PHYSICAL REVIEW A VOLUME 45, NUMBER 7 1 APRIL 1992

Ramsey fringes in atomic Rydberg wave packets

L. D. Noordam, D. I. Duncan, and T. F. Gallagher
Department of Physics, University of Virginia, Charlottesville, Virginia 22901
(Received 24 October 1991)

We examine the dynamics of the wave function of a hydrogen Rydberg electron for the case in which the Rydberg states are created by two temporally separated laser pulses. We study the influence of the phase difference between the two pulses. For instance, the electron can make one orbit as a Rydberg wave packet created by the first laser pulse, after which it is sent back to the ground state by the second pulse. The time dependence of the wave packet created by the first pulse can be probed by examination of the total population after the two-pulse sequence. The influence of the relative phase of the two pulses on the total population can be seen as Ramsey fringes. We describe a scheme of wave-packet detection based on this effect.

PACS number(s): 32.80.Rm

I. INTRODUCTION

One of the challenging aspects of Rydberg atoms is their classical behavior. The size of the wave function is almost macroscopic (160 nm radius for $n=40$), and their characteristic oscillation, or orbit time, is no longer infinitely small (10 ps for $n=40$). Recently it has been predicted [1,2] and observed [3,4] that a Rydberg electron can be prepared such that it is radially localized and, almost like a classical particle, bounces back and forth between the core region and the outer turning point of the Coulomb potential. These radial wave packets are created by a short laser pulse, with a bandwidth large enough to overlap several adjacent Rydberg levels and hence create a coherent superposition of the levels. Since the initial ground state is confined to a small region close to the core, the excitation of the wave packet by the short pulse also takes place close to the core. Once launched in the core region, the wave packet created starts to oscillate radially in the classically allowed region of the potential. The oscillation time of this wave packet is given by $T=2\pi n^3$, which is simply the inverse of the spacing between the adjacent Rydberg levels. In order to create a wave packet, more than one state has to be excited. However, the orbiting time is not the same for different states; as a result the localized wave packet will spread in the course of time (dispersion). After a certain time interval the different orbiting times will again coincide and the wave packet is relocalized (revival).

We describe how the time dependence of the wave function can be probed in a two-pulse experiment. The first pulse creates a wave packet, while the second delayed pulse can either excite another wave packet or deexcite the first wave packet. As in a Ramsey-fringe experiment, the phase difference between the pulses is crucial. The total population of the Rydberg state after the pulse sequence is modulated by the phase difference. The amplitude of the modulation is a measure of the amount of Rydberg wave function created by the first pulse that is close to the core when the atom is exposed to the second pulse. For instance, for a time delay that equals the oscillation time of the Rydberg electron, the first pulse can create a wave packet that is entirely pumped back to the ground state by the second pulse. This special case has already been discussed by Alber, Ritsch, and Zoller [1]. They showed that the third pulse can be applied to study the influence of the second pulse on the first pulse. Here we discuss the influence of the phase in more detail and propose a simple technique to study the influence of the phase difference between the first and second pulse. We will give a detailed interpretation of the results of the calculations.

Besides being of fundamental interest, studying wave packets in this manner has experimental applications. All the observations of wave packets to date have a limited signal-to-noise ratio. For the same reason, while a large number of time-dependent Rydberg wave functions have been predicted, the corresponding experiments have not yet been carried out. For instance, it has been calculated that a Rydberg wave packet created in a magnetic field will split into several orbits, each with its own oscillation time [5]. Recently we have calculated that wave packets created by an intense laser pulse can be compressed to a width less than the duration of the exciting laser pulse [6]. We describe a way to detect the wave packets more efficiently than the presently used technique. This method may allow us to observe the abovementioned and other fascinating phenomena.

II. THEORY

We study the situation sketched in Fig. 1. The initially populated ground state is coupled to Rydberg p states by two laser pulses. We investigate the total population of the Rydberg states as a function of the delay τ_d between the two pulses. The first pulse pumps some of the amplitude into the Rydberg states, while the second pulse either enhances the Rydberg population or pumps the previously created amplitude back to the ground state. We assume the pulses to be Gaussian, i.e., the electric field of the laser beam where it interacts with the atoms is given by

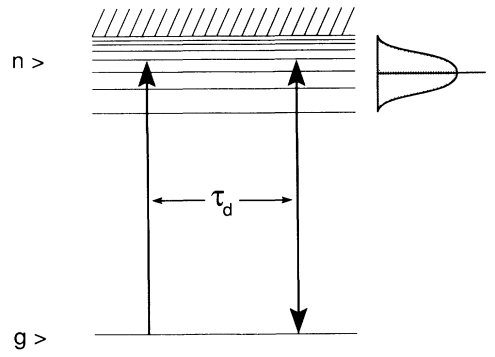

FIG. 1. Schematic representation of the two-pulse coupling of the ground state to the Rydberg series. Initially all the wave function is in the ground state. The first pulse creates a radial wave packet, a coherent superposition of Rydberg states, while the second pulse can enhance the wave-packet amplitude or send all the wave function back to the ground state.

$$E(t)=E_0\sin(\omega_L t)e^{-2\ln 2(t/\tau_p)^2} + E_0\sin[\omega_L(t-\tau_d)]e^{-2\ln 2[(t-\tau_d)/\tau_p]^2}, \quad (1)$$

in which ω_L is the angular laser frequency. As noted above, the associated bandwidth of each of the pulses is large enough to coherently excite several Rydberg states. For simplicity the intensity of the pulses is chosen to be low. In this way the following effects can be neglected: depletion of the ground state [7], ionization, and ac Stark shift of the Rydberg states with respect to the ground state [6].

Starting from the Schrödinger equation for a hydrogen atom and applying the rotating-wave approximation, we find for the amplitude of the Rydberg state n

$$a_n(t)=-ie^{-i\omega_n t}\mu n^{-3/2}\int_{-\infty}^{t}dt'E(t')e^{-i(\omega_1-\omega_n)t'}, \quad (2)$$

in which $h\omega_1$ and $h\omega_n$ are the energies of the ground state and of the state n, and μ is the matrix element $\langle 1s|z|np\rangle$ where np are the Rydberg wave functions per unit energy. For hydrogen $\mu=1.25ea_0$. The radial and temporal parts of the wave function are described by

$$\Psi(r,t)=\sum_n a_n(t)R_n(r), \quad (3)$$

in which $R_n(r)$ is the radial part of the Rydberg wave function [8]. Since we start from the ground state $1s$, the Rydberg state will be a p state, and we ignore the angular part of the Rydberg state wave function which is the same for all n. From Eq. (2), the amplitude of the states a_n can be calculated after the pulses. Assuming that both pulses are identical but delayed with respect to each other by a time τ_d the total amplitude after the pulse sequence is found by integrating Eq. (2):

$$a_n(t)=-ie^{-i\omega_n t}\mu n^{-3/2}\left[\frac{\pi}{2\ln 2}\right]^{1/2} \times\tau_p E_0 e^{[-(\Delta_n\tau_p)^2/8\ln 2]}(1+e^{-i\Delta_n\tau_d}e^{i\omega_L\tau_d}), \quad (4)$$

where $\Delta_n=\omega_L+\omega_1-\omega_n$.

III. RAMSEY FRINGES

Let us focus on the last term in parentheses in Eq. (4). The square of this term is proportional to the Rydberg population, and oscillates between 0 and 4. There is a rapidly oscillating term ($\omega_L\tau_d$) related to the central frequency of the pulse and a slowly varying term ($\Delta_n\tau_d$) related to the oscillation time of the wave packet. This is graphically shown in Fig. 2, a plot of the dependence on the delay time of the total population of all Rydberg states, $\sum_n|a_n(t)|^2$. For all the calculations presented in this paper the central laser frequency corresponds to excitation of $n=40$ ($T=10$ ps), and the excitation is realized by two sequential 2.0-ps pulses. For clarity, in Fig. 2 the laser frequency is scaled down by several orders of magnitude (in fact there are about 70 000 rapid oscillations within the slow envelope oscillation). The envelope of Fig. 2 shows the radial oscillations of the wave packet between the core region ($\tau_d=0T,1T,\ldots$) and the outer turning point ($\tau_d=0.5T,1.5T,\ldots$). Since the spacing between adjacent Rydberg states is not constant the wave packet broadens in course of time, seen by the broadening and shrinking height of the peaks of the envelope.

We now discuss the fast oscillations in Fig. 2. For $\tau_d=0$ the pulses overlap and the production of Rydberg states is at a maximum. For a delay corresponding to half an oscillation of the optical field (very close to $\tau_d=0$) the two pulses are out of phase and there is almost no production of Rydberg states: the two optical pulses cancel and the atom is not exposed to any light. This can be seen directly if we prepare the pulse pair in a Michelson interferometer. If the two merged beams are out of phase in one arm, all the energy will be in the other arm of the interferometer. For $\tau_d=0.5T$ the effect of the phase has almost disappeared. The first pulse creates a wave packet which is close to the outer turning point by the time the

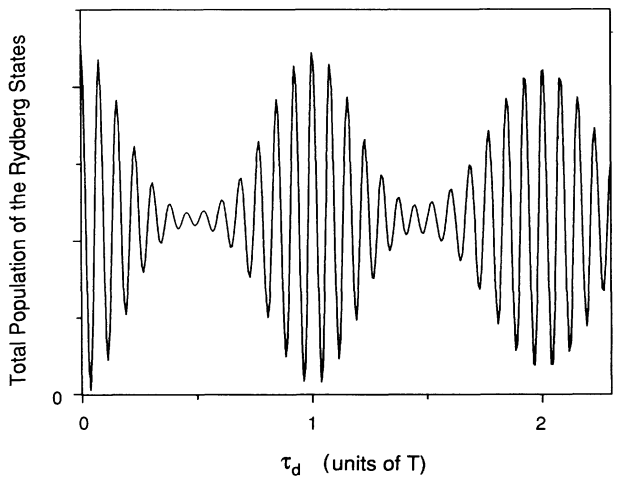

FIG. 2. Total population of the Rydberg states (around $n=40$) after the two pulses as a function of the delay between the pulses. The time is given in units of the round-trip time of the wave packet. The envelope shows the orbiting and broadening of the radial wave packet, while the fast oscillations indicates the influence of the phase difference between the two pulses.

second laser pulse interacts with the atom. Since excitation to the Rydberg states takes place close to the core, the two pulses can in this case be viewed as acting independently on the atom. The total population is just the incoherent sum of the contributions of each pulse and the phase difference between the pulses is not relevant. At $\tau_d = 1T$, the wave packet launched by the first pulse has returned to the core when the second laser pulse interacts with the atom. Apart from some dispersion the situation for $\tau_d = 0$ is restored.

The fast oscillations in Fig. 2 can be seen as Ramsey fringes: the first pulse excites the Rydberg states, while the phase of the second delayed pulse determines whether the Rydberg amplitudes are enhanced or diminished. Another perspective on the same phenomena can be obtained by Fourier transforming the two pulses. The Fourier transform of one Gaussian pulse gives a Gaussian frequency spectrum which overlaps several adjacent Rydberg states. The Fourier transform of two pulses, separated by the wave-packet oscillation time, gives the same spectrum with a modulation superimposed on it. For a separation time τ_d of T, the spacing of the peaks in the frequency domain corresponds to the spacing between the Rydberg states. The peaks of the modulation shift as a function of the phase difference between the two pulses. Whenever the peaks overlap in frequency with the eigenfrequency of the Rydberg states, the states will be populated by the pulse pair. On the other hand, if the peaks are in between the Rydberg levels there is no net population of the Rydberg states.

IV. WAVE PACKET FOR ONE ROUND TRIP

We focus on the minima close to $\tau_d = 1T$. While there is no population of the Rydberg states after the pulse sequence, there is population during the sequence. This point can most easily be seen by following the amplitude during the pulses. We calculated the average distance from the core of the wave function as a function of time using Eqs. (2) and (3). The results clearly show (Fig. 3) that part of the wave function is excited by the first pulse and forms a wave packet. This wave packet makes one oscillation after which it is deexcited to the ground state by the second pulse: a radial wave packet is created that makes exactly one round trip in the Rydberg orbit. The amplitude of the radial oscillation, as shown in Fig. 2, is proportional to the population of the Rydberg states, and thus to the coupling between the ground state and the Rydberg states: $(n^{-3/2}\tau_p E_0)^2$. As can be seen from Fig. 3, after the two pulses some amplitude remains in the Rydberg states. Due to dispersion, the wave packet launched by the first pulse is broadened and not all the wave function can be pumped back while the second pulse is on. Note that for $\tau_d = 2T$ and the two laser pulses out of phase a similar behavior occurs. The first pulse creates a wave packet that makes two oscillations before it is sent back to the core by the second pulse.

V. APPLICATION

Finally, we discuss a general way to improve experimental observations of Rydberg wave packets. In all experiments performed to date, one pulse has been used to excite the wave packet from the ground state, and a second delayed pulse to ionize the wave packet. Since photoionization takes place close to the core, the ionization probability is proportional to the amount of wave function close to the core. The oscillations of the wave packets are observed by monitoring the ionization as a function of the delay. Every time the wave packet returns to the core, the ionization is enhanced. It is well known that the photoionization probability of Rydberg states is fairly low. Therefore, once the wave packet is created, there is only a small probability that it can be detected by the second pulse. It is our belief that this small detection efficiency causes the, at best, moderate signal to noise ratio in the experiments. In the pump-probe technique presented in this paper it is sufficient to measure the total population of the Rydberg wave packets after the pulse. This can be realized by field ionization, a technique with almost 100% detection efficiency. The amplitude of the oscillations of the population of the Rydberg states, as shown in Fig. 2, is a proper measure for the amount of wave function close to the core at the time the second laser pulse is applied.

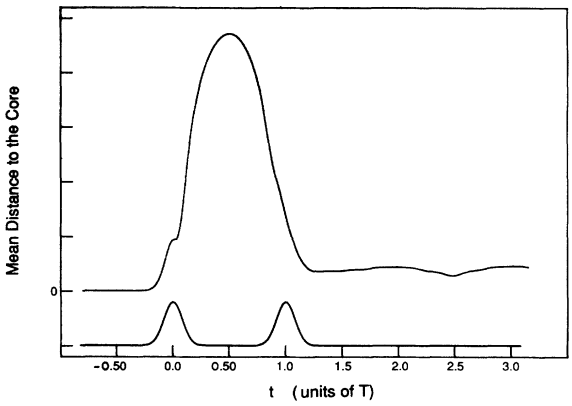

FIG. 3. Mean distance to the core of the wave function as a function of time, for the case in which the two pulses are out of phase and separated by the round-trip time of the wave packet. The wave packet created by the first pulse makes one round trip in the Rydberg orbit before it is sent back to the ground state by the second pulse. The lower trace gives the intensity profile of the optical pulse sequence.

VI. CONCLUSIONS

We have shown that an atomic electron can be prepared in a Rydberg orbit for one radial oscillation, after which almost all the wave function returns to the ground state. By means of field ionization this effect, and more generally, time-dependent Rydberg wave functions, can be measured efficiently. Compared to the scheme used in experiments to date, the proposed scheme has the same excitation probability, but a much higher detection efficiency.

ACKNOWLEDGMENTS

The authors would like to acknowledge careful reading of the manuscript by H. Stapelfeldt and M. J. Renn, and the support of the National Science Foundation under Grant No. PHY-9001126.

[1] G. Alber, H. Ritsch, and P. Zoller, Phys. Rev. A **34**, 1058 (1986).
[2] J. A. Yeazell and C. R. Stroud, Jr., Phys. Rev. Lett. **56**, 716 (1986).
[3] A. ten Wolde, L. D. Noordam, A. Lagendijk, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).
[4] J. A. Yeazell, M. Mallalieu, and C. R. Stroud, Jr., Phys. Rev. Lett. **64**, 2007 (1990).
[5] G. Alber, Z. Phys. D **14**, 307 (1989).
[6] L. D. Noordam, H. G. Muller, A. ten Wolde, and H. B. van Linden van den Heuvell, J. Phys. B **23**, L115 (1990).
[7] G. Alber and P. Zoller, Phys. Rev. A **37**, 377 (1988).
[8] M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A **20**, 2251 (1979).

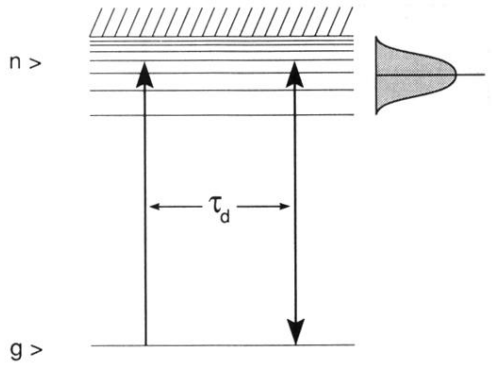

FIG. 1. Schematic representation of the two-pulse coupling of the ground state to the Rydberg series. Initially all the wave function is in the ground state. The first pulse creates a radial wave packet, a coherent superposition of Rydberg states, while the second pulse can enhance the wave-packet amplitude or send all the wave function back to the ground state.