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Rydberg-atom wave packets localized in the angular variables

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Calculations are presented that show the behavior of a wave packet composed of alkali-metal-atom Rydberg states. We demonstrate that it is possible to localize such a wave packet in the polar and azimuthal angles. The evolution of this wave packet is equivalent to the precession of a classical Kepler orbit under the influence of a perturbing potential. In this case, the perturbation is the quantum defect of the alkali-metal atoms. An excitation scheme is proposed for producing these states and the results of a numerical model based on this scheme are presented. Finally, the interaction of these angularly localized wave packets with external fields is discussed and a method of detection is suggested.

I. INTRODUCTION

The quantum theory of atoms is based on a Hamiltonian which is just that of the classical Kepler theory of planetary orbits. In spite of this common foundation, the predictions of the quantum theory ordinarily do not much resemble those of the classical theory. Even in the limit of large quantum numbers the energy eigenstates of quantum theory do not look at all like the classical elliptical orbits. Recently, Parker and Stroud[1] have shown that it is possible to excite atomic states which do resemble the classical ideal of a particle traveling in an elliptical orbit. These states are wave packets made up of a linear superposition of different Rydberg states with the same angular-momentum quantum numbers, but with principal quantum numbers n distributed over a range Δn. The wave packets are localized in r, the radial coordinate, but are spread in the polar and azimuthal coordinates. The similarity between the wave packet and the classical particle appears in those effects which depend on the localized coordinate. For example, the quantum-beat fluorescence from the wave packet exhibits a periodicity which agrees with the orbital period of the classical particle. Such studies help to define the regions of validity for classical interpretations of quantum-mechanical systems.

In this paper, we discuss the formation of a wave packet in the polar and azimuthal angles (θ and Φ) by the coherent excitation of Rydberg states with the same principal quantum number, but with a distribution of the angular-momentum quantum numbers. When this wave packet is formed of the Rydberg states of an alkali-metal atom the connection with classical theory is again striking. For high-angular-momentum Rydberg states, the quantum defect of the alkali-metal atoms stems directly from the polarizability of the core. The wave packet precesses due to this core polarization, just as the classical elliptical orbit precesses due to an added r^{-4} perturbing potential.

The study of coherent superpositions of eigenstates is far from new. Schrödinger performed some of the initial studies in this area which resulted in the discovery of the coherent states of the harmonic oscillator.[2] These states are minimum uncertainty wave packets that move in classical trajectories. The large volume of work concerning atomic coherent states[3–6] is the result of a desire to form an electronic wave packet which moves classically. These studies, however, have not treated the excitation of the atomic coherent states. Here, we examine the excitation and behavior of a wave packet, made up of high-angular-momentum Rydberg states, whose motion can be described classically.

In Sec. II we will show that there are wave packets with the required properties, and show some numerical examples. In Sec. II A we will discuss the manner in which such states might be formed using a laser and an rf-dressing field. In Sec. II B we will discuss methods by which these states might be detected and studied. Finally, in Sec. III we will discuss our results and their connection with classical theory.

II. THE ANGULAR WAVE PACKET

The coherent excitation of high-angular-momentum Rydberg states presents some serious difficulties. However, before discussing these problems we will consider which of these states we need to excite in order to form a localized wave packet. Let us consider the wave packet formed from a linear superposition of Rydberg states with the same principal quantum number and with the Zeeman quantum number equal to the angular-momentum quantum number (e.g., $n=50$, $l=35$, $m=l$). Hereafter, we will refer to these Rydberg states as the aligned states, i.e., the angular-momentum vector is aligned along the quantization axis. One of the desirable features of this set of states is that the θ dependence of the aligned states is $\sin^m\theta$ which serves to confine the electron to the x-y plane. And so, for a sufficiently large value of m the localization in θ is accomplished. This allows us to focus our attention on the Φ dependence of the wave packet. A general expression for the wave function as a function of azimuthal angle and time is given by

$$\Psi(r,\theta,\Phi,t)=\sum_m A_m e^{im\Phi} e^{-i\omega_m t}, \tag{1}$$

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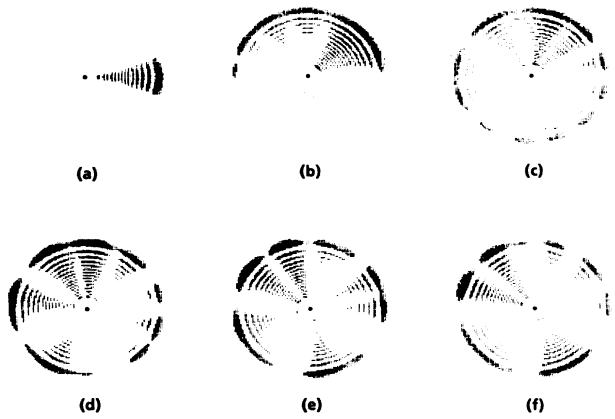

FIG. 1. The evolution of the wave packet (i.e., its position and extent) are described by a series of gray-level plots of the probability in the x-y plane. The circle, at center, indicates the position of the nucleus. The distance from the nucleus to the edge of the plot is 5000 Bohr radii. The wave packet has just been formed in (a). The subsequent plots are spaced by 1.4 ms. The initially well-defined wave packet produced by the coherent superposition, of a flat distribution of 15 eigenstates ($n=50$, $27 \leq m \leq 41$, $l=m$) disperses quickly.

where the A_m's include the amplitudes of the aligned states and the r and θ dependence of their eigenfunctions. The energy of the mth-aligned state above the ground state is $\hbar\omega_m$. The time dependence of the aligned states, ignoring spontaneous decay, is free oscillatory motion as indicated above. So the wave packet evolves at a rate which is on the order of the difference between the maximum and minimum values of the ω_m's.

When the wave packet is initially formed ($t=0$), Eq. (1) is simply a spatial Fourier series. It follows that a flat distribution of the A_m's over a large range of m's will produce a narrow wave packet. In Fig. 1 we show the results of a numerical calculation, under such conditions, for the formation and evolution of the wave packet. The wave packet, at time zero, is quite narrow, but it disintegrates before it can begin to move. If the aligned states were equally spaced in energy this disintegration would not occur. Unfortunately, the difference in energy between neighboring aligned states varies like m^{-6}. Still for a sufficiently small range of m's and a large average value of m, the spacing between aligned states is nearly uniform. Figure 2 shows a wave packet formed from four eigenstates ($n=50$, $33 \leq m \leq 36$, $l=m$). The smaller range of m's produces a broader wave packet, but one that remains intact for a precessional period.

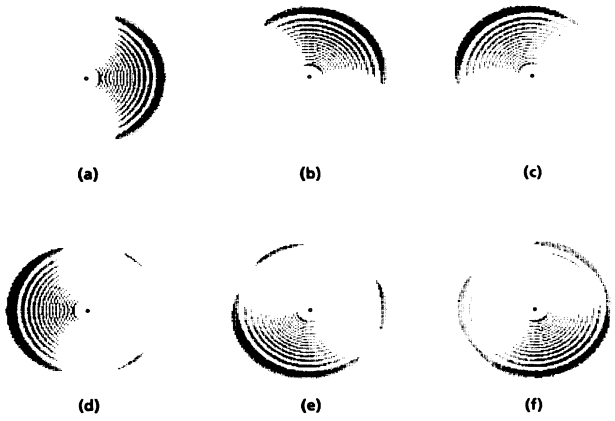

FIG. 2. The wave packet formed by the superposition of a flat distribution of 4 eigenstates ($n=50$, $33 \leq M \leq 36$, $l=m$) is less well defined, but remains intact as the wave packet precesses. The gray-level plots of the probability are again spaced by 1.4 ms.

A. Formation of the wave packet

The formation of the angular wave packet involves the excitation of several high-angular-momentum Rydberg states. The problem of exciting these Rydberg states has been addressed in several papers.[7-9] Here, we will consider the scheme discussed in our previous paper,[7] which will also allow the coherent excitation of a range of these states. It involves the optical excitation of an atom that has been dressed by an rf field. The rf field supplies the additional angular-momentum required to reach the high-angular-momentum states. We will show that by making suitable choices for the characteristics of these fields the excitation of the wave packet described above is possible. That is, it is possible to produce a relatively flat distribution of the amplitudes over the desired range of aligned states. The optical excitation consists of two parts. First, the transition between the nS ground state of the alkali-metal atom and the nP excited state is optically pumped by a circularly polarized field from a cw laser. This procedure[10] produces a significant steady-state population in only one of the magnetic sublevels of the excited state ($l=1$, $m=1$). This allows us to ignore the ground state and treat the p state as the initial state in our calculations. The remainder of the optical excitation is by a short transform-limited pulse whose field is also circularly polarized in the same sense. The shortness of the pulse gives it sufficient bandwidth to overlap several states within the Rydberg manifold (the eigenstates with the same principal quantum number). However, dipole selection rules would allow only the d state of the manifold to be excited. The purpose of the rf field is to lift this restriction. The interaction of the rf field with the Rydberg manifold produces dressed states which are linear combinations of the atomic eigenstates. Now, it is possible for the optical pulse to coherently excite several dressed states since they each contain part of the zero-rf-field d state. Each dressed state is linked adiabatically to an atomic eigenstate. That is, as the rf field is turned off adiabatically, the population in the dressed state goes directly into the atomic eigenstate to which it is linked. And so, a coherent superposition of eigenstates with the same principal quantum number, but with a spread in the orbital angular momentum and Zeeman quantum numbers, is formed.

Before proceeding further, let us introduce a simplification of the basis needed to describe the dressed states. We

have shown in our previous paper[7] that only the aligned states need to be considered. This is due, in part, to the polarizations we have chosen for our optical and rf fields. It is also due to the large dipole moments which link aligned states. We can also drop the s and p Rydberg states from our basis since their energies are quite different from the rest of the manifold. This leaves us with $n-3$ dressed states to consider in our calculations.

The excitation of the dressed states may be described by the following equations:

$$\frac{d}{dt}a_n(t) = -\tfrac{1}{2}i\Omega_n a_i(t) f(t) e^{i\Delta_n t}, \tag{2a}$$

$$\frac{d}{dt}a_i(t) = -\tfrac{1}{2}i\sum_n \Omega_n a_n(t) f(t) e^{-i\Delta_n t}, \tag{2b}$$

where a_i is the amplitude of the initial state and a_n is the amplitude of the nth dressed state. The pulse envelope is denoted $f(t)$ and the Rabi frequencies and detunings for the various transitions are Ω_n and Δ_n, respectively.

Following the adiabatic turn off of the rf field, the amplitude of each dressed state goes directly to the amplitude of the aligned state to which it is linked. So the wave packet is formed of a coherent superposition of aligned states. The conditions for such an adiabatic turn off were discussed in the previous paper.[7]

For appropriate choices of the amplitude and frequency of the rf field and of the center frequency and bandwidth of the optical pulse, the angular wave packet described in the preceding section can be formed. As an example, we have performed a numerical calculation modeling the excitation of such a wave packet in sodium Rydberg states ($n=50$, $33 \leq m \leq 36$, $l=m$). The frequency and amplitude of the rf field (65 MHz and 0.3 V/cm, respectively) were chosen to give a nearly flat distribution of the A_m's over the range of m's excited by the optical pulse (pulse length is 100 ps). In Fig. 3 we show the results of this numerical calculation. The initial orientation of the wave packet lies along the direction of the rf field during the optical pulse. At large r, where the probability is a maximum the distribution is flat. However, this proved to be impossible for the entire range of r so that near the core the probability is isotropic, though the amplitude is small. The necessary quantities (eigenvalues, matrix elements, etc.) were found using an effective potential based on a quantum-defect model of sodium.[11] This potential was used in the time-independent Schrödinger equation which was then solved by a Numerov algorithm.[12,13] Though we have chosen to solve Eq. (2) numerically, it should be noted that analytic solutions can be obtained by such methods as those presented in the papers by Parker and Stroud[14] and Alber *et al.*[15]

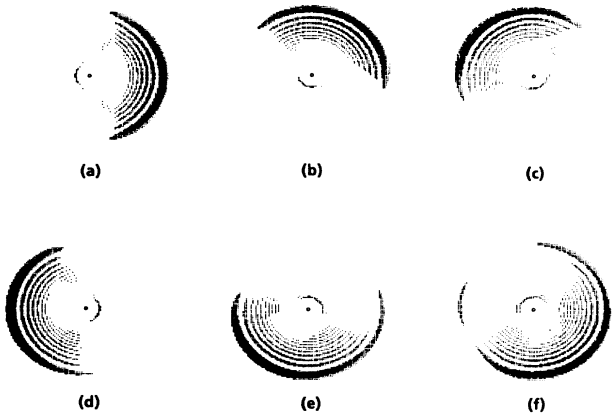

FIG. 3. Evolution of wave packet composed of sodium Rydberg states as described by the numerical model. A coherent superposition of 4 eigenstates ($n=50$, $33 \leq m \leq 36$, $l=m$) produces the wave packet. The gray-level plots occur at 1.4 ms intervals. The wave packet remains intact throughout the precessional orbit.

B. Interaction with external fields

We now examine what effects a localization of the wave packet in Φ has on its interaction with external fields. For transitions which involve large changes of energy the transition rate exhibits little dependence on localization in Φ. The dipole moments for such transitions depend largely on the overlap of the eigenfunctions near the core and the wave packet, described above, is not localized near the core. However, the dipole moments between neighboring Rydberg states ($\Delta n = \pm 1$) depend on the overlap of their wave functions for large values of r where the wave packet is localized. And so, the localization in Φ significantly affects the rates of transitions between adjacent Rydberg levels. The spontaneous emission spectrum is dominated by transitions of the first type and so will give little information about the wave packet. An effective probe of this wave packet must deal with transitions between neighboring states.

One example of such a probe is electric field ionization. Recent studies[11,16–18] of the electric field ionization of alkali-metal atoms have concluded that, for a sufficiently slow slew rate of the electric field, ionization occurs through successive transitions between states of nearly the same energy. This results in a greater ionization rate when the directions of the wave packet and of the electric field are aligned. A particularly attractive field ionization technique was developed by Fabre *et al.*[18] in order to obtain a direct measurement of the size of Rydberg atoms. They pass a beam of Rydberg atoms through micrometer-sized slits in a gold foil and measure the transmission for various Rydberg levels. The transmission of the foil was found to decrease linearly as the size of the orbit increases (i.e., as n^2). If a wave packet localized in Φ were passed through such a "sieve" (the plane of the sieve parallel to the x-y plane) the transmission would vary with the orientation of the slits. Alternatively, the motion of the wave packet may be analyzed by fixing the orientation of the slits and varying the time (or the distance from the interaction region) at which they encounter the sieve.

III. COMPARISON WITH CLASSICAL PARTICLE

The classical motion of the electron may be found by solving the Kepler problem with a perturbing potential.

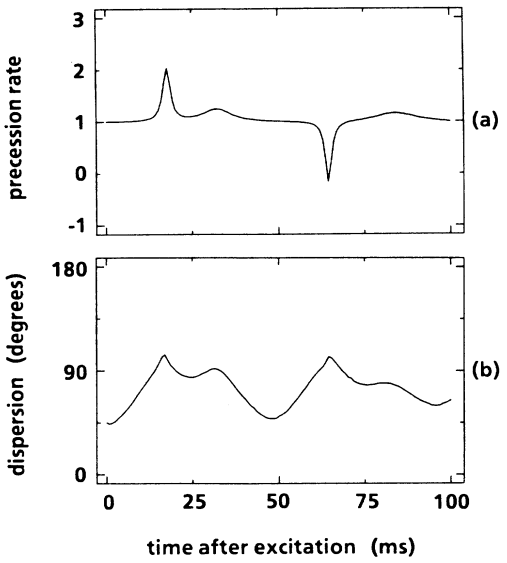

FIG. 4. (a) The precession rate of the average position of the wave packet in units of the classical rate is shown as a function of time. While the wave packet is localized the precession rate agrees with the classical precession rate. As the wave packet spreads (b) the wave packet departs from classical motion. However, when the wave packet reforms, it reaches nearly the initial tightness and again moves as a classical particle. Also, the average rate of motion of the wave packet remains the classical rate.

In the Kepler problem ($1/r$ potential), the Runge-Lenz vector, which points along the major axis of the elliptical orbit, is an integral of the motion. With a perturbation on this potential, the Runge-Lenz vector becomes time dependent and indicates that the orbit precesses. The equation of motion for the Runge-Lenz vector is the following:[19]

$$\frac{d}{dt}\mathbf{A}=\left[-\frac{1}{r}\frac{dV_p}{dr}\right](\mathbf{L}\times\mathbf{r}) . \tag{3}$$

The perturbation (V_p) on the Coulomb potential is due to the quantum defects of the alkali metals. For high-angular-momentum Rydberg states the quantum defect is due primarily to the polarizability of the core. In the simplest model,[20] this produces a perturbing potential that varies like r^{-4}. The solution of this problem for initial conditions corresponding to the wave packet of Sec. II A yields a precession rate which agrees well with the motion of the wave packet. In Fig. 4(a) we show the precession rate of the average position of the wave packet in units of the classical rate. The wave packet spreads until the probability is uniformly dispersed [Fig. 4(b)]. However, the wave packet reforms (the recurrence time is ~ 50 ms) and the dispersion decreases to nearly the original value. While the wave packet is localized it precesses as if it were a classical particle. Thus we have the unusual situation of a quantum state which evolves in a way describable by classical Kepler mechanics.

IV. CONCLUSIONS

Our calculations have shown that it is possible to localize the electron wave function in the angular coordinates. The excitation of such a wave packet formed from sodium Rydberg states was examined. The results of this numerical calculation showed a rather well-localized wave packet which precessed like a classical particle moving in a perturbed Kepler orbit. The interaction of the wave packet with external fields was discussed. This led to a detection scheme which took advantage of the physical size of the Rydberg-atom wave packet. The precession of the wave packet can be studied by measuring the time-dependent transmission of these atoms through a metallic sieve.

It should be noted that if such a wave packet were formed from hydrogenic Rydberg states the wave packet would not precess. This offers a different perspective on quantum-defect theory. The alkali-metal states can be thought of as hydrogenic states which are precessing. That is, the effects of the Coulomb potential and of the quantum-defect potential have been separated. The motion of the wave packet is due solely to the quantum-defect potential.

ACKNOWLEDGMENT

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