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Spatially Resolved Transitions to Autoionizing States

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We have observed transitions to autoionizing states in Mg using two short optical pulses. Mg atoms are initially prepared in a high lying Rydberg wave packet with the first ps laser. A second ps laser is then used to excite the inner electron, producing an autoionizing state. The dependence of the transition probability on the delay between the two lasers shows that when the second laser is tuned away from the ionic resonance, the inner electron can make a transition only when the Rydberg wave packet is near the core.

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The isolated core excitation (ICE) [1] method has been used to gather a wealth of spectral information about doubly excited autoionizing states of alkaline earth atoms [2]. The essence of the ICE method is easily understood using Mg as an example [3]. Atoms are first excited to a bound Rydberg state, e.g., $3snd$ in Mg, then optically excited to the autoionizing $3pnd$ state. The $3snd \rightarrow 3pnd$ transition is essentially the Mg^+ $3s \rightarrow 3p$ transition with the outer nd electron remaining a spectator. The absorption has a maximum near the frequency of the Mg^+ $3s \rightarrow 3p$ resonance line and, to a first approximation, the position and width of the maximum give the energy and spectral width of the autoionizing $3pnd$ state.

The above isolated resonance description accurately describes the spectrum for small detunings from the ionic line. It is not, however, useful for larger detunings. When the light is detuned from the ionic transition, the binding energy of the Rydberg electron must be altered by the amount of the detuning to conserve energy. When the detuning exceeds the energy interval ΔW_n between the Rydberg states, shakeup satellites corresponding to the excitation of the adjacent autoionizing Rydberg states appear clearly in the spectrum [4,5]. Applying quantum defect theory (QDT) to the above notion of an ionic transition with a spectator electron has been successful in reproducing the entire ICE spectrum, including the shakeup satellites.

A point which is not transparent in the QDT description is that the Rydberg electron is located in different places for on and off resonant absorption. In on resonant absorption the Rydberg electron need not be present near the Mg^+ ionic core when the photon is absorbed. In fact, it is most likely to be near its outer turning point since that is where the electron spends most of its time during its orbit. In contrast, in off resonant absorption the photon's energy is shared between the ion and the Rydberg electron, requiring that they be close together when the photon absorption occurs.

Here we report the results of an experimental test of the above notions using short pulse ICE of a bound wave packet. The utility of using short pulse ICE to probe the dynamics of autoionization was first suggested by Wang and Cooke [6]. They later demonstrated that with short pulse ICE it was possible to make a notch in a bound Rydberg state wave function, creating an antiwave packet [7]. A related method of reproducing antiwave packets has been reported by Noordam *et al.* [8], and short pulse ionization of the core but not the Rydberg electron has been reported by Jones and Bucksbaum [9] and Stapelfeldt *et al.* [10]. Our experiment is done by using a ps laser to produce a localized radial wave packet of bound Mg $3snd$ states so that the nd electron oscillates between the core and its outer turning point [11–14]. With a second synchronized ps laser tuned on or off the Mg^+ $3s \rightarrow 3p$ transition, we can drive the $3snd \rightarrow 3pnd$ transition when the Rydberg electron is either near to or far from the ion core. The experiments show that on resonant absorption occurs whether the Rydberg electron is far from or near to the core, but that off resonant absorption occurs only when the Rydberg electron is near the core. We show that the observed dependence on the spatial position of the Rydberg electron can be derived from a generalization of the QDT used to describe the ICE shakeup spectrum. Our theoretical approach is similar to the one developed by Wang and Cooke to describe short pulse ICE [6,7,15]. A more general QDT description of wave packets in two electron systems has been given by Henle, Ritsch, and Zoller [16].

In the experiment an effusive Mg beam from a heated oven passed between a pair of plates 10 cm from the oven. Between the plates atoms were excited by two synchronized uv ps laser pulses as shown in Fig. 1. The first pulse, at 325 nm, drove the $3s^2 \rightarrow 3snd$ two photon transition creating a radial wave packet of $3snd$ states. The second laser pulse, at 280 nm, drove the $3snd \rightarrow 3p_{3/2}nd$ transition after a variable delay. Following the laser pulses, the ions from the decay of autoionizing $3pnd$ states were detected by applying a 60 V/cm field pulse to the plates to drive the Mg^+ ions to a microchannel plate detector.

The synchronized ps uv pulses were produced as follows. A frequency doubled Coherent Antares mode locked Nd:YAG laser was used to pump two Coherent 700 mode locked tunable dye lasers. The first dye laser, tuned to 650 nm, had a temporal width of 3.2 ps (FWHM), and the second, tuned to 560 nm, had a tem-

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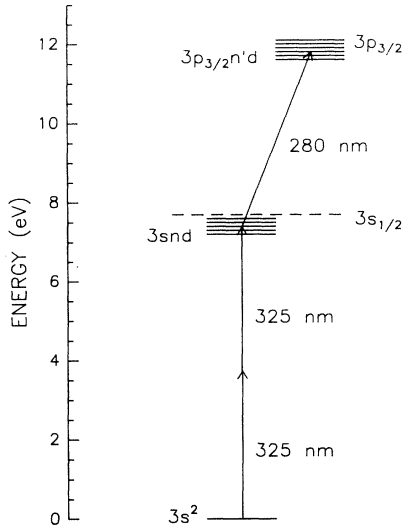

FIG. 1. The excitation path used in the experiment is shown. Two photons of doubled 650 nm light excited Mg to a $3snd\ ^1D_2$ Rydberg wave packet state, where n is the central state of the wave packet. One photon of the doubled 560 nm light then excited the $3s$ core electron, producing $3p_{3/2}n'd$ autoionizing states, where n' is the central state of the final Rydberg wave packet.

poral width of 5.2 ps. Because the two dye lasers were pumped by the same source, the timing jitter between the two pulses was less than 2 ps. The two dye laser beams were amplified in three stage dye amplifiers pumped by a Nd:YAG regenerative amplifier and then frequency doubled using KDP crystals. The temporal width of the two laser pulses after doubling was 3.3 and 4.7 ps. The delay between the two laser pulses was adjusted using a delay line in the 560 nm laser beam path which could be scanned continuously. The uv beams were focused when they crossed the Mg atomic beam inside the vacuum chamber. Data were taken by fixing the detuning of the second laser from the ion resonance and scanning the delay between the two lasers. Positive delay corresponds to the second laser pulse coming after the first.

Figure 2 shows typical data obtained with this method. For these data the first laser was tuned to excite a wave packet centered at $n_0=54$ with a classical orbital period of 24 ps. The second laser was tuned approximately 15 cm^{-1} above the Mg^+ $3s \rightarrow 3p_{3/2}$ transition. The large initial peak in the data is due to enhanced multiphoton ionization of the ground state of Mg when the two lasers are temporarily overlapped. The width of the initial peak is due mainly to the temporal width of the two lasers, where the effective width including the 2 ps timing jitter is given by $\Gamma_{\text{eff}}=2+(\Gamma_1^2+\Gamma_2^2)^{1/2}$ ps $=7.7$ ps. There are two additional peaks, at 25 and 50 ps, which correspond to the first and second returns of the Rydberg wave packet to the core. The third peak, the second return, is broadened by dispersion of the wave packet. Beyond the

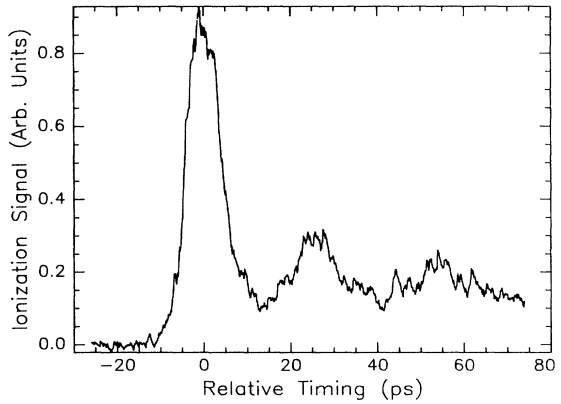

FIG. 2. A time resolved excitation spectrum is shown where the initial Rydberg state is centered around $n=54$ and the core laser is tuned approximately 15 cm^{-1} to the blue of the ionic resonance. The time axis represents the relative timing between the Rydberg and core laser. Peaks are seen in the excitation spectrum when the relative timing is equal to an integral number of classical orbital periods of the central Rydberg state of the wave packet. For $n=54$ the classical orbital period is 24 ps.

second return, the wave packet is sufficiently dispersed so that no additional structure can be observed. These data clearly show that the Rydberg electron must be near the core in order for the core electron to make an off resonant transition.

Data were taken with the first laser tuned to different n states and, as expected, the signal oscillated with the orbital period of $2\pi n_0^3$. Data were also taken for different detunings of the second laser from the ionic transition, and examples are shown in Fig. 3 along with calculated spectra. The laser detunings from resonance are accurate

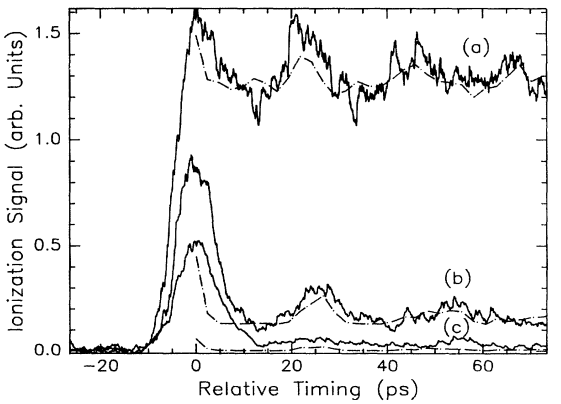

FIG. 3. Measured and calculated spectra are shown for several tunings of the core laser with the Rydberg laser tuned to $n=54$. Near the ionic resonance, (a), a large baseline signal results from the resonant excitation of the core electron which is largely independent of the position of the Rydberg electron. Away from the ionic resonance (b) and (c), the shakeup excitation exhibits a strong dependence on the Rydberg electron position.

to 10 cm^{-1}. In Fig. 3(a) we show the ionization signal for the core laser tuned to the $3s \rightarrow 3p_{3/2}$ ion resonance. As shown, the signal goes from zero for negative delay to a plateau for positive delay, and there are 20% oscillations due to the first and second returns of the Rydberg electron to the core at the classical orbital period. In other words the atom has a high probability of making the $3snd \rightarrow 3p_{3/2}nd$ transition irrespective of the spatial position of the Rydberg electron. As the detuning is increased to 15 cm^{-1}, in Fig. 3(b), the signal for positive delays exhibits the same structure as shown in Fig. 2: a large peak at zero delay followed by two peaks due to the first and second returns of the wave packet to the core. Figure 3(c) shows data taken with a detuning of 30 cm^{-1}. Except for the multiphoton peak at zero delay, the signal level has dropped considerably. However, the increases due to the first and second return of the wave packet are still discernable. These experimental data indicate that the Rydberg electron must be near the core for off resonant excitation of the ionic transition.

To show the connection between the time resolved spectra of Fig. 3 and the theory used to describe time independent ICE spectra, we briefly describe the generation of the synthetic spectra of Fig. 3. A bound Rydberg $3snd$ state has the energy $W_n = -\frac{1}{2}n^{*2}$ relative to the Mg^+ $3s$ state, implicitly defining the effective quantum number n^*. Similarly, at an energy $W_\nu = -\frac{1}{2}\nu^2$ relative to the Mg^+ $3p$ state, the $3pnd$ wave function has an effective quantum number ν. Unlike n^*, ν is continuous since the autoionizing $3pnd$ states are coupled to the $3s\ \varepsilon f$ continua. The ICE transition matrix element is easily given in terms of the effective quantum numbers n^* and ν as [17]

$$T_n(\nu) = A(\nu)\mu\left[\frac{\sin[\pi(n^*-\nu)]}{\pi(\frac{1}{2}n^{*2}-\frac{1}{2}\nu^2)}\right]. \quad (1)$$

In Eq. (1), μ is the constant ionic dipole matrix element, and $A^2(\nu)$ is the density of the final autoionizing states. $A^2(\nu)$ is peaked at the locations of the Mg $3pnd$ states, at $\nu = n'$, the widths of the peaks giving the autoionization rates. The bracketed term of Eq. (1) represents the overlap of the bound nd wave function with the autoionizing νd wave function. Squaring Eq. (1) gives the frequency dependence of the ICE spectrum. For $|n^*-\nu| < 1$, the bracketed term is constant and the spectrum is determined entirely by $A^2(\nu)$, which depends on the energies and widths of the nd autoionizing states. In our experiment the bound intermediate state is not a stationary state but a wave packet with a wave function given by

$$\psi_I(t) = \sum C_n \psi_n e^{-it/2n^{*2}}, \quad (2)$$

where C_n is the amplitude of the nth state which is determined by the tuning and spectral width of the first laser. If the central frequency of the first laser excites the state with principal quantum number n_0 and the bandwidth of the laser is Γ_1, C_n is given by

$$C_n = \exp[-(W_{n_0}-W_n)^2/\Gamma_1^2]. \quad (3)$$

To describe the excitation by the two short pulses, we combine the Rydberg wave packet of Eq. (2) with the transition matrix element of Eq. (1) for each final state energy or, equivalently, each value of ν. The transition probability to the final state energy W_ν from the ground state is given by

$$T_g(\nu) = \left|\sum C_n C_{n\nu} T_n(\nu) e^{-it/2n^{*2}}\right|, \quad (4)$$

where $C_{n\nu}$ reflects the number of bound states coupled to the final state energy W_ν due to the bandwidth of the second laser. For a Gaussian pulse, $C_{n\nu}$ is given by

$$C_{n\nu} = \exp[(W_\nu - W_n - \omega)^2/\Gamma_2^2], \quad (5)$$

where ω is the second photon's energy and Γ_2 is the bandwidth of the second laser pulse. Squaring Eq. (3) gives the ionization signal for any value of final state energy W_ν, and integrating over ν we obtain an expression proportional to the observed ionization signal S. Explicitly,

$$S = \int |T_g(\nu)|^2 d\nu. \quad (6)$$

The calculated curves shown by the dashed lines in Fig. 3 have been obtained using Eq. (6).

The one parameter which was adjusted to fit the data was the detuning from the ion resonance. In the calculations, the size of the resonant plateau for positive delays was rather sensitive to the detuning of the core laser from the ionic resonance. Since the tuning of the core laser was known only to within 10 cm^{-1}, we adjusted the detuning to best fit the data. The fit detunings were 8, 13, and 25 cm^{-1}, in reasonable agreement with the measured values of 0, 15, and 30 cm^{-1}. With this single adjustable parameter, the theoretical spectra show excellent agreement with the data for positive delays. The calculation does not agree with the measurement at zero delay because it does not include multiphoton ionization of the ground state due to the temporal overlap of the lasers.

These data clearly show that the Rydberg electron must be near the core to excite the inner electron away from the ionic resonance, whereas for core excitation near the ionic resonance the position of the Rydberg electron is relatively unimportant and, in theory, should be far from the core. We note that the theory predicts that if the core laser is tuned on the ionic resonance there should be a decrease, not an increase, in the ionization signal at the returns of the wave packet to the core due to broadening of the ionic transition beyond the laser linewidth. We have not yet observed this phenomenon, but the good agreement between theory and experiment leads us to conclude that the QDT model is valid for excitation by short pulses.

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