Validity of the independent-processes and isolated-resonance approximations for electron-ion recombination

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We examine the physics behind the continued success of the independent-processes and isolated-resonance approximations in predicting the high-resolution recombination cross sections observed in the current generation of electron-ion experiments. First-step extensions of theory to include the effects of radiative-dielectronic recombination interference, overlapping-resonance structures, and scattering-channel coupling are shown to be relatively small for these recombination experiments.

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In recent years atomic physics experiments carried out using heavy-ion traps, accelerators, and storage-cooler rings have produced high-resolution mappings of the resonance structures associated with electron-ion recombination. The experiments have been carried out using a wide range of facilities and technologies, such as the test storage ring (TSR) at Heidelberg University, the experimental storage ring (ESR) at GSI, the EN tandem accelerator-cooler in Aarhus, the electron-beam ion trap (EBIT) at Lawrence Livermore National Laboratory, the electron-beam ion source (EBIS) at Kansas State University, and the Bevalac at Lawrence Berkeley Laboratory. For the most part, the agreement between the high-resolution measurements and theoretical calculations based on the independent-processes and isolated-resonance (IP-IR) approximations has been quite good [1–11]. On the one hand, this result is reassuring to the astrophysical and laboratory plasma physics community, which bases many of its density and temperature diagnostics on theoretically generated recombination rates obtained in the IP-IR approximations. On the other hand, there has been a great deal of effort in recent years to develop a more general theory of electron-ion recombination which would go beyond the IP-IR approximations to include radiative-dielectronic recombination interference, overlapping-resonance structures, and scattering-channel coupling [12–19]. In an attempt to find an explanation for the success of the IP-IR approximations, we examine in this article the three major extensions of the theory and show that for current recombination experiments one should expect relatively small corrections. We also give our own ideas on the physical effects that need to be included in order to make more accurate predictions of electron-ion recombination processes.

Electron-ion recombination into a particular final recombined state, in the presence of one projectile continuum, may be schematically represented as

$$e^- + A_i^{q+} \rightarrow A_f^{(q-1)+} + \omega \ , \tag{1}$$

and

$$e^- + A_i^{q+} \rightleftarrows [\, A_j^{(q-1)+}\,] \rightarrow A_f^{(q-1)+} + \omega \ , \tag{2}$$

where q is the charge on the atomic ion A, ω is the frequency of the emitted light, and the brackets in Eq. (2) indicate a doubly-excited resonance state. The first process is called radiative recombination (RR), while the second is called dielectronic recombination (DR). In the widely used IP approximation the two paths for recombination are treated as independent processes. The radiative recombination cross section for Eq. (1), in lowest order of perturbation theory, is given by

$$\sigma_{\rm RR} = \frac{8\pi^2}{k_i^3} |\langle \Phi_f | D | \Psi_i \rangle|^2 \ , \tag{3}$$

where $|\Psi_i\rangle$ is the initial state of target ion plus continuum electron with total energy E, $|\Phi_f\rangle$ is the final bound recombined state,

$$D = (2\omega^3/3\pi c^3)^{1/2} \sum_{s=1}^{N+1} \mathbf{r}_s$$

is the dipole radiation field interaction, and k_i is the linear momentum of the initial free electron. The continuum normalization is chosen as 1 times a sine function, and atomic units are used. In the IR approximation the dielectronic recombination cross section for Eq. (2), in lowest-order of perturbation theory, is given by

$$\sigma_{\rm DR} = \frac{8\pi^2}{k_i^3} \sum_j \left| \frac{\langle \Phi_f | D | \phi_j \rangle \langle \phi_j | V | \Psi_i \rangle}{E - \varepsilon_j + i\Gamma_j/2} \right|^2 , \tag{4}$$

where $|\phi_j\rangle$ is the doubly-excited state with energy ε_j,

$$V = \sum_{s=1}^{N} |\mathbf{r}_s - \mathbf{r}_{N+1}|^{-1}$$

is the electrostatic interaction between electrons, and Γ_j is the total resonance width.

By the principle of detailed balance, the RR cross section of Eq. (3) is proportional to the photoionization cross section from the bound state $|\Phi_f\rangle$, while the energy-averaged DR cross section may be written as

$$\langle \sigma_{\rm DR} \rangle = \frac{2\pi^2}{\Delta\varepsilon k_i^2} \sum_j \frac{A_a(j\rightarrow i) A_r(j\rightarrow f)}{A_a(j\rightarrow i) + A_r(j\rightarrow f)} , \quad (5)$$

where the autoionization decay rate $A_a(j\rightarrow i) = 4/k_i |\langle \Psi_i | V | \phi_j \rangle|^2$, the radiative decay rate $A_r(j\rightarrow f) = 2\pi |\langle \Phi_f | D | \phi_j \rangle|^2$, and $\Delta\varepsilon$ is the energy bin width. Of course, for calculations involving degenerate states associated with either *LS* terms or *LSJ* levels, one must sum over magnetic quantum numbers. Then the equations for radiative [Eq. (3)] and dielectronic [Eqs. (4)–(5)] recombination must be divided by $2g_i$, the statistical weight of the incident electron times that of the target state. In addition, within the sum over j, the dielectronic recombination cross section must be multiplied by g_j, the statistical weight of the resonance state.

We first consider the interference between a single DR resonance and the RR background. In the limit of $A_a \gg A_r$, one can write the energy-averaged total recombination cross section as [15,20]

$$\langle \sigma_T \rangle \approx \sigma_{\rm RR} + \frac{2\pi^2}{\Delta\varepsilon k_i^2} \left[1 - \frac{1}{q^2} \right] A_r , \quad (6)$$

where q is the familiar Fano-resonance profile factor, given by

$$\frac{1}{q^2} = \frac{A_a}{A_r} \frac{k_i^2}{4\pi} \sigma_{\rm RR} . \quad (7)$$

Typically, the dominant resonances have $q^2 \gg 1$ and interference effects are less than 1% [20–22]. Weaker resonances can be perturbed by more, but they contribute proportionately less to a sum over a group of resonances. In the case of a very weak resonance ($q^2 \ll 1$) $\sigma_T \rightarrow 0$ on-resonance with a width of A_a. This would require that an energy resolution in the meV range to be observable, assuming it is not buried under numerous stronger resonances. Such a resolution is obtainable in photoionization experiments but not in photorecombination, to date. The effect on the overall cross sections is small since a typical RR cross section being suppressed is much weaker than a typical nonvanishing DR cross section. Furthermore, many of the final recombined states differ from the initial state by two electron pairs because of the two-body electrostatic operator, and so there is no RR background and no perturbation of the DR resonance. The radiative-dielectronic recombination interference cases for which $A_a \approx A_r$ and $A_a \ll A_r$ can be analyzed in a similar manner [20].

We have carried out calculations over a wide range of values for A_a, A_r, and $\sigma_{\rm RR}$, using more general expressions than Eq. (6) (see Refs. [16] and [20]), looking for possible observable effects. The largest effects are generally found in the extremes, namely *KLL* resonances in very highly charged ions and *LLL* or *MMM* resonances in neutrals. Previously [20], we found only a 3% reduction due to interference for the case of U^{90+}, although it may prove possible to observe an effect due to a factor of 2 reduction of one of the weaker *KLL* resonances. In the other extreme, we find a 5% reduction in the DR cross section from the $2s2p^3\,{}^1D$ resonance in neutral C and a 15% reduction for the $3p^2\,{}^1D$ resonance in neutral Mg. In both cases, the resonances lie just above the ground continuum and so would be important for low-temperature recombination. But these reductions are comparable with, or less than, the level of accuracy of the calculations due to uncertainties in the atomic structure for low-lying resonances in neutrals. The n^{-6} dependence of $1/q^2$ means the interference effects rapidly decrease with increasing n and, thus, charge state. We conclude that, in general, we can neglect the interference between a DR resonance and the RR background, if any exists.

Recently Nahar and Pradhan [23] have employed the close-coupling method to calculate photoionization cross sections from ground and excited states, and then by the principle of detailed balance extracted the total electron-ion recombination rate. However, they are incorrect in their assertion that the difference of up to a factor of 2 found between their close-coupling calculation for the total recombination rate of O^{3+} and earlier IP-IR calculations is due to the breakdown of the independent-processes approximation. The differences found are due to other factors. First, they should have added together the IP-IR low-temperature DR [24] and IP-IR high-temperature DR [25] contributions in their Fig. 2. This, together with the RR contribution [26], reduces the difference between the IP-IR and close-coupling results to +20% at low temperatures and −30% at high temperatures. Second, the reason for the cross over in the two sets of results is that the RR contribution to the close-coupling results appears to fall off more slowly with temperature ($\sim T^{-0.3}$) compared to the RR results of Aldrovandi and Pequignot [26] ($\sim T^{-0.67}$). At high temperatures, the RR contribution to the close-coupling results can be determined unambiguously from their Fig. 2 and it is a factor of 10 larger than the RR rate coefficient of Aldrovandi and Pequignot [26]. We note that at high temperatures the radiative recombination into the ground configuration of O^{2+} contributes 50% of the total RR rate coefficient and, for this contribution, Aldrovandi and Pequignot [26] used the previous close-coupling results of Henry [27]. We have also calculated the total RR rate coefficient for O^{3+} over the full temperature range in the IP approximation, and can confirm the results of Aldrovandi and Pequignot [26] to within the accuracy of the fitting formula used to present their results (which is typically better than 10%). Third, different N-electron and $(N+1)$-electron atomic structures were used in the IP-IR and close-coupling calculations.

With the validity of the independent-processes approximation in hand, we next consider the effect of overlapping DR resonances. A general reaction theory [28], including the effects of overlapping resonance structures, has been applied for many years to study photocapture in, and neutron scattering from, nuclei. A related configuration-interaction theory [29, 30] has also been applied to study photoabsorption in, and electron scattering

from, atoms and molecules. Recently, the projection-operator approach has been applied to electron-ion recombination [16–18]. For overlapping doubly excited resonances, the dielectronic recombination cross section for Eq. (2) is given by

$$\sigma_{\rm DR}=\frac{8\pi^2}{k_i^3}\left|\sum_j\sum_k\frac{\langle\Phi_f|D|\phi_j\rangle(\Omega^{-1})_{jk}\langle\phi_k|V|\Psi_i\rangle}{E-\varepsilon_k+i\Gamma_k/2}\right|^2, \tag{8}$$

where LaGattuta's dimensionless coupling matrix [17] is given by

$$\Omega_{jk}=\delta_{jk}-(1-\delta_{jk})\frac{\Lambda_{jk}}{E-\varepsilon_j+i\Gamma_j/2}, \tag{9}$$

and δ_{jk} is the Kronecker delta. In the pole approximation,

$$\Lambda_{jk}=-\frac{i}{2}[\tilde{A}_a(j\to i\to k)+\tilde{A}_r(j\to f\to k)], \tag{10}$$

where the generalized autoionization coupling rate $\tilde{A}_a(j\to i\to k)=4/k_i\langle\phi_k|V|\Psi_i\rangle\langle\Psi_i|V|\phi_j\rangle$, and the generalized radiative coupling rate $\tilde{A}_r(j\to f\to k)=2\pi\langle\phi_k|D|\Phi_f\rangle\langle\Phi_f|D|\phi_j\rangle$. When $\Omega_{jk}=\delta_{jk}$ is substituted in Eq. (8), one recovers the isolated-resonance DR cross section of Eq. (4), since one sum vanishes and the other sum yields no cross terms when squared ($\Lambda_{jk}=0$ for $j\neq k$). We note that in the paragraphs below we follow convention in labeling resonances whose widths are larger than their energy separations as "overlapping" if they interact, while only "superimposed" if they do not interact.

In order for doubly-excited resonances to interact there must be a small energy separation along with electron-continuum coupling and (or) photon-continuum coupling, as given by Eqs. (8)–(10). Resonance states that differ by more than two units of angular momentum, or have opposite parities, yield $\tilde{A}_a=\tilde{A}_r=0$. Resonance states that differ in total angular momentum yield $\tilde{A}_a=0$, while resonance states that do not have strong configuration-interaction mixing cannot couple effectively through core radiative transitions. We have performed a series of elementary numerical studies with Eqs. (8)–(10), and they show that only small differences beyond the isolated-resonance approximation are observed on line shapes when two DR resonances are separated in energy by more than five times their mean natural widths. For $A_a=\tilde{A}_a=10^{13}$ Hz and $A_r=\tilde{A}_r=10^{12}$ Hz, this corresponds to a separation of about 0.05 eV. With current DR experiments, these small effects would be "washed out" by the width of the electron beam. Furthermore, the integrated cross sections differ by less than 10% when the resonances are separated by more than twice their mean natural widths. Thus the combination of electron and photon continuum coupling selection rules and the requirement of near energy degeneracy will make it difficult to observe overlapping-resonance effects in current DR cross-section experiments.

To illustrate the difficulty in finding strong overlapping-resonance effects, we consider several cases. At energies far from the high-n accumulation point of a Rydberg series of resonances, there is only the accidental near degeneracy of two resonances. The photoionization calculations of Fliflet and Kelly [31] on neutral Zn suggest that dielectronic recombination of Zn^+ through the $3d^9 4s^2 4p$ resonances will produce a window feature due to the interaction between the ^1P and ^3P terms. Although observable in photoabsorption experiments [32], the window width of 0.05 eV is too small for current electron-ion experiments. Sakimoto [33] suggests that there should be observable effects for the $2s3s$ ($J=1$) and $2\bar{p}3\bar{p}$ ($J=1$) resonances in the dielectronic recombination of highly-charged hydrogenic ions, in particular Xe^{53+} ($n\bar{p}\equiv np\,\frac{1}{2}$). If we use the data of Sakimoto's [33] Table I for Xe^{53+} in Eqs. (8)–(10), we can reproduce Sakimoto's overlapping-resonance results in his Fig. 3 reasonably well. However, Sakimoto [33] has taken energies and rates from Nilsen [34], and it appears that he has incorrectly converted the rates to widths. On converting Nilsen's data, we find Sakimoto's values to be exactly a factor of 10 too large for all of the widths. For Xe^{53+}, this means that the $2s3s$ ($J=1$) and $2\bar{p}3\bar{p}$ ($J=1$) energy separation is still over four times the mean of the total resonance widths; we find very small differences in the line shapes and a 1% difference in the integrated cross section on using Nilsen's data in our isolated and overlapping-resonance calculations. Also, we have calculated energies and rates for Xe^{53+}, and they are in broad agreement with Nilsen's results [34]. Even at U^{91+}, we find the energy separation to be three times the resonance widths. It should also be noted that the $2s3s$ ($J=0$) resonance sits at virtually the same energy as the $J=1$ resonance, and its DR cross section is two orders of magnitude larger than the $J=1$ resonance.

We have also reexamined our IP-IR calculations of the KLL resonances in the U^{89+} and U^{90+} atomic ions [20] and have found no examples of overlapping resonances. The corresponding $1s2s^2$ ($J=\frac{1}{2}$) and $1s2\bar{p}^2$ ($J=\frac{1}{2}$) resonances in the dielectronic recombination of U^{90+} have strong configuration-interaction mixing, which allows the possibility of photon continuum coupling through the core transition ($1s\to 2\bar{p}$), but which also leads to a relatively large energy separation of 381 eV. With increased energy resolution at the storage-cooler rings, one may observe the rare overlapping resonance case, but the isolated-resonance approximation is certainly applicable for the vast majority of low-n DR resonances.

A second case is found in the recent work of LaGattuta [17] and of Bell and Seaton [14]. Near the high-n accumulation point of a single Rydberg series of resonances for a low charged atomic ion, the radiative widths are much larger than both the autoionization widths and the energy separations. If configuration-interaction mixing is small, however, then the photon continuum coupling through the strong core transition is nearly zero. Thus the resonances are merely superimposed, since they couple to mutually orthogonal photon continua, and the isolated-resonance approximation should be reasonably accurate. In LaGattuta's [17] Figs. 1 and 2, the overlapping resonance DR spectrum calculated using Eq. (8) is

compared with the isolated resonance DR spectrum calculated using Eq. (4) for a single Rydberg series of resonances. The two spectra would differ little except for the fact that the autoionization rates have been artificially chosen so large that they violate unitarity, which affects the high-n limit of LaGattuta's IR results. If large autoionization rates are found in real atomic systems, they may be corrected for unitarity by

$$A_a'(j\rightarrow i)=\left[1+\frac{\pi A_a(j\rightarrow i)}{2\delta E}\right]^{-2} A_a(j\rightarrow i)\ , \qquad (11)$$

where $\delta E=Z^2/n^3$ is the separation between resonances. The isolated-resonance approximation with unitarized autoionization rates may thus be applied over an even wider range of cases. As pointed out by LaGattuta [17], the overlapping resonance DR cross section results from Eq. (8) are indistinguishable from the multichannel-quantum-defect-theory results of Bell and Seaton [14] for the single Rydberg series case which, in turn, agree reasonably well with the IP-IR results of Bell and Seaton [14]. Our only reservation is the case of strong configuration interaction along the series, which will increase the photon-continuum coupling rates; however, it may at the same time remove any near energy degeneracies.

A third case is that of a low-lying resonance of one Rydberg series interacting with high-lying resonances of a second series. This situation occurs in the DR of metastable He-like ions [3,5,6]. In particular, the $1s2p\ ^1P\ 9l$ resonances formed by the DR of O^{6+} are buried in the resonances attached to the $1s2p\ ^3P$ limit. It has been suggested (Ref. [6], and references therein) that interactions between these two series could cause the $1s2p\ ^3P\ nl$ series to contribute significantly to the recombination and so explain the broadened experimental feature [3] observed below the $1s2p\ ^3P$ limit, which is not predicted by the IR approximation. We have carried out a calculation for O^{6+}, using Eqs. (8)–(10), in which we let pairs of resonances interact (a $1s2p\ ^1P\ 9l$ with a member of $1s2p\ ^3P\ nl'$ series, noting that within the series the resonances are well isolated) and sum over $n=20-100$. We find no difference from the IR result. On considering a much more highly charged case where the $2\ ^3P_1\rightarrow 1\ ^1S$ radiative rate is comparable with the $2\ ^1P_1\rightarrow 1\ ^1S$ rate, we still find no differences. The generalized autoionization rates for the $1s2p\ ^3P\ nl'$ resonances ($n>20$) interacting with the $1s2p\ ^1P\ 9l$ resonances are just too weak.

A fourth case of two overlapping-resonance series has been addressed in a recent model calculation for the dielectronic recombination of C^{5+} [18]. LaGattuta finds an approximate 15% difference in the integrated cross section for interacting versus noninteracting resonance series. But this is probably somewhat of an overestimate since all of the autoionization rates were chosen to satisfy $n^3A_a=0.2$, and we find that a significant fraction of the DR cross section comes from levels with $n^3A_a\ll 0.2$. Photoionization of complex ions, such as neutral Ca [35], neutral Yb [36], or neutral Ar [37], with multiple resonance series (and large autoionizing widths) attached to large numbers of LS terms or fine-structure levels, give excellent examples of the need for overlapping-resonance theory. With a further increase in the energy resolution found in current electron-ion experiments, the same type of overlapping-resonance features in the dielectronic recombination of complex ions might be uncovered.

Finally, we consider the effects of scattering-channel coupling within an overall IP-IR calculation. The close-coupling method has been shown to be clearly superior to the distorted-wave method in the calculation of background electron-impact excitation cross sections for transitions in low-charged or neutral atomic systems, although the two methods are often in fair agreement for strong dipole-allowed transitions. However, for all types of transitions in moderate to highly charged atomic systems, the close-coupling and distorted-wave methods have been shown to give similar background excitation-cross-section results [38]. We may extend such general statements to the calculation of autoionization rates since they involve similar Coulomb matrix elements; in fact, quantum defect theory relates the two in the limit of high-n and small energies [39]. The reason the distorted-wave approximation has been so successful in describing dielectronic recombination cross sections for any atomic ion is that for low charged ions the DR cross section is proportional to the radiative rate, while for highly charged ions the DR cross section is proportional to the autoionization rate. Thus the weakness of the distorted-wave method in calculating accurate autoionization rates for low charged ions is masked by a DR cross section that is highly dependent on radiative atomic structure. Even for the very sensitive case of O^+ [19,40], the accuracy of the final DR result is dominated by the accuracy of the N-electron structure (errors of up to a factor of 2 are possible), while $(N+1)$-electron correlation has about a 10% effect and close-coupling versus distorted-wave for a fixed $(N+1)$-electron expansion has a 1% effect. As one moves to more highly charged ions, the DR cross section becomes more sensitive to the autoionization rates, but at the same time, the distorted-wave method becomes increasingly more accurate.

Within the context of the independent-processes, isolated-resonance, and distorted-wave approximations for the calculation of electron-ion recombination cross sections, we conclude with a short discussion of the main limits to the accuracy of the method. One limit is the quality of the $(N+1)$-electron atomic structure. For example, in quite a few atomic systems, LS terms and LSJ levels within a single configuration are found to straddle the zero energy of the DR cross-section spectrum. Whether they are bound or resonance states depends on a sometimes difficult atomic-structure calculation. A second limit is the influence of the environment on the high-n states in a Rydberg series. Small electric fields in the interaction region of the experiment may substantially alter the DR cross sections at high n. However, this environmental effect is not limited to isolated-atom DR experiments. In fact, the main obstacle to applying overall DR rate coefficients calculated in the zero-field, zero-density limit to the modeling of astrophysical or laboratory plasmas is the substantial influence of collisions and static fields. Thus, in light of the apparent small

effects of quantum-mechanical interference on electron-ion recombination, the primary emphasis should now be on (1) the calculation of recombination cross sections and rate coefficients for heavy ions in relatively low stages of ionization, where the atomic structure is complex and overlapping-resonance effects might be more pronounced, and (2) the thorough investigation of plasma density effects, which have the potential for strongly affecting the recombination rate coefficient.

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