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Magnetic field enhancement of dielectronic recombination from a continuum of finite bandwidth

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We report the results of a study of the effects of combined electric and magnetic fields on dielectronic recombination (DR) from a continuum of finite bandwidth. Specifically, we have examined the process Ba $6p_{3/2}8g \rightarrow$ Ba $6p_{1/2}nk \rightarrow$ Ba $6s_{1/2}nk+h\nu$ in the presence of electric fields from 0 to 7 V/cm and magnetic fields from 0 to 250 G. Our observations elucidate the requirements for magnetic field enhancement of the DR rate. In particular, they demonstrate that the magnetic coupling must be comparable not only to the electric field splitting of the intermediate autoionizing Rydberg states, but also to their decay rates.

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Dielectronic recombination (DR) is the process by which an ion and an electron recombine via a doubly excited autoionizing state [1]. While it can be an important recombination mechanism in low-temperature plasmas [2], it is critical for recombination in high-temperature plasmas. In the latter case the predominant contribution is via doubly excited Rydberg states. For example, a ground-state Ba^+ ion can recombine with an electron via the process

$$\mathrm{Ba}^+\ 6s_{1/2} + e^-\epsilon\ell' \rightarrow \mathrm{Ba}\ 6p_{1/2}n\ell \rightarrow \mathrm{Ba}\ 6s_{1/2}n\ell + h\nu. \quad (1)$$

We follow the convention that n, ℓ, and m are the principal, orbital angular momentum, and azimuthal angular momentum quantum numbers of the Rydberg electron. The incoming electron, which has an energy just below that required to excite the Ba^+ $6p_{1/2}$ state, excites the ionic $6s_{1/2} \rightarrow 6p_{1/2}$ transition and is itself captured, resulting in a Ba $6p_{1/2}n\ell$ atom. If this atom radiatively decays to the bound $6s_{1/2}n\ell$ state, DR has occurred.

When DR passes through the intermediate autoionizing Rydberg states, the DR rates are very sensitive to external perturbations [3]. In particular, Burgess and Summers noted that long-range collisions with background ions and electrons in the plasma redistribute or mix the intermediate Rydberg angular momentum states, leading to DR rates in excess of those that might normally be expected. Jacobs *et al.* made the seminal observation that the slow-moving ions in a plasma lead to quasistatic microfields, which have the same effect as a macroscopic electric field [4]. While we are here concerned with DR, we note that the insights into field effects on DR are directly applicable to zero-kinetic-energy spectroscopy [5] and fluorescence yield spectroscopy from doubly excited states [6].

Though the practical importance of DR is in plasmas, detailed insight is more easily obtained by other means. For example, crossed beam, merged beam, storage ring, and laser spectroscopy experiments have shown clearly that a macroscopic electric field increases the DR rate [7–11], as predicted by Jacobs *et al.* [4]. A magnetic field by itself is expected to have no effect on DR rates unless it is strong enough that diamagnetism comes into play [12]. For this reason, the omnipresent magnetic fields in storage ring DR experiments were ignored until Robicheaux and Pindzola suggested that, in the presence of an electric field, a perpendicular magnetic field strong enough to couple Stark states of m and $m\pm1$ can create a mixture of m states [13]. Analogous to the ℓ mixing by an electric field, the mixing of the m states by the magnetic field should further enhance the DR rate, and this problem has been explored theoretically in some detail [14,15]. Experiments have shown that magnetic fields have an effect on DR rates, in some cases increasing the DR rate [16] and in others decreasing it [17,18]. Furthermore, the experiments to date have not clarified the the precise requirements for magnetic field enhancement of the DR rate.

Here we report the results of an experiment in a low-electric-field regime which demonstrates that enhancement of the DR rate requires that the B field coupling exceed not only the E field splitting of the states of adjacent m, but the autoionization and radiative decay rates of the intermediate Rydberg states as well. In the sections that follow, we briefly recount why fields enhance the DR rate and outline the requirements for B field enhancement. We then describe our experiments on DR from a continuum of finite bandwidth in combined E and B fields, which demonstrate the importance of the magnetic coupling's exceeding both the Stark splitting and the autoionization rate. Unless stated otherwise, atomic units are used throughout.

If we assume the Ba $6s\epsilon\ell'$ continuum to be the only continuum, the DR rate via the $n\ell$ state, $\Gamma(n\ell)$, for the process of Eq. (1) is given by the product of the capture rate and the branching ratio for radiative decay to the $6sn\ell$ state. Explicitly,

$$\Gamma(n\ell) = \beta A_I(n\ell)\frac{A_R}{A_I(n\ell)+A_R} \approx \beta A_<. \quad (2)$$

Here $A_I(n\ell)$ and A_R are the autoionization and radiative decay rates of the Ba $6p_{1/2}n\ell$ state, and β is a constant that relates the capture rate to the rate of its inverse process, autoionization. A_R is the radiative decay rate of the excited Ba^+ ion. The $6p_{1/2}n\ell$ state decays radiatively to the $6sn\ell$ state at this rate, since the $n\ell$ electron is simply a spectator in this process. Unless $A_I(n\ell) \approx A_R$, $\Gamma(n\ell) = \beta A_<$, where $A_<$ is the smaller of $A_I(n\ell)$ and A_R. Since $A_I(n\ell) = \gamma(\ell)n^{-3}$ [19], and $\gamma(\ell)$ falls approximately exponentially with ℓ, the energy-integrated DR rate Γ is given by

$$\Gamma \approx \beta N_> A_R, \quad (3)$$

where $N_>$ is the number of states for which $A_I(n\ell) > A_R$, and we assume that $A_I(n\ell) > A_R$ for low n. We are ignoring the

contributions of high-n states for which $\Gamma(n\ell)=\beta\gamma(\ell)n^{-3}$ [11].

Using Eq. (3) it is straightforward to understand how E and B fields can increase the DR rate. An electric field converts the zero-field $n\ell$ states into nk Stark states, which have approximately the average autoionization rate of all the ℓ states of the same n and m. Often, $A_I(n\ell)\gg A_R$ for low ℓ and $A_I(n\ell)<A_R$ for high ℓ, with the result that $A_I(nk)>A_R$ for all k. Thus in an electric field $N_>$ increases, thereby increasing the DR rate. In an E field m is still a good quantum number, and the high-m states do not contribute to the DR rate since they are all high-ℓ states. If $A_I(nk)\gg A_R$ for the low-m states then mixing of the m states when $B\perp E$ leads to a further increase in the DR rate. Of course, if $B\|E$ the B field has no effect since different m states cannot be coupled.

For the $B_\perp$ field to alter the DR rate, several conditions must be met. As pointed out by Robichaeux and Pindzola, it must couple Stark states differing in m by 1 [13]. We assume a hydrogenic model where the nkm Stark states are even mixtures of the zero-field nlm states and the spacing of the m and $m+1$ Stark states is given by $3nE/2Z$ where E is the electric field strength and Z is the charge of the hydrogenic ion. Then the minimum field required to mix low-m states, B_{mix}, can be written in terms of the L_x matrix element of the Stark states, which is given by

$$\langle nkm|\mu_B B_{mix}L_x|nk'm\pm 1\rangle \simeq \frac{\mu_B B_{mix}n}{4}=\frac{3nE}{2Z} \tag{4}$$

or

$$\mu_B B_{mix}=\frac{6E}{Z}. \tag{5}$$

Here μ_B is the Bohr magneton.

The magnetic coupling must also exceed the autoionization rate $A_I(nk)$ to affect the DR rate. In simple physical terms, the magnetic field must cause the atom to precess to more stable k states of higher m before it reionizes after being captured. We assume that in an electric field all states with $\ell\geq\ell_c$ are uniformly spread over the Stark manifold, which does not include the states of $\ell<\ell_c$ since their quantum defects are too large, so $A_I(nk)$ can be approximated by the average autoionization rate of the $n\ell$ states in the Stark manifold, or [10]

$$A_I(nk)=\gamma_E n^{-4}, \tag{6a}$$

where

$$\gamma_E=\sum_{\ell=\ell_c}^{n-1}\gamma(\ell). \tag{6b}$$

The requirement for the autoionization rate can then be written using Eqs. (4) and (6a) as

$$\mu_B B_{mix}=\frac{4A_I(nk)}{n}=\frac{4\gamma_E}{n^5}. \tag{7}$$

Finally, just as the magnetic coupling of Eq. (4) must exceed the autoionization rate, it must exceed the radiative decay rate, i.e.,

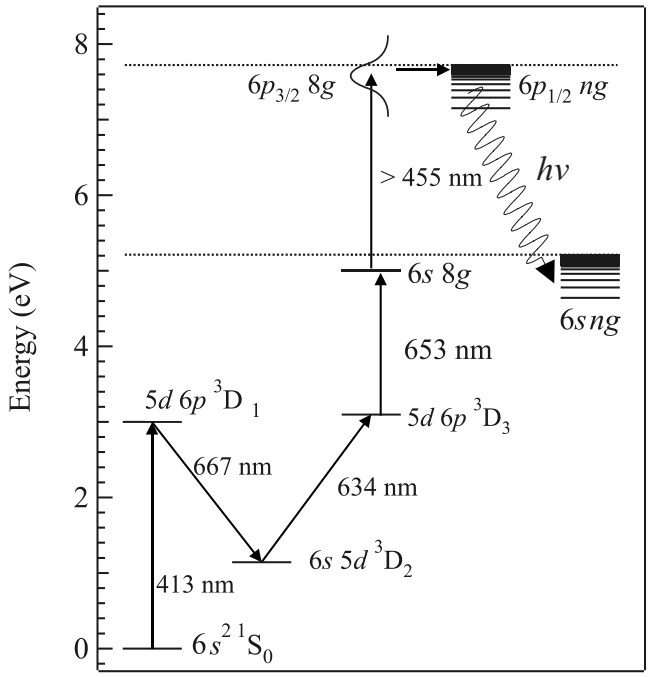

FIG. 1. Energy level diagram for DR from a continuum of finite bandwidth (CFB). The five solid arrows indicate the five laser pumping steps. The laser driving the last transition to the $6p_{3/2}8g$ CFB is typically tuned to the energy of a $6p_{1/2}ng$ state. DR occurs by "capture" from the CFB into the $6p_{1/2}ng$ states, shown by the horizontal arrow, followed by radiative decay to the bound $6sng$ state, shown by the wavy arrow.

$$\mu_B B_{mix}=\frac{4A_R}{n}. \tag{8}$$

For the states that contribute substantially to DR this requirement is necessarily lower than the one imposed by the autoionization rate in Eq. (7), and as suggested by the discussion preceding Eq. (3), it is usually the requirement of Eq. (7) that is important. The dependences of B_{mix} on n and E in Eqs. (5), (7), and (8) are obviously quite different, allowing us to determine which is most important.

Our experiment is done using not a true continuum but a continuum of finite bandwidth (CFB), the broad Ba $6p_{3/2}8g$ state that straddles the Ba^+ $6p_{1/2}$ limit [20]. As shown in Fig. 1, we use five dye laser pulses to excite Ba atoms from the ground state to a well-defined energy in the $6p_{3/2}8g$ state. The $8g$ electron collides with the Ba^+ $6p_{3/2}$ ion core and drives the quadrupole transition to the $6p_{1/2}$ state. The $8g$ electron gains energy in this superelastic collision and is "captured" into the Ba $6p_{1/2}ng$ state. If an atom in this state radiatively decays to the bound $6sng$ state, DR has occurred, which we detect by field ionization of the bound $6sng$ atom. An important aspect of using the $6p_{3/2}8g$ state as the CFB is that DR occurs via the $6p_{1/2}ng$ states or the $6p_{1/2}ng$ component of the $6p_{1/2}nk$ Stark states. Since the $6p_{1/2}ng$ states have a small quantum defect of $\delta=0.02$, very small E fields, ~5 V/cm, are required to mix them with the $6p_{1/2}n\ell$ states of higher ℓ to produce E field enhancement of the DR rates. The relative effect of an electric field scales as n^5, so the

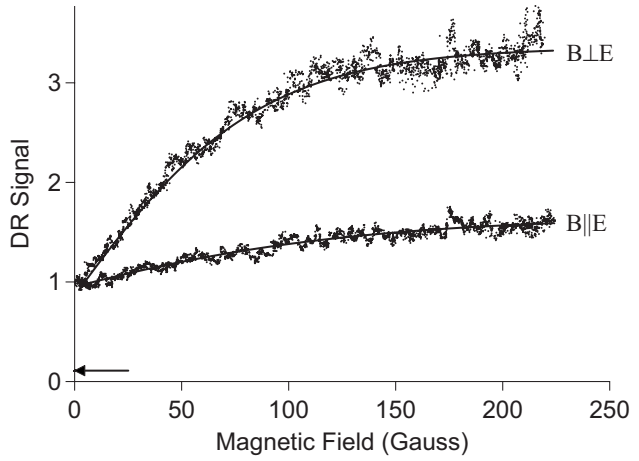

FIG. 2. DR signals obtained via the $6p_{1/2}42k$ states in an E field of 3.3 V/cm with $B\|E$ and $B\perp E$. The signals are normalized to indicate the magnetic field enhancement factor for an electric field of E=3.3 V/cm. With $B\|E$ there is little enhancement, but with $B\perp E$ the magnetic field produces a maximum enhancement factor of 3. The arrow shows the signal obtained when both E and B are 0, indicating that the E field alone produces a tenfold enhancement.

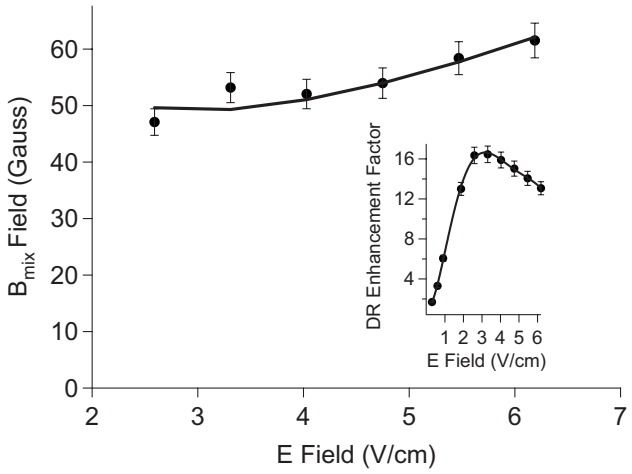

FIG. 3. Experimental (●) values of B_{mix} obtained vs E for n =42 with $E\perp B$ and the values calculated from the model of Eq. (9) (—), which takes into account the dependence of the autoionization rate and the DR rate, shown in the inset, on the E field alone. The theoretical curve is normalized to the data.

5 V/cm fields we use have negligible effect on the $6p_{1/2}8g$ state. However, the bound $6sng$ states are converted to $6snk$ Stark states analogous to the $6p_{1/2}nk$ states. What we observe is analogous to forced autoionization [21], which implies that the external fields do not affect DR through low-lying autoionizing states.

In the experiment a thermal beam passes down the axis of a set of four rods which are 0.24 cm in diameter and spaced vertically and horizontally by 1.00 cm [22]. Applying a voltage to the right two rods produces a horizontal E field, and applying the voltage to the bottom two rods produces a vertical E field. The vertical magnetic field is provided by a pair of 54-turn Helmholtz coils of 5.08 cm diameter spaced 2.54 cm apart. We discharge a 5 μF capacitor through the coils, producing a current pulse 100 μs long, leading to peak field of 250 G, which we have calibrated using a gaussmeter. The Nd:YAG (yttriuum aluminum garnet) laser pumping the dye lasers is fired at the peak of the magnetic field pulse. The negative voltage field ionization pulse is applied to the lower pair of rods 1 μs after the laser pulses, and the resulting electrons are ejected vertically, along the B field, and strike a microchannel plate (MCP) detector. We record the signal from the MCP with a gated integrator. We set the last laser to the energy of a specific $6p_{1/2}ng$ state and record the field ionization, or DR, signal as the magnetic field is swept over many shots of the laser, with a fixed parallel or perpendicular E field.

Figure 2, obtained with the laser tuned to the energy of the $6p_{1/2}42g$ state, is typical of our data. We show the DR signals obtained by scanning the field from 0 to 250 G with $E\|B$ and $E\perp B$ for E=3.3 V/cm. For reference, the signal obtained with B=0 and E=0 is indicated on the graph by the arrow. For B=0 and E=3.3 V/cm there is approximately a factor of 10 increase from the signal with E=0 and B=0. When $E\perp B$ the enhancement factor, relative to the B=0 signal, increases rapidly and saturates at a maximum enhancement factor greater than 3. For $E\|B$ there is minimal enhancement. A diamagnetic enhancement is possible [15], but should not be as large as that seen in Fig. 2 for $E\|B$. The apparent enhancement we observe for $E\|B$ could be due to stray electric fields, 50 mV/cm, or motional electric fields, 100 mV/cm at the highest B field, producing a component of $E\perp B$. It is also possible that the magnetic field increases the collection efficiency for the electrons from field ionization. In any case, the effect of $E\|B$ is minimal compared to the increase in the signal when $E\perp B$.

In the absence of a more sophisticated model it is reasonable to take the field at which the $B_\perp$ field produces half of its maximum enhancement as B_{mix}. For Fig. 2, B_{mix}=53(5) G. We have made measurements similar to the ones shown in Fig. 2 for n=32, 37, 42, and 47, for $2<E<7$ V/cm.

Before we consider the n dependence, it is useful to examine the dependence of B_{mix} on E for a given n state, and in Fig. 3 we show B_{mix} vs E for n=42. As shown, B_{mix} increases with E, which suggests that the magnetic field might need to be increased to match the increase in the Stark splitting. However, B_{mix} is certainly not proportional to E as in Eq. (5). In fact, the increase in B_{mix} with E is due to the fact that as the E field is raised beyond 3 V/cm the $\ell<4$ states join the truncated Stark manifold, raising the autoionization rate and reducing the DR rate, as shown in the inset of Fig. 3 [23]. At the peak of the DR rate enhancement $A_I(nk)\simeq 0.06n^{-4}$ [24], which occurs at $E\simeq 2.5$ V/cm, but at the Inglis-Teller field of E=13.1 V/cm all ℓ states are mixed into the Stark manifold and $A_I(nk)\simeq 0.31n^{-4}$ [25]. The change in the autoionization rate substantially alters the DR rate, and we can take this phenomenon into account by writing a variant of Eq. (2) to express the DR rate $\Gamma(nk)$ via the nk Stark state from the CFB. Explicitly,

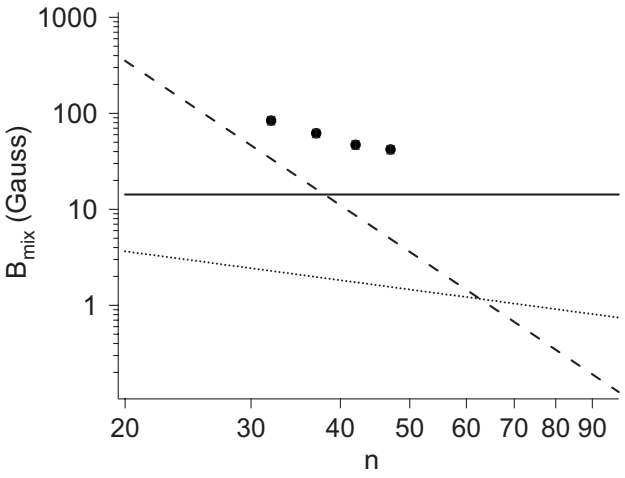

FIG. 4. Requirements for B field enhancement of the DR rate vs n of the intermediate $6p_{1/2}nk$ states. From the $\Delta m=1$ Stark splitting for Eq. (5) (—), from the autoionization rate Eq. (7) (---), and from the radiative decay rate Eq. (8) (···) of the $6p_{1/2}nk$ states. The experimental values of B_{mix} (●) obtained at $E=2.6$ V/cm indicate that both the requirements of Eqs. (5) and (7) are important. The uncertainty in B is indicated by the size of the data points.

$$\Gamma_{nk} = R(nk)\frac{A_R}{R(nk) + A_I(nk) + A_R}, \tag{9}$$

where $R(nk)$ is the coupling of the $6p_{3/2}8g$ state to the $6p_{1/2}nk$ Stark states. In this experiment, $A_R=3.88\times10^{-9}$ [26]. $R(nk)=0.02n^{-4}$ [27] and $A_I(nk)\geqslant 0.06n^{-4}$. In this context, $A_I(nk)$ represents the autoionization rate into all continua other than the continuum of finite bandwidth. Because $A_I(nk)\gg R(nk)\gg A_R$, $\Gamma_{nk}\propto 1/A_I(nk)$, and we can extract the relative autoionization rate from the DR rate in electric fields. In Fig. 3 we show $1/A_I(nk)$ normalized to our experimental data points. It is evident that the increase of B_{mix} with E is due solely to the increase in the autoionization rate with E.

Confirmation that the B field requirement is highly dependent on the autoionization rate is obtained by examining the n dependence of the magnetic field required for enhancement. In Fig. 4 we plot the magnetic field requirements imposed by Eqs. (5), (7), and (8). We use the radiative rate $A_R=3.88\times10^{-9}$ for Eq. (8), and we use $\gamma_E=0.06$ for Eq. (7), which is the correct value for the $m\leqslant 4$ states of the truncated Stark manifold. For comparison, we show our experimental values for B_{mix} obtained at $E=2.6$ V/cm.

It is evident that the experimental points for B_{mix} lie substantially above the line for Eq. (5) at 2.6 V/cm and are not n independent. At the same time the n dependence of the data is certainly not the n^{-5} dependence expected from Eq. (7). It appears that the data are consistent with B_{mix} needing to meet both the requirements of Eqs. (5) and (7). The requirement of Eq. (8) is easily met.

In conclusion, these measurements show clearly that several criteria must be met by the magnetic field to enhance the DR rate in the presence of a perpendicular electric field. In particular, not only must the magnetic coupling exceed the Stark splitting between adjacent Rydberg states, but it must exceed the total energy width of these states as well.

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