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High-order harmonic generation from a confined atom

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The order of high harmonics emitted by an atom in an intense laser field is limited by the so-called cutoff frequency. Solving the time-dependent Schrödinger equation, we show that this frequency can be increased considerably by a parabolic confining potential, if the confinement parameters are suitably chosen. Furthermore, due to confinement, the radiation intensity remains high throughout the extended emission range. All features observed can be explained with classical arguments.

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Typical features of the emission spectra of an atom in a strong laser field, known as ''the plateau'' and ''the cutoff,'' are a wide frequency region with harmonics of comparable intensities, and an abrupt intensity decrease at the high-energy end of the plateau. For a monochromatic driving field, the cutoff energy is given by $\varepsilon_{\max}=|\varepsilon_0|+3.17U_p$, where $|\varepsilon_0|$ and U_p are the ionization potential and the ponderomotive energy, respectively. This simple cutoff law, derived by classical means only [1,2], or using more refined methods [3], corresponds to the physical picture referred to as the ''three-step model'' [1–3]: A bound electron exposed to the laser field leaves the atom through tunneling at a time t_0 (step 1), propagates in the continuum, being driven back towards its parent ion at a later time t_1 (step 2), and finally falls back preferentially to the ground state under emission of high harmonics (step 3). This scenario describes the spectral features observed experimentally very well [4]. The cutoff frequency, in quantitative agreement with the experiment, is related to the maximum kinetic energy the electron has upon return, $E_{\rm kin}(t_1,t_0)$.

According to this picture, in order to increase the cutoff energy, one must increase the kinetic energy of the returning electron. Indeed, the existing proposals to extend the plateau towards higher energies reach a higher value of $E_{\rm kin}(t_1,t_0)$ by different means. However, this does not necessarily imply an efficient generation of high-order harmonics up to this larger cutoff energy.

For instance, a rather complex situation with several ''cutoffs'' [5] emerges by using bichromatic fields with driving waves of comparable intensities. An illustrative example is presented in [6], using a driving field of linearly polarized monochromatic light of frequency ω and its second harmonic. Under such conditions the monochromatic cutoff, as a function of the field-strength ratio between the two driving waves, splits into two branches. Thereby, the upper branch extends up to $|\varepsilon_0|+5U_p$. However, the harmonics emerging up to the cutoff of the upper branch are weak compared to those from the lower branch and therefore irrelevant to the emission spectrum. The reason is simple: The intensity of the harmonics is strongly influenced by step 1, which is the tunneling process out of the binding potential under the influence of the field. If the field amplitude is small at the emission time t_0 (which is the case for the upper branch), then the tunneling barrier is large and the generated harmonics will be weak compared to those that originate from an effective tunneling process (as is the case for the lower branch).

Another idea to increase the cutoff energy is to use a static electric field. It provides an additional force that accelerates the electron towards the atomic core, resulting in a higher kinetic energy $E_{\rm kin}(t_1,t_0)$. Indeed, it has been demonstrated that with an electric field whose strength is only a few percent of the amplitude of the laser field one can considerably enlarge the cutoff energy [7,8]. However, the scheme suffers from two principal limitations. First, the increased kinetic energy occurs mainly for electrons with long excursion times. Due to wave-packet spreading, the overlap between the returning electronic wave packet and the ground-state wave function is not significant, such that those trajectories have negligible influence on the harmonic spectra. This problem has been overcome by introducing an additional magnetic field to restrict the spreading [8]. A second, more severe limitation is the pronounced bound-state depletion caused by the static electric field: the atom is irreversibly ionized within a few field cycles, such that no appreciable high-harmonic generation takes place.

The bound-state depletion that prevents an effective extension of the high-harmonic frequency points to the principle dilemma easily described in the picture of the returning electron: To extend the plateau and increase the cutoff, a kinetic energy of the returning electron, as large as possible, is desirable. On the other hand, an electron with such a high energy will leave the atom and will be lost for the possible generation of high harmonics in consecutive laser cycles.

Hence, we need a mechanism that brings an electron back to the nucleus, despite the fact that it has a kinetic energy so high that it would be irreversibly driven away from the core. Naively, a simple wall for the electron should already do this. However, one must avoid the abrupt reflection of the charged electron at a wall, since this would lead to bremsstrahlung that masks the desired high-order harmonic generation of the atom [9].

In the following we will show that the idea of bringing back the fast electron by an additional confinement and thereby extending the cutoff for the spectrum without additional depletion does indeed work for a suitably soft confinement potential.

We consider a one-dimensional situation, which is a reasonable approximation for linearly polarized light. Atomic units are used throughout. The binding of the electron is described by the potential

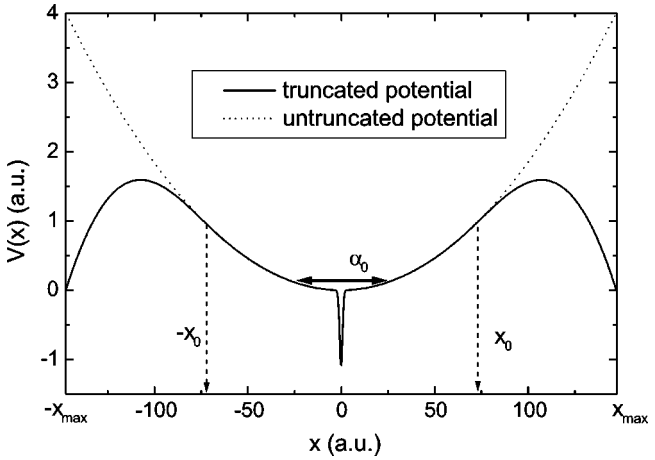

FIG. 1. Schematic representation of an atom in an external confining potential $V_h(x)$ [cf. Eq. (2a)]. The parameter x_0 for which the potential is truncated and the electron excursion amplitude α_0, for the parameters of Fig. 2(a), as well as the nontruncated potential, are indicated in the figure.

$$V_a(x) = -1.1\exp(-x^2/1.21), \tag{1}$$

which supports a single bound state $|0\rangle$ at energy $\varepsilon_0 = -0.58$ a.u., corresponding to the argon ionization potential. The system is exposed to a monochromatic laser field $E(t) = E_0 \sin \omega t$ and the additional harmonic potential (Fig. 1)

$$V_h(x) = \frac{\Omega_h^2}{2} x^2 h(x), \tag{2a}$$

$$h(x) = \begin{cases} 1, & |x| < x_0, \\ \cos\left(\dfrac{\pi}{2}\theta\right), & x_0 \leqslant |x| \leqslant x_{\max}, \\ 0, & |x| > x_{\max}, \end{cases} \tag{2b}$$

with $\theta = (|x| - x_0)/(x_{\max} - x_0)$, truncated at $x = x_0$. Setting $h(x) = 1$ in Eq. (2a) gives the corresponding non-truncated potential. The parameter $x_0 = nE_0/\omega^2$ is chosen to be a multiple of the electron excursion amplitude, and $x_{\max} = 2x_0$. Parabolic potential shapes are taken as a first approximation in several physical systems, as for instance electromagnetic traps [10] or solid-state devices [11]. Note that, for the parameter range chosen, identical emission spectra are obtained with and without truncation of the harmonic potential, indicating that even in the truncated potential depletion has a negligible influence. Thus, the electron does not reach the edges of $V_h(x)$, which indicates an effective confinement. Furthermore, this shows that the confining potential does not generate harmonics itself. Therefore, high-harmonic generation still takes place only near the atomic core, for which the coordinate x is considerably smaller than the electron excursion amplitude.

The evolution of the electronic wave packet is described by the time-dependent Schrödinger equation

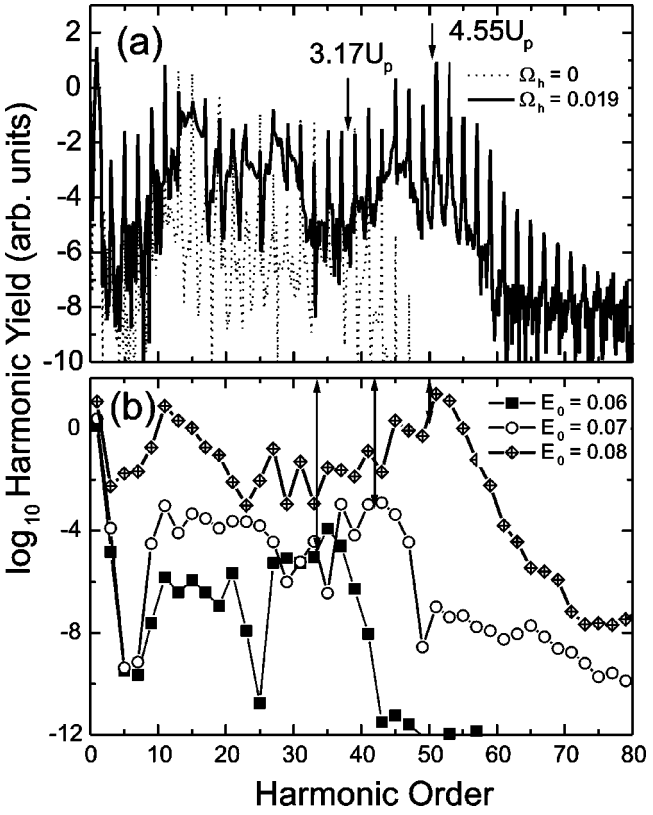

FIG. 2. Harmonic spectra calculated using the time-dependent Schrödinger equation (TDSE) [cf. Eq. (4)]. (a) Field amplitude $E_0 = 0.08$ a.u., field frequency $\omega = 0.057$ a.u., without (dashed line) and with (solid line) confinement ($\Omega_h = 0.019$ a.u.). (b) Field strengths $E_0 = 0.06$ a.u., $E_0 = 0.07$ a.u., and $E_0 = 0.08$ a.u., confinement curvature $\Omega_h = 0.019$ a.u., and the same frequency as in the previous part. The classical cutoff energies, given by the cutoff law $|\varepsilon_0| + 4.55U_p$, correspond to the harmonic orders $n = 33$, $n = 41$, and $n = 49$ and are indicated by arrows in the figure. In part (b), only the harmonic intensities are given, connected by lines.

$$i\frac{d}{dt}|\psi(t)\rangle = \left[\frac{p^2}{2} + V(x) - pA(t)\right]|\psi(t)\rangle, \tag{3}$$

with $V(x) = V_a(x) + V_h(x)$, and the emission spectra are given by

$$\sigma(\omega) = \left| \int_0^\infty d(t)\exp[-i\omega t] \right|^2, \tag{4}$$

where the dipole acceleration

$$d(t) = \langle \psi(t)| -dV(x)/dx + E(t)|\psi(t)\rangle \tag{5}$$

is computed by means of Ehrenfest's theorem [9]. We take the atom initially in the ground state $|0\rangle$. Furthermore, in the results to be presented we chose $x_0 = 73.87$ a.u., which corresponds to three times the excursion amplitude of an electron in a monochromatic field with $E_0 = 0.08$ a.u. and $\omega = 0.057$ a.u. With these field parameters and a reasonable choice of Ω_h, one indeed finds that the high-order harmonic spectrum extends beyond the cutoff energy $\varepsilon_{\max} = |\varepsilon_0|$

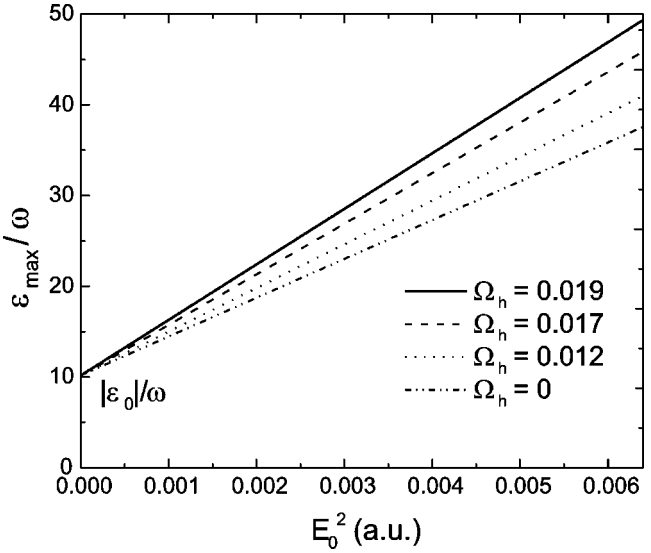

FIG. 3. Cutoff energies computed using the classical model, as functions of the field intensity E_0^2, for confinement constants $0 \leqslant \Omega_h \leqslant 0.019$ a.u. and $\omega = 0.057$ a.u.

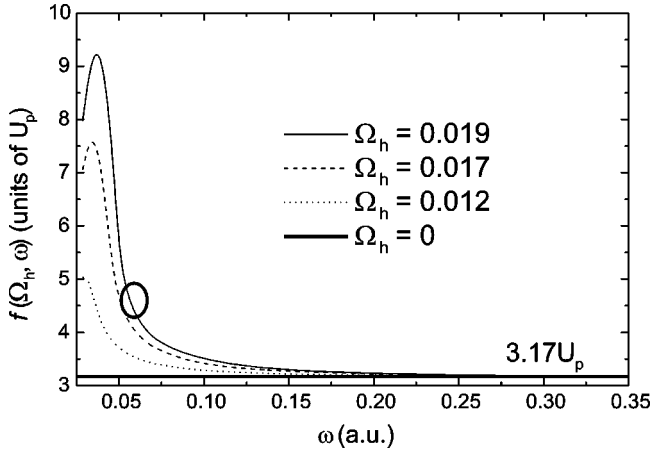

FIG. 4. Cutoff energies computed using the classical model, as functions of the frequency ω of the driving field, for confinement curvatures $0 \leqslant \Omega_h \leqslant 0.019$ a.u. The circle in the figure corresponds to $\omega = 0.057$ a.u. and $\Omega_h = 0.019$ a.u., for which the spectra in Fig. 2 have been calculated.

$+3.17U_p$ without significant loss of intensity; see Fig. 2. More specifically, we have determined a cutoff energy of $|\varepsilon_0| + 4.55U_p$, which is a 50% increase compared to the case without trapping. The classical argument for the cutoff energy applies to the situation with confinement as well and we find very good agreement between the cutoff in the quantum spectra (e.g., Fig. 2) and the classical cutoff. The latter has been determined in analogy to the situation without confinement [4,5]: Starting with an electron of velocity zero, its trajectory is propagated under the influence of the laser field and the confinement potential V_h [but without the atomic potential $V(x) = V_a$]. We vary the initial time t_0 for which the electron leaves the atom within a field cycle, computing $E_{\rm kin}(t_1, t_0)$ for return times $t = t_1$ satisfying the condition $x(t_1) = 0$. The local maxima in $E_{\rm kin}(t_1, t_0)$ yield the classical prediction for the cutoffs in the harmonic spectra.

The good agreement of the classical cutoff with the one found in the quantum spectra allows us to predict, with the classical model, the behavior of the cutoff as a function of the external parameters, i.e., the confinement constant Ω_h, the frequency and the amplitude of the external field. We find that in the parameter range of interest the cutoff law can be written in the form

$$\varepsilon_{\rm max} = |\varepsilon_0| + f(\Omega_h, \omega) U_p, \tag{6}$$

where $f(\Omega_h, \omega)$ in general neither exhibits a simple functional form nor can be derived analytically. However, the linear dependence on the field intensity E_0^2 through U_p in Eq. (6) is preserved just as in the case without confinement; see Fig. 3. Only for large confinement constants or electron trajectories with long excursion times does $f(\Omega_h, \omega)$ become slightly intensity dependent.

The general behavior of $f(\Omega_h, \omega)$ is rather complex. Nevertheless, asymptotically a simple and familiar behavior is recovered: For very high frequency, the monochromatic cutoff constant is approached, i.e., $f(\Omega_h, \omega \to \infty) \to 3.17$, as can be seen in Fig. 4. For finite frequency ω the cutoff energy increases with growing Ω_h. In fact, the lower the frequency, the more sensitively the cutoff law depends on Ω_h. This property is the actual reason why one can obtain an increased cutoff energy with a confinement. For very low frequencies, the cutoff energy can be easily extended beyond $|\varepsilon_0| + 9U_p$. In practice, however, there is a lower frequency limit to generate an appreciable intensity of high harmonics in the present context. If the confinement frequency is comparable to the laser frequency, $\Omega_h \sim \omega$, the confinement potential itself starts to contribute to the harmonic generation process, ceasing to be a passive element. Hence, the condition for high-harmonic generation (HHG) under a confinement potential can be written as $\Omega_h / \omega \ll 1$. However, there is also the usual upper limit in frequency ω that comes from the requirement that the atom in the laser field must be in the tunneling regime [12].

Typical frequencies used in HHG experiments, and for which a long plateau is obtained, are in the vicinity of $\omega = 0.057$ a.u. For this frequency a confinement indeed leads to a larger cutoff energy, as demonstrated in Fig. 2.

In conclusion, we have presented a scheme for increasing the cutoff energy of the high-harmonic spectra of an atom under the influence of a strong laser field. Placing the atom in a confining parabolic potential, we have shown that the cutoff energy can be increased by more than 50%. An effective increase of the cutoff requires a careful choice of the confinement strength. The confinement curvature Ω_h must be strong enough for the electron to be appreciably accelerated towards the parent ion, but weak enough for it to move in a ''quasicontinuum.'' If Ω_h is too weak, the conventional cutoff law $|\varepsilon_0| + 3.17U_p$ is not altered by it. If Ω_h is too strong, the electron moves as a bound particle that does not generate higher harmonics. In the extreme case, one observes the dipole response of a harmonic oscillator, i.e., equally spaced resonances. A rough indication of whether the electron is in a ''quasicontinuum'' is given by the ratio of the energy dif-

ference between two consecutive levels of the confinement potential, $\Delta\varepsilon_h=\Omega_h$, and the ionization potential of the atom in question. If $\Omega_h/|\varepsilon_0|\ll 1$, this condition is fulfilled. Also, as already discussed, the ratio between the frequency ω of the external field and the confinement curvature Ω_h plays an important role. If $\Omega_h/\omega\sim 1$, the parabolic potential contributes too actively to the harmonic generation process, and the plateau and cutoff are not present in the spectra. The best results have been obtained for $x_0\sim 100$ a.u., $\Omega_h \sim 0.02$ a.u., and $\omega\sim 0.04$ a.u. In this case, the energy difference between two consecutive levels of the confinement potential is still of the order of one tenth of the ionization potential $|\varepsilon_0|$ and $\Omega_h/\omega\sim 0.5$. For this parameter range, the cutoff energy can be extended until approximately $|\varepsilon_0| + 6U_p$.

On a more technical level, yet very interesting from the theoretical point of view, we have seen that the cutoff law is given by the classical picture of an electron moving under the influence of the laser field and the confinement potential. Very good agreement between the quantum-mechanical full calculation and the classical model occurs for a wide range of field strengths, frequencies around $\omega\sim 0.05$ a.u., and confinement curvatures of the order of $\Omega_h\sim 10^{-2}$ a.u. Thereby we have found that the cutoff law strongly depends on the confinement curvature Ω_h and the frequency ω of the laser field, but only linearly on the field intensity E_0^2.

The proposed setup presents several advantages over the schemes using static fields. For instance, using a confining potential, one can achieve a considerable extension of the cutoff energy already for the trajectories corresponding to *short* electron excursion times, whereas using static fields one mainly affects electron trajectories with *long* excursion times. Due to wave-packet spreading, the former trajectories are far more important for the harmonic spectra than the latter. In order to reduce the spreading one needs very strong magnetic fields [8]. Another noteworthy feature of a confinement potential is that one can obtain stronger harmonics than in the static field, or even in the monochromatic case. In fact, a serious disadvantage concerning static electric fields is an appreciable decrease in the harmonic intensities compared to the field-free case, due to depletion, i.e., irreversible ionization. This problem is not present in our scheme.

However, similarly to the so far proposed extension of the cutoff energy by using a combination of a static electric field and magnetic fields, we are not aware of a direct possibility for an experimental realization of our scheme. In the former case the necessary magnetic field is unrealistically large for a laboratory application [13]. For our situation, a true electromagnetic trap is too macroscopic compared to the paramater range we need. On the other hand, there might be exciting possibilities in the future to design a confined atom as described in a quantum-dot like device, for instance as an impurity. An important issue here, however, is the limitation in the radiation intensity in order to avoid the damage threshold. Recently, solid-state materials that can survive our parameter range, namely fields of wavelength $\lambda=790$ nm and intensities above 10^{14} W/cm^2, have been observed [14].

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