Redistribution of Rydberg States by Intense Picosecond Pulses

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Starting with Ba atoms in one Rydberg state we have observed the population of adjacent Rydberg states when the atom is exposed to an intense 1.5-ps laser pulse. The initial Rydberg state population is redistributed by a Raman-like two-photon process, rather than simply being photoionized. This population trapping, which is associated with stabilization against ionization, is described in terms of Rydberg anti-wave-packets.

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Recently there has been a great deal of theoretical interest in the possibility of stabilization against ionization of atoms in high-intensity radiation fields. There are two general types of stabilization. First, if the radiation is linearly polarized and its intensity and frequency are very high, the electron wave function splits into parts spatially removed from the ion core [1]. Since ionization occurs by the absorption of photons when the electron is near the core, the ionization rate is suppressed [2,3]. The suppression is not limited to linear polarization but occurs with circular polarization as well [3]. This form of stabilization can be viewed as steady-state stabilization. In the second mechanism of stabilization [4–6], a short laser pulse forms a coherent superposition of states in which the electron's wave function has vanishing amplitude at the ion core at the end of the pulse. At this time, and at periodic recurrences of vanishing amplitude at the core, the possibility of substantial ionization is precluded. In general, at later times the electron will be found near the ion core, so the suppression of ionization which occurs in this case is only periodic.

Our interest here is in the second form of stabilization. Specifically, we are interested in atoms in a single Rydberg state exposed to an intense laser pulse of duration short compared to the classical round-trip time of the electron in the Rydberg state. In the simplest picture the short laser pulse ionizes only those atoms in which the Rydberg electron is near the core when the laser pulse passes by. For most atoms, in which the Rydberg electron is far from the core during the pulse, the Rydberg electron is unaffected by the laser pulse. In more quantum-mechanical terms, the laser pulse burns a hole, at the ionic core, in the wave function of the initial state. The resulting wave function can no longer be described as a single stationary state, but is instead a coherent superposition of many states. This superposition is formed by a Raman process via the continuum, the Λ process shown in Fig. 1. The fact that the laser pulse duration is short compared to the round-trip time ensures that the bandwidth of the laser is adequate to drive the transition from the initial state to the continuum and back down to a different final Rydberg state. The hole formed in the wave function might be called an anti-wave-packet since it oscillates back and forth between small radius and large radius in time, just as does a normal Rydberg wave packet [7]. In any case, when the hole is at the ionic core, the ionization is suppressed and the atom is stabilized against ionization. An equivalent statement in the frequency domain is that together with the continuum the bound coherent Rydberg states form a Λ system in which the upper state, the continuum, is not populated [8].

One of the manifestations of the second form of stabilization is the transfer of population from the initial Rydberg state to adjacent Rydberg states. In this paper we present the experimental observation of the transfer of population from an initial single Rydberg state to adjacent Rydberg states by an intense picosecond laser pulse. The experiment is performed in three steps. First, a sin-

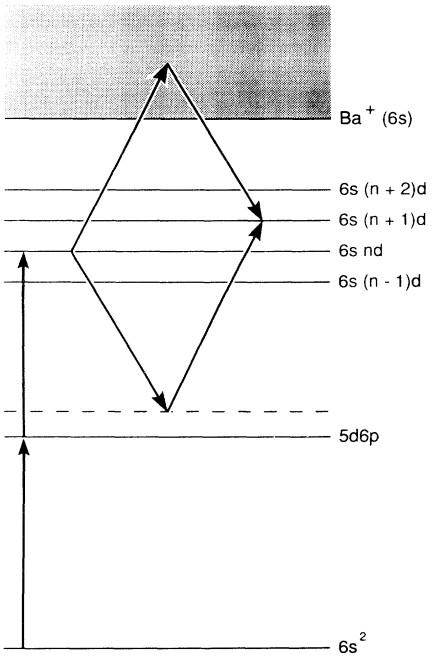

FIG. 1. Schematic energy-level diagram of the two ways of populating neighboring states starting from one Rydberg state. The Λ path corresponds to coupling via the continuum while the V path represents coupling via a virtual bound state (---). As shown, the initial Rydberg state is created by two-step resonant ns laser excitation from the ground, $6s^2$, state via the $5d6p$ state prior to the exposure to the intense ps laser.

gle Rydberg state is prepared by ns laser excitation of ground-state Ba atoms. Second, the Rydberg atoms are exposed to the intense ps laser pulse. Third, the final Rydberg-state distribution is determined by means of field ionization. The experimental setup is described briefly since a more detailed description is given elsewhere [9]. An effusive atomic barium beam passes between two electric-field plates where it is crossed by laser beams focused with a 50-cm lens. Using two ns dye lasers we excite the Ba atoms from the ground, $6s^2\,{}^1S_0$, state to the $5d6p\ {}^3D_1$ state, and then to a $6snd$ Rydberg state of $n \approx 26$, the vicinity of the $5d7d\ {}^1D_2$ perturber. After 100 ns the Rydberg atoms are exposed to the ps laser pulse. This delay is considerably longer than the $5d6p$ state lifetime and sufficiently short to avoid decay of the Rydberg state. The polarizations of all lasers are linear and parallel to each other. The ps laser system consists of a dual-jet synchronously pumped dye laser which is amplified in a dye amplifier pumped by the second harmonic (532 nm) of a 100-ps Nd-doped yttrium aluminum garnet laser. The system produces 1.5-ps pulses of 0.5-mJ energy at a 15-Hz repetition rate. Based on the measured spot size we estimate the intensity at the focus to be 4×10^{12} W/cm^2. After the ps laser pulse an adjustable ramped extraction field is applied to the interaction region, ionizing Rydberg-state atoms and accelerating the resulting electrons towards the microchannel plate detector. Since different Rydberg states ionize at different field strengths of the electrical pulse, the arrival times of the electrons at the detector yield the Rydberg-state distribution.

As mentioned above, the Ba $6snd$ states are perturbed at $n \approx 26$ by having the $5d7d$ state inserted into the series. The effect is to admix $5d7d$ character into the Rydberg states and vice versa. The wave function Ψ of a $6snd$ state or of the nominal $5d7d$ state is given by $\Psi = \sqrt{\epsilon_R}\Psi_{6snd} + \sqrt{\epsilon_P}\Psi_{5d7d}$, where $\epsilon_R + \epsilon_P = 1$, and ϵ_R and ϵ_P are termed the Rydberg and perturber fractions. The largest perturber fraction, $\epsilon_P = 30\%$, occurs in the nominal $5d7d$ state, so all these states, including the $5d7d$ state, are predominantly Rydberg states [10]. In principle, the admixture of the $5d7d$ state is not important, but in practice it is. Classically, on each orbit the Rydberg electron spends more time near the core than it would in a pure Rydberg state, raising the photoionization cross section.

In Fig. 2 we compare the resulting field ionization spectrum, with and without the intense ps laser, when the second ns laser is tuned to the $5d7d$ state. Without the intense ps pulse the spectrum consists basically of a single peak, corresponding to field ionization of the initially populated Rydberg state, as shown in Fig. 2(b). With the ps laser population appears in neighboring states, as shown in Fig. 2(a). In order to make the redistribution of the Rydberg states more clear the vertical scale of Fig. 2 is expanded to the point that the peak corresponding to the initially populated $5d7d$ state is truncated. In reality the population of the neighboring states is at most a few percent of that of the initial state.

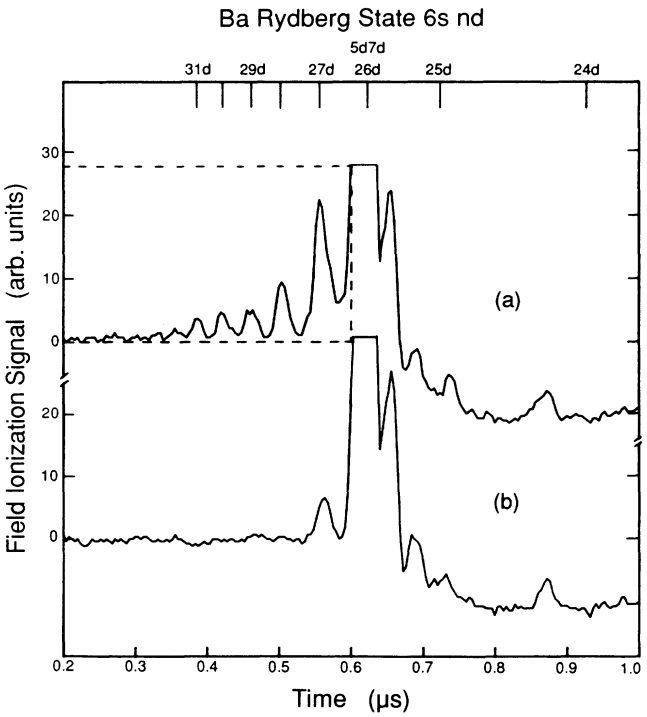

FIG. 2. Field ionization spectrum of Ba Rydberg states when the initial state is the $5d7d$ state. The $5d7d$ and $6s26d$ states ionize at the same field and are indistinguishable in this spectrum. Trace (a) is with the ps laser while trace (b) is without the ps laser. The peaks to the left of the saturated $5d7d$ signal in trace (a) correspond to excitation of the $27d$–$31d$ states by the ps laser.

Without the ps laser a small peak at the position of the $6s27d$ state is visible. Its presence is due to the fact that the second ns laser has some intensity at the excitation frequency of the $6s27d$ state. We do not observe any population transfer to states more bound than the initial state. The nearest lower-lying state, $6s26d$, has the same ionization field as the initially populated $5d7d$ state, and can therefore not be distinguished from it in our field ionization spectra. Furthermore, the channel plate is less sensitive to electrons arriving after the main peak due to saturation. Thus it cannot be concluded that there is no population of lower-lying states: We are simply not able to detect them.

Now we would like to address two questions. First, is more than one Raman transition needed to reach the outermost observed states such as $6s30d$ and $6s31d$? Second, which of the two Raman transitions shown in Fig. 1 populates the neighboring states: the Λ transition via the continuum, or the V transition via the virtual bound state?

To answer the first question we performed the follow-

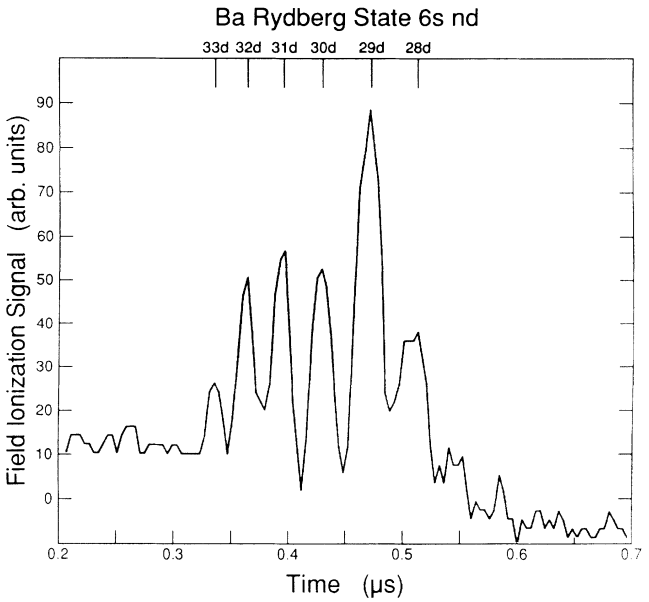

FIG. 3. Field ionization spectrum of Ba Rydberg states when the second ns laser is replaced by the ps laser. From the number of excited states we estimate the effective bandwidth to be 50 cm^{-1}.

ing experiment. The second ns laser, used to populate the Rydberg states from the intermediate $5d6p$ state, was replaced by the ps laser. A resulting field ionization spectrum is presented in Fig. 3. In the spectrum the coherent population of six different states ($6s28d$-$6s33d$) can be seen, corresponding to a width of 50 cm^{-1}. This width has two origins: first, the bandwidth of the ps laser, about 10 cm^{-1} and, second, the ac Stark shift of the Rydberg states with respect to the intermediate state. During the pulse the intensity changes, and the Rydberg states shift in and out of resonance, and, as a result, the total bandwidth of the radial wave packet created exceeds the laser bandwidth [11]. Note that the total number of states excited by the ps laser, shown in Fig. 3, is approximately the same as the number of states populated when the ps laser redistributes the population of the initial Rydberg state, as shown in Fig. 2. This observation is consistent with the population transfer occurring by a single Raman transition rather than by a sequence of Raman transitions. Furthermore, this conclusion is in agreement with the observations that only a small fraction of the population is redistributed and that the amount of population transfer does not diminish rapidly with increasing change in n.

To answer the second question we have observed the redistribution of the $5d7d$ Rydberg state for different wavelengths of the ps laser. The tuning range of 400 cm^{-1}, equal to 8 times the effective laser bandwidth, includes the wavelength of the $5d6p$-$5d7d$ transition. As shown in Fig. 4, no significant change in the final-state populations is observed as the ps laser is tuned. Since no influence of a bound resonance is observed we believe that

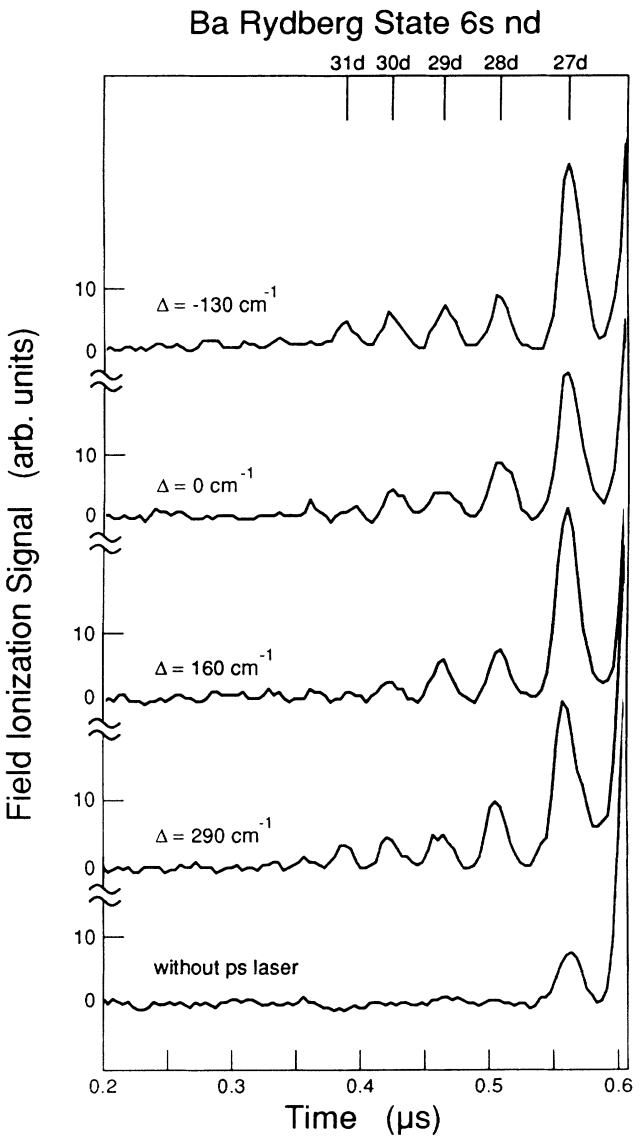

FIG. 4. Field ionization spectrum of Ba Rydberg states when the initial $5d7d$ Rydberg state is exposed to different wavelengths of the ps laser. The detuning Δ is given with respect to the $5d6p\,^3D_1$-$5d7d$ transition at 17650 cm^{-1}. Since no enhancement by the resonance is observed, we conclude that the two-photon coupling occurs via the continuum.

the redistribution occurs by a Raman process via the continuum, the Λ transition of Fig. 1. We also performed the experiment for initial states other than $5d7d$ but found no significant population of adjacent states, presumably due to too weak a coupling to the continuum.

We now discuss the time evolution of the wave function created by the redistribution of the population. We have calculated the amplitudes of the hydrogen Rydberg states after a $42s$ state is exposed to an intense 2-ps laser pulse. The duration of the intense pulse is short compared to the round-trip time T of the $42s$ state ($T=2\pi n^3$; 11 ps for $n=42$). This state is coupled to the ϵp continuum from

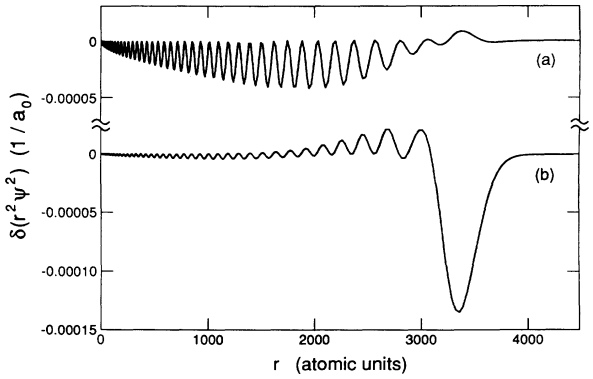

FIG. 5. Calculated difference $\delta(r^2\Psi^2)$ between $r^2\Psi^2$ for the hydrogen 42s state before and after exposure to an intense 2-ps laser pulse. (a) Immediately after the pulse has burned a hole in the initial-state 42s wave function near the ionic core. (b) After 5.5 ps the hole or anti-wave-packet has evolved to the outer turning point of the Coulomb potential.

which s and d Rydberg states can be populated. Since population of the s series is 20 times more likely, we ignore the d series. We first calculate the amplitudes a_n of the adjacent states following the method of Parker and Stroud [5]; i.e., we solve their Eq. (5) for the case of Rydberg states. From the amplitudes we construct the total wave function $\Psi(r,t)=\sum_n a_n \exp(i\omega_n t)R_{nl}(r)$. Although a more detailed description of the calculations will be given elsewhere, here we briefly summarize the results. In Fig. 5 we plot the difference, $\delta(r^2\Psi^2)$, between $r^2\Psi^2$ before the pulse, when the wave function is a pure 42s state, and at two times after the pulse, when the wave function is a coherent superposition of states around 42s. The intense pulse ionizes some of the wave function close to the core, and a hole is burned in the initial wave function at the origin, hence the large value of $\delta(r^2\Psi^2)$ near the origin immediately after the pulse, as shown in Fig. 5(a). This anti-wave-packet oscillates radially back and forth between the core and the outer turning point (3500 a.u.) of the potential, just as does a Rydberg wave packet [7]. In Fig. 5(b) we show the evolution of the anti-wave-packet after 5.5 ps, corresponding to about half of the oscillation time of the Rydberg electron, and indeed the anti-wave-packet, as manifested by $\delta(r^2\Psi^2)$, is close to the outer turning point.

In summary, we have observed the population of adjacent Rydberg states when a Rydberg atom is exposed to a short intense laser pulse. This population redistribution is one manifestation of the coherent population trapping mechanism of stabilization. We have, in addition, carried out time-dependent calculations of the wave function showing that the part of the wave function close to the core is ionized, creating a Rydberg wave packet with a hole. This anti-wave-packet oscillates in the atomic potential with the classical orbiting time.

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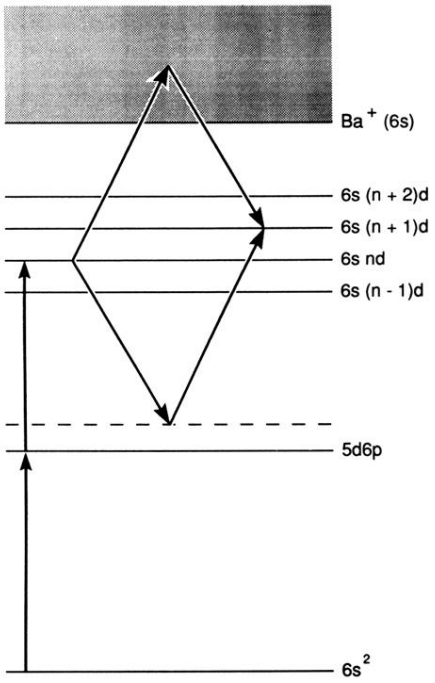

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