Ionization Suppression of Stark States in Intense Laser Fields

R. R. Jones and P. H. Bucksbaum
Department of Physics, University of Michigan, Ann Arbor, Michigan 48109-1120
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We have observed suppression of ionization in Rydberg atoms irradiated by an intense short-pulse laser field. We find that single-photon ionization of the outer electron in a Ba $6snk$ Stark state can be severely suppressed relative to high-order (≥ 3 photons) nonresonant multiphoton ionization of the tightly bound $6s$ core electron. This population trapping occurs in intense short laser pulses, where the bandwidth of the ionizing laser is greater than the spacings between adjacent Stark states, and may be due to a substantial modification of the initial Stark state via stimulated Raman processes through the continuum. The Ba^{+**} ion should be left in a highly excited superposition state, or wave packet.

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The development of intense, short-pulse lasers has spurred interest in laser-atom interactions in optical fields comparable to electron binding fields. Perturbation theory is not generally valid in this new regime. The intensity (I) dependence of n-photon processes is therefore not simply I^n. Nonetheless, until recently it was widely held that multiphoton ionization probabilities should increase with intensity, aside from trivial exceptions due to intensity-dependent resonances [1], at least until the rate becomes comparable to the laser frequency. Likewise, single-photon ionization probabilities were expected to depend on pulse fluence rather than intensity, so long as special cases such as autoionizing resonances were avoided.

These ideas have recently been called into question. Several theories now maintain that photoionization rates may saturate, or even actually decrease, with increasing laser-pulse intensity under general conditions of high frequency (i.e., single-photon ionization is possible) and high intensity. Several mechanisms have been proposed for this contrary behavior: laser-induced modification of the atomic wave function (so-called dichotomy) [2], destructive interference of ionization amplitudes [3], or Raman mixing in the continuum [4,5].

We report the observation of highly stable Stark states of barium subjected to intense 355-nm, 70-psec light pulses. These states remain stable at intensities where the nonresonant multiphoton ionization of the $6s$ core electron is saturated. We believe these experimental results can most easily be understood in terms of the model presented by Burnett *et al.* [5], in which Raman mixing processes between bound states and the continuum redistribute the atomic state into a superposition that cannot ionize. Their model suggests redistribution among high n states which are excited during the ionization of ground-state atoms by an intense laser; however, we have chosen to study redistribution among k states in an initially populated Stark manifold. The experimental technique we introduce is quite general, and may be used in the future to investigate other types of high-intensity atomic stabilization as well.

The experiment proceeds as follows: A Rydberg Stark state is ionized in an intense, high-frequency focused laser. Following ionization, the state of the atoms is examined. The presence of Rydberg ions indicates that the core electron, and not the Rydberg electron, has been ejected. The stability of the outer electron is consistent with the model of population trapping due to Raman redistribution among Stark states [4,5].

We use atomic Ba in this experiment, principally because its second electron may be multiphoton ionized in our laser pulse, a focused ($f/8$) 70-psec, 10-mJ, 355-nm third harmonic of a mode-locked and amplified neodymium-doped yttrium aluminum garnet (Nd:YAlG) laser. We have chosen the third harmonic of Nd:YAlG, rather than lower harmonics, because this frequency reduces the possibility of coupling between the core electron and our Rydberg electron via doubly excited resonances [6,7]. Correlation effects such as autoionization suppress the effect we want to observe by opening rapid ionization channels for the Rydberg electron. We have observed that the number of stabilized Rydberg atoms is reduced by nearly an order of magnitude if 1064-nm radiation is used instead of 355-nm radiation.

A thermal beam of ground-state Ba atoms ($\rho \approx 10^9$ cm^{-3}, chamber base pressure $\approx 10^{-7}$ torr), in the presence of a static electric field of ~ 100 V/cm, is excited in two steps: The $6s^2$ ground state is excited through the $6s6p$ resonance line using a 10-nsec, 553.7-nm, 100-μJ dye laser, and then to a $6snk$ Rydberg Stark state with $n > 20$ using a 10-nsec, 420-nm, 2-mJ dye laser. Both dye lasers are weakly focused through the same 500-mm lens ($\sim f/100$). This procedure provides sufficient laser fluence to substantially deplete the ground-state atoms in the interaction region.

The short-pulse laser propagates antiparallel to the dye lasers, and both are at right angles to the Ba beam. The static electric field is along the third orthogonal axis, and is produced by parallel field plates (see inset in Fig. 1). The ions created with the 70-ps laser drift in the static field through a hole in the upper field plate into a second field region defined by a third field plate. Any Ba^+ Rydberg states are field ionized in this region to form Ba^{2+}, and then all ions accelerate through a hole in the third field plate into an 8-cm field-free drift region before striking a dual microchannel plate detector. In this way, single ions, double ions, and Rydberg ions strike the detector at different times and may be counted separately.

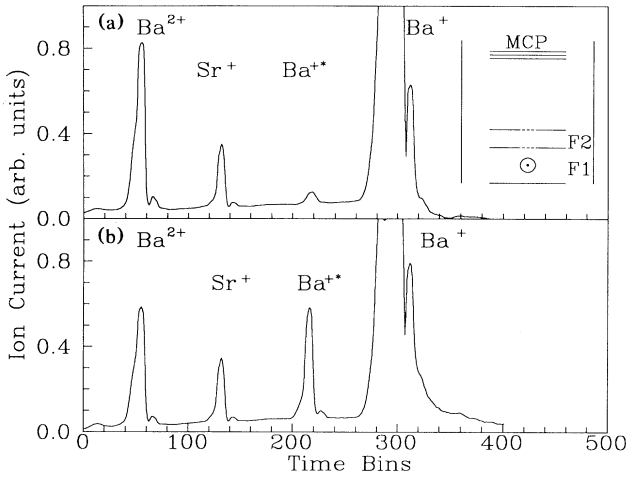

FIG. 1. Time-of-flight spectra showing the production of Ba^{2+}, Ba^+ Rydberg states, and ground-state Ba^+ ions using an (a) $n=25$ or (b) $n=35$ initial state. Both sets of data were taken with 162 V/cm in the interaction region. Inset: Schematic of the interaction region.

Two typical time-of-flight spectra are shown in Fig. 1. The most prominent feature by far is singly ionized Ba in its ground (or low-lying) state. A substantial but smaller population of doubly ionized Ba is also visible. Between these two Ba peaks are two additional features: singly ionized Ba Rydberg states, which have been field ionized during their flight to the detector; and Sr^+, which appears due to impurities in the Ba metal. The Sr^+ peak is unaffected by the dye lasers, and so provides a good normalization. The Ba^{+*} signal indicates suppression of photoionization of the Stark Rydberg states.

Figure 1(a) shows excitation of a state with principal quantum number $n=25$ and Fig. 1(b) shows a state with $n=35$. The field in the interaction region is $F_1=162$ V/cm, and the field that ionizes surviving Rydberg ions as they drift to the detector is $F_2=4.260$ kV/cm. Note that the increase in the population of the Ba^+ Rydberg states is matched by a decrease in the Ba^{2+} signal. This indicates that both signals originate in the same focal volume and that a substantial fraction of the Ba^{2+} is due to double ionization of Stark states and not simply of ground-state atoms.

Figure 2(a) shows the ratio of the Ba^+ Rydberg state production to the Ba^{2+} production as a function of the $6s6p$-$6snk$ transition frequency. The pulse energy in the 70-psec laser was 10 and 5 mJ for the upper and lower traces, respectively. The electric field F_1 was 220 V/cm. Structure in the ratio is visible as the laser is scanned over adjacent Stark manifolds. Figure 2(b) shows the $6s6p$-$6snk$ excitation probability over the same wavelength range as the data in Fig. 2(a). Stark states with larger than average s- or d-state components are more prominent because of the excitation scheme. Clearly the maxima in the Ba^{+*} signal does not always correspond to maxima in Ba^+ excitation. Figure 2(c) shows a calculation of the Ba energy levels in a 220-V/cm field. Note that the d states in Ba do not mix strongly with the inner manifold states until adjacent manifolds cross.

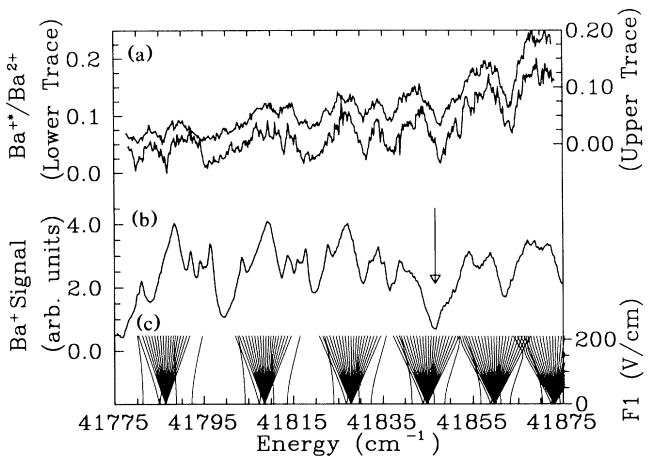

FIG. 2. (a) Ratio of the production of Ba^+ Rydberg states to Ba^{2+} as a function of initial-state energy for different values of 355-nm pulse energy. The upper and lower traces correspond to pulse energies of 10 and 5 mJ, respectively. A constant value of 0.05 was added to the upper trace so that it could be distinguished from the lower one. (b) Ba^+ field-ionization signal showing the relative amounts of initial-state population for the signal in (a). Note that a maximum in the Ba Rydberg cross section does not necessarily correspond to a maximum in the ratio of (a). The arrow designates the energy of a member of the $5d7d$ multiplet which perturbs the character of the $6snd$ series near this energy by introducing a non-Rydberg component to the wave function [8]. There is a decrease in the excitation cross section to the Stark state, and also an increase in finding both electrons near the core. (c) Calculation of Ba Stark energy levels ignoring fine structure and the $5d7d$ perturber. The quantum defects for the s, p, and d states are 4.21, 3.90, and 2.75, respectively.

We will now show that there is a connection between electron stabilization in our experiment and angular momentum mixing in an electric field. The connection is straightforward: A Stark state with principal quantum number n contains all angular momentum components with $l<n$, but only the low angular momentum components contribute to photoionization. (The photoabsorption probability is only appreciable near the atomic core where the nucleus is available to conserve momentum.) The suppressed ionization that we observe comes from a reduction of the low angular momentum character of the wave function.

The fluence in our short laser pulse is $\approx 10^3$ J/cm^2, which is sufficient to one-photon ionize any state with cross section greater than ~ 500 b. At this fluence we observe total ionization of $6sns$ and $6snd$ states at the values of n used in this experiment. An external electric field introduces a new time scale τ_s in the system. τ_s is the characteristic time for mixing between l states. Ionization of the mixed (i.e., Stark) $6snk$ states is suppressed when the laser pulse is shorter than this time.

The characteristic rate $2\pi/\tau_s$ for mixing within a Stark state is equal to $3Fn$ a.u., which is the average of the static field dipole matrix elements between the angular momentum states. Here F is the electric field and n is the principal quantum number of the manifold states. (Unless otherwise noted atomic units will be used for the remainder of the paper.) This rate has a simple spectroscopic meaning: It is the energy of separation between Stark states with different k (the parabolic quantum number) in a single Stark manifold [9].

A laser pulse much shorter than the mixing time τ_s can only project the low angular momentum components of the Stark state into the continuum, because the high angular momentum components can only couple to the continuum by mixing to lower l states first. The laser pulse therefore leaves behind a "trapped" nonstationary state in the atom consisting of high l orbitals. As a result, *the ionization probability versus laser intensity saturates at a level which can be much less than unity. This is laser-enhanced stabilization.*

The nonstationary state created by the laser evolves in time similar to a standard parabolic wave packet [10]. After a time shorter than but on the order of τ_s, low angular momentum components are established and further ionization is possible with another light pulse. On the other hand, since the parabolic wave packets are nondispersive, the low l character should oscillate with period τ_s.

This population trapping has also been considered in the frequency domain [4,5]. When the laser pulse width is less than τ_s, the bandwidth is greater than the separation between adjacent states in the Stark manifold. Stimulated Raman processes then begin to compete with ionization, coherently redistributing population to the other nearby bound states. Others have shown that this process rapidly produces a superposition state whose ionization is suppressed through destructive interference [4,5].

The results shown in Fig. 1 are now easily understood. The $n=25$ state shown in Fig. 1(a) has a Stark mixing time, $\tau_s=60$ ps, which is slightly less than the laser-pulse duration; and the $n=35$ state of Fig. 1(b) has a mixing time, $\tau_s=230$ ps, which is much longer than the laser pulse. The probability of finding the electron in a low angular momentum state is nearly unity in Fig. 1(a) and is almost negligible in Fig. 1(b).

Since the data in Figs. 1 and 2 were taken in static fields near a field value of $F=1/3n^5$ where adjacent Stark manifolds cross, it is difficult to assign a mixing time; the level spacing is no longer constant across the manifold. Instead, the Stark mixing time near the crossing is approximately equal to the inverse of the level splitting at the crossing, $\tau_s=2\pi n^4$.

The excitation of lower-lying Stark states on the left-hand side of Fig. 2 is dominated by low angular momentum ($6sns$ and $6snd$) states. These have large quantum defects and do not mix thoroughly with higher l states in the electric field; as a result they contribute little to the trapped Ba^+ production. In the higher-lying states on the right-hand side of the spectrum, the angular momenta are more thoroughly mixed by the electric field, and the ratio in Fig. 2(a) closely follows the excitation signal. Figure 2 also shows the increase in the production of ion Rydberg states as the Stark mixing time increases with increasing principal quantum number. The ratio of Ba^{+*} to Ba^{2+} approaches unity at high n.

The Rydberg ions formed in this experiment are not eigenstates of the Ba^+ Hamiltonian. When the core electron is ionized, the Rydberg electron sees a sudden change in the ion potential. If we assume that the expectation value for the radius of the Rydberg orbit remains unchanged, then Bohr theory predicts a new principal quantum number $n'=\sqrt{2}n$. The sudden approximation predicts a coherent superposition of new states in the Rydberg ion. The results of a calculation assuming hydrogenic Stark states are shown in Fig. 3 for an initial state with parabolic quantum numbers $n=20$ and $n_1=10$ in a field of 150 V/cm. Note that the maximum of the overlap integral squared occurs very near to $n'=\sqrt{2}n$. Thus, the parabolic oscillation period $\tau_s\approx 2\pi n^4/Z^2$ a.u. is nearly unchanged following ionization.

We have verified this "frozen electron" approximation by detecting Ba^+ Rydberg states while the field in the second ionization region is varied. Signal originating from Ba^+ Rydberg levels disappears completely if the field in the ionization region has a value of $F\approx 1/8n^4$, in agreement with the classical field-ionization threshold, $F_c=Z^3/16n'^4$, for $n'=\sqrt{2}n$ [9]. This agrees with the overlap calculation; virtually no states with $n'>\sqrt{2}n$ are populated.

In conclusion, we have studied the production of Ba^+ Rydberg states via the photoionization of the $6s$ core electron in an initially populated $6snk$ Stark state. The existence of the Ba^+ Rydberg ions is a clear indication of inhibition of ionization for these states.

Finally, we note that this experimental technique can also be used to search for more exotic forms of high-

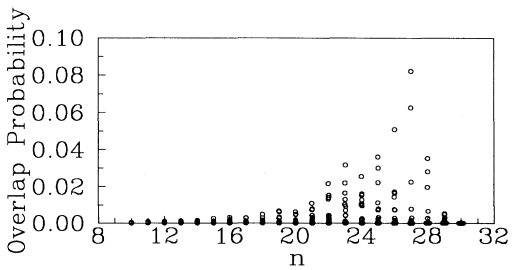

FIG. 3. Calculation of the overlap matrix elements $|\langle n=20, n_1=10, Z=1|n',n_1', Z=2\rangle|^2$ as a function of the parabolic quantum numbers n' and n_1', assuming hydrogenic Stark states in a field of 150 V/cm. Each data point corresponds to a different final state $|n,n_1\rangle$. Note that the maximum in the integral occurs for $n'\approx\sqrt{2}n$.

intensity stabilization. The formation of a Rydberg wave packet is a convenient way to "shelve" the outer electron during a time when the laser pulse is rising in intensity. We might then observe a different form of stabilization (e.g., "dichotomy" [2]), at the peak intensity, when the wave function returns to the core. We already have a test of this in the present data, since Fig. 2 includes states with mixing times from $\tau_s = 27$ ps at the low-frequency side of the plot to $\tau_s = 72$ ps at the high-frequency end. In this range there is ample time for the electron probability to reestablish itself at the core. Although there are minor amplitude discrepancies in the data taken at high and low laser intensity, particularly at the low-frequency end of Fig. 2(a), there is no strong evidence for any intensity dependence in the production of Ba^+ Rydberg states for any of the initial states studied. We estimate that the peak intensity was 10^{14} W/cm^2. Therefore, if high-intensity stabilization does occur, it has a lower threshold at some intensity above 10^{14} W/cm^2 in this system. Future work will explore other forms of stabilization, using this technique.

While preparing this work we became aware of recent experiments in which similar effects were observed in high n Rydberg states, where the characteristic time scale is the Kepler orbit time, and ultrashort pulses are necessary in order to observe suppressed ionization [11]. In this case, the laser creates a superposition state with vanishing probability density at small r, rather than low l.

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