Recollisions, bremsstrahlung, and attosecond pulses from intense laser fields

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We investigate quantum-mechanical effects in the recollision picture of high-harmonic generation, relate these to its classical counterpart, and discuss the generation of attosecond pulses from recollision bremsstrahlung. [S1050-2947(96)50405-1]

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Over the last two decades the interaction of atoms with intense laser light has been a rich area of research with many interesting and important discoveries. One of these major findings has been high-harmonic generation (HHG); in this process the strongly driven atomic system reradiates energy at odd multiples (owing to the symmetry of the process) of the driving frequency. Using HHG it has been possible to generate very-short-wavelength light [1] with excellent coherence properties (which is a consequence of the generation process). This has attracted a great deal of interest because of the many possible applications of such light; for example, in atom lithography or imaging of biological samples [2]; one current goal is to provide a sufficiently intense source of these photons.

All HHG spectra show the same generic behavior: there is a sharp fall from the driving frequency to a plateau, followed by a cutoff. The position of the cutoff has received much attention as it essentially determines the maximum frequency that can be emitted. A quasiclassical approach involving tunnel ionization, ponderomotive acceleration, and recollision of the electronic wave packet with the parent ionic core, proposed by Kulander *et al.* [3] and by Corkum [4] predicts the cutoff energy to be

$$E_c = I_p + 3.17U_p, \tag{1}$$

where I_p is the ionization potential of the atom and U_p the ponderomotive energy of the electron in the incident field. It was specifically found that this empirical law was independent of the form of the potential and so was only really dependent on the form of the field [recently it has been found that by adding a second commensurate frequency it is possible to extend the cutoff energy to beyond that defined in Eq. (1) [5]]. This classical recollision picture has since been extended to a fully quantum-mechanical treatment of the electron motion in the laser field; according to this treatment the harmonics are emitted at every recollision [6]. However, since the time scale for a ‘‘single’’ recollision is only a fraction of the fundamental laser period, problems arise with the onset of coherence in the emission of radiation.

In this Rapid Communication we formulate a mathematical model in which the radiation that is emitted during each recollision (on an attosecond time scale) is simply bremsstrahlung with an appropriate cutoff. The harmonics emerge from the broad bremsstrahlung spectrum through the interference in time of all the single encounters within the incident pulse duration. We investigate the role of quantum-mechanical effects in the recollision picture of HHG. We first construct a simple model, and compare its predictions with those from both classical and quantum-mechanical wave-packet treatments. In our approach, the ultrashort time scale of the radiation emerges from a single encounter of the wave packet with the core; such an encounter generates a broad spectrum with no harmonic substructure. Other approaches [7] have centered on the utilization of the whole ‘‘harmonic comb.’’

The recollision picture is quasiclassical in that the ionization process is purely quantum mechanical (tunneling ionization [8]); however, once ionized the electron wave packet is treated as a free electron in a laser field. Harmonics are then generated by a sequence of single collisions of these electron wave packets formed by the tunneling process near the peaks of the incident laser electric field. One has for the complete electron acceleration

$$a(t) = \sum_{l=1}^{2N} a_l(t), \tag{2}$$

where $2N$ is the total number of collisions during an N-cycle laser pulse. The individual component accelerations $a_l(t)$ over half a cycle and including one recollision are assumed to be identical, but with different phases due to the different birth times of the wave packets, i.e., $a_l(t) = a_B(t)e^{il\pi}$, for $t_{l-1} < t < t_l$, and zero otherwise. The lth collision occurs between the times t_{l-1} and t_l, where $t_l = lT/2$ and T is the laser period. The Fourier amplitude from the initial time t_0 to the final time t_f is then

$$\tilde{a}(\omega) = \int_{t_0}^{t_f} dt\ e^{i\omega t} a(t) = \sum_{l=1}^{2N} e^{i\omega t_{l-1}} e^{il\pi} \tilde{a}_B(\omega). \tag{3}$$

Here, $\tilde{a}_B(\omega)$ is the Fourier amplitude of the acceleration during a single collision event,

$$\tilde{a}_B(\omega) = \int_0^{T/2} d\tau\ e^{i\omega\tau} a_B(\tau), \tag{4}$$

i.e., it is the strength of the bremsstrahlung radiation [9] at frequency ω. Then the spectrum is

$$S(\omega) = |\tilde{a}(\omega)|^2 = 2N|\tilde{a}_B(\omega)|^2 \times \left\{ 1 + \frac{1}{N} \sum_{\substack{n,m=1 \\ n<m}}^{2N} \cos\left[\left(\frac{\omega}{\omega_L} + 1 \right)(n-m)\pi \right] \right\}, \tag{5}$$

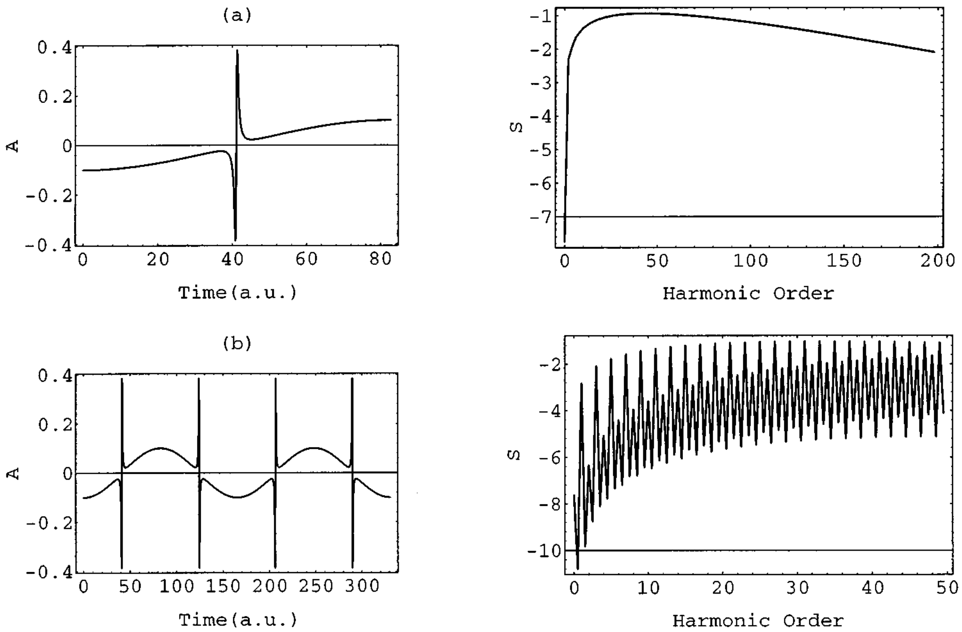

FIG. 1. Dipole acceleration of a single electron (A) with the corresponding power spectrum (S) on a logarithmic scale for (a) one and (b) four encounters with the nucleus. The electric field strength was 0.1 a.u., the angular frequency 0.038 a.u., and the electron was initially placed 70 a.u. away from the nucleus with zero velocity. Note that for all the power spectra the fundamental frequency directly from the laser field has been removed. a.u. stands for atomic units.

where $\omega_L = 2\pi/T$ is the frequency of the incident laser. For odd harmonics, $\omega/\omega_L = 2s+1$, where $s = 0,1,2, \ldots,$ one finds maxima $S[(2s+1)\omega_L] \sim N^2|\tilde{a}_B(\omega)|^2$, while for even harmonics the sum in Eq. (5) tends to zero for large N, yielding $S(2s\omega_L) \sim N|\tilde{a}_B(\omega)|^2$. This would mean that as the number of collisions increases then the spectrum would become better defined.

Next we show how the above intuitive picture is verified by both classical and quantum simulations of the laser-atom interaction in the tunneling regime. An interesting way of obtaining an insight into the dynamics of the recollision picture is to eliminate the ionization process and start with a wave packet initially at some distance away from the *bare* atomic core. We then follow this system under the influence of a monochromatic laser field. We first investigate this by using a simple classical one-electron system and calculating its dipole acceleration and then its spectrum. In Fig. 1 we consider the dynamics of a single classical electron that is initially displaced some distance away from the atomic core with zero velocity and we calculate the spectrum after 1 and 4 collisions. Clearly, as N increases then the HHG spectrum becomes sharper and more defined, as would be expected from our previous calculation. The spectrum of N collisions by N *different* electronic wave packets produced by periodic tunneling events is equivalent to one electron wave packet recolliding N times. This is a subtle fact that is often overlooked and can lead to misinterpretion of results. A single encounter generates a bremsstrahlung supercontinuum with a broad width and an associated ultrashort temporal duration (the electronic recollision time with the core). The bremsstrahlung cuts off at a frequency given by the maximum quiver energy [9].

To model a wave packet truly, an ensemble of these individual classical electron trajectories must be considered and the results averaged [10]. This approach is able to model wave-packet spreading, but is, of course, unable to simulate quantum-mechanical interference effects. We have calculated the classical HHG spectrum for $E = 0.1$ a.u., $\omega = 0.038$ a.u. and an initial Gaussian wave packet simulated by an ensemble in phase space centered at 70 a.u. away from the core in the spatial direction with width 10 a.u., with zero momentum and width 0.1 a.u. in momentum space. The field is phased such that it initially accelerates the electron towards the core with maximal strength. This produces a spectrum with an exponential decrease in harmonic strength up to the 20th order, followed by a plateau that has a cutoff at $I_p + 2U_p$ (the 91st harmonic). However, the cutoff is far less pronounced than in the quantum-mechanical calculations that follow.

To study the importance of quantum-mechanical interference, we have chosen to integrate the one-dimensional Schrödinger equation using a well-known method based on the unitary transformation into the Kramers-Henneberger frame, and the smoothing of the Coulomb potential [11]. Assuming the same parameters as for the classical case and the same initial conditions, we find an interesting wave-packet evolution as a function of time (Fig. 2). As expected, the wave packet spreads and encounters the core; however, for the first few encounters the quantum-mechanical interference is not very large and these collisions are ‘‘classical-like.’’ However, once the wave packet has spread enough, the core interaction leads to the wave packet acquiring sharp, fine-structured features. The underlying physics of the additional structures can be understood as arising from the inter-

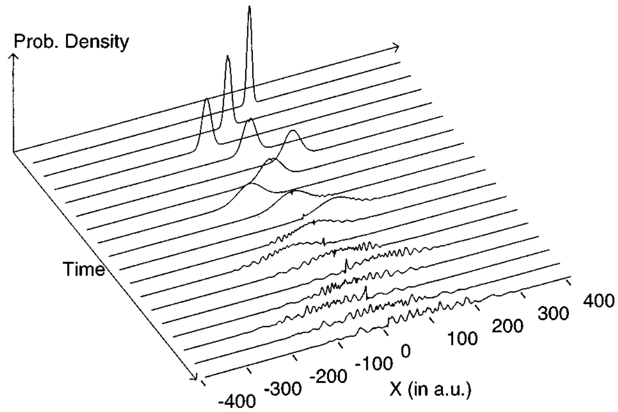

FIG. 2. Quantum-mechanical evolution of the wave packet for the same parameters as for Fig. 1, but taken over four full cycles of the incident laser field. The first snapshot is the initial wave packet of width 10 a.u., and consequent snapshots are taken every quarter of a cycle.

ferences between those parts of the wave packet that are *reflected* by the core and those parts that are still incoming. This is a purely quantum-mechanical effect and is missing from the previous classical treatment. Its implications are quite important and will be discussed in what follows.

In order to demonstrate the importance of quantum-mechanical interference, we show in Fig. 3 the calculated dipole acceleration. It is easy to see that the first three collisions resemble quite closely those of the single-electron trajectory of Fig. 1, with, however, a fast amplitude decay due to the wave-packet spreading. If one takes a windowed Fourier transform of the first collision and the associated power spectrum, one sees just a bremsstrahlung supercontinuum spectrum [Fig. 3(a)]. After the second and third collisions we see the beginnings of harmonic structure [Fig. 3(b)], but only for the low-order harmonics (again the wave packet spreading is the cause for this classical bremsstrahlung mechanism being inefficient). Only when the fast frequency components of the dipole acceleration are included do we begin to see anything resembling a true harmonic spectrum [Fig. 3(c)]. If we take the spectrum of the whole dipole acceleration, this now has all the features associated with normal harmonic generation [Fig. 3(d)]. It is possible to observe, in the progression from Fig. 3(a) to Fig. 3(d), how the overall efficiency is enhanced, producing a very *clear* cutoff around the expected harmonic order.

It would seem that to obtain a ''good'' harmonic spectrum it is important to have these fast oscillations in the dipole acceleration. The question is, how do they occur? The answer lies in the wave-packet evolution (Fig. 2) and the form of the dipole acceleration operator [12],

$$\langle a(t)\rangle = -\int \frac{\partial V(x)}{\partial x}|\psi(x,t)|^2 dx, \tag{6}$$

where $V(x)$ is the atomic potential and $\psi(x,t)$ the wave function. A highly structured spatial wave packet generates via Eq. (6) high temporal frequencies [13] in the radiated field. Therefore, any wave packet with many features will produce an acceleration that has fast oscillations and, consequently, efficient harmonic generation. These spatial features only occur when the previously mentioned interference between incoming and outgoing parts of the wave packet takes

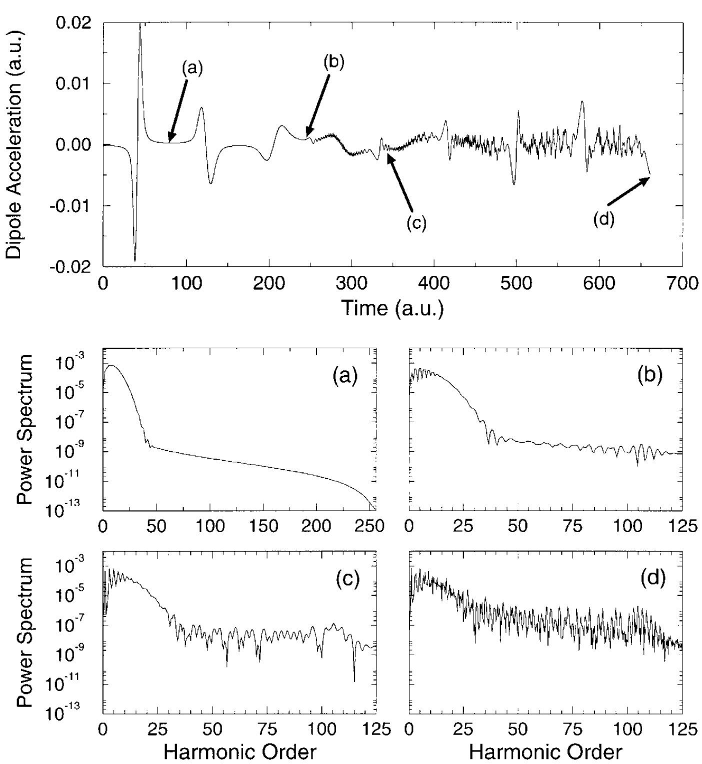

FIG. 3. Dipole acceleration (top) and the corresponding power spectra at various points in time as indicated by the labels (a)–(d). The parameters are the same as in Fig. 2.

place. This idea can be corroborated by observing that the fast oscillations in the dipole acceleration appear at the same time as the spatial features in the wave packet. If it were possible to control this interference it would provide a very powerful way of controlling the harmonic spectrum.

A further possibility is to engineer a single encounter (using time-dependent ellipticity of the superintense light [7]), which leads to a broadband bremsstrahlung supercontinuum generated within the attosecond time scale of the recollision. We have shown that a single encounter certainly generates a bremsstrahlung continuum, but with a spectral envelope that is *not* precisely that of the HHG comb of frequencies. Multiple reencounters and interference are responsible for increasing the spectral weight around the cutoff. Nevertheless, a single encounter has, of necessity, the attosecond time scale relevant to the recollision process. The duration of emission of the radiated field, whose intensity is given by the square of the acceleration in Eq. (6), is directly related to the time length of the single electron-core encounter. The brevity of the emission process can be read off directly from the time dependence of the acceleration shown in Fig. 3 (one atomic unit of time is 24 attoseconds). Narrow wave packets take substantially less time to traverse the core and if constructed in an appropriate way can produce emission over an extremely short time scale. Therefore, *only* sufficiently narrow wave packets can yield attosecond pulses. We address in a later paper the question of phase matching of this supercontinuum in a true macroscopic source.

In summary, we have demonstrated that classical dynamics associated with the free-electron recollision picture is capable of reproducing many aspects in the harmonic spectrum. It reproduces the increased clarity of the harmonics with a rising number of recollisions and simulates the wave-packet spreading. However, it cannot, naturally, take into account the quantum interferences within the wave packet. These interferences are quite important for the very high harmonics, as we have pointed out, and lead to significant differences between the classical and quantum results.

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