Very-high-n Stark wave packets generated by an electric-field step

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Wave packets comprising a coherent superposition of $n \simeq 390$ Stark states have been created in potassium by rapid application of a dc field. Their properties are examined using a half-cycle probe pulse that is applied following a variable time delay and that ionizes a fraction of the excited atoms. The survival probability exhibits pronounced oscillations (quantum beats) that are associated with the time evolution of the wave packet. Interestingly, even in the present regime of complete overlap of different Stark manifolds, a single dominant beat frequency is observed, and this is explained quantum mechanically in terms of energy-level statistics. Similar behavior is predicted by classical trajectory Monte Carlo simulations that reproduce well the experimental observations demonstrating classical-quantum correspondence in high-n systems. [S1050-2947(97)50802-X]

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In recent years there has been increasing interest in the study of wave packets formed from a coherent superposition of high-lying Rydberg states. Such wave packets frequently display novel dynamical behavior that mimics the classical motion of the excited electron, thereby providing a bridge between quantum and classical physics. Rydberg wave packets were first created by photoexcitation of ground-state atoms using ultrashort laser pulses whose bandwidth exceeded the level spacing in the final Rydberg manifold [1–3]. Recent studies, however, have shown that Rydberg wave packets can also be formed by excitation from a single initial (stationary) high Rydberg state using a so-called ''half-cycle pulse'' (HCP) [4,5], a pulsed unidirectional electric field whose duration is much shorter than the classical electron orbital period of the initial state [6–8]. We demonstrate here that Rydberg wave packets comprising a coherent superposition of Stark states with very large values of principal quantum number n ($n \sim 390$), can be generated by rapid application of a dc field. The time evolution of the resulting wave packet is examined using a half-cycle probe pulse that is applied following a variable time delay and that ionizes a fraction of the atoms. The survival probability exhibits pronounced oscillations (quantum beats) that have a single dominant frequency.

Quantum beats from a coherent superposition of Stark states with $n \sim 23$ have been observed previously using picosecond laser pulses [9] and with $n = 2$ in beam-foil spectroscopy [10]. The present study using very-high-n atoms is unique both from an experimental and from a conceptual point of view. Classically, a simple quasiperiodic evolution of the wave packet is expected corresponding to the precession of the angular momentum and the Runge-Lenz vector of the electron in the electric field. However, it is not obvious *a priori* that quantum mechanics will predict such simple behavior because the present dc fields are sufficiently strong that many n manifolds overlap, resulting in a broad range of energy-level separations. Nonetheless, our calculations show that in the high-n limit the weighted quantum-statistical distribution of eigenenergies is such that the quantum evolution closely mirrors the classical limit.

The apparatus used in the present experiments is described in detail elsewhere [5]. Briefly, K(np) Rydberg atoms are created by photoexciting ground-state potassium atoms in a thermal-energy beam using a frequency-doubled Coherent CR699-21 dye laser. Excitation occurs near the center of an interaction region (defined by three pairs of planar copper electrodes each 10×10 cm^2), where the residual field can be locally reduced to $\lesssim 50$ μV cm^{-1} [11]. To minimize motional electric fields, the magnetic field is reduced to $\lesssim 20$ mG by use of μ-metal shields. Measurements are conducted in a pulsed mode. The laser output is formed into a train of pulses of 4-μs duration using an acousto-optic modulator. Excitation occurs in (near) zero electric field and the laser is tuned to create np Rydberg atoms with $n \approx 388$, for which the classical orbital period T_n is ≈ 9 ns. However, the probability that a Rydberg atom is formed during any laser pulse is small, $\lesssim 0.02$, and data must be accumulated over many laser pulses. After each laser pulse, a ''rectangular'' voltage pulse of several microseconds duration with a rise time adjustable down to ≈ 10 ns is applied to a circular electrode 4 cm in diameter that is inset into the upper large electrode. This pulse establishes a dc field in the experimental volume. However, because the pulse must be ac coupled (to remove dc offsets present in the output of the pulse generator), its amplitude (and that of the applied field) decreases by $\sim 10\%$ over a typical measurement period of 500 ns. The time development of the Stark wave packet produced by application of the ''dc'' field is examined using a half-cycle probe pulse of duration $T_p \approx 2.4$ ns full width at half maximum (FWHM) that is applied following a variable time delay. The probe HCP is superposed on the ''dc'' pulse using a power combiner. The pulse shapes and amplitudes at the circular electrode are directly measured using a fast probe and sampling oscilloscope. The number of Rydberg atoms

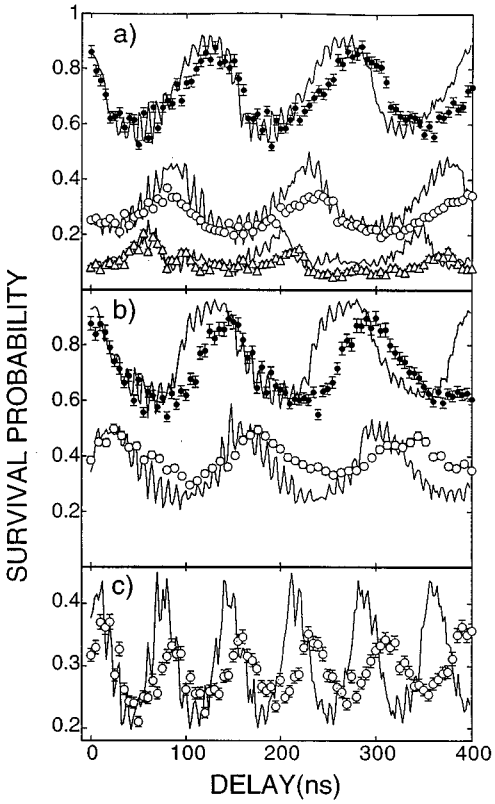

FIG. 1. Rydberg-atom survival probability as a function of time delay between application of the ''dc'' and half-cycle pulses for ''dc'' fields with a 10-ns rise time and amplitudes of (a), (b) 5 mV cm^{-1} and (c) 10 mV cm^{-1}. In (a) the ''dc'' and HCP fields are in the same direction; in (b) and (c) they are in opposite directions. ⏚, ⏚, and ⏚, experimental data obtained with HCP amplitudes of ~80, ~170, and ~290 mV cm^{-1}, respectively; —, results of the CTMC calculations.

remaining in the experimental volume after application of the HCP is determined by field ionization [5]. Measurements with no HCP's applied are interspersed at routing intervals during data acquisition to monitor the number of Rydberg atoms initially created by the laser. The Rydberg atom survival probability is determined by taking the ratio of the Rydberg atom signals observed with and without HCP application.

Data obtained with a ''dc'' field of ~5 mV cm^{-1} and 10-ns risetime are presented in Fig. 1, which shows the survival probability as a function of the time delay between application of the ''dc'' and the HCP pulses for different HCP amplitudes and polarities. (Classically, the dc field required to ionize Rydberg atoms with $n\sim388$ is ~14 mV cm^{-1}.) The error bars indicate the statistical uncertainties and do not include possible systematic errors associated with uncertainties in the applied field amplitudes, estimated to be $\lesssim\pm10\%$. Sizable periodic oscillations (quantum beats) in the survival probability are evident, which, measurements show, continue for at least 1.5 μs. The positions of the maxima and minima depend on the size of the HCP as does the overall survival probability. Figure 1 includes results obtained with a ''dc'' field of ~10 mV cm^{-1} (and 10-ns rise time), which demonstrate that the quantum beat frequency is proportional to the magnitude of the ''dc'' field.

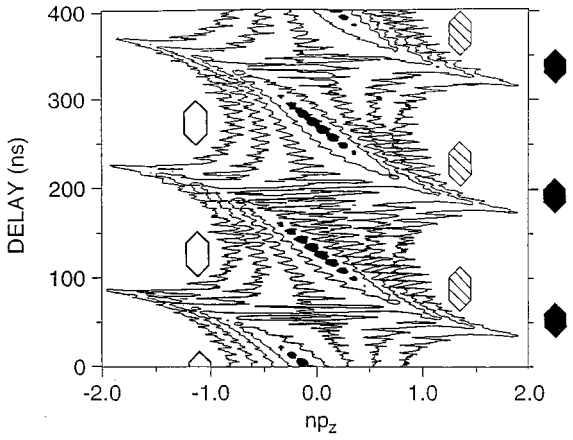

FIG. 2. Time evolution of the distribution of the z component, p_z, of electron momentum, plotted as np_z, following application of the ''dc'' pulse. The contours denote 0.04, 0.08, 0.16, and 0.32 of the peak value. Regions above 0.64 of the peak value are shown in black. The vertical bars are discussed in the text.

Figure 1 also shows the results of classical trajectory Monte Carlo (CTMC's) calculations [12] that use the actual measured ''dc'' and half-cycle pulse profiles and that incorporate the estimated initial excited-state distribution present immediately after laser excitation (p states with a statistical distribution of m) [5,8]. The interaction between the electron and K^+ core ion is represented by a model potential that yields accurate quantum defects and satisfies the correct boundary conditions at small and large distances. Even without recourse to any adjustable parameters, the general agreement between theory and experiment is good. The calculations reproduce well the observed beats and the small differences in magnitude and period can be attributed to the uncertainty inherent in determining the value of the ''dc'' and HCP fields. Classically, the time development of the survival probability is governed by two quite dissimilar frequencies: one corresponding to the classical electron orbital period in the atom ($w_{\rm orb}\simeq n_i^{-3}$), the other the classical precession of the electron orbit in the applied electric field F, whose frequency is given in first-order hydrogenic Stark theory by $w_s=3nF$. For $n\sim388$ and $F=5(10)$ mV cm^{-1}, this corresponds to quantum beat periods $T_s\sim134(67)$ ns, which are close to the values measured experimentally. Theory also predicts high-frequency oscillations, with period $2\pi n_i^3\sim9$ ns, due to electron orbital motion. The experimental data, however, contain only a hint of these oscillations, possibly because of small field inhomogeneities in the experimental region.

The periodic oscillations in the survival probability can be explained classically in terms of the time evolution of the z component, p_z, of the momentum of the excited electron (the z axis is taken to be parallel to the dc field). Figure 2 shows the calculated distribution of the scaled electron momentum, np_z, as a function of time following the application of the ''dc'' pulse. At early times, the distribution is sharply peaked at small negative values of p_z. As time advances, the distribution broadens. The peak moves toward more negative values and decreases in size before finally disappearing. A small peak then appears in the distribution at positive values of p_z that grows and moves toward $p_z=0$ as the distribution

narrows. Ultimately the peak crosses to negative values of p_z, and the whole cycle repeats.

Consider now the effect of applying the half-cycle probe pulse to the evolving system. Since the HCP is short, it delivers an impulsive momentum transfer $\Delta p_z = -\int_{-\infty}^{\infty} F_z(t)dt$ to the excited electron where $F_z(t)$ is the electric field generated by the HCP. The resulting energy transfer is given by $\Delta E=(\Delta p_z)^2/2+p_z\Delta p_z$. If $p_z\Delta p_z>0$, i.e., if the impulse delivered by the HCP is in the same direction as the initial z component of electron momentum, $\Delta E>0$ and this increase in electron energy can lead to ionization. The situation is more complex when $p_z\Delta p_z<0$. If $|\Delta p_z|$ is small, the final energy of the electron will typically be lower. However, for sufficiently large $|\Delta p_z|$, ΔE becomes positive, i.e., the $(\Delta p_z)^2/2$ term is dominant; thus large HCPs can induce ionization even if $p_z\Delta p_z<0$. Consider now application of HCPs for which, as for the data in Fig. 1(a), $\Delta p_z<0$. If the impulse is small, only atoms initially having large negative p_z can be ionized. The overall survival probability should therefore be large, and should be maximum at those times indicated by the unshaded bars in Fig. 2 where the p_z distribution has its smallest negative excursions. These times correspond well with the maxima in the upper data set in Fig. 1(a). In contrast, for large HCPs all but atoms with large initial positive p_z will be ionized. The overall survival probability is therefore low, but should be greatest at those times indicated by the solid bars in Fig. 2 where the p_z distribution has its largest positive values. These times match well the maxima in the lower data set in Fig. 1(a). For intermediate HCP amplitudes, the cutoff for ionization will be near $p_z\sim 0$. Atoms with $p_z\lesssim 0$ will be ionized and the survival probability should be greatest at those times indicated by the shaded bars in Fig. 2 where the p_z distribution peaks at positive p_z; these times correspond well with the maxima in the middle data set in Fig. 1(a). Similar arguments can be used to explain the data in Fig. 1(b), for which $\Delta p_z>0$. For example, if a HCP of intermediate amplitude is employed, the survival probability should be *smallest* at times near those shown by the shaded bars in Fig. 2, as is observed.

The effect of varying the rise time of the ''dc'' pulse is illustrated in Fig. 3. Quantum beats are observed even when the rise time becomes greater than the final quantum beat period, i.e., when the bandwidth associated with the turn on of the ''dc'' pulse becomes less than the final spacing between adjacent Stark levels (of the same n). This is not unexpected, however, because the level separation depends on applied field and is much smaller at early times during the turn on of the ''dc'' field.

Quantum mechanically, if the rise time of the ''dc'' pulse is very short, the coherent superposition of Stark states can be written in the sudden approximation as

$$|\Phi(t)\rangle \simeq \sum_{\alpha} e^{-i\epsilon_\alpha t}\langle\alpha|i\rangle|\alpha\rangle \qquad (1)$$

where ϵ_α and $|\alpha\rangle$ are the eigenenergies and eigenvectors in the dc field and $|i\rangle=|n_i l_i m\rangle$ is the initial Rydberg state. However, only states with a narrow distribution of n values $(\Delta n\lesssim 4)$ centered at $\sim n_i=388$ contribute significantly to the summation in Eq. (1). Expressed in parabolic quantum numbers, the eigenenergies for hydrogen are given to first order by

$$\epsilon_\alpha = -\frac{1}{2n^2}+\frac{3}{2}Fn(n_1\text{-}n_2). \qquad (2)$$

The relevant energy differences governing the time development of the wave packet are given therefore in terms of the classical frequencies $w_{\rm orb}$ and w_s approximately by

$$\Delta\epsilon \approx w_{\rm orb}\delta n + w_s k, \quad k=1,2,\ldots,(n_i-|m|-1). \qquad (3)$$

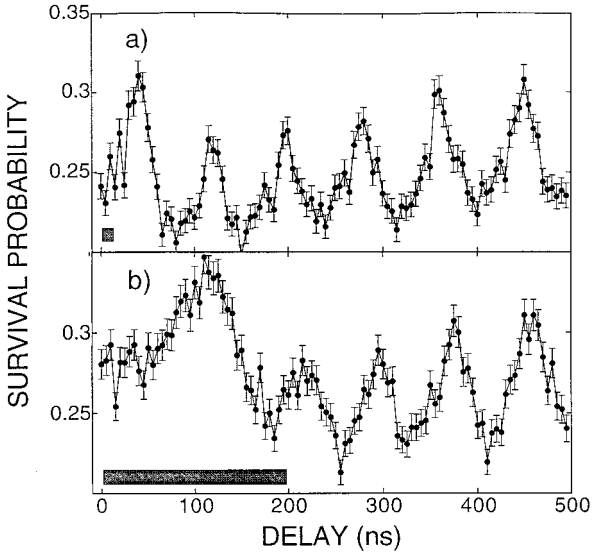

FIG. 3. Rydberg-atom survival probability as a function of time delay between the onset of the ''dc'' pulse and application of the HCP for a ''dc'' pulse amplitude of 10 mV cm^{-1} and rise times of (a) 10 ns and (b) 200 ns.

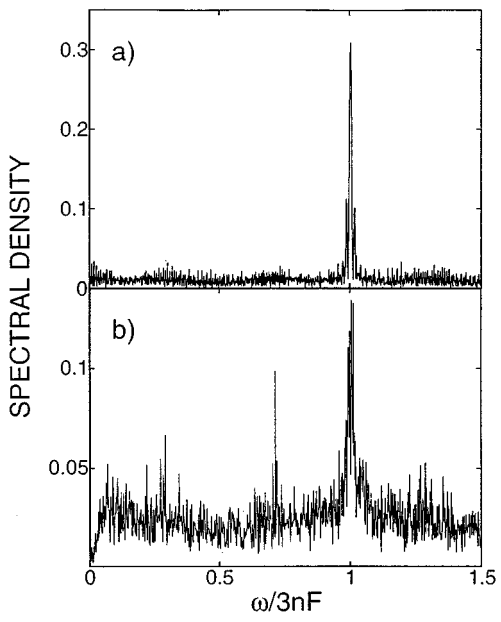

FIG. 4. Weighted statistical spectrum of frequencies near $n\sim 100$ for (a) hydrogen and (b) potassium in an applied dc field of 2 V cm^{-1}.

The constant energy differences are reminiscent of a harmonic oscillator for which quantum and classical expectation values agree.

For potassium and the present dc field strengths Eq. (3) breaks down because many Stark manifolds overlap and mixing occurs. To determine the quantum-mechanical origin of a single dominant Stark frequency in the fully mixed regime, we analyzed the weighted statistical spectrum of frequencies $(\epsilon_\alpha - \epsilon_\beta)$ near $n \simeq 100$ for a few thousand Stark levels and an applied field of 2 V cm^{-1}, sufficient to cause overlap of several adjacent n manifolds. Motivated by Eq. (1) we used as weight the overlap factor with the initial state $|\langle\alpha|i\rangle\langle i|\beta\rangle|$ (only those Stark states that have a significant overlap with the initial state should carry a sizable occupation amplitude). The resulting frequency spectrum for potassium is strongly peaked and, as shown in Fig. 4, closely resembles that of hydrogen, i.e., the peaks are as given by Eq. (3). (These peaks would not be present had an unbiased distribution of nearest spacings been used.)

The dephasing of the Stark wave packet is directly related to the width of the peak at $\Delta\epsilon = w_s = 3nF$ in the frequency spectrum, which is somewhat larger for potassium than for hydrogen. Thus, quantum mechanics predicts that dephasing, i.e., damping, of the quantum beats will be more rapid for potassium. Classically, core effects are to be expected because, during each Stark period T_s, the electron angular momentum changes from $l=1$ to $l=n-1$ and back. The electron penetrates the core during every Stark period thereby introducing small deviations from hydrogenic behavior. However, even for potassium, the time scale for damping is long, extending over many Stark periods. This suggests that it might be possible to observe external damping induced by dephasing in collisions with a target gas. If so, such studies would provide a new tool for investigating elastic electron-molecule scattering at electron energies down to a few micro-electron-volts.

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