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Population trapping in Kr and Xe in intense laser fields

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We have observed trapping of population in Rydberg states of atoms and ions exposed to intense laser radiation. The trapping occurs in nonstationary "wave-packet" superposition states, created, via ac Stark-shifted multiphoton resonances during the laser pulse. The formation of these highly excited bound states requires the nonresonant absorption of up to 20 or more photons; some of the ionic Rydberg states lie nearly 32 eV above the ionic ground state. All of the trapped states having binding energies much less than the photon energy but remain stable against ionization even when the multiphoton ionization probability of the atom is saturated.

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Numerous experiments have now shown that the cross section for above-threshold ionization (ATI) in atoms is greatly enhanced when one or more excited bound states have energies which lie an integer number of photons above the ground state [2,3]. Such resonances do not generally exist in the absence of the laser field, but are created by intensity-dependent ac Stark shifts during the laser pulse [3].

In typical studies, rare-gas atoms have been used as laser targets [2,3]. These atoms have relatively large ionization potentials (12–25 eV) compared to available photon energies (1–2 eV) and have no low-lying excited states [4]. Under these conditions, the ac Stark shifts between the ground and excited states are comparable to the ponderomotive shift of a free electron, which gains energy in the laser field at the rate of about $\lambda^2 I$ eV, where the wavelength λ is expressed in μm and the intensity I is in units of 10^{13} W/cm^2. For example, a Kr atom (ionization potential of 14.0 eV) exposed to $\lambda=0.77$-μm radiation ($h\nu=1.6$ eV) requires nine photons to ionize at low intensity. When the intensity exceeds 10^{13} W/cm^2, however, high-lying Rydberg states can be resonantly excited with nine photons and ionization proceeds through the absorption of one additional photon.

The main evidence for this ionization enhancement due to ac Stark-shifted resonances comes from ATI photoelectron spectra [3]. There has been no direct observation of highly excited Rydberg-state population ($n > 10$) during the photoionization process. In fact, for linearly polarized light, the single-photon ionization probability for a Rydberg state is saturated for intensities required to drive the nine-photon absorption; a natural conclusion is that the excited states ionize immediately.

Despite this reasoning, there are circumstances where population may be transferred from the ground state to Rydberg states and trapped there in a strong laser field [1]. By "trapped," we mean any transfer of population from one atomic state that can be ionized by an intense laser to other states that cannot be ionized by the laser, via some interaction with the same laser pulse. For example, recent experiments have shown that the single-photon ionization probability for Rydberg states in multielectron atoms can be substantially suppressed relative to the probability for ionizing one of the core electrons through high-order multiphoton absorption [5].

In the present work, we investigate the stability of Rydberg states in strong laser fields, where the intense laser must both excite the Rydberg state from the ground state, and also trap it so that it cannot ionize. We have observed population trapping in highly excited Rydberg states ($n > 14$) in Kr and Xe atoms which have been exposed to laser intensities greater than 5×10^{15} W/cm^2. In the following paragraphs we describe our experimental approach and discuss the results. We present a qualitative model that agrees with our data. We conclude with a comment on the compatibility of our results with recent studies of population transfer during ATI [2,3,6].

Intense laser pulses for this study are generated using chirped pulse amplification of a self-mode-locked Ti:Al_2O_3 oscillator [7–9]. This system produces 100-fs, 8-mJ laser pulses. A vacuum chamber with a base pressure of 2×10^{-8} Torr is backfilled with Kr or Xe to a pressure of 10^{-7} to 10^{-6} Torr. The laser is focused with a 250-mm lens to a ~20-μm spot between two parallel field plates as shown in the inset in Fig. 1. A small posi-

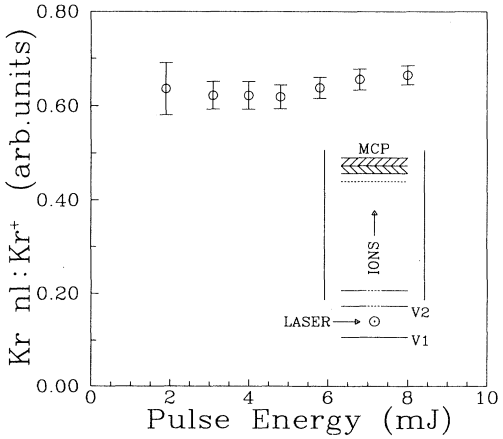

FIG. 1. Ratio of Kr Rydberg states observed to Kr^+ ions as a function of laser pulse energy for a fixed temporal pulse width. The actual ratio has a constant value < 1% Inset: Schematic of the laser-atom interaction region.

tive voltage ($V_2 \sim 100$ V) applied to the upper field plate after the laser pulse directs the photoions to the lower conducting plate. After the ions have been removed from the interaction region, a large positive voltage pulse V_1 is applied to the lower field plate. The field produced by this voltage is capable of ionizing any neutral Rydberg state with $n > 14$. Ions created by field ionization are pushed by the same field toward a dual microchannel plate detector. Therefore, we detect only ions formed by the field ionization of neutral atoms which are left in highly excited states after the laser pulse. We can, however, detect all of the ions created by simply setting the clearing voltage $V_2 = 0$. Then all ions in the interaction region are pushed toward the detector by the voltage V_1. Different charge states are distinguished by their flight times.

Our first measurement allows us to determine which Rydberg states remain after the laser pulse. By varying the amplitude of the field-ionization pulse, we selectively ionize only those states with principle quantum numbers above some critical value. Using this technique we have observed that the laser pulse creates a uniform distribution of states with $50 > n > 14$.

Figure 1 shows the ratio of Rydberg states to singly ionized Krypton detected as a function of laser pulse energy, for constant pulse width ($\tau_L \sim 100$ fs). The ratio has a constant value of less than 1%. At these intensities, the Kr^+ signal is actually saturated, so it is proportional to the focal volume as the pulse energy increases. Clearly, the number of field-ionized Rydberg states is proportional to the number of Kr^+ ions over this intensity range, indicating that the Rydberg excitation cross section is saturated as well.

Figure 2 shows the same ratio shown in Fig. 1, but in this case the laser intensity is varied by changing the laser pulse duration at a constant pulse fluence. The temporal

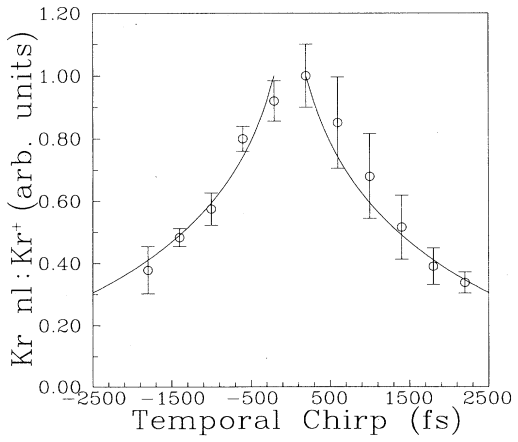

FIG. 2. Same ratio as in Fig. 1 plotted as a function of temporal chirp in the laser pulse. The chirp was created by introducing positive (+ sign) or negative (− sign) group velocity dispersion in the laser pulse using a diffraction grating. By comparing this figure with Fig. 1 it is clear that the decrease in the ratio for a broad temporal pulse is due to the length of the laser pulse and not to a decrease in the laser intensity. The solid curve is the result of a simple model as discussed in the text.

pulse width is increased by introducing a positive or negative frequency chirp in the laser pulse by adjusting the grating spacing in the pulse compressor. The ratio of the Rydberg signal to the Kr^+ ion signal is almost identical for both positively and negatively dispersed pulses, so the sense of the frequency chirp has no apparent effect on the results. There is, however, a dramatic pulse width dependence: Far more Rydberg states are present when the pulse is the shortest.

Combining the results of Figs. 1 and 2, we conclude that the ratio of Rydberg states to Kr^+ ions has no intensity dependence but has a large pulse duration dependence. This is a signature of a coherent nonstationary superposition state in the atom. We will now show that the decrease in trapped population with increasing pulse width is in quantitative agreement with the formation of a spatially localized radial wave packet [10].

The solid curve shown in Fig. 2 is the result of a calculation based on a simple model where the ionization rate is independent of pulse fluence due to saturation, but is proportional to the amount of time that the Rydberg wave packet spends near the ion core during its classical orbit. The probability P for photoionization of a given excited state with principle quantum number n is [11]

$$P = \begin{cases} \tau_L/\tau_k , & \tau_L < \tau_k \\ 1 , & \tau_L > \tau_K \end{cases}$$

where $\tau_k = 2\pi n^3$ is the classical period of the Rydberg orbit in atomic units, and τ_L is related to the laser pulse duration, also in atomic units. We can detect any Rydberg state whose energy is high enough so that it can be ionized by the high voltage pulse but low enough so that it is not ionized by the small clearing field pulse. Therefore, the number of states that are field ionized following the laser pulse is

$$N = N_0 \sum_{n_{\min}}^{n_{\max}} (1 - \tau_L/\tau_k) . \quad (1)$$

The sum extends over all states $n > (\tau_L/2\pi)^{1/3}$ which can be ionized by the high voltage pulse but not by the clearing pulse. We measure a uniform distribution of excited Rydberg states, so N_0 is a constant. The constant τ_L in Eq. (1) is the time interval during which the laser pulse has sufficient amplitude to saturate the production of Rydberg states. The value of τ_L used in Fig. 2 is 1.7 times the full width at half maximum (FWHM) of the temporal pulse profile. This is longer than the FWHM, due to (1) the relatively low threshold intensity for the Stark-shifted nine-photon resonance with Rydberg states($\approx 1 \times 10^{13}$ W/cm^2) and (2) saturation of the Rydberg state excitation cross section relatively early in the temporal pulse profile.

The data in Fig. 2 were acquired with a clearing field of 60 V/cm and an ionization field with a peak amplitude of 8 kV/cm. Equation (1) predicts that increasing V_2 to 400 V/cm should decrease N by approximately a factor of 2. Experimentally, we observe only a 25% decrease in the signal. Evidently, our model is only qualitatively correct. Equation (1) also predicts that with larger clearing fields

the number of Rydberg states should decrease faster with increasing pulse width. This is because the average Kepler period of the Rydberg states that survives the clearing pulse decreases for larger clearing fields. Hence, each term in Eq. (1) is more sensitive to increases in the laser pulse duration, τ_L. Again, our observations are in qualitative but not quantitative agreement.

Trapped Rydberg population is also observed in Xe, but the ratio of Rydberg states to Xe^+ ions decreases slowly with increasing laser intensity. Again, we have varied the laser intensity using two methods. We find that the ratio of Rydberg states to Xe^+ ions is more sensitive to a decrease in the laser pulse energy at a constant pulse duration than to an increase in the laser pulse duration at constant pulse energy. Our measurements at constant pulse duration show the intensity dependence of Rydberg excitation. This can be used to remove the intensity dependence from the measurements at constant pulse energy. When this rescaling is performed, the production of Xe Rydberg states shows a pulse duration dependence very similar to that seen in Kr. The different intensity dependence of the Rydberg state production in Xe vs Kr is not currently understood, but could be a simple difference in the saturation intensity for multiphoton excitation in the two atoms.

When circular polarization is used we observe no Rydberg population following the intense laser pulse. This result is in perfect agreement with previous ATI experiments, where no evidence for intermediate resonances was observed when atoms were ionized with circular polarizaton [3]. The angular momentum selection rules that exist for circular polarization forbid the excitation of any bound states which have non-negligible spatial overlap with the ground state. Therefore, no Rydberg states are excited during the laser pulse.

At the laser intensities used in these experiments, we observe many stages of ionization both in Kr and in Xe. We have detected the presence of ion Rydberg states in Kr and Xe which have energies up to 32 eV above the ionic ground state. Such levels can be populated either by direct multiphoton excitation via a Stark-shifted ion resonance or as a daughter product of the autoionization of a high-lying doubly excited state of the neutral atom. Rydberg ions are observed using a detection scheme that utilizes two electric-field regions [5]. The first field draws all ions created by the laser pulse into a second field region, which contains a strong stripping field of ~ 5 kV/cm. Ground-state ions experience only an increased acceleration, but ion Rydberg states ionize further, so that their charge state and acceleration rate increase. Thus, ground-state ions and Rydberg ions have different flight times to the MCP detector.

The Kr^+ Rydberg states we observe have principle quantum numbers $n > 20$. The number of highly excited ion states we detect is much less than 1% of the number of Kr^{2+} ions that are created. Although we expect a small population of ion Rydberg states, the number we observe is possibly less than the actual number that reaches our detector. The large amplitude of the Kr^{2+} peak can cause saturation of the channel plate detector and limit its efficiency for ions which have longer flight times. We have also observed Rydberg states of Xe^+ and Xe^{2+}. These highly excited Xe^{2+} states lie 32 eV above the Xe^{2+} ground state and their production requires the absorption of 20 photons by the Xe^{2+} ion [4]. We believe that Rydberg states of higher ion stages are also created by the laser pulse, but unfortunately, the large background of ground-state ions reduces our detection efficiency for these states to an unsatisfactory level. Also, our present apparatus limits the static electric fields, which we can produce in the second field region, to ~ 6 kV/cm. Therefore, the number of ion Rydberg states that we can detect drops sharply with the ion charge, Z, due to the Z^3 dependence of the field-ionization thresholds.

Our Xe results should be compared with those of another recent experiment [6]. de Boer and Muller have observed that up to 50% of low-lying excited states ($n < 8$) created during ATI of Xe by a 100-fs laser pulse remain trapped. As in the current experiment, population was transferred to excited states via ac Stark-shifted resonances during the laser pulse. Unlike the experiment described here, the intensities used by de Boer and Muller were insufficient to saturate the single-photon ionization cross section for the excited states. Hence, a very large (10–50 %) population remained in the excited states after the pulse.

The trapping observed in this experiment is not a steady-state stabilization, such as proposed by Gavrila and co-workers [12]. Both the intensity and the laser frequency are far too low to approach the regime of validity for these theories in low-l, low-m Rydberg states.

In summary, we find that neutral and ionic Rydberg states of Kr and Xe are populated during the interaction of ground-state atoms with intense 100-fs, 770-nm laser pulses. These results are consistent with the production of spatially localized Rydberg wave packets by the laser pulse, and with previous suggestions that ATI is dominated by Rydberg intermediate resonances [3]. Rydberg states of ions that are bound by only a fraction of the photon energy have been detected, thereby demonstrating the stability of these states at laser intensities capable of producing Xe^{3+} (i.e., capable of efficiently driving a nonresonant 20 photon process) [4].

Note added. In subsequent work, de Boer *et al.* have repeated their experiment under conditions more closely resembling ours [13]. They confirm our observation of $\sim 1\%$ trapped population in high-n states.

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