Bunching and Focusing of Tunneling Wave Packets in Enhancement of High-Order Above-Threshold Ionization

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Numerical solution of the time-dependent Schrödinger equation for a model argon atom subject to an intense laser pulse quantitatively reproduces the resonance enhancements seen in the high-order part of the above-threshold ionization (ATI) electron spectrum in earlier experiments. The calculation reveals that the envelope of the ATI spectrum is due to bunching of the ionized wave packet on the first reencounter with its parent ion core, followed by backscattering of such bunches on subsequent encounters. The resonant structure is caused by interference between bunches produced in different cycles. [S0031-9007(98)06757-X]

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Photoionization of atoms by intense lasers can produce photoelectrons with energies corresponding to absorption of dozens of photons [1], even though only a few photons would be sufficient to overcome the ionization potential. At high light frequencies and low intensities the absorption of such an excess of photons by a photoelectron [known as above-threshold ionization (ATI) [2]] can be explained satisfactorily by perturbation theory [3]. At higher intensities, the perturbation series tends to diverge, especially at low frequencies ω. This reflects the physical fact that a free electron cannot resist acceleration by an electric field E. In an ac field this leads to an oscillatory ("quiver") motion of such an electron. The average kinetic energy associated with this motion (the ponderomotive energy U_P) can be many times the photon energy.

Recent high-sensitivity experiments [1,4] have revealed that the ATI spectrum extends to energies much above $2U_P$, the maximum energy that can be acquired by a photoelectron after its release in the field by the original ionization event [5]. Such fast electrons are thought to originate by backscattering from the core of their parent ion, when the electron reencounters this core in the course of its quiver motion [6]. Since photoelectron production is phase locked with the light, the electron spectrum will consist of a sequence of peaks an integer number of photon energies above the initial-state energy, but according to the correspondence principle, the envelope of the peaks can usually be well explained in terms of the underlying classical behavior.

High-order ATI has proven to be a highly structured process. The envelope of the ATI spectrum does not decrease regularly, but shows regions where the peak intensities stay roughly constant or even increase with peak order [4]. In analogy with the high-harmonic generation [7], these regions have been called plateaus, and their occurrence was attributed to rescattering of photoelectrons by their parent ion [8,9].

A recent high-resolution experiment [10] has demonstrated that the very pronounced plateau that occurs in the ionization of argon with 800 nm light is due to resonant processes; i.e., the production of photoelectrons in that energy region occurs at well defined light intensities, and drops by at least an order of magnitude for intensities only a few percent different. Similar results were reported for xenon [11]. This type of behavior is well known from resonance-enhanced multiphoton ionization at lower intensities [12], where it occurs because (easily ionizable) excited states are Stark shifted to an energy an integer number of photons above the ground state, where they are resonantly populated by a multiphoton transition. Such behavior was, however, unexpected at the high intensity used in the experiment (70 TW/cm^2), since all excited states lie very far above the potential barrier created by the combined action of the nuclear Coulomb attraction and the laser field [13]. Such states were thought to ionize instantly ("barrier-suppression ionization" [14]), and indeed the perturbative lifetimes of most states extrapolate to values much shorter than an optical cycle.

The nature of the resonances causing the high-order enhancement so far have remained a mystery, although it has been suggested [11,15] that two-electron effects are involved. Another mystery is why the enhancement predominantly occurs in a few high orders of the ATI spectrum making up a plateau.

To address those questions, we performed a numerical integration of the time-dependent Schrödinger equation for a model argon atom in the single active-electron approximation [16]. The atom was represented by a (three-dimensional) potential well,

$$V(\mathbf{r}) = [1 + Ae^{-Br} + (17 - A)e^{-Cr}]/r \qquad (1)$$

(atomic units are used throughout). For $A = 5.4$, $B = 1$, and $C = 3.682$, the eigenenergies of an electron bound in this potential faithfully reproduce the configuration averages [17] of the binding energies of the singly excited states [18], as well as the K-shell, L-shell, and $3s$ ionization potentials.

To avoid numerical problems due to the rapid time evolution of the inner shells, a hard-core boundary condition [$\Psi(R) = 0$] was imposed on the wave function Ψ at $R = 0.5$ bohr, and the time integration was only performed outside this core. Since the first node of the p wave and the first two nodes of the s wave lie within the core, this eliminates the K and L shells. Although the surface of the hard core lies very close to the natural position of the nodes of the s and p eigenfunctions, in a region where higher angular momenta hardly penetrate, it does disturb the energy levels [19]. To counteract this disturbance, a potential

$$W(r) = F\{[(R_x - r)/G]^5 - [(R_x - r)/G]^4\} \quad (2)$$

was added for $R < r < R_x$. With $R_x = 3$, $F = 2.5$, and $G = 2.01785$, this potential repaired the quantum defects of all s and d states to within 0.01 of the original ones, and that of the p states to within 0.04, with an exact representation of the $3p$ energy. In the relevant energy range (-1 to $+1$ hartree) the scattering behavior of $V(r)$ is hardly affected by these modifications. It was assumed that the ionization process itself and subsequent processes that help shape the electron spectrum occur outside R_x, so that the model provides a similarly accurate description of argon in an electromagnetic field. Unlike more common pseudopotential models [20], this model allows easy transformation to the velocity gauge in which the calculation is done.

The time-dependent Schrödinger equation,

$$i\partial_t\Psi = \{-1/2p^2 + \mathbf{p}\cdot\mathbf{A}(t) + V(r) + W(r)\}\Psi, \quad (3)$$

was integrated using a method that was fourth order in the spatial grid spacing Δr, and second order in the temporal spacing Δt, which will be described in detail elsewhere. At $r = 1000$ an absorbing wall removed photoelectrons that reached the end of the radial grid. An initial $3P$ state oriented along the laser polarization [21] was subjected to an 18-cycle laser pulse $A(t) = A_o(1 - \cos^{60}\omega t/36)\hat{z}\sin\omega t$, ($\omega = 0.05773$), which effectively included a 2-cycle turn-on and turn-off to 99% of the peak intensity. After this pulse the wave function was energy analyzed for each angular-momentum channel l with a fourth-order energy window [16]. At each energy, both the total ionization yield and the yield along the polarization were calculated by adding the populations in the l components incoherently and coherently with the phase prescribed by the corresponding spherical harmonic, respectively. The results converged to four digits at $\Delta r = 0.2$ and $\Delta t = 0.1$. Because of the velocity-gauge formulation [22], only 20 partial waves were needed.

The inset of Fig. 1 shows a typical electron spectrum resulting from this procedure, containing peaks up to 40 eV (faster electrons do reach the absorber before the turn-off is completed and are lost). For the intensity shown ($E_o = 0.044$, i.e., 68 TW/cm^2), the spectrum displays a pronounced plateau at about 25 eV. At 5% higher or lower intensities, the corresponding peaks shrink an order of magnitude, showing that the plateau is due to a resonance. This behavior closely resembles the high-order enhancement observed in the experiment. By stepping the peak laser field $E_o = A_o\omega$ from 0.030 to 0.047, three such resonances were identified.

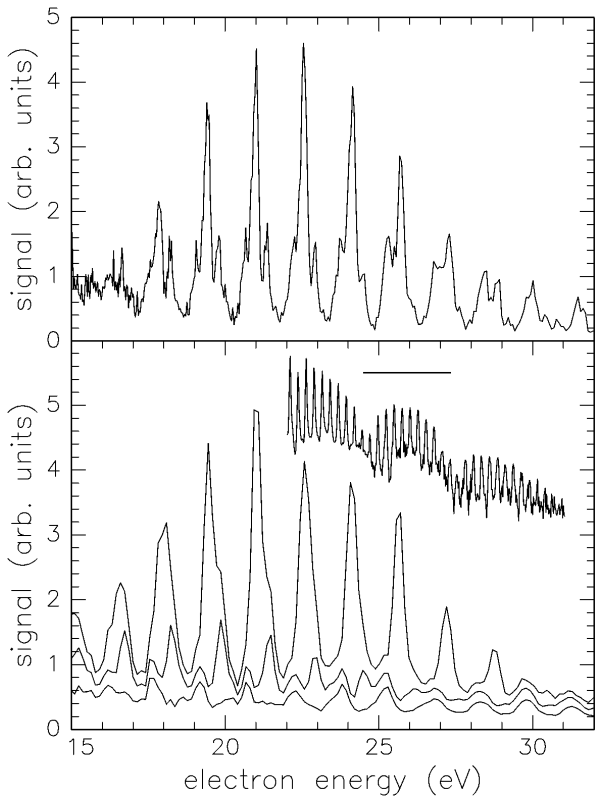

FIG. 1. Comparison between an experimental argon ATI spectrum from [10] [upper trace ($I = 70$ TW/cm^2, $\lambda = 800$ nm)], and some calculated spectra (lower traces) in the energy range of the plateau (indicated by the bar above in the full calculated spectrum in the inset). The calculations are for a Gaussian focus, focal peak intensities 69.5, 64.9, and 56.2 TW/cm^2, corresponding to $E_o = 0.0445$, 0.043, and 0.04 (top to bottom). The uppermost calculated trace shows a (barely resolved) substructure of three resonances in each ATI peak, which becomes obvious at lower intensities where the resonances disappear one by one.

To compare our numerical results with the experimental spectra of Hertlein *et al.* [10], we summed the electron spectra calculated at different intensities, weighted by a factor representing the intensity distribution in a three-dimensional Gaussian focus. The results for three different focal peak intensities are presented in Fig. 1. Apart from the lower energy resolution in the calculation (mostly due to the loss of electrons from the far end of the grid) the correspondence with the experimental spectra of Ref. [10] is excellent. It thus seems that the occurrence of high-order enhancements is well explained as a one-electron effect.

To clarify the nature of the resonances that are responsible for the observed enhancement, the wave function in the presence of the laser was analyzed. Since the large population in the (dynamically polarized) ground state largely obscures anything that goes on near the nucleus, this state was first removed from the wave function [23]. Figure 2 shows the residual wave function 0.15 cycle after a zero crossing of $E(t)$, after transient effects due to the turn-on had subsided. At this point a quasistationary state has developed, the wave function decreasing three parts in 10^4 of each cycle through ionization, but otherwise repeating its evolution periodically. The most noticeable feature of the evolving wave function is a string of probability "bunches" ejected in the preceding quarter cycle of decreasing $E(t)$. Most of these bunches escape to infinity, but the two innermost ones have almost zero drift velocity. They are driven back towards the atom and collide with it in the next half cycle, to produce a burst of high-energy electrons on backscattering. Figure 2 shows that the population in these inner bunches is strongly enhanced at the resonance.

The bunches leave the atom around a time that bunches produced in an earlier cycle collide with it from the other side. The quasistationary picture thus does not reveal whether the bunches are created by the initial tunneling event, by forward scattering of the infalling electron flux, or by interference of both.

To get a clearer picture of the events that lead to the observed bunching, the time evolution of a wave packet resulting from a single half cycle of $E(t)$ (preceded by a virtually ionizationless, twice continuously differentiable, half cycle turn-on) was studied. To this end the calculation was interrupted at a zero crossing of the electric field after half a cycle, at which time the ground-state population was removed. After that, the time evolution was continued for several laser cycles.

Figure 3b shows the history of the resulting electron density on the polarization axis, plotted in the nonstationary Kramers-Henneberger (KH) frame [24] (which greatly simplifies electron trajectories by eliminating the quiver motion). Figure 3 reveals that the initial ionization (tunneling) event produces a single nearly unstructured wave packet. The tail of this packet is sucked back towards the nucleus, to appear at the other side in the next half cycle. During this forward scattering event it gets apparently bunched and focused onto the axis. Most high-energy electrons are formed when the last appearing bunch, which has near-zero drift velocity, is scattered from the ion core at the beginning of the third half cycle after ionization.

Because of the compactness of the bunches, this scattering occurs over only a limited range of impact energies just below $2U_P$, resulting in high-energy backscattered electrons in a correspondingly limited range below $8U_P$. The envelope of the ATI spectrum is a direct image of this electron bunching, the high- and low-energy cutoffs delimiting the plateau corresponding to nodal planes or other minima between the bunches.

Part of the bunch survives the encounter by scattering in the forward direction, to reencounter the ion core on later cycles in a similar way. In addition, the nucleus seems to recapture part of the probability from an earlier, outward moving bunch when the field peaks, to emit it in a similar

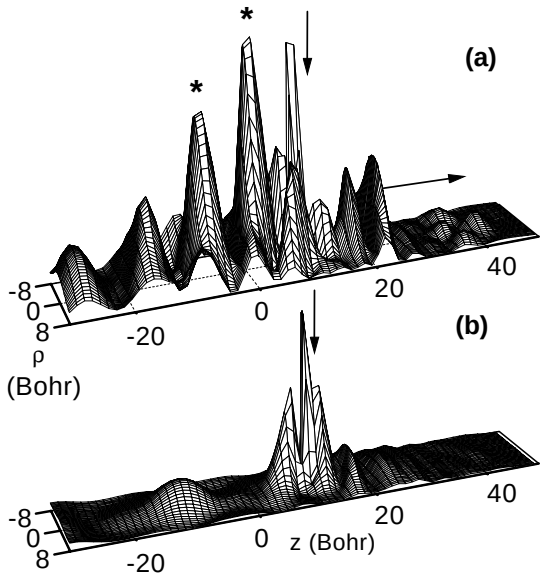

FIG. 2. Electron density left after removal of the ground state at (a) $E_o = 0.0440$ (on resonance) and (b) $E_o = 0.0425$ (off resonance). The main difference between the two cases is the presence of two lobes (marked *) with near-zero drift velocity, from which fast electrons (visible at about $z = 20$) have been scattered as the lobes encountered the nucleus (vertical arrows).

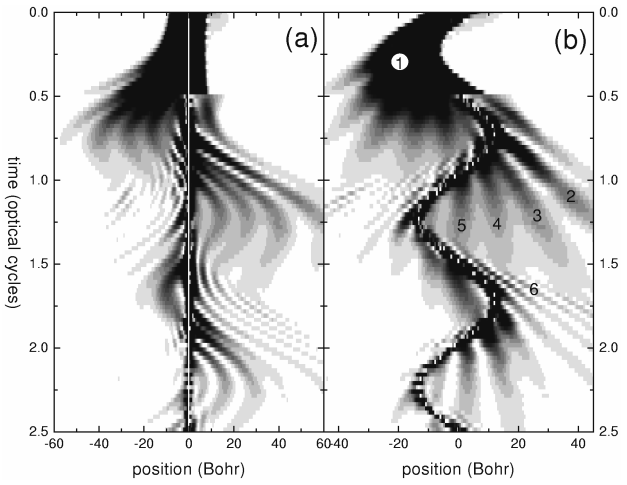

FIG. 3. Gray-scale image of the electron density on the polarization axis, for the tunneling wave packet (1) produced in a single half cycle ($t = 0 - 0.5$), plotted as a function of time (a) in the lab frame and (b) in the KH frame. The packet breaks up into bunches (2–5) on first passing the nucleus (the dark wavy band). In (b) plateau electrons can be seen speeding to the right (6) after a bunch (5) scatters from the nucleus at $t = 1.5$.

string of bunches as on the previous cycle. This process seems to repeat a number of times.

In real life, when new wave packets are created not only in a single but on each half cycle, the contribution from all of these packets has to be added coherently. This will usually result in destructive interference, except when the contribution to the bunch from all previous wave packets will be in phase. Since the phase difference between wave packets emitted in successive cycles is determined by the (intensity-dependent) Stark shift of the ground state, this condition can be fulfilled only at specific intensities. At such a resonant intensity the inner bunches build up to a large density (the two lobes in Fig. 2a), producing the high-order enhancement, while at other intensities it is virtually absent. Because of the poor overlap with the wave packet emerging from the initial tunneling (which happens at the side of the nucleus opposite to the lobes), the population can be fed only into the lobes by the complicated sequence of tunneling, recapture, and bunching.

The remarkable bunching starts when part of the electron density gets trapped about 3 to 4 bohr from the nucleus as $E(t)$ reverses. Being increasingly pushed towards the nucleus it is somehow able to survive there until $E(t)$ reaches its peak [25]. The field ramps up fast compared to the time scale of excited states, and apparently this nonadiabatic perturbation results in the formation of a radial wave packet, bouncing between the nucleus and the up-sloping $E \cdot r$ laser potential. On collision with the nucleus, part of the packet scatters forward, to appear as a bunch, and part scatters back (at fairly low energy) for the next round.

In conclusion, numerical simulations reveal that the photoionization dynamics of even a single electron near the tunneling regime shows extremely rich and interesting behavior that is capable of explaining many features of high-order ATI spectra, specifically the occurrence of plateaus and high-order enhancements.

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