Inner-Electron Multiphoton Ionization of Barium Rydberg States with Picosecond Pulses

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We have studied multiphoton ionization of Ba $6snl$ Rydberg states by intense ps pulses and have shown that the duration of the laser pulse compared to the orbit time of the Rydberg electron determines the laser-atom interaction. For high-n states with a long orbit time, the loosely bound Rydberg electron is not affected by the intense laser pulse. Instead the inner electron is ejected by absorption of five photons, leaving the Ba^+ ion in a Rydberg state. For low-n states the outer electron is ejected and the residual Ba^+ ion can subsequently be multiphoton ionized to Ba^{++}.

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Several aspects of correlation in two-electron or two-valence-electron systems have been studied extensively over the past decade. One of these is the question of whether or not both electrons are ejected simultaneously when the atom is exposed to a high-intensity short laser pulse. Most of the latter experiments have been one-color laser irradiation of alkaline-earth atoms in the ground state [1,2]. The overall conclusion from these investigations is that the dominant mechanism for double ionization is a sequential removal of the electrons with a low-lying state of the singly charged ion acting as an intermediate state.

In the present work we have studied multiphoton ionization with a 5-ps laser pulse starting from bound barium $6sns$ and $6snd$ Rydberg states instead of from the ground state. While we see no evidence for simultaneous ejection, we do see a novel phenomenon. For low-n states in which the radial round-trip time for the Rydberg electron is short compared to the duration of the laser pulse, Ba^{++} is produced by the now familiar sequential multiphoton-ionization process [Fig. 1(b)]. The Rydberg electron is ejected by one-photon absorption, and the resulting Ba^+ ion absorbs five photons to produce Ba^{++}. In contrast, for high-n states with a round-trip time longer than the laser pulse, the intense laser field does not result in ejection of the weakly bound Rydberg electron. For these high-n states only a small fraction of the wave function will pass by the core during the laser pulse and since ionization takes place close to the core, ionization of the Rydberg electron is unlikely. Instead the strongly bound $6s$ electron is multiphoton ionized and the Rydberg electron is projected onto the Ba^+ $n'l$ Rydberg states of the same radial size [Fig. 1(a)]. Its quantum state changes due to the electron shakeup associated with the sudden change in the Coulomb potential of the core [3]. We have determined experimentally the relation between the principal quantum number of the initial Ba Rydberg state and that of the final Ba^+ Rydberg state.

The basic experimental setup consists of a crossed atomic-laser-beam arrangement, which is described in detail elsewhere [2]. An effusive atomic barium beam passes between two electric-field plates where it is crossed at 90° by the laser beams focused with a 50-cm lens. The Rydberg states of barium are created by two-step resonant laser excitation via the $6s6p\ ^1P_1$ state. Since both beams are linearly polarized, Ba $6snd$ as well as Ba $6sns$ Rydberg levels are populated. The ns dye lasers pumped by the harmonics of an 8-ns Nd-doped yttrium aluminum garnet (Nd:YAlG) laser have a linewidth of 1 cm^{-1}, allowing resolution of Rydberg states up to $n\sim45$. Since the radiative lifetimes of the excited Rydberg states are long, typically >1 μs, the ps laser is applied 50 ns after the ns lasers. The linearly polarized amplified ps laser has a 70-μm-diam focal spot and a peak intensity $\sim1\times10^{12}$ W/cm^2. Approximately 200 ns after the ps laser pulse, an adjustable extraction field of up to 3.6 kV/cm is applied to the interaction region, ionizing atoms and ions in Rydberg states and accelerating Ba^+ and Ba^{++} ions into a 6-cm-long field-free flight tube. Adjusting the amplitude of the extraction field pulse allows us to analyze the Ba and Ba^+ Rydberg state distributions. The detector consists of a dual microchannel-plate arrangement, and data acquisition follows standard time-of-flight procedures using gated integrators and computer recording. The temporal gate for the Ba^{++} signal is set small enough to exclude signals from contaminant strontium but sufficiently long to include the signal from field ion-

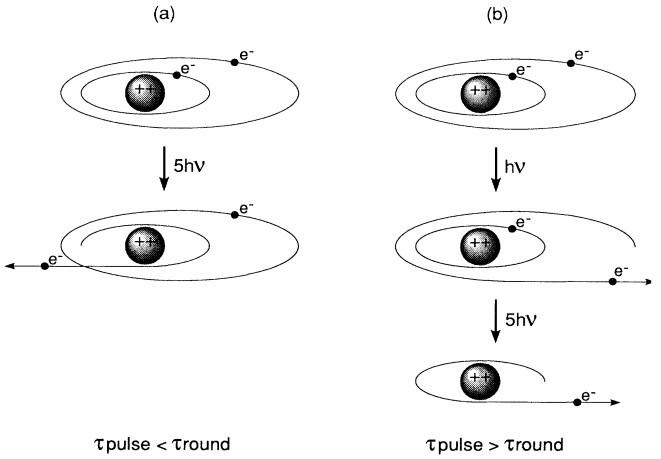

FIG. 1. Schematic representation of the interaction between the ps laser pulse and Ba $6snl$ atom for (a) high-n state and (b) low-n state.

ization of Ba^+ Rydberg states.

The observations consist of spectra of Ba^{++} recorded while scanning the wavelength of the second ($6s6p$-$6snl$) ns laser over the Rydberg series. Spectra are taken at different wavelengths of the ps laser and different extraction fields. Starting well above the $Ba^+(6s)$ limit the second ns laser wavelength is scanned over Ba $6snl$ states down to $n\sim 20$. The frequency of the ps laser is tuned to a three- or four-photon bound-bound resonance in the Ba^+ ion originating from the $Ba^+(6s)$ or $Ba^+(5d)$ states, otherwise very little Ba^{++} is observed. A detailed description of the resonances in the Ba^{++} production is given in Ref. [2]. In Fig. 2(a) a typical Ba^{++} spectrum, obtained with the ps laser at the Ba^+ $6s_{1/2}\rightarrow 7p_{3/2}$ three-photon resonance at 600.5 nm, and an extraction field of 3.6 kV/cm, is shown. At the beginning of the scan the Ba^{++} signal is at a high level due to the copious quantity of $Ba^+(6s)$ ions produced by the ns lasers. As the second ns laser is scanned into the bound region the signal exhibits a smooth decrease to a plateau equal to the signal level with no ns lasers. Figure 2(b) contains a section of the wavelength scan of Fig. 2(a) recorded at a slower scan rate. This scan reveals that the structure in the Ba^{++} signal corresponds to the Ba Rydberg states. Figure 2(a) shows that the excitation of high Rydberg states of Ba enhances the production of Ba^{++} by a factor of 2, but when the wavelength of the second ns laser is increased towards $n\sim 30$ the enhancement gradually disappears.

A most useful insight into the mechanism by which the Ba^{++} is produced is obtained by lowering the amplitude of the extraction field pulse. If we lower the amplitude from 3.6 to 1.0 kV/cm, there is no effect on the Ba^{++} signal. The data are identical to those of Fig. 2. Reducing the field to 590 V/cm, however, produces a threshold in the Ba^{++} signal at $\lambda=418.69$ nm, corresponding to a Ba $6snl$ state of effective quantum number $n_l^*=35.3$, as shown in Fig. 3(c). The redmost part of the enhancement of the Ba^{++} signal of Fig. 2 is no longer observed. Further reductions in the field lead to shifts of the threshold in the Ba^{++} signal to the blue, approaching the ionization limit at $\lambda=417.15$ nm, as shown in Figs. 3(b) and 3(a). The dependence of the Ba^{++} signal on the extraction field indicates that the ps laser is driving the Ba Rydberg atoms to Ba^+ Rydberg states, which are only detected as Ba^{++} if the field pulse is high enough to ionize them. A more quantitative analysis of the variation of the wavelength, or effective quantum number of the $6nsl$ states, with the threshold field shows that the Ba and Ba^+ Rydberg orbits are of the same size. For each value of the field pulse below 1 kV/cm, inspection of spectra such as those of Fig. 3 gives the value of the effective quantum number n_l^* of the Ba $6snl$ Rydberg state at which the threshold occurs (Table I). From the value of the field we can determine the effective quantum number n_{ll}^* of the Ba^+ Rydberg state to which the threshold corresponds using the classical ionization field,

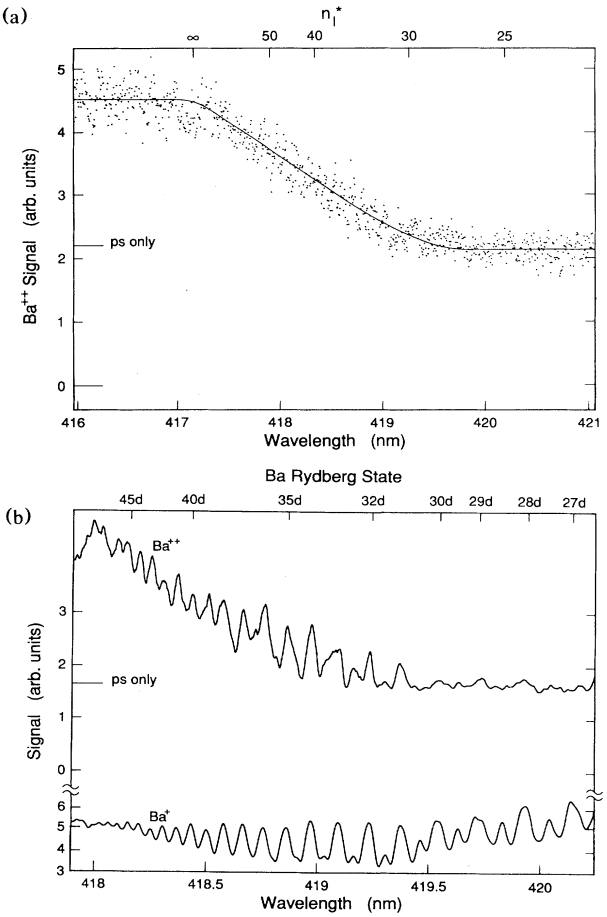

FIG. 2. The Ba^{++} signal recorded as a function of the wavelength of the ns laser. (a) A scan from well above the $6s$ continuum to $n\sim 22$. (b) A section of the wavelength region recorded at a slower scan rate. The bottom curve in (b) displaying the Ba^+ signal serves as a reference for the positions of the Ba $6snl$ Rydberg states.

$$E_{FI}=Z^3/16(n_{ll}^*)^4 \quad (1)$$

(in a.u.), where $Z=2$ is the charge of the Ba^{++} core. Using Eq. (1) we can determine n_{ll}^* of the highest Ba^* Rydberg state that is populated. These values are given in Table I, as are the values of the ratio n_{ll}^*/n_l^*. The average ratio of the two effective quantum numbers is $n_{ll}^*/n_l^*=1.32$.

If the inner $6s$ electron is ejected rapidly and the outer electron is simply projected from the initial Ba $6snl$ Rydberg state onto the Ba^+ $n'l'$ Rydberg state, the probability of populating the $Ba^+(n'l')$ state is given by the square of the overlap integral $|\langle nl|n'l'\rangle|^2$ [4]. Using one-electron Rydberg wave functions, constructed by a numerical integration of the radial differential equation for a Coulomb potential [5], we have calculated the overlap integrals, where $l=l'$ by orthogonality. Assuming that the

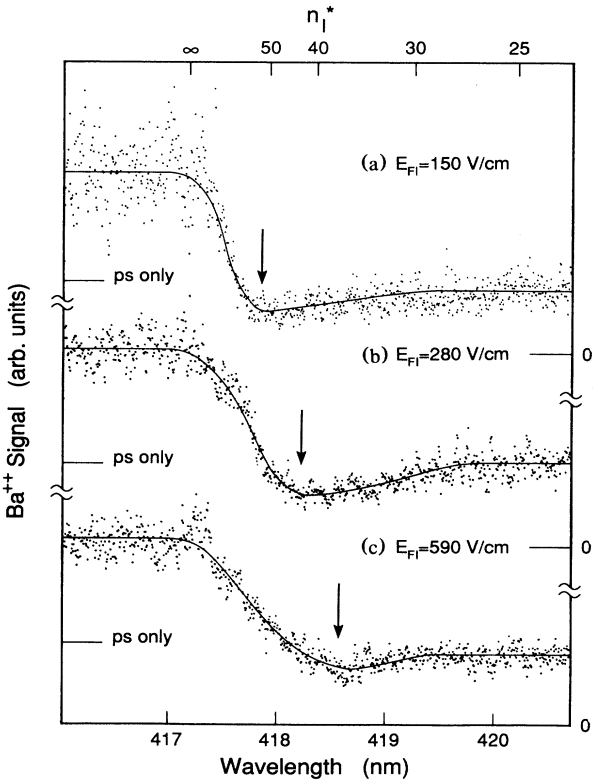

FIG. 3. Same as in Fig. 2, but for three different extraction fields. The arrows mark the positions of the thresholds. The curve drawn through the experimental points serves to guide the eye.

outer electron is projected onto the Ba^+ Rydberg states in this way is equivalent to assuming that there is minimal correlation between the two electrons, which is certainly the case in the $6snl$ state. For both s and d states the calculations resulted in the ratio

$$n_{II}^*/n_I^* = 1.40\,, \tag{2}$$

where n_{II}^* is the effective quantum number for the highest Ba^+ $n'l$ state that is populated. Alternatively, the expectation values of the orbital radii of the Ba and Ba^+ Rydberg states are the same if $n_{II}^*/n_I^* = \sqrt{2}$. The good agreement with the average value of Table I indicates that the Ba nl electron is projected onto the Ba^+ states. To determine any dependence of the ratio on the angle between the polarization of the ps laser and the ns lasers, the experiment was performed with the ps laser polarization parallel as well as perpendicular to the ns laser polarization. Since mostly $6snd$ states are excited, one would in the parallel case expect a higher probability for interaction between the two electrons, because the $6s$ electron is likely to be ejected in the direction of the $m=0$ nd state. However, no significant change in the ratio was observed for the two polarization cases supporting that no interaction between the two electrons occurs.

We now return to the question of why the excitation of the Ba^+ Rydberg states occurs only for initial Rydberg states of $n > 30$. To understand this point we recall that absorption of photons involving the outer electron to produce Ba^+ only takes place when the outer electron is close to the nucleus [6]. A Rydberg electron in a high-n state spends most of its time far away from the core. If the laser pulse is short compared to the radial round-trip time, the electron has a very small probability of passing near the core during the laser pulse. Consequently, the probability of its absorbing a photon is small even if the laser intensity is high. At higher intensities the ps laser will remove the small r part of the wave function, forming a dark wave packet by resonant two-photon excitation, via the continuum, of nearby Rydberg states, as suggested by several authors [7]. The classical round-trip time is given by

$$\tau_{\text{round}} = 2\pi(n_I^*)^3 \tag{3}$$

(in a.u.) and is 5.0 ps for $n_I^* = 32$. For $n_I^* \gg 30$, τ_{round} is much larger than the 5-ps duration of the laser pulse, meaning that most of the wave function of the nl electron is not affected by the laser field. Instead, the inner $6s$ electron is multiphoton ionized by five photons, leaving the residual core in a Ba^+ Rydberg state.

When Ba $6snl$ states with lower-n quantum numbers are excited the round-trip time is decreased, and the probability for photoionizing the outer electron to produce Ba^+ $6s$ and $5d$ ions during the pulse increases. Ba^{++} can now be produced by subsequent five-photon absorption from the $Ba^+(6s)$ state but *not* from the $Ba^+(5d)$ state since the wavelength of the ps laser does not match any Ba^+ $5d$ resonances. During the ps pulse a fraction of the Ba $6snl$ population is transferred to the metastable $Ba^+(5d)$ state, which explains the overall decrease in the Ba^{++} signal when the second ns laser is scanned towards lower-n states [Fig. 2(a)].

For states with $n^* \ll 30$ where $\tau_{\text{round}} < \tau_{\text{laser}}$, the Ba^{++} signal is approximately the same as without the ns lasers and no pronounced Rydberg structure is present any longer [Fig. 2(b)]. This suggests that the ratios of $Ba^+(6s)$ to $Ba^+(5d)$ ions produced are almost equal for direct three-photon ionization of $Ba(6s^2)$ and for photoionization of a low Rydberg state.

TABLE I. Experimental determination of effective quantum number for initial Ba Rydberg state (n_I^*) and final Ba^+ Rydberg state (n_{II}^*).

E_{FI} (V/cm)	n_I^*	n_{II}^*	n_{II}^*/n_I^*
82	54.2	74.8	1.38
146	48.0	64.8	1.35
198	44.9	60.0	1.34
280	40.1	55.0	1.37
400	38.4	50.4	1.31
433	40.6	49.4	1.22
590	35.3	45.7	1.29

The present investigations have been carried out with the ps laser set to four different $6s$ resonances [2] ($6s_{1/2} \rightarrow 7p_{3/2}$, $6s_{1/2} \rightarrow 7p_{1/2}$, $6s_{1/2} \rightarrow 8d_{5/2}$, $6s_{1/2} \rightarrow 9s_{1/2}$). The observations are identical to the ones already discussed. In particular, the onset of the increased Ba^{++} signal at $n^* \sim 30$ is perfectly reproduced in all four cases, strongly supporting the relation between the pulse duration and the radial round-trip time. The experiment was also carried out with the ps laser fixed at various Ba^+ $5d \rightarrow n_1 l_1$ resonances. In this case the Ba^{++} signal observed with the ps laser alone is decreased when the ns lasers are added, the second tuned just above the $Ba^+(6s)$ limit. The ns lasers convert many of the ground-state Ba atoms into $Ba^+(6s)$ ions, which are not affected by the ps laser. As the second ns laser is scanned towards lower-n states a slight increase in the signal was observed, indicating a sequential production of Ba^{++} from Ba $6snl$ via $Ba^+(5d)$.

As noted, we only observe the excitation of the Ba^+ Rydberg states when the ps laser is set to Ba^+ $6s \rightarrow n_1 l_1$ resonance. The multiphoton absorption from Ba $6snl$ is presumably resonantly enhanced by $n_1 l_1 n_2 l_2$ states lying just below the $Ba^+(n_1 l_1)$ state. For very-high-n states the $6s \rightarrow n_1 l_1$ resonance is negligibly affected by the presence of the Rydberg electron [8]. Without resonant enhancement, increased laser intensity is required to effect multiphoton ionization of the inner $6s$ electron, and an experiment along these lines is under way [9].

Finally, we would like to address the similarities between our observations of the absence of the ionization of high Rydberg states and a previous experiment [10]. In the present experiment the Rydberg electron is not ejected because most of its wave function does not return to the core during the short laser pulse. In the previous experiment, multiphoton ionization of Xe ground-state atoms was studied with Rydberg states acting as intermediate levels. For Rydberg states with an orbit time long compared to the pulse duration, the wave packet created did not return to the core during the pulse and the Rydberg electron was not ejected. In contrast, for low Rydberg states the wave function returned several times and the ionization was enhanced.

In conclusion, we have demonstrated the existence of two different regimes for the interaction between an intense ps laser field and a Ba $6snl$ Rydberg atom. The crucial factor determining the character of the interaction is the ratio between the radial round-trip time of the Rydberg electron and the pulse duration. For low-n states where $\tau_{\text{round}} < \tau_{\text{laser}}$ the Rydberg electron is ejected under absorption of one photon followed by multiphoton ionization of the residual Ba^+ ion. In the case of high-n states where the orbit time exceeds the pulse duration the inner $6s$ electron is ejected under multiphoton absorption and the Rydberg electron is projected onto Ba^+ Rydberg states of the same radial size.

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