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Interference effects in electron-ion recombination. I. Resonance channels only

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The Feshbach formalism is employed in investigations of the effects of interacting and/or overlapping resonances on dielectronic recombination. Model systems are considered, and the problem of singly charged target ions is discussed. Small reductions in dielectronic recombination cross sections are found to occur for these systems. Comparison is made with the results of multichannel quantum-defect theory calculations.

INTRODUCTION

Although the effect of overlapping resonances on the cross section for dielectronic recombination (DR) has attracted some attention[1-3] in the past, it has been thought that overlaps occurred but rarely and that a detailed treatment was not generally needed. It is only recently that this subject has begun to seem of a more practical significance.

Of course, it is almost always true that overlapping resonances will intrude into DR calculations if very high Rydberg states (HRS) are included for the captured electron, i.e., in those doubly excited states for which the radiative rate is much larger than the Auger rate $\Gamma_r \gg \Gamma_a$. But these cases are usually of only small importance in the calculation of a total DR cross section (σ^{DR}), for singly or few-times charged ions.[4] For large target charge (Z_I) one may find cases for which $\Gamma_r \gg \Gamma_a$, even in doubly excited states which do not include a very HRS electron,[5] e.g., for intershell transitions, where $\Gamma_r \propto Z_I^4$ while $\Gamma_a \propto Z_I^0$. However, at large Z_I, captures to very-low-lying states usually dominate the total σ^{DR}, so that the effects of overlapping resonances involving even moderately HRS's can often be neglected. Resonances involving captures to low-lying states are usually, although now always, well separated.

Lately, the importance of the enhancing effects of extrinsic electric fields (F) on σ^{DR} for low to moderate Z_I ions has been discovered.[6-12] These effects, the existence of which had been suggested earlier,[13,14] arise mainly from an electric-field-induced mixing of HRS's of fixed principal quantum number n, but different orbital angular-momentum quantum number l. The mixing is such that states evolved from large l, which play essentially no role in DR at $F=0$ (since for these, $\Gamma_a \cong 0$), can make contributions to σ^{DR} comparable to those of states evolved from small l. The overall effect of the mixing is that the number of states "accessible" to DR is increased.[9] However, as a consequence of this increase in the number of participating resonances, the probability of encountering an overlap, at any electron beam energy (e_c), also increases. Even though it remains doubtful that, for $F>0$, overlaps will be extensive and come to dominate σ^{DR} (except just below the threshold for field ionization), it seems worthwhile to recast the DR theory into a form in which the effects of overlaps can be readily determined.

The theory now in wide use for calculations of σ^{DR} was introduced originally by Feshbach[5] for nuclear-physics applications, and then later adapted for atomic scattering problems by Hahn.[16] In the following, we briefly review the Feshbach theory in its usual form, and then introduce a modification of the notation which allows one to access directly the effect of overlapping resonances in DR. We emphasize that the Feshbach theory, as originally formulated, already fully encompasses the possibility of resonance overlaps.[17] Our aim here is to clarify this point, and in so doing, to make the theory more useful for DR calculations.

An alternate approach to the calculation of σ^{DR} involves the use of multichannel quantum-defect theory[18] (MQDT). The predictions of MQDT have already been compared with those of the standard DR theory.[4] For further remarks, see the Discussion section. We note that interfering resonances have been studied only just recently for bound states in the continuum,[19] and for the autoionizing states of molecules.[20] The effects of overlapping resonances on atomic photoabsorption spectra[21] have been considered, using the theory of configuration interaction with continua.[22]

FORMALISM

We write the Hamiltonian for $N+1$ electrons and an ionic core as

$$H = H_0 + D \ , \tag{1}$$

where H_0 is the Hamiltonian for $N+1$ electrons interacting with each other via $V = \sum_{\substack{i,j \\ (i<j)}} (1/r_{ij})$, and with a nucleus of charge Z_c via $\sum_i (Z_c/r_i)$, and $D \propto \sum_i (\mathbf{r}_i \cdot \boldsymbol{\varepsilon}_i)$ is the electron-photon interaction. Following Feshbach[3] and Hahn[16] we define the idempotent projection operators P, Q, and R such that $1_{\mathrm{op}} = P+Q+R$, where 1_{op} is the unit operator, and $PQ = QP = QR = RQ = RP = PR = 0$. P projects onto states of N electrons bound, with one electron in a continuum state of the N electron ion of charge $Z_I = Z_c - N$, and no pho-

tons; Q projects onto doubly excited states of $N+1$ electrons "bound," and no photons; and R projects onto the ground and singly excited states of $N+1$ electrons bound, plus one photon. The wave function Ψ satisfies the equation

$$H\Psi=H(P+Q+R)\Psi=E\Psi=E(P+Q+R)\Psi \quad (2)$$

for the total energy E. This may be written instead as three equations, viz.,

$$\begin{aligned} &PH_0P\Psi_P+PVQ\Psi_Q=E\Psi_P \ , \\ &QH_0Q\Psi_Q+QVP\Psi_P+QDR\Psi_R=E\Psi_Q \ , \\ &RH_0R\Psi_R+RDQ\Psi_Q=E\Psi_R \ , \end{aligned} \quad (3)$$

where $\Psi_P\equiv P\Psi$, $\Psi_Q\equiv Q\Psi$, and $\Psi_R\equiv R\Psi$, and we ignore here the coupling between the R and P states, mediated by D. This coupling is not expected to be important for those HRS's which can most readily give rise to overlapping resonances. The last line in Eq. (3) may be rewritten in terms of Ψ_Q as

$$\Psi_R=(E-RH_0R)^{-1}RDQ\Psi_Q\equiv g_RRDQ\Psi_Q \ . \quad (4)$$

Similarly, the first line in Eq. (3) may be written as

$$\Psi_P=\Phi_P+(E-PH_0P)^{-1}PVQ\Psi_Q\equiv\Phi_P+g_pPVQ\Psi_Q \quad (5)$$

upon inclusion of the homogeneous solution Φ_P, i.e., $(PH_0P-E)\Phi_P=0$. If Eqs. (4) and (5) are substituted back into the second line in Eq. (3), one obtains an equation for Ψ_Q which is

$$\begin{aligned} (E-QH_0Q)\Psi_Q=&QDRg_RRDQ\Psi_Q \\ &+QVP(\Phi_P+g_PPVQ\Psi_Q) \end{aligned} \quad (6)$$

with solution

$$\begin{aligned} \Psi_Q=&(E-QH_0Q-QDRg_RRDQ \\ &-QVPg_PPVQ)^{-1}QVP\Phi_P \\ \equiv&G_QQVP\Phi_P \ . \end{aligned} \quad (7)$$

Since $\Psi_Q=Q\Psi$ is the projection onto doubly excited states of the exact system wave function Ψ, then the matrix element of the DR probability amplitude is[16]

$$M^{\mathrm{DR}}=\langle\Phi_RRDQ\Psi_Q\rangle=\langle\Phi_RRDQG_QQVP\Phi_P\rangle \quad (8)$$

with $(RH_0R-E)\Phi_R=0$. Expanding Q in eigenstates of QH_0Q as

$$Q=\sum_\alpha|\alpha\rangle\langle\alpha| \ , \quad (9)$$

where

$$QH_0Q|\alpha\rangle\equiv\varepsilon_\alpha|\alpha\rangle \ , \quad (10)$$

the DR probability, becomes

$$\begin{aligned} P^{\mathrm{DR}}=&|M^{\mathrm{DR}}|^2 \\ =&\left|\sum_\alpha\langle\Phi_RRD|\alpha\rangle\Phi_{Q\alpha}\right|^2 \\ =&\left|\left\langle\Phi_RRD\sum_\alpha|\alpha\right\rangle\left\langle\alpha|G_Q\sum_{\alpha'}|\alpha'\right\rangle\langle\alpha'|VP\Phi_P\rangle\right|^2 \end{aligned} \quad (11)$$

up to a statistical factor, where integration over incoming electron and outgoing photon momenta is understood. In a region of energies where resonances are nonoverlapping, Eq. (11) may be written as

$$\begin{aligned} P^{\mathrm{DR}}=&\sum_\alpha|\langle\Phi_RRD|\alpha\rangle\langle\alpha|G_Q|\alpha\rangle\langle\alpha|VP\Phi_P\rangle|^2 \\ \cong&\sum_\alpha|\langle\Phi_RRD|\alpha\rangle(E-\varepsilon_\alpha-\langle\alpha|DRg_RRD|\alpha\rangle \\ &-\langle\alpha|VPg_pPV|\alpha\rangle)^{-1}\langle\alpha|VP\Phi_P\rangle|^2 \\ \equiv&\sum_\alpha|\langle\Phi_RRD|\alpha\rangle G_{Q\alpha}\langle\alpha|VP\Phi_P\rangle|^2 \ , \end{aligned} \quad (12)$$

which is the usual form of the DR probability in the isolated resonance approximation (IRA).

If resonances are overlapping, then we have recourse to the following procedure. We rewrite Eq. (6) as an explicit q-component equation, where q is the dimension of the full Q space, as

$$\begin{aligned} (E-\varepsilon_\alpha)\Psi_{Q\alpha}=&\sum_\beta\langle\alpha|DRg_RRD|\beta\rangle\Psi_{Q\beta}+\langle\alpha|VP\Phi_P\rangle \\ &+\sum_\beta\langle\alpha|VPg_pPV|\beta\rangle\Psi_{Q\beta} \end{aligned} \quad (13)$$

and where β (and α) range from 1 to q. Then Eq. (7) becomes

$$\begin{aligned} \Psi_{Q\alpha}=&(E-\varepsilon_\alpha-\langle\alpha|DRg_RRD|\alpha\rangle \\ &-\langle\alpha|VPg_pPV|\alpha\rangle)^{-1} \\ &\times\left[\langle\alpha|VP\Phi_P\rangle+\sum_{\substack{\beta\\(\beta\neq\alpha)}}(\langle\alpha|VPg_pPV|\beta\rangle\right. \\ &\left.+\langle\alpha|DRg_RRD|\beta\rangle)\Psi_{Q\beta}\right] \\ \equiv&G_{Q\alpha}(\langle\alpha|VP\Phi_P\rangle+\sum_{\substack{\beta\\(\beta\neq\alpha)}}\Lambda_{\alpha\beta}\Psi_{Q\beta}) \end{aligned} \quad (14)$$

with solution

$$\Psi_{Q\alpha}=\sum_\beta(\Omega^{-1})_{\alpha\beta}G_{Q\beta}\langle\beta|VP\Phi_P\rangle \ , \quad (15)$$

where Ω^{-1} is the inverse of a matrix Ω with elements

$$\Omega_{\alpha\beta}=\delta_{\alpha\beta}-(1-\delta_{\alpha\beta})G_{Q\alpha}\Lambda_{\alpha\beta} \ . \quad (16)$$

The DR probability is then

$$P^{\mathrm{DR}}=\left|\sum_\alpha\sum_\beta\langle\Phi_RRD|\alpha\rangle(\Omega^{-1})_{\alpha\beta}G_{Q\beta}\langle\beta|VP\Phi_P\rangle\right|^2 \ , \quad (17)$$

where, again, integration over incoming electron and outgoing photon momenta is understood. We refer to this formula, which describes Q-space mixing, but with no explicit continuum-continuum coupling,[17] as the "multiple interacting resonance approximation" (MIRA). The effects of interacting continua, in the con-

text of both IRA and MIRA calculations, will be considered in a future publication.

DISCUSSION

According to Eq. (16), the $\Lambda_{\alpha\beta}$ act to couple the Q-space states to each other through common P-space and/or R-space states, whenever such common states exist. Hence, this result describes a kind of configuration interaction among the Q-space states. In this context, we note that the Q-space states are eigenstates of QHQ ($\langle\alpha\,|\,H\,|\,\beta\rangle \propto \delta_{\alpha\beta}$), so that each is, generally, already a multiconfiguration state. Presumably, the total $P^{\rm DR}$ (MIRA) for N states in the Q-space [Eq. (17)] may be either greater than or less than $P^{\rm DR}$ (IRA) for these same N states, depending on the detailed values of the $\Lambda_{\alpha\beta}$. The following special cases are of interest.

(i) We suppose that there is no coupling whatsoever between the Q-space states, i.e., $\Lambda_{\alpha\beta}=0$, for $\alpha\neq\beta$. This condition implies, from Eq. (16), that $\Omega_{\alpha\beta}=\delta_{\alpha\beta}=(\Omega^{-1})_{\alpha\beta}$. Consequently, the DR probability, from Eq. (17), reduces immediately to Eq. (11). Then, if the condition $\Lambda_{\alpha\beta}=0$ ($\alpha\neq\beta$) is again applied, Eq. (12) is obtained. This is the IRA result, independent of whether or not the N resonances overlap.

(ii) We next suppose that the $\Lambda_{\alpha\beta}$ are identical and nonzero, for all α and β. This is a particularly simple case, for which the DR probability is given by

$$P^{\rm DR}=A_a A_r\,|\,G_Q\,|^2\,|\sum_\alpha\sum_\beta(\Omega^{-1})_{\alpha\beta}\,|^2\ , \tag{18}$$

having assumed a complete degeneracy in energy, where

$$A_a\equiv -2\langle\alpha\,|\,VP\ {\rm Img}_p PV\,|\,\alpha\rangle\ ,$$

$$A_r\equiv -2\langle\alpha\,|\,DR\ {\rm Img}_R RD\,|\,\alpha\rangle\ ,$$

and

$$G_Q\equiv G_{Q\alpha}\ .$$

One has that

$$\Omega_{\alpha\beta}=\delta_{\alpha\beta}+a\,(1-\delta_{\alpha\beta})\ , \tag{19}$$

$$(\Omega^{-1})_{\alpha\beta}=\{[1+(N-2)a\,]\delta_{\alpha\beta} -a(1-\delta_{\alpha\beta})\}/[1+(N-2)a-(N-1)a^2]\ , \tag{20}$$

$$a\equiv[i(A_a+A_r)/2]G_Q =[i(A_a+A_r)/2]/[E-\varepsilon+i(A_a+A_r)/2]\ , \tag{21}$$

where, for simplicity, we have ignored the real part of $\Lambda_{\alpha\beta}$. The double sum in Eq. (18) becomes

$$\sum_\alpha\sum_\beta(\Omega^{-1})_{\alpha\beta} =\{N[1+(N-2)a\,] -N(N-1)a\,\}/[1+(N-2)a-(N-1)a^2] =N/[1+(N-1)a] \tag{22}$$

and the total DR probability for these N states is

$$P^{\rm DR}({\rm MIRA})=N^2A_aA_r/|\,(E-\varepsilon)+iN(A_a+A_r)/2\,|^2 \sim 2\pi NA_aA_r\delta(E-\varepsilon)/(A_a+A_r)\ , \tag{23}$$

which is the same as the IRA prediction. A similar result was obtained by Feshbach [Eq. 34 of Ref. 17].

(iii) We suppose again that N states in the Q space are completely degenerate in energy, the $\langle\alpha\,|\,VPg_pPV\,|\,\beta\rangle$ are identical for all α and β, the $\langle\alpha\,|\,DSg_SSD\,|\,\alpha\rangle$ are identical for all α, but now $\langle\alpha\,|\,DSg_SSD\,|\,\beta\rangle=0$, if $\alpha\neq\beta$. These conditions model the important case of DR for low Z_I target ions, where the interacting Q-space states are enumerated by n, the principal quantum number of the captured HRS electron ($\alpha=n\geq 100$). Since radiative stabilization involves essentially the inner-shell excited electron only, then $A_r\propto\delta_{nn'}$. Also, $\langle n\,|\,VP\ {\rm Img}_pPV\,|\,n'\rangle\propto 1/(nn')^{3/2}\cong 1/n^3\cong 1/n'^3$, if $1\ll N\ll n$. Reduction of Eq. (17) follows along the lines of the preceding example. The total DR probability for N states is found to be

$$P^{\rm DR}({\rm MIRA})=NA_aA_r/|\,(E-\varepsilon)+i(NA_a+A_r)/2\,|^2 \sim 2\pi NA_aA_r\delta(E-\varepsilon)/(NA_a+A_r)\ . \tag{24}$$

Comparison of Eq. (24) with the IRA result of Eq. (23) shows the following.

(a) If $A_r\ll NA_a(n)$, then $P^{\rm DR}({\rm MIRA})/P^{\rm DR}({\rm IRA})\sim 1/N$.

(b) If $A_r\cong NA_a(n)$, then $P^{\rm DR}({\rm MIRA})/P^{\rm DR}({\rm IRA})\sim 1/2$.

(c) If $A_r\gg NA_a(n)$, then $P^{\rm DR}({\rm MIRA})/P^{\rm DR}({\rm IRA})\sim 1$.

The significance of example (iii) may be understood in the light of IRA calculations of $\sigma^{\rm DR}$ performed for the Mg^{1+} target ion,[23] which have shown that, for this ion, Q-space states are nonoverlapping until values of $n\sim 100$ are reached. For larger n, one has that $A_r>A_a$, states become completely overlapped, and recourse should be had to a theory of interacting resonances. Based on the results of such a theory, and on example (iii), we predict a reduction of $P^{\rm DR}$ (MIRA), relative to the $P^{\rm DR}$ (IRA), for large n. However, the contribution of the large-n domain to the total $P^{\rm DR}$ is small, so that the overall reduction should be very small. Similar remarks should apply to the DR probability integrated over energy, denoted $\mathcal{P}^{\rm DR}$. Our preliminary calculations of $\mathcal{P}^{\rm DR}$ (MIRA) for Mg^{1+}, in *LSJ* coupling, have shown this reduction to be approximately 10%, when n is summed from 10 to 500, *if* one assumes complete degeneracy in energy of all interacting states within a subset of states $3pnl$ of fixed n, and only states of the same n are interacted. This should be close to the maximum possible reduction in $\mathcal{P}^{\rm DR}$ due to interference. More realistically, if one allows the nl states to have energies split by the appropriate quantum defect, then the calculated reduction is only $\sim 1\%$. We point out that the only restriction on those Q-space states which interact through the P space is that they be of the same parity and J value. For example, among the doubly excited states of 12 electron systems, $3p\,10d\,(^1F_3)$ and $3p\,10g\,(^3F_3)$ may interact, or $3p\,10d\,(^3P_2)$ and $3p\,10d\,(^3F_2)$ may interact.

The question then arises as to whether or not this

reduction becomes more important when a static electric field ($F>0$) is applied in the interaction region.[7–9,24] In this case, the Auger probability[9] falls off at large n as $A_a \sim 1/n^4$, i.e., faster than when $F=0$. Moreover, the number of states making significant contributions to $\mathcal{P}^{\mathrm{DR}}$ is increased[9] relative to the $F=0$ case, so that the number of overlaps may be greater. That is, the value of n beyond which A_r exceeds A_a is smaller when $F>0$ than when $F=0$, and the number of interacting states is potentially larger. Hence, the reduction of $\mathcal{P}^{\mathrm{DR}}$ (MIRA) relative to $\mathcal{P}^{\mathrm{DR}}$ (IRA) is likely also to be larger. The overall reduction in the total $\mathcal{P}^{\mathrm{DR}}$ is, however, still expected to be small since the n value at which interaction becomes appreciable is still quite large. More precise statements must await the outcome of explicit calculations, which we are now performing. We remark that the problem just discussed has nothing to do with the mixing of the eigenstates of QH_0Q induced by F, e.g., the mixing of levels corresponding to different l values for fixed n, or the mixing of levels belonging to different n values above the Inglis-Teller limit. This is a separate issue which still deserves separate consideration.

In order to illustrate the differences and similarities between the MIRA, IRA, and MQDT theories, we performed model calculations of P^{DR} (MIRA) [Eq. (17)], P^{DR} (IRA) [Eq. (12)], and P^{DR} (MQDT) (Eq. 6.76 of Ref. 18). The model was basically an extension of example (iii). It involved a single Rydberg series of resonances, and used the parameter values quoted in Fig. 1a of Ref. 18, i.e., $A_r=2\times10^{-5}$ and $A_a=0.2/n^3$ (energies in a.u.). The principal quantum number was restricted to the range $10\leqq n\leqq 65$, in order to limit computer time for the MIRA calculation. Such a restriction was not necessary with MQDT, since this approach "automatically" includes effects of the entire Rydberg series. This seems to be the primary advantage of the MQDT. Plots of P^{DR} (MIRA), P^{DR} (IRA), and P^{DR} (MQDT) versus energy appear in Figs. 1, 2, and 3, respectively.

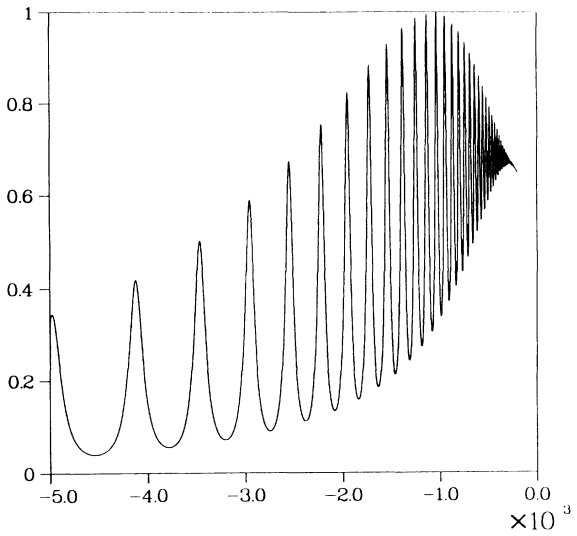

FIG. 1. P^{DR} (MIRA) vs energy, for $A_r=2\times10^{-5}$ and $A_a=0.2/n^3$, all in a.u., and with $10\leqq n\leqq 65$.

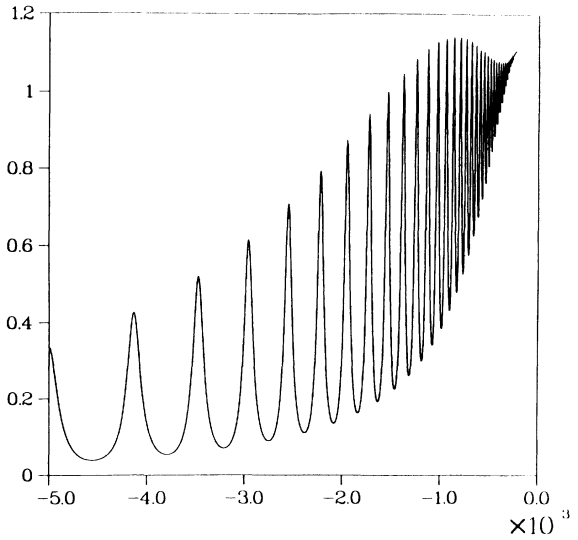

FIG. 2. Same as Fig. 1, but for P^{DR} (IRA).

From Fig. 2, it is clear that P^{DR} (IRA) does not, in general, conserve probability. In fact, one can show[18] that, as $E\to0$, P^{DR} (IRA)$\to 2\pi n^3 A_a$ which can have values greater than 1. Hence, the IRA can be a bad choice of algorithm when resonances overlap. On the other hand, the curves described by P^{DR} (MIRA) and P^{DR} (MQDT) are essentially indistinguishable, except in the neighborhood of the largest n values. This is simply a consequence of the fact that we chose to omit values of $n>65$ from the MIRA calculation. Further work showed P^{DR} (MIRA) and P^{DR} (MQDT) to be essentially indistinguishable regardless of the values chosen for A_r and A_a. But this is hardly surprising since both the MIRA and MQDT should contain all of the essential physics needed to correctly formulate this DR model problem.

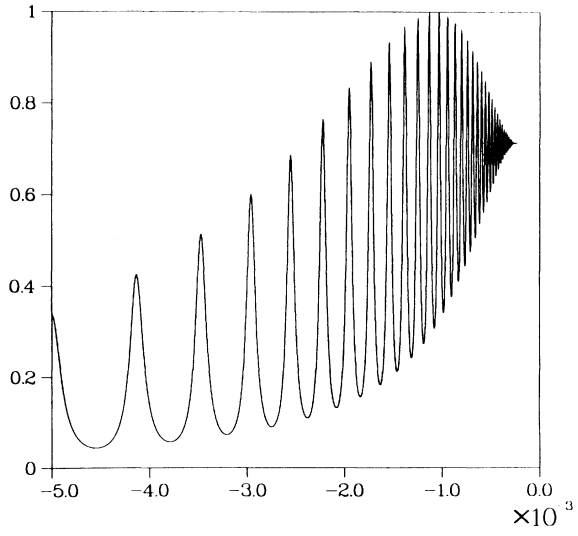

FIG. 3. P^{DR} (MQDT) vs energy, for $A_r=2\times10^{-5}$ and $A_a=0.2/n^3$, all in a.u.

Whether the MIRA or MQDT will ultimately prove to be more amenable to actual calculations involving interacting DR resonances still remains to be seen. Presumably, in the near term, the MIRA will be the preferred approach when the effects of a limited number of resonances are dominant, while the MQDT will be more successful at describing situations in which it is really essential to consider one or more entire Rydberg series. Taking a longer view, it seems likely that the MIRA and MQDT theories will be shown, in detail, to be exactly equivalent. In this case, there may be little to choose between them.

Finally, as an example of a case in which Q-space states interact through both the P and R spaces, we consider the processes

$$\begin{aligned} &1s+k_c(l_c=0)\leftrightarrow 2s^{2*}(^1S_0)\rightarrow 1s2p(^1P)+\gamma\ , \\ &1s+k_c(l_c=0)\leftrightarrow 2p^{2*}(^1S_0)\rightarrow 1s2p(^1P)+\gamma\ , \end{aligned} \tag{25}$$

where $2s^{2*}(^1S_0)$ and $2p^{2*}(^1S_0)$ are eigenstates of QH_0Q obtained as multiconfiguration states from the single configurations $2s^2$ and $2p^2(^1S_0)$. Application of Eq. (17) to the calculation of $\mathcal{P}^{\mathrm{DR}}$ (MIRA) for these processes, in LSJ coupling, shows a reduction of $\sim 5\%$ for O^{7+} targets relative to $\mathcal{P}^{\mathrm{DR}}$ (IRA), having taken realistic multiconfiguration Hartree-Fock values for the energies of the doubly excited states. If, artificially, one assumes a degeneracy of these two states, then the $\Lambda_{\alpha\beta}$ values are such as to lead to a large increase of $\mathcal{P}^{\mathrm{DR}}$ (MIRA). Hence, it is not correct to assume that interactions between DR resonances lead inevitably to decreases in $\mathcal{P}^{\mathrm{DR}}$.

Note added in proof. In the preceding examples, the approximation $\mathrm{Re}\Lambda_{\alpha\beta}=0$ was applied.

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