Surface-Hopping Model for Near-Resonant
Electronic Energy Transfer

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A surface-hopping model was developed to account for the large near-resonant electronic energy transfer cross sections in systems such as NF-Bi and O₂-I, in which the dipole-dipole mechanism is formally forbidden. Multiple surface crossings are predicted to occur in NF-Bi at distances of ≈8 Å, because of long-range (r⁻³) dipole-induced dipole terms in the potential. This corresponds well with the measured transfer cross section of ≈200 Å². These terms are absent in O₂-I, so that the leading
term is the London dispersion interaction with an R dependence of \( R^{-6} \).
The predicted crossing distance is 3.5 Å, in agreement with the smaller transfer cross section for this system. A Landau-Zener crossing probability is used to predict the temperature dependence of the thermally averaged cross section \( Q(T) \) for the \( I \)-atom laser pumping reaction,

\[
O_2*(a^1\Delta) + I(2P_{3/2}) \rightarrow O_2(X^3\Sigma^-) + I*(2P_{1/2})
\]

Requirements for a more accurate calculation are discussed.
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I. INTRODUCTION

A recent measurement\(^1\) of the rate for the near-resonant electronic energy-transfer reaction

\[
\text{Bi}(^{4}S_{3/2}) + \text{NF}^*(a^{1}\Delta) \rightarrow \text{Bi}^*(^{2}D_{3/2}) + \text{NF}(X^3\Sigma) (\Delta E = 16 \text{ cm}^{-1})
\]  \(1\)

has yielded a value of \(1 \times 10^{-9} \text{ cm}^3\text{ molecule}^{-1}\text{ sec}^{-1}\), corresponding to a thermally averaged cross section \(\approx 200 \text{ Å}^2\), which is much greater than gas kinetic. The theoretical problem posed by this result is that the electronic transitions involved in this process are both strongly dipole forbidden \((A_{21} = 31 \text{ and } 1 \text{ sec}^{-1} \text{ for the Bi and NF transitions, respectively})\). Thus, the model of interacting transition dipoles, usually employed for such processes, cannot account for the observed efficiency. In this report, we discuss an alternative model, based on potential surface crossings at large interparticle distances, which successfully accounts for the observed rate.

An understanding of these processes is important for several reasons. Transfer reactions, e.g., Reaction (1), are the basis for the recently developed metastable transfer emission spectroscopy technique,\(^2\) which is a powerful method for determining trace components in gas mixtures.\(^3\) A similar reaction,

\[
I(^{2}P_{3/2}) + O_2^*(a^{1}\Delta) \rightarrow I^*(^{2}P_{1/2}) + O_2(X^3\Sigma^-) (\Delta E = 279 \text{ cm}^{-1})
\]  \(2\)

is the basis for the chemically pumped O\(_2\)-I transfer laser.\(^4\) In order to optimize the performance of this system and analogous potential laser systems, it is necessary to be able to model the dependence of the key transfer step, Reaction (2), on such parameters as energy defect, reactant states, and gas temperature.
The inadequacy of the transition dipole-dipole interaction mechanism was previously pointed out by Breckenridge and co-workers\textsuperscript{5} for the case of Cd*-NO and Zn*-NO energy transfer. They suggested an alternative mechanism, such as direct or charge-transfer curve crossing, as an explanation. A similar model has been proposed by Setser and co-workers\textsuperscript{6} for quenching of metastable rare-gas atoms. In this report, NF*-Bi and O$_2^*$-I transfer processes are discussed in terms of a mechanism of this type.
II. SURFACE CROSSINGS IN NF*-Bi

The electronic energy levels associated with the low-lying states of Bi and NF are shown schematically in Fig. 1. These states will effect a variety of triatomic potential surfaces at small Bi-NF distances; in addition, a strongly attractive surface will result from the Bi$^+$-NF$^-$ charge-transfer state. This attractive ionic surface will cut through the neutral surfaces arising from Bi($^4$S) + NF($^4$Δ) and Bi($^2$D) + NF($^3$Σ) at a Bi-NF separation $R_0$ given by

$$U(R_0) = [I. P. (Bi) - E. A. (NF)] - E_{exc}$$

where I. P. is ionization potential, E. A. is electron affinity, and $E_{exc}$ is the energy of the electronically excited levels (1.42 eV). The interaction potential $U(R)$, in this point-charge approximation, includes contributions from both coulomb attractive and charge-induced-dipole terms, and, thus is given by

$$U(R) = -\frac{e^2}{R} - \frac{e^2\alpha}{R^4}$$

In order to find $R_0$, we need to know the ionization potential of bismuth, the electron affinity of NF, and the polarizability $\alpha$ of the most polarizable species in the system, which will be the negative ion NF$^-$. The ionization potential of Bi is known spectroscopically$^7$ to be 7.28 eV, but the other two quantities are not experimentally determined. An ab initio calculation$^8$ gives E. A. (NF) $\approx$ 0.6 eV; we accept this value because the ionization potential of NF, calculated from the same wave functions, is 13.2 eV, in excellent agreement with the experimental value (13.1 eV).$^9$ The polarizability of
Fig. 1. Low-Lying Electronic States of Bi and NF and Attractive Curve Resulting from Interaction of Bi$^+$ and NF$^-$
NF" is taken to be slightly larger than that of O₂, \( ^{10} \text{O}_2 \), i.e., \( 2 \times 10^{-24} \text{ cm}^3 \).

Combining Eqs. (3) and (4), we obtain

\[
- \frac{e^2}{R_o} - \frac{e^2 \alpha}{R_o^3} = 7.28 - 0.6 - 1.42 \text{ eV}
\]

\[
= 5.26 \text{ eV}
\]

\[
= 8.43 \times 10^{-12} \text{ erg}
\]

The required value of \( R_o \) is 2.95 Å, which is much too small to account for the observed efficiency of Reaction (1). The corresponding cross section, 27 Å², is nearly an order of magnitude smaller than the measured cross section. Thus, the charge-transfer surface cannot be important in the non-adiabatic surface-crossing process, and we focus attention instead on the direct crossing of the two surfaces correlating with \( \text{NF}^*(a^1 \Delta) + \text{Bi}(^4S_{3/2}) \) and with \( \text{NF}(X^3\Sigma) + \text{Bi}(^2D_{3/2}) \).

A schematic representation of these surfaces, at a fixed N-F distance, is shown in Fig. 2. The symmetries of the intermediate nonlinear Bi-NF complexes must be determined from adiabatic correlation rules. Since the heavy atoms are in a j-coupled limit (only one spin-orbit component interacts with each molecular state), Hund's Case C coupling is appropriate for the complex. For \( \text{Bi}(^4S_{3/2}) + \text{NF}(^1 \Delta) \), \(^4A'\) and \(^4A''\) surfaces will be obtained. The \( \text{Bi}(^2D_{3/2}) + \text{NF}(\Sigma) \) interaction will result in \(^4A', \; ^4A'', \; ^2A'\), and \(^2A''\) states. Thus, multiple surface crossings are possible. Attention is focused on one possible crossing, shown in Fig. 2, in order to estimate the crossing distance \( R_o \).

The leading term in the long-range potential between these two species will be the dipole-induced-dipole interaction,
Fig. 2. Avoided Crossing of Bi-NF* and Bi*-NF Potential Surfaces. The adiabatic surface sections, shown as dotted lines, lie in the region indicated by the circle in Fig. 1.
The dipole moments and polarizabilities are needed for both the NF and Bi ground and electronically excited states. We take $\alpha(Bi) \approx 2 \times \alpha(I)$ $[Z = 53] \approx 2 \times (5.1 \times 10^{-24} \text{cm}^3)^{12} \approx 10^{-23} \text{cm}^3$ and $\alpha(Bi^*) \approx 3 \times \alpha(Bi) \approx 3 \times 10^{-23} \text{cm}^3$. The dipole moment of NF$^1(\Delta)$ has been measured to be $0.37 \text{D} = 3.7 \times 10^{-19} \text{esu-cm}$. The previously cited calculation gives $\mu(NF^3\Sigma) - \mu(NF^1\Sigma) = 0.24 \text{D}$. We, thus, take $\mu(NF^3\Sigma) \approx 6.1 \times 10^{-19} \text{esu-cm}$ (the experimental value is not known). With the use of these values, $R_0$ was estimated from

$$\frac{e}{R^5} (\mu*\alpha + \mu\alpha*) = 16 \text{cm}^{-1}$$

$$= 3.18 \times 10^{-15} \text{erg}$$

where the $*$ refers to the property of the electronically excited species.

With the above values, it is found that $R_0 \approx 8 \text{Å}$, corresponding to a cross section of $200 \text{Å}^2$.

The agreement of this calculated value with the experimental one (200 $\text{Å}^2$) is, of course, fortuitous. Most of the parameters used to determine this value are rough estimates, and the experimental value itself has an error bracket of $\pm 50\%$. Nevertheless, the fact that the two numbers are the same indicates that the basic model for the process is essentially correct. This model also accounts satisfactorily for the roughly five times smaller rate for the similar exchange process

$$I^*(2P_{3/2}) + NF^1(\Delta) \rightarrow I^*(2P_{3/2}) + NF^*(\Sigma^+)(\Delta E = 110 \text{cm}^{-1})$$

(6)
The explanation is simply that the larger energy defect in Reaction (6) results in a smaller crossing distance $R_o$ and a correspondingly smaller transfer cross section.
III. NEAR-RESONANT ELECTRON ENERGY TRANSFER
IN O₂*-I SYSTEM

The successful demonstration of the O₂*-I transfer laser system has provided a strong motivation for understanding near-resonant electronic energy-transfer processes, such as the excitation transfer, Reaction (2). A model for this process similar to that employed for the NF*-Bi system is considered, and Landau-Zener theory is then used to estimate a temperature dependence for the rate of this reaction.

A. POTENTIAL SURFACE CROSSINGS

The electronic energy levels associated with the low-lying states of I and O₂ are shown schematically in Fig. 3. As in the NF*-Bi case treated earlier, a charge-transfer surface (arising from I⁻ and O₂⁺) will intersect the neutral surfaces (arising from I⁺ + O₂, I + O₂*). The crossing distance \( R₀ \) is estimated from Eqs. (3) and (4), with I.P. \((O₂) = 12.06 \text{ eV}, \) \( E.A. (I) = 3.06 \text{ eV}, \) \( E.A. (O₂) = 0.5 \text{ eV}, \) the surface arising from \( I^+ + O₂^- \) lies \( \approx 1 \text{ eV} \) above that from \( I^- + O₂^+ \), and thus intersects the \( \text{I}^*-\text{O}_2 \) surfaces at an even smaller \( R₀ \). The polarizability of \( I^- \) is \( 6.2 \text{ Å}^3 \). Thus, we have

\[
\frac{\frac{2}{R₀}}{\frac{2}{R₀}} - \frac{\frac{2}{R₀}}{\frac{4}{R₀}} = 12.06 - 3.06 - 0.94 \text{ eV}
\]

\[
= 8.06 \text{ eV}
\]

\[
= 1.29 \times 10^{-11} \text{ erg}
\]

or a value of \( R₀ = 2.5 \text{ Å} \). Whereas this does yield an estimate of the cross section for Reaction (2) in agreement with the experimental value of \( 15 \text{ Å}^2 \), it is more likely that the process is dominated by the direct surface crossing at larger \( R₀ \), as shown in Fig. 3.
Fig. 3. Low-Lying Electronic States of I and O₂ and Attractive Curve Resulting from Interaction of I⁻ and O₂
For $\text{I} + \text{O}_2$, none of the species has a permanent dipole moment, so that the leading term in the long-range interaction is the London dispersion energy\(^\text{17}\)

$$U_{AB}(R) = -\frac{3}{2} \frac{(\text{IP})_A(\text{IP})_B}{(\text{IP})_A + (\text{IP})_B} \frac{\alpha_A \alpha_B}{R^6} \tag{7}$$

From the adiabatic correlation rules,\(^\text{11}\) it is found that the interaction of $\text{I}^*(2\,^2\text{P}_{1/2})$ with $\text{O}_2(3\Sigma^-)$ results in $^4\text{A}'$ and $^2\text{A}'$ surfaces, whereas $\text{I}^*(2\,^2\text{P}_{3/2}) + \text{O}_2^*(a^1\Delta_g)$ results in several $^2\text{A}'$ and $^2\text{A}''$. Thus, the avoided crossing will be between surfaces of $^2\text{A}'$ symmetry. A suggested model for the crossing region is shown in Fig. 4.

The surface sections $U_1(R)$ and $U_2(R)$, shown in Fig. 4, are given by Eq. (7), with the appropriate molecular parameters. In order to find the required ionization potentials, it is necessary to subtract the correct excitation energy; thus,

$$\text{I. P.} (\text{O}_2^*) = \text{I. P.} (\text{O}_2) - E(a^1\Delta)$$

$$= 12.06 \text{ eV}^{15} - 0.98 \text{ eV} = 11.08 \text{ eV}$$

and

$$\text{I. P.} (\text{I}^*) = \text{I. P.} (\text{I}) - E(2\,^2\text{P}_{1/2})$$

$$= 10.45 \text{ eV}^{7} - 0.94 \text{ eV} = 9.51 \text{ eV}$$

The polarizability of O$_2$ is $1.6 \times 10^{-24}$ cm$^3;^{10}$ we take $\alpha(\text{O}_2^*) \approx 2\alpha(\text{O}_2) \approx 3.3 \times 10^{-24}$ cm$^3.^{13}$ The polarizability of an I atom is $5.1 \times 10^{-24}$ cm$^3.^{12}$
Fig. 4. Avoided Crossing of I-O₂* and I*-O₂ Potential Surfaces. The adiabatic surface sections, shown as dotted lines, lie in the region indicated by the circle in Fig. 3.
Since the $^2P_{1/2}$ state is a spin-orbit component with the same configuration as the ground ($^2P_{3/2}$) state, we take $\alpha(I^*)$ only slightly larger than $\alpha(I)$, viz., $6 \times 10^{-24}$ cm$^3$. These choices give

$$U_1(R) = \frac{1.095 \times 10^{-42} \text{ cm}^{-1}}{R^6} \quad (8a)$$

and

$$U_2(R) = \frac{6.17 \times 10^{-43} \text{ cm}^{-1}}{R^6} \quad (8b)$$

with R in cm. To find $R_0^2$, we set $U_1(R_0^2) = U_2(R_0^2) + 279 \text{ cm}^{-1}$, which gives $R_0 = 3.45$ Å. The cross section $\pi R^2$ is, thus, 37.4 Å$^2$, compared to the experimental value\textsuperscript{19-21} of $(15 \pm 5)$ Å$^2$. A more precise comparison of this model with experiment requires the use of Landau-Zener theory for the actual surface-crossing efficiency, which is discussed in the following section.

B. CALCULATION OF MAGNITUDE AND TEMPERATURE DEPENDENCE OF EXCITATION TRANSFER PROBABILITY IN I + O$_2^*$

The Landau-Zener model\textsuperscript{22,23} has been widely used to calculate non-adiabatic curve-crossing probabilities in molecular collisions. Several reformulations of the model have recently been presented;\textsuperscript{24,25} the model has also been compared with the method of analytic continuation across a crossing seam.\textsuperscript{26} The quantities pertinent to this model are defined in Fig. 5. "Splitting" of the adiabatic surfaces in the vicinity of $R_0$ is identified as $V_{12}$. As the system approaches on surface 1, the probability of remaining on that surface is given by
Fig. 5. Definitions of Quantities Required in Landau-Zener Calculation
\[ P_{LZ} = \exp \left( - \frac{2\pi |V_{12}|^2}{\hbar |U'_1 - U'_2| v} \right) \]  

(9)

where \( U'_1 = \frac{dU_1(R)}{dR} \bigg|_{R=R_0} \), \( U'_2 = \frac{dU_2(R)}{dR} \bigg|_{R=R_0} \), and \( v \) is the relative velocity of the colliding species (I and \( O_2 \), in this case). Since a net transition from surface 1 to surface 2 requires two traversals of the region shown in Fig. 5, with the system "crossing" on one passage and following the adiabatic surface on the other, the net probability of excitation transfer is

\[ P_{12}(v) = 2P_{LZ} (1 - P_{LZ}) \]  

(10)

The thermally averaged transfer cross section is then

\[ Q(T) = \pi R_0^2 \frac{\int_{v_1}^{v_2} P_{12}(v) f(v) \, dv}{\int f(v) \, dv} \]  

(11)

with \( f(v) \) given by the Maxwell-Boltzmann velocity distribution,

\[ f(v) \, dv = 4\pi \left( \frac{\mu}{2\pi kT} \right)^{3/2} \exp \left( - \frac{\mu v^2}{2kT} \right) v^2 \, dv \]  

(12)

For the \( O_2-I \) system considered here, with \( U_1(R) \) and \( U_2(R) \) given by Eqs. (8a, b), \( U'_1 = 2.43 \times 10^{-5} \) dyn, \( U'_2 = 1.40 \times 10^{-5} \) dyn, so that \( |U'_1 - U'_2| \approx 10^{-5} \) dyn, evaluated at \( R_0 = 3.45 \times 10^{-8} \) cm. Since there is no value for \( V_{12} \) estimates of 10, 30, and 100 cm\(^{-1}\) are used (i.e., of the order of the London dispersion interaction itself). The reduced mass \( \mu \) of the I-\( O_2 \) collision pair is \( 4.24 \times 10^{-23} \) g.

The results of a numerical integration of Eq. (11) are shown in Fig. 6. It is clear that a value of \( V_{12} \approx 30 \) cm\(^{-1}\) gives the correct magnitude for the cross section, whereas estimates much above or much below this figure lead
Fig. 6. Temperature Dependence of Thermally Averaged Cross Section $Q(T)$ for $O_2^*$-I Excitation Transfer
to gross underestimates. The temperature dependence predicted by this model is particularly striking. The temperature variation in $Q(T)$ over the entire temperature range, 100 to 500 K, is smaller than the stated uncertainty in the measured value at 300 K. Thus, before embarking on an experimental program designed to measure this temperature dependence, it would be advisable to reduce the experimental uncertainties in the room-temperature measurement, in order to obtain meaningful results.

The performance of the $O_2$-I transfer laser is a function of the excitation transfer reaction, Reaction (2), and the quenching reaction

$$I^*\left(^2P_{1/2}\right) + O_2(a^1\Delta) \rightarrow I\left(^2P_{3/2}\right) + O_2(b^1\Sigma^+) (\Delta E = 2364 \text{ cm}^{-1}) \quad (13)$$

The rate of this reaction is reported to be $k_{13} = 1.6 \times 10^{10} \text{ cm}^3 \text{ mole}^{-1} \text{ sec}^{-1}$, or $Q(300 \text{ K}) \approx 5 \times 10^{-19} \text{ cm}^2$. Whereas a calculation similar to the foregoing could be attempted for the quenching cross section, the many uncertainties in the required molecular parameters would make the calculation of such a small cross section of doubtful significance. Qualitatively, it is found that cross sections of this type, with magnitudes much less than $1 \text{ Å}^2$, are steeply rising functions of temperature. Thus, it is suspected that the efficiency of the $O_2$-I transfer laser will be improved by operation at reduced temperatures, with the small increase in the transfer cross section (shown in Fig. 6) being accompanied by a much larger relative decrease in that for the quenching reaction, Reaction (13).
In this report, a simple surface-crossing model is discussed, which accounts, at least qualitatively, for the efficiencies of such near-resonant electronic-energy-transfer processes as the excitation of Bi by NF*, Reaction (1), or of I by O₂*, Reaction (2). A more exact quantitative treatment would involve integration of classical collision trajectories on an assumed set of potential surfaces, with the probability of crossing from one electronically adiabatic surface to another given by a Landau-Zener, Eq. (9), or equivalent expression. Such surface-hopping trajectory calculations have been successfully carried out for such systems as H⁺ + H₂,²⁷ H⁺ + D₂,²⁸ I₂ + rare gas (→2I + rare gas),²⁹,³⁰ N₂(B³Π) + Ar [→N₂(W³Δ) + Ar],³¹ and He₂⁺ + Ne.³² In order to describe such energy-transfer processes as Reactions (1) or (2), a surface-hopping inelastic trajectory calculation would have to be carried out. For such a calculation to be meaningful, much more accurate potential surfaces will be required than the simple models that were used in this study. Such surfaces can, in principle, be constructed by a method such as that of diatomics in molecules.³³ If such surfaces, along with improved accuracy in experimental measurements of these rates, were available, then a calculation of these rates by surface-hopping inelastic trajectories would be warranted.
REFERENCES


13. The polarizabilities of electronically excited atoms or molecules are frequently much larger than those of the ground-state species. For example, the polarizability of Ar(1S0) is $1.64 \times 10^{-24}$ cm$^3$ [A. Dalgarno and A. E. Kingston, Proc. R. Soc. of London, A259, 424 (1961)], whereas that for Ar*(3P) is $48.4 \times 10^{-24}$ cm$^3$ [B. Bederson and E. J. Robinson, "Molecular Beams," Adv. Chem. Phys. 10, and J. Ross, ed., Interscience Publishers, New York (1966), p. 25]. Thus, the estimate of $a(Bi*) \approx 3 \times a(Bi)$ is a conservative one.


33. See, for example, the recent calculations of potential energy surfaces for Ne₂F, Ar₂F, Kr₂F, and Kr₂Cl by this method. D. L. Huestis and N. E. Schiotter, J. Chem. Phys. 69, 3101 (1978).
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