Ionization of very-high-*n* Rydberg atoms by half-cycle pulses in the short-pulse regime

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The ionization of potassium Rydberg atoms with $n\sim388$ and $n\sim520$ by pulsed electric fields whose durations, which range from ~2 to ~110 ns, span the transition from the short-pulse to the long-pulse regime is investigated experimentally and the data compared with the results of classical trajectory Monte Carlo calculations. We observe excellent agreement between theory and experiment providing a benchmark test for the validity of the classical limit of ionization. Very-high-*n* atoms provide an excellent tool with which to probe the response of atoms to pulsed electric fields in the short-pulse regime. [S1050-2947(96)50305-7]

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The ionization of atoms in high-lying Rydberg states by pulsed electric fields has been investigated extensively. In the majority of these studies the rise time and width of the applied pulse are much greater than the classical Kepler orbital period of the Rydberg electron, given by $T_n = 1.5\times10^{-16}n^3$ s, where n is the principal quantum number. In such a slowly varying field, ionization results primarily from tunneling through the potential barrier generated by the atomic and applied fields or from over-barrier escape [1–3]. The field required to induce such ionization decreases rapidly with increasing n, scaling as $1/n^4$. In contrast, application of a very short electric-field pulse with duration $T_p < T_n$ lowers the Coulomb barrier only momentarily. Ionization can still occur, however, if the impulse delivered to the excited electron by the pulsed field is sufficient to increase its energy by an amount that is greater than its original binding energy. Consequently, the n scaling of the ionization threshold crosses over from the $1/n^4$ dependence characteristic of the adiabatic limit to a $1/n$ dependence in the limit of ultrashort pulses, $T_p \ll T_n$ [4].

Ionization in the short-pulse regime has been investigated using short, fixed-duration (~0.5 ps) unidirectional electromagnetic field pulses, termed half-cycle pulses (HCPs), produced by illuminating photoconductive semiconductor switches with output pulses from a femtosecond laser system [5,6]. These experimental advances have stimulated a large number of theoretical investigations [4,7–13] of excitation and ionization by HCPs. Measurements on sodium Rydberg atoms with $13\lesssim n\lesssim35$ suggest that in the range $0.1 \lesssim T_p/T_n \lesssim 1$ the threshold field for ionization varies approximately as $\sim1/n^2$. Theoretical analyses reproduce this behavior, but detailed comparisons between theory and experiment are difficult because of uncertainties inherent in determining the absolute amplitude and shape of the HCPs [14]. Unresolved discrepancies between experiment and theory have raised questions as to the validity of the classical trajectory Monte Carlo (CTMC) method [4,7] and of the classical-quantum correspondence for high n levels. Moreover, since most quantum calculations for ionization to date [4,7,9,11,12] are performed for hydrogen rather than for alkali-metal atoms, possible deviations due to core effects have to be considered.

In the present work, we have minimized uncertainties in the pulse height and shape by undertaking measurements at very much higher n, $n\gtrsim400$. For such n, the classical electron orbital period is quite large, allowing ionization to be investigated, even in the short-pulse regime, using conventional fast pulse generators. Here we report studies of the ionization of potassium Rydberg atoms with $n\sim388$ ($T_n \sim9$ ns and $n\sim520$ ($T_n\sim21$ ns) by pulsed electric fields whose durations T_p, which range from ~2 to ~110 ns, span the transition from the short-pulse to the long-pulse regime, yet allow them to be accurately characterized. The fields required to induce ionization increase rapidly as T_p is reduced below T_n. The measured ionization probabilities are compared with the results of fully three-dimensional CTMC calculations, and the agreement on an absolute scale and without any adjustable parameters is excellent.

The present apparatus is shown schematically in Fig. 1 and is similar in many respects to that used in earlier studies of very-high-*n* atoms in this laboratory [15]. Rydberg atoms are created by photoexciting potassium atoms contained in a collimated, thermal-energy (600 K) beam using the focused output of an intracavity-doubled CR699-21 dye laser. Excitation occurs at the center of an interaction region defined by three pairs of copper electrodes, each 10×10 cm^2. Use of large electrodes, well separated from the experimental volume, minimizes the effect of patch fields associated with nonuniformities in the electrode surfaces. Any residual fields are locally reduced to $\lesssim50$ μV cm^{-1} by application of small bias potentials to the electrodes [16].

The HCP is created by applying a voltage pulse to the electrode indicated in Fig. 1, a circular copper disk 4 cm in diameter inset into the upper electrode. The voltage pulse is produced by an Avtech model AVI-V pulse generator that can provide output pulses of ~25 V amplitude with widths adjustable from $T_p\sim2$ to ~110 ns. To maintain a constant pulse shape, the amplitude of the pulse applied to the HCP electrode is varied using broadband attenuators connected in

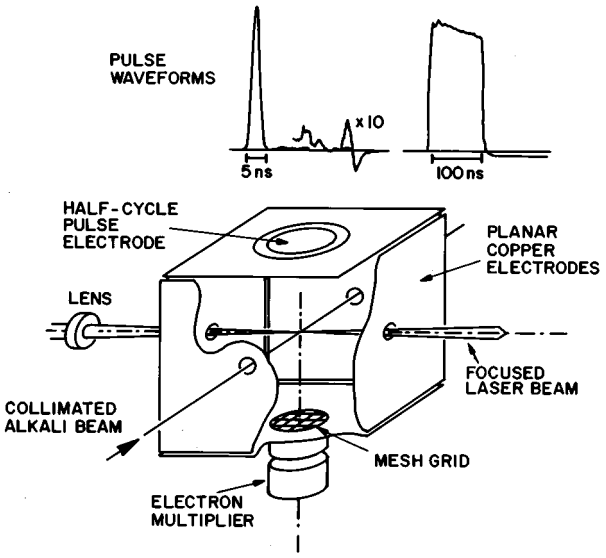

FIG. 1. Schematic diagram of the apparatus. The inset shows the profiles of the shortest and longest HCPs used in this work.

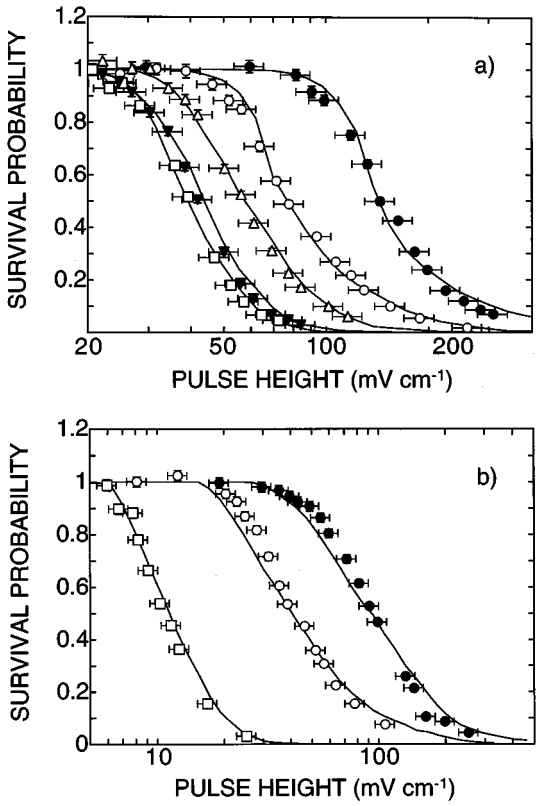

FIG. 2. Measured and calculated survival probabilities for atoms with (a) $n\sim388$ and (b) $n\sim520$ as a function of pulsed field amplitude for HCPs of $\sim$2 ns (●), $\sim$5 ns (○), $\sim$10 ns (△), $\sim$20 ns (▼), and $\sim$110 ns (□) duration.

series with the pulse generator. The pulse shape and amplitude at the HCP electrode are directly measured using a high-bandwidth dc–3.5 GHz probe and fast sampling oscilloscope. The applied pulses contain Fourier components whose frequencies ($\lesssim$350 MHz) are all much smaller than the characteristic frequencies associated with the cavity created by the interaction region electrodes (the lowest accessible mode in the present configuration is $\sim$2.1 GHz). Calculations show that under these conditions the electric field generated in the experimental volume by the pulse essentially maps the applied voltage and is very nearly equal to that expected for application of a dc potential of the same size [17]. Spatial variations in the field produced by the HCP electrode, and uncertainties inherent in the measurement of the HCP amplitude, introduce a small uncertainty, $\lesssim\pm10\%$, in the applied fields. The measured profiles of the longest and shortest HCPs used in the present work are included in Fig. 1. The applied voltage does not immediately return to zero following each HCP, but this small residual offset is taken into account in the calculations, and its effects are discussed later.

Measurements are conducted in a pulsed mode. The laser output is formed into a train of pulses of ~4 μs duration with a pulse repetition frequency of $\sim$10 kHz. (The probability that a Rydberg atom is formed during any laser pulse is very small, $\lesssim$0.01, and data must be accumulated following many laser pulses.) Excitation occurs in (near) zero electric field. Approximately 200 ns after the end of the laser pulse the HCP is applied. The number and excited-state distribution of Rydberg atoms remaining in the excitation region are determined, after a time delay of 6 μs, by selective field ionization (SFI). The time delay discriminates against free low-energy electrons resulting from ionization by the HCP which, tests revealed, have a residence time of $\lesssim5$ μs in the interaction region. For SFI, a slowly varying voltage ramp is applied to the bottom interaction-region electrode. Electrons resulting from field ionization are accelerated to, and detected by, a particle multiplier. Because atoms in different Rydberg states ionize at different applied fields, measurement of the field ionization signal as a function of time, i.e., of applied field, provides a measure of the excited-state distribution of those atoms present at the time of application of the ramp. Measurements in which no HCP is applied are interspersed at routine intervals during the data acquisition to monitor the rate of production of Rydberg atoms and their subsequent decay. The Rydberg atom survival probability is obtained by comparing the Rydberg atom signals observed with and without HCPs present. This approach eliminates possible systematic errors associated with, for example, drifts in laser power or Rydberg atom destruction by collisions with background gas.

The measured survival probabilities for atoms with $n\sim388$ and $n\sim520$ are shown in Fig. 2 as a function of the pulse height for several different durations of the HCP and are compared with the classical theory. The calculation uses the actual measured HCP profile and incorporates the estimated initial-state distribution after the laser excitation. Because of the presence of small stray fields, at $n\sim388$ it is assumed that p states with a statistical distribution of m are formed. This corresponds in the CTMC method to a subset of a microcanonical ensemble with the classical angular momentum restricted to $[l,l+1]$ with $l=1$. For $n\sim520$ the presence of the residual field causes states in adjacent Stark manifolds to overlap, and a mixture of Stark states is assumed. Tests revealed, however, that the theoretical predictions are not particularly sensitive to the exact choice of parent state or m distribution (at most 15% in the field). Each

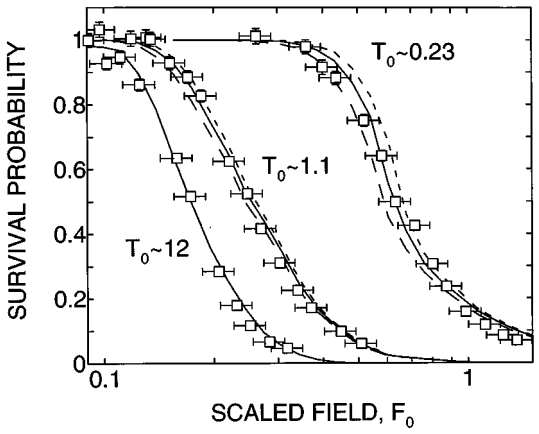

FIG. 3. Calculated survival probabilities as a function of the scaled pulse field amplitude $F_0=n^4F$ (a.u.) for different $T_0=T_p/T_n$ and varying cutoff: $n_c=800$ (– – –); $n_c=1200$ (—), $n_c=\infty$ (- - -). Experimental data (□) are included for comparison.

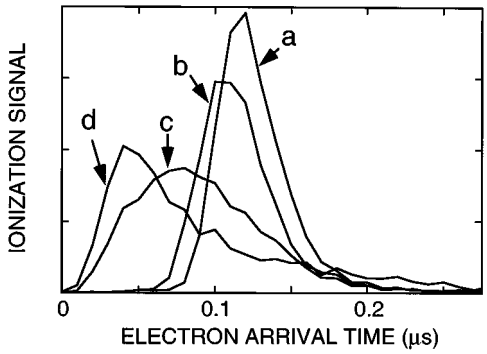

FIG. 4. SFI profiles observed for (a) parent np atoms with $n\sim388$, and following application of HCPs of (b) ~110 ns, (c) ~10 ns, and (d) ~2 ns duration.

initial condition in phase space is propagated according to Hamilton's equation of motion taking into account the experimentally determined time dependence of the HCP field, and the final energy E of the evolved electron is determined. True ionization corresponds to $E>0$ while $E_n<E<0$ corresponds to excitation to higher Rydberg states. The interaction between the electron and the K^+ core is represented by a model potential that yields accurate quantum defects and satisfies the correct boundary conditions at small and large distances. However, core effects are found to be negligible for the present high-n values.

The excellent agreement between theory and experiment demonstrates the validity of a classical description of ionization, i.e., the classical limit is reached for large n. An immediate consequence is that the experimental data satisfy the classical scaling invariance. Plotted as a function of the scaled field strength $F_0=n^4F$ (in a.u.), the survival probabilities display a universal behavior governed only by the scaled duration $T_0=T_p/T_n$ of the pulse (see Fig. 3). The monotonic dependence by the survival probability on F_0 indicates that semiclassical path interference effects are negligible. The latter become visible for lower n and polarized initial states [7,10,13]. Since the ''frequency'' of oscillations associated with such interferences scale as $\sim n$, they are averaged out as $n\rightarrow\infty$.

The calculations indicate that as T_p/T_n changes from the adiabatic to the sudden limit impulsive energy transfer to the electron increases. To verify this experimentally, SFI spectra were recorded after application of a number of different HCPs to Rydberg atoms with $n\sim388$ and the resulting data are presented in Fig. 4. In each case, the amplitude of the HCP pulse was adjusted to achieve ~10% ionization, thereby providing the best opportunity to observe possible energy transfer (i.e., excitation). In the absence of an applied pulse, a single relatively narrow SFI feature is observed that corresponds to ionization of parent atoms with a well-defined initial energy. The SFI profile is little changed following application of a HCP ~110 ns long, demonstrating that in the long-pulse regime ($T_p/T_n\sim12$) very little energy is transferred to the excited electron. In fact, highly excited states cannot exist in the presence of the HCP because the electron has enough time to escape the atom over the potential barrier. The situation changes dramatically, however, when $T_p/T_n\lesssim1$. The SFI profile measured following application of a HCP pulse ~10 ns long ($T_p/T_n\sim1$) is significantly broader than that for the parent atoms and extends towards lower ionizing fields, pointing to significant population of higher-n states. The production of high-n states becomes even more pronounced as T_p is decreased further. Indeed, after application of a HCP ~2 ns long ($T_p/T_n\sim0.2$) the peak SFI signal is observed at field strengths well below those characteristic of parent state ionization.

One consequence of impulsive excitation to n levels much higher than those initially prepared by the laser is that the measured ionization probability may be larger than the true ionization probability because small residual fields present after the HCP can ionize very-high-n Rydberg states. The importance of excitation to higher-n states is illustrated in Fig. 3, which shows the apparent survival probability for different scaled fields F_0 and durations $T_0=T_p/T_n$, assuming cut-off quantum numbers n_c for field ionization of $n_c=800$, 1200, and ∞. Clearly, in the short-pulse regime, the presence of residual fields sufficient to ionize very-high-n atoms can introduce a systematic error in measured threshold ionization fields, especially when $T_p/T_n\ll1$. This effect is included in the calculated survival probabilities shown in Fig. 2.

The present work demonstrates that very-high-n atoms provide an excellent vehicle with which to probe the response of atoms to applied fields in the short-pulse regime and show that for high n ($n\gtrsim400$) the classical limit of ionization is reached and quantum corrections are negligible. CTMC theory is able to accurately predict absolute ionization and survival probabilities. The ionization probabilities satisfy the classical scaling invariance and display the predicted crossover from a $1/n^4$ to a $1/n$ scaling in threshold fields as the pulse duration changes from the adiabatic to the sudden limit. In the future, high-n atoms will allow detailed study of atomic response to well-defined sequences of applied field pulses. For example, the evolution of the perturbed wave packet can be investigated by a ''pump-probe'' scheme if the frequency of the pulse sequence is matched to orbital frequencies (or fractions thereof).

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