Observation of the Collapse and Revival of a Rydberg Electronic Wave Packet

John A. Yeazell, Mark Mallalieu, and C. R. Stroud, Jr.
Institute of Optics, University of Rochester, Rochester, New York 14627
(Received 20 November 1989)

We report an observation of the long-term evolution of a radially localized electronic wave packet formed by the coherent superposition of Rydberg states of atomic potassium. Initially, the wave packet can be described classically. Subsequent dephasing of the discrete states in the superposition leads to a loss of spatial localization so that the evolution can no longer be described classically. However, the wave packet revives at a later time. Theory and experiment show good agreement including an accurate measurement of the phase shift of the wave packet on revival.

PACS numbers: 32.90.+a, 31.50.+w, 32.60.+i

Quantum-mechanical wave-packet states can approach the classical ideal of a spatially localized particle traveling along a well-defined trajectory. The uncertainty principle places a limitation on this localization. But, for bound systems excited appreciably above their ground states, this is not a very stringent limitation. A more serious limitation on the quasiclassical nature of the states is the fact that for most systems the wave packet does not remain localized, but rapidly spreads.

Normally, as time goes on the wave packet continues to spread and becomes less and less classical in its nature. There are a few cases, however, in which the spreading reverses itself and the wave packet relocalizes, again approximating a particle moving on a classical trajectory. A closely related decay and revival of classical coherence has been predicted,[1-3] and observed[4] recently in the micromaser realization of the Jaynes-Cummings problem. There it is the coherent Rabi oscillations of the inversion of a Rydberg atomic transition that decay and revive. In both cases the revival is possible only due to the discreteness of the quantized energy states. In the Jaynes-Cummings problem it is the quantized nature of the cavity field, while in the present case it is the quantized nature of the atomic energy. A second necessary feature in both cases is the fact that the coherent superposition state is made up of frequency components that are almost equally spaced.

In this paper, we report the observation of the decay and revival of a spatially localized Rydberg electronic wave packet. Methods for exciting such wave packets have been reported in several papers[5-9] and several different types of these wave packets have been observed.[10-13] In this experiment, a radially localized wave packet is formed by the coherent excitation of Rydberg states by a short, optical pulse. The Rydberg states have a range of values for the principal quantum number n with an average value of $\bar{n}$. The resulting wave packet has the appearance of a shell oscillating between the nucleus and the outer turning point. The oscillations are at the classical orbital period. The classical orbital period is inversely proportional to the first derivative of the average energy of the wave packet with respect to $\bar{n}$ ($\tau_0 = 2\pi\bar{n}^3$ a.u.).[5] These oscillations have been observed experimentally for a few periods.[11,12]

The long-term evolution of the wave packet is more complex. If the Rydberg states that compose the superposition were equally spaced in energy, the localization and motion of the wave packet would continue unchanged. Such an atomic coherent state would be exactly like a coherent state of a harmonic oscillator. This wave packet would have many of the properties of a classical atom and retain them. If the states were randomly spaced in energy, then the superposition would be almost periodic with an infinite Poincaré recurrence time.[14] The wave packet would quickly spread and never revive. Any classical characteristics of the wave packet would also be quickly lost. However, the Rydberg states are neither equally nor randomly spaced. For a superposition with a large value of $\bar{n}$ compared to the number of states in the superposition, the energy levels are nearly equally spaced. The deviation from equal energy spacing causes the wave packet to spread slowly and after a time the wave packet is no longer localized. However, the nearly equal spacing and the finite number of states in the superposition results in a rephasing of the states of the superposition and the wave packet again becomes localized. The wave packet periodically loses and regains its classical appearance. The period of this decay and revival is inversely proportional to the second derivative of the average energy of the wave packet ($\tau_{dr} = \frac{2}{3}\pi\bar{n}^4$ a.u.). This temporal behavior of the wave packet has been predicted in several papers.[5,9,15,16]

The exact nature of the excitation and subsequent evolution of the wave packet can be calculated by the numerical integration of Schrödinger's equation for this system. The equations for the amplitudes of Schrödinger's equation are[5]

$$\dot{a}_n(t) = -\frac{i}{2} a_g(t)\Omega_n f(t) e^{i\Delta_n t}, \tag{1a}$$

$$\dot{a}_g(t) = -\frac{i}{2}\sum_n a_n(t)\Omega_n f(t) e^{-i\Delta_n t}. \tag{1b}$$

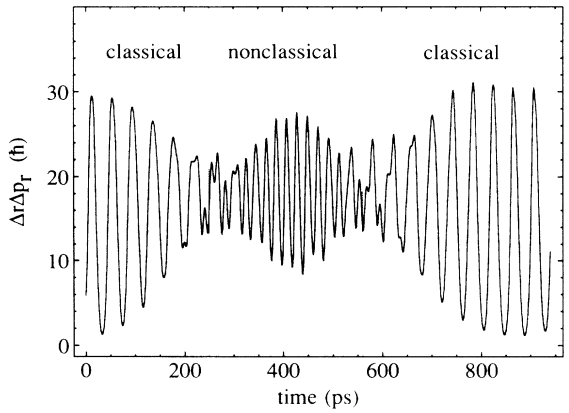

FIG. 1. The evolution of the uncertainty product of the radial wave packet. The uncertainty product reaches a minimum as it approaches the outer turning point. Near the inner turning point the wave packet spreads and the uncertainty product reaches a maximum. The oscillations of the product are damped as the wave packet disperses. However, at approximately 700 ps after the exciting pulse, the oscillations have nearly regained their original amplitude. There is also a region in which the oscillations show a doubling of the frequency. This is the result of the wave packet breaking up into two distinct parts.

The amplitudes of the ground state and the excited states are a_g and a_n, respectively. The Rabi frequencies and the detunings are denoted by Ω_n and Δ_n, respectively. The calculations were performed using a Gaussian pulse shape, $f(t)$.

The solution of these equations determines the evolution of the wave packet. This evolution is displayed in Fig. 1, which is a theoretical plot of the uncertainty product $\Delta r \Delta p_r$ as a function of time. A pulse $f(t)$ of duration 10 ps FWHM forms a wave packet made up primarily of five eigenstates centered on $\bar{n}=65$. The rapid periodic oscillations with a period of approximately 50 ps are due to the radial motion of the wave packet in its classical trajectory. In the first few orbits the uncertainty product approaches its minimum possible value $\hbar/2$, but then the wave packet decays so that the product remains greater than $10\hbar$ for the entire orbit. At this point the wave packet is spread throughout the orbit and the system is nonclassical. It can be seen in the figure that the wave packet becomes very well localized again at 700–800 ps. The atom may be said to have undergone a free evolution from a classical, to a nonclassical, and back to a classical state. Of course, the system is not really periodic, so the revival is not perfect. During the revival, all five significant states of the superposition are nearly, but not exactly, in phase. This results in a wave packet that is not quite as well localized as it was initially. Another interesting feature of the long-term evolution is the rapid oscillations that occur between the major revivals. The oscillations are at twice the orbital frequency, and can be attributed to the breakup of the wave packet into two discrete parts. During this intermediate revival, every other state of the superposition is in phase. Such an intermediate revival has been predicted by several authors and has been denoted a fractional revival.[5,9,15]

The wave packets are radially localized, but not angularly localized. There is a simple classical way of understanding this radially localized wave packet. The corresponding classical system is not a particle moving around a particular elliptical Kepler orbit. Instead, the wave packet corresponds to an ensemble of classical orbits with various orientations in space around the nucleus. The short pulse has the effect of synchronizing the phase of the electron in each classical realization, so that they are all at the same point (e.g., outer turning point) at the same time. The entire ensemble then appears to be a shell oscillating in and out from the nucleus. However, the classical model holds only while the wave packet is intact, so that the evolution of the wave packet alternates between regions of classical and nonclassical behavior.

The classical model can be used to describe the interaction of the wave packet with external fields and suggests the method used to detect the wave packet. When the electron is near the outer turning point it moves slowly so that the electron current $\mathbf{J}(t)=e\mathbf{v}(t)$ is small. On the other hand, when the electron is near the nucleus it is moving rapidly so that the corresponding current is large. If a second laser pulse is applied after the wave packet has been formed, then the rate of absorption of energy from the second pulse will be given by $R(t)=\mathbf{J}(t)\cdot\mathbf{E}(t)$. Thus, when the wave packet is near the nucleus it can efficiently absorb energy from a probe pulse and the atom will be ionized; when the wave packet is near the outer turning point it will not absorb much energy and the atom will not be ionized. This pump-probe method of detecting the wave packet was first proposed by Alber, Ritsch, and Zoller.[6]

In this experiment, the Rydberg electronic wave packet is formed in the excited states of atomic potassium. The wave packet is excited by a one-photon process from the ground state to the Rydberg series by a short, ultraviolet pulse ($\tau_{\text{pump}}=15$ ps, $\lambda=2858$ Å). The wave packet is allowed to freely evolve for a delay time, τ_d. Following this delay time, the wave packet interacts with a short, optical pulse ($\tau_{\text{probe}}=20$ ps, $\lambda=5716$ Å). The evolution of the wave packet is followed by analyzing the photoionization signal as a function of the delay time. In general, the photoionization signal is larger when the wave packet is localized near the nucleus. This technique allows us to observe not only the short-term evolution, but also the dephasing or collapse of the wave packet and its subsequent revival.

In previous experiments,[11,12] the rapid oscillations of the wave packet between the nucleus and the outer turning point have been observed in a similar manner. However, these experiments excited the wave packet by a two-photon process. The two-photon excitation led to a

large ac Stark shift of the ground state that chirped the transition frequency. This chirp only slightly modifies the short-term evolution observed in these experiments. However, this chirp would play a more significant role in the observation of the revival of the wave packet. If the Stark shift exceeds the energy separation of the states, then the visibility of the revival is reduced. Also, the same frequency was used for the pump and probe pulses in these experiments. Using the same frequency to probe the wave packet connects the wave-packet states to both the continuum and the ground state. This second interaction with the ground state can complicate the behavior of the wave packet. Both these potential problems are eliminated in the scheme described above.

The apparatus used to perform this experiment is shown in Fig. 2. A detailed description of much of this apparatus can be found in Ref. 12. Here, we will focus on the changes that have been made to implement the one-photon excitation of the wave packet and the different frequency probe of the wave packet. The synchronously pumped mode-locked, cavity-dumped dye laser produces a short, optical pulse (20 ps, 5716 Å, 10 μJ). The ultraviolet pump pulse is produced by second-harmonic generation in an 8-mm crystal of beta-barium-borate crystal (15 ps, 2858 Å, 1 μJ). The pump pulse is tuned to the single-photon resonance between the ground state and the $n=65$ p state. The remaining first-harmonic field is used for the probe pulse. The optical probe pulse passes through the dichroic mirror and enters a variable delay line. A half-wave plate is adjusted so that the polarization of the probe pulse has the same orientation as that of the pump pulse. The uv pump pulse is reflected by the dichroic mirror and directed to the second dichroic mirror where it is recombined with the delayed probe pulse. The pulses are focused by a 20-cm lens onto a beam of atomic potassium. The 1-m lens in the delay arm is used to adjust the divergence of the optical probe pulse, so that the overlap of the foci of the pump and probe pulses is maximized. The resulting focal spots are approximately 100 μm in diameter. The peak intensities of the pump and probe pulses are 400 MW/cm^2 and 3.5 GW/cm^2, respectively.

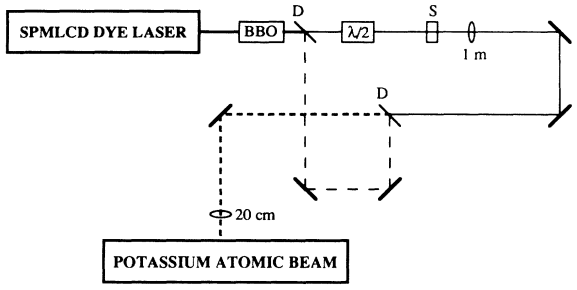

FIG. 2. A schematic diagram of the apparatus. The dye laser produces optical pulses (20 ps, 5716.8 Å) at a repetition rate of 800 Hz. The uv pump pulse is produced by second-harmonic generation in the beta-barium-borate (BBO) crystal. A dichroic mirror separates the fundamental field (probe pulse) from the second-harmonic field. The probe pulse enters a delay arm, where its polarization is adjusted, with a half-wave plate, to match that of the pump pulse. The two pulses are recombined at a dichroic mirror and then focused onto the potassium atomic beam. A 1-m lens is introduced into the delay arm to adjust the divergence of the probe pulse. A shutter is also present in the delay arm so that a background measurement may be taken without the probe pulse.

In order to produce an intense atomic beam, the beam is only weakly collimated. The beam is defined by apertures 1 mm in diameter and separated by a distance of 2 cm (divergence angle of 0.03 rad). The interaction region lies immediately behind the second aperture. The oven operates at a temperature of 280 °C, and produces a density of 10^{11} atoms/cm^3 in the interaction region.

The photoions produced by the pulses are collected and amplified by an electron multiplier. Its output is amplified and fed to a photon counter. At each setting of the delay time, the signals from 10000 pulses are averaged. The delay time is incremented in steps of 5 ps. A background signal is obtained by blocking the probe pulse and recording the photoion signal at each delay step. The variation of this background signal provides a

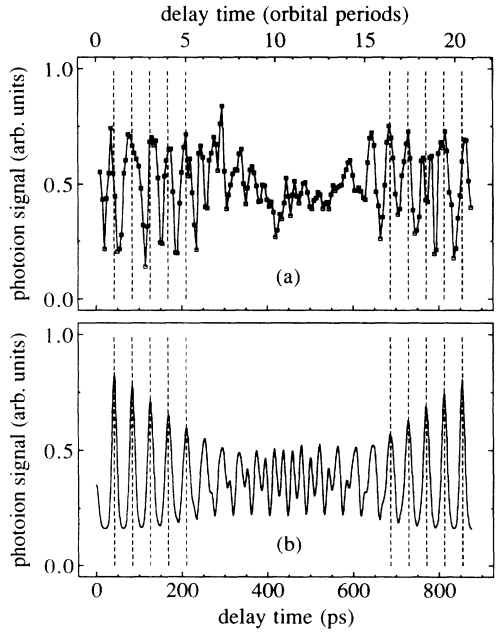

FIG. 3. The photoionization signal as a function of the delay time shows the evolution of the radial wave packet. (a) The results of the experiment. A wave packet is excited with $n=65.2$. Oscillations at the orbital period are evident. The vertical dashed lines are separated by the orbital period. The oscillations are damped, but then revive. In the revival the oscillations are again at the orbital period, but a phase shift has occurred; i.e., the vertical dashed lines no longer coincide with the peaks of the photoionization signal. (b) A theoretical prediction. The characteristics of the experimental results, discussed in text, are apparent in this prediction.

measure of the drift of atomic- and laser-beam intensities throughout the experiment.

Figure 3(a) shows the results from the excitation of a wave packet with $\bar{n} \approx 65$. The pump pulse occurs at zero time delay. For approximately five orbital periods after the pump pulse, the photoionization signal exhibits clear oscillations at the classical orbital period. The amplitude of these oscillations decay, but subsequently revive to approximately the original value. These results are compared with the theoretical predictions for such a wave packet in Fig. 3(b). Note that the orbital period and the position of the revival are in excellent agreement. A phase shift of the orbital oscillation is evident in the revival. The top horizontal axis is scaled in orbital periods. Before the wave packet decays the peaks are aligned with the tick marks, but during the revival the peaks are not aligned. The phase shift can be seen in both the theoretical and experimental results. The phase shift is an indication that the revived wave packet is not exactly the same as the original wave packet. The position of the revived wave packet in the orbit is not the position that the original wave packet would be at if the wave packet had not decayed. This orbital position or phase shift is dependent on the characteristics of the excitation. The theoretical plot was obtained with a Gaussian-shaped pump pulse (15 ps FWHM) and a slight detuning ($\bar{n}=65.2$).

The long-term evolution of the wave packet offers some other interesting features. In particular, the theoretical predictions indicated a doubling of the orbital frequency during the fractional revival of the wave packet. Other fractional revivals are also possible where the oscillations occur at higher harmonics of the orbital frequency. Experiments are currently in progress to observe these effects.

Effects of the type described here are by no means limited to Rydberg atomic states. Recently, wave packets have also been produced and their decay observed in a molecular system.[17,18] It would appear that similar behavior should also be observable in semiconductors.

We would like to acknowledge the support of the Joint Services Optics Program and the University Research Initiative through the Army Research Office. We would also like to acknowledge conversations with Jonathan Parker whose theoretical calculations stimulated much of this research.

[1]F. W. Cummings, Phys. Rev. **140**, A1051 (1965).

[2]P. Meystre, E. Geneux, A. Quattropani, and A. Faist, Nuovo Cimento **25B**, 521 (1975).

[3]N. B. Narozhny, J. J. Sanchez-Mondragon, and J. H. Eberly, Phys. Rev. A **23**, 236 (1980).

[4]G. Rempe, H. Walther, and N. Klein, Phys. Rev. Lett. **58**, 353 (1987).

[5]J. Parker and C. R. Stroud, Jr., Phys. Rev. Lett. **56**, 716 (1986).

[6]G. Alber, H. Ritsch, and P. Zoller, Phys. Rev. A **34**, 1058 (1986).

[7]J. Parker and C. R. Stroud, Jr., Phys. Scr. **T12**, 70 (1986).

[8]J. C. Gay, D. Delande, and A. Bommier, Phys. Rev. A **39**, 6587 (1989).

[9]A. ten Wolde, L. D. Noordam, H. G. Muller, and H. B. van Linden van den Heuvell, in *Fundamentals of Laser Interactions II,* edited by F. Ehlotzky (Springer-Verlag, New York, 1989).

[10]J. A. Yeazell and C. R. Stroud, Jr., Phys. Rev. Lett. **60**, 1494 (1988).

[11]A. ten Wolde, L. D. Noordam, H. G. Muller, A. Lagendijk, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).

[12]J. A. Yeazell, M. Mallalieu, J. Parker, and C. R. Stroud, Jr., Phys. Rev. A **40**, 5040 (1989).

[13]A. ten Wolde, L. D. Noordam, A. Lagendijk, and H. B. van Linden van den Heuvell, Phys. Rev. A **40**, 485 (1989).

[14]N. Wiener, *The Fourier Integral and Certain of Its Applications* (Cambridge Univ. Press, Cambridge, 1933), Sec. 24.

[15]I. Sh. Averbukh and N. F. Perelman, Phys. Lett. A **139**, 449 (1989).

[16]M. Nauenberg, Phys. Rev. A **40**, 1133 (1989).

[17]R. B. Bernstein and A. H. Zewail, J. Chem. Phys. **90**, 829 (1989).

[18]R. M. Bowman, M. Dantus, and A. H. Zewail, Chem. Phys. Lett. **156**, 131 (1989).