Streak-Camera Probing of Rubidium Rydberg Wave Packet Decay in an Electric Field

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The decay of autoionizing electronic wave packets of rubidium in a static electric field of 2.0 kV/cm is studied. Using an atomic streak camera, the escape of the laser-excited Rydberg wave packets over the saddle point is measured. New information on the ionization dynamics is obtained that cannot be retrieved from optical recurrence spectra. For laser excitation perpendicular to the electric field at scaled energy $\epsilon = -1.74$, the main ionization is delayed by 12 ps. For parallel polarization ($\epsilon = -1.32$) the ionization occurs in two bursts, 7 ps separated.

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Several aspects of the dynamics of alkali-metal Rydberg atoms in a static electric field have been studied extensively over the past decades [1–9]. If the energy of the Rydberg electron is above the saddle point of the combined Coulomb and static field potential, it escapes on a picosecond time scale. A wave packet above the saddle point, created by a short laser pulse, shows a few oscillations in angular momentum (l) and radial coordinate (r) before escaping from the atomic potential. Up to now, experimental studies on the dynamics of the wave packet focused on recurrences of the wave packet to its initial conditions (r_0, l_0). These studies have been carried out in the time [8,10] and frequency [11] domain. In optical pump delayed-probe studies the overlap of the evolved wave packet with the initial wave packet is directly measured as a function of time. The recurrences of the launched wave packet manifest themselves as resonances in the photoionization spectrum. As discussed below these two established optical techniques provide identical, but not complete, information of the Rydberg electron dynamics. We present a new way of probing the electron dynamics which distinguishes different electron trajectories that are seen as identical by the optical techniques. Let us consider the following two classical electron trajectories as an example. In the first trajectory the electron is directly launched towards the saddle point, giving rise to a prompt emission of the electron. In a second trajectory the electron is not encountering the saddle point immediately. The angular momentum will oscillate between zero and $n - 1$ as a function of time due to the presence of the static electric field. Once the angular momentum is low again, the nonhydrogenic core can scatter the electron into the direction of the saddle point. Also, the ionization is most likely when the angular momentum is low. In such an alternative trajectory the electron escapes over the saddle point after some delay, without returning to its initial conditions (r_0, l_0). Since in the two trajectories described above there is no return to the initial conditions, they will not be seen as recurrences in a pump-probe study, or, equivalently, in the Fourier transformed domain, as resonances in the photoionization spectrum. Despite the different ionization times, the electron trajectories are indistinguishable in these optical measurements. In this Letter we introduce a new device, the atomic streak camera, which measures directly the escape of the Rydberg electron in a static electric field with picosecond resolution. By measuring the time dependent leaking of the wave packet over the saddle point, instead of measuring recurrences to its initial conditions, trajectories such as the two mentioned above are distinguished: New information on the dynamics is obtained. We have compared the results of the electron escape, as measured with the atomic streak camera, with Fourier transformed absorption spectra obtained under identical circumstances. The completely different features measured by these two techniques demonstrate that indeed complementary information on the Rydberg electron dynamics is obtained: The lifetime as measured by an optical technique is not the same as the time it takes the electron to leave the atom.

Using a short laser pulse the ground state s electron of an alkali-metal atom can be excited above the saddle point energy, $E_c = -2\sqrt{F}$, where F is the static electric field (atomic units are used, unless stated otherwise). Using the scaled energy $\epsilon = E/\sqrt{F}$ ($\epsilon = 0$ at the zero field ionization limit and $\epsilon = -2$ at the saddle point energy) the position of the excitation relative to the saddle point energy is determined independent of the electric field. Because of the bandwidth of the short laser pulse, a coherent superposition of several Stark states is excited, thus creating a wave packet. At the time of excitation the angular momentum of the wave packet is low ($l = 1$), but due to the static electric field the angular momentum will oscillate as a function of time. The beating time of the angular momentum is determined by the inverse of the energy spacing, ΔE, of the excited Stark states. The energy spacing can be calculated for hydrogen [12], resulting in a recurrence time $\tau_k = 2\pi/\Delta E$ of

$$\tau_k = \frac{2\pi\sqrt{-2\epsilon}}{3} F^{-3/4}. \quad (1)$$

Although Eq. (1) is derived for hydrogen, it is still a good approximation for alkali-metal atoms [8]. At a sufficiently

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large bandwidth of the exciting laser, k states belonging to different n manifolds can be excited, resulting in radial oscillations of the wave packet [10]. For the dynamics of the electron above the saddle point studied in this paper, there are about six radial oscillations during one oscillation of the angular momentum.

In Fig. 1 is shown the principle of the atomic streak camera that is used to measure the electron escape over the saddle point. Atoms in a static electric field (typically 2–5 kV/cm) are excited by a short laser pulse. The time of autoionization of the excited atoms determines the temporal shape of the electron pulse. The electrons are accelerated by the electric field and pass through a slit, entering the deflection region. The voltage of the upper deflection plate is swept as a function of time (typically 2.5 kV in 500 ps). The deflection of the electrons depends on the time of arrival between the deflection plates; i.e., the temporal profile of the electron pulse is transformed into a spatial profile which is measured using a position sensitive detector. The time calibration of the camera is obtained by applying two ionizing laser pulses to the atoms, with a known delay with respect to each other. The measured distance between the two resulting electron peaks on the detector provides a calibrated time axis.

To create the fast sweep pulse a GaAs photoswitch is used [13]. When the GaAs photoswitch is illuminated with a femtosecond laser pulse the resistance is lowered 7 orders of magnitude; i.e., the GaAs is used as a switch to charge up the deflection plates to the high voltage. The shape of the voltage pulse is fully determined by the geometry of the circuit, giving a rise time of 500 ps. Since the same laser is used to ionize the atoms, the shot to shot time jitter is extremely small (<2 ps). Details on the atomic streak camera will be published elsewhere [14].

The laser used in this experiment to create the Rydberg wave packets is based on a colliding pulse mode locked laser. The pulses are amplified using a four-stage dye cell amplification chain which is pumped by the second harmonic of a Nd:YAG (where YAG denotes yttrium aluminum garnet) laser operating at 10 Hz. Most of the light (100 μJ) is used to trigger the photoswitch while a small fraction (10 μJ) is used for continuum generation in a water cell. After amplification and the use of a shaper, tunable pulses are obtained (590–605 nm) with a bandwidth between 0.1 and 1.2 nm [15]. The laser light is frequency doubled using a potassium dihydrogen phosphate crystal. The autoionizing Rydberg wave packets are excited using a single photon excitation step from the 5s ground state of rubidium.

In order to obtain absorption spectra the frequency-doubled output of a pulsed nanosecond dye laser (bandwidth 0.1 cm^{-1}) is used to excite the rubidium ground state atoms to highly excited states above the classical field ionization limit. The ionization yield is determined as a function of the laser wavelength [11]. The spectra are recorded under identical circumstances; i.e., the same interaction region and static field are used as with the atomic streak camera measurement. A so called recurrence spectrum is obtained by Fourier transforming these spectra after multiplication with the Gaussian spectrum of the short pulse used for the excitation of the wave packet. The recurrence spectrum does not depend significantly on the actual shape of the optical spectrum [11]. This recurrence spectrum is compared with the decay as observed by the atomic streak camera.

In Fig. 2 an ionization spectrum is shown of an electronic wave packet in a static electric field of 2.0 kV/cm.

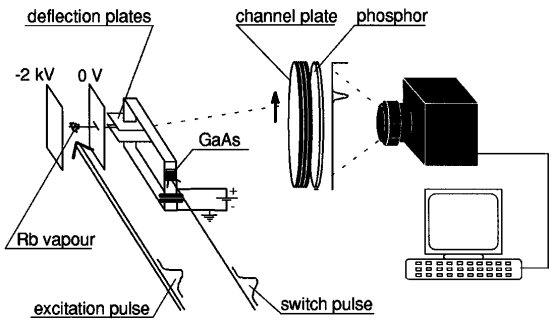

FIG. 1. Principle of the atomic streak camera. Using a short laser pulse rubidium atoms are excited in a strong electric field. Once the electrons have escaped over the saddle point, they are accelerated and pass through a slit entering the deflection region. The voltage over the deflection plates is swept in time (typically 2.5 kV/500 ps), transforming the temporal profile of the electron pulse into a spatial profile, which is measured using a position sensitive detector.

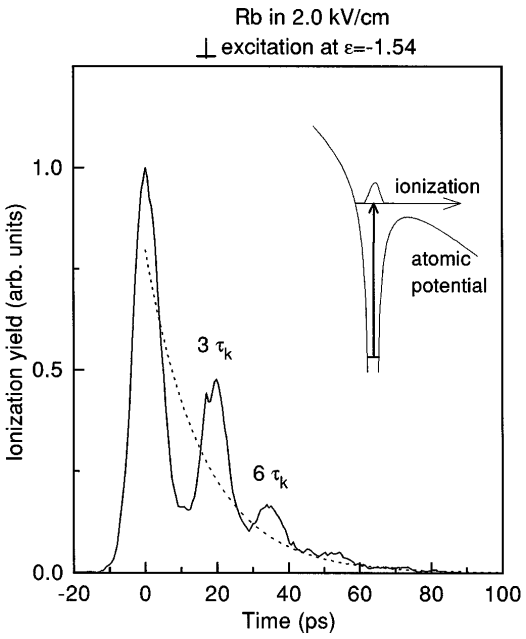

FIG. 2. Measured decay of an autoionizing electronic wave packet (full line). The wave packet is created above the classical field ionization limit at a scaled energy of $\epsilon = -1.54$. The laser polarization is perpendicular to the electric field of 2.0 kV/cm. The calculated angular recurrence time using the hydrogenic model is $\tau_k = 5.7$ ps. The dotted line is an exponential fit with a decay time of 16 ps.

The polarization of the exciting laser is perpendicular to the electric field. A number of k states in the Stark manifold around $n = 22$ are excited by a 4 ps laser pulse (bandwidth 0.13 nm), creating an angular wave packet. Since the excitation is well above the saddle point in the potential ($\epsilon = -1.54$), the main fraction of the Rydberg population is ionized immediately, i.e., the first peak in the ionization spectrum. However, some of the Rydberg population does not escape over the saddle point immediately, but makes some recursions in the potential before escaping. According to Eq. (1) the angular recurrence time τ_k at this field strength and excitation energy is 5.7 ps. A number of conclusions can be drawn from Fig. 2. (1) The time resolution of the developed streak camera is sufficient to monitor the decay dynamics. (2) Escaping over the saddle point at this particular energy is favorable at every third oscillation of the angular momentum of the wave packet (the observed peak spacing is about $3\tau_k$). (3) Under these conditions a fixed fraction of the remaining wave packet is ionized (the peak height is decaying exponentially).

In Fig. 3 an ionization spectrum is shown of a wave packet created just above the classical field ionization limit ($\epsilon = -1.74$). The laser polarization is chosen perpendicular to the electric field of 2.0 kV/cm. Under identical experimental conditions, the photoionization spectrum is measured. In the lower part of Fig. 3 the Fourier transform of the relevant part of the photoionization spectrum is shown. This Fourier transform represents the recurrence spectrum of the angular wave packet to the initial conditions ($r_0 \approx 0, l_0 = 1$) [11]. Note that the time scales of the two figures are obtained independently of each other.

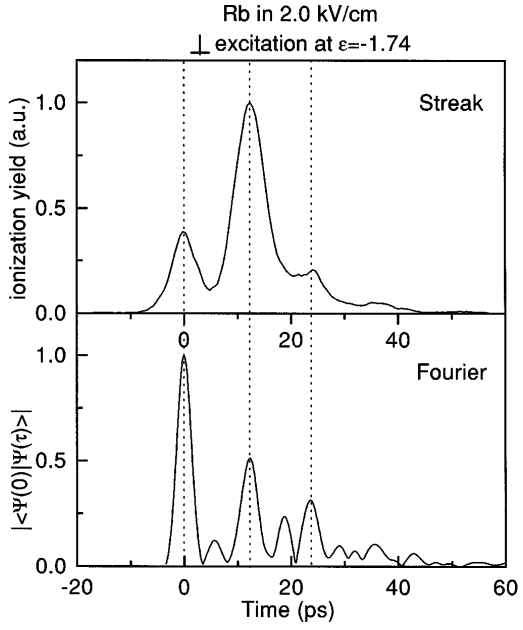

FIG. 3. Comparison between time resolved ionization spectrum (upper graph) as measured by the atomic streak camera and recurrence spectrum (lower graph). The excitation is at a scaled energy of $\epsilon = -1.74$. The laser polarization is perpendicular to the electric field of 2.0 kV/cm.

The upper scale is obtained by calibrating the atomic streak camera directly in the time domain, whereas the lower scale is obtained by the inverse of the frequency scale of the recorded photoionization spectrum.

The main observation presented in this paper is shown in Fig. 3. The dynamics observed by the optical probe (recurrence at starting condition) are not the same as the dynamics probed by the atomic streak camera (escape over the saddle point). From the recurrence spectrum it can be seen that at the even oscillations of the angular momentum ($2\tau_k = 12.3$ ps) the evolved wave packet mimics the initial conditions best in both angular momentum and radial coordinate. Maxima in the ionization spectrum are also observed when the angular momentum is low. We will argue why low angular momentum is required for efficient ionization using semiclassical arguments. (1) Scattering from the ionic core is efficient at low angular momentum since $r_{\min} \sim l(l + 1)$. Therefore, the electron initially directed perpendicular to the electric field can be scattered towards the saddle point when its angular momentum is low. (2) For an efficient escape over the saddle point a high radial velocity compared to the angular velocity is required and hence a low angular momentum. (3) In order to probe the saddle point the outer turning point of the electron trajectory must be large and hence the angular momentum needs to be low. The most striking observation in Fig. 3 is that the ionization of the main fraction of the wave packet (60%) is delayed until the second angular recurrence of the wave packet. *From the recurrence spectrum alone it is impossible to draw this conclusion*. Directly after excitation the wave packet is ejected in the direction of the laser polarization, perpendicular to the electric field. It is therefore unlikely that the wave packet will approach the saddle point, and the observed prompt ionization is indeed small. In a short while the angular momentum of the wave packet increases and the scattering from the ionic core upon radial recurrences is strongly reduced. After $2\tau_k = 11.4$ ps, the angular momentum of the wave packet is low again as well as the radial distance. As a result core scattering will change the direction of the electron into the direction of the saddle point. While moving from the core to the saddle point, the angular momentum of the scattered electron is still low, and the ionization is efficient.

In Fig. 4 an ionization spectrum is shown of a wave packet created with the laser polarization *parallel* to the electric field at $\epsilon = -1.32$. The created wave packet consists of two subpackets oriented along the electric field and laser polarization. One subpacket is located near the saddle point (downhill) leading to prompt ionization, i.e., the first ionization peak, whereas the sub-wave-packet located at the uphill side will have to make a recursion before escaping. The observed spacing between the ionization peaks and the recurrence to the initial condition (7.3 ps) are reasonably well predicted [16] by the hydrogen model ($\tau_k = 6.1$ ps), calculated for the bluest Stark states, at this field strength. From the recurrence spectrum (lower graph), we observe that the recurrence amplitude

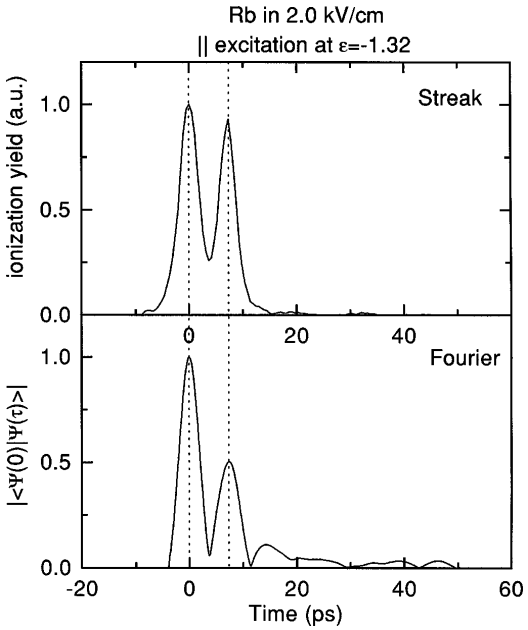

FIG. 4. Time resolved ionization spectrum of a wave packet created at a scaled energy of $\epsilon = -1.32$. The laser polarization is parallel to the electric field of 2.0 kV/cm (upper graph). Also plotted (lower graph) the corresponding recurrence spectrum at this excitation energy and field strength.

after τ_k is 50% of the initial wave packet, i.e., the wave packet created uphill returns to its initial conditions. The uphill subpacket scatters from the core in the downhill direction of the saddle point and ionizes: the second peak in the ionization spectrum.

In conclusion, we have shown that a detailed understanding of the dynamics of short lived electron wave packets above the saddle point is obtained by measuring both the optical recurrence spectrum and the atomic streak-camera ionization spectrum. For *perpendicular* polarization of the laser we have seen that the conventionally determined lifetime is not the same as the time of ionization. Optical lifetime measurements indicate the recurrences to the initial conditions and not the time of escape over the saddle point. In the case of *parallel* polarization of the laser the downhill subpacket ionizes promptly while the uphill packet ionization is delayed. After a full oscillation of the angular momentum the uphill subpacket scatters in the direction of the saddle point and ionizes.

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[16] In order to understand fully the evolution of the wave packet and the coupling to the continuum, a quantum calculation involving a large number of Stark states is needed, a challenging theoretical effort beyond the scope of this Letter.