 fs, whereas they are smeared out on the level of a single atom due to interference effects [16].

Given the regularity of the phase modulation, it should be possible to compensate for (or increase) the dynamically induced harmonic chirp by introducing an appropriately designed frequency chirp on the laser beam. We have calculated the macroscopic harmonic spectrum generated in argon by a laser pulse presenting the same spectral width as the 27 fs pulse considered above, but with a quadratic phase in frequency. The latter fact induces a temporal broadening of the pulse (to 38 fs in the case considered), together with a quadratic phase in time (linear chirp). Figure 2 presents the spectra obtained from adiabatic theory for positive and negative chirps in the case $z = 0$. For this position, the dynamically induced

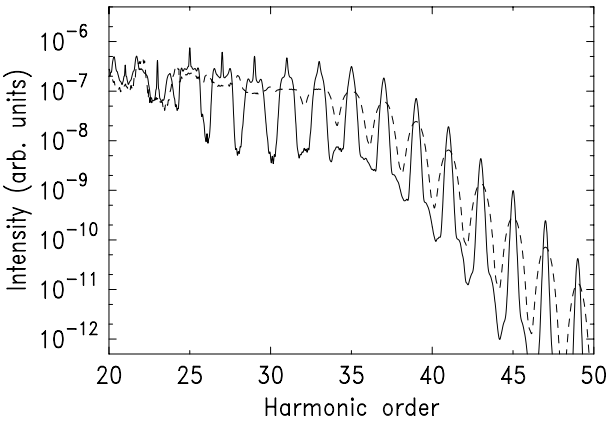


FIG. 2. Harmonic spectrum generated by a 38 fs laser pulse presenting a positive chirp (solid line) or a negative chirp (dashed line).

harmonic chirp, which is negative, is relatively big, and the spectrum in the absence of laser chirp is white in the plateau [see Fig. 1(b)]. When the laser chirp is negative (blue before the red, dashed line), it adds to the negative dynamic chirp, and does not modify significantly the spectrum as compared to the Fourier limited 27 fs case. On the other hand, the positive laser chirp gets subtracted from the dynamic chirp, leading to very well-resolved harmonics at the end of the plateau, and significantly narrower peaks in the cutoff. This asymmetry has been observed in experiments in argon [5], and more recently in helium [4]. Our theory provides the first explanation of those effects.

One can then wonder whether it is possible to compress temporally the *macroscopic* harmonic pulses, as was done for the single atom response in the cutoff region [3]. Indeed, the phase modulation is not spatially homogeneous and can be significantly perturbed by the emission from points experiencing different peak intensities. Moreover, in the plateau region, harmonics can overlap. What would be the result of the compression in such a case? To simulate the compression by a pair of gratings, we subtracted a mean quadratic phase, centered on a given frequency, from the phase of the spectrum emitted at each spatial point at the exit of the medium, and analyzed the resulting temporal macroscopic profiles. The result of such a procedure is shown as a solid line in Fig. 3 in the case $z = 0$, and for the frequency range $30\omega_L$–$36\omega_L$. The profile consists of a train of three ultrashort peaks of duration $\simeq$1.5 fs separated by 4.4 laser periods $T$. They correspond to the three harmonics contained in this frequency range, that are individually compressed. Indeed they present similar phase modulations but centered on each harmonic frequency $\omega q$: $\Phi_q(\omega) = -a(\omega - \omega_q)^2$. Removing the chirp of harmonic $q'$ by adding the phase $a(\omega - \omega_{q'})^2$ results for harmonic $q$ in both a compression and a time shift $\Delta t = 2a(\omega_q - \omega'_q)$. In the case considered here, $a \simeq 3$ eV$^{-2}$ results in $\Delta t = 4.4T$ for

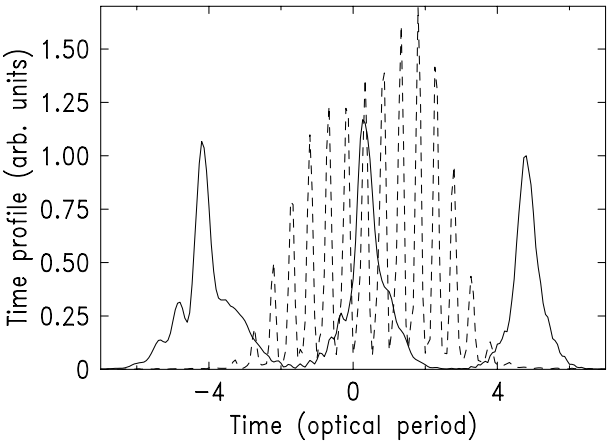


FIG. 3. Temporal profile of the macroscopic harmonic emission in the frequency range $30\omega_L - 36\omega_L$ before (dashed line) and after (solid line) compression.

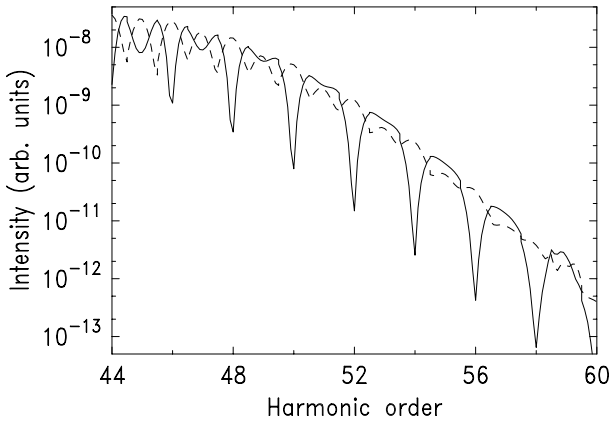


FIG. 4. Macroscopic harmonic spectra generated with a 10 fs laser such that $\phi = 0$ (dashed line) or $\phi = \pi/2$ (solid line).

neighboring harmonics. Note that this train is very different from the train obtained before compression, associated to the phase relationship between harmonics [14]. The latter is presented as a dashed line in Fig. 3. The peaks are narrower but their number and periodicity (equal to $T/2$) are fixed. On the contrary, the compression brings a unique possibility of tailoring the harmonic temporal profile. We have shown that the harmonic spectrum (and chirp) could be controlled thanks to the jet/focus position and to the laser chirp. We thus have a direct control of $\Delta t$ through the value of $a$, while keeping relatively short pulses. Moreover, by filtering an appropriate number of harmonics out of the spectrum, one can choose the number of peaks into the train. This pulse shaping at the femtosecond scale provides a rare opportunity for performing coherent control of ultrashort wave packet dynamics in the XUV.

We have shown that the adiabatic theory explains the main features of harmonic spectra generated with pulses as short as 27 fs. We now investigate what happens for shorter pulse durations. A striking nonadiabatic effect appears for 10 fs laser pulses: the spectrum changes dramatically depending on the phase of the driving field [17], as illustrated in Fig. 4 in the case $z = 0$. When the field is maximum at the center of the laser envelope $t = 0$ ($\phi = 0$, dashed line), even harmonics as intense as the odd are generated, leading to an almost white spectrum also in the cutoff region. On the contrary, when the field is minimum at $t = 0$ ($\phi = \pi/2$, solid line), only odd harmonics quite broad but discrete are generated. In this case, the behavior is very similar to that described by the adiabatic calculation. The reason is that a certain amount of symmetry is preserved close to $t = 0$ where the harmonics are generated: $\vec{x}(t + T/2) \simeq -\vec{x}(t)$. Note that the distortion of the spectrum in the case $\phi = 0$ is even larger in the single atom response and survives the propagation. The experimental observation of such behavior would be the signature of the first truly nonadiabatic phenomenon in HHG.

In conclusion, for pulses as short as 27 fs, the regular behavior of the dynamically induced harmonic chirp can be employed to tailor the temporal as well as the spectral properties of the macroscopic harmonic emission. This unprecedented capability of pulse shaping in the XUV at the femtosecond scale opens wide perspectives for the applications of HHG in physics.

We thank M. Gaarde, A. L'Huillier, H. Kapteyn, K. Kulander, K. Schafer, and M. Murnane for discussions and sharing with us results prior to publication.

*Also at Fonds national de la Recherche Scientifique de la Communauté Française de Belgique.

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