Excitation of the Classical-Limit State of an Atom

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(Received 29 December 1993)

We describe a technique designed to excite a classical-limit state of an atom. A picosecond electric field pulse converts a circular state into a Rydberg wave packet which is localized in all three dimensions and travels along a classical Kepler orbit with arbitrary ellipticity.

PACS numbers: 32.80.Rm, 42.50.Hz

The development of picosecond and femtosecond laser technology has provided the tools to control the internal state of atoms and molecules to an extent not previously possible. In laser chemistry a major effort is underway to control vibrational states so as to selectively make or break particular bonds in a molecule [1]. In the case of atoms, it is the state of the electrons that is controlled to produce spatially localized wave packets that may be stationary in space, or move along well-defined orbits. There are several goals that one might hope to achieve by producing and controlling such electronic wave packets: studying the correspondence principle limit of an atom, producing and studying states of an atom that are stable against ionization by intense electromagnetic fields, producing atomic states with large electric dipole moments, producing and studying Schrödinger-cat-like states, studying the connections between the orbits of a classically chaotic system and the motion of quantum wave packets, and studying the orbits of multielectron wave packets in complex atoms.

Some progress has been made in the production and control of electronic wave packets made up of superpositions of Rydberg atomic states. Wave packets that are localized in the radial coordinate, oscillating in and out over an orbit of micron extent in a breathing motion, but not at all localized in the angular coordinates, have been studied in several laboratories [2–4]. Yeazell and Stroud have produced a wave packet that is well localized in the angular coordinates, but not well localized in the radial coordinate [5]. This wave packet is in the form of a pie-shaped wedge sitting nearly stationary on one side of the nucleus; it possesses a very large electric dipole moment. But many of the goals that we listed above require that the wave packet be localized simultaneously in all three coordinates, and this has proved a difficult task using the experimental tools that have been used in this research thus far: picosecond laser pulses along with radio- and microwave-frequency fields. In this Letter we propose a new approach to these problems using a picosecond electric field pulse with no carrier wave, and show how it could be used to produce a three-dimensionally localized electron wave packet moving along a Kepler orbit.

The technique we present here can be used to excite a wave packet that travels along an ellipse of arbitrary eccentricity [6]. However, the basic idea is best described using a circular-orbit wave packet [7,8] as an example. A circular-orbit wave packet is a coherent superposition of circular Rydberg states which belong to several neighboring n-manifolds. In order to excite such a superposition of states an atom must be supplied with enough energy to almost ionize, and with many units of angular momentum. This cannot be achieved in an interaction with a single external electromagnetic field, and has thus far not been achieved with more complex field combinations.

The proposed technique consists of two distinct steps. In the first step a circular eigenstate in a desired n-manifold is excited. In the second step a short, strong electric pulse [9] is applied to populate the circular states in the neighboring n-manifolds.

Any of the existing techniques for the excitation of a circular state [10–13] can be used in the first step. In all of these techniques a low-frequency or static field is applied to dress the atom; a laser is then tuned precisely to a particular dressed state that evolves into a circular state as the dressing field is turned off adiabatically. The crossed-fields method [12] has been especially popular because of its high efficiency [14] and broad applicability in experiments with heavier alkali atoms [15] and two-electron atoms [16]. The probability distribution of a circular state is a ring of radius n^2 and width $(n/2)\sqrt{2n+1}$ in the plane orthogonal to the quantization axis. We refer to this plane as the plane of the orbit.

The excitation of a circular-orbit wave packet from a circular state is easy to understand with the help of a classical analogy. When an ensemble of classical electrons distributed evenly around a circle, all moving with the same angular velocity (Fig. 1), interacts with a short electric pulse, the change in momentum for each particle is proportional to the component of its velocity antiparallel to the field. Immediately after the short pulse the particles are at approximately the same positions as before the interaction, but their energies have changed. The spread in momentum space will be "translated" into a spread in coordinate space over time—faster particles will catch up with the slower ones and at some moment most of the particles will be found in a relatively small segment of the orbit. As we will see, quantum theory confirms the basic correctness of this model.

The exact shape of the electric pulse is not of essential importance but the shorter the pulse, the more sym-

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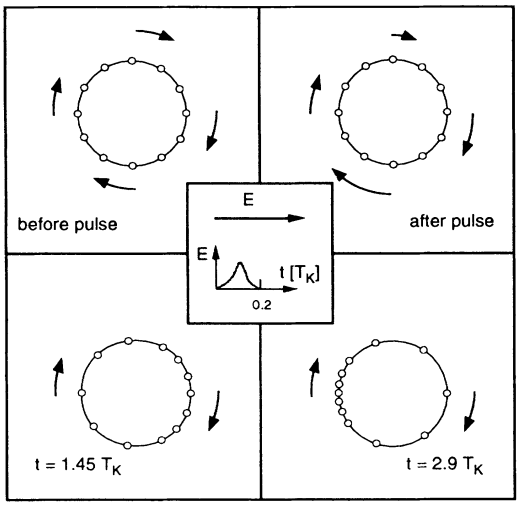

FIG. 1. An ensemble of classical electrons traveling along a circle before and after the interaction with a short pulse. Dispersion in velocities that results from the interaction causes the ensemble to bunch up in a small segment of the orbit after about three orbits.

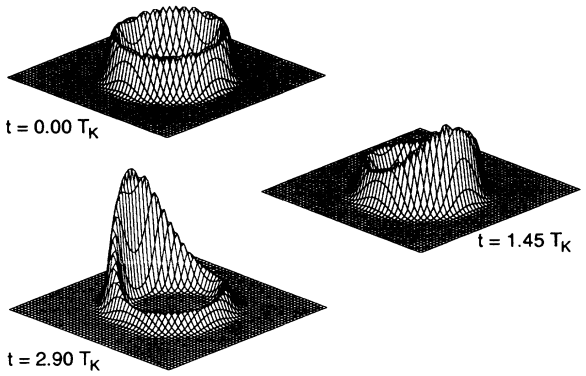

FIG. 2. Evolution from circular eigenstate to wave packet. The short electric field pulse is applied just before time $t = 0$, and the wave packet in coordinate space is not yet noticeably affected. However, in the first three orbits the probability distribution forms up into a well-localized wave packet following a classical Kepler trajectory.

metric the population of the circular states will be with respect to the initial state, because a short pulse does not "see" the differences in the energy separations and in the dipole moments between different circular states. In order to excite a circular-orbit wave packet with principal quantum number $n \sim 50$ a picosecond pulse is required. Such a pulse has no carrier wave; the field itself begins from zero, increases to a maximum value, and decreases back to zero with the field vector constantly pointing in one direction. Pulses as short as several hundred femtoseconds and the bandwidth of the order of 1 THz have been produced using optoelectronic switching devices [17], which utilize photoconductive properties of semiconductors.

The interaction of a circular state with a short electric pulse can be described by a simple quantum theory. The dipole moments that connect two circular states in neighboring manifolds are greater by at least a factor of $\sqrt{n}$ than those connecting a circular state with any other state. Most of the population from the initial circular state will thus reside in circular states at the end of the short pulse. If the pulse is too short to resolve the differences in the energy separations and in the dipole moments for different circular states, the energy-level structure of hydrogen can be reduced to a one-dimensional ladder of equidistant states connected only to the nearest neighbors via dipole moments that are constant throughout the entire ladder.

The interaction of circular states with a short electric pulse is described by

$$\dot{C}_{\bar{n}\pm k} = i\Omega(t)C_{\bar{n}\pm(k-1)} + i\Omega(t)C_{\bar{n}\pm(k+1)}, \qquad (1)$$

where $\bar{n}$ is the principal quantum number of the initial circular state $|\bar{n}, \bar{n}-1, \bar{n}-1\rangle$, $C_{\bar{n}\pm k}(t)$, $k = 0, 1, 2, ...$, are the slowly varying Schrödinger coefficients of the circular states (it is justified to drop the exponents with time-dependent phase because the interaction time is very short), and $\Omega(t)$ is the time-dependent Rabi frequency, equal to the product of the dipole moment and the field strength at time t. Coefficients C_n can be calculated in closed form by solving the linear system of the Laplace-transformed Eq. (1) under the assumption that the ladder of states is infinite in both directions from $\bar{n}$. This assumption is justified for short enough interaction times. The solutions are given in the form of Bessel functions:

$$C_{\bar{n}\pm k}(t) = i^k J_k(\Theta(t)), \qquad (2)$$

where $\Theta(t) = \int_0^t \Omega(t')dt'$ is the area of the pulse.

The population distribution at the end of the pulse in this simple theoretical model is a function of the area of the pulse and deviates only slightly from those calculated numerically with all the possible transitions included. At the end of the electric pulse used to calculate the circular-orbit wave packet in Fig. 2 [the pulse shape was $\sin^2(\pi t/\tau)$, with $\tau = 0.1T_K$ and peak strength 30 V/cm] the population distribution is approximately Gaussian in n, with the standard deviation equal to 0.8. Note that after the interaction with the pulse about 95% of the population resides in circular states while the expectation values of the angular momentum and principal action are changed by less than 0.1%. For stronger pulses the population spreads out into an increasing number of n-manifolds and out of the circular states into other states.

Immediately after the pulse the spatial probability distribution is still that of the initial circular eigenstate. We can see in Fig. 2 that the wave packet localizes over time

in a way similar to the classical ensemble in Fig. 1, and it reaches the point of tightest localization after about $3T_K$. The motion of the localized circular-orbit wave packet over one orbital period is shown in Fig. 3.

By taking advantage of the hidden symmetry of the Kepler-Coulomb Hamiltonian [18,19], the technique described above can be expanded to excite a wave packet that travels along a Kepler ellipse of arbitrary eccentricity. When a hydrogen atom is placed in a weak static field, either electric or crossed electric and magnetic, there exists a good quantum number λ which corresponds to an angular-momentum-like observable $\boldsymbol{\lambda}$; $\boldsymbol{\lambda}$ consists of two components of the Runge-Lenz vector, A_x and A_y, and one component of angular momentum, L_z. During the interaction the expectation values of the two components of $\boldsymbol{\lambda}$ orthogonal to the electric field, L_z and A_y (if the x axis is defined along the field), oscillate with the Stark frequency, $\omega_S = \frac{3}{2}Fn$, where F is the field strength and n is the principal quantum number. For an atom initially in a circular state $|n, n-1, n-1\rangle$ and the field applied parallel to the plane of the orbit, λ has the maximum value of $n-1$; the expectation values of L_z and A_y at time t are equal to $(n-1)\cos\omega_S t$ and $(n-1)\sin\omega_S t$, respectively. The corresponding classical orbit is deformed from a circle into an ellipse whose large semiaxis is orthogonal to the field and whose eccentricity is proportional to the Runge-Lenz vector. The uncertainty product of the two orthogonal components of the Runge-Lenz vector in the plane of the orbit maintains a minimum possible value, $\Delta A_x \Delta A_y = (n-1)|\cos\omega_S t|/2$, during the entire interaction. This result coincides with that of Nauenberg [6] for a coherent elliptic state and assures that the wave packet remains on a Kepler orbit during the interaction, despite the fact that the orbit itself is changing shape.

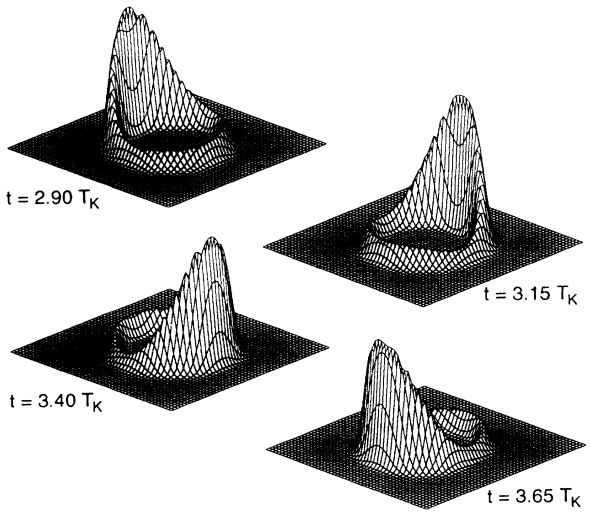

FIG. 3. The motion of the circular-orbit wave packet over one orbital period.

In an actual experimental situation a weak electric field [9] is turned on to deform the orbit from a circle to an ellipse and turned off when the desired eccentricity is achieved. Typically, the peak field strength is less than 1% of the Coulomb field at the relevant Rydberg state, which is equal to $1/n^4$ in atomic units; the pulse is several Kepler periods long. The eccentricity varies as $\sin\Theta$, where Θ is the area of the pulse, so the effect does not critically depend upon the particular shape or length of the pulse, as long as the pulse is weak enough not to mix states from different n-manifolds. The pulse used to prepare the elliptic state which served as initial state for the wave packet in Fig. 4 had a sine squared shape and peak strength of 6 V/cm; the time from the beginning to the peak of the pulse was $9.37T_K$. The eccentricity of the elliptic state is approximately equal to 0.6 and more than 99% of the population resides in aligned states.

At the end of the weak electric pulse a strong, short pulse [9] is applied to populate the neighboring n-manifolds. The pulse used to prepare the elliptic-orbit wave packet in Fig. 4 had the shape $\sin^2(\pi t/\tau)$, with $\tau = 0.1T_K$ and peak strength 40 V/cm. The change in the expectation values of the angular momentum and principal action due to the interaction is less than 1%.

At the end of the short pulse the wave packet has the same shape as the initial elliptic state. About three Kepler periods later (Fig. 4, $t = 2.9T_k$) the wave

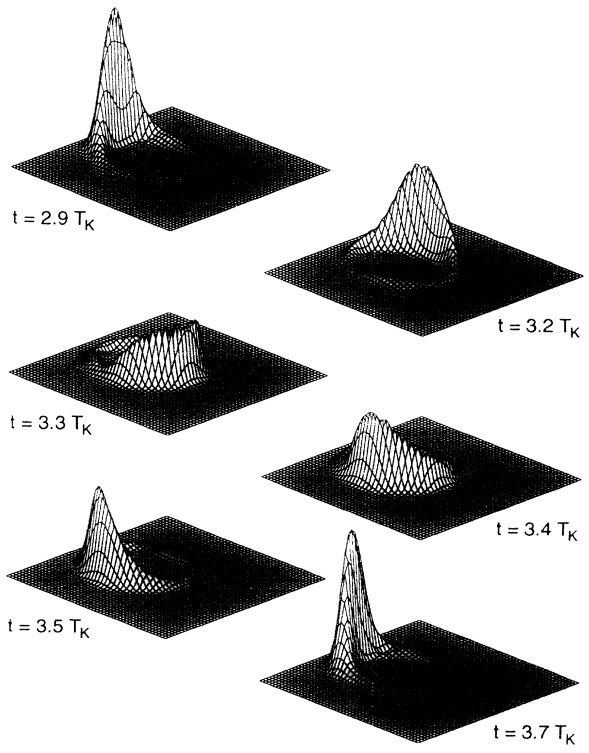

FIG. 4. The motion of the elliptic-orbit wave packet over one orbital period.

packet reaches the point of tightest localization. Note that the wave packet now spreads along the orbit and speeds up every time it passes near the nucleus (Fig. 4, $t = 3.3T_k$, $3.4T_k$, and $3.5T_k$) only to narrow up and slow down when it moves away from it (Fig. 4, $t = 3.7T_k$).

The exact timing of the short pulse is not of critical importance because the elliptic state that is the final state of the weak interaction is stationary. However, since stray fields, which may rotate the orbit and change its eccentricity [20], are bound to be present in the experimental setup, the time lapse between the end of the weak and the beginning of the strong pulse should be short enough for the interaction to be completed before unwanted stray-field effects become noticeable.

Theoretically it is possible to excite a hydrogenic elliptic-orbit wave packet with eccentricity equal to one. In that case the wave packet travels along a straight line. However, few Rydberg-atom experiments are ever done in hydrogen because of the lack of convenient UV lasers. Instead, single-electron-atom experiments are performed in alkali atoms which exhibit excellent resemblance to the hydrogenic energy-level structure for high-angular-momentum states. The presence of core electrons in alkali atoms is responsible for deviations from hydrogenic behavior in low-angular-momentum states. This sets an upper limit on the eccentricity of the elliptic state that can be achieved experimentally; nevertheless, the theory presented here is valid when core-penetrating states are not mixed into the state vector.

We thank J. West, J. Corless, and M. Van Leeuwen for useful discussions. This work was supported by the Army Research Office.

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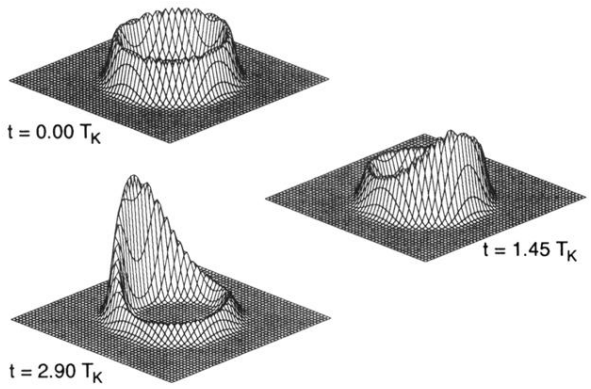

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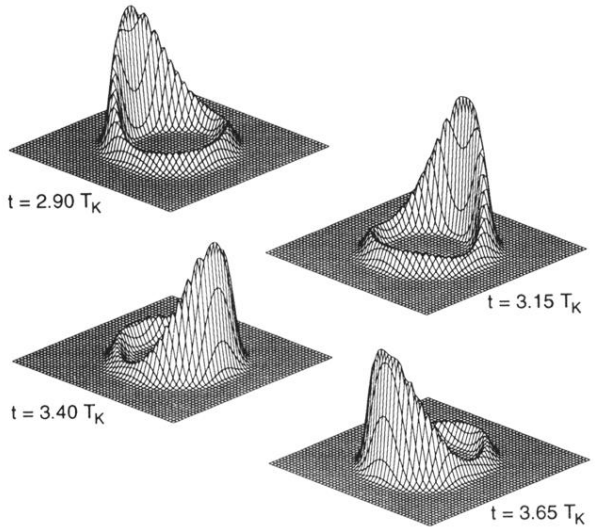

FIG. 3. The motion of the circular-orbit wave packet over one orbital period.

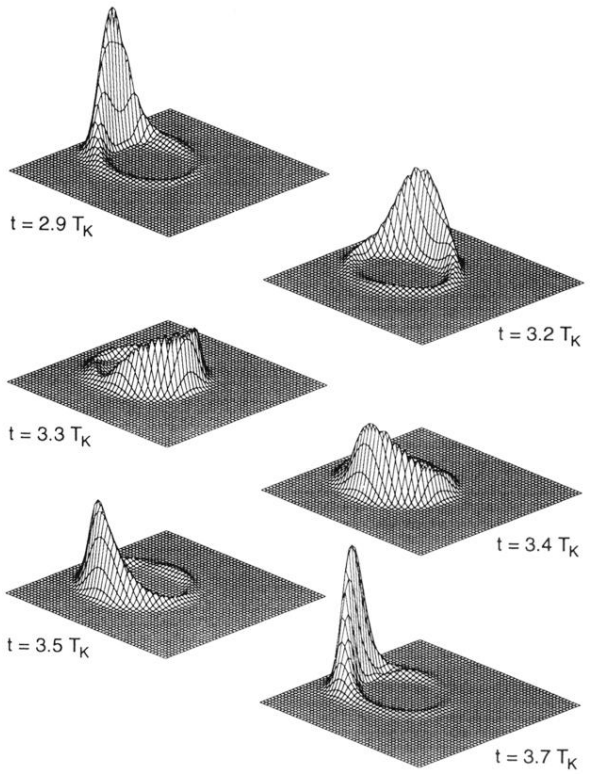

FIG. 4. The motion of the elliptic-orbit wave packet over one orbital period.