Time-Resolved Dynamics of Electronic Wave Packets above the Classical Field-Ionization Threshold

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In a picosecond time-resolved study, Rb atoms in an electric field are excited above the classical field-ionization threshold by a laser. Part of the electron wave function is observed to return to the core before escaping over the saddle point. At energies just above the saddle point all Stark states in the wave packet contribute to the returning part. Close to the zero-field-ionization limit, only bluest Stark components are involved. Also, a wave packet is observed performing simultaneous quantum beats in the radial coordinate and angular momentum. Simple calculations reproduce the orbit times.

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During the last decade, the possibility to produce ultrashort laser pulses has led to an increasing interest in the dynamical behavior of wave packets and its relation to classical mechanics. A wave packet is formed by coherent excitation of several nondegenerate eigenstates, and therefore is a nonstationary state. The wave function generating this quantum beat is localized in some spatial coordinate, in contrast to the wave function of an eigenstate, which fully extends over the classically accessible region. The time evolution of the wave packet along that particular coordinate axis mimics a classical trajectory in the corresponding classical potential. In a binding potential well, a superposition of eigenstates gives rise to a nearly time-periodic wave function. The characteristic period is given by the inverse of the frequency spacing of the excited levels and equals the classical oscillation time of a particle in that same potential well. In atomic systems, coherent superpositions of electronic wave functions have been made, forming wave packets localized in, e.g., the angular [1] or radial [2] coordinates.

All atomic wave packets studied so far were superpositions of *bound* eigenstates of a potential. Their classical motion then corresponds to closed, periodic orbits. In these cases the lifetimes of the states were orders of magnitude longer than the observation times in time-resolved measurements. Consequently, a probe of the system in the frequency domain seems more useful. However, in the case of short-lived continuum states in a classically free region of a potential, the lifetimes can be fully covered by the experimental observation times, so time domain measurements seem more appropriate [3].

In this Letter we report on a time-resolved study of such a wave packet formed out of continuum states, namely the rubidium atom in a static electric field, excited above the classical field-ionization threshold. From a classical point of view, field ionization corresponds to electron trajectories that escape over the saddle point at E_c (see Fig. 1). In a hydrogenic approximation [4], the energy levels of an atom in an electric field F can be written as (atomic units are used)

$$E_{nk} = E_0 - \frac{1}{2n^2} + \frac{3}{2}nkF. \tag{1}$$

The third term is the well-known (linear) Stark effect, where n is the principal quantum number and k is the quantum number connected to the electric dipole moment of the wave function ($k = -n+1, -n+3, ..., n-1$) [5]. The states with $k > 0$ are often called "blue" Stark states because their energy is lifted by the electric field. In the classically bound region ($E < E_c$), two types of electronic wave packets that are related to our situation have been studied. Without electric field, a superposition of states with different principal quantum number n results in a "radial wave packet." The localized electron cloud oscillates between the turning points of a classical Kepler orbit [2]. Its orbit time is given by the inverse of the frequency spacing of levels: $\tau_n = 2\pi/\Delta E_n = 2\pi n^3$. With an electric field, a superposition of Stark states with different quantum number k but the same principal quantum number n has been made. It was shown

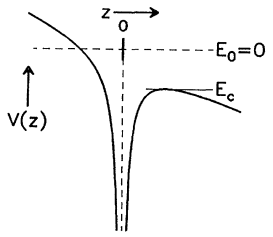

FIG. 1. Potential energy of an electron in a combined Coulomb and electric field. The electric field, directed along the z axis [$V(r) = -1/r - Fz$], lowers the ionization potential to $E_c = -2\sqrt{F}$, the potential energy at the saddle point.

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that the resulting time dependence corresponds to an oscillation of the angular momentum between the extreme values $l = 0$ and $l = n - 1$ [6]. Since low l values mean cigarlike orbits and high l values circular orbits, this implies an oscillation of the eccentricity of the orbit. The time period is given by $\tau_k = 2\pi/\Delta E_k = 2\pi/3nF$.

In our time-resolved measurements we observe that the electron, starting out in the core region with $E > E_c$, can return to that region a few times before escaping over the saddle point. Furthermore, we conclude that for energies just above the saddle point ($E \gtrsim E_c$) almost all Stark states are part of the localized electron density that returns, while for energies close to the zero-field-ionization threshold ($E \approx E_0 = 0$) only the bluest Stark components are involved. The observed periods of the (nearly closed) orbits will be shown to obey simple scaling laws with respect to energy and field strength. In addition we have observed a wave packet that performs *two simultaneous* quantum beats, corresponding to the motion of a classical particle on a Kepler orbit of which the eccentricity changes periodically.

Since the idea underlying the used experimental technique, namely, a phase sensitive measurement without light detection, has been published [7] and the experimental details will be given elsewhere [8], we will describe both only briefly here. Short (4 ps), wavelength-tunable (around 595 nm) light pulses, obtained from a cavity-dumped, synchronously pumped dye laser (repetition rate 3.8 MHz, average power 150 mW), are frequency doubled in a nonlinear crystal (efficiency $\sim 1\%$) and divided into two equal parts on a beam splitter. This beam splitter is part of a Michelson-interferometer-like setup, of which the path length of one arm can be varied in order to delay one of the pulses that comes out of the Michelson with respect to the other. The phase between the two pulses is modulated at about 6 kHz by wiggling a glass plate around Brewster's angle in the other, "fixed-delay" arm of the Michelson. This pulse pair is then used to excite ground state ($5s$) rubidium atoms between electric field plates (spaced by 5.1 mm). The polarization of the light is parallel to the electric field (selection rule $\Delta m = 0$).

The effect of the two pulses can be pictured as follows. The first pulse excites a superposition of Stark states; i.e., it creates a wave packet localized in the interaction region that starts to move out. The delayed second pulse creates a similar wave packet. If, at that moment, the first wave packet is in the interaction region, the two wave packets interfere, and it depends on the relative phase between the two pulses whether this interference is constructive or destructive. This means that the total "amount of wave packet," and thus the resulting population of excited states after the two pulses, is modulated as the phase of the light. If, on the other hand, the first wave packet is out of the interaction region when the second pulse arrives, there will be no interference between the wave packets, so the total amount of excited atoms does not depend on the phase of the light. Since the wave packet is made above the field-ionization limit, the electron escapes over the saddle point after a limited number of returns to the interaction region, and we detect the resulting ion. So the behavior of the wave packet can be monitored by looking at the 6 kHz modulation on the ion signal.

The idea to use the dependence on the relative phase of a pulse pair was used earlier in phase-*locked* detection of a vibrational wave packet [9]. In our phase-*modulated* version, the ion signal is passed through a bandpass filter at the modulation frequency, and led into a PC. There the root mean square (rms) of the filtered phase-modulated signal is computed. When the time evolution of the wave packet after one pulse is written as $|\psi(t)\rangle = \sum_n a_n e^{-i\omega_n t}|n\rangle$, the rms signal is proportional to $|\langle\psi(0)|\psi(t)\rangle|$, the absolute value of the time correlation function of the wave function. This expresses the notion that we directly measure the resemblance between the wave packet at time "t" and the initial wave packet, $|\psi(0)\rangle$. It also helps to specify the "interaction region." For a pure radial wave packet, $|\psi(0)\rangle$ is confined to the core region since the ground state, where it is excited from, is small. This implies that peaks will be observed whenever $|\psi(t)\rangle$ is localized near the core. In the case of a pure superposition of k states, peaks will be seen whenever $|\psi(t)\rangle$ predominantly has p character (angular momentum $l = 1$), because $|\psi(0)\rangle$ is excited from an s state and the selection rule $\Delta l = \pm 1$ holds.

Measurements have been performed at field strengths ranging from 98 V/cm to 3137 V/cm and excitation energies between E_c and E_0. Two results are given in Fig. 2. They illustrate some of the features that were seen at all field strengths. First, peaks were observed for $t > 0$, indicating that the electron does not escape over the saddle point "at once," but has a chance of returning to the

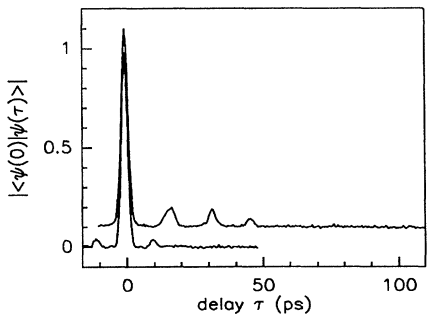

FIG. 2. Two delay scans at F=555 V/cm. The upper trace is measured at an energy near the saddle point ($E = 0.87E_c$), the lower trace at an energy near E_0 ($E = 0.18E_c$). Note that the number of recurrences of the wave packet decreases with increasing energy. The observed time periods correspond to the oscillation times of the angular momentum.

core a few times. Second, the number of observed peaks (recurrences) gradually decreases from typically four for energies just above E_c to zero above E_0. This implies that the lifetimes of the states decrease with increasing energy above E_c. Third, when the peak spacing (recurrence time) is normalized to the peak spacing at $E = E_c$ and plotted as a function of normalized energy E/E_c, the results for all field strengths are similar [see Fig. 3(a)] [10]. This result shows that the basic wave packet dynamics do not depend on the absolute energy above the saddle point ($E - E_c$), but only on this energy difference relative to E_c. Fourth, all recurrence times decrease with increasing field strength. This point is illustrated in Fig. 3(b) for the recurrence times both closest to E_c (τ_c) and closest to E_0 (τ_0).

For the relative heights and shapes of the observed recurrence peaks, no simple trends were found. This is understandable since the Stark manifolds belonging to different principal quantum numbers n largely overlap above the classical field-ionization threshold, and it is the precise way in which they are interwoven which determines those peak heights. Here we shall concentrate on the regularities in the peak *spacings*. These might

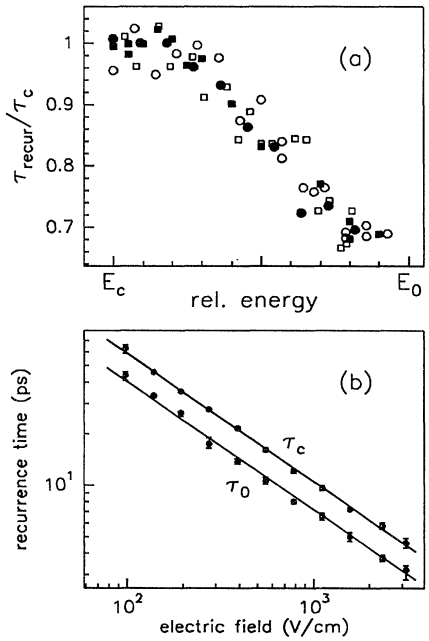

FIG. 3. (a) Observed peak spacing ("recurrence time"), normalized to the peak spacing at energies just above E_c, plotted as a function of the relative energy above E_c for four different values of the electric field. Filled circles: 139 V/cm; filled squares: 392 V/cm; open circles: 784 V/cm; open squares: 2353 V/cm. (b) Points: measured peak spacing ("recurrence time") for energies just above E_c (τ_c, upper set) and near E_0 (τ_0, lower set) as a function of field strength. Lines: electric-field dependence of τ_c and τ_0, calculated from Eqs. (2) and (3), respectively.

indicate that, in spite of the strong n mixing, the two basic types of wave packet motion described earlier (corresponding to radial oscillations and eccentricity oscillations) are still applicable to some extent. We note that $\tau_k \propto n^{-1}$ decreases when n gets larger, while the opposite is true for $\tau_n \propto n^3$. Furthermore, n increases with energy E. The observed recurrence times decrease with energy E [Fig. 3(a)], suggesting that τ_k is the most prominent time scale in the experiments. From the observed dependence of the recurrence time on energy, $\tau(E)$ [see Fig. 3(a)], an "effective value" of n as a function of energy, $\overline{n}(E)$, can be defined using $\tau_k(E) \equiv 2\pi/3F\overline{n}(E)$.

The resulting dependence of $\overline{n}$ on E can be interpreted as follows. For energies right above E_c all Stark states in the superposition, i.e., within the laser bandwidth, contribute to the part of the wave function that returns to the interaction region. The relevant $\overline{n}(E_c) \equiv \overline{n}_c$ therefore is the average n of all $|nk\rangle$ states for which $E_{nk} = E_c = -2\sqrt{F}$ [5]. Setting $k_{\max} = n - 1$ and $k_{\min} = 1 - n$, the lowest and highest values of n are found as a function of electric field F, respectively. For the electric-field dependence of τ_c one then obtains (in atomic units)

$$\tau_c = \frac{2\pi}{3\overline{n}_c F} = 4.023F^{-3/4}. \tag{2}$$

When the energy is increased from E_c to E_0, less and less Stark states contribute to the returning part of the wave function. This gradual loss of Stark states starts with the reddest component and ends (above E_0) with the bluest, in agreement with the observation that the lifetimes of Stark states increase with increasing k [11]. The relevant value of $\overline{n}(E_0) \equiv \overline{n}_0$ can be found by considering that at E_0 only the bluest Stark components are relevant. From $E_{nk} = E_0 = 0$ and $k_{\max} = n - 1$, it follows that

$$\tau_0 = \frac{2\pi}{3\overline{n}_0 F} = 2.756F^{-3/4}. \tag{3}$$

The measured dependence of τ_c and τ_0 on the electric field is in very good agreement with the calculated dependence, as can be judged from Fig. 3(b). The $F^{-3/4}$ dependence of the recurrence time is also consistent with results of frequency-resolved measurements of the photoabsorption cross section of an atom in an electric field above the zero-field-ionization threshold (E_0) [12]. These results were theoretically reproduced with closed-orbit theories [13]. Our observations show that this $F^{-3/4}$ dependence can also be found for energies below E_0.

The gradual loss of Stark states from the returning part of the wave packet, starting with the reddest one, can be understood classically as well. The electron density of red Stark states is concentrated between the core and the saddle point, that of the blue Stark states on the opposite side of the core [5]. The magnitude of $|k|$ gives the degree to which the electron density is peaked in the direction towards ($k < 0$) or away from ($k > 0$) the saddle point.

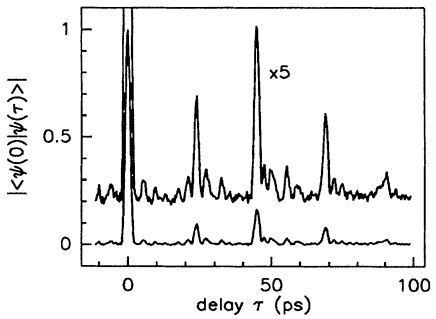

FIG. 4. Delay scan at $F = 392$ V/cm and $E = 0.9E_c$, showing two simultaneous quantum beats. The short time period (4.0 ps) corresponds to a radial oscillation, and the long period (22 ps) to an oscillation of the angular momentum.

The results then illustrate that for energies just above E_c almost every classical trajectory, independent of starting direction, will form a nearly closed orbit and return to the core region before escaping over the saddle point. As the energy is increased, the starting direction needs to be more and more in the direction pointing away from the saddle point in order to give a trajectory that returns. Finally, only the trajectory that starts out exactly in the $-z$ direction (Fig. 1) will come back once.

The observed dynamics are thus dominated by a quantum beat that corresponds to the oscillation of the angular momentum. It reflects the fact that the dominant energy spacing in the superposition can be written as $3\overline{n}F$, where $\overline{n}$ is the average n value of the relevant $|nk\rangle$ states in the superposition. However, the effect of the *radial* oscillations can also be observed. In the presented experiments, the radial oscillation time $(2\pi\overline{n}^3)$ is always a few times shorter than the eccentricity oscillation time $(2\pi/3\overline{n}F)$. This implies that the eccentricity motion is modulated by the radial motion, which is indeed seen in the measurements (see Fig. 4). The observed time scales of 22 ps and 4.0 ps are agreement with τ_k and τ_n for $\overline{n} \approx 30$. This wave packet thus performs two simultaneous quantum beats and can classically be pictured as a particle traveling on a Kepler orbit with a slowly changing eccentricity.

In conclusion, we have performed a time-resolved study of a rubidium atom in an electric field above the classical field-ionization threshold. Although in rubidium no states are stable against field ionization, we observe that part of the electronic wave function can return to the core region before escaping over the saddle point. The observed scaling laws for the recurrence times have been interpreted as follows: At energies just above the saddle point all Stark states in the superposition contribute to the part of the wave function that returns. For energies close to the zero-field-ionization threshold, only the bluest Stark components are involved. In addition, we have observed a wave packet performing two simultaneous quantum beats: one in the radial coordinate and one in the angular momentum.

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