01 May 1993

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(Received 9 February 1993)

When exposed to intense light of ~580 nm, the ground state of K shifts up in energy, passing through two-photon resonances with Rydberg states and finally crossing the two-photon ionization limit. Using laser pulses of 0.5 to 13 ps duration, we have shown experimentally that ionization occurs for short pulses, but for long pulses the population is diverted into the Rydberg states where some population survives the peak intensity of the pulse, in excellent agreement with a dynamic Floquet model.

PACS numbers: 32.80.Rm, 32.80.Wr

The energy shift of atomic levels in an intense laser field plays an important role in multiphoton ionization. For example, the ponderomotive shift, $e^2E^2/4m\omega^2$, of the ionization limit to higher energies means that more photons must be absorbed to effect ionization [1,2]. This shift can be directly observed as the suppression of the lowest energy peaks in the above-threshold-ionization (ATI) electron energy spectrum, and ponderomotive shifts several times the photon energy are common. Perhaps more important, the energies of excited states also shift in intense fields, often by an amount equal to the ponderomotive shift, so that these states are brought into multiphoton resonance with the ground state during the pulse [3]. Freeman *et al.* have demonstrated that virtually all multiphoton ionization by linearly polarized light passes through these resonant intermediate states [4]. While the importance of this process is clear, the details of how it occurs are apparently not. Freeman and co-workers [4–6] have proposed that the excitation to the resonant state and the ensuing photoionization both occur at the temporal peak of the pulse, while de Boer and Muller [7] suggest that the resonant excitation can occur on the rising edge of the pulse with ionization occurring later.

Here we report an experimental investigation of multiphoton excitation and ionization of K atoms by ps pulses as well as a Floquet analysis of the process. Together they show that the two above mentioned scenarios are limiting cases of the possible evolution of the Floquet (or dressed) atomic states during the laser pulse. More precisely, we show that both scenarios may be understood in terms of the diabatic or adiabatic evolution during the laser pulse [8–11].

Specifically, we have investigated the excitation and ionization of K atoms by 0.5 to 13 ps pulses. As shown by Fig. 1 the laser is tuned so that the ground state energy plus two photons lies just below the ionization limit in zero field [12]. The laser frequency is ~4000 cm^{-1} above the $4s$-$4p$ resonance line, so the $4s$ ground state shifts up in energy, quadratically in the field. In Fig. 1 we show the shifts of the levels in a pulse of 1 ps duration. In this experiment the ac Stark shift of the $4s$ level, which can be as great as 2000 cm^{-1}, is 1.4 times the ponderomotive shift. As shown by Fig. 1, the $4s$ level plus two photons can be shifted above the ionization limit during the laser pulse. Consequently, as the laser field rises, the lowest order ionization process changes from a three-photon to a two-photon process. Although this situation would seem to ensure virtually complete ionization, we shall see that the fact that the ac Stark shift brings the ground state into multiphoton resonance with the Rydberg states on the rising edge of the pulse can lead to trapping of the population in the Rydberg states and the inhibition of ionization. A roughly analogous experiment which leads to the population of lower lying excited states has recently been carried out by Vrijen *et al.* [13].

The most straightforward way of understanding how the population can be trapped in the Rydberg states is to use a Floquet approach. The essential idea of the Floquet

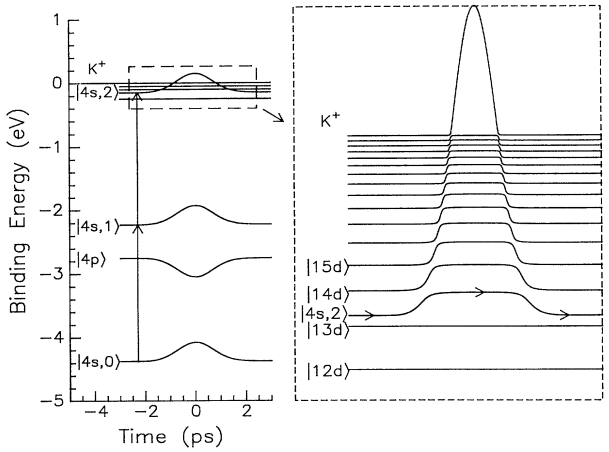

FIG. 1. The relevant energy levels of K are shown along with the two-photon interaction. The states $|4s,1\rangle$ and $|4s,2\rangle$ represent the dressed ground state with one and two photons, respectively. During the laser pulse, interaction between the $|4s,1\rangle$ and $|4p\rangle$ states causes an upward shift of the ground state. An expanded view of the $|4s,2\rangle$ state shows that as the ground state energy increases, the $|4s,2\rangle$ state passes through a series of anticrossings due to interaction with the *nd* Rydberg series. For a sufficiently large shift, the $|4s,2\rangle$ state crosses into the continuum so that two photons can ionize the ground state.

approach is that the energy of any state can be shifted by an integer times the photon energy, so that all states lie in an energy band spanning $h\nu$. With electric dipole coupling, even-parity states which have $Nh\nu$ added to their energies are coupled to odd-parity states with $(N \pm 1)h\nu$ added to their energies. For a fixed laser frequency we calculate the adiabatic Floquet energies at each laser field amplitude and from these construct the Floquet energy level diagram as shown in the box in Fig. 1. For simplicity, we shall refer to the states by their diabatic labels. With this convention, only the $4s$ Floquet state has an ac Stark shift. With the enlarged scale in the box of Fig. 1, we can also see that there are avoided crossings between the $4s$ state and the nd Rydberg states due to the two-photon $4s$-nd couplings, V_n, which lift the degeneracy of the $4s$ and nd Floquet states. The two-photon coupling matrix element is given, to a good approximation, by

$$V_n = \frac{\langle nd|\mu|4p\rangle\langle 4p|\mu|4s\rangle}{W_{4p} - W_{4s} - \hbar\omega} E^2, \tag{1}$$

where $\langle nd|\mu|4p\rangle$ and $\langle 4p|\mu|4s\rangle$ are electric dipole matrix elements, E is the laser field amplitude, and the denominator is the detuning of the laser from the resonance line. The only variation of Eq. (1) with n is due to the $\langle nd|\mu|4p\rangle$ matrix element, which scales as $n^{-3/2}$. Thus the avoided crossings decrease with n less rapidly than the n^{-3} level spacing, a potential complication which we here ignore. When the $4s$ Floquet state is shifted above the limit, it is broadened by the two-photon $4s \rightarrow \epsilon d$ coupling. However, we do not show the broadening explicitly in Fig. 1. All the states shown in Fig. 1 are of course degenerate with the ϵf continuum minus one photon and exhibit a small broadening due to the resulting coupling.

Armed with the Floquet energy level diagram of Fig. 1 we can now ask what happens during the laser pulse. Initially, the atoms are all in the $4s$ Floquet state. As the laser field rises, the $4s$ Floquet energy shifts up in energy and immediately encounters the avoided crossing with the $14d$ state. Whether the atoms traverse the crossing diabatically or adiabatically depends upon how quickly the field rises. Specifically, the probability of the atoms traversing the avoided crossing with the nd state diabatically and remaining in the $4s$ Floquet state is given by [14]

$$P_n = \exp\left[-\frac{2\pi V_n^2}{dW/dt}\right], \tag{2}$$

where W is the difference in energy between the $4s$ and nd Floquet energies. A very slow traversal leads to purely adiabatic evolution into the Rydberg state, as shown by the solid arrow. A fast traversal leads to diabatic evolution, and the atoms remain in the $4s$ state. With a laser pulse of sufficient intensity to shift the ground state above the ionization limit, the $4s$ Floquet state can pass through anticrossings with all Rydberg states lying above its zero-field location. The population left in the $4s$ Floquet state after passing through all of these anticrossings is given by

$$P_g = \prod_{n=i}^{\infty} p_n \tag{3}$$

and these atoms can be ionized by absorbing two photons.

The net effect of the shift through the Rydberg states is that some or all of the atomic population is transferred to the Rydberg states on the rising edge of the laser pulse. As a result of the small cross section for photoionization of the Rydberg states, population which is transferred into these states is likely to survive the peak intensity of the laser pulse without being ionized [15–18], whereas virtually all population left in the ground state will be photoionized when the intensity passes above the two-photon threshold. On the falling edge of the laser pulse, atoms which have been transferred to the nd Rydberg states, as shown by the solid arrow of Fig. 1, pass through the $4s$-nd avoided crossing again. An adiabatic traversal, shown by the solid line, takes them back to the ground state while a diabatic traversal leaves them in the Rydberg state after the pulse. For a temporally symmetric laser pulse, the crossings on the rise and fall must be traversed partially adiabatically to leave population in the Rydberg states after the pulse.

The experiment was performed by exciting K atoms in an effusive atomic beam with a focused ps dye laser beam. We used a Coherent 700 mode locked tunable dye laser pumped by an Antares mode locked Nd:YAG laser. A regenerative Nd:YAG amplifier was used to pump a three-stage dye amplifier which produced 1 mJ pulses. The length of the dye laser pulse could be varied from 420 fs to 13 ps. The dye laser beam was circularly polarized and focused to a waist of approximately 30 μm diameter at the intersection with the atomic beam, producing a peak intensity of up to 6×10^{13} W/cm^2. Using a time-of-flight electron energy analyzer we could discriminate between two- and three-photon ionization of the ground state. Alternatively, the population left in the Rydberg states was detected using field ionization plates. In the latter case a ramped voltage with a 1 μs rise time was applied to the plates 100 ns after the laser pulse, producing a time resolved field ionization signal from individual Rydberg states. The electrons produced by both photoionization and field ionization were detected with microchannel plate detectors.

In Figs. 2(a), 2(b), and 2(c) we show the low energy electron yields from two-photon ionization as a function of laser wavelength for pulse durations of 4.5, 1.0, and 0.42 ps, respectively. These signals represent ionization of atoms which did not make a transition to a Rydberg state, but are ionized by two photons from the ground state when it passes above the two-photon limit at the peak of the pulse. In Figs. 2(a)–2(c), the ionization signal shows a rapid decrease when the laser is tuned below the two-photon ionization limit at 17505 cm^{-1} and is constant above it. Note that ionization is observed farthest below the limit for the shortest pulse. In this

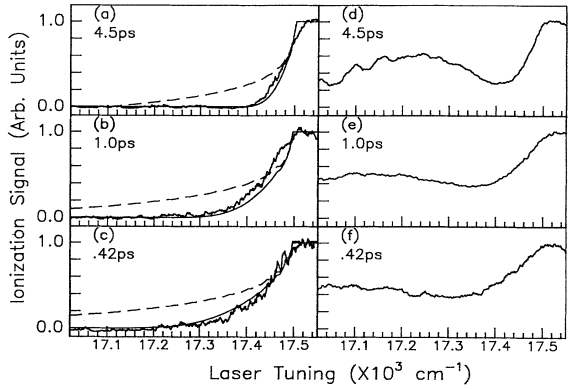

FIG. 2. The photoionization signal for 4.5, 1.0, and 0.42 ps pulse lengths vs frequency tuning of the laser, 17505 cm^{-1} being the two-photon ionization limit. (a), (b), and (c) are the two-photon ionization signals. The rough curves are the data, the smooth solid curves are the calculated spectra which include transitions to the Rydberg states, and the dashed curves are a calculation which neglects the Rydberg state interaction. (d), (e), and (f) show data of the total ionization signal including the photoionization of the Rydberg states. The maximum shift at the peak of the pulse for the three pulse times is 800, 1600, and 2000 cm^{-1} for the 4.5, 1.0, and 0.42 ps pulses, respectively.

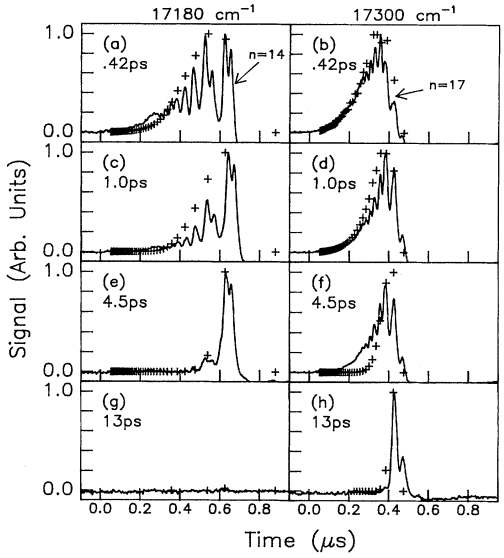

FIG. 3. The residual Rydberg state population detected by field ionization is shown for pulse widths of 0.42, 1.0, 4.5, and 13 ps, with laser frequencies of 17180 and 17300 cm^{-1}. The x axis shows the time at which the atoms were ionized by the electric field which was ramped on after the laser pulse. Also shown is the calculated spectra (+). For the 17180 cm^{-1} tuning in (a), (c), (e), and (g), the zero-field position of the $|4s,2\rangle$ Floquet state is just below the $14d$ state; for the 17300 cm^{-1} tuning in (b), (d), (f), and (h), the $|4s,2\rangle$ state is just below the $17d$ state. The doublet structure is due to different m_l values which ionize at slightly different fields.

case, atoms in the $4s$ Floquet state are more likely to pass diabatically through the avoided crossings with the Rydberg states and be ionized by two photons.

With a model based on Eqs. (1)–(3), it is straightforward to calculate numerically the evolution of the atomic system during the laser pulse. Assuming that the atoms are all initially in the $4s$ Floquet state we have calculated the two-photon ionization spectra for the three pulse lengths of Figs. 2(a)–2(c). The results, which include spatial averaging, are shown by the solid lines of Figs. 2(a)–2(c). The only adjustable parameter in the calculation is the overall normalization. As can be seen in Fig. 2, the signal agrees well with the calculation. We have also done the calculation assuming a purely diabatic traversal of the Rydberg states (dashed line), ignoring the possibility of population transfer to the Rydberg states. This assumption leads to the prediction of a larger two-photon photoionization yield, which does not agree with the data. The effect is more pronounced for longer pulse times where the passage through the Rydberg anticrossings is slower so that more population is transferred to the Rydberg states. Figures 2(d), 2(e), and 2(f) show the total photoionization signal, i.e., the sum of two-photon ionization and three-photon ionization via the Rydberg states as a function of laser wavelength for 4.5, 1.0, and 0.42 ps pulse lengths. The pronounced minimum in Fig. 2(d) is due to population being trapped in high Rydberg states which have a low rate of photoionization (scaling as n^{-3}). For the shorter pulses the minimum is less pronounced but still present.

Figure 3 shows the Rydberg state population after the laser pulse for four different pulse times and two laser frequencies, 17180 and 17300 cm^{-1}. The horizontal axis shows the time at which the atoms were field ionized by the ramped voltage so that increasing time corresponds to decreasing values of n. The doublet structure seen at lower n is due to two different m values which ionize at slightly different fields. Although the laser was circularly polarized to produce $l=2$, $m=2$ Rydberg states, the ionizing electric field was orthogonal to the laser propagation direction, which created a superposition of m states. The + symbols represent the calculated Rydberg spectra with the only adjustable parameter in the calculation being the overall normalization. As can be seen in Fig. 3, the 0.42 ps pulse populated a large range of Rydberg states. This large range is due to the relatively fast passage through the avoided crossings on the rising edge of the pulse, resulting in a small transfer of population to any one state. Large population transfer to any given state is only possible near the temporal peak of the pulse where the atom traverses the avoided crossings sufficiently slowly. The major contribution to the population of any given state therefore occurs at specific spatial locations in the profile of the laser beam [6,7]. Even though the transition probabilities to individual Rydberg states during the rising edge of the laser pulse are small, the accumulated effect of the entire Rydberg series produces a

dramatic decrease in ground state population by the time it reaches the limit.

As the pulse time increases, the final Rydberg state population becomes more concentrated in the lower n states. As the laser pulse is made longer, the atoms traverse the first few avoided crossings more adiabatically, depleting the $4s$ population and leaving no atoms to populate higher states or cross the two-photon ionization limit. The depletion of the $4s$ state can be easily seen in Fig. 3(e), where population is seen only in the first three states, even though there was sufficient intensity to shift the ground state above the two-photon ionization limit. In this case all the atoms make transitions to the Rydberg states early in the pulse. The ionization of the Rydberg states can then occur at any subsequent time during the pulse. The longest pulse, 13 ps, shows the fully adiabatic passage through the anticrossing with the $14d$ state in Fig. 3(g). In this figure, no population is observed in any Rydberg state. The lack of signal in Fig. 3(g) is not due to low peak intensity; the intensity is sufficient to shift the ground state through three Rydberg state resonances. The lack of population is due to the almost completely adiabatic transitions both from the $4s$ state to the $14d$ state on the rising edge of the pulse and from the $14d$ state back to the $4s$ state on the falling edge of the pulse, as shown by the arrow in Fig. 1. These atoms were in the $14d$ state at the peak of the pulse, but as was seen in Fig. 2, the Rydberg states are less likely to be photoionized than the ground state. If the coupling to the Rydberg states is reduced, as is the case for the high tuning in Fig. 3(h), the passage becomes partially diabatic so that population is again seen in the Rydberg states.

These experiments and those of Vrijen *et al.* [13], demonstrate that multiphoton excitation and ionization by ps laser pulses can be well understood in terms of the Floquet picture presented here. For very short pulse lengths, all avoided crossings except those traversed at the peak of the pulse are traversed diabatically. For very long pulse lengths, the first encountered avoided crossing is traversed adiabatically, and much of the population is returned to the ground state. Between these two extremes population is left in the excited states.

This work was supported by the National Science Foundation.

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