Stepwise Electron Emission from Autoionizing Magnesium Stark States

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We report on investigations of time dependent autoionization of doubly excited magnesium atoms in a strong electric field. A narrow band laser excites one electron towards a $3s16k$ Stark state and, subsequently, the second electron is excited with a short laser pulse. The autoionizing electron yield shows a stepwise decay which is detected by a streak camera with picosecond resolution. Quantum calculations are in good agreement with the experimental results.

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Decay of an autoionizing two-electron atom involves a doubly excited state coupled to a state in which one electron is ejected while the second electron relaxes to a deeper bound state. If one of the excited electrons is in a Rydberg state, only the fraction of the Rydberg wave function close to the nucleus can exchange energy with the excited core electron. At higher principle quantum number n the wave function spends less time at the core; hence, the decay time scales with the oscillation period of the Rydberg electron. The emitted electron flux of such an autoionization process is most often *exponential* in nature.

In this Letter we present a combined experimental and theoretical study of a *stepwise* decay of the electron flux. A long pulse dye laser excites the Mg ground state atoms ($3s^2$) to a stationary Rydberg state ($3snl$). The excitation pulse is much longer than the classical orbit time of the Rydberg state and the electron density is spread over the complete trajectory. Since only one electron is excited, autoionization is not yet possible.

The key element for stepwise decay is that the second electron is excited with a laser pulse of short duration with respect to the classical period of the spectating Rydberg electron. The initial Rydberg eigenstate $3snl$ is projected onto final autoionizing states in the $3pn'l'$ Rydberg series. The Rydberg electron experiences a change in potential upon excitation of the core electron which induces a shakeup from the initial nl Rydberg state to final $n'l'$ state. If the excitation pulse is short compared to the oscillation period, a coherent superposition of autoionizing final states, or a wave packet, is created.

An intuitive picture of stepwise decay is given in Fig. 1. Most of the initial Rydberg wave function is far away from the nucleus during the excitation of the core electron as depicted as the gray ribbon in Fig. 1a. The fraction of the initial Rydberg wave function that happens to be near the core during excitation can autoionize ($3pn'l' \rightarrow 3s\epsilon l''$) which results in electron emission. We define the parameter γ to be the probability for the Rydberg electron to autoionize during one collision with the core electron (50% in Fig. 1). The remainder $(1 - \gamma)$ will leave the core unaffected and persists in its bound orbit. The outgoing electron flux will remain constant during one oscillation period since the Rydberg density far from the nucleus is unaffected by the short excitation (Fig. 1b). After one oscillation period all of the Rydberg density is reduced and the fraction originally near the core returns. At that time the incoming density is reduced and the autoionization flux suddenly decreases to $\gamma(1 - \gamma)$. This reduced electron emission remains constant for a second Rydberg oscillation period, and so on (Fig. 1c). The result is a series of steps in the outgoing electron flux.

In 1991, Wang and Cooke [1] predicted this stepwise decay behavior for a Rydberg electron that initially oscillates in the radial coordinate [2], after sudden isolated core excitation (ICE) [3]. In the original proposal by Wang and Cooke, one electron is excited into a high n ($n = 65$) Rydberg state where the energy spacing between Rydberg states is nearly constant. Although starting from an eigenstate, the sudden ICE creates a superposition of final, autoionizing, states. The emitted electron flux exhibits a stepwise decay with time steps of the radial oscillation period, $\tau = 2\pi n^3$.

In this Letter we report on the observation of stepwise decay [4] by probing the electron emission of an autoionizing Rydberg atom in a strong, static electric field after ICE. The angular momentum l of a Rydberg electron in

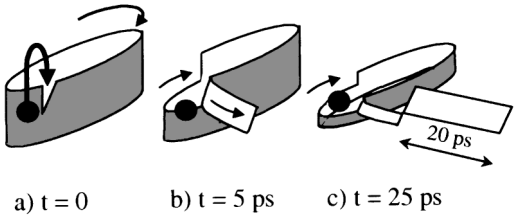

FIG. 1. Intuitive picture for stepwise decay. In the initial state one electron is in a high Rydberg orbit. The electron density is depicted as the gray ribbon around the black core. At $t = 0$ a short laser pulse excites the inner electron creating an exit for the outer electron. The autoionization probability γ (50% here) is constant and the electron emission has a constant intensity during a complete round-trip period of 20 ps in this example. After one round-trip time there is less electron density to autoionize and the outgoing flux will suddenly drop and remain constant again during the second round-trip period.

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an electric field is no longer conserved and the Rydberg state splits up into a manifold of n Stark states. We excite the initial Stark state $3snk$ to an autoionizing Stark state $3pn'k'$ with a short pulse. A Stark state is a superposition of all angular momenta in that manifold. All angular momenta of the final Stark wave packet oscillate between the extreme values $l = 0$ and $l = n - 1$ [5]. Only low angular momentum wave packets penetrate into the core region [$r_{\min} \sim l(l + 1)$] and thus can interact with the, now excited, inner electron [6]. As a result, only the fraction of the wave packet with low angular momentum character has a chance to autoionize. Since all angular momenta eventually evolve to low angular momentum, the autoionization rate remains constant for a complete angular oscillation period after which it drops suddenly with γ. The intuitive picture in Fig. 1 is now in l space. The electron emission will show a stepwise decay with a step time given by the angular momentum oscillation rather than the radial oscillation period as in the proposal by Wang and Cooke [1].

In order to measure the electron flux, we have used a streak camera with picosecond resolution. A detailed description of the streak camera is given in Ref. [7]. The principle is as follows. An effusive beam of Mg atoms, from a resistively heated oven, is excited in a crossed laser beam arrangement. A 7-ns laser pulse with a frequency in the region 650 to 655 nm and a bandwidth of 0.2 cm^{-1} is frequency doubled and excites the Mg atoms in a nonresonant two-photon process from the ground state to a Stark state $3s16k$. A second ($\lambda \sim 560$ nm) pulse with a duration of 2 ps is frequency doubled and drives the inner electron $3s \rightarrow 3p$ transition. The interaction region lies between two plates (separation = 10.0 mm) over which a static electric field is applied. The second plate has a slit of 10×0.3 mm parallel to the beam direction through which the electrons can leave the interaction region. A set of multichannel plates and a phosphor screen viewed by a CCD camera images the signal. In the trajectory of the electrons two deflection plates are placed with a fast sweeping electric field perpendicular to the direction of the electrons. Electrons arriving at different times in the deflection region experience a different deflection field and the time coordinate is converted to a position coordinate on the phosphor screen.

In the experiment, the initial states are various Stark states of the $3s16k$ manifold in a field of 1400 or 1600 V/cm. In these fields, the spacing between the Stark states of a hydrogen atom is 2.9 and 3.3 cm^{-1}, respectively, corresponding to angular oscillation periods of 12 and 10 ps, respectively. We expect the angular oscillation period of the final Stark wave packet in Mg to be close to these values. For comparison, the radial oscillation time for a $n = 16$ Rydberg state is 0.6 ps.

To classify the initial Stark states a calculated Stark map around the Mg $3s16k$ manifold in fields up to 1600 V/cm is shown in Fig. 2a. In zero field, the singlet

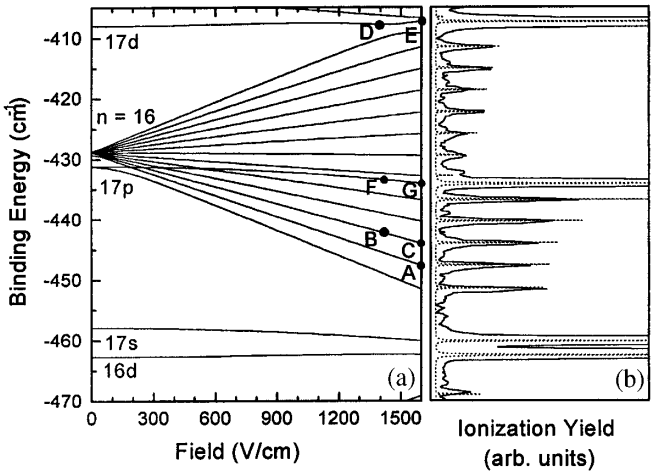

FIG. 2. (a) A calculated Stark map from the Mg $n = 16$ manifold in fields up to 1600 V/cm. (b) Experimental three-photon resonant ionization yield spectrum at 1600 V/cm as a function of binding energy. The dotted line is the calculated two-photon absorption cross section. The letters indicate the experimental positions in Figs. 3 and 4.

s, p, and d quantum defects near $n = 16$ are 1.52, 1.05 and 0.60, respectively. The $17s$ and $16d$ states are about halfway between the $n = 15$ and 16 manifolds and only start mixing into the manifolds at high fields. The $17p$ state is almost in the middle of the $n = 16$ manifold. Since it gains only s or d character at higher fields, it does not mix with the Stark states until relatively high fields. In Fig. 2b the experimental three-photon resonant ionization yield spectrum at 1600 V/cm is shown as a function of binding energy. The s, p, and d states are an order of magnitude stronger than the Stark states and off scale in the figure. The dotted lines in Fig. 2b are the calculations of the two-photon absorption cross section. The positions of all peaks are accurately reproduced and gives us confidence that, concerning the initial states, the theory matches the experiment.

The narrow band excitation laser is fixed on a selected Stark state and the second UV laser pulse is applied to induce the $3s \rightarrow 3p_{1/2}$ transition at 280.4 nm. The near transform-limited pulse has a time duration of 2 ps which is short compared to the Stark period of the Rydberg electron. Figure 3 presents the main observation of this Letter: The electron emission of the $3s16k$ state at the initial position A in Fig. 2 after sudden ICE is shown as a function of time. After a rise time of about 5 ps the autoionization rate remains constant for 7 ps and then drops by 60% in 4 ps. It remains constant again for 5 ps before a second, equal drop in emission. The apparent shorter second and third period are caused by time smearing in the detector due to the large excess kinetic energy of the autoionizing electrons. From Fig. 3 we can deduce that the angular oscillation time is about 9 ps and the probability to autoionize is 60% per core passage.

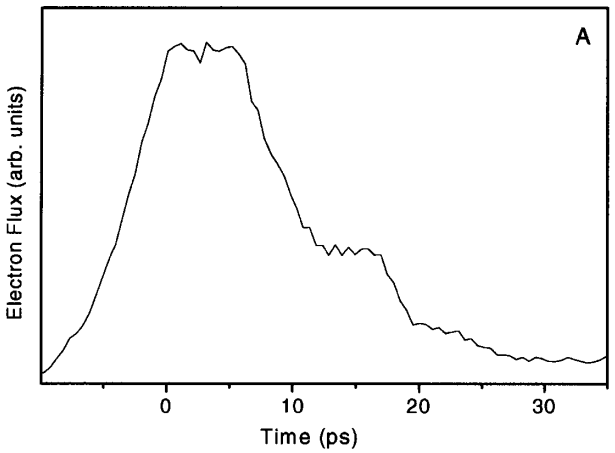

FIG. 3. The observed stepwise decay of the outgoing electron flux from the $3s16k$ state at position A in Fig. 2.

Our calculations are based on the multichannel Stark theory developed in Ref. [8], the formulation of isolated core excitations in Ref. [9], and the theory of time dependent ejection of electrons from autoionizing states in Ref. [10]. The main idea of the calculation is to use the energy eigenstates in the field as a basis to expand the time dependent wave function, $\Psi(\mathbf{r}, t)$,

$$\Psi(\mathbf{r}, t) = \int A(E) e^{-iEt} \psi_E(\mathbf{r})\, dE\,, \qquad (1)$$

where ψ_E is the eigenstate and $A(E)$ is the amplitude of the final state E. The amplitude, $A(E)$, for exciting a state at energy E is the dipole matrix element times the amplitude that the laser contains a photon at the correct frequency. The basic part of the calculation uses the semiclassical treatment of the Stark problem in parabolic coordinates [11] to obtain the initial state and the final eigenstates in the field. The final state is an autoionizing continuum state in which the only path for the electron to escape from the atom is by scattering from the core. As in Ref. [8], all of the zero-field scattering parameters for both initial and final states are obtained from an R matrix calculation in LS coupling where the spin-orbit effects are incorporated by using an LS to jQ frame transformation [10].

There are two major difficulties that must be overcome in applying Eq. (1). The first is that the dipole matrix elements cannot be obtained by direct numerical integration from an initial state. Instead we use the idea of Ref. [9] that the dipole matrix element between initial and final state is the *projection* of the initial Rydberg wave function onto the final eigenstates [12]. The second difficulty is that Eq. (1) is easiest to use for atomic distances ($<10^5$ a.u.) and we must obtain the time dependent electron flux through a slit that is a macroscopic distance from the atom. Fortunately, the electric field hardly affects the ejected electron's trajectory within 10^4 a.u. of the nucleus because the ejected electron has a relatively high kinetic energy compared to the potential energy from the external field. This allows us to use the formulation of Ref. [10] to obtain the time dependent flux in any direction; we can use a linear combination of regular and irregular Coulomb functions for the energy eigenstates out to 10^4 a.u. From the time dependent wave function, $\Psi(\mathbf{r}, t)$, we calculate the angular dependence and the time dependence of the electron flux through a sphere of radius 10^4 a.u. A classical calculation uses the angle and time dependent flux at small distances $\sim 10^4$ a.u. as input to give the flux into the detector which is at $\sim 10^9$ a.u. from the atoms. The fraction of the total autoionizing flux that enters the streak camera mirrors the total flux.

To illustrate the differences in electron emission flux after excitation of the $3s \rightarrow 3p$ transition with a laser pulse of 2 ps, various $3s16k$ states are probed and compared to low-l states in fields of 1400 and 1600 V/cm. The characters in Fig. 4 are the experimental positions indicated in Fig. 2. In Figs. 4B and 4C the electron emission after sudden ICE is depicted as a function of time, starting

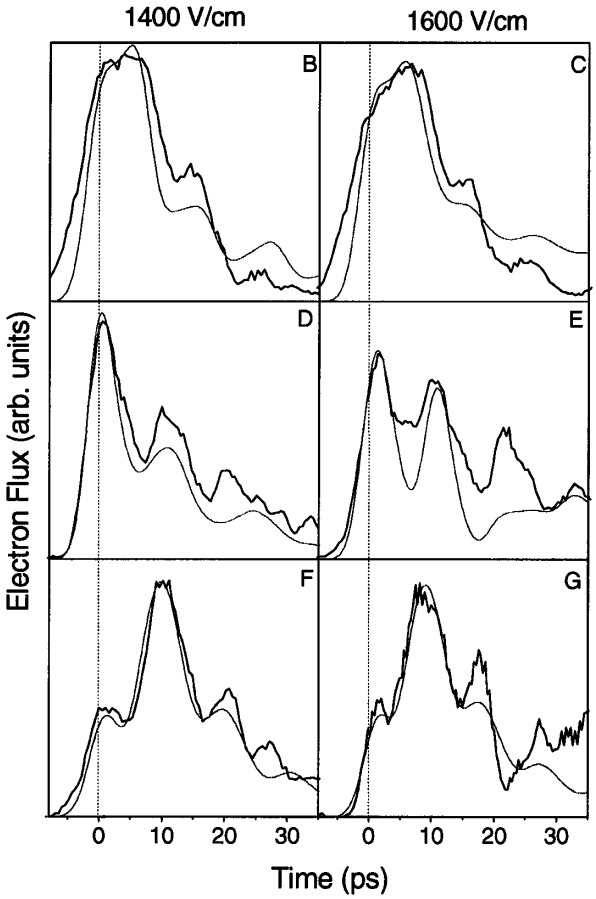

FIG. 4. The outgoing electron flux starting from various states in fields of 1400 and 1600 V/cm. One can see a clear difference between the flux after excitation of an initial Stark state on one hand (B, C) and starting from the $17d$ (D, E) and $17p$ (F, G) states on the other hand. The letters refer to the labels in Fig. 2. All show the same period which is 11 ps at 1400 V/cm and 9 ps at 1600 V/cm. The smooth lines are the calculations.

from a red Stark state. From this state we observe stepwise emission such as in Fig. 3. There is no qualitative difference in the observed flux starting from the red Stark states. Only the reddest state shows a less clear stepwise emission. All spectra show the same step period of 11 ps at 1400 V/cm and 9 ps at 1600 V/cm. Blue Stark states were too weak to be observed. The ICE calculations (smooth lines) qualitatively agree with the experimental data. We observe the same period of decay. Intensity differences might be due to difficulties in simulating the time smear of the detector.

If the initial state is the $17d$ state (Figs. 4*D* and 4*E*), the electron emission, after sudden ICE, appears dramatically different. Unlike a Stark wave packet, which has a constant fraction of low-l character, this wave packet has, at $t = 0$, predominantly d character. The angular momentum starts to oscillate in the electric field up to $n - 1$ and back. Since only the low-l character can autoionize, the emitted flux exhibits recurrences separated by the oscillation period. The decay behavior is reminiscent of the decay of a one-electron autoionizing Stark wave packet [7]. The experiments and calculations agree well in the first 20 ps and start to deviate at later times.

Also starting from the $17p$ state (Figs. 4*F* and 4*G*) the electron emission after ICE does not show the stepwise decay such as from an initial Stark state. The electron emission shows recurrences similar to those from an initial d state, with the same periods. There is no intuitive reason that the second peak of electron flux should be larger than the first. Note the fact that the p state lies right in the Stark manifold but its electron emission in time is still different from the Stark states that are only a few wave numbers away. This indicates that the p state indeed does not have strong Stark character.

In conclusion, we have performed sudden isolated core excitation of highly excited Rydberg Stark states and observed a stepwise electron emission. The time dependent electron flux from an initial Stark state after ICE is qualitatively different than from an initial low-l state, such as a p or d state. A Stark state has, after sudden ICE, a constant low-l fraction giving a constant autoionization rate during one angular oscillation period followed by a drop in outgoing electron flux. The electron emission shows a stepwise decay. A low-l state has predominantly low-l character after ICE and oscillates between high and low angular momentum, resulting in recurrences of electron emission separated by the angular oscillation time.

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