Density-matrix theory of recombination

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A general formulation for calculating the recombination probability from a system consisting of many autoionizing states interacting with many continuum states is given. The formulation is applied to obtain the recombination probability from two continuum states coupled to an autoionizing state. Explicit expressions for various fundamental processes contributing to recombination probability are also given.

I. INTRODUCTION

The physics of the dielectronic-recombination (DR) process, which involves the capture of a free electron by an ion to form a doubly excited state that then radiatively stabilizes to end up as a bound highly excited Rydberg state, has attracted much attention in recent years. An excellent review was given in 1976 by Seaton and Storey[1] summarizing the theory and importance of DR in applications. A general theory of DR was developed by Bell and Seaton.[2] The DR process in a system involving a single-electron continuum state interacting with a single discrete state has been studied in detail by Alber, Cooper, and Rau,[3] and it was shown that the total recombination probability has three contributions coming from (i) direct decay of the electron in the continuum state to the bound state (RR), (ii) decay of the continuum electron to the bound state via the autoionizing state (DR), and (iii) the interference between the above two processes.

A unified description of radiative and dielectronic recombination in a system consisting of single-electron and photon continua interacting with a single autoionizing state was developed by Jacobs, Cooper, and Haan,[4] using the projection-operator techniques to construct the transition operator for the electron-ion photorecombination process. In this method one needs to calculate the transition amplitude for each individual process and obtain the total transition amplitude by summing over all the allowed channels. The Feshbach formalism of projection-operator techniques was recently utilized by LaGattuta[5] to study the effects of overlapping resonances in electron-ion recombination, and it was found that these systems show a small reduction in the DR cross section. Haan and Jacobs[6] have also shown how projection-operator methods can be used to obtain the T matrix for electron-ion photorecombination processes in systems having a limited number of discrete states and continua. As a further application of this work, Haan[7] showed how generalized Fano parameters can be introduced to characterize a line profile for a system consisting of a single autoionizing state, a single-electron continuum, and one final state. Haan also applied the T-matrix formulation to study the recombination profiles when more than one electron or photon continuum was involved.

The effects of external fields on the DR process has also been extensively studied.[8–12] Using the techniques of quantum-defect theory, Harmin[8] extended the work of Bell and Seaton to include the influence of an arbitrary dc electric field, and he found that there was a considerable enhancement of the DR probability. The effect of plasma electric microfields was studied by Davis and Jacobs,[9] and it was found that the DR probability is strongly field dependent owing to the mixing of different angular momentum states. In a previous study[13] we had examined the effects of an external dc electric field on the recombination from coupled states. We showed that the recombination would also exhibit many interference effects, which occur in autoionization via coupled states.

For many situations, particularly those involving many events of radiative decay, the density-matrix framework is better suited. For example, if one were studying recombination in the presence of laser fields, then one has the possibility of the following processes repeatedly occurring: recombination, absorption of laser photon, autoionization, recombination. Agarwal, Haan and Cooper[14] studied a closely related system using the density-matrix method. Density matrix methods are also advantageous if one is interested in considerations of photon statistics. Note that recently there is considerable interest in the study of laser action[15–18] using autoionizing states.[19] It is known that a complete study of laser action can best be carried out in the density matrix framework. Thus the problem of laser action using autoionization and recombination processes would require density matrix treatment of recombination.

In this paper we present a general density-matrix formulation for calculating the recombination probability from a system consisting of many autoionizing (AI) states interacting with many continuum states. We demonstrate how the density-matrix framework can be used to account for different radiative decay processes. To keep the analysis simple, we neglect the presence of external fields. The organization of the paper is as follows: In Sec. II we present the density-matrix formulation of the recombination. In Sec. III we present the solution of the density-matrix equation. In Sec. IV we apply the results of Secs. II and III to obtain the recombination probability for a system involving two continuum states coupled to an AI state and decaying to a bound state. We also give the physical interpretation of various terms in the recom-

bination probability. Finally, in Sec. V we calculate the total recombination probability for the two continuum systems. The relation of our results to recent works is also established.

II. DENSITY-MATRIX THEORY OF RECOMBINATION

In this section we present a general formulation to obtain the recombination probability in a system consisting of many continuum states interacting with many autoionizing states and decaying to a single final bound state. We will derive a master equation describing the time evolution for the atomic density operator and use this master equation for calculating the recombination probabilities.

We will assume that the diagonalization of the part of the Hamiltonian that accounts for the coupling of the autoionizing states with the continuum states has been carried out with the result that we have many orthogonal continuum states denoted by $|\alpha E_\alpha\rangle$ (α represents the continuum index and E_α the energy of the continuum) decaying via spontaneous emission to the bound state $|f\rangle$. In recombination processes it is assumed that initially the incoming electron is captured into a high-lying state of the ion; this electron now makes a transition to the bound state by spontaneous emission of photons. For our model system if we assume the initial state of the atomic system to be the continuum state $|\alpha E_\alpha\rangle$, then the various available channels through which the electrons can go to the bound state $|f\rangle$ are as follows: direct transition from the state $|\alpha E_\alpha\rangle$ to $|f\rangle$ (this process is referred to as radiative recombination) and transition from $|\alpha E_\alpha\rangle$ to one or many AI states via configuration interaction followed by a decay from the AI state to the state $|f\rangle$ (this process is dielectronic recombination). In the other process which arises due to the presence of many continuum states, the electrons make transitions from $|\alpha E_\alpha\rangle$ to one or many continuum states by configuration interaction and then via spontaneous emission decays to the bound state $|f\rangle$. Thus we see that there are many channels available for the incident electron. The recombination probability to the state $|f\rangle$ in our approach can be obtained by calculating the density-matrix element ρ_{ff}. The formulation automatically includes all the interference effects between different channels of radiative decay. In Sec. III it will be shown that the recombination probability apart from having contributions from the fundamental processes also has contributions which arise due to the interference between different processes. The total Hamiltonian for our system has the form

$$H = H_A + H_R + H_{AR} , \tag{2.1}$$

where H_A and H_R are the unperturbed atomic and radiation part of the Hamiltonian and H_{AR} gives the interaction part which is responsible for recombination. The different parts of H can be written in the form

$$H_A = \sum_\alpha \int E_\alpha |\alpha E_\alpha\rangle\langle \alpha E_\alpha| dE_\alpha + E_f |f\rangle\langle f| , \tag{2.2}$$

$$H_R = \sum_{ks} \omega_{ks} a_{ks}^\dagger a_{ks} , \tag{2.3}$$

$$H_{AR} = -\mathbf{d}\cdot\mathbf{E} , \tag{2.4}$$

where

$$\mathbf{d} = \sum_\alpha \int \mathbf{d}_{\alpha f} |\alpha E_\alpha\rangle\langle f| dE_\alpha + \text{H.c.} \tag{2.5}$$

is the dipole matrix element connecting the continuum state and the state $|f\rangle$ and

$$\mathbf{E} = i \sum_{ks} \left[\frac{2\pi ck}{L^3} \right]^{1/2} \epsilon_{ks} a_{ks} e^{i\mathbf{k}\cdot\mathbf{r}} + \text{H.c.} \tag{2.6}$$

is the mode expansion for the quantized electric field. Here L^3 is the volume in which the mode is quantized, ϵ_{ks} is the polarization, $\mathbf{k}$ the wave vector, and a_{ks} and $a_{ks}^\dagger$ are the photon annihilation and creation operators. In Eq. (2.5) the dipole matrix elements are implicit functions of the energy E_α. The Hamiltonian (2.4) can be written using (2.5) and (2.6) in the interaction picture after making the rotating-wave approximation as

$$H'_{AR}(t) = -\sum_{ks} \epsilon_{ks}\cdot \mathbf{A}_f^\dagger a_{ks} g_{ks} e^{-i\omega_{ks} t} + \text{H.c.} , \tag{2.7}$$

$$\mathbf{A}_f^\dagger(t) = \sum_\alpha \int \mathbf{d}_{\alpha f} |\alpha E_\alpha\rangle\langle f| e^{i(E_\alpha - E_f)t} dE_\alpha , \tag{2.8}$$

$$g_{ks} = i \left[\frac{2\pi ck}{L^3} \right]^{1/2} e^{i\mathbf{k}\cdot\mathbf{r}} . \tag{2.9}$$

Thus in the recombination process the atomic system makes a transition from the "diagonalized" continuum state $|\alpha E_\alpha\rangle$ to the state $|f\rangle$ and a photon is emitted.

Let $\rho_{A+R}(t)$ represent the density operator characterizing the statistical state of the combined system of atom and radiation field. It obeys the Liouville equation

$$\dot{\rho}_{A+R}(t) = -i[H, \rho_{A+R}(t)] = -iL\rho_{A+R}(t) , \tag{2.10}$$

where L is the Liouville operator defined as $L\cdots = [H, \ldots]$. The reduced density operator corresponding to the atomic system alone $[\rho_A(t)]$ is obtained by tracing over the radiation field variables, i.e.,

$$\rho_A(t) = \text{Tr}_R \rho_{A+R}(t) . \tag{2.11}$$

We will see that the knowledge of $\rho_A(t)$ is sufficient to calculate the recombination probability. At the beginning of the interaction, we assume that the total density matrix is a product of atomic and field density matrices, i.e.,

$$\rho_{A+R}(0) = \rho_A(0)\rho_R(0) , \tag{2.12}$$

where $\rho_A(0)$ and $\rho_R(0)$ are the initial states of the atomic system and radiation field, respectively. Initially, the radiation field is in a vacuum state, i.e.,

$$\rho_R(0) = |\{0\}\rangle\langle\{0\}| , \tag{2.13}$$

i.e., all the modes are empty. The initial atomic state $\rho_A(0)$ is left arbitrary. We solve for the total density operator of (2.10) using the projection-operator techniques and obtain an equation for the atomic density operator $\rho_A(t)$ alone. The reduced density-matrix equation is obtained under two standard assumptions: (i) The

interaction between the atomic system and the radiation field is weak and (ii) the reservoir has no memory effects, i.e., the atomic time scales of interest are much larger than the correlation time of the reservoir. Both these assumptions generally hold. Note that the emitted photon is not reabsorbed by the atomic system. We work in the interaction picture and use these approximations, and then obtain the following equation for the atomic density operator ρ:[20]

$$\frac{\partial}{\partial t}\rho'_A(t)+\lim_{t\to\infty}\int_0^t d\tau\,\mathrm{Tr}_R[L'_{AR}(t)L'_{AR}(t-\tau)\rho^0_R\rho'_A(t)]=0\ , \tag{2.14}$$

where the operators in the interaction picture are denoted by ρ' and L' and where $L'_{AR}(t)=[H'_{AR}(t),\ ]$. For the sake of completeness the derivation of (2.14) is given in Appendix A.

We will now apply the basic equation (2.14) to the recombination process. Using (2.7), we have

$$L'_{AR}(t-\tau)\rho^0_R\rho'_A(t-\tau)=-\sum_{ks} g_{ks}e^{-i\omega_{ks}(t-\tau)}[a_{ks}[\epsilon_{ks}\cdot\mathbf{A}^\dagger_f(t-\tau)]\ ,\rho^0_R\rho'_A(t-\tau)]$$
$$-\sum_{k's'} g^*_{k's'}e^{i\omega_{k's'}(t-\tau)}[a^\dagger_{k's'}[\epsilon^*_{k's'}\cdot\mathbf{A}_f(t-\tau)]\ ,\rho^0_R\rho'_A(t-\tau)]\ . \tag{2.15}$$

Now,

$$L'_{AR}(t)L'_{AR}(t-\tau)\rho^0_R\rho'_A(t-\tau)=-\sum_{k''s''}[[\epsilon_{k''s''}\cdot\mathbf{A}^\dagger_f(t)]a_{k''s''}g_{k''s''}e^{-i\omega_{k''s''}(t)}$$
$$+[\epsilon^*_{k''s''}\cdot\mathbf{A}_f(t)]a^\dagger_{k''s''}g^*_{k''s''}e^{i\omega_{k''s''}(t)}\ ,L'_{AR}(t-\tau)\rho^0_R\rho'_A(t-\tau)]\ , \tag{2.16}$$

Using (2.15) and the relation

$$\mathrm{Tr}_R(\rho^0_R a^\dagger_{ks}a_{k's'})=\mathrm{Tr}_R(a_{k's'}\rho^0_R a^\dagger_{ks})=\mathrm{Tr}_R(a^\dagger_{ks}a_{k's'}\rho^0_R)=0\ ,$$
$$\mathrm{Tr}_R(\rho^0_R a_{k's'}a^\dagger_{ks})=\delta_{kk'}\delta_{ss'}\ , \tag{2.17}$$

we find that

$$\mathrm{Tr}_R L'_{AR}(t)L'_{AR}(t-\tau)\rho^0_R\rho'_A(t-\tau)=\sum_k|g_{ks}|^2\{[\epsilon_{ks}\cdot\mathbf{A}^\dagger_f(t)][\epsilon^*_{ks}\cdot\mathbf{A}_f(t-\tau)]\rho'_A(t-\tau)$$
$$+\rho'_A(t-\tau)[\epsilon_{ks}\cdot\mathbf{A}^\dagger_f(t-\tau)][\epsilon^*_{ks}\cdot\mathbf{A}_f(t)]\}e^{-i\omega_{ks}\tau}$$
$$-\sum_{ks}|g_{ks}|^2\{[\epsilon^*_{ks}\cdot\mathbf{A}_f(t-\tau)]\rho'_A(t-\tau)$$
$$\times[\epsilon_{ks}\cdot\mathbf{A}^\dagger_f(t)]+[\epsilon^*_{ks}\cdot\mathbf{A}_f(t)]\rho'_A(t-\tau)[\epsilon_{ks}\cdot\mathbf{A}^\dagger_f(t-\tau)]\}e^{i\omega_{ks}\tau}\ . \tag{2.18}$$

Substituting Eqs. (2.8), (2.9), and (2.18) in (2.16) together with the relation

$$\sum_s(\epsilon_{ks})_\alpha(\epsilon_{ks})_\beta=(\delta_{\alpha\beta}-\hat{k}_\alpha\hat{k}_\beta) \tag{2.19}$$

and upon simplification, we find the atomic density operator satisfies the following equation in the Schrödinger representation:

$$\frac{\partial\rho_A}{\partial t}=-i[H_0,\rho]-\sum_{\alpha\beta}\int dE_\alpha dE_\beta s_\beta\mathbf{d}^\dagger_{\alpha f}\cdot\mathbf{d}_{\beta f}(|\alpha E_\alpha\rangle\langle\beta E_\beta|\rho_A+\rho_A|\alpha E_\alpha\rangle\langle\beta E_\beta|-2|f\rangle\langle f|\langle\beta E_\beta|\rho_A|\alpha E_\alpha\rangle)\ , \tag{2.20}$$

with

$$H_0=\sum_\alpha\int E_\alpha|\alpha E_\alpha\rangle\langle\alpha E_\alpha|dE_\alpha+E_f|f\rangle\langle f|\ , \tag{2.21}$$

$$s_\alpha=\frac{2}{3c^3}(E_\alpha-E_f)^3\ . \tag{2.22}$$

We have now transformed back to the Schrödinger picture. From now on we deal only with $\rho_A(t)$ and hence we drop suffix A from ρ. Using (2.20), various matrix elements of the atomic density operator satisfy the equations

$$\dot{\rho}_{\alpha'\alpha''}=\langle\alpha' E_{\alpha'}|\dot{\rho}|\alpha'' E_{\alpha''}\rangle$$
$$=-i(E_{\alpha'}-E_{\alpha''})\rho_{\alpha'\alpha''}-\sum_\beta\int s_\beta\mathbf{d}^\dagger_{\alpha' f}\cdot\mathbf{d}_{\beta f}\rho_{\beta\alpha''}dE_\beta$$
$$+\sum_\alpha\int s_{\alpha''}\mathbf{d}^\dagger_{\alpha f}\cdot\mathbf{d}_{\alpha'' f}\rho_{\alpha'\alpha}dE_\alpha\ , \tag{2.23}$$

$$\dot{\rho}_{f\alpha'}=-i(E_f-E_{\alpha'})\rho_{f\alpha'}-\sum_\alpha\int s_{\alpha'}\mathbf{d}^\dagger_{\alpha f}\cdot\mathbf{d}_{\alpha' f}\rho_{f\alpha}dE_\alpha\ , \tag{2.24}$$

$$\dot{\rho}_{ff}=2\sum_{\alpha\beta}\int s_\alpha\mathbf{d}^\dagger_{\alpha f}\cdot\mathbf{d}_{\beta f}\rho_{\beta\alpha}dE_\alpha dE_\beta\ . \tag{2.25}$$

Equation (2.25) gives the recombination rate to the state $|f\rangle$. This is related to the dipole matrix elements connecting the discrete state $|f\rangle$ to the new diagonalized states, $|\alpha\rangle$'s and $|\beta\rangle$'s. The recombination probability to the state $|f\rangle$ can be obtained from the relation

$$P_f=\lim_{t\to\infty}\rho_{ff}(t)\ . \tag{2.26}$$

We next present the solutions of the density-matrix elements.

III. SOLUTION OF THE DENSITY-MATRIX EQUATION

In order to solve the coupled equations, it turns out to be convenient to introduce quantities ψ's defined by

$$\rho_{\alpha\beta}(t)=\psi_\alpha(t)\psi^*_\beta(t)\ . \tag{3.1}$$

The wave functions ψ_α's are found to satisfy the equations

$$\dot{\psi}_\alpha=-i\Delta_\alpha\psi_\alpha-\sum_\beta\int \mathbf{d}^\dagger_{\alpha f}\cdot\mathbf{d}_{\beta f}\psi_\beta s_\beta dE_\beta\ , \tag{3.2}$$

where

$$\Delta_\alpha=(E_\alpha-E_f)\ , \tag{3.3}$$

$$s_i=\frac{2}{3c^3}(E_i-E_f)^3\ . \tag{3.4}$$

Taking the Laplace transform of (3.2), we get

$$\hat{\psi}_\alpha(z)=\frac{\psi_\alpha(0)}{z+i\Delta_\alpha}-\sum_\beta\int\frac{\mathbf{d}^\dagger_{\alpha f}\cdot\mathbf{d}_{\beta f}\hat{\psi}_\beta s_\beta dE_\beta}{(z+i\Delta_\alpha)}\ , \tag{3.5}$$

where $\psi_\alpha(0)$ represents the initial continuum state. The integral equation (3.5) has the separable form and can be solved by standard methods. For simplicity we ignore in the following the vectorial nature of $\mathbf{d}$, though for problems involving magnetic degeneracies, the vectorial nature of $\mathbf{d}$ is very important. Defining quantities

$$k_\alpha(E_\alpha)=\frac{d^\dagger_{\alpha f}}{(z+i\Delta_\alpha)}\ ,\quad L_\beta(E_\beta)=d_{\beta f}s_\beta\ , \tag{3.6}$$

$$\hat{\chi}_\beta(z)=\int L_\beta(E_\beta)\hat{\psi}_\beta dE_\beta\ , \tag{3.7}$$

Eq. (3.5) can be written as

$$\hat{\psi}_\alpha(z)+k_\alpha(E_\alpha)\sum_\beta\chi_\beta(z)=\frac{\psi_\alpha(0)}{z+i\Delta_\alpha}\ . \tag{3.8}$$

Multiplying this equation by $L_\alpha(E_\alpha)$ and integrating over dE_α, we have

$$\hat{\chi}_\alpha(z)+m_{\alpha\alpha}\sum_\beta\hat{\chi}_\beta(z)=\hat{f}_\alpha(z)\ , \tag{3.9}$$

where

$$\begin{aligned} m_{\alpha\alpha}&=\int k_\alpha(E_\alpha)L_\alpha(E_\alpha)dE_\alpha\ ,\\ \hat{f}_\alpha(z)&=\int\frac{\psi_\alpha(0)L_\alpha(E)_\alpha dE_\alpha}{z+i\Delta_\alpha)}\ . \end{aligned} \tag{3.10}$$

Equation (3.9) also leads to

$$\sum_\alpha\hat{\chi}_\alpha(z)=\left[1+\sum_\beta m_{\beta\beta}\right]^{-1}\sum_\alpha\hat{f}_\alpha(z)\ . \tag{3.11}$$

We next introduce ϕ, defined by

$$\hat{\phi}_\beta(z)=\int d_{\beta f}\hat{\psi}^*_\beta(z)dE_\beta\ . \tag{3.12}$$

Using (3.1), (3.11), and (3.12) the recombination rate (2.25) can be written as

$$\dot{\rho}_{ff}=2\sum_\alpha\chi^*_\alpha(t)\sum_\beta\phi_\beta(t)\ . \tag{3.13}$$

Note that the quantity $\hat{\phi}_\beta(z)$ involves evaluation of $\hat{\psi}_\beta(z)$ which can be done by substituting (3.11) in (3.8). Equation (3.13) thus gives the recombination rate in a system involving many AI states interacting with many continuum states and decaying to the bound state $|f\rangle$. Substituting (3.6) in (3.7), we have

$$\hat{\chi}_\beta(z)=\int d_{\beta f}s_\beta\hat{\psi}_\beta dE_\beta\ . \tag{3.14}$$

For the case when s_β can be taken to be energy independent,

$$s_\beta=(2/3c^3)(E_\beta-E_f)\cong s\ . \tag{3.15}$$

Upon using Eqs. (3.12) and (3.15), we find that

$$\hat{\psi}_\beta(z)=s\hat{\phi}_\beta(z)\ ; \tag{3.16}$$

thus Eq. (3.13) in this limit can be written in a simple form as

$$\dot{\rho}_{ff}=\frac{2}{s}\left|\sum_\beta\chi_\beta(t)\right|^2\ . \tag{3.17}$$

The recombination probability can be obtained using (3.17) as

$$P_f=\lim_{t\to\infty}\int_0^t\frac{2}{s}\left|\sum_\beta\chi_\beta(\tau)\right|^2 d\tau\ . \tag{3.18}$$

Explicit determination of the recombination probability to a particular problem of interest would require the form of the diagonalized states $|\alpha E\rangle$ and various dipole matrix elements. We next apply these results to a system involving two continuum states interacting with a single AI state.

IV. APPLICATION OF THE GENERAL RESULT TO RECOMBINATION INVOLVING TWO CONTINUA

In Fig. 1 we represent schematically the model system of interest. Here $|\psi_E\rangle$ and $|\chi_E\rangle$ are unperturbed continuum states which are coupled to the autoionizing (AI) state $|a\rangle$ via configuration interaction. The wavy lines represent the spontaneous decay of the AI and continuum states to the bound state $|f\rangle$. The Hamiltonian for the system has the form

$$H=H_0+H_c+H_I\ , \tag{4.1}$$

$$\begin{aligned} H_0=&\int E|\psi_E\rangle\langle\psi_E|dE+\int E|\chi_E\rangle\langle\chi_E|dE+E_a|a\rangle\langle a|\\ &+E_f|f\rangle\langle f|+\sum_{ks}\omega_{ks}a^\dagger_{ks}a_{ks}\ , \end{aligned} \tag{4.2}$$

$$H_c=\int V_{Ea}|\psi_E\rangle\langle a|dE+\int W_{EA}|\chi_E\rangle\langle a|dE+\text{H.c.}\ , \tag{4.3}$$

$$H_I=\left[\int|\psi_E\rangle\langle f|(\mathbf{V}_{Ef}\cdot\mathbf{E}_{\text{vac}}^{(+)})dE+\int|\chi_E\rangle\langle f|(\mathbf{W}_{Ef}\cdot\mathbf{E}_{\text{vac}}^{(+)})dE+|a\rangle\langle f|(\mathbf{v}_{af}\cdot\mathbf{E}_{\text{vac}}^{(+)})+\text{H.c.}\right]. \tag{4.4}$$

Various terms in Eq. (4.1) have the following meaning: H_0 is the unperturbed Hamiltonian of the atom field system. H_c accounts for the autoionization of state $|a\rangle$ into channels $|\psi_E\rangle$ and $|\chi_E\rangle$, whereas H_I is responsible for the spontaneous decay of the states $|\psi_E\rangle$, $|\chi_E\rangle$, and $|a\rangle$. In (4.3) and (4.4) terms involving $V(W)$ represent coupling to the continuum $|\psi_E\rangle(|\chi_E\rangle)$ and v_{af} is the dipole coupling between $|a\rangle$ and $|f\rangle$. Further, $E_{\text{vac}}^{(+)}$ is the positive frequency part of the electric-field operator and consists of all the modes of the radiation field. This is the part responsible for recombination. In the recombination process an electron from a doubly excited state stabilizes to a bound state by emission of photons. We now diagonalize the part of the Hamiltonian H_c which is responsible for configuration interaction.[19] The diagonalization produces new continuum states which we denote by $|E\rangle$ and $|G\rangle$. The old basis states and the new basis states are related to each other by the following relations:

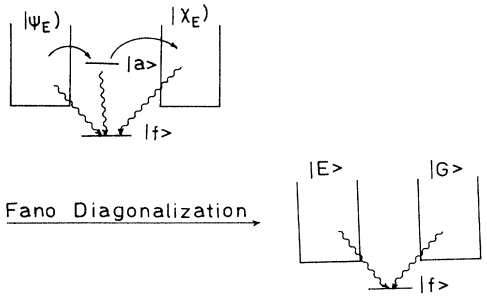

FIG. 1. Schematic energy-level diagram for the model system of interest.

$$|E\rangle=\left[\frac{2}{\Gamma\pi}\right]^{1/2}\sin\Delta|a\rangle+\int\left[\frac{2\pi}{\Gamma}\right]^{1/2}V_{E'a}\left[\frac{\sin\Delta}{\pi(E-E')}-\cos\Delta\delta(E-E')\right]|\psi_{E'})dE'+\left[\frac{2\pi}{\Gamma}\right]^{1/2}\int W_{E'a}\left[\frac{\sin\Delta}{\pi(E-E')}-\cos\Delta\delta(E-E')\right]|\chi_{E'})dE'\ , \tag{4.5}$$

$$|G\rangle=\left[\frac{2\pi}{\Gamma}\right]^{1/2}[W_{Ea}^*|\psi_E)-V_{Ea}^*|\chi_E)]\ , \tag{4.6}$$

$$\Gamma=2\pi(|V_{Ea}|^2+|W_{Ea}|^2)\ , \tag{4.7}$$

$$\tan\Delta=-\frac{\Gamma}{2(E-E_a)}\ . \tag{4.8}$$

The Hamiltonian (4.1) can be written in the new basis as

$$H=\tilde{H}_0+\tilde{H}_1\ , \tag{4.9}$$

$$\tilde{H}_0=\int E|E\rangle\langle E|dE+\int G|G\rangle\langle G|dG+E_f|f\rangle\langle f|+\sum_{ks}\omega_{ks}a_{ks}^\dagger a_{ks}\ , \tag{4.10}$$

$$\tilde{H}_1=-\left[\int|E\rangle\langle f|(\mathbf{d}_{Ef}\cdot\mathbf{E}_{\text{vac}}^{(+)})dE+\int|G\rangle\langle f|(\mathbf{d}_{Gf}\cdot\mathbf{E}_{\text{vac}}^{(+)})dG+\text{H.c.}\right]. \tag{4.11}$$

In (4.11), d_{ij}'s are the dipole matrix elements connecting the states $|i\rangle$ and $|j\rangle$ and these are found to have the following explicit form:

$$d_{Ef}=q_fC_{1f}\sin\Delta\left[1+\frac{2}{\Gamma q_f}(E-E_a)\right], \tag{4.12}$$

$$d_{Gf}=C_{2f}\ , \tag{4.13}$$

$$C_{1f}=\left[\frac{2\pi}{\Gamma}\right]^{1/2}(V_{Ef}V_{Ea}^*+W_{Ef}W_{Ea}^*)\ ,\qquad C_{2f}=\left[\frac{2\pi}{\Gamma}\right]^{1/2}(V_{Ef}W_{Ea}-W_{Ef}V_{Ea})\ , \tag{4.14}$$

$$q_f=\frac{\langle a|V|f\rangle|}{\pi[V_{Ea}^*(\psi_E|V|f\rangle+W_{Ea}^*(\chi_E|W|f\rangle]}\ . \tag{4.15}$$

We are now in a position to compute all the quantities of interest. For the two-continuum case the expression (3.6) reduces to

$$K_1(E)=\left[\frac{d_{Ef}}{z+i\Delta_G}\right],\quad L_1(E)=sd_{Ef}^*\ ,\qquad K_2(G)=\left[\frac{d_{Gf}}{z+i\Delta_G}\right],\quad L_1(G)=sd_{Gf}^*\ , \tag{4.16}$$

while the expression (3.9) has the form

$$\hat{\chi}_1(z)=f_1-m_{11}(\hat{\chi}_1+\hat{\chi}_2)\ , \tag{4.17}$$

$$\hat{\chi}_2(z)=f_2-m_{22}(\hat{\chi}_1+\hat{\chi}_2)\ , \tag{4.18}$$

where

$$\hat{f}_1(z)=\int \frac{L_1(E)\psi_E(0)dE}{z+i\Delta_E}\ ,$$
$$\hat{f}_2(z)=\int \frac{L_2(G)\psi_G(0)dG}{z+i\Delta_G}\ , \tag{4.19}$$

$$m_{11}=\int L_1(E)K_1(E)dE\ ,$$
$$m_{22}=\int L_2(G)K_2(G)dG\ . \tag{4.20}$$

In (4.19), $\psi_E(0)$ and $\psi_G(0)$ are the initial states of $|E\rangle$ and $|G\rangle$, respectively. Equations (4.17) and (4.18) can be cast in a matrix form,

$$\begin{bmatrix}\hat{\chi}_1(z)\\ \hat{\chi}_2(z)\end{bmatrix}=\begin{bmatrix}1+m_{11} & m_{11}\\ m_{22} & 1+m_{22}\end{bmatrix}^{-1}\begin{bmatrix}\hat{f}_1\\ \hat{f}_2\end{bmatrix}\ . \tag{4.21}$$

Equation (2.25) for the recombination rate to the state $|f\rangle$ can be written as

$$\dot{\rho}_{ff}(t)=2|\chi_1(t)+\chi_2(t)|^2\ . \tag{4.22}$$

Thus, to determine the recombination probability to the state $|f\rangle$ [Eq. (2.26)], one needs to compute the various matrix elements elements of m and determine the quantities f.

To determine f, we need to know the initial states $|\psi_{E_1}(0)\rangle$ and $|\psi_{G_1}(0)\rangle$ of the new diagonalized states. Following Davies and Seaton,[21] we assume a wave-packet structure for the initial state of the electron, i.e.,

$$|\psi(0)\rangle=\int dE|\psi_E\rangle\zeta(E)\ , \tag{4.23}$$

with

$$\int dE|\zeta(E)|^2=1\ . \tag{4.24}$$

The form of ζ is taken to be

$$\zeta(E)=\left[\frac{2m\beta^3}{\pi k\hbar^2}\right]^{1/2}\frac{e^{ikr_0}}{(k-k_0)^2+\beta^2}\ , \tag{4.25}$$

where r_0 is large, β is small, k_0 gives the center of the wave packet, and $r_0\beta\gg 1$. Note that in (4.25) the parameter k depends on the energy.

Now,

$$\psi_{E_1}(0)=\langle E_1|\psi(0)\rangle$$
$$=\int dE''\langle E_1|\psi_{E''}\rangle\zeta(E'') \tag{4.26}$$

and

$$\psi_{G_1}(0)=\int dE''\langle G_1|\psi_{E''}\rangle\zeta(E'')\ . \tag{4.27}$$

Using (4.5), (4.6), (4.26), and (4.27) and the orthogonality property between the unperturbed states $|\psi_E\rangle$ and $|\chi_E\rangle$, we have

$$\psi_{E_1}(0)=\int dE''\zeta(E'')\left[\frac{2\pi}{\Gamma}\right]^{1/2}V^*_{E''a}$$
$$\times\left[\frac{\sin\Delta}{\pi(E_1-E'')}-\cos\Delta\delta(E_1-E'')\right]\ , \tag{4.28}$$

$$\psi_{G_1}(0)=\int dE''\zeta(E'')\left[\frac{2\pi}{\Gamma}\right]^{1/2}W^*_{E''a}\delta(E_1-E'')\ . \tag{4.29}$$

Substituting (4.28) and (4.29) in (4.19), we have

$$\hat{f}_1(z)=\int dE\frac{s}{(z+i\Delta_E)}d^*_{Ef}\int dE'\zeta(E')\left[\frac{2\pi}{\Gamma}\right]^{1/2}V^*_{E'a}\left[\frac{\sin\Delta}{\pi(E-E')}-\cos\Delta\delta(E-E')\right]$$
$$=s\left[\frac{2\pi}{\Gamma}\right]^{1/2}\int dE'\zeta(E')V^*_{E'a}\int\frac{dE\,d^*_{Ef}}{(z+i\Delta_E)}\left[\frac{\sin\Delta}{\pi(E-E')}-\cos\Delta\delta(E-E')\right]\ , \tag{4.30}$$

$$\hat{f}_2(z)=s\left[\frac{2\pi}{\Gamma}\right]^{1/2}\int\frac{d^*_{Gf}W_{Ea}\zeta(E)dE}{(z+i\Delta_E)}\ . \tag{4.31}$$

Substituting for the dipole matrix elements of (4.12) and (4.13) in (4.30) and (4.31) and upon simplification, we find

$$\hat{f}_1(z)=s\left[\frac{2\pi}{\Gamma}\right]^{1/2}q_fC_{1f}\int\zeta(E')V^*_{E'a}dE'\left[\frac{(1-i/q_f)}{(\delta_{E'}+i)[z+i(E_a-E_f)+\Gamma/2]}+\frac{(1+\delta_E/q_f)}{(\delta_{E'}+i)[z+i(E'-E_f)]}\right]\ , \tag{4.32}$$

$$\hat{f}_2(z)=s\left[\frac{2\pi}{\Gamma}\right]^{1/2}\int\frac{\zeta(E')W_{E'a}C_{2f}}{[z+i(E'-E_f)]}\ , \tag{4.33}$$

where $\delta_{E'}=(2/\Gamma)(E'-E_a)$.

The matrix elements m_{11} and m_{22} can be computed using (4.12) and (4.16) in (4.20). These are found to be

$$m_{11}=s\pi q_f^2|C_{1f}|^2\left[\frac{(1-i/q_f)^2}{2z/\Gamma+1-i\alpha}+\frac{1}{q_f^2}\right]\ , \tag{4.34}$$

$$m_{22}=s\pi|C_{2f}|^2\ , \tag{4.35}$$

$$\alpha=\frac{2}{\Gamma}(E_f-E_a)\ . \tag{4.36}$$

Having obtained the exact expressions for f and m, we use these now to compute the recombination probability

to the state $|f\rangle$. The recombination probability to the state $|f\rangle$ in this case has the form

$$P_f=\frac{2}{s}\int_0^\infty[|\chi_1(t)|^2+|\chi_2(t)|^2]dt\ , \tag{4.37}$$

Using (4.21), we have

$$\hat{\chi}_1(z)=[(1+m_{22})\hat{f}_1-m_{11}\hat{f}_2]\frac{1}{\|m\|}\ , \tag{4.38}$$

$$\hat{\chi}_2(z)=[-m_{22}\hat{f}_1+(1+m_{11})\hat{f}_2]\frac{1}{\|m\|}\ , \tag{4.39}$$

where

$$\|m\|=\det m=1+m_{11}+m_{22}\ . \tag{4.40}$$

Quantities such as $\chi(t)$ can be obtained on taking the inverse Laplace transform of $\hat{\chi}(z)$. From (4.32) we see that $\hat{f}_1(z)$ has poles at values of z given by $z=-i(E'-E_f)$ and $z=-i(E_a-E_f)-\Gamma/2$, while $\hat{f}_2(z)$ has a pole at $z=-i(E'-E_f)$. Also, $\|m\|(z)$ has complex zeroes z_i. Using (4.32), (4.33), and (4.34), it is evident that $\chi(t)$ has the form

$$\chi_1(t)=\lim_{z\to -i(E'-E_f)}\int\frac{\zeta(E')dE'}{(\|m\|)}\left[(1+m_{22})q_fC_{1f}\frac{V^*_{E'a}(1+\delta_{E'}/q_f)}{(\delta_{E'}+i)}-m_{11}W_{E'a}C_{2f}\right]\left[\frac{2\pi s^2}{\Gamma}\right]^{1/2}e^{-i(E'-E_f)t}+\cdots\ , \tag{4.41}$$

$$\chi_2(t)=\lim_{z\to -i(E'-E_f)}\int\frac{\zeta(E')dE'}{(\|m\|)}\left[(-m_{22})q_fC_{1f}\frac{V_{E'a}(1+\delta_{E'}/q_f)}{(\delta_{E'}+i)}+(1+m_{11})W_{E'a}C_{2f}\right]\left[\frac{2\pi s^2}{\Gamma}\right]^{1/2}e^{-i(E'-E_f)t}+\cdots\ . \tag{4.42}$$

In the above expressions terms $\cdots$ represent the contributions from the complex zeroes. These contributions go to zero in the long-time limit and hence we neglect these. From (4.41) and (4.42) we have

$$\chi_1(t)+\chi_2(t)=\lim_{z\to -i(E'-E_f)}\int\frac{\zeta(E')dE'}{(\|m\|)}\left[\frac{q_fC_{1f}V^*_{E'a}(1+\delta_{E'}/q_f)}{(\delta_{E'}+i)}+W_{E'a}C_{2f}\right]\left[\frac{2\pi s}{\Gamma}\right]^{1/2}e^{-i(E'-E_f)t} \tag{4.43}$$

Using (4.43) in (4.37), we have

$$P_f=2\lim_{z_1\to 0}\int\frac{dE_1\zeta(E_1)}{S_{E_1}S^*_{E_2}}\int\frac{dE_2\zeta^*(E_2)}{[z+i(E_1-E_2)]}\left[\frac{q_fC_{1f}V^*_{E_1a}(1+\delta_{E_1}/q_f)}{(\delta_{E_1}+i)}+W_{E_1a}C_{2f}\right]\times\left[\frac{q_fC^*_{1f}V_{E_2a}(1+\delta_{E_2}/q_f)}{(\delta_{E_2}+i)}+W^*_{E_2a}C^*_{2f}\right]\frac{2\pi s}{\Gamma}\ , \tag{4.44}$$

with

$$S_{E_i}=\|m\|(z)\Big|_{z=-i(E_i-E_f)}\ , \tag{4.45}$$

$$\delta_{E_i}=\frac{2}{\Gamma}(E_i-E_a)\ . \tag{4.46}$$

Note that in writing (4.44) we had taken the unperturbed matrix elements to be energy independent. The expression (4.44) can be simplified using the procedure of Davies and Seaton.[21] To carry out the E_1 integral, we first carry out a complex integral in the k plane, remembering that $\zeta(E)$ has a factor e^{ikr_0} and in the limit $r_0\to\infty$, $z_1\to 0$, only the pole at $E_1=E_1+iz$ contributes. Thus, (4.44) reduces to

$$P_f=4\pi\int\frac{dE_2\zeta(E_2)\zeta^*(E_2)}{|S_{E_2}|^2}\left|\frac{q_fC_{1f}V^*_{Ea}(1+\delta_{E_2}/q_f)}{(\delta_{E_2}+i)}+W_{Ea}C_{2f}\right|^2\frac{2\pi s}{\Gamma}\ . \tag{4.47}$$

Substituting (4.25) in (4.47) and using the relationship

$$\delta(x)=\lim_{\epsilon\to 0}\frac{\epsilon}{\pi(x^2+\epsilon^2)}\ , \tag{4.48}$$

we obtain (for small values of β)

$$P_f \simeq 4\pi \left| \frac{q_f C_{1f} V^*_{E_0 a}(1+\delta_{E_0}/q_f)}{(\delta_{E_0}+i)} + W_{E_0 a} C_{2f} \right|^2 \frac{2\pi s}{\Gamma |S_{E_0}|^2} , \tag{4.49}$$

where

$$\delta_{E_0} = (2/\Gamma)(E_0 - E_a) . \tag{4.50}$$

In (4.50), E_0 is the energy of the incident wave packet. Upon using Eqs. (4.14), (4.15), (4.34), and (4.35) and defining quantities

$$q_{f1} = \frac{V_{fa}}{\pi V_{E_0 a} V_{f E_0}} , \tag{4.51}$$

$$\Gamma_1 = 2\pi |V_{E_0 a}|^2 , \tag{4.52}$$

we find that the Eq. (4.49) can be written in the form

$$P_f = \frac{4\gamma_f}{\Gamma_1 q_{f1}^2} \left[\left[\delta_{E_0} + \frac{\Gamma_1}{\Gamma} q_{f1} \right]^2 + \left[1 - \frac{\Gamma_1}{\Gamma} \frac{q_{f1}}{q_f} \right]^2 \right] \times \frac{1}{\psi^2 [\eta^2 + (\delta_{E_0} - \Delta_a)^2]} , \tag{4.53}$$

where

$$\psi = 1 + \frac{\gamma_f}{\Gamma q_f^2} + s\pi |C_{2f}|^2 = 1 + s\pi (|V_{f E_0}|^2 + |W_{f E_0}|^2) ,$$

$$\Delta_a = -\frac{2\gamma_f}{\Gamma q_f \psi} ,$$

$$\eta = 1 + \frac{\gamma_f}{\Gamma \psi} \left[1 - \frac{1}{q_f^2} \right] , \tag{4.54}$$

$$\gamma_f = 2s |V_{af}|^2 ,$$

$$s = \frac{2}{3} \frac{\omega^3}{c^3} .$$

The expression (4.53) was also recently obtained by Haan and Jacobs[6] using the projection-operator approach. From (4.54) we see that while η depends on the coupling of the unperturbed continuum states $|\psi_E\rangle$ and $|\chi_E\rangle$ and the autoionizing state to $|f\rangle$, ψ soley depends on the coupling of $|\psi_E\rangle$ and $|\chi_E\rangle$ to $|f\rangle$. Note that parameters such as Γ and q_f given in (4.7) and (4.15) depend on the coupling of the autoionizing state to the continuum states $|\psi_E\rangle$ and $|\chi_E\rangle$. The physical interpretation of Δ_a is that it represents a shift in the energy of the autoionizing state due to the continuum-continuum coupling.

The plot of P_f as a function of δ_{E_0} is shown in Figs. 2 and 3. In Fig. 2 we present the variation of the recombination probability P_f as a function of the incident energy parameter δ_{E_0} for various values of spontaneous emission rate γ_f. From the figure we see an increase in the value of P_f with increase in γ_f. This is to be expected, as increase in spontaneous decay leads to a greater transfer

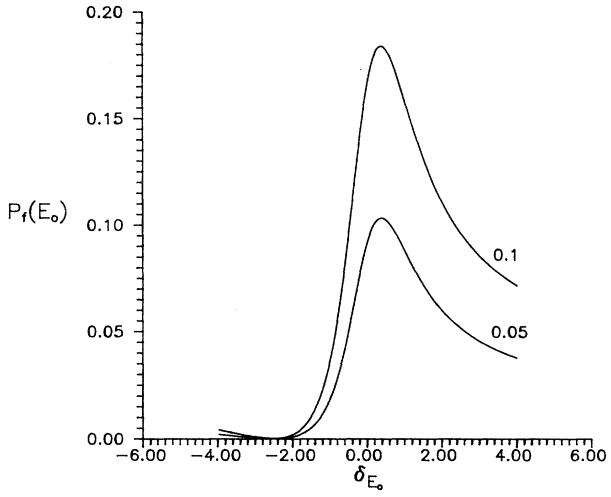

FIG. 2. Variation of recombination probability as a function of incident energy for indicated values of γ_f; other parameters are taken to be $q_f=2$, $q_{f1}=5$, $\Gamma=1$, $\Gamma_1=0.5$.

of population from the initial state $|\psi_E\rangle$ to the bound state $|f\rangle$. Figure 3 shows the comparison of the values of P_f for recombination involving a single continuum ($\Gamma_2=0$) and for two continua ($\Gamma_2\neq 0$). From the figure it is evident that the recombination probability for the latter case is less; a similar result was found by Haan.[7] Increase in Γ_2 leads to an increase in transfer of population into the second continuum and hence a decrease in population to the state $|f\rangle$. We next discuss the various fundamental processes that have contributed to the recombination probability of Eqs. (4.53). These processes are shown in Figs. 4(a)–4(c):

(a) The situation shown in Fig. 4(a) corresponds to the

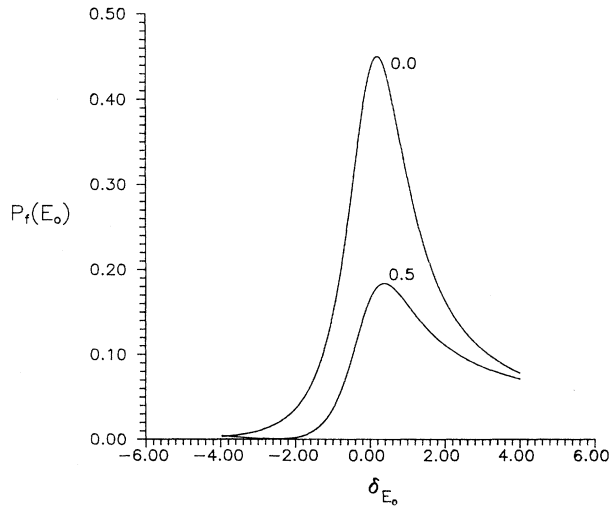

FIG. 3. Comparison of recombination probability as a function of incident energy for a single continuum ($\Gamma_2=0$) and for a two-continuum ($\Gamma_2=0.5$) system for parameters $q_f=2$, $q_{f1}=5$, $\gamma_f=0.1$.

case where the spontaneous emission from the initial state $|\psi_E\rangle$ as well as from the autoionizing state $|a\rangle$ is zero; in such a case the only contribution to the recombination probability comes from the transition $|\chi_E\rangle \rightarrow |f\rangle$. The recombination probability for this case $[P_{f1}(E_0)]$ is obtained by putting

$$V_{E_0 f} = V_{af} = 0 , \tag{4.55}$$

whereby Eq. (4.53) reduces upon using (4.54) to

$$P_{f1}(E_0) = \frac{4\pi s \Gamma_1 (\Gamma - \Gamma_1) |W_{E_0 f}|^2}{\Gamma^2 \psi^2 (\eta^2 + \delta_0^2)} , \tag{4.56}$$

with the appropriate limits [Eq. (4.55)] taken for ψ and η.

(b) The other process is shown in Fig. 4(b). In this the electrons from the state $|\psi_E\rangle$ make a transition to the AI state $|a\rangle$ and from there decay to the state $|f\rangle$ (DR process). To obtain the recombination probability $[P_{f2}(E_0)]$ for this process, we put

$$W_{E_0 a} = W_{E_0 f} = V_{E_0 f} = 0 , \tag{4.57}$$

in which case the expression (4.53) now has the form

$$P_{f2}(E_0) = \frac{8s |V_{af}|^2}{\Gamma(\eta^2 + \delta_0^2)} . \tag{4.58}$$

(c) Another possibility shown in Fig. 4(c) corresponds to the case where the electrons from the initial state $|\psi_E\rangle$ directly decay to the state $|f\rangle$ (RR process) To obtain the recombination probability $[P_{f3}(E_0)]$ for this process, we put the matrix elements

$$W_{E_0 a} = V_{E_0 a} = W_{E_0 f} = 0 \tag{4.59}$$

to obtain the expression

$$P_{f3}(E_0) = 4\pi s |V_{E_0 f}|^2 / (1 + \pi s |V_{E_0 f}|^2) . \tag{4.60}$$

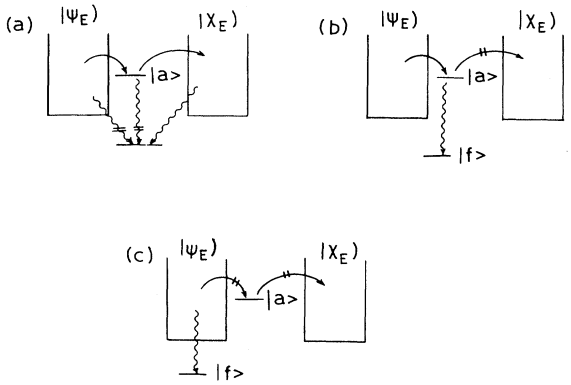

FIG. 4. Schematic diagram of the various fundamental processes leading to recombination.

We see on comparison of (4.18) with (4.56), (4.58), and (4.60) that

$$P_f(E_0) \neq P_{f1}(E_0) + P_{f2}(E_0) + P_{f3}(E_0) . \tag{4.61}$$

From (4.61) it is evident that the recombination probability apart from having contributions from the fundamental processes shown in Fig. 4 also has terms that arise due to interference between these fundamental processes.

V. TOTAL RECOMBINATION PROBABILITY

We next calculate the total recombination probability to the state $|f\rangle$ defined by the relation

$$\langle P_f \rangle = \frac{1}{\Delta E} \int_{-\Delta E/2}^{\Delta E/2} P_f(E) dE , \tag{5.1}$$

where ΔE is taken to be much greater than the total autoionization width, i.e., $\Delta E \gg \Gamma/2$. Substituting (4.53) in (5.1), we have

$$\langle P_f \rangle = \int_{-\Delta\delta_{E_0}/2}^{\Delta\delta_{E_0}/2} \frac{4\gamma_f}{\Gamma_1 q_{f1}^2} \left[\left[\delta_{E_0} + \frac{\Gamma_1}{\Gamma} q_{f1} \right]^2 + \left[1 - \frac{\Gamma_1}{\Gamma} \frac{q_{f1}}{q_f} \right]^2 \right] \frac{(\Delta\delta_0)^{-1} d\delta_0}{\psi^2 [\eta^2 + (\delta_{E_0} - \Delta_a)^2]}$$

$$= \frac{4\gamma_f}{\Gamma_1 \psi^2 q_{f1}^2} + \frac{4\gamma_f (\Delta\delta_{E_0})^{-1}}{\Gamma_1 q_{f1}^2 \psi^2} \int_{-\infty}^{\infty} \frac{d\delta_{E_0} \{\delta_{E_0} [(2\Gamma_1/\Gamma) q_{f1} + 2\Delta_a] + [(\Gamma_1/\Gamma) q_{f1}]^2 - \eta^2 - \Delta_a^2 + [1 - (\Gamma_1 q_{f1}/\Gamma q_f)]^2\}}{[\eta^2 + (\delta_{E_0} - \Delta_a)^2]} . \tag{5.2}$$

Carrying out the integral in (5.2), we get

$$\langle P_f \rangle = \frac{4\gamma_f}{\Gamma_1 \psi^2 q_{f1}^2} + \frac{4\pi\gamma_f}{\Gamma_1 \psi^2 q_{f1}^2 (\Delta\delta_{E_0}) \eta} \times \left[\Delta_a \left[\Delta_a + \frac{2\Gamma_1}{\Gamma} q_{f1} \right] + \left[\frac{\Gamma_1}{\Gamma} q_{f1} \right]^2 + \left[1 - \frac{\Gamma_1 q_{f1}}{\Gamma q_f} \right]^2 - \eta^2 \right] . \tag{5.3}$$

Using Eqs. (4.56), (4.58), and (4.60) in (5.1), we find that

$$\langle P_{f1}(E_0) \rangle = \frac{4\pi^2 s \Gamma_1 (\Gamma - \Gamma_1) |W_{E_0 f}|^2}{\Delta\delta_0 \Gamma^2 \psi^2 \eta} , \tag{5.4}$$

$$\langle P_{f2}(E_0) \rangle = \frac{8\pi s |V_{af}^2|^2}{\Delta\delta_0 \Gamma \eta} , \tag{5.5}$$

$$\langle P_{f3}(E_0) \rangle = 4\pi s |V_{E_0 f}|^2 / (1 + \pi s |V_{E_0 f}|^2) . \tag{5.6}$$

Here the first term (5.4) gives the contribution to the total recombination probability due to the presence of the second continuum, the second term (5.5) is the contribu-

tion coming from the dielectronic-recombination process, and the third term (5.6) represents the radiative recombination contribution. Note that the total recombination probability (5.3) is not equal to the sum of (5.4), (5.5), and (5.6). This is due to the interference among different pathways leading to the final state $|f\rangle$.

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APPENDIX A: DERIVATION OF EQ. (2.14)

In this appendix we present briefly the derivation of (2.14) even though master equations of the type (2.14) are well known in quantum optics literature.[20] In the interaction picture (2.10) can be written as

$$\dot{\rho}'_{A+R}(t)=-iL'_{AR}(t)\rho'_{A+R}(t) , \tag{A1}$$

where

$$L'_{AR}(t)=\exp[+i(L_A+L_R)t]L_{AR}\exp[-i(L_A+L_R)t] ,$$
$$\rho'_{A+R}(t)=\exp[+i(L_A+L_R)t]\rho_{A+R}(t) . \tag{A2}$$

In order to obtain a dynamical equation for the reduced density matrix, we introduce a projection operator $\mathcal{P}$ defined by

$$\mathcal{P}\ldots=\rho_R(0)\mathrm{Tr}_R\cdots . \tag{A3}$$

On multiplying (A1) on the left by $\mathcal{P}$ and by $(1-\mathcal{P})$ and on using $\mathcal{P}+(1-\mathcal{P})=1$, we obtain

$$\mathcal{P}\dot{\rho}'_{A+R}(t)=-i\mathcal{P}L'_{AR}(t)\mathcal{P}\rho'_{A+R}(t) -i\mathcal{P}L'_{AR}(t)(1-\mathcal{P})\rho'_{A+R}(t) , \tag{A4}$$

$$(1-\mathcal{P})\dot{\rho}'_{A+R}(t)=-i(1-\mathcal{P})L'_{AR}(t)(1-\mathcal{P})\rho'_{A+R}(t) -i(1-\mathcal{P})L'_{AR}(t)\mathcal{P}\rho'_{A+R}(t) . \tag{A5}$$

Clearly, (A5) can be integrated for $(1-\mathcal{P})\rho'_{A+R}(t)$ in terms of $\mathcal{P}\rho'_{A+R}(t)$. This result when used in (A4) will lead to an exact closed equation for $\mathcal{P}\rho'_{A+R}(t)$. However, for recombination problems we do not need an equation for $\mathcal{P}\rho$ to all orders in coupling between the atom and the radiation field. We therefore simplify using straightaway the approximations mentioned in Sec. II. Note that the initial condition for the recombination problem is such that $\rho_{A+R}(0)=\rho_A(0)\rho_R(0)$, which implies that $(1-\mathcal{P})\rho_{A+R}(0)=0$. Moreover, the initial state of the field is vacuum and hence $\mathcal{P}L'_{AR}(t)\mathcal{P}=0$. Assuming that the interaction between the vacuum field and the atom is weak, we restrict our analysis to second order in the interaction H_{AR}. From (A5) we get

$$(1-\mathcal{P})\rho'_{A+R}(t)=-i\int_0^t(1-\mathcal{P})L'_{AR}(\tau)\mathcal{P}\rho'_{A+R}(\tau)d\tau , \tag{A6}$$

which on substitution in (A4) leads to

$$\mathcal{P}\dot{\rho}'_{A+R}(t)=-\int_0^t d\tau\,\mathcal{P}L'_{AR}(t)(1-\mathcal{P})L'_{AR}(\tau)\mathcal{P}\rho'_{A+R}(\tau) . \tag{A7}$$

On using (A3), Eq. (A7) simplifies to

$$\dot{\rho}'_A(t)=-\int_0^t d\tau\,\mathrm{Tr}_R L'_{AR}(t)L'_{AR}(t-\tau)\rho_R(0)\rho'_A(t-\tau) . \tag{A8}$$

The range of time integration in (A8) is determined by the time-correlation function—in particular, correlation time τ_c of the vacuum field. For the radiation field it is known that the correlation function is nonvanishing over a time interval which is much smaller than the time over which the atomic system evolves. Hence it is a good approximation to replace $\rho'_{A+R}(t-\tau)$ by $\rho'_A(t)$ and take the limit $t\to\infty$. Note further that for the spontaneous emission problem τ_c is so small that one can hardly expect to observe the behavior of the system for times of the order of τ_c. Thus times of interest are much bigger than τ_c and Eq. (A8) reduces to Eq. (2.14), i.e.,

$$\dot{\rho}'_A(t)+\lim_{t\to\infty}\int_0^t d\tau\,\mathrm{Tr}_R L'_{AR}(t)L'_{AR}(t-\tau)\rho_R(0)\rho'_A(t)=0 . \tag{A9}$$

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