Observation of a Nondecaying Wave Packet in a Two-Electron Atom

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(Received 6 July 1998)

We excite and manipulate wave packets in the Rydberg states of atomic calcium. The manipulation is via the electron-electron correlations between a core electron and the Rydberg electron and mediated by a strong excitation of the core transition. Striking changes in the wave packet are observed particularly when the Rabi oscillations of the core transition are synchronized with the natural, classical oscillation frequency of the Rydberg wave packet. In that case, the wave packet is shaped into a form that inhibits decay or autoionization as demonstrated by a coherent probe of the resulting wave packet. This work impacts on both coherent control and quantum measurement studies. [S0031-9007(98)08062-4]

PACS numbers: 32.80.Qk, 32.80.Dz, 32.80.Rm

We have observed the formation of a nondecaying wave packet. A Rydberg electronic wave packet was excited in atomic calcium and further manipulated via a strong core excitation into this nondecaying form.

The creation, control, and measurement of wave packets are central topics in a broad range of research areas. Specially designed wave packets have been proposed (and to a lesser extent implemented) to control key processes in atomic [1], molecular [2], and solid state systems [3]. Such wave packets also play a significant role in understanding measurement aspects of quantum mechanics [4,5].

Wave packets are nonstationary states that are phased superpositions of highly excited atomic eigenstates. Among the simplest of these to excite is the radially localized wave packet. These are routinely created by short pulse excitation in a single-electron atom that produces a superposition of states with different principal quantum numbers (n). A typical distribution is Gaussian with a FWHM of seven states centered on an average n of $\bar{n} = 50$. On a short time scale, these wave packets demonstrate nearly classical behavior [6]. That is, the radial wave packet oscillates at the classical period, $\tau_{\mathrm{cl}} = 2\pi\bar{n}^3$, between the inner and outer turning point of the classical electron's orbit. At longer times, quantum mechanics plays a distinct role and the wave packet disperses and displays a regular interference pattern that contains both fractional [7] and full revivals [8] of the wave packet. Recently, radially localized wave packets in two-electron systems have been excited that demonstrate nearly identical temporal behavior [9].

In this paper, we control the behavior of a radially localized wave packet in a two-electron system by driving the remaining, correlated valence electron. One of the valence electrons is excited by a short pulse to a radially localized wave packet state. The remaining core electron is driven so that it oscillates between its ground state and its first excited state. The states of this core electron are much like the states of the singly ionized atom or like that of a single electron atom so that this type of excitation is described as an isolated core excitation (ICE) [10]. When the core electron is in the ground state the two-electron system is bound. In contrast, if the core electron is excited the system may autoionize. Thus, the manipulation of the wave packet involves the entanglement of the two valence electrons and the related autoionization process. This entanglement can shape the wave packet into a nondecaying form. A related aspect of this work is the control of the decoherence associated with the decay of an electron to a continuum via autoionization.

Although the alkaline earth atoms are well understood spectroscopically [11], the above control ideas are quite recent and are experiencing a tremendous growth. ICE has been used to excite and modify Rydberg wave packets (e.g., enhancement of photoionization) [10,12,13]. Recent experiments have explored the variation in the autoionization cross section due to the detuning of the ICE [14]. Dynamics of wave packets in this two-electron system have also been studied [15]. Some effects of oscillating bound configurations on wave packet behavior have been observed [16]. Quantum measurement studies have also been performed using a perturbative ICE [17].

The experiments studied here are related to the theoretical papers that have explored strong-field ICE processes [18–22]. One of the more dramatic predictions of these works is the shaping of nondispersing or nondecaying wave packets [18,22]. The shaping effects are striking when the Rabi frequency, Ω, for the ICE is equivalent to and synchronized to the classical orbital frequency, ω_{cl}, of the radial wave packet. Consider the situation in which the goal is to suppress decay (autoionization) [18]. The objective is to time the wave packet's motion so that it is localized away from the core when the core is in the excited state. The resulting lack of overlap between the wave packet and the excited core limits the autoionization. As the evolution of the wave packet progresses by half a period, it will approach the core. However, if the Rabi and classical oscillations are synchronized the core electron will have completed its cycle back to the ground state and again there is little autoionization. This physical picture gives a clear description of the suppression of decay. However, a more complete model is needed to discuss the shaping of the wave packet. It is the channel

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interaction driving the autoionization process that provides the shaping. The channel interaction provides a coherent coupling, via the continuum, between the states making up the wave packet. This allows the amplitudes and the relative phases of these states to be modified. In the situation described above, this modification results in the formation of a nondecaying state. The autoionizing Rydberg states are resistant to decay due to the precise phasing of the states that allows destructive quantum interference between the decay routes to the continuum. However, the dispersion of the wave packet would disrupt this precise phasing, if the synchronization were to be removed. The synchronizing field compensates the effects of dispersion and preserves the nondecaying form of the wave packet. The physical origin of this compensation is that the wings of the wave packet, that have grown due to dispersion, experience a strong interaction with the continuum [18,22]. This interaction tends to rearrange the amplitudes and phases of the states into a nondecaying form. Of course, a portion of the population is also lost to the continuum. Note that these nondecaying, multilevel superposition states are analogous to those known as "dark" states [23] that have been studied in three-level atomic systems.

The experiment, described here, explores these nondecaying wave packets produced by a strong, synchronized ICE. For comparison, wave packets with no ICE are excited. Also for comparison, nonsynchronized wave packets for which Ω is not equal to $\omega_{\rm cl}$ are excited.

Figure 1(a) indicates the laser interactions with the Ca atom. A cw laser is tuned to the resonance of the isolated core transition. Note that until the pulsed laser excites the Rydberg wave packet the cw field is far from any resonance in the Ca atom. The two-photon process driven by the pulsed laser ($\sim$406 nm) excites a wave packet with both *nd* and *ns* character. The *nd* dominates the process described in this paper and therefore only a single series is displayed. The simplified energy level diagram of Fig. 1(a) shows that the Rydberg wave packet sees a core that oscillates at Ω between a ground state (bound series) and an excited state configuration (autoionizing series).

Figure 1(b) displays a schematic diagram of the experimental apparatus. The short pulse is derived from the output of a Ti:sapphire laser (a nearly transform-limited pulse of 5 ps duration and an energy of 10 nJ). This pulse is frequency doubled in a BBO crystal and split into a pump and a variably delayed, probe pulse by the Michelson interferometer setup shown in Fig. 1(b). The interaction of this pump pulse and the delayed probe pulse can greatly modify the population in the Rydberg states [24]. If the wave packet is near the nucleus when the probe pulse occurs constructive (coherent increase in the population) or destructive interference (zero population) can occur in the total Rydberg state population. This variation or fringes in the population depends upon the optical phase delay between the two pulses. If the wave packet is not near the nucleus when the probe occurs, the interference does not occur and leads to low visibility fringes. Therefore, the visibility of these fringes (commonly known as Ramsey fringes) as a function of time reveal the evolution of the wave packet as it approaches and recedes from the nucleus.

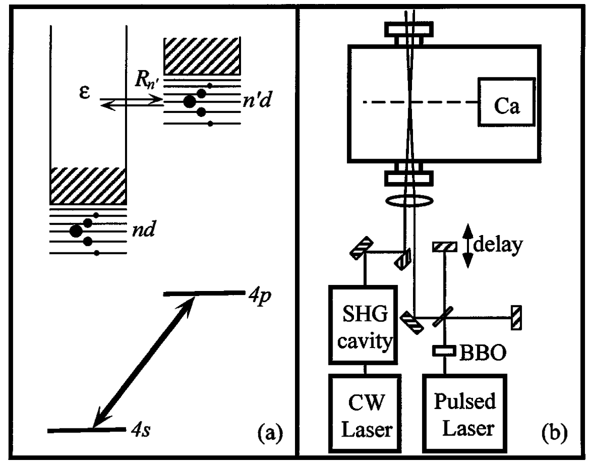

FIG. 1. (a) Energy level diagram describing the modification of the wave packet by the core laser interaction. The Rabi oscillations of the core transition cause the wave packet to oscillate between a bound and autoionizing series. The superposition of states that comprises the wave packet is depicted by the weighted dots. At the moment shown, the core transition has gone through $\pi/2$ of a Rabi oscillation. The wave packet has equal weight in the two series. (b) Optical and atomic beam layout.

The core laser is derived from a Coherent 899 Ti:sapphire laser producing 2W at 794 nm. This output is frequency doubled in a stabilized traveling wave cavity. The design is similar to that described in [25] and produces 200 mW of light at 397 nm. Both laser beams are collimated and are made to travel parallel paths ($<$10 μrad). They are focused by the same lens ($f = 150$ mm) so that their focal spots overlap on the path of the Ca atomic beam. The focal spot of the core laser (15 μm) is slightly larger than that of the pulsed laser (10 μm). This size difference, in conjunction with the fact that the excitation of the wave packet is the result of a two-photon process, insures that the majority of excited wave packets see the same core laser intensity. After the probe pulse, an electrical pulse (width of 5 ns and a peak field of 50 V/cm) synchronized to the laser pulse is applied to the interaction region. This field ionizes a portion of the bound Rydberg states. Thus, the ionization signal is a combination of the autoionized and field-ionized populations. Note that this type of pulsed-field ionization has an $\bar{n}$-dependent efficiency that increases by a factor of 2 from $\bar{n} = 50$ to $\bar{n} = 80$.

Prior to the main experiment, the core laser resonance is found by spectroscopic means and the core laser is locked to a stabilized Fabry-Perot interferometer that holds the frequency of the laser to the peak of the core transition for the duration of the main experiment.

The time evolution of the resulting two-electron wave packets (without the ICE) was observed by the Ramsey fringe technique, described above, and was similar in character to those reported in [9]. However, for the purposes of this paper a variation of that Ramsey method is more efficient and appropriate. Here, the Ramsey fringes are recorded as a function of the detuning of the pulsed laser. This method of recording the Ramsey fringe data is chosen to emphasize the effect of the synchronization of the Rabi and classical oscillations. As the central frequency of the laser pulse is increased, the wave packet created has a larger $\bar{n}$ and a smaller classical frequency ($\omega_{\mathrm{cl}} = 1/\bar{n}^3$). Therefore, as the detuning or $\bar{n}$ is increased, a smooth transition from nonsynchronized wave packets to a synchronized wave packet and back is observed. The probe pulse occurs at a long fixed time delay (long in comparison with the autoionization lifetime of a single Rydberg state) that allows the different effects of synchronized and nonsynchronized ICE to develop. These effects are observed in the Ramsey fringes produced in the Rydberg atom population by introducing phase delays ranging from zero to 2π (spatial delays of zero to 203 nm) into the probe pulse. The Michelson interferometer is locked to a stabilized helium-neon laser and by locking at various levels of these helium-neon fringes the necessary delays are created. The resulting modulation in the Rydberg population is extracted by an FFT algorithm yielding the amplitude of the Ramsey fringes. Thus, as the detuning is scanned the fringe visibility displays both the general nonsynchronized behavior and the more singular synchronized behavior of the wave packets. For the laser and atomic parameters of this experiment the synchronization should occur in the vicinity of $\bar{n} = 67$.

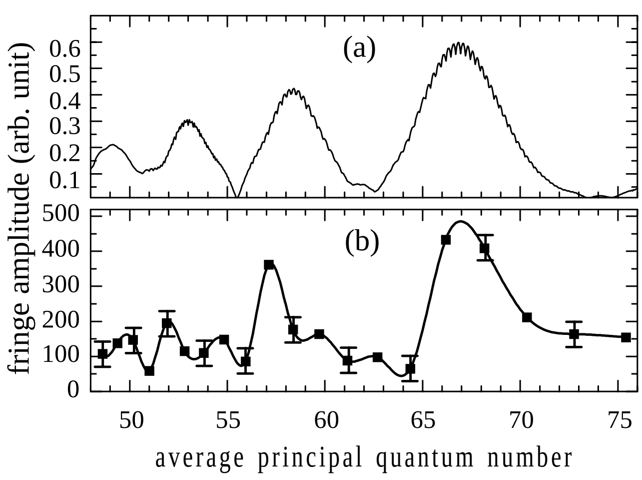

FIG. 2. Visibility of Ramsey fringes versus the average principal quantum number of the wave packet. In (a) a theoretical model is presented for comparison. The experimental results are shown in (b). Each peak corresponds to a wave packet that has gone through an integral number of orbits to reach the fixed delay time. The errors are estimates of systematic deviations observed during data collection.

Two sets of data are taken during the main experiment. The visibility is measured with and without the strong ICE at each detuning step. A theoretical model of the ICE-off case is shown in Fig. 2(a). This and subsequent theoretical descriptions were developed in detail in [22]. Briefly, it assumes that only a single autoionizing series exists and that only a single continuum channel plays a significant role. Despite these simplifications, it produces excellent qualitative agreement with the experimental results [Fig. 2(b)]. Note that the peaks correspond to wave packets that have completed an integral number of orbits in the fixed delay time. A wave packet with a short τ_{cl} will go through many orbits before reaching that delay time. The τ_{cl} grows as $\bar{n}^3$ so that as $\bar{n}$ increases the wave packets reach the delay time by going through a progressively smaller number of orbits. This nonlinear relationship between τ_{cl} and $\bar{n}$ also leads to the progressively larger spacing between the peaks.

When the strong ICE is turned on we observe the signal shown in Fig. 3(b). The theoretical model for this case is given in Fig. 3(a), for comparison. Again, the qualitative agreement is excellent. In general, the Rabi oscillations driven by the core laser are not synchronized with the wide range of τ_{cl}'s that appear in this figure. Without synchronization, the decay due to autoionization destroys much of the coherence and therefore the visibility of the fringes by the fixed delay time of 86 ps. That is, this delay time is long compared to autoionization decay time of a single Rydberg state, which for this system is approximately one classical period [11]. The advantage of this measurement scheme is that it allows direct comparison of the decay of a synchronized wave packet with that of nonsynchronized wave packets.

In Fig. 2(b), three high visibility peaks of comparable height were observed. In contrast, only a single high

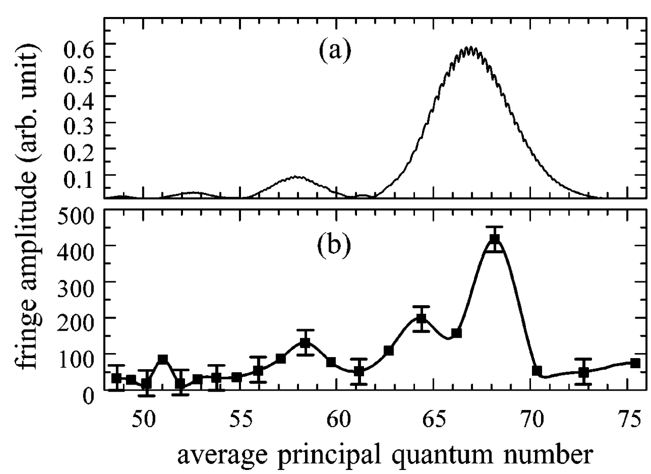

FIG. 3. Visibility of Ramsey fringes as modified by core laser. In (a) a theoretical model is presented for comparison. The experimental results are shown in (b). Note that only the synchronized case ($\bar{n} = 67$) producing a nondecaying wave packet has significant visibility at the fixed delay time.

visibility peak remains in Fig. 3(b), i.e., the peak associated with $\bar{n} = 67$. This preserved visibility is the signature of a nondecaying wave packet. It is the synchronization of the ICE process with the classical frequency that enables the channel interaction to maintain the wave packet in a nondecaying form. The fixed delay time of 86 ps is twice τ_{cl} for this synchronized wave packet. Without this synchronization, the visibility of this peak would have been greatly reduced. For example, the peak for $\bar{n} = 57$ is a factor of 4 smaller than the synchronized peak. This peak corresponds to a wave packet that has executed three orbits to reach the set delay time. The existence of this experimentally observed remnant strengthens the agreement with the theoretical predictions and fits with the experimental estimates for the autoionization decay time.

We have observed striking modifications of the behavior of a wave packet due to the presence of a strong ICE. In particular, when the Rabi oscillations of the core are synchronized to the classical oscillations of the wave packet, the decay of the visibility of this synchronized wave packet is markedly less than for a nonsynchronized wave packet. This nondecaying aspect of the wave packet is associated theoretically [22] with a nondispersing characteristic. Explicit verification of this nondispersing character requires following this wave packet for many classical periods. However, the technical challenges of such an experiment are considerable. Also, for the present experimental system, such delay times would approach the excited state lifetime of the core ($\sim$6 ns), and so introduce a new decoherence mechanism to the problem.

This and other recent experiments open several interesting areas of investigation. For example, these experiments may also be viewed as a study of the control of decoherence. This work controls or limits the irreversible decay of an electron to continuum. The preservation of coherence in the presence of a strong decay process (e.g., autoionization) is of great interest to such fields as quantum computing. Also, these electron-electron correlations experiments address not only these control and shaping issues, but also offer great promise for pushing the limits of quantum measurement.

This work was supported by the National Science Foundation under Grant No. PHY-9733643.

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