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Laser-induced stabilization of autoionizing states

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Stabilization of autoionizing states of barium by laser-induced, stimulated emission of light is demonstrated. Relative to purely flourescent stabilization, the data clearly show an enhancement of the stabilization process for laser pulses short compared to the flourescent lifetime of the autoionizing states. Shakeup spectra in which the principal quantum number of both electrons changes during the stimulated emission process are also clearly demonstrated. [S1050-2947(99)07711-2]

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I. INTRODUCTION

Doubly excited states of atoms with energies above the first ionization limit play an important role in dielectronic recombination (DR), a process in which an electron is captured from the continuum by an ion. The doubly excited states provide the intermediate states into which electrons are initially captured through an electron-ion collision [1]. Once the doubly excited state is produced, the atom can be stabilized through the emission of a photon leaving the atom in a singly excited bound state. If, on the other hand, the atom decays through autoionization, the captured electron is released back into the continuum. The fluorescence branching ratio determines what percentage of the doubly excited states will be stabilized. The DR rate is determined by the product of the collision excitation rate times the fluorescence branching ratio. In the case in which one of the electrons is in a high-lying Rydberg state and the other electron is in a relatively low-lying excited state, it has been shown that the presence of a static electric field can enhance the fluorescence branching ratio [2–13]. The enhancement is due to mixing of the angular momentum in the Stark Rydberg states, which decreases the autoionization rate of the doubly excited states. The electric field has no effect on the photoemission rate which is determined by the fluorescence rate of the low-lying electron.

In the following it will be shown that under certain conditions the presence of a laser field can lead to enhanced stabilization of doubly excited states due to stimulated emission from the low-lying electron. The presence of the laser field does not change the nature of the doubly excited states, as is the case with a static field, but simply increases the photoemission rate, effectively increasing the fluorescence branching ratio. The laser can also have the undesirable effect of reexciting the atoms to the doubly excited state after photoemission has occurred. In this case the laser will have the opposite effect of lowering the yield of stabilized atoms. For this reason the temporal parameters of the laser are critical factors in determining the effects of the presence of the laser. Specifically, the laser pulse used to induce stabilization must have a pulse length less than the fluorescent lifetime of the doubly excited states.

A second effect of the laser interaction with the doubly excited states which will be discussed in the following is the presence of satellite resonances in the stimulated emission spectra. Satellite resonances are the result of a two-electron transition in which the inner electron undergoes a dipole transition and the outer, Rydberg electron is ‘‘shaken’’ into a state with a different principal quantum number. Satellite resonances are well known in the excitation spectra of doubly excited states [14,15]. To our knowledge, this is the first observation of these resonances in a stimulated emission spectra.

II. EXPERIMENTAL SETUP

Figure 1 shows the excitation scheme used in the experiment. The experiment used the isolated core excitation technique [16] in which atoms are initially prepared in a singly excited Rydberg state followed by excitation of the core electron to produce a doubly excited state. Two lasers were used to prepare barium atoms in the $6s11d\,^1D_2$ singly ex-

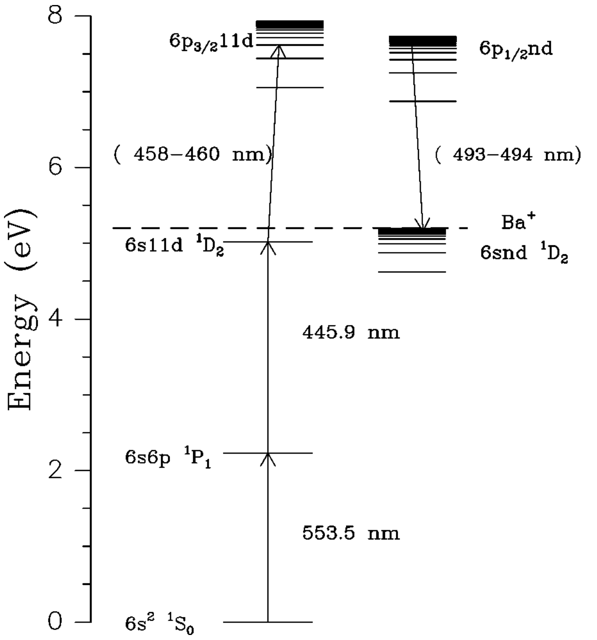

FIG. 1. Energy levels of barium involved in the experiment are shown along with the relevant laser transitions. Two photons were used to excite the $6s11d\ ^1D_2$ state. A third laser then excited a doubly excited state in the energy range of $6p_{1/2}nd$ Rydberg series. A fourth laser was used to drive the transition from the $6p_{1/2}nd$ states to the $6snd$ states.

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cited state. A third laser tuned near the 6s to $6p_{3/2}$ ionic transition was then used to excite the $6p_{3/2}11d$ doubly excited state which overlaps in energy with the $6p_{1/2}nd$ doubly excited Rydberg series. The energy eigenstates produced by this excitation have both $6p_{3/2}11d$ and $6p_{1/2}nd$ character due to interaction between the two electrons. This excitation scheme has previously been used to study fluorescent stabilization of doubly excited states [17] and the effects of static electric fields on the DR process [13]. Stabilization of the atoms can occur when a photon is emitted by the ionic core, which returns atoms to a singly excited state. The previous experiments have examined the fluorescence process in which the barium atoms emit a photon on the $6p_{1/2}nd$ to $6snd$ transition and in the case of Stark states the $6p_{1/2}nk$ to $6snk$ transition. In the present experiment, a fourth laser was tuned near the $6s$ to $6p_{1/2}$ ionic transition for the purpose of driving the $6p_{1/2}nd$ to $6snd$ transition through stimulated emission of a photon. The driven process has two main differences from the fluorescent process. Re-excitation of the atoms can occur in the driven process, which makes the results of the stabilization process highly dependent on the temporal parameters of the laser pulses. Second, the increased strength of the transition due to the driving field allows the examination of weak transitions such as the $6p_{1/2}nd$ to $6sn'd$ transition in which the initial and final n states are different.

The four lasers used in the experiment were pulsed tunable dye lasers, pumped by either the second or third harmonic of a Q-switched Nd:YAG laser which produced 8 ns pulses at a 20 pulse per second repetition rate. The two dye laser pulses used to excite the $6s11d\,^1D_2$ state were timed to arrive at the interaction region slightly before the two pulses used to perform the ionic core transitions (~ 10 ns). The two pulses used in the ionic transition were temporally overlapped and had pulse durations in the range of 1.2–1.6 ns. The four laser beams were focused and crossed in the interaction region. The first three lasers were tightly focused in the interaction region. The fourth laser was slightly defocused in the interaction region so that the intensity of this laser was relatively uniform over the range in which doubly excited states were produced.

The atomic beam was produced by a resistively heated barium oven. A 1 mm aperture was used to collimate the atomic beam which crossed the laser beams in the interaction region at a right angle inside a vacuum chamber. Capacitor plates were placed above and below the interaction region with a screen mesh in the upper plate which allowed electrons to pass. A pair of microchannel plate charged particle detectors located above the interaction region was used to detect electrons produced in the experiment and data were collected on a digital oscilloscope and stored on a computer.

Stabilization of the doubly excited states through photoemission was detected by measuring the final-state distribution of barium atoms in the high n, $6snd$ states. These states could be populated only by the emission of a photon via the $6p_{1/2}nd$ to $6snd$ transition. Detection was accomplished by field ionizing the Rydberg atoms using a ramped negative voltage which was applied to the lower capacitor plate. Electrons produced by the field ionization process were driven through the screen in the top capacitor plate into the charged particle detector. The ramped voltage allowed the measurement of the final n state distribution, since the different n states field-ionize at different values of the electric field. Calibration of the n versus time measurement was accomplished using only the first three lasers responsible for the excitation of the doubly excited states. The laser used to drive the $6s$ to $6p_{3/2}$ transition was tuned to different known energies which overlapped with the $6p_{1/2}nd$ series. The final bound-state population produced by fluorescence of the $6p_{1/2}nd$ to the $6snd$ states was then detected. In the case of fluorescence, the probability is extremely small that the Rydberg electron will change its value of n during the core transition since the transition moment is heavily weighted near the ion resonance where the Rydberg state remains unchanged [18].

Measurements of the enhancement of stabilization of the autoionizing states were performed by comparing the final $6snd$ population for data taken both with and without the presence of the fourth laser. In the first measurement, the fourth laser was tuned to the $6s$ to $6p_{1/2}$ ionic transition and measurements were made over a range of Rydberg states. A second measurement consisted of detuning the fourth laser from the ion transition to drive the $6p_{1/2}nd$ to $6sn'd$ transition, where the final n' was determined by the specific laser tuning. These transitions correspond to ‘‘shakeup’’ transitions in which both electrons change state. These measurements were accomplished by detecting the change in the final-state population distribution using the ramped field technique described above.

III. THEORY

For a case in which atoms are excited to an autoionizing state and left to themselves, the fraction of atoms which are stabilized through fluorescent decay can be determined by calculating the fluorescence branching ratio given by

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

where R_{fl} is the fluorescence rate and R_{ai} is the autoionization rate. Consider a simple case such as the $6p_{1/2}nd$ doubly excited states of barium in zero electric field, where n represents the principal quantum number of a Rydberg state. The fluorescence rate is equal to the fluorescence rate of the $6p_{1/2}$ 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 how the autoionizing states are excited. It is also independent of the time scale over which the autoionizing states are produced. If a laser is used to stimulate the emission of a photon in the stabilization process, a time-independent branching ratio cannot be defined. The fraction of atoms which will be stabilized is strongly dependent on the temporal parameters of the excitation and stabilization lasers. There are three separate time scales of interest which are determined by the laser pulse time. The first time scale involves laser pulse durations which are short compared to the autoionization lifetime of the doubly excited states. In this temporal regime a fully quantum-mechanical treatment of the atoms must be employed to account for the wave-packet nature of the doubly excited states [19–22]. The second temporal regime involves laser pulse durations which are long compared to the autoionizing lifetime but short relative to the fluorescent lifetime of the autoionizing states. In this regime, the atom-laser interaction can accurately be modeled by transition rate equations. The third temporal regime in which the laser pulses are long compared to the fluorescent lifetime of the doubly excited states is of little interest due to the process of reexcitation of stabilized atoms to the autoionizing states. The net effect of reexcitation is a large reduction of the final number of atoms which are stabilized. In this paper we will examine the intermediate temporal regime for the $6p_{1/2}nd$ autoionizing states, which requires laser pulses which are short compared to the 6.2 ns fluorescent lifetime of these states. The rate equations which govern the evolution of the atoms in this regime can be stated as follows:

$$\frac{dN_1(t)}{dt}=P_1(t)-R_{\mathrm{fl}}N_1(t)-R_{\mathrm{ai}}N_1(t)-P_2(t)N_1(t)+P_2(t)N_2(t) \tag{2}$$

and

$$\frac{dN_2(t)}{dt}=R_{\mathrm{fl}}N_1(t)+P_2(t)N_1(t)-P_2(t)N_2(t), \tag{3}$$

where N_1 represents the population in the doubly excited state and N_2 represents the population in the final singly excited state. $P_1(t)$ is the excitation rate into the doubly excited state which depends on the excitation laser pulse. For large values of $P_1(t)$, depletion of the initial singly excited Rydberg population would have to be included. In the present experiment the excitation rate was kept sufficiently low that depletion could be neglected. $P_2(t)$ is the rate of population transfer between the doubly and singly excited states due to a laser pulse tuned to the $6s$ to $6p_{1/2}$ ionic transition. $P_2(t)$ is given by the product of the photoemission cross section times the laser flux. R_{fl} represents the fluorescence rate between the $6p_{1/2}$ and $6s$ ionic states and R_{ai} represents the autoionization rate of the doubly excited states. R_{fl} is simply given by one over the fluorescence lifetime of the ionic states. R_{ai} depends on the particular n state and on the amount of Stark mixing of the Rydberg states.

If the fourth laser is detuned slightly from the $6p_{1/2}$ to $6s$ ionic transition, then the Rydberg electron must change n during the transition, to conserve energy. This type of transition, known as a shakeup transition, is commonly seen in photoabsorption experiments [14,15]. The change in n is a result of the changing ionic charge distribution seen by the Rydberg electron during the core transition [19]. The transition moment for the shakeup transition is given by [18]

$$T=\mu A(n_f^*)\frac{\sin[\pi(n_f^*-n_i^*)]}{\dfrac{1}{2(n_f^*)^2}-\dfrac{1}{2(n_i^*)^2}}, \tag{4}$$

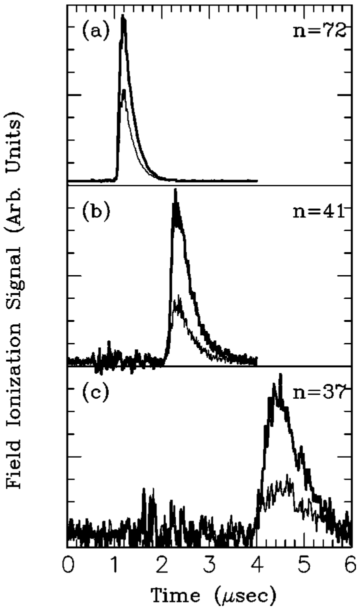

FIG. 2. Field ionization signal vs time is shown for three tunings of the third laser. The data represent atoms which were stabilized via photon emission. The bold curve represents data taken with the fourth laser tuned to the $6s$ to $6p_{1/2}$ ionic transition. The light curve represents data taken with the fourth laser blocked. The corresponding n states produced by the three tunings are (a) $n=68$, (b) $n=40$, and (c) $n=37$. The data in (b) are magnified by a factor of 15 relative to the data in (a), and the data in (c) are magnified by a factor of 60 relative to the data in (a). The data clearly show an enhancement of stabilization due to the presence of the forth laser.

where μ is the ionic dipole moment, $A(n_f^*)$ is the density of final states, and n_i^* and n_f^* are the effective quantum numbers of the initial and final Rydberg states, respectively. The final Rydberg state is determined by the detuning of the core laser from the ionic resonance, where zero detuning would give $n_f^*=n_i^*$. The transition moment drops off rapidly with detuning from the ionic resonance. This expression can also be used to give the final-state Rydberg distribution due to fluorescence decay of a doubly excited state. In a fluorescent decay event, it is possible for the atom to emit a photon slightly off of the ionic resonant frequency leading to a shakeup transition. This type of transition has never been observed in fluorescence due to the dominance of the on-resonance transition moment. In an absorption transition it is possible to increase the size of the shakeup spectra relative to the on-resonance signal by saturating the on-resonance transition. In the present experiment, saturation can also be used to enhance the shakeup spectra in the stimulated emission process.

IV. RESULTS AND DISCUSSION

Figure 2 shows stabilization data for three tunings of the third laser corresponding to three different values of n in the $6p_{1/2}nd$ series. The light curve shows data corresponding to fluorescence to the $6snd$ states without the presence of the fourth laser. The bold curve shows data with the fourth laser tuned to the $6s$ to $6p_{1/2}$ ion transition. In these data the third and fourth lasers were temporally overlapped and had a pulse

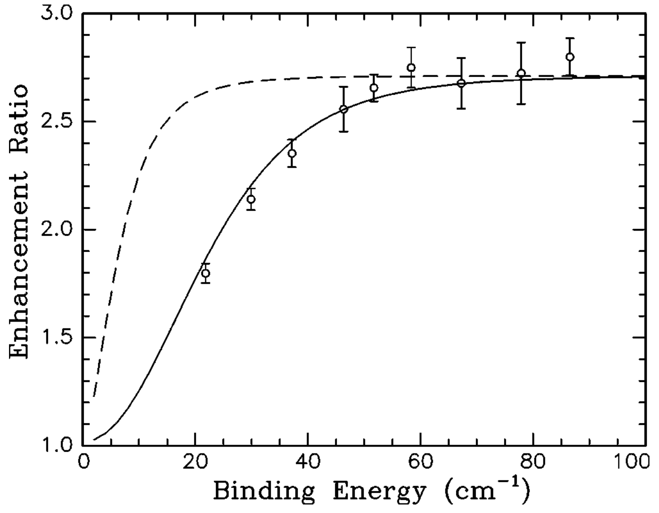

FIG. 3. Circles represent the ratio of the stabilization signals with and without the presence of the fourth laser for a number of tunings of the third laser. The solid line represents the calculated ratio assuming complete mixing of the *l* states by a static electric field. The dashed line represents the calculated ratio without field mixing. Both calculations used laser pulse durations of 1.6 ns and maximum laser field strengths of five times the saturation intensity for the $6p_{1/2}$ to $6s$ ion transition. The data are in good agreement with the mixed *l* calculation, which is consistent with the estimated stray fields in the interaction region of 5 V/cm.

duration of 1.6 ns. The data clearly show an enhancement due to the presence of the fourth laser for the three cases shown.

Figure 3 shows a plot of the enhancement versus binding energy of the Rydberg electron. The enhancement factor is simply given by the ratio of the two signals, with and without the fourth laser, as shown in Fig. 3 . The circles represent the measured enhancement factors. For very-high-lying Rydberg states, corresponding to small binding energy, the enhancement approaches 1. This limit is due to the fact that the autoionization rate goes to zero for high *n* states so that all atoms are stabilized regardless of whether the fourth laser is present or not. The dashed line represents the calculated enhancement factor using the rate equation given in Eqs. (2) and (3), with an autoionization rate appropriate for the $6p_{1/2}nd$ states. The two equations were numerically integrated to obtain the final-state population in the singly excited state. The solid line represents the calculated enhancement factor using an autoionization rate appropriate for the $6p_{1/2}nk$ Stark states. Both calculations used Gaussian laser pulses for $P_1(t)$ and $P_2(t)$. Comparison of the two calculated curves shows that there is only an appreciable difference for the high *n* states. The excellent agreement between the data and the Stark calculation suggests the presence of stray electric fields in the interaction region. The fields necessary to produce Stark mixing down to a binding energy of 40 cm^{-1} are of the order of 5 V/cm. These fields may be produced by stray fields from the capacitor plates.

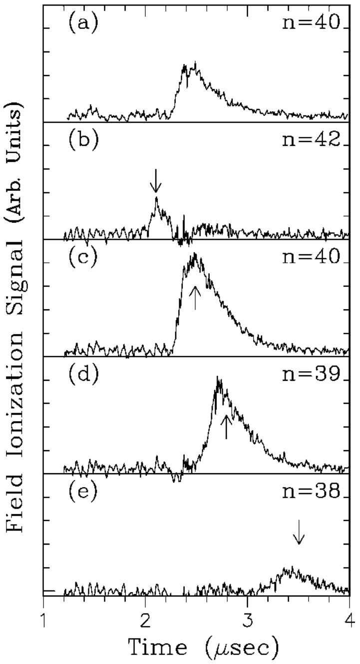

FIG. 4. Stabilization signal is shown for several tunings of the fourth laser near the $6s$ to $6p_{1/2}$ ion transition. In all of the figures the third laser was tuned to 446 nm, which produced $n=40$ Rydberg states. (a) shows the purely flourescent $n=40$ signal with the fourth laser blocked. In (b)–(e) the fluorescence signal is subtracted from the stabilization signal so that only the signal due to stimulated emission is shown. The arrows correspond to the calculated positions of the laser-induced stabilization, which are in good agreement with the actual positions for all tunings of the stabilization laser. The four tunings of the laser shown correspond to detunings from the $6s$ to $6p_{1/2}$ ion resonance of (b) -6.4 cm^{-1}, (c) 0.0 cm^{-1} , (d) 3.6 cm^{-1} , and (e) 7.4 cm^{-1}.

The one free parameter in the calculation was the intensity of the fourth laser. It was found that the final result of the calculation was relatively insensitive to the intensity of the fourth laser. The intensity of the fourth laser was defined in terms of the saturation intensity, where saturation is defined as the point at which the peak value of $P_2(t)$ is equal to the fluorescence rate R_{fl}. The calculation showed little change in the enhancement factor for intensities between four and eight times the saturation intensity. For intensities both above and below this range, the enhancement was found to decrease. The decrease at lower intensity is simply due to too small of a pump rate out of the doubly excited state. The decrease at higher intensity is due to repumping of the stabilized atoms during the tail of the Gaussian temporal pulse. This effect was also observed in the experiment. The data shown in Fig. 3 represent the optimal intensity for the pulse durations used in the experiment. The intensity used in the calculation shown in Fig. 3 was five times the saturation intensity, which maximized the enhancement and gave the best agreement with the experiment. The intensity of the fourth laser was not measured, since the actual value depended not only on the spatial parameters of the laser but on the exact spatial overlap between the fourth laser and the other three lasers, which is extremely difficult to measure with any accuracy. The spatial overlap was particularly difficult to determine in this experiment since the alignment was set to maximize the stabilization signal. As discussed above, the stabilization signal has an optimal intensity range, so that for a given peak intensity at the center of the beam, a slight misalignment may produce a maximum stabilization signal. It was found that an increase in the intensity of the fourth laser did reduce the stabilization signal in agreement with the calculation.

The stabilization enhancement was found to be extremely sensitive to the duration of the laser pulses and the temporal overlap of the laser pulses. Longer laser pulses reduced the enhancement due to repumping of the stabilized atoms. Also, as one would expect, the third and fourth laser pulses needed to be precisely overlapped in time to see any enhancement.

Figure. 4 shows the effect of detuning the fourth laser from the ionic resonance. As discussed earlier, these transitions require the Rydberg electron to change energy in order to conserve the total energy in the stimulated emission process. This process represents a shakeup transition in the stimulated emission process. For clarity, the purely fluorescent signal has been subtracted from the data, leaving only the signal due to stimulated emission. The arrows in the figure correspond to the calculated position of the field ionization signals using the measured detunings and the calibrated Rydberg state versus time data. The signal drops off rapidly with detuning. However, the signal drops off much slower with detuning than the cross section would predict. This slower rate of decrease is due to the fact that the stimulated emission process is saturated. This effect is common in absorption spectra, in which the satellite resonances are increased relative to the central resonance due to saturation of the central resonance. In principle, these satellite resonances should appear as very small sidebands in a purely fluorescent signal. They are sufficiently small that they have never been observed in a purely fluorescent transition. This experiment represents, to the best of our knowledge, the first direct observation of the shakeup transition in a photoemission process.

V. CONCLUSION

In the preceding it was demonstrated that under certain conditions it is possible to enhance the stabilization of autoionizing states through laser-induced stimulated emission. Unlike the enhancement due to Stark mixing in a static electric field, the laser-induced stabilization does not change the atomic structure, but simply increases the probability of photoemission. It was also shown that by using a laser to induce photoemission, it was possible to change the final-state principal quantum number by tuning the laser to a satellite resonance.

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