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Two-step stabilization of autoionizing states

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A two-step process using internal conversion and fluorescence to stabilize the $6P_{3/2}11d$ autoionizing state of barium has been observed. The internal conversion produces very highly excited autoionizing Rydberg states, which then emit fluorescence to produce singly excited, bound states. Inclusion of this process should bring calculations closer to agreement with recent measurements of dielectronic recombination.

Recently, there have been advances in the study of autoionization from both temporal directions. Laser excitation schemes have made it possible to populate, efficiently and selectively, a wide variety of autoionizing states of neutral atoms.[1] The time-reversed process, dielectronic recombination (DR), has now been observed also in crossed-beam experiments.[2-4] These two sets of experiments have usually produced complementary information: The laser-based experiments produce easily interpreted atomic characteristics, such as state positions and autoionization rates; the crossed-beam experiments measure total cross sections for DR. Technologically, the cross sections are more important since DR is a useful solar plasma diagnostic and an important charge-reduction process in controlled thermonuclear reactor plasmas. However, since the cross sections are not measured under the same conditions as exist in the plasmas of interest, it is important that models which use atomic parameters as input be able to reproduce the measured cross sections. So far, the measured cross sections have been considerably larger than those calculated.

The most recent crossed-beam work has shown that part of the discrepancy is certainly due to the presence of ambient electric fields; however, the history of the recognition of this effect shows the interrelationship between theory, atomic parameter measurements, and DR cross-section measurements. In 1976, Jacobs and Davis calculated that electric fields would enhance DR since high-l states begin to autoionize as the electric field mixes in low-l contributions.[5] In 1978, Freeman and Bjorklund used a four-wave mixing experiment to demonstrate this increased autoionization of high-l states in strontium.[6] The recent DR measurements have completed the cycle.

Yet, even after accounting for electric field effects, the measured cross sections are still larger than the calculated ones, suggesting that still other effects must be taken into account. In this work we show that a two-step stabilization process can significantly enhance the production of highly excited, bound Rydberg states produced by an autoionizing state. The two-step process of internal conversion to a second autoionizing state followed by radiative stabilization has been directly observed in a laser excitation experiment, and is presumably present in the crossed-beam experiments.

An atom in a doubly excited state above the first ionization limit has two possible decay processes: autoionization producing an ion and free electron, and fluorescent decay to a bound, singly excited state. For highly excited states, the large separation of the spatial wave functions of the two electrons reduces their interaction and produces a low autoionization rate A_{AI}, which is proportional to n^{-3}, where n is the principal quantum number of the more highly excited electron. Since the other electron fluoresces, the fluorescence rate does not depend on n. Thus, for sufficiently high n, the fluorescence rate can become the dominant decay process. Such highly excited states are usually not accessible because of their small oscillator strengths (again, due to little overlap of the high n wave function with the lower lying wave functions). However, a two-step process can efficiently populate these high Rydberg states which then will fluoresce.

Since the internal conversion conserves energy, one can equivalently view this process as the time-independent configuration interaction (CI). In this view, configuration mixing causes the states to be linear combinations of two components, one having a large oscillator strength and the other having a large fluorescence yield. If one examines the temporal evolution after exciting a band of these states using a short pulse, then the excited wave packet would evolve from one which had primarily the large oscillator strength character into one which had the large fluorescent yield character. This behavior suggests the two-step nomenclature even though the time independent CI analysis is equivalent.

We have observed this two-step process in barium using the isolated core excitation (ICE) method,[1,7] as shown in Fig. 1. Two pulsed, circularly polarized lasers stepwise excite an effusive beam of barium to the $6s11d\,^1D_2$ state using the $6s6p\,^1P_1$ intermediate state. A third pulsed, circularly polarized laser then excites the $6s$ core electron, producing the $6P_{3/2}11d$, $J=3$ autoionizing state. This state is centered just above the $6P_{1/2}$ ionization limit; however, the tail of its excitation profile extends below that limit. We measured the total excitation profile by exciting the barium atoms in a field free region between two parallel plates, separated by 1 cm. After the third laser pulse, a 50-V pulse was applied to the bottom plate to drive the ions produced by autoionization through a hole in the top plate and into a particle detector. In a second measurement, we mounted an optical detection system (an $f=2$ lens with a 10-nm bandpass, 493-nm interference filter) to collect fluorescence from any excited $Ba^+(6P_{1/2})$ ions. Both pro-

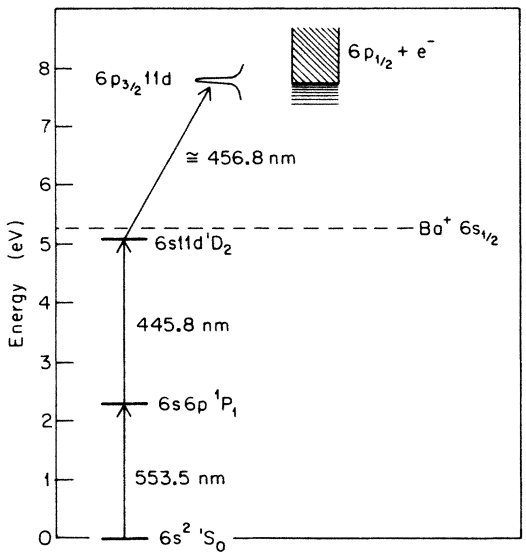

FIG. 1. Energy-level diagram of Ba showing the relevant transitions for populating the $6p_{3/2}11d$ state.

files were measured as a function of the third laser's tuning, as shown in Fig. 2. When the third laser's photons had sufficient energy ($\lambda < 458.31$ nm) then it was possible, and even likely, for the $6P_{3/2}11d$ state to produce an excited $6P_{1/2}$ ion and a slow electron after autoionization. This produced the fluorescence signal seen in Fig. 2. Just to the left of the dashed line in Fig. 2, the internal conversion that produced $6P_{1/2}$ ions and slow electrons still occurred, but the product was then $6P_{1/2}$ ions and bound electrons.

The fluorescing autoionizing states naturally divide into three regions of principal quantum number n. For the highest n states ($n \geqq 600$), the spacing between Rydberg states is smaller than the fluorescence broadened linewidth. It is no surprise that these states are as efficient in producing fluorescence as the ion itself, since the core fluorescence occurs in a time shorter than the classical Rydberg electron orbit period. This region extends to only 0.006 nm below the ionization limit. In the second region, $n \sim 200$, the fluorescence rate and the autoionization rate are comparable so that interference effects can become important. Bell and Seaton have treated this problem in detail in their treatment of DR.[8] For the final region, small $n (\leqq 100)$, the fluorescence rate is smaller than the autoionization rate and the two processes can be described using simple rate equations. In this case, the fluorescence yield is proportional to the autoionizing state's population, times the branching ratio factor,

$$A_{FL}/(A_{AI}+A_{FL}) ,$$

where A_{FL} and A_{AI} are the fluorescence and autoionization rates, respectively.

In Fig. 3, we show an expanded view of the region just below the $6P_{1/2}$ ionization limit. The solid line is a model which used an ICE excitation line shape[7] and the branching ratio factor

$$S = S_0 \frac{|\langle n_i | n_f \rangle|^2}{|\sin[\pi(n_f+\delta_f+i\gamma_f)]|^2} \frac{A_{FL}}{(A_{FL}+A_{AI})} . \quad (1)$$

Here, δ_f is the quantum defect of the $6P_{3/2}11d$ state ($\delta_f = 2.75$), γ_f is n_f^3 times the linewidth of that state ($\gamma_f = 0.12$), and n_i and n_f are the effective quantum numbers of the initial $6s11d\,^1D_2$ state ($n_i = 8.4107$)[9] and the final $6P_{3/2}11d$ state, respectively. (The value of n_f varies with the third laser's tuning.) The function $\langle n_i | n_f \rangle$ represents the overlap of the radial part of the initial and final wave functions. S_0 is a normalization constant.

The model fits the data well, above the ionization limit, and for low $n (\leqq 60)$. From the low-n behavior, we can determine that the $6P_{1/2}nl$ states that are fluorescing must have a linewidth of at least $0.005n^{-3}$, but certainly no more than $0.02n^{-3}$. This strongly suggests that at those quantum numbers, the $6P_{1/2}ng$ configuration is producing most of the fluorescence.[10] The model fails badly in the

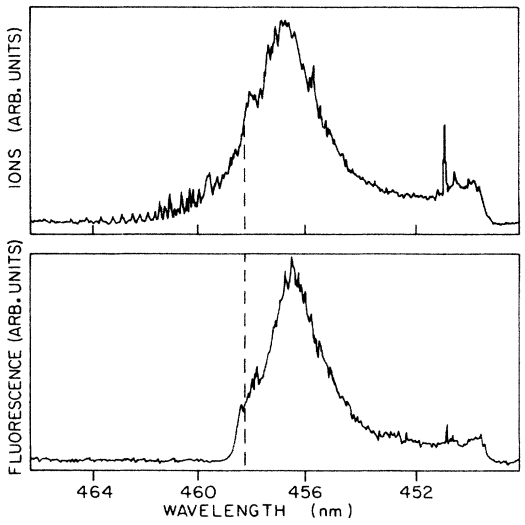

FIG. 2. Ion and fluorescence data showing a cutoff of fluorescence below the $6p_{1/2}$ ionization limit (dashed line).

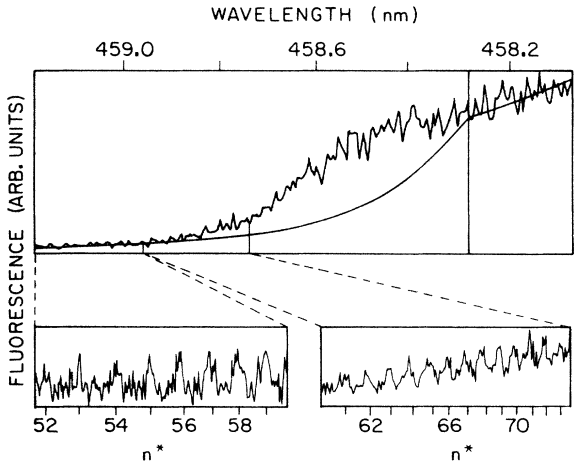

FIG. 3. Expanded view of below-threshold fluroescence. The solid verticle line represents the $6p_{1/2}$ ionization limit and the solid curve is the model described in the text. Rydberg structure is evident in the lower-n region.

region between $n \approx 65$ and the ionization limit. We ascribe this to the effects of Stark field mixing of the $6P_{1/2}ng$ states with configurations having high l values and lower zero-field autoionization rates. Such a mixing would enhance the branching ratio. We have estimated these fields to be of the order of 0.25 V/cm from the disappearance of the Rydberg structure near $n \approx 100$. Such a field would mean that only for states with $n \leqq 63$ would the field be small enough to preserve the angular momentum quantum number of a g state. This ambient field is presumably due to plasma fields generated by the large number of autoionizing states produced.

This Stark mixing, as mentioned above, was one of the first complications to be considered in models of DR. The cross section for DR through a specific autoionizing state can be simply related to that state's autoionization rate through detailed balancing arguments such as:

$$\sigma \propto A_{\mathrm{AI}} A_{\mathrm{FL}} / (A_{\mathrm{AI}} + A_{\mathrm{FL}}) \approx A_{<} , \qquad (2)$$

where the proportionality constant depends on the electron and ion densities, and $A_{<}$ corresponds to the smaller of the two rates. Under these conditions, the total DR cross section usually can be well approximated by summing the partial cross sections over only those states where A_{AI} is larger than A_{FL}. Consequently, those states where fluorescence dominates do not contribute much to the DR cross section. High n states, for example, will not contribute to the DR cross section, because although they would fluoresce very efficiently once populated, their autoionization rates are sufficiently small that the states are never populated in the first place.

The two-step process described above, however, *can* efficiently populate the high n states so that their contribution to DR will be significant. In terms of rate arguments, the configuration mixing enhances A_{AI} well above the normal n^{-3} scaling law, and the (usually large) number of high-n states then have contributions according to Eq. (2). This type of effect has been considered by McLaughlin and Hahn in highly ionized iron (Fe^{23+}) by introducing configuration mixing into the autoionizing state basis;[11] however, they only considered mixing between states of nearly the same principal quantum number. (For highly ionized states the fluorescence rates are much faster, so that A_{AI} is greater than A_{FL} only for the lowest-lying states.) In the case of lower ionization stages, such as the measurements in Mg, the ionic fine structure is of the right magnitude to maximize the two-step process. For example, calculations of DR in Mg show that only states with $n < 50$ should contribute to the DR cross section.[2] However, the $\mathrm{Mg}(3P_{1/2}nl)$ states with high n can be strongly mixed with the $\mathrm{Mg}(3P_{3/2}34l)$ states and thus produce *more* high-n bound states through fluorescence than otherwise expected.

Jacobs, Rogerson, Chen, and Cowan have recently proposed a similar two-step process to enhance DR.[12] In this model, other free electrons cause transfers from an initially populated autoionizing state to a high-l fluorescing state through collisions. This effect, like the Stark mixing effect, serves to enhance the production of high-l states with moderate n values. The internal conversion process described here, however, will not produce high-l states, since angular momentum must still be conserved, but will produce high-n states. Both of these processes will probably only be of importance in tenuous plasmas, since high l and high-n states are both likely to be ionized through collisions or background radiation before they radiatively stabilize to a low-lying state. The conditions in the crossed-beam experiments, however, will allow these states to contribute to the total DR cross sections.

We have shown that one can efficiently excite high-n autoionizing states by using internal conversion after first exciting a lower-n state that converges to a higher ionization limit. This method may be useful for high precision studies of autoionizing states' lineshapes.[13] The two-step process has probably contributed to recent DR cross section measurements, although it has not been included in the calculations of these cross sections.

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