Short-pulse laser-induced stabilization of autoionizing states

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Atoms in doubly excited states above the first ionization limit can decay via autoionization in which an electron is emitted leaving an ion, or by photoemission which leaves the atom in a singly excited state. In this paper, it is demonstrated that interaction between the atoms and a laser pulse that is short compared to the autoionization lifetime can lead to large enhancement of the photoemission process by stimulating the atoms to emit a photon. Since the resultant singly excited atoms do not autoionize, this process can be viewed as an enhancement of the stabilization of the doubly excited atoms against autoionization. A simple theoretical model is outlined that shows good agreement with the experimental results.

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I. INTRODUCTION

The use of the isolated core excitation [1] technique to study the complex spectra of many doubly excited states above the first ionization limit has been proven to be a valuable tool (a few examples can be found in Refs. [2–5]). In this method, one of the ground-state electrons is optically excited to a high-lying Rydberg state, thus isolating the Rydberg electron from the core. After some time, the second electron or ''core electron'' is optically excited, producing the doubly excited state. If the combined energies of both electrons is above the first ionization limit, then a collision between these two electrons can lead to an autoionization event. In this case, one electron is ejected and the atom is left in an ionic state. In addition to autoionization, doubly excited states above the first ionization limit can also decay through fluorescence; in this case the core electron emits a photon that brings the total energy of the atom below the first ionization limit, thus stabilizing the atom. The percentage of atoms that is stabilized can be calculated using the fluorescence branching ratio, which is simply the ratio of the fluorescence rate to the total decay rate out of the doubly excited states.

Recently, a few experiments have been performed in which the recombination between ejected electrons and ions through an autoionizing state (a process referred to as dielectronic recombination [6,7]) were probed in cases when the atom was subjected to a small static electric field [8,9]. The application of the static electric field induced angular momentum mixing for the high-lying Rydberg electron. As a result, the autoionization rate of the doubly excited states was decreased. On the other hand, the electric field had no effect on the photoemission rate that was determined by the fluorescence rate of the core electron. As a result, the fluorescence branching ratio was increased. In a more recent experiment, the effect of a combination of electric and magnetic fields on the fluorescence branching ratio was measured [10–12]. The application of the magnetic-field-induced mixing of the different azimuthal angular momentum components, which resulted in a further enhancement in the fluorescence branching ratio for the case when the magnetic field was perpendicular to the electric field.

In a previous work, we have shown that the application of a pulsed laser field to stimulate the core electron back to a singly excited stable configuration enhances the fluorescence branching ratio [13]. The presence of a pulsed laser field did not change the nature of the doubly excited states, as in the case of the previously described experiments, but simply increased the photoemission rate. In the previous experiment, we investigated the case in which the laser pulse was long compared to the autoionization lifetime but short relative to the fluorescence lifetime. In this time domain, the results could be accurately modeled using simple rate equations.

In the present experiment, it will be shown that the application of a short laser pulse (shorter than the autoionization lifetime) will greatly increase the stabilization of the doubly excited states.

II. THEORY

For an atom prepared in a doubly excited state above the first ionization limit, the fraction of atoms that are stabilized through fluorescence decay can be determined by calculating the fluorescence branching ratio given by

$$B(n)=\frac{R_{\mathrm{fl}}}{R_{\mathrm{fl}}+R_{\mathrm{ai}}(n)}, \tag{1}$$

where R_{fl} is the fluorescence rate, $R_{\mathrm{ai}}(n)$ is the autoionization rate, and n is the principle quantum number of the Rydberg electron. As an example, consider the $4p_j nd$ doubly excited states of calcium in a zero electric field, where j represents the total angular momentum of the ionic electron. The fluorescence rate is equal to the fluorescence rate of the $4p_j$ ionic state and is independent of n. The autoionization rate, on the other hand, scales as n^{-3}. In the presence of a static electric field sufficiently strong to form Stark states, the fluorescence rate is unchanged but the autoionization rate scales as n^{-4}. In both cases, the branching ratio approaches unity for large values of n.

The branching ratio defined in Eq. (1) is independent of the time scale over which the autoionizing states were produced. In the case when a laser pulse is used to stimulate the emission of a photon, the stabilization process is strongly

dependent on the temporal parameters of the exciting and stabilizing lasers [13]. To understand the effect of a short-pulse laser (short relative to the autoionization lifetime) on the enhancement of the fluorescence branching ratio, consider the case when a calcium atom in a high Rydberg state $4s20d$ is exposed to a short-pulse laser tuned to excite the core electron from the $4s$-$4p_{3/2}$. In calcium, the $4p_{3/2}20d$ doubly excited state is energy degenerate with a few $4p_{1/2}n'd$ states (n' ranging between 35 and 42) and several continuum states. In other words, the true eigenstate of the system in this energy range is a superposition of all these states. Since the excitation with the short pulse happens rapidly, the Rydberg electron does not have enough time to react to the changes that happened to the core electron [14,15]. So, immediately after the laser excitation, the atom is in a pure $4p_{3/2}20d$ state. This state is not an energy eigenstate of the atom but a nonstationary two-electron wave packet. A short time after the laser excitation, the interaction between the core electron and the Rydberg electron will spread the population among all the degenerate bound and unbound states. The population that ends up in the unbound states is lost via autoionization. But the population that goes into the bound $4p_{1/2}n'd$ states can be stabilized using a second short-pulse laser that is tuned to the $4p_{1/2}$-$4s$ ionic resonance, which stimulates the atom back to the singly excited energy configuration.

For a sufficiently short laser pulse, very few atoms in the $4p_{1/2}n'd$ states have enough time to autoionize during the stimulated emission process. As a result, the total number of atoms in the $4p_{1/2}n'd$ states that are stabilized can be written as

$$N_L = N_0 P + N_0(1-P)B(n'), \tag{2}$$

where N_0 is the initial number of atoms in the $4p_{1/2}n'd$ state, P is the percentage of atoms that are stimulated from the $4p_{1/2}n'd$ doubly excited states to the $4sn'd$ singly excited states using the second short-pulse laser, and $B(n')$ is the branching ratio defined in Eq. (1). If no laser was applied, $P=0$ and the percentage of atoms that are stabilized is just the fluorescence branching ratio defined in Eq. (1). The first term in Eq. (2) is the number of atoms that are stabilized using the short-pulse laser; this process happens on a time scale that is very short relative to any other process in the atom (fluorescence or autoionization). The number of atoms that are left in the $4p_{1/2}n'd$ states are now $N_0(1-P)$; the percentage of these atoms that will be stabilized through fluorescence is simply given by the fluorescence branching ratio defined in Eq. (1). Under saturation condition, in the case of high laser intensity, the possibility of redistribution of population among the n' states could lead to further complications in Eq. (2). In the present case, this possibility is not considered. Direct excitation and stimulated emissions by the Rydberg laser could also produce high n Rydberg electrons. However, this process is extremely weak compared to the core transitions and is neglected here. Thus the enhancement factor in the stabilization process due to the application of the second short pulse laser is given by

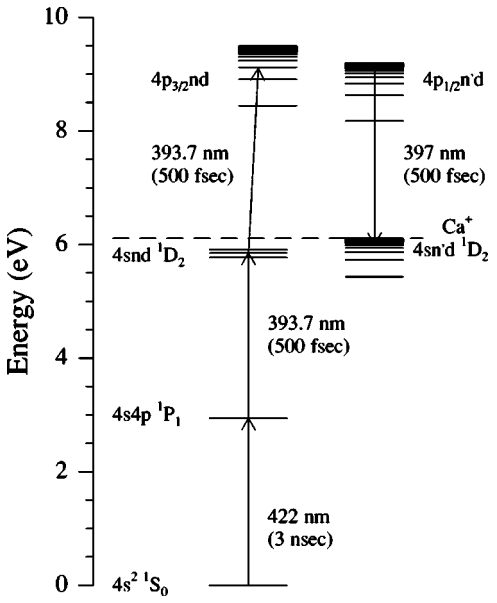

FIG. 1. Excitation diagram for calcium used in the experiment. The first nanosecond dye laser excites the $4s^2\ ^1S_0$ ground state to the $4s4p\ ^1P_1$. About 4 nsec later, a 500-fsec laser pulse promotes the atoms via a two-photon resonant transition to a pure $4p_{3/2}nd$ doubly excited state, where $n=20$, 21, and 22. A second 500-fsec laser short pulse was then used to drive the transition from the $4p_{1/2}n'd$ states to the $4sn'd$ states.

$$S(n') = \frac{P + B(n')(1-P)}{B(n')}. \tag{3}$$

The enhancement factor $S(n')$ is a function of the principle quantum number (n'). Thus the enhancement as a function of n' or the binding energy can be calculated using this equation. The parameter (P) depends on the second short-pulse laser intensity and on the spatial overlap between this laser and the other two lasers; thus for this simple model, P is a fitting parameter.

III. EXPERIMENTAL SETUP

Ground-state $(4s^2\ ^1S_0)$ calcium atoms in a thermal atomic beam were excited to the $4s4p\ ^1P_1$ excited state using a nanosecond dye laser as shown in Fig. 1. The dye laser used in the excitation was pumped by the third harmonic of a pulsed Nd:YAG (yttrium aluminum garnet) laser. Approximately 4 nsec after the excitation, a 500-fsec laser pulse was used to excite the $4p$ electron to a high Rydberg state. Due to the large bandwidth of the short-pulse laser (30 cm^{-1}), the state of the atom was not a single Rydberg state but a coherent superposition of several Rydberg states (wave packet). For the case when the laser central wavelength was tuned to 393.7 nm, the wave packet was composed of three Rydberg states with principle quantum number $n=20$, 21, and 22. In calcium, the $4s$-$4p_{3/2}$ ionic resonance wavelength is 393.5 nm. The spectral width of the short-pulse laser was large enough to include this resonant wavelength so that atoms excited to a Rydberg wave packet could also be promoted to the $4p_{1/2}nd$ doubly excited states. These states are

not energy eigenstates of the atom, but form a nonstationary two-electron wave packet. A short time after the short-pulse laser excitation, some electrons will evolve into the degenerate bound $4p_{1/2}n'd$ states. These electrons can be stabilized using a second 500-fsec laser pulse that is tuned to the $4p_{1/2}$-$4s$ ionic resonance ($\lambda_{\text{central}}=397$ nm) that stimulates the atoms back to a singly excited stable configuration. The atoms in these states live for a long time (~ 10 μsec), which allows a selective field-ionization measurement to be made.

The short laser pulses were produced using a self mode-locked Ti:sapphire laser that produced about 100-fsec laser-pulse widths, with a repetition rate at 100 MHz. These short pulses were amplified using regenerative amplification which produced energies in excess of 3 mJ with a repetition rate of 20 Hz. The full width at half maximum of the amplified short pulse laser was about 160 cm^{-1}, with a central wavelength at 790 nm. With this much spectral width, it was possible to produce both the core laser wavelengths from a single laser beam simply by splitting the beam into two parts and using two separate 1.2-cm KDP doubling crystals, with each tuned to the proper core laser wavelength. By doing so, we were assured of almost zero temporal jitter between the two short pulses. Each of the laser beams had a variable optical path length that allowed an accurate timing of the two laser pulses. The KDP doubling crystal limits the pulse bandwidth to about 30 cm^{-1}, which corresponds to a minimum pulse duration of about 500 fsec. The actual pulse length is slightly larger due to group velocity dispersion in the KDP crystal.

The calcium atoms were prepared using a resistively heated stainless steel oven with a 1-mm hole on the side. Atoms leaving the oven were crossed at a right angle with the laser beams in the interaction region. A pair of capacitor plates were located above and below the interaction region, with the bottom plate connected to a negative ramped electric field, which was used to field ionize the stabilized Rydberg atoms. The top capacitor plate had a 1-cm hole with a copper mesh to allow the ionized electrons to reach a multichannel plate detector, which was located above the interaction region. The ramped voltage on the bottom capacitor plate was applied at about 100 nsec from the firing of the short-pulse laser. The electric field E that is required to field ionize a state with principle quantum number n' can be calculated using $E=51.3\times 10^8/[16(n')^4]$ (V/cm). For the case when a ramped electric field $E(t)$ was used, the atom was subjected to an increasing electric field value. As a result, states with larger principle quantum number n' will field ionize earlier in time than states with smaller n' values. This technique allowed the measurement of the population distribution in the final n' states to be made.

The experiment consisted of tuning the central wavelength of the first short pulse to several different tunings near the $4s$-$4p_{3/2}$ ionic resonance. In each tuning, the timing of the second short pulse was adjusted to maximize the stabilizing signal. The enhancement in the stabilization of the autoionizing states was measured by comparing the final $4sn'd$ population for data taken with and without the second short pulse. Maximum enhancement of the stabilization signal was achieved with a time delay of approximately 2 psec between the two short pulses. This delay was sufficient for the atoms to evolve from the initial $4p_{3/2}nd$ state to the $4p_{1/2}n'd$ states.

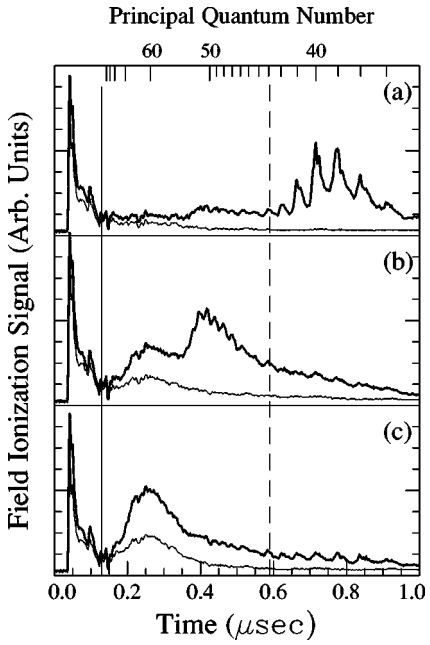

FIG. 2. The field-ionization signal of the final $4sn'd$ states is shown. In (a)–(c), the laser tuning was chosen to maximize the population in one of the three possible Rydberg states, $n=20$, 21, and 22, respectively. The large signal at about 50 nsec represents the autoionization signal. The ramped electric field was applied $\sim$100 nsec after the short-pulse laser was fired and had a rise time of 1 μsec that produced the time-resolved Rydberg signal seen in (a)–(c). The beginning of the ramp is marked on the figure by the solid vertical line. The bold signal is the population in the $4sn'd$ states when the second short pulse was applied. The light signal represents the population that was stabilized when the second short pulse was blocked. The vertical dashed line represents the limit at which the field-ionization method can resolve individual Rydberg states.

IV. RESULTS AND DISCUSSIONS

Figure 2 represents the field-ionization signal versus time for three different tunings of the first short pulse. The large signal at about 50 nsec in Figs. 2(a)–2(c) represents the autoionization signal. The ramped electric field was applied $\sim$100 nsec after the short-pulse laser was fired. The tunings in Figs. 2(a)–2(c) were chosen to maximize the population in one of the three possible Rydberg states, $n=20$, 21, and 22, respectively. In each of the tuning positions, the timing of the second short pulse was adjusted to maximize the stabilized signal. The bold signal in Figs. 2(a)–2(c) represents the stabilization signal when all three pulses were used in the experiment. The light curves in Figs. 2(a)–2(c) represent the stabilization signal when the second short pulse was blocked. From the data in Fig. 2, it can be seen that a large enhancement in the stabilization process was achieved when the second short pulse was used. To measure the enhancement factor due to the application of the second short pulse, the area under the curve for each tuning position was calcu-

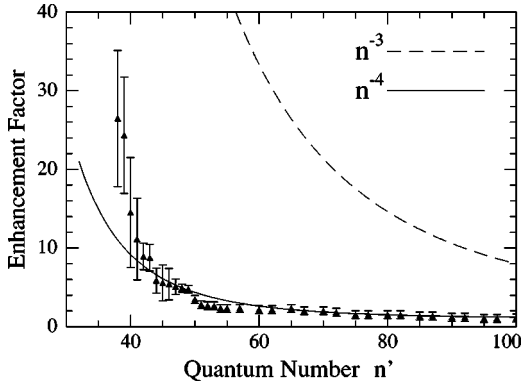

FIG. 3. The enhancement factor in the stabilization of the doubly excited states versus the final Rydberg state principle quantum number (n') due to the application of the second short laser pulse is shown. The dashed curve represents our theoretical calculation for the case when no static electric field was present in the interaction region. The solid curve represents the calculation when a small static field ($\sim$5 V/cm) was present in the interaction region.

lated. The ratio between the area with the second short pulse to the area without the second short pulse is a measure of the enhancement factor in the stabilization process. From the data in Fig. 2, it can be seen that the field-ionization method can resolve individual Rydberg states for $n' < 43$ (marked by the vertical dashed line), as a result, the area under each Rydberg state was measured, but for higher Rydberg states we could not resolve individual states and for that reason the area under the whole signal was divided into a discreet set of regions and the enhancement in each region was measured.

Figure 3 represents the enhancement factor versus the principle quantum number of the final Rydberg states (n') when the second short pulse was applied. As expected, very large enhancement factors were achieved, especially for the low-lying Rydberg states. This was primarily due to the fact that very few autoionization events occurred during the excitation and stabilization process. The sharp increase in the enhancement factor as n' was reduced was due to the fact that the autoionization lifetime decreases very sharply as n' decreases, so without the stabilizing laser most of the electrons autoionize before having a chance to fluoresce. As a result, very little population was detected in the absence of the second short-pulse laser. In a previous experiment, we have shown that the percentage of electrons that reached the $4p_{1/2}n'd$ states after the atom being excited in the pure $4p_{3/2}nd$ state was independent of the initial Rydberg state n, over the same range at which this experiment was performed [16]. The percentage of the population was calculated to be about 60%. As a result, it was possible to calculate the enhancement factor for each tuning position of the first short pulse in Fig. 2 and then average the results to get the overall enhancement factor shown in Fig. 3.

The dashed and solid lines in Fig. 3 are the calculations using Eq. (3) with the autoionization rate scaled as n^{-3} and n^{-4}, respectively. From the results, we can see that the theoretical calculation for the case when using n^{-4} autoionization scaling gives a much better fit to the data. This result suggests that a static electric field ($\sim$5 V/cm) was present in the interaction region during the experiment. This static electric field could have been produced by stray fields from the capacitor plates. For n states below $n=50$, the static field was not sufficient to produce complete mixing of the angular momentum states and the data shows the beginning of a shift to the field free values of enhancement. The fluorescent rate used in the calculation was 3.58×10^{-9} a.u. [17], the scaled autoionization rate (multiplied by n^3) for the calculation when no static electric field was applied was 0.1 a.u., and the scaled autoionization rate (multiplied by n^4) for the case when a small static electric field applied was 0.3 a.u. Both autoionization rates were estimated using Refs. [8,18]. The parameter P in Eq. (3) was found to be about 0.25, suggesting that 25% of the population was stabilized, which produced very large enhancement factors. This shows that without the stabilizing laser, very little of the population would be stabilized.

In this experiment, it was not possible to start from any lower initial Rydberg state (n), the reason being that the purely fluorescent signal was sufficiently small that an accurate measurement could not be made. Also, it was not possible to use simple rate equations out-lined in Ref. [13] to model the experimental results, this was due to the fact that simple rate equations predict a simple exponential decay of the autoionizing states, which was only true for laser pulses long relative to the autoionization life time. In the short pulse case the decay of the autoionizing states was not a simple exponential decay as shown in Refs. [16,19–21]. On the other hand, it was possible to use rate equations to find the limit at which the enhancement factor would level off. This limit corresponds to the case when the short pulse was no longer considered short relative to the autoionization lifetime. From the calculation outlined in Ref. [13], we found that the limit at which rate equations can be used was about $n'=12$, and the maximum enhancement factor that can be achieved using a 500-fsec laser pulse was about 8.8×10^3 at $n'=6$.

V. CONCLUSION

We have shown that using a short-pulse laser to excite and stimulate a doubly excited state, it was possible to achieve very large enhancement factors in the stabilization process. A simple theoretical model was presented that showed relatively good agreement with our experimental results. Even though in this experiment we could not start from a lower initial Rydberg state (n), we have shown that by doing so, a much larger enhancement factor was expected to be produced simply because the atoms have very little chance to fluoresce on their own.

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