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Harmonic generation by a classical hydrogen atom in the presence of an intense radiation field

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We present the results of a calculation of high-order harmonic generation by a classical hydrogen atom, submitted to an intense, single-mode, radiation field. The spectra showing these harmonics associated with different kinds of trajectories as well as with an ensemble average of trajectories are discussed.

Classical mechanics has proven to be a valuable tool in the study of the response of excited hydrogen atoms in microwave fields.[1-3] It has been observed also that, as a result of the scaling properties of the classical hydrogen atom,[1] this analysis of the atom-radiation processes can in principle be extended to the low-lying atomic states, provided the field frequency and intensity are properly modified.[4] Although such approaches are unable to account for details resulting from the quantal properties of real atoms, they are expected to provide interesting insights on the dynamics of such processes, especially in the limit of very strong fields, i.e., when the atom-field coupling will be comparable to the atom's potential energy. This remark has given rise to several classical-mechanical studies of the strong-field ionization of hydrogen,[5-7] and multielectron atoms.[8] The purpose of this Rapid Communication is to report on a calculation of higher-order harmonic generation by a classical hydrogen atom, driven by an intense monochromatic (classical) field.

High-order harmonic generation has been observed recently when relatively dense samples of rare-gas atoms are irradiated by strong laser radiation.[9-11] Generation of harmonics up to 33rd order has been reported in the (relatively) low-frequency regime (Nd laser, $\omega = 1.17$ eV $= 0.043$ a.u.). Although collective effects appear to be essential to account for the properties of the emitted light, the fundamental process originates at a microscopic level. Lowest-order perturbative,[12-14] as well as nonperturbative, either Floquet-like[15] or numerical,[16] quantal treatments have been reported. We present here a classical, in essence nonperturbative, counterpart of these calculations.

Our analysis is based on the well documented Monte Carlo classical trajectory method.[1] It allows us to follow the time evolution of an ensemble of atomic dipole moments, with z component $\langle \mu_z \rangle$, evolving from an initial three-dimensional microcanonical distribution associated with ground-state hydrogen atoms. The switching on of the laser is simulated by a linear ramp, with the maximum field intensity being reached at $t \sim 300$ a.u. (1 a.u. $= 2.42 \times 10^{-17}$ s). Given the laser frequency considered here, $\omega_L = 0.043$ a.u., the maximum intensity is reached after about two laser cycles. At this frequency, such that $\omega_L \ll 1$ a.u., we are in the adiabatic domain, and accordingly, transient contributions are expected to be small, even in the case of a sudden turn on of the field. After the initiation stage, the intensity is kept constant for a large number of laser cycles. We then compute the power spectrum, i.e., the time average of the square of the Fourier transform of $\langle \mu_z \rangle$, which provides us with the spectrum of frequencies emitted by the ensemble of atomic dipoles. Specifically,

$$D(\omega) = \lim_{T \to \infty} \frac{1}{T} \left| \int_0^T \langle \mu_z(t) \rangle e^{i\omega t} dt \right|^2 . \quad (1)$$

This analysis furnishes the relative intensities of the generated harmonics.[16] Note that we have sampled our Fourier transforms over large enough numbers of cycles in order to avoid the occurrence of transients induced by the initial response to the field. As discussed next, in addition to considering the ensemble average, we have also considered the spectra associated with individual trajectories.

Without external field the ground-state electron follows a periodic Kepler orbit confined in a region of space of about 2 a.u. around the nucleus.[17] In the presence of the laser field, the movement of the electron is no longer confined to the immediate vicinity of the nucleus. Depending on the initial conditions and field strength intensity, it can experience different kinds of trajectories, as described in Ref. 1. Some of them can lead to the ionization of the atom, ionizing trajectories being associated with asymptotically positive compensated energy[1] (atomic energy corrected for the quiver motion) states: The electron moves far apart from the nucleus, and in the asymptotic regime, oscillates at the laser frequency.[18] It is also known that even at quite large field intensities (typically a fraction of atomic unit) some of the trajectories remain bound for a surprisingly long time. In the context of the current discussion on the possible relation between harmonic generation and above threshold ionization,[15,19] it was of interest to compare the spectra associated with these different kinds of trajectories.

Shown in Fig. 1 are the temporal variations of the atomic dipole moment μ_z along the laser polarization direction, associated with a typical trajectory which leads to ionization at $t \sim 7000$ a.u., i.e., after ~ 48 laser cycles at $\omega_L = 0.043$ a.u. For short times, namely during the turn on of the field, the dipole still oscillates at the atomic frequency ($T = 2\pi$ a.u.). Then the presence of the intense external field [here $F = 0.15$ a.u. (7.875×10^{14} W cm^{-2})] destroys the coherence of the fast atomic oscillations and superimposes a slower periodic motion at the laser frequency.

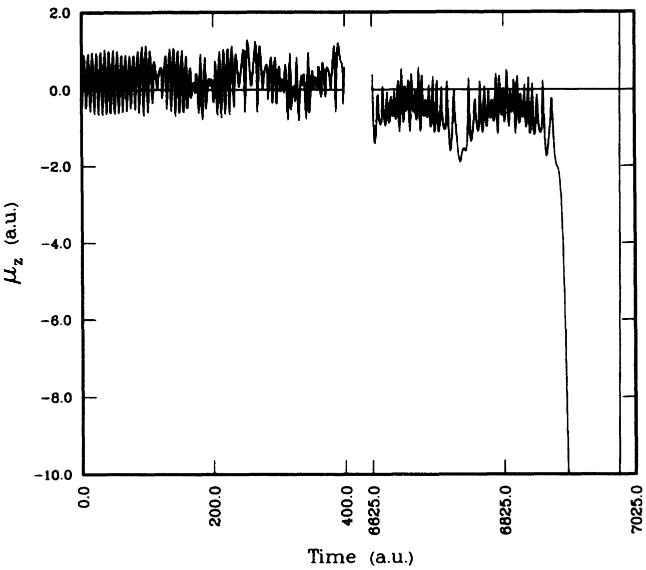

FIG. 1. Temporal variations of the atomic dipole μ_z moment of a trajectory along the laser polarization direction: $\omega_L = 0.043$ a.u., $F = 0.15$ a.u.

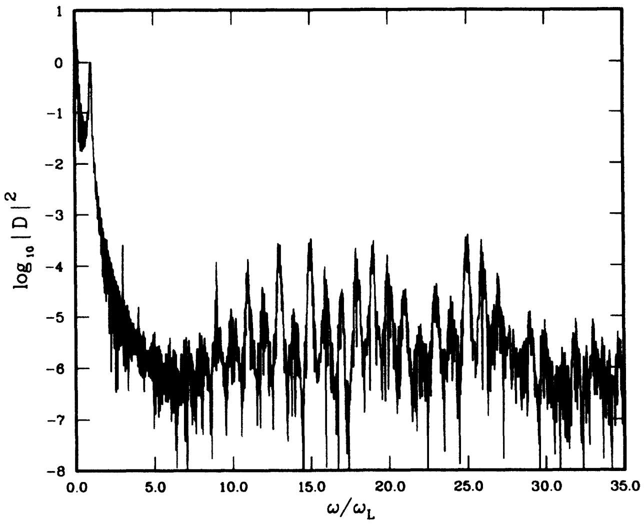

FIG. 3. Power spectrum of μ_z corresponding to the bound motion plus some unbound motion of the trajectory in Fig. 1. Note the growing continuous background at the lower end of the spectrum.

The power spectrum corresponding to the nonionizing part of this trajectory [i.e., with T in Eq. (1) equal to the time at which ionization occurs, see Fig. 1] is displayed in the Fig. 2. The spectrum is dominated by a Rayleigh scattering component at the laser frequency $\omega = \omega_L$ and one observes the presence of harmonics of the laser frequency up to orders larger than 30. Even harmonics are also present since such a trajectory does not possess an inversion center. One does not observe any significant component at the characteristic atomic frequency $\omega_{at} \sim 23.4\omega_L$, which confirms that at such a field intensity the system rapidly loses the memory of its initial atomic motion.[20]

The inclusion of the ionizing part of the trajectory entails the appearance of a continuous background in the power spectrum as well as a dominant component at the laser frequency corresponding to Rayleigh scattering (see Fig. 3). The occurrence of this background corresponds to the contribution of the linear component of the motion of the asymptotically free electron on its way far from the nucleus. Note that if one follows such an ionizing trajectory over a large number of laser cycles, the corresponding spectrum is dominated by this continuous background which ultimately washes out the harmonics. Such an observation supports the view that harmonic generation is more efficient for bounded trajectories or, more precisely, when the electronic motion is close to the nucleus that it

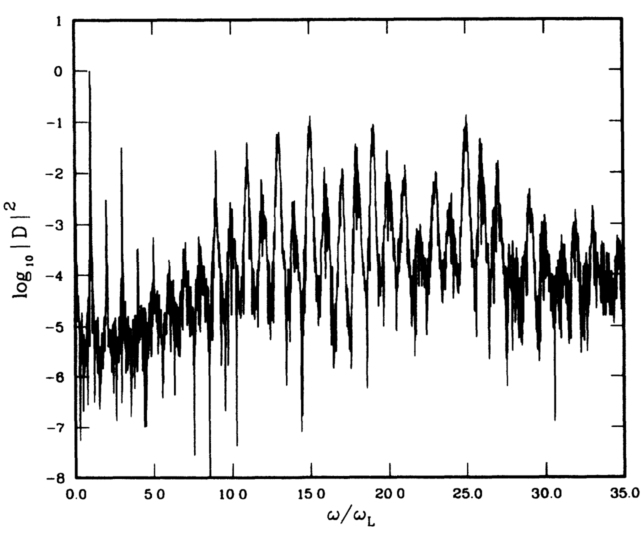

FIG. 2. Power spectrum of μ_z corresponding to the bound motion of the trajectory in Fig. 1. Spectrum is normalized with respect to the first peak.

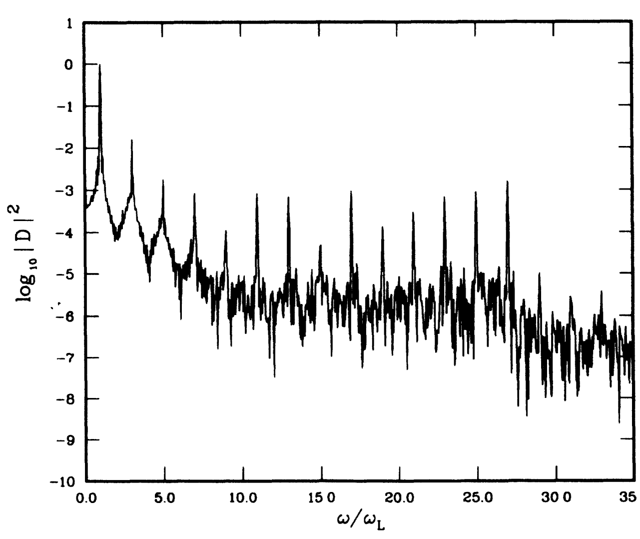

FIG. 4. Harmonic spectrum of a ground-state hydrogen atom in an intense laser field. 10000 Monte Carlo trajectories were used: $\omega_L = 0.043$ a.u., $F = 0.15$ a.u.

suffers sufficient acceleration to generate the harmonics.

The power spectrum associated with an ensemble of atomic dipole moments, averaged over 10000 trajectories, is shown in Fig. 4. The initial conditions have been chosen at random as usual,[1] with the additional constraint that the average dipole moment was zero at $t=0$, in order to mimic the spherical symmetry of real H atoms. Performing the ensemble average allows us to get rid of the unphysical even harmonics. As expected, increasing the number of trajectories reduces the relative contribution of the ionizing background. We have, however, discarded the contribution of the ionizing parts of the trajectories (such that the compensated energy is positive *and* the distance to the nucleus $r > 20$ a.u.) in order to reduce the computing time.

A remarkable result of this classical analysis is the presence of a broad plateau between the harmonics 7–27, followed by a significant drop of 2 orders of magnitude in intensity. Although we have not attempted here to draw quantitative conclusions from such a classical model, it is interesting to note that this result seems to parallel the main features of the quantal calculations.

For computational convenience the results presented in this Rapid Communication correspond to a quite high intensity regime, such that the laser electric field strength is $F=0.15$ a.u. This allowed us to keep the computational effort at a relatively modest level. We have checked, of course, that high-order harmonic generation takes place also at much lower intensities: One has, however, to perform the ensemble average over more trajectories in order to suppress the even harmonics. At still lower intensities one notes also a component at the characteristic atomic frequency (based on the period of the orbit) $\omega_{\text{at}}=1$ a.u. $\sim 23.4\omega_L$, corresponding to the survival of the initial atomic motion.[20] In the scattering of light from an ensemble of atoms the component due to initial oscillations at the atomic frequency is usually referred to as the "redistributed or fluorescent component" in contrast to Rayleigh scattering which occurs at the laser frequency. A more detailed discussion of these peculiarities of the classical model will be published elsewhere.

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