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TRENDS IN THE ELECTRIC FIELD ENHANCEMENT OF DIELECTRONIC RECOMBINATION CROSS SECTIONS*

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Abstract The effect of external electric fields on the dielectronic recombination cross section of selected ions has been studied in the configuration-average, distorted wave approximation. By applying the linear-Stark approximation to the doubly-excited Rydberg states formed from resonant recombination, we examine the systematics of field-mixing effects on dielectronic recombination and determine the maximum field enhancement of dielectronic recombination cross sections.

INTRODUCTION

Dielectronic recombination (DR), being a resonant process involving the formation of doubly-excited Rydberg states, is quite sensitive to the presence of external electric fields. For example, in the case of the dielectronic recombination reaction associated with the 2s → 2p excitation in the Li-like ions, the continuum electron collisionally excites the N electron ion in its 2s ground-state configuration and is simultaneously captured (resonant recombination) into the doubly-excited $2pn\ell$ configuration:

$$e^- + X_{Z,N}(2s) \rightarrow X_{Z,N+1}(2pn\ell). \qquad (1)$$

The N+1 electron ion can autoionize back to the ground-state configuration of the N electron ion. However, it may also decay to an excited, bound state of the N+1

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electron ion through one of two possible types of radiative transitions

$$X_{Z,N+1}(2pn\ell) \rightarrow X_{Z,N+1}(2sn\ell) + h\nu \ , \tag{2}$$

$$X_{Z,N+1}(2pn\ell) \rightarrow X_{Z,N+1}(2pn'\ell') + h\nu \ , \tag{3}$$

thereby completing the DR process.

When an external electric field is applied, the DR reaction can be affected in two ways. First of all, the field can ionize the high Rydberg states, and in so doing, reduce the DR cross section. Secondly, the electric field can cause a redistribution of the angular momentum among the $2pn\ell$ Rydberg states, which increases the number of such states for which the resonant recombination rates are appreciable, and thereby, enhances the DR cross section. Comparison of experiment and theory[1,2] indicates that this effect is substantial, at least in low stages of ionization.

THEORETICAL CONSIDERATIONS

For DR transitions of the type discussed above, reasonably accurate values of the cross section can be obtained from the configuration-average (CA) approximation.[3,4] In this approximation, the energy-averaged cross section for DR transitions through all doubly-excited states within the configuration with an $n\ell$ Rydberg electron is given by the expression:

$$\sigma_{CA} = \frac{4.95 \times 10^{-30}}{\Delta\varepsilon \ \varepsilon_i} \frac{G_C}{2G_I} (4\ell+2) \frac{A_a(n,\ell) \cdot A_r(n,\ell)}{A_a(n,\ell) + A_r(n,\ell)} \ . \tag{4}$$

In this equation, $\Delta\varepsilon$ is an energy bin width chosen to be larger than the largest resonance width; ε_i is the energy of the continuum electron; G_C is the statistical weight of the core electrons within the doubly-excited configuration; G_I is the statistical weight of the initial configuration; the 2 in the denominator is the intrinsic statistical weight of the continuum electron; and $A_a(n, \ell)$ and $A_r(n, \ell)$ are the autoionizing rate and the radiative rate, respectively, averaged over all states of the doubly-excited configurations. The energies are in units of eV and the rates are in $\sec^{-1}$. Expressions for the rates are given in Ref. 3.

The CA approximation will overestimate the DR cross section for cases in which the autoionizing rates and the radiative rates for individual levels are comparable in magnitude. However, this is only true for a limited range of n and ℓ, and for the cases considered here it should provide cross sections in fair agreement with those calculated in intermediate coupling.[3]

The effects of field ionization may be approximated by including only those doubly-excited states up to some maximum value of the principal quantum number, n_{max}. The value of n_{max} can be estimated from various approximate formulae[1,5] which depend on the stage of ionization and the electric field. A discussion of the accuracy of this approach is given in Ref. 4.

In high members of the Rydberg series of doubly-excited states, configurations for which the Rydberg electron has the same value of n but different values of ℓ become nearly degenerate. This is especially true for configurations with high values of ℓ; but as n increases, it becomes nearly true for configurations with lower values of ℓ. Thus, it may be reasonable to estimate field-mixing effects by employing the linear-Stark approximation.

Several methods for estimating the onset of the linear-Stark region (where the separation between levels with the same value of n and different values of ℓ should be less than the energy splitting due to the electric field) as a function of n only and as a function of n and ℓ have been developed.[2,6,7] However, the amount of field mixing depends on the details of the atomic structure, and should be determined by diagonalizing a Hamiltonian matrix which includes the internal electrostatic and spin-orbit terms as well as the Stark matrix elements. Until this is done, it may be more meaningful to assume that the resonant doubly-excited configurations are all within the linear-Stark region. This does not allow one to study field mixing as a function of field strength; however, it does provide physical insight into field-mixing effects, and allows one to examine important trends in the maximum field enhancement of the DR cross sections.

In the linear-Stark approximation, the wavefunction for the Rydberg electron in an external electric field can be determined from a simple Clebsch-Gordon transformation from spherical to parabolic coordinates.[8] In the resulting Stark representation, the Rydberg electron states are characterized by the quantum numbers n, k and m, where k is the electric quantum number. It is defined in terms of the parabolic quantum numbers n_1 and n_2 as $k=n_1-n_2$, where $n_1+n_2+|m|+1=n$.

When exchange is neglected, the CA autoionizing rates transform to the Stark representation according to the equation:

$$A_a(n,k,m) = \sum_{\ell=|m|}^{n-1} \left| C^{\;(n-1)/2\;\;(n-1)/2\;\;\ell}_{\;(m-k)/2\;\;(m+k)/2\;\;m} \right|^2 A_a(n,\ell), \quad (5)$$

where C is the Clebsch-Gordon coefficient. This equation is exact for the p $\rightarrow$ s autoionizing transitions considered here, even when exchange is included.

The radiative rates associated with transitions of the type shown in Eq. (2) are nearly independent of n and ℓ; thus in the Stark representation, they are nearly independent of n, k and m. However, this is not true of transitions of the second type shown in Eq. (3). For low stages of ionization, this type of radiative transition is only significant for low values of n and ℓ. Conversely, for high stages of ionization, the rates for such transitions can be large up to relatively high values of n and ℓ, and must be included in the cross section calculation. The radiative rates for transitions of this type transform to the Stark representation in a way identical to that for the autoionizing rates given in Eq. (5).

Thus we can obtain the total CA cross section for an n manifold of degenerate states in the presence of an external electric field from the equation:

$$\sigma_{CA} = \frac{4.95 \times 10^{-30}}{\Delta\varepsilon\ \varepsilon_i} \frac{G_C}{2G_I} \sum_m \sum_k 2 \frac{A_a(n,k,m)\cdot A_r(n,k,m)}{A_a(n,k,m)+A_r(n,k,m)}, \quad (6)$$

where the 2 inside the sums is the statistical weight due to the spin of the Rydberg electron. We shall apply this equation to all resonant doubly-excited configurations to determine the DR cross sections with maximum field enhancement.

SAMPLE CALCULATIONS AND RESULTS: C^{3+} and Fe^{23+}

Details regarding the calculation of autoionizing and radiative rates are provided elsewhere.[3,4] Here we first illustrate some of the ideas discussed in the last section with some sample calculations for the DR cross section associated with the 2s $\rightarrow$ 2p excitation in C^{3+}. In particular, we shall examine recombination through the doubly-excited configurations 2p4d, 2p5d and 2p20ℓ. An energy-level diagram showing the DR transitions through these configurations is given in Fig. 1.

The 2p4d and 2p5d configurations serve as examples of resonances lying relatively close to threshold for which radiative transitions of the second type (Eq. 3) are important. The variables used to calculate the cross section for these two configurations from Eq. (4) are

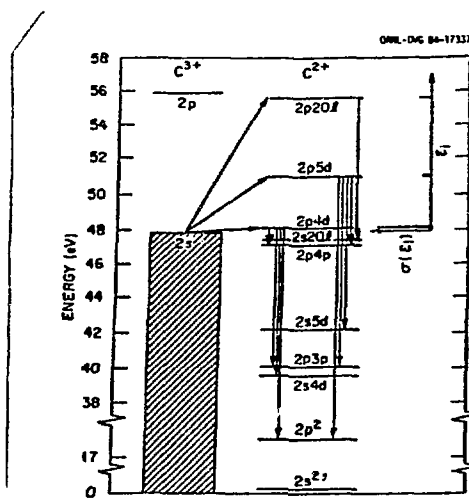

FIGURE 1. Energy level diagram showing several DR transitions in C^{3+}

given in Table I, along with the resulting cross sections. For both of these configurations, the autoionizing rates are much larger than the radiative rates so that the cross section is directly proportional to $A_r(n,\ell)$. The relatively large cross section for the 2p4d configuration is due to the large rate associated with radiative transitions of the second type, as well as the small value of ε_i. The 2p5d calculation illustrates how the DR cross section for individual configurations falls off rapidly as we move up a Rydberg series.

The calculation of the cross section for DR transitions through the manifold of nearly degenerate 2p20ℓ states shown in Table II serves as an example of relatively high Rydberg states for which only the radiative transitions of the first type (Eq. 2) are important. In the first calculation we apply Eq. (4) and sum over the $2n^2$ degenerate states. The variation of the autoionization rate with ℓ is shown in Fig. 2. We see that by ℓ = 9, the autoionizing rate has fallen below the radiative rate, and by ℓ = 10, the cross section is nearly proportional to the autoionizing rate, which is quite small.

The above considerations suggest an approximation which is the basis of the second calculation in Table II. Since $A_a(n,\ell)$ falls off rapidly with ℓ near the point where it crosses $A_r(n)$, we are justified in assuming that the cross section is proportional to $A_r(n)$ before the crossing, and negligible after the crossing. This reduces the cross section calculation to a simple count of the statistical weight of the Rydberg electron before $A_a(n,\ell)$ drops below $A_r(n)$, which for this case equals

TABLE I Calculations of DR cross sections for transitions: $C^{3+}(2s)+e^- \rightarrow C^{2+}(2pnd)$ with n=4,5.

$\Delta\varepsilon = 0.136$ eV	$G_I = 2$	$G_C = 6$
Variable	$C^{2+}(2p4d)$	$C^{2+}(2p5d)$
ε_i	0.256 eV	3.08 eV
A_a	1.27×10^{14} sec^{-1}	5.55×10^{13} sec^{-1}
A_r	3.09×10^{9} sec^{-1}	1.77×10^{9} sec^{-1}
σ_{CA}	6.59×10^{-18} cm^2	0.314×10^{-18} cm^2

TABLE II Calculation of DR Cross section for transitions: $C^{3+}(2s)+e^- \rightarrow C^{2+}(2p20\ell)$

$$\sigma_{CA} = \frac{4.95 \times 10^{-30}}{\Delta\varepsilon\ \varepsilon_i} \frac{G_C}{2G_I} \sum_{\ell} (4\ell+2) \frac{A_a(n,\ell)\cdot A_r(n,\ell)}{A_a(n,\ell)+A_r(n,\ell)}$$

$\Delta\varepsilon = 0.316$ eV $\quad \varepsilon_i = 7.75$ eV $\quad G_C = 6 \quad G_I = 2$

$A_a(20,\ell)$ varies as shown in Fig. 2

$A_r(20,\ell)$ is nearly independent of ℓ

$\approx A_r(20) = 2.80 \times 10^8$ sec^{-1}

$\sigma_{CA} \approx 0.308 \times 10^{-18}$ cm^2

$$\sigma_{CA} \approx \frac{4.95 \times 10^{-30}}{\Delta\varepsilon\ \varepsilon_i} \frac{G_C}{2G_I} A_r(20) \sum_{\ell}{}' (4\ell+2)$$

$\sum_{\ell}'$ signifies sum over ℓ up to the point where $A_a(20,\ell) < A_r(20)$

$\sum_{\ell}' (4\ell+2) = 2+6+10+14+18+22+26+30+34 = 162$

(out of a possible $2n^2 = 800$ states)

$\sigma_{CA} \approx 0.320 \times 10^{-18}$ cm^2

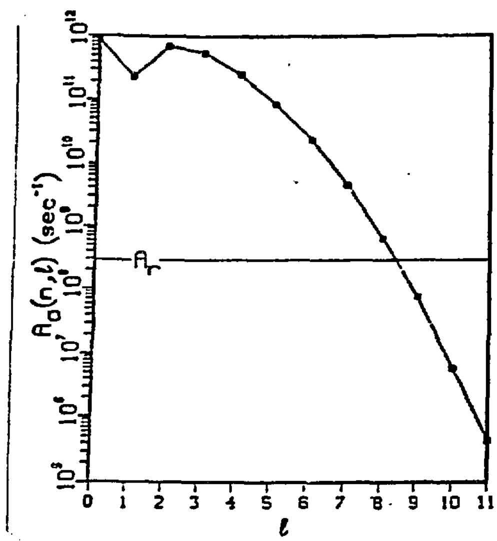

FIGURE 2. Autoionizing rates for the autoionizing transition $C^{2+}(2pn\ell) \rightarrow C^{3+}(2s) + e^-$ for n=20 as a function of ℓ. $A_r(n,\ell) = A_r(n)$ is shown by the line marked A_r.

162 out of a possible $2n^2 = 800$ states. The result of this approximate calculation is in good agreement with its more exact counterpart in the first part of the table. However, we only use this method for interpretative purposes.

In the presence of an electric field, the high Rydberg electron states with the same value of n but different values of ℓ will be mixed. This will tend to lower the autoionizing rate for lower values of ℓ, but enhance the autoionizing rate for higher values of ℓ, thereby increasing the number of states for which the autoionizing rate is greater than the radiative rate. As we have seen from our approximate calculation, this will increase the cross section. A quantitative estimate of this enhancement can be obtained by employing the linear-Stark approximation.

A sample calculation of the field enhanced cross section using Eq. (6) for the n=20 manifold of states in C^{3+} is shown in the first part of Table III. The variation of the autoionizing rate with $|m|$ and k is shown in Fig. 3. We see that by $|m|=8$, all autoionizing rates are below the radiative rate; furthermore, by $|m|=9$, nearly all the autoionizing rates are so small that the cross section is proportional to $\sum_m \sum_k A_a(n,k,m)$, and this will provide only a negligible contribution to the total cross section.

TABLE III Calculation of field-enhanced DR cross section for the transitions: $C^{3+}(2s)+e^- \rightarrow C^{2+}(2p20km)$

$\Delta\varepsilon = 0.316$ eV $\quad \varepsilon_i = 7.75$ eV $\quad G_C = 6 \quad G_I = 2$

$A_a(20,k,m)$ varies as shown in Fig. 3

$A_r(20,k,m)$ is nearly independent of k and m

$\quad\quad \approx A_r(20) = 2.80 \times 10^8\ \mathrm{sec}^{-1}$

using Eq. (6) we obtain:

$\sigma_{CA} = 0.901 \times 10^{-18}\ \mathrm{cm}^2$

$$\sigma_{CA} \approx \frac{4.95 \times 10^{-30}}{\Delta\varepsilon\ \varepsilon_i} \frac{G_C}{2G_I} A_r(20) \sum_m{}' \sum_k{}' 2$$

$\sum_m{}' \sum_k{}'$ signifies a sum over m and k up to a point where $A_a(20,k,m) < A_r(20)$

$\sum_m{}' \sum_k{}' 2 = 40+76+72+68+64+60+48+28 = 456$

$\quad\quad$ (out of a possible $2n^2 = 800$ states)

$\sigma_{CA} \approx 0.900\times10^{-18}\ \mathrm{cm}^2$

TRENDS IN ELECTRIC FIELD ENHANCEMENT OF DR

The variation of $A_a(n,m,k)$ suggests the approximation which we employ in the second part of Table III. We assume that the cross section is proportional to $A_r(n)$ for all Stark states for which $A_a(n,k,m) > A_r(n)$, and ignore the contributions from all states for which the reverse is true. Again this reduces the calculation to a count of states, but now the count is over Stark states. As we see, the approximate method agrees very well with the result of the more exact treatment in the first part of the table, and both predict a field enhancement of nearly a factor of 3. When the field is turned on, many more states become accessible to resonant recombination and the cross section increases. A measure of this increase is provided by dividing the count of states for

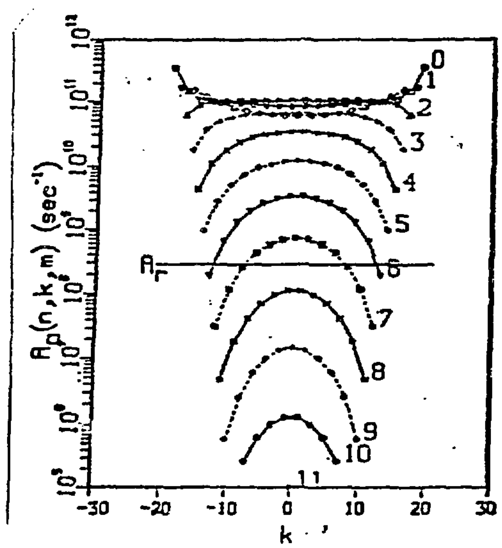

FIGURE 3. Autoionizing rates in the Stark representation for the transitions $C^{2+}(2pnkm) \rightarrow C^{3+}(2s) + e^-$ for n=20 as a function of k. Values of |m| are given to the right of each curve.

ment is more than a factor of 6, and then begins to decrease for higher values of n. The overall effect of this on the cross section with n_{max} = 50 is seen to be an enhancement of nearly a factor of 4 in the high energy peak.

Now we shall consider one of the most interesting Li-like ions, Fe^{23+}. The autoionizing rate curves in both the Stark and spherical representation for n=20 are shown in Fig. 5. Radiative transitions of the second

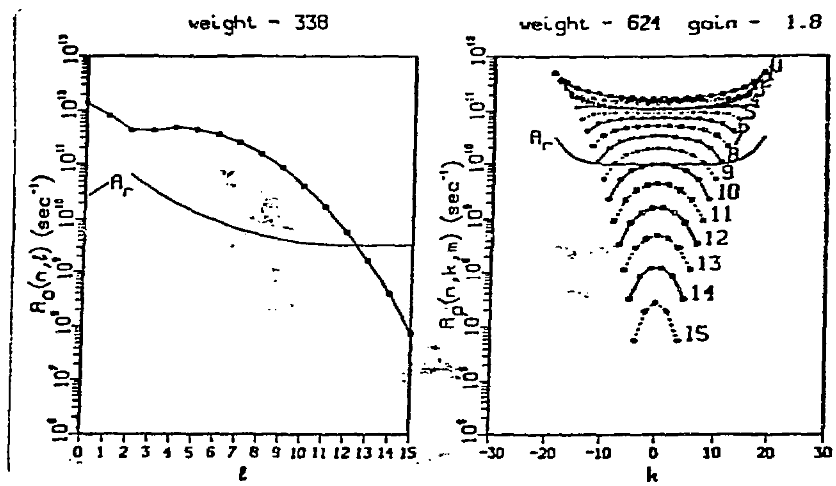

FIGURE 5. Autoionizing rate curves for autoionization from the n=20 Rydberg states of Fe^{22+} to the 2s states of Fe^{23+}. In the curves on the right the radiative rates are shown for m = 0 only; for higher values of |m|, these rates decrease and become less dependent on the electric quantum number k.

type are quite important in this highly ionized system, and as a result, the radiative rates are a function of ℓ. In the presence of a field, the radiative rates are a function of both m and k, and this will have an effect on the field enhancement.

We also notice from this figure that $A_a(n,\ell)$ drops below $A_r(n,\ell)$ at much higher values of ℓ, than in the case of C^{3+}. Thus in the absence of a field there are

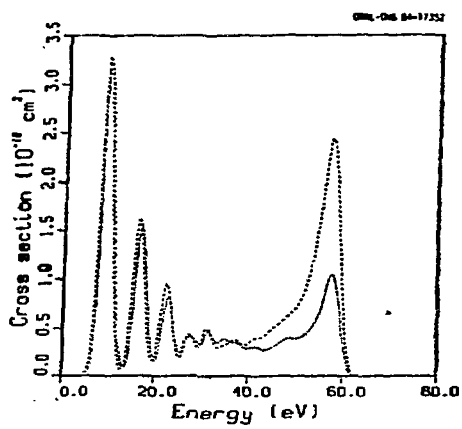

FIGURE 6. The DR cross section for Fe^{23+} including all resonances with n < 100 convoluted with a 3.0 eV FWHM Gaussian. The solid curve is with no field mixing and the dotted curve includes complete field mixing. The lowest energy peak is due to DR transitions through the n=12 manifold of doubly-excited configurations.

already many states (338 in the case of n=20) that contribute in a major way to the cross section. Thus when the field is turned on the relative increase (gain) in the number of states for which resonant recombination is appreciable is smaller than in the corresponding case in C^{3+}. As n increases the crossing of the two curves in the spherical basis occurs at a lower value of ℓ, and as a result the gain increases. However, overall, we would expect a decrease in gain, and thereby a decrease in the field enhancement of the DR cross section as we move up an isoelectronic sequence.

The cross section for Fe^{23+} with and without field mixing is shown in Fig. 6. We see that the curves are dominated by the low energy resonances near threshold. The strength of these low energy peaks is again due to large radiative rates for transitions of the second type. These rates are quite large and persist to relatively high values of n in highly ionized systems for two reasons: (1) the doubly-excited configurations $2pn\ell$ are bound up to fairly high values of n and therefore transitions of the type $2pn\ell \rightarrow 2pn'\ell'$ ($\ell'=\ell\pm1$) are allowed to high values of ℓ; (2) the radiative rate is proportional ω^3, and ω, the transition frequency, increases with ionization stage. It is obvious that such transitions must be included in the calculation of the DR cross sections or rate coefficients, especially for high stages of ionization.

We see from the figure that the low energy resonances are not affected by the electric field and the field enhancement in the high energy peak is approximately 2.3. Thus, as expected, the field enhancement of the DR cross section decreases with ionization stage, even though the number of Rydberg states included in the Fe^{23+} case is much larger than the number included in the C^{3+} calculation. Our calculated field enhancement of 2.3 for Fe^{23+} appears to be compaoable to the enhancement in the rate coefficient for this ion obtained by Jacobs et. al.[6]

CONCLUSIONS

When our field-enhanced cross sections are compared with experimental results, it is apparent that they overestimate the cross section, at least in the lower stages of ionization where experimental data are available. However, this is expected since all the doubly-excited resonant states were assumed to be mixed by the field. The actual amount of field mixing will depend on the separation between individual intermediate-coupled, doubly-excited levels and therefore will require a much more sophisticated calculation. This should be our next step in the study of dielectronic recombination.

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