Vacancy sharing in strongly asymmetric heavy-ion collisions

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(Received 15 August 1986)

A complex-energy, complex-interaction Nikitinlike model is proposed to describe the vacancy-sharing process when many quasimolecular states are strongly coupled. The vacancy-sharing ratio is obtained in closed analytical form. The general result is used to calculate the L-K vacancy sharing in the $C^+ + Ar$, $N^+ + Ar$, and $O^+ + Ar$ collisions.

The inner-shell vacancies created in close ion-atom collisions are subject to a redistribution between the colliding partners in the outgoing stage of the collision. This vacancy-sharing process, which usually occurs at larger internuclear distances R, is governed by radial couplings between occupied and unoccupied inner-shell molecular orbitals (MO) of the system. The vacancy sharing has been extensively studied both experimentally and theoretically in the last 10—15 years (see, e.g., Refs. 1—3). The investigations have shown that in near-symmetric colliding systems, the $2p\sigma$ vacancy created in the united atom region ($R \rightarrow 0$) (by a $2p\pi$-$2p\sigma$ rotational coupling, for instance), is shared between K shells of collision partners due to the $2p\sigma$-$1s\sigma$ radial coupling.[1-3] Two-state close-coupling models of Demkov's[4] or Nikitin's[5] type were found to be appropriate for describing the K-K vacancy sharing.[1-3] In strongly asymmetric systems, however, when the K shell of the lighter partner may become degenerate (or nearly degenerate) with the L, M, or N shells of the heavier collision partner, the vacancy created in the $R \rightarrow 0$ region is subject to a sharing process which usually involves many mutually strongly coupled states. Thus, in the case of an asymmetric system in which the K shell of the lighter partner is more strongly bound than the L shell of the heavier partner (as in the N+Ar, O+Ar, and Ne+Ar systems, for instance), the $3d\sigma$ vacancy is shared between the 4σ, 3σ, and 2σ adiabatic MO's at large R, while at smaller R (e.g., $R < 0.5a_0$ for the Ne-Ar system, a_0 being the Bohr radius), the $3d\sigma$ vacancy passes through strong-coupling (avoided crossing) regions involving 6σ-5σ, 5σ-4σ, and 4σ-3σ adiabatic MO's. In the case of K-M or K-N level-matching asymmetric systems the number of radially strongly coupled states is even larger. A similar multistate coupling situation takes place also in the L-K, L-M, etc., vacancy-sharing processes. In all these situations, for calculating the distribution of vacancies among different adiabatic MO's, one has to solve numerically a multistate dynamical problem. The only such calculations using realistic radial couplings have so far been performed for the K-L vacancy sharing in the Ne-Kr system[6] ($3d\sigma$ vacancy essentially shared between 4σ and 3σ adiabatic MO's) and for the L-K vacancy sharing in the Cl+Kr and Ar+Kr systems[7] (4σ-3σ-$2s$ MO's included). As pointed out in Ref. 3 (cf. p. 227), the fair agreement with the experiments[7,8] in the case of L-K sharing may be somewhat fortuitous.[9] For the L-K vacancy sharing in the C+Ar, N+Ar, and O+Ar systems, three-state close-coupled calculations have been carried out by using scaled hydrogenic radial coupling matrix elements,[10] but poor agreement with the experimental data[11,12] for the R_{LK} vacancy-sharing ratio was obtained. Attempts[2,10] to interpret the L-K vacancy sharing in C+Ar, N+Ar, and O+Ar systems (and the K-L for B+Ar) in terms of the two-state Nikitin model have also failed, pointing to the inadequacy of this model to describe the multistate vacancy-sharing process.

In the present paper we propose a different approach for describing the multistate vacancy-sharing process. The approach utilizes the idea[13,14] of reducing the multistate close-coupling problem to the one for two strongly coupled quasistationary states interacting via a complex interaction. The reduction procedure has recently been accomplished elsewhere[13,14] under rather general conditions (sufficiently high density of interacting states). Written in a diabatic representation of molecular states, the coupled equations for the amplitudes C_1 and C_2 of the hole states $|\psi_1^d\rangle$ (which correlates to a given hole state in the $\mathscr{L}_K$ shell of one of the partners) and $|\psi_2^d\rangle$ (which correlates to a given hole state in the $\mathscr{L}_L$ shell of the other partner) have the form[13,14]

$$i\dot{C}_1 = -\frac{i}{2}\Gamma_1(t)C_1 + \left[H_{12} - \frac{i}{2}\Gamma_{12}\right] \exp\left[-i\int_0^t w(t')dt'\right] C_2 , \tag{1a}$$

$$i\dot{C}_2 = -\frac{i}{2}\Gamma_2(t)C_2 + \left[H_{21} - \frac{i}{2}\Gamma_{21}\right] \exp\left[i\int_0^t w(t')dt'\right] C_1 , \tag{1b}$$

$$w = H_{22} - H_{11} . \tag{1c}$$

The quantities Γ_i and Γ_{ij} are, respectively, the decay width of the state $|i\rangle$ due to its interaction with all the states from the group $\{\psi_j\}$, and the interaction of $|i\rangle$ and $|j\rangle$ through other (virtual) states. The diabatic representation $\{\psi_{1,2}^d\}$ in a half-collision problem like ours is inadequate to represent the molecular states in the region of small R (or $t \approx 0$) due to their large adiabatic mixing. It is therefore natural to transform Eqs. (1) into an adiabatic basis $\{\psi^a_{1,2}\}$, using the unitary transformation $\{\psi^a\}=U\{\psi^d\}$, where[3,10]

$$U=\begin{bmatrix}\cos(\chi/2) & \sin(\chi/2)\\ -\sin(\chi/2) & \cos(\chi/2)\end{bmatrix},\quad \tan\chi=\frac{2H_{12}}{H_{22}-H_{11}}, \tag{2}$$

and $H_{ij}=\langle\psi^d_j|H|\psi^d_i\rangle$. Choosing now that at $t=0$ the vacancy is on the adiabatic state ψ^a_2, our initial conditions for the system (1) are

$$C_1(0)=-\sin\left[\frac{\chi(0)}{2}\right],\quad C_2(0)=\cos\left[\frac{\chi(0)}{2}\right]. \tag{3}$$

With the above initial conditions, the vacancy-sharing ratio between the states $|1\rangle$ and $|2\rangle$ is given by

$$R=\frac{|C_1(+\infty)|^2}{|C_2(+\infty)|^2}. \tag{4}$$

A natural generalization of the Nikitin model to quasistationary states is the following:

$$\omega(t)=\Delta\epsilon[1-\beta\exp(-\alpha|t|)],\quad H_{12}=H_{21}=V\exp(-\alpha|t|), \tag{5a}$$

$$\tfrac{1}{2}[\Gamma_1(t)-\Gamma_2(t)]=-\Delta\Gamma\exp(-\alpha|t|),\quad \Delta\Gamma=\Gamma^0_2-\Gamma^0_1, \tag{5b}$$

$$\tfrac{1}{2}\Gamma_{12}(t)=\tfrac{1}{2}\Gamma_{21}(t)=\Lambda\exp(-\alpha|t|),\quad \Lambda=(\Gamma^0_1\Gamma^0_2)^{1/2}, \tag{5c}$$

where Γ^0_1 and Γ^0_2 are the half-widths of Γ_1 and Γ_2 at $t=0$, $\Delta\epsilon=|I_1-I_2|$ is the difference of ionization potentials I_1 and I_2 of considered states, $V=$const is the strength of the direct coupling between $|1\rangle$ and $|2\rangle$ at $t=0$, and $\alpha=\gamma v_R$, with $\gamma=2^{-1/2}(I_1^{1/2}+I_2^{1/2})$ and v_R being the radial component of collision velocity v. (Hereafter we use atomic units). Equation (5a) constitutes the standard Nikitin's model, with the parameter β related to the "Nikitin angle" θ by $\tan\theta=2V/\beta\Delta\epsilon$. In the straight-line trajectory approximation and for zero impact parameter ($v_R=v$), the coupled equations (1) with the generalized Nikitin model (5) can be solved exactly.[15] For the ratio $C_1(+\infty)/C_2(+\infty)$ one obtains

$$\frac{C_1(+\infty)}{C_2(+\infty)}=\frac{\Gamma(2i\lambda)}{\Gamma(-2i\lambda)}e^{2\pi\lambda\xi}\frac{1+\nu}{1-\nu}\frac{\Gamma[-i\lambda(1+\nu)]}{\Gamma[i\lambda(1-\nu)]}e^{-2i\lambda\ln(-2i\lambda B/\nu)}\times\frac{T^0_1-T^0_2e^{iQ}\dfrac{1-\nu}{1+\nu}\dfrac{\Gamma[i\lambda(1-\nu)]}{\Gamma[i\lambda(1+\nu)]}e^{-i\pi\lambda\xi}e^{i\lambda\nu[\ln(2i\lambda B/\nu)+\ln(-2i\lambda B/\nu)]}}{\tilde{T}^0_1-\tilde{T}^0_2e^{iQ}\dfrac{1+\nu}{1-\nu}\dfrac{\Gamma[i\lambda(1+\nu)]}{\Gamma[-i\lambda(1-\nu)]}e^{\pi\lambda\xi}e^{i\lambda\nu[\ln(2i\lambda B/\nu+\ln(-2i\lambda B/\nu)]}}, \tag{6}$$

$$\lambda=\frac{\Delta\epsilon}{2\alpha},\quad B=\beta+i\frac{\Delta\Gamma}{\Delta\epsilon},\quad \xi=\operatorname{sgn}\arg\left[2i\lambda\frac{B}{\nu}\right], \tag{7a}$$

$$\nu=B\left[B^2+4\frac{(V-i\Lambda)^2}{\Delta\epsilon^2}\right]^{-1/2}, \tag{7b}$$

$$Q=-\frac{\Delta\epsilon}{\alpha}\left\{\left[\frac{B^2}{\nu^2}-2B+1\right]^{1/2}-\nu\ln\left[\frac{B}{\nu}-\nu+\left[\frac{B^2}{\nu^2}-2B+1\right]^{1/2}\right]-\ln\left[1-B+\left[\frac{B^2}{\nu^2}-2B+1\right]^{1/2}\right]+\nu+\nu\ln\left[\frac{2B}{\nu}\right]+\ln\left[B\left[\frac{1}{\nu}-1\right]\right]\right\}, \tag{7c}$$

$$T^0_{1,2}=\left[\frac{1+\cos[\chi(0)]}{2\eta(0)}\right]i\Gamma^a_{12}(0)+\frac{\sin[\chi(0)]}{2\eta(0)}[\eta(0)\mp\Delta E(0)], \tag{7d}$$

$$\tilde{T}^0_{1,2}=\frac{1+\cos[\chi(0)]}{2\eta(0)}[\eta(0)\mp\Delta E(0)]\pm\frac{\sin[\chi(0)]}{2\eta(0)}i\Gamma^a_{12}(0), \tag{7e}$$

$$\sin[\chi(0)]=-\frac{2V}{[4V^2+(1-\beta)^2\Delta\epsilon^2]^{1/2}},\quad \cos[\chi(0)]=\frac{(1-\beta)\Delta\epsilon}{[4V^2+(1-\beta)^2\Delta\epsilon^2]^{1/2}}, \tag{7f}$$

$$\Delta E(0)=[4V^2+(1-\beta)^2\Delta\epsilon^2]^{1/2}-i\frac{\Delta\Gamma\Delta\epsilon(1-\beta)+4\Lambda V}{[4V^2+(1-\beta)^2\Delta\epsilon^2]^{1/2}}, \tag{7g}$$

$$\Gamma^a_{12}(0)=2\frac{\Lambda\Delta\epsilon(1-\beta)-V\Delta\Gamma}{[4V^2+(1-\beta)^2\Delta\epsilon^2]^{1/2}}, \tag{7h}$$

$$\eta(0)=\{\Delta E^2(0)-[\Gamma^a_{12}(0)]^2\}^{1/2}. \tag{7i}$$

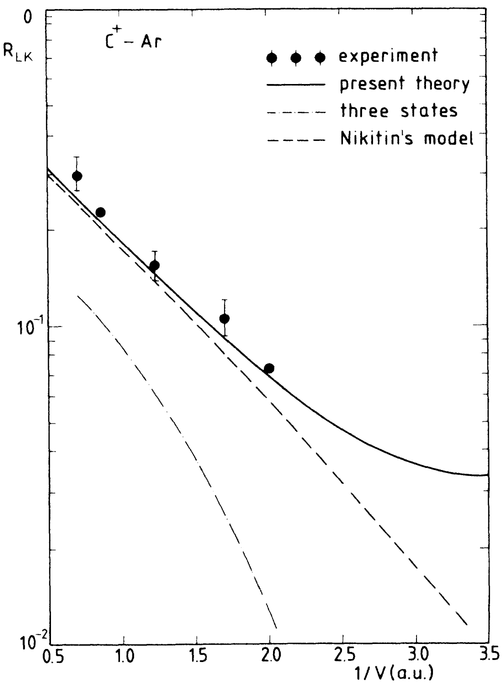

FIG. 1. L-K vacancy-sharing ratio for the $C^+ + Ar$ system as a function of $1/v$. Solid circles are experimental data (Refs. 11 and 12). Theoretical calculations: solid line, present model: dashed line, Nikitin's model (Ref. 11); dot-dashed line, three-state close-coupled calculations (Ref. 10).

For $\Gamma_1^0 = \Gamma_2^0 = 0$, from Eqs. (6), (7), and (4) one obtains

$$R = e^{-2\pi\lambda}\frac{\sinh[\pi\lambda(1+\mu)]}{\sinh[\pi\lambda(1-\mu)]}, \quad \mu = \frac{\beta}{(\beta^2 + 4V^2/\Delta\epsilon^2)^{1/2}}, \tag{8}$$

which is the result of the standard Nikitin model.[5]

The complexity of the result (6) and (7) with respect to Eq (8) arises due to the fact that in case of decaying states it is not sufficient to give only the initial conditions in the adiabatic representation, but also to solve the adiabatic-transformed Eqs. (1) in the region of small R (or t) and then to match these solutions with those of Eqs. (1). (The matching procedure, as usual, is accomplished by using the asymptotics of the corresponding solutions in the region of t where they overlap.) By setting $\beta = 0$ in Eqs. (7) and (6), one obtains a result corresponding to the complex-energy, complex-interaction Demkov model. The ratio R, calculated with the expression (6) for $C_1(+\infty)/C_2(+\infty)$, exhibits very fast oscillations as a function of $1/v$. These oscillations (coming mainly from the terms $e^{\pm iQ}$) are the result of interference effects due to the interaction Γ_{12} between the states $|1\rangle$ and $|2\rangle$ through intermediate (or virtual) states. Writing $\mathrm{Re}Q = q/v$ and

$$\frac{C_1(+\infty)}{C_2(+\infty)} = \frac{A - Be^{iq/v}}{C + De^{iq/v}}, \tag{9}$$

where A, B, C, and D are some slowly varying functions

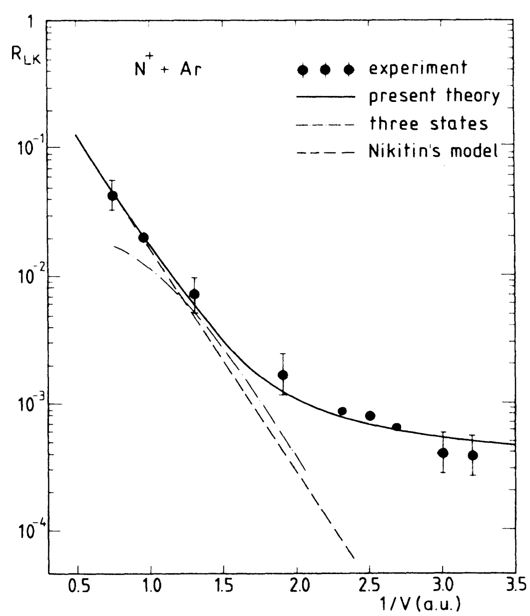

FIG. 2. Same as Fig. 1, but for the $N^+ + Ar$ system.

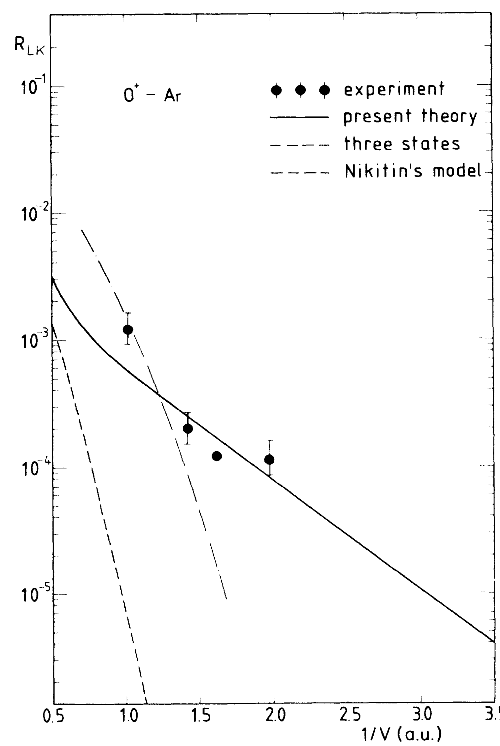

FIG. 3. Same as Fig. 1, but for the $O^+ + Ar$ system.

of $1/v$, the averaging over the oscillations gives the following expression for R:

$$R=\frac{|A|^2+|B|^2}{||C|^2-|D|^2|}\times\left[+\frac{2|A|^2|D|^2}{(|A|^2+|B|^2)(|C|^2+|D|^2)}\mathrm{Re}\left[\frac{BC}{AD}\right]\right]. \quad (10)$$

The application of the present model to real physical situations presumes in principle a sufficient density of neighboring states to which the considered ones, $|1\rangle$ and $|2\rangle$, are coupled, so that the decay widths Γ_i can be introduced as in the case of interaction of a discrete state with a quasi-continuum of states.[14,16] When there is only a finite number of decay channels l for the state $|i\rangle$, Γ_i can be calculated as[16]

$$\Gamma_i=2\pi\sum_l |H_{i,l}|^2\frac{1}{\Delta_{l,l'}}, \quad (11)$$

where $\Delta_{ll'}$ is the energy difference between the neighboring states in the group of states $\{\psi_l\}$. $\Gamma_i(R)$, calculated from (11), has to be adjusted to the form (5b). For two given states $|i\rangle$ and $|j\rangle$, the parameters β and V in the Nikitin model (5a) can be determined[10,2] by fitting the adiabatic energy splitting of the model

$$\Delta E_{ij}(R)=[\Delta\epsilon^2(1-\beta e^{-\gamma R})^2+4V^2e^{-2\gamma R}]^{1/2} \quad (12)$$

to accurate molecular-energy calculations, now available for many systems (see, e.g., Refs. 3 and 10). Using such a procedure we have determined the parameters in the generalized Nikitin model for the 4σ-2σ vacancy sharing in the C+Ar, N+Ar, and O+Ar systems (β and V have earlier been determined[11]), and calculated the sharing ratio R from Eqs. (6)—(10). The results are shown in Figs. 1—3 and compared with experimental data.[10,12] The results of the standard Nikitin model[2] and three-state calculations with scaled hydrogenic coupling matrix elements[10] are also shown in these figures. Although the considered L-K vacancy-sharing case is at the edge of applicability of the present model (small number of strongly coupled states), the obtained agreement with the experimental data may be considered as satisfactory. Calculations within the present model of other, more representative collision systems is underway.[15]

Partial support of this work by the Joint U.S.-Yugoslav Board for scientific and technological cooperation under Contract No. NBS(G)-691-*Yu*, is gratefully acknowledged.

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