Nonadiabatic Effects in High-Harmonic Generation with Ultrashort Pulses

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High-harmonic generation using ultrashort laser pulses with pulse durations 25 to 200 fs is studied theoretically and experimentally. We observe that the harmonic spectrum of argon taken with 25 fs laser pulses contains harmonics up to 20 orders higher than for 100 fs laser pulses with the same intensity. We show that this increase in harmonics is because the atom survives to higher laser intensities, due in part to the nonadiabatic response of the atomic dipole to the fast rise time of our pulse. [S0031-9007(96)01018-6]

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The shortest x-ray pulses which can be generated to date are obtained by high-harmonic conversion of intense laser pulses in gas jets. This scheme has been used to generate harmonics up to orders exceeding 100, using either picosecond [1] or femtosecond pulses [2]. High-harmonic generation thus represents an attractive technique for generating ultrafast coherent radiation in the UV and soft x-ray region of the spectrum.

For high intensity laser illumination, the ionization of atoms occurs via tunneling through the core potential [3–5]. Once free, the electron moves in the laser field, and when the laser field reverses, the electron can return to the core with a maximum kinetic energy of $3.17U_p$. Here, $U_p = E^2/4\omega^2$ is the ponderomotive or quiver energy (atomic units) of a free electron in a electric field E with frequency ω. Thus the energy of the highest harmonic emitted from an atom of ionization potential I_p is predicted to be $I_p + 3.17U_p$. With the exception of our experiments, this prediction is in good agreement with experimental data to date for the width of the plateau region and the cutoff wavelength of the harmonic spectra [1,2]. Here, U_p corresponds to the maximum field that an electron may experience before ionization, even though the laser field may subsequently increase. This simple law has also been confirmed by numerical [6] and analytic calculations [7,8].

In all the experiments performed by others, laser pulse widths >100 fs have been used. In the visible region of the spectrum, this implies that the field amplitude changes little between successive optical cycles. Thus when the laser pulse reaches the intensity required for tunneling, there is sufficient time for the electron to escape from the atomic core before the field increases further. We recently reported [9] the generation of high harmonics from noble gases, which for the case of 25 fs pulses, exhibited unusually high orders in comparison with previous work using longer pulses. We also reported that the harmonics could be tuned in wavelength by adjusting the chirp of the excitation pulse.

In this paper, we report new results on the direct comparison of harmonic emission as the transform-limited pulse width of the excitation laser is varied over the range 25–200 fs. This avoids any complications due to the introduction of chirp [9]. Our experiments show that the harmonic spectrum of argon taken with 25 fs laser pulses contains harmonics up to 20 orders higher than for 100 fs excitation pulses with the same intensity. We also report on quantum calculations which consider the interaction of a 1D atom with an intense laser pulse. We show that the ionization process can be strongly affected by the ultrashort rising edge of the pulse, and that, in the presence of finite ionization rates, the atom can survive to higher laser intensities prior to ionization. The electron is then exposed to a stronger, rapidly increasing, laser field, which allows the electron to gain even more energy prior to reencountering the parent ion. Quantum mechanical calculations show a further effect for very short pulses. For rapid rise-time pulses, the phase of the atomic dipole lags that of the field, which can result in reduced ionization due to the nonsinusoidal shape of the electric field in time.

A simple intuitive picture of tunneling, described quantum mechanically by the Keldysh theory [3], can be seen as follows. In the case of a field with an amplitude E, comparable to the amplitude which suppresses the Coulomb barrier to the level of I_p (the appearance amplitude $E_{\text{app}} = I_p^2/4$) [10], the barrier width is given by $L = 2(E_{\text{app}} - E)^{0.5}/E$. Classically, we assume the bound energy of the electron to be kinetic with velocity $v = (2I_p)^{0.5}$, to obtain a tunneling time

$$T_{\text{tun}} = \sqrt{E_{\text{app}} - E}\,/(EE_{\text{app}}^{0.25}). \tag{1}$$

On the other hand, quantum mechanically for weak fields ($E \ll E_{\text{app}}$), the ratio of the tunneling time to the laser period is given by the Keldysh parameter $\gamma = 2E_{\text{app}}^{0.25}\omega/E$. Thus a simple classical model retains the qualitative features of the more exact calculation. In the case of argon, for example ($I_p = 0.58$ a.u.), at a field of $0.5E_{\text{app}}$, we find a tunneling time of $\approx$3 fs. (The period of our laser light is $\approx$2.7 fs, while the pulse width is 25 fs.) We will show that when the tunneling time is close to an optical cycle ($\gamma \approx 1$) and for rapid rise-time pulses, the electron wave

function can experience a strongly delayed response. This effect is in addition to the theoretically predicted increase of harmonic orders for short pump pulses recognized by others in the past [5,11]. However, none of the previous work considers the dynamics of wave function within a few periods of the optical field, in the transient regime where the atom responds to the nonsinusoidal individual half period of the increasing electric field, and when the Keldysh parameter is close to unity. Moreover, although many recent calculations on high-harmonic generation and stabilization of atoms used simulated pulses with a rise time of a few optical periods, followed by a region with constant amplitude to avoid the "death valley" problem of atomic ionization [12,13], only recently have lasers advanced to the point that such high-power ultrashort pulses can be generated in the laboratory [14,15].

In order to model the interaction of a single atom with an ultrashort pulse, we solve the Schrödinger equation for a 1D Coulomb potential [16]

$$i\frac{\partial\Psi(x,t)}{\partial t} = \left[-\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{|x| + \alpha} - xE(t)\right]\Psi(x,t). \tag{2}$$

For small $\alpha \approx 0$, the eigenvalues and eigenfunctions of Eq. (2) approach those of the 1D hydrogen atom. (For no external field, the 1D and 3D hydrogen atom energy levels are the same, with zero probability of finding the electron at the core.) The lowest-energy state has energy of -0.5 a.u. (ignoring a single infinitely bound state peaked at the core, but which does not couple to other states through dipole transitions) [16]. In contrast, when the smoothing parameter α is large, this state is no longer infinitely bound, and it can interact with other states through dipole transitions. For example, the soft potential $-1/(2 + x^2)^{0.5}$ supports a ground state of $I_p = 0.5$ a.u. which is peaked at the core [17,18]. We use a small value of $\alpha = 0.037$ a.u., which allows the core-peaked wave function to be ignored because it is deeply bound. This modest degree of smoothing gives for the next lowest state (the *de facto* ground state) a binding energy of $I_p = 0.443$ a.u., and the corresponding appearance field is 0.049 a.u. ($I_{\text{app}} = 8.4 \times 10^{13}$ W/cm^2). We calculate the harmonic spectra by using the dipole acceleration approach [18]. In order to determine the time dependence of a specific harmonic, we Fourier transform the corresponding spectral peak. The ionization probability was calculated by projecting the wave function over the first 50 bound states of the atom, since the contribution of the remaining states was found to be negligible.

Our calculations show that for weak pulses ($E \ll E_{\text{app}}$) the harmonic spectra do not depend on the pulse width, since the highest intensity the atom can experience is always the peak intensity of the pulse. On the other hand, for strong pulses ($E \approx E_{\text{app}}$) the ionization can depend on the pulse width. Figures 1(a), 1(b), and 1(c) show the time dependence of the ionization probability, the third-harmonic field, and the phase of the atomic dipole, respectively, for 100 fs excitation. These figures show that the peak of the third-harmonic field corresponds to an ionization probability of 42%, occurring at a field amplitude of $0.74E_{\text{app}}$, and that the dipole phase is nearly constant. Figures 1(d), 1(e), and 1(f) correspond to excitation by a 25 fs duration pulse, of the same peak intensity. In this case, at the peak of the third harmonic the ionization probability is 32% while the amplitude of the field is $0.86E_{\text{app}}$. Therefore the atom is exposed to higher fields for shorter duration excitation pulses ($<$100 fs). Because of the higher field, the kinetic energy of the electron significantly increases, and this results in the generation of more intense and higher-energy harmonic peaks.

It can also be seen from Fig. 1(f) that the phase of the dipole changes significantly for 25 fs illumination when compared with 100 fs illumination, particularly when the ionization exceeds 50%. Thus for weak fields or long pulses, the atomic dipole adiabatically follows the oscillations of the laser field, but, for ultrashort pulses of high intensity, the response of the wave function is delayed with respect to the laser field. The onset of this phase delay coincides with the stabilization of the

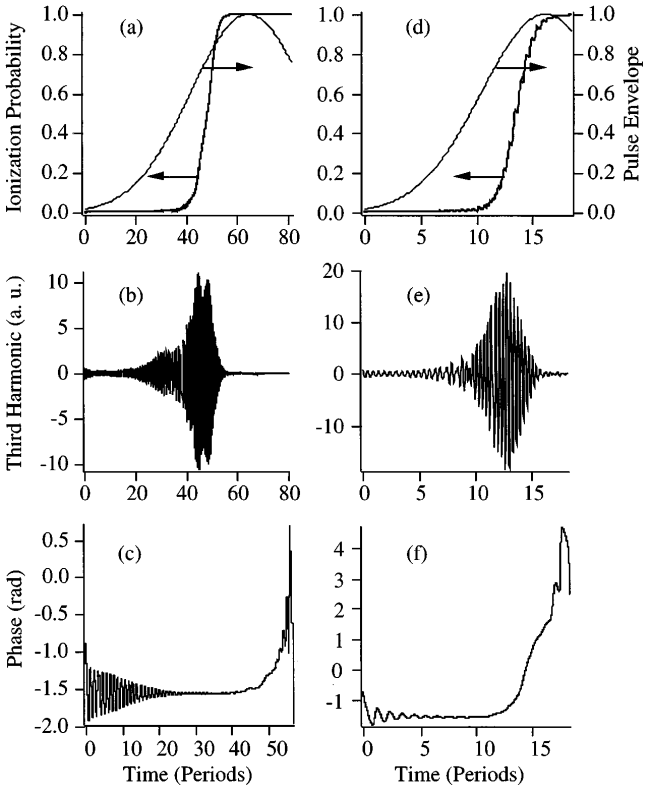

FIG. 1. Time dependence of (a) the pulse envelope and ionization probability for a 100 fs excitation pulse, with peak amplitude $E = E_{\text{app}} = 4.9 \times 10^{-2}$ a.u.; (b) the generated third harmonic; and (c) the phase of the atomic dipole at the fundamental frequency; (d), (e), and (f) correspond to the case of a 25 fs pulse, with the same peak amplitude.

atom against fast ionization on the leading edge of the pulse. Our results can also be considered in the context of tunneling theory. For cw radiation, the tunneling ionization ($\gamma \ll 1$) can be described by a "rate" which is dominated by the exponential factor [19,20]

$$W \propto \exp[-2(2I_p)^{3/2}/3E]. \quad (3)$$

For long pulses, when the laser intensity varies slowly during an optical period, one may assume from above that the atomic dipole adiabatically follows the field oscillations, with no dependence on the pulse history. In that approximation, the ionization rate depends on the instantaneous value of the field envelope $E(t)$, and we may substitute $E(t)$ for E in Eq. (3). After integration of (3) over time, we find that the ionization probability is proportional to the pulse duration, which for weak fields is consistent with Fermi's golden rule. Thus the ionization probability is a function of the pulse energy (photon number), and thus for shorter pulses the ionization saturates at higher intensities than for longer pulses, as predicted previously [11].

In contrast, in the case of an ultrashort pulse (<25 fs), the ionization and harmonic generation occur within a few periods on the leading edge of the pulse. Therefore it is not precise to introduce an ionization "rate," since the ionization probability is nonlinear, and may even decrease with time (see Fig. 1 and [21]). Thus the application of a quasiclassical rate like Eq. (3) to a rapid rise-time pulse is limited. Equation (3) does not consider the shape of the incident field, including the carrier frequency, which means that it is rigorously valid only for low frequencies and long pulses [3]. Moreover, Eq. (3) gives the ionization rate averaged over a half period of the carrier wave, which cannot describe the nonadiabatic effects due to the return of the electron wave packet to the core, when the particular cycle where the harmonic generation is occurring may have a nonzero integrated electric field (rapidly rising leading edge). In order to observe the pure nonadiabatic response, we calculate the unsaturated ionization of an atom irradiated by an exponentially increasing pulse given by $E(t) = E_0 e^{t/T} \sin(\omega t)$, where ω corresponds to 800 nm, and E_0 is chosen to ensure equal incident energy for different values of the rise time T, for each fixed number of periods in the pulse. Here equal energy means that the time integral of the square of the field has the same value in each case. First, we show in Fig. 2(a), that for a half period of the carrier wave, the ionization is almost independent of the rise time T. Then, Fig. 2(b) shows the case of an incident field consisting of one full optical period. The ionization in Figs. 2(a) and 2(b) differ since the pump field which ensures equal energy differs in time in all cases. The effect of ionization suppression due to the nonadiabatic response of the dipole can be seen by comparing the ionization of the 5 fs rise-time pulse with that of the 50 fs case, even though we have irradiated the

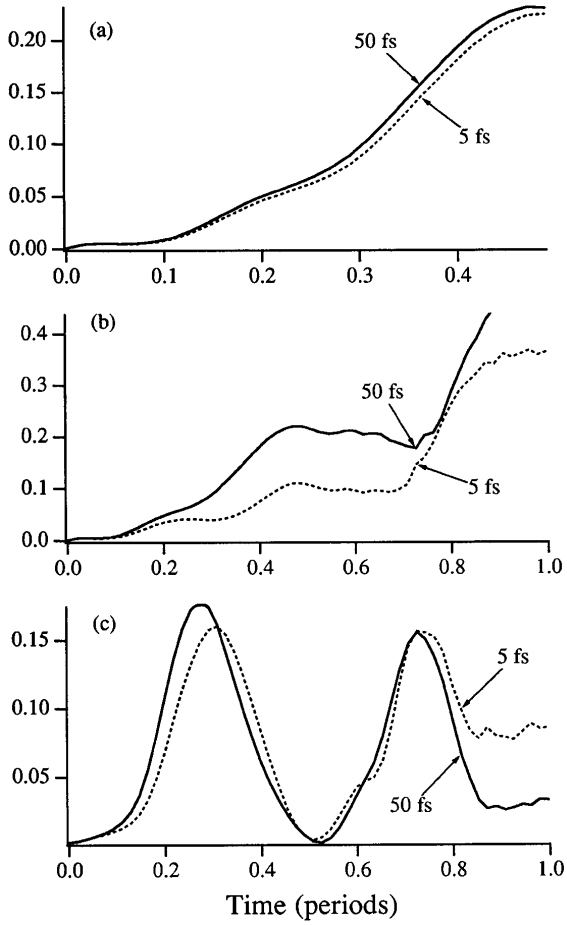

FIG. 2. Ionization probability of the model atom irradiated by an exponentially increasing field which ensures the same energy for a given number of cycles but different rise times T. (a) One half period: $T = 50$ fs (solid), $T = 5$ fs (dashed); (b) one full period: $T = 50$ fs (solid), $T = 5$ fs (dashed); (c) population of the first excited state for 50 fs (solid) and for 5 fs (dashed) rise times.

atom with the same "number of photons" in the same period of time. This difference disappears when 15 fs pulses are compared with 50 fs pulses. It is important to point out that this effect of ionization suppression appears when there is a significant ionization (>10%) during the first half period of the field, which ensures that there is sufficient penetration of the electron wave packet into the Coulomb barrier. Since the atom responds to each individual half period of the increasing electric field, the different depth of penetration of the electron wave packet into the potential barrier due to the large difference in amplitudes of the subsequent half periods leads to an increasing delay of dipole response with shorter pulses (<25 fs), and to a decrease of the total ioniation.

In frequency space, the short rise time and broad spectrum of the pulse leads to population of intermediate states. This defies the adiabatic assumption that only the initial and final states of the electron are significant in the tunneling process, while the intermediate states play

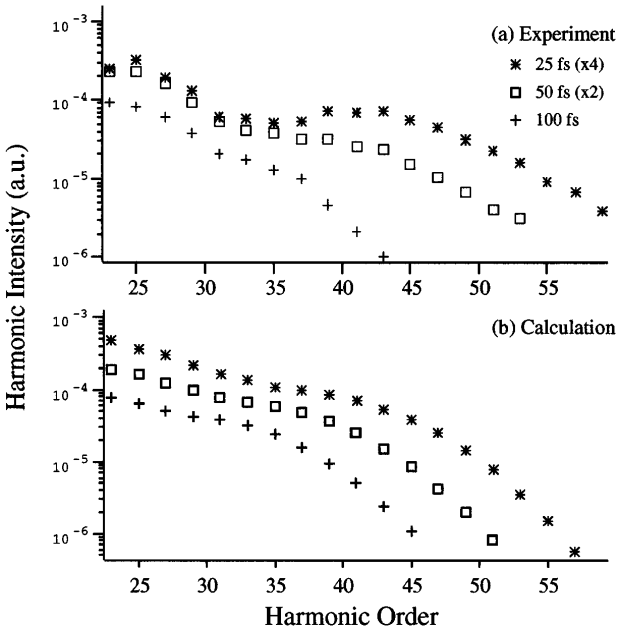

FIG. 3. (a) Experimental and (b) theoretical results for high-harmonic generation in argon by 25 fs (*), 50 fs (□), and 100 fs (+) excitation pulses, with a peak intensity of 4×10^{14} W/cm^2.

no role [3,19]. We found that the population of the first excited state for rise times of 5 fs is ≈9%, while for rise times of 50 fs it is less than 3.5% [see Fig. 2(c)]. Thus for very short rise times, a portion of the population is captured in intermediate states, which again leads to suppression of the ionization of the atom.

Figure 3(a) shows the experimental results for high-harmonic generation in argon by pulses of durations 25, 50, and 100 fs, and with a fixed peak laser intensity of 4×10^{14} W/cm^2. The setup is described elsewhere [9,14]. The pulse duration was varied by varying the bandwidth of the laser to obtain near transform-limited pulses with varying duration. The results of the corresponding simulations are shown in Fig. 3(b). This simulation was done using the soft Coulomb potential $-1/(1.40 + x^2)^{0.5}$, which has a core-peaked ground state with a binding energy the same as that for argon, 0.58 a.u. Both experiment and theory clearly show an increase in number of harmonics with decreasing pulse duration. A comparison of these results, and those of others [1,2] made with longer pulses and much higher peak intensity, shows that the higher saturation intensities achieved by using shorter excitation pulses results in higher ponderomotive energy and more intense higher harmonics than with longer pulses. Thus the nonadiabatic behavior of the atom when illuminated by very short, intense light pulses offers the possibility for greatly enhanced generation of short wavelength radiation.

In conclusion, we have shown that we can generate significantly more high-harmonic orders using very short excitation pulses, with rise times under 100 fs. This enhanced emission is a result of the rapid rise time of the pulse, which in the presence of the rapidly increasing field and the nonadiabatic atomic dipole response, allows the atom to survive to higher laser intensities. Thus our results indicate that the laser pulse shape itself can be used to control the ionization of atoms. It is likely that, with pulses of duration ≈10 fs, significant soft-x-ray flux in the "water window" (>270 eV) may be generated.

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