Classical periodic motion of atomic-electron wave packets

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An experiment is described in which a coherent superposition of the Rydberg states of atomic potassium is excited by a short optical pulse. The coherent superposition forms a wave packet localized in the radial coordinate. The radial motion of the wave packet is periodic with the period of the classical Kepler orbit. The time evolution is probed by a second short pulse. The resulting photoionization signal, as a function of the delay between pulses, shows the classical periodicity.

In the very earliest days of quantum mechanics, Schrödinger[1] and Lorentz[2] attempted to find atomic coherent-state wave packets that were localized and moved in classical Kepler orbits. Recently, several authors have shown that states closely approximating these states can be formed by use of a short laser pulse to excite a coherent superposition of Rydberg states.[3–7]

In a recent paper[8] we have reported the observation of one variety of these states in which the electron is angularly localized with the wave packet sitting stationary in space on one side of the nucleus. Although this state is quite different from the normal energy eigenstate in its properties, it does not orbit periodically like a classical Kepler particle. Quite recently, ten Wolde *et al.*[9] have reported the observation of another type of wave packet (radially localized). Our work confirms their basic results while exploring a different part of the regime, in which such a wave packet can be formed.

In this paper we report the production and detection of a Rydberg electron wave packet that is radially localized and oscillates in and out about the nucleus with an oscillation period that is exactly the orbital period of a classical particle with the same energy. The motion of this wave packet is illustrated in Fig. 1. The wave packet is of the form of a shell oscillating periodically in radius from a few Bohr radii to the classical turning point (8000 Bohr radii). A detailed theoretical analysis of the time evolution of such wave packets has been presented in Refs. 3–5. There it is shown that such wave packets can approach localization in r, the radial coordinate, and p_r the conjugate momentum, to within a few percent of the uncertainty principle limit. The wave packet spreads after several orbits but reassembles later in a form of quantum revival.

The radially localized wave packet does not represent the ideal classical limit of a single particle orbiting in a particular Kepler ellipse, because the localization is in the radial coordinate only and not in the angular coordinates. It corresponds to an ensemble of different classical particles each traveling in a classical orbit, each with exactly the same phase in its orbital motion but with the directions of the ellipses distributed about all of the different possible angular orientations in space. This classical ensemble is illustrated in Fig. 2. Each particle approaches the nucleus at the same time and each reaches the outer turning point at the same time, so the ensemble as a whole produces a radial shell wave packet oscillating at exactly the period of the individual Kepler orbits.

The detection of the wave packet makes use of the fact that the rate of absorption of energy by a particle from an optical field is given by[10] $W = e\mathbf{v}\cdot\mathbf{E}$, where $\mathbf{v}$ is the velocity of the particle, and $\mathbf{E}$ is the electric field. Thus the electrons will not absorb energy when they are near the outer turning point of the orbit moving slowly, but will absorb energy from a laser pulse applied when they are near the nucleus moving rapidly. One pulse excites the wave packet, and a second pulse, after a variable delay, probes it. If the second pulse occurs when the wave packet is near the nucleus, energy will absorbed and there will be a significant enhancement of the probability that

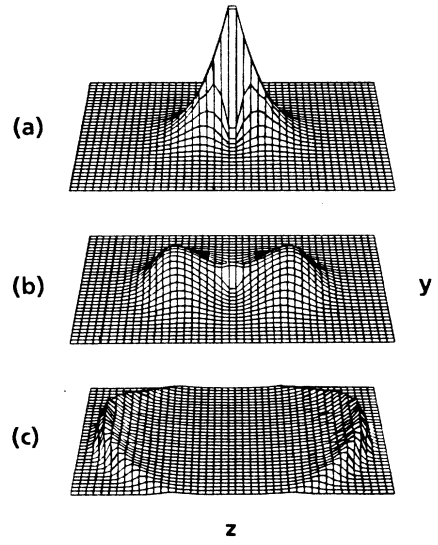

FIG. 1. The evolution of the wave packet is described by this series of plots. The projection, onto the y-z plane, of the probability density of the radial wave packet is shown ($n=85$). The distance from the nucleus to the edge of the plot is 8000 Bohr radii. The wave packet has just been formed (a). (b) At one quarter of the classical orbital period, the wave packet has moved away from the nucleus. It has reached the outer classical turning point (c) at one-half the classical orbital period.

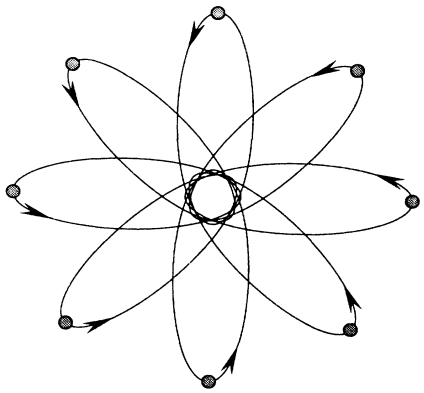

FIG. 2. An ensemble of classical states (Kepler-type orbits) is shown. The orbital motion of all the particles is in phase, i.e., their position on the orbital path is the same at a given time. The ensemble average of the motion is a shell oscillating at the Kepler orbital period.

the atom will be ionized. If the second pulse occurs when the wave packet is moving slowly at the outer turning point, the probability of ionization is much less. This pump-probe ionization process has been analyzed by Alber *et al.*[3]

The experiment of ten Wolde *et al.*[9] was carried out in the Rydberg states of atomic rubidium. A superposition of $\sim 2-3$ states (Δn) was formed, with an average value of n equal to $42(\bar{n})$. The resulting wave packet was detected by the method described above. Their photoionization signal exhibited a periodicity that agreed with the classical orbital period (~ 10 ps). They observed the first and second return of the wave packet to the core.

As $\bar{n}$ and Δn change, the characteristics and the behavior of the radial wave packet, also, change. An increase in the value of $\bar{n}$, reduces the rate at which the wave packet evolves and reduces the rate at which it spreads. The slower rate of spreading follows directly from the more nearly equal spacing of the energy levels. An increase in the value of Δn has two effects: (1) The initial localization of the wave packet is increased; (2) the rate of spreading is also increased. The localization of the wave packet determines the sharpness of the quantum beats that are observed in the photoionization signal.

In our experiment we optically excite the two-photon resonance from the ground state of potassium to a Rydberg state with $\bar{n}=89$. Both s and d states are excited by the pulse, but with the d states dominating by a ratio of 4:1. This small mixing of angular momentum states produces little angular localization.[8] Overall, the behavior of the wave packet should be governed by the characteristics of the d states which dominate the mixture. The coherent bandwidth of the excitation overlaps a number of levels ($\Delta n \sim 5$). The orbital period of this wave packet is 107 ps. We anticipate that a well-localized wave packet will be produced by this excitation (the position-momentum uncertainty product should equal $\hbar$, at the outer turning point). Sharp quantum beats should result from this localization. The characteristics and behavior of the wave packet can be predicted by a relatively simple model. The model includes a second pulse, identical to the first, that probes the excited-state wave packet after a variable delay τ.

We can represent the field as

$$E(t)=\varepsilon\{[f(t)e^{i\omega t}+f(t-\tau)e^{i\omega(t-\tau)}]+\text{c.c.}\}\ , \tag{1}$$

where ε is the amplitude of the field and $f(t)$ is a Gaussian pulse shape. The probe pulse couples the excited-state population to both the ground state and the continuum. The process can be described by the following set of differential equations:

$$\frac{da_g}{dt}=i\Delta_g|f(t)+f(t-\tau)e^{i\omega\tau}|^2a_g + i[f(t)+f(t-\tau)e^{-i\omega\tau}]^2\sum_n \Omega_n a_n\ , \tag{2a}$$

$$\frac{da_n}{dt}=-i\delta_n a_n+i\Omega_n[f(t)+f(t-\tau)e^{i\omega\tau}]^2a_g + i\sum_n \Delta_n|f(t)+f(t-\tau)e^{i\omega\tau}|^2a_n + \sum_z i\Omega_{nz}[f(t)+f(t-\tau)e^{-i\omega\tau}]a_z\ , \tag{2b}$$

$$\frac{da_z}{dt}=i\sum_n \Omega_{nz}[f(t)+f(t-\tau)e^{i\omega\tau}]a_n\ , \tag{2c}$$

where a_g is the ground-state amplitude and the many Rydberg levels are represented by the amplitudes a_n. The equation for the intermediate state of the two-photon transition was adiabatically eliminated. The continuum is represented by the amplitudes a_z. The coupling to the continuum states is assumed to be flat, so that there are no Rabi oscillations of the population between the bound states and the continuum. The amplitudes are in a rotating frame and the rotating-wave approximation has been made. The detuning of each Rydberg level is denoted δ_n. The Rabi frequency for the two-photon transition is Ω_n, Δ_g is the ground-state Stark shift, and Δ_n is the excited-state Stark shift.[11] It should be noted that the ground-state Stark shift can be very large and acts as a frequency chirp upon the interaction.

We numerically integrate these equations and compare the solution with our experimental results. The photoionization signal is

$$P_i=\lim_{t\to\infty}\sum_z|a_z|^2\ . \tag{3}$$

The solution for P_i, as a function of the delay time, contains two beat patterns. One is due to the motion of the wave packet, while the other is due to the relative phase between the pump and the probe pulses. This second beat pattern is rapid (varying at the optical frequency). In this experiment, an averaging over the relative phase occurs so that this second beat pattern is not observed.

The experimental setup is shown in Fig. 3. The source of the short, optical pulse is a cavity-dumped, synchronously-pumped dye laser similar to that developed by Wisoff *et al.*[12] The dye laser is sync-pumped by a mode-locked, Q-switched (MLQS) Nd:YAG (yttrium aluminum garnet) laser. A streak camera trace

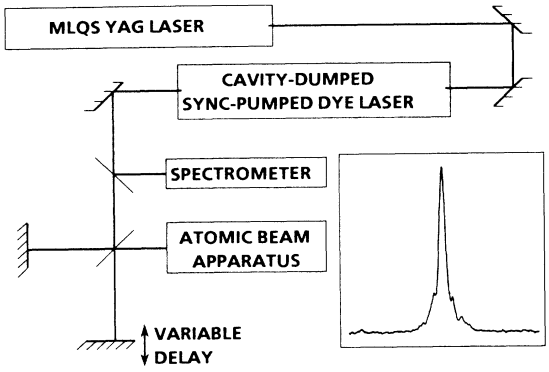

FIG. 3. The experimental apparatus is shown. The short pulse is generated by a synchronously pumped dye laser. A streak camera trace of the dye laser pulse is shown in the inset. The pulse has a width of 25±2 ps.

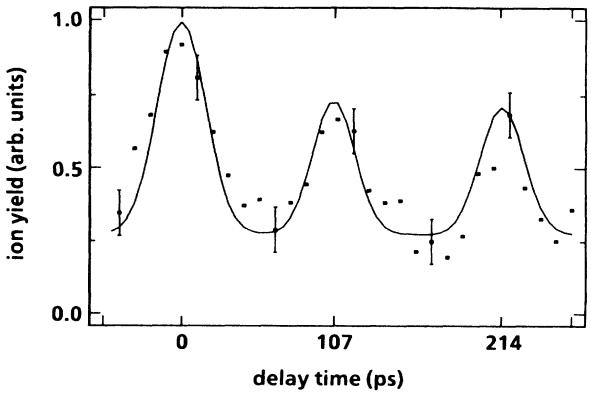

FIG. 4. The experimental and theoretical results are compared (squares and solid curve, respectively). The ionization signal shows a periodic enhancement as a function of the time delay between pulses. The period of this signal agrees with the classical orbital period (107 ps) of the Rydberg state to which the laser was tuned ($n=89$). The width of the peaks indicate that about five states were significantly excited ($87 < n < 91$).

of the dye laser pulse is shown in the inset of Fig. 3. The pulse has a FWHM of 25 ps and has an energy of 5 μJ. The dye laser is tuned to excite a coherent superposition of states with an average n of 89. The initial pump pulse and the delayed probe pulse are derived from the same laser pulse. These pulses are focussed to a 100-μm spot within a beam of potassium atoms. The interaction took place within a grounded box to eliminate stray electric fields. Following the interaction, an electric field is applied to sweep the photoions out of the interaction region. An electron multiplier collects and amplifies the photoionization signal.

The ion signal is measured as a function of the delay of the probe pulse. A computer-controlled translation stage changes the time delay in steps of 10 ps. At each step, the ion signals from 500 pulses were averaged. A 1-m Czerny-Turner spectrometer was used, in second order, to analyze each pulse so that only pulses of nearly the same center frequency ($\Delta\lambda_0 \approx 0.1$ Å) contributed to the ion signal average. This reduces the jitter of the center frequency of the optical pulse to less than the coherent bandwidth of the pulse ($\Delta\lambda \approx 0.5$ Å). An averaging over the frequency fluctuations of the laser ($\Delta\lambda_0 \approx 0.5$ Å) would greatly reduce the depth of modulation expected in the ion signal.

The results of the experiment are shown in Fig. 4. The ionization signal shows sharp peaks at the classical orbital period (107 ps). The error bars are obtained by blocking the pump pulse and recording just the signal produced by the probe laser. Our theoretical model provides an accurate description of the results. However, there is a significant drop in the height of the peak corresponding to the wave packet's second return to the core. At this point, the theoretical curve departs somewhat from the experimental results. The decrease in the height of the peak has two causes. In the case shown, the decrease is due primarily to the residual frequency fluctuations of the laser pulse. These fluctuations cause a slightly different wave packet to be formed from shot-to-shot. The higher-frequency pulses produce an ionization signal with a longer period than the lower-frequency pulses. The averaging of these different signals causes a fairly rapid decrease in the height of succeeding peaks. These fluctuations are not included in the theoretical model and account for the departure of the theory from the experiment. The decrease in height is also due to the dephasing of the coherent superposition state. The wave packet is not as well localized at each successive return to the core. This reduces the enhancement of the photoionization signal. However, this is a small effect which causes the slight reduction of the peak height (see the theoretical curve).

We have observed a radially localized wave packet evolving in a way that agrees with classical theory. The wave packet discussed in this paper was well localized due to the relatively large number of states in the superposition. The motion of this wave packet produced beats in the photoionization signal and the beats were separated in time by the classical orbital period. The sharpness of the beats is the result of the strong localization of the wave packet.

The evolution of such a wave packet over a long period of time should exhibit a region of nonclassical behavior, as the wave packet spreads, then a return to a classical region, upon revival of the wave packet. The contrast between these two regions of behavior will also depend upon the localization of the wave packet. An experiment to observe the dephasing and revival of the wave packet is in progress.

ACKNOWLEDGMENTS

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