Field Effects on the Rydberg Product-State Distribution from Dielectronic Recombination

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The effects of state mixing by extrinsic fields in the collision region have been investigated for the dielectronic recombination process $Mg^+(3s)+e \rightarrow Mg(3p,nl) \rightarrow Mg(3s,nl)+h\nu$. By field ionization of the Rydberg atoms produced, cross sections $\sigma(n_f)$ have been measured. The observed large changes of $\sigma(n_f)$ with alteration of the extrinsic field provide the first incontrovertible experimental evidence that dielectronic recombination can be changed by external fields.

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Dielectronic recombination (DR) has been an object of great attention during the past three years. The process is the resonant capture of an electron by an ion to a doubly excited state of a once-less-charged ion followed by radiative stabilization. The recent interest is ascribable in large measure to the first direct observations and measurements[1-4] of DR in colliding-beam experiments after twenty years[5] of theoretical development, and recognition of the importance of the process in the modeling of high-temperature plasmas. Even keener excitement was elicited by the fact that all the experimental cross-section results were substantially larger (by factors of several times) than predicted by conventional theory.

This disparity between experiment and theory evoked a flurry of effort[6] to reexamine the basic theoretical approaches to the problem, the approximations used, the coupling schemes, the inclusion of fine-structure effects, and various details of the collision. It was shortly recognized,[7] however, that earlier workers[8] had predicted an enhancement of DR cross sections in the presence of an electric field; and the hypothesis was advanced that this phenomenon could explain the incongruous results.

With the effects of fields as the paramount issue, renewed fervor has brought good progress[7,9] in the theory. On the experimental side, significant data[10] came from plasma experiments on DR and from heavy-particle experiments on the DR-related process of resonant transfer and excitation. Both of these experimental situations involved transitions which should be insensitive to fields—and agreement with theory was found. Merged-beam measurements on additional species were made at Oak Ridge National Laboratory, and earlier measurements[3] there were redone with improved techniques.

Regrettably, for all the DR beam experiments except that on Mg^+, the fields in the collision region have not been directly measurable nor calculable, but have had to be guessed or estimated. By making "proper" guesses (and for the Mg^+ case, by using the known value) for fields and invoking theory for the DR process in the presence of a field (DRF), all of the experimental results have been "rationalized" with theoretical results.[9,11,12] It is now becoming generally accepted that fields can play a major role in DR—a concept advanced by Jacobs *et al.*[8] nearly ten years ago.

Despite the evolving acceptance of DRF as important and different from DR, the glaring fact stands out that there is really no incontrovertible experimental verification of DRF. There have been no experiments where the fields in the collision region were known and varied to produce measurable changes in the DRF cross sections. The purpose of this paper is to present new experiments which do constitute compelling evidence for DRF.

First we review with the reader some approximate physical arguments providing a perspective of why DRF is different from DR. The cross section for dielectronic recombination can be written $\sigma=\sigma_c[A_r/(A_r+A_a)]$, where σ_c is the capture cross section and the bracketed quantity represents the branching ratio between radiative stabilization of the core excited electron (rate A_r) and autoionization (rate A_a). With recognition of the relationship between σ_c and A_a due to detailed balancing, this becomes for capture into a state (nl),

$$\sigma_{nl}=\sigma_0 2(2l+1)\{A_a(n,l)A_r[A_a(n,l)+A_r]\},$$

where σ_0 involves statistical weights of the initial state and the electron core after capture, the resonance energy width, and various constants, and where $2(2l+1)$ is the statistical weight of the final Rydberg state (n,l).

Now, generally A_r is nearly constant with n and l, since the high Rydberg electron is just a spectator to the core transition, while $A_a(n,l)$ varies as $1/n^3$. For low n's and l's, $A_a \gg A_r$; numbers may be $A_a \sim 10^{14}$ s^{-1}, $A_r \sim 10^8$ s^{-1}. Under these conditions $\sigma(n,l) \propto A_r$. At high enough n's, $A_a = A_r$, and beyond that,

$A_r > A_a$, so that $\sigma(n,l) \propto A_a(n,l) \propto 1/n^3$. Similarly, $A_a(n,l)$ decreases rapidly with l [e.g., $A_a \propto \exp(-al^2)$, where a is a constant]. The total DR cross section is a sum over the resonances covering the combinations of n and l. For a given (low to moderate) n, and for low l, again $A_a >> A_r$, so that $\sigma(n,l) \propto (2l+1)A_r$, and for high l, $\sigma(n,l) \propto (2l+1)\exp(-al^2)$, becoming negligible for $l > l_c$, where $A_a(n,l_c) \approx A_r$. Thus, though the number of resonances which could contribute to DR increases as $\sum_{l=0}^{n-1} 2(2l+1) = 2n^2$, only l's for $l \leq l_c$ will typically contribute, and states with $l_g > l_c$ form a substantial reservoir which, if their autoionization rates could be increased, could contribute to DR, thus enhancing the cross section.

The effective increase of $A_a(n,l_g)$ is precisely the role played by an electric field. The field mixes states with l lower and higher than l_c, and since generally $A_a(n,l)$ for low l is so much greater than A_r, the net effect is $A_a > A_r$ for many more of the Stark states than the pure spherical states. Thus, the electric field provides a "knob" on the reaction by tuning in and out the number of states with $A_a > A_r$—and hence this knob tunes the size of the cross section.

In the experiments reported here, DRF cross sections σ have been measured as a function of electron energy for a fixed and selected "principal quantum number," and these measurements have been done for two values of electric field in the collision region. Also, at the same two fields and for fixed electron energy (where σ is a maximum), σ has been measured as a function of "principal quantum number"; and some data were obtained at a third value of electric field.

The experiments were performed for the process $Mg^+(3s) \rightleftarrows Mg^{**}(3p,n_f) \rightarrow Mg^*(3s,n_f) + h\nu$ with the apparatus illustrated schematically in Fig. 1. A beam of mass-selected $^{24}Mg^+$ ions (2 keV, ~ 300 nA) is crossed by a magnetically confined beam of electrons (~ 25 μA, ~ 0.5 eV energy width). The electric field in the collision region is $\mathbf{E}_c = \mathbf{v}_i \times \mathbf{B}$, and can be varied by a change of the magnetic field B or the ion velocity v_i.

Both E_c and an applied field E_s downstream from the interaction serve to separate the product Rydberg atoms from the ions which are directed into a Faraday cup as shown. Product Rydberg atoms continue in the z direction into a Rydberg-state analyzer with an electric field E_l in the x-z plane (shown rotated 90° in Fig. 1) given by $\mathbf{E}_l = (V/\beta R)\hat{\theta}$, where $\pm V$ is the voltage applied to plates at an angle 2β with respect to each other, and R is the distance from the apex of the wedge. The spatially varying field leads to Rydberg atoms in different states being field ionized at different locations, and thus electrons (or ions, depending on polarity of V) from given quantum states will hit or miss a detector, depending on the values of V and β. In Fig. 1, electrons from $n_f = 44$–48 are shown hitting the detector.

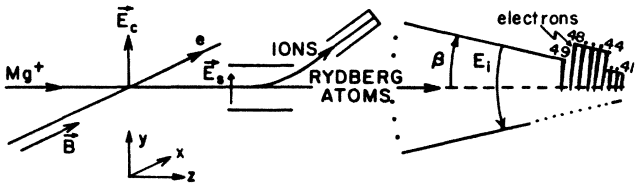

FIG. 1. Schematic of the experimental apparatus.

More experimental details will be published separately, but the measurements were carried out with use of well-established crossed-beam procedures. Numerous possible systematic errors were tested for and—where found—were minimized to insignificance and/or corrected for. Cross sections were calculated from

$$\sigma(n_f) = \frac{[N(n_f)/T]e^2 v_i v_e F}{I_e I_i (v_e^2 + v_i^2)^{1/2} \epsilon \Delta n_f}, \quad (1)$$

where Δn_f is the range of n_f for which resultant electrons (or ions) hit the detector as determined from particle-trajectory calculations, $N(n_f)$ is the number of signal counts at n_f for time T, v_i and v_e are ion and electron velocities, F is the beam overlap form factor, I_i and I_e are the beam currents, and ϵ is the absolutely measured detector efficiency.

To this point n_f has been used freely without a definition of it, as if it were the same as the principal quantum number n. Such is not the case; instead n_f is basically a number identifying the electric field value at which a Rydberg atom is field ionized. The classical saddle-point field-ionization relationship is used here, with E_i in volts per centimeter,

$$n_f = (3.2 \times 10^8 / E_i)^{1/4}. \quad (2)$$

In a real atom the presence of an electric field leads to Stark splitting, and the field-ionization lifetime $\tau(n_1,n_2,m,n)$ depends on the Stark state quantum numbers n_1, n_2, m, n and on the electric field E_i. Assuming hydrogenic states, and assuming a statistical population of all Stark levels, we calculated the detection probability of our detector versus n, and found $\bar{n} = 1.23 n_f$ for the mean value of n detected, and $\Delta n = 0.0455 n_f$ (compared to $\Delta n_f = 0.0369 n_f$) for the detector width in n. Thus, an expression analogous to Eq. (2), $n = (7.3 \times 10^8 / E_i)^{1/4}$ could have been chosen to index the field (possibly) more directly to n, but the assumptions of hydrogenlike atoms and statistical state populations are special and not necessarily correct. Thus, to emphasize the fact that any theoretical result must be "mapped" to compare with experiment, the simple field "index" n_f was chosen. The Stark state populations within an n manifold may be altered some

as the atoms traverse the fields and field gradients between the collision region and detector. Thus, for detailed mapping of theory to experiment, this field distribution may be important. It has already been presented elsewhere (see Ref. 9, p. 414) and will also be amplified upon when experimental details of this experiment are published separately.

With the techniques outlined above, we measured DRF cross sections $\sigma(n_f=33)$ as a function of electron energy at $E_c=23.5$ V cm^{-1} and $E_c=7.24$ V cm^{-1} and the results are shown in Fig. 2. The energy width of the electron beam for the higher field E_c was $\Delta E_e=0.52$ eV, while for the lower E_c it was $\Delta E_e=0.48$ eV, the difference being due to different space-charge potentials in the electron beam. The uncertainties given in Fig. 2 are statistical standard deviations only. Systematic uncertainties resulting from possible errors in the measurements of detection efficiency, total particle currents, ion velocity, bin width Δn_f, and other sources are the same for both measurements and amount to about $\pm 8\%$.

The data displayed in Fig. 2 show a large difference in the DRF cross sections obtained for the two different extrinsic fields E_c. The strength of the $n_f=33$ resonance represented by

$$S(n_f)=\int\sigma(n_f=33,\ E_e)\,dE_e$$
$$=\sigma(n_f=33,\ 4.43\text{ eV})\Delta E_e$$

is 3.70×10^{-19} cm^2 eV for $E_c=23.5$ V cm^{-1} and is 2.23×10^{-19} cm^2 eV for $E_c=7.24$ V cm^{-1}. The ratio is 1.66, and the total uncertainty of these integral values is less than $\pm 10\%$.

We also measured the resonance strength as defined above versus n_f, and the results are presented in Fig. 3. The data show that not only are the magnitudes of DRF cross sections different when different extrinsic fields are applied, but also the final Rydberg state distributions are changed. For the fields E_c used here the cross sections for the lower Rydberg states appear to increase rapidly with E_c [a dependence $S(33)\propto E_c^{1/2}$ is a reasonable estimate for $n_f=33$], whereas for the high Rydberg states, no significant changes are observed. This trend is protracted with two separate absolute measurements of resonance strength for $n_f=18$ and $n_f=33$ at an extrinsic field $E_c=3.62$ V cm^{-1}. The data are in arrant contrast to the dot-dashed curve, which is the DR (no fields) cross section calculated by Burgess[13] and mapped to the n_f coordinate by use of the hydrogenic-statistical assumption discussed earlier. The cutoff at high n_f is determined by field ionization by the separation field E_s (see Fig. 1). From the classical viewpoint for $E_s=36$ V/cm, Rydberg atoms with $n_f>55$ will be "lost" in the separator—consistent with the observations.

The observed dependences in Fig. 3 are concordant with the general theoretical predictions of LaGattuta, Nasser, and Hahn[11] and of Bottcher, Griffin, and Pindozola.[12] Both groups have shown that their theoretical predictions can be mapped to the n_f coordinate to give agreement with these measurements if suitable *assumptions* are made about the populations within a Stark manifold. Whether these assumptions follow physically from field-time histories of the particles is a theoretical mapping issue still not addressed in full detail.

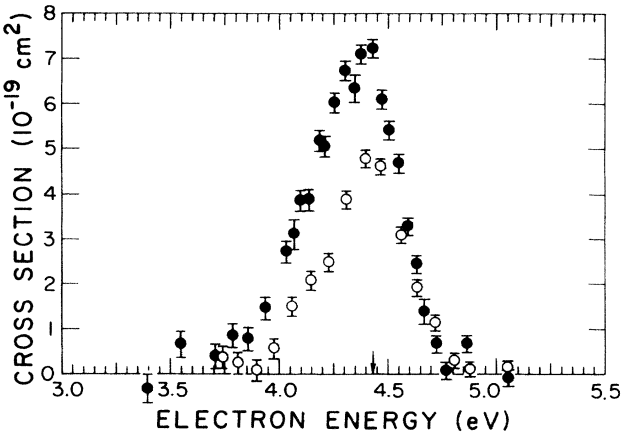

FIG. 2. Cross section $\sigma(n_f=33)$ vs electron energy for two different extrinsic fields: solid circles, $E_c=23.5$ V cm^{-1}; open circles, $E_c=7.24$ V cm^{-1}. Uncertainties are one statistical standard deviation. The arrow indicates the excitation threshold energy.

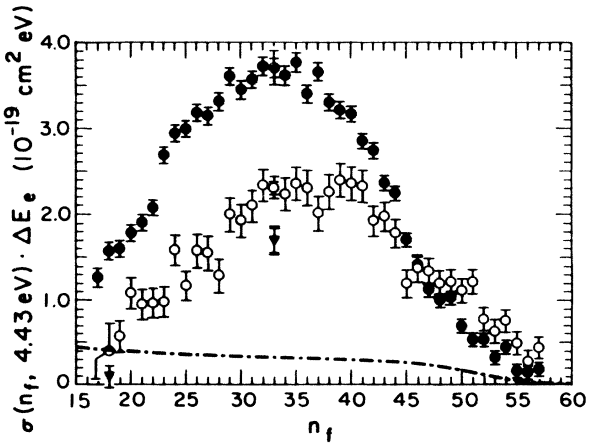

FIG. 3. Resonance strength $S(n_f)$ (see text) vs field-ionization quantum number n_f [see Eq. (2)]. Solid circles, $E_c=23.5$ V cm^{-1}; open circles, $E_c=7.24$ V cm^{-1}; triangles, $E_c=3.62$ V cm^{-1}. The curve is the DR (no fields) calculation of Burgess (Ref. 13) mapped to n_f. Uncertainties are one statistical standard deviation. For $n_f=33$, the relative uncertainties are also indicated. Absolute uncertainties at $n_f=33$ are less than 10%.

Summing the measured $S(n_f)$ values from $n_f = 18$ to $n_f = 57$ yields 8.94×10^{-18} cm^2 eV at $E_c = 23.5$ V cm^{-1} and 5.78×10^{-18} cm^2 eV at $E_c = 7.24$ V cm^{-1}. By extrapolation of the distribution to account for n_f below $n_f = 17$ additional contributions can be estimated for the total. At $E_c = 23.5$ V cm^{-1}, the resulting total $S = \Sigma_{n_f} S(n_f)$ is 9.5×10^{-18} cm^2 eV, which can be compared to the previous photon-neutral coincidence experiment[2] that yielded $(1.1\times10^{-17}$ cm$^2)\times(0.3$ eV$) = 3.3\times10^{-18}$ cm^2 eV. There is significant uncertainty in "integrating" the cross section from the previous experiment to get S, and this has been approximated simply by multiplying by the 0.3-eV width of the e beam. Apart from that, the big uncertainty in the early measurement was in the *estimated* efficiency of the Rydberg detector, which was taken to be 0.65, a factor of 2.32 higher than *measured* for ions on the detector. With use of the efficiency measured for ions, a resonance strength of 7.7×10^{-18} cm^2 eV results, in modest agreement with the present $S = 9.5\times10^{-18}$ cm^2 eV where *all* efficiencies were measured. LaGattuta, Nasser, and Hahn[11] obtain theoretical values of 7.4×10^{-18} cm^2 eV and 6.3×10^{-18} cm^2 eV at E_c's of 24 and 8 V cm, respectively. The latter number compares with the presently measured value (including an estimated 0.4×10^{-18} cm^2 eV for $n_f < 18$) of 6.2×10^{-18} cm^2 eV. In all cases, agreement seems to be good.

The results presented here constitute the only compelling experimental evidence for DRF, a process proffered nearly ten years ago but seemingly only recently receiving acceptance. Though many states (e.g., $\Delta n_{\text{core}} = 1$) may not be sensitive to fields, it is incumbent on experimentalists to design future DR (or DRF) experiments to make the fields well known—and preferably variable. Plasma workers must seriously consider the effects of plasma microfields as emphasized by Jacobs, Davis, and Kepple,[8] as well as ambient magnetic (as in fusion devices) and electric fields.

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