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Anticorrelation between yields of recombination luminescence and recombination-induced defect formation in alkali-metal halides

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Coupled rate equations are used to examine the premise that the anticorrelation of *F*-center formation yield and of self-trapped exciton luminescence in alkali-metal halide crystals is a consequence of thermally activated motion of a halogen interstitial atom (*H* center) relative to its complementary vacancy site (*F* center). In our treatment, the anticorrelation depends on relative rates of processes occurring on the lowest adiabatic potential surface connecting self-trapped excitons and *F*-*H* pairs, but is essentially independent of how that surface is populated from higher states. Thermally activated nonradiative decay to the electronic ground state must be disallowed if the anticorrelation is to hold. Conversely, a distinguishing feature of those alkali halides which do not exhibit the anticorrelation is the existence of a nonradiative channel for self-trapped exciton decay to the ground state. Since the present treatment was developed for conditions of thermal equilibrium, it cannot fully address the prompt (~ 10 ps) formation of *F* centers at liquid-helium temperature or the ejection of energetic halogen atoms from the surface, as observed in some alkali halides of the latter group.

I. INTRODUCTION

F-center generation in alkali-metal halides is a classic problem among recombination-induced defect processes. It is known to occur upon nonradiative decay of a self-trapped exciton or recombination of an electron with a self-trapped hole. Progress in understanding the mechanism has been discussed in several reviews.[1,2] It was shown by Pooley *et al.*[3] that the temperature dependence of the yields of radiative transitions from the lowest triplet state of the self-trapped exciton (π-polarized luminescence) and of *F*-center formation are anticorrelated in some alkali halides, as illustrated in Fig. 1(a). These include KI, RbI, NaBr, LiBr, and NaCl, and are called group I in this paper. Based partly on the observation of anticorrelation, Pooley suggested that the *F*-*H* pairs evolve from the lowest electronic state of the self-trapped exciton (STE) because of highly energetic vibrational excitation upon nonradiative transitions to the ground state.[3]

However, the anticorrelation fails to hold in other alkali halides, including KCl and KBr. As shown in Fig. 1(b), the *F*-center yield in these alkali halides is nonzero at low temperature and increases further with increasing temperature. The luminescence is quenched at a temperature lower than that where the *F*-center yield starts to increase. These alkali halides are called group II in this paper. Moreover, the experimental observation that the delay in *F*-center formation in these alkali halides is much smaller than the lifetime of the π-luminescent state[4,5] has ruled out the lowest self-trapped exciton state as the precursor of *F*-center formation at low temperature. Thus it has

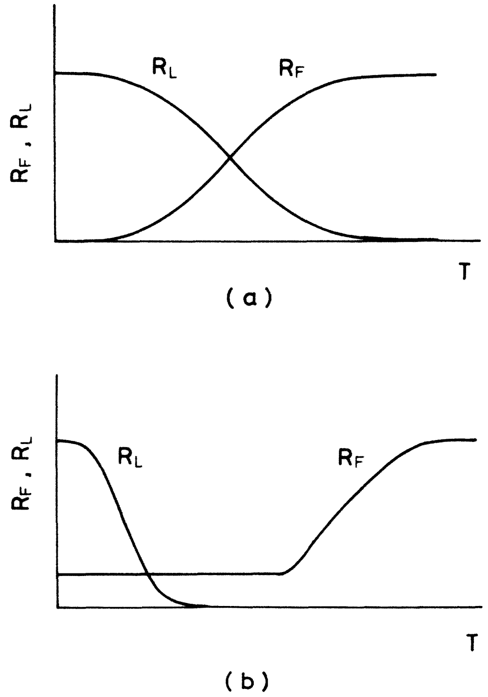

FIG. 1. Schematic diagram for the temperature dependence of the intensity of the π-polarized STE luminescence ($\propto R_L$) and the primary yield of *F* centers ($\propto R_F$) for the two categories of alkali halides labeled in this paper as group I [represented in (a)] and group II [represented in (b)]. The behavior illustrated in (a) is termed "anticorrelation" in this paper.

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been suggested earlier by Kabler,[6] Itoh and Saidoh,[7] and Toyozawa[8] that the *F-H* pairs evolve from an excited state of the self-trapped exciton. This has been referred to as the local excitation mechanism.[9] Recent cascade-excitation experiments have identified the next higher state ($^3\Pi_g^+$, the state with the electron excited to a $2p\pi_u$ orbital) as the most efficient precursor of defects formed at low temperature.[10] This result has been interpreted as indicating that the *F-H* pairs are generated after an Auger transition populates the hole-excited $^3\Pi_g^+$ state.[11] However, this interpretation cannot account for the anticorrelation illustrated in Fig. 1(a) for the group-I alkali halides. Williams, Craig, and Faust have pointed out that the important thermal activation barriers controlling the production of observable *F* centers above $T=4$ K must lie on the adiabatic potential surface connecting the lowest STE and close *F-H* pair configurations.[12,13] The compatability of an excited-state mechanism with the known characteristics of thermally activated defect formation is examined in this paper, using the steady-state solutions of coupled rate equations to describe the populations of neighboring minima on the lowest STE/*F-H* adiabatic potential surface after that surface is populated from higher electronic states.

II. THE MODEL

A. Potential curves

In Fig. 2 we illustrate the adiabatic potential surface which includes the π-luminescent STE state and ground-state *F-H* pairs. The surface contains the activation barriers determining the temperature dependence of *F-H* pair formation and the π-luminescence quenching. In the figure the abscissa, R_{FH}, is the separation between the *F* center and the *H* center. It will be noted that Fig. 2 presents two different possible shapes for the potential curve near $R_{FH}=0$. Figure 2(a) corresponds to the "V_K+e^-" or on-center model of the self-trapped exciton. It is the model first suggested by Kabler[14] and by Murray and Keller,[15] and has been widely used in interpreting STE data. It assumes that the lowest-energy configuration of the STE luminescent state occurs when the diatomic halogen molecular ion is situated symmetrically about the midpoint between two halide sites along a [110] direction ($R_{\text{FH}}=0$). The defect point symmetry is then D_{2h}, as in the V_K center. The V_K center, being stable at low temperature, has been thoroughly characterized and is known to exist in the D_{2h} symmetric site.

However, optically detected ENDOR (electron-nuclear double resonance) of the STE π-luminescent state in KCl,[16] together with excited-state absorption spectra in a variety of alkali halides, have led to the suggestion of an "off-center" model of the lowest-energy STE state.[17] This model has been supported by the calculations of STE electronic structure in the work of Song *et al.*,[18] and has recently been discussed in relation to *F-H* pair production by Williams, Song, Faust, and Leung.[19,20] In the treatment of Ref. 18, the π-luminescent STE state and the nearest-neighbor *F-H* pair are virtually identical and correspond to the first minimum in Fig. 2(b). The V_K+e^- on-center configuration at $R_{FH}=0$ is actually a local

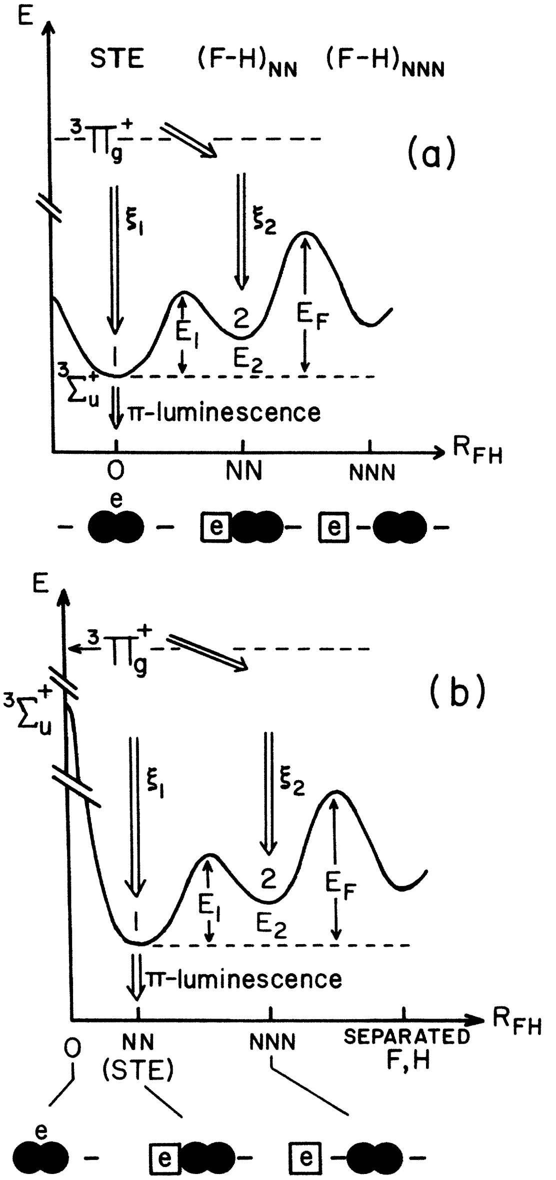

FIG. 2. (a) Adiabatic potential curves connecting the configurations for the self-trapped exciton and the close *F-H* pair along the reaction coordinate R_{FH} describing the separation of the *F-H* pairs. The solid curve is the adiabatic potential curve connecting the lowest state of the self-trapped exciton ($^3\Sigma_u^+$ in the V_K+e^- on-center model) with the ground states of the *F* center and *H* center at pair configurations labeled nearest-neighbor (NN) and next-nearest-neighbor (NNN). The dashed curve is broadly representative of excited states of the STE and defect pair; for example, the expectation of a rather flat curve passing through the STE excited state $^3\Pi_g^+$ is represented here. At the bottom of the figure the schematic configurations for STE, $(F\text{-}H)_{\text{NN}}$ and $(F\text{-}H)_{\text{NNN}}$ are shown, where (-) denotes a halide ion; the square, a vacancy; the closed double circle, a $(\text{halogen})_2^-$ molecular ion; and *e*, an electron. (b) Alternative view of the potential curves based on the off-center model of the STE, as developed in Refs. 16—20. The V_K+e^- configuration is in this case an energy maximum at $R_{FH}=0$. The STE luminescent state is identified with the first minimum (NN or STE), and is virtually identical with a nearest-neighbor *F-H* pair (Ref. 20).

maximum of energy in the treatment of Ref. 18, about 1 eV above the minimum. It is not the purpose of this paper to prove or disprove either of the above-mentioned models for the STE. In fact, the conclusions of the present paper will hardly be affected by which of the two figures is assumed, because both lead to the same set of rate equations, to be described below. The differences in the two diagrams lie mainly in the labeling of the minima and in the behavior when $R_{FH}=0$. In Fig. 2(a), $R_{FH}=0$ is the configuration identified as the self-trapped exciton. In Fig. 2(b) it corresponds to a local energy maximum, and the adjacent minimum at approximately the nearest-neighbor *F*-*H* separation is the off-center STE suggested to be responsible for the recombination luminescence.[18-20] The similarities of both part (a) and part (b) are that the first energy minimum at the left is the STE, and successive minima proceeding to the right represent successively larger separations of the *F* center and *H* center.

We denote the lowest-excited state at the self-trapped configuration by 1 and that at the second minimum by 2. We assume that the electron transition at the third minimum and beyond exhibits the *F*-band optical absorption. We denote the potential energy at the saddle-point configuration between the states 1 and 2 by E_1, the energy maximum between the second and third minima by E_F, and the minimum at state 2 by E_2. For the group-II alkali halides, *F*-*H* pairs are known to be formed when the next higher ${}^3\Pi_g^+$ state is populated upon laser excitation of ${}^3\Sigma_u^+$. In the present discussion we intend to examine a model wherein the data for alkali halides of both groups I and II at various temperatures can be consistent with defect formation proceeding out of an excited state (e.g., ${}^3\Pi_g^+$), by way of population in states 1 and 2. We therefore assume for the present discussion that the *F*-center formation process begins with population of self-trapped excitons (state 1) and of intermediate defect pairs (state 2) from the same initial state in all alkali halides, as denoted by the double arrows in Fig. 2. We investigate how the magnitudes of E_1, E_2, E_F, and E_{1g} modify the properties relevant to STE luminescence and *F*-*H* pair formation.

B. Rate equations

Upon formation of a highly excited self-trapped exciton or recombination of an electron with a self-trapped hole,[21] a series of nonradiative transitions will lead to the π-luminescent state, the intermediate defect pair, and the σ-luminescent state. We denote the number of these states populated per unit time under continuous irradiation by ξ_1, ξ_2, and ξ_3, respectively, where $\xi_1+\xi_2+\xi_3=1$. It is known that all of the group-I alkali halides exhibit the σ-polarized luminescence, while some of the group-II alkali halides do not. We assume that the branching ratios into the intermediate defect pair (state 2), the σ-luminescent state, and the π-luminescent state (state 1) are not temperature dependent. This assumption should be valid since the branching occurs at highly excited states where no thermal equilibrium is reached.[22]

We denote the transition rate to the ground state from state 1 by ν_{1g} and from state 2 by ν_{2g}. We denote also the transition rate from state 1 to 2 by ν_{12}, from state 2 to 1 by ν_{21} and from state 2 to the separated *F*-*H* pair by ν_{2F}. The populations n_1 and n_2 of the excited states at 1 and 2 are given by solving the following differential equations:

$$\frac{dn_1}{dt}=\xi_1-\nu_{1g}n_1-\nu_{12}n_1+\nu_{21}n_2 , \tag{1}$$

$$\frac{dn_2}{dt}=\xi_2-\nu_{2g}n_2-(\nu_{21}+\nu_{2F})n_2+\nu_{12}n_1 . \tag{2}$$

We will assume that quasiequilibrium is established in the populations n_1 and n_2 under continuous irradiation, so that

$$dn_1/dt=dn_2/dt=0 .$$

Then we obtain

$$n_1=A^{-1}[\xi_1(\nu_{2g}+\nu_{21}+\nu_{2F})+\xi_2\nu_{21}] , \tag{3}$$

$$n_2=A^{-1}[\xi_1\nu_{12}+\xi_2(\nu_{1g}+\nu_{12})] , \tag{4}$$

where

$$A=\nu_{1g}(\nu_{2g}+\nu_{21}+\nu_{2F})+\nu_{12}(\nu_{2g}+\nu_{2F}) . \tag{5}$$

The luminescence intensity is proportional to the product of the population n_1 and the *radiative* transition rate ν_{1g}^R,

$$I_L \propto \nu_{1g}^R n_1=R_L ,$$

where

$$\nu_{1g}=\nu_{1g}^R+\nu_{1g}^N(T) \tag{6}$$

is the sum of a temperature-independent radiative rate and a thermally activated nonradiative rate ν_{1g}^N,

$$\nu_{1g}^N=\nu_{1g}^0 \exp\left[-\frac{E_{1g}}{kT}\right] . \tag{7}$$

The barrier E_{1g} is difficult to show in Fig. 2, which depicts only the coordinate R_{FH}. E_{1g} is the energy of the attempted crossing of the STE luminescent state and the electronic ground state, relative to the energy minimum of the luminescent state.

Thus,

$$R_L=\nu_{1g}^R n_1=A^{-1}[\xi_1\nu_{1g}^R(\nu_{2g}+\nu_{21}+\nu_{2F})+\xi_2\nu_{1g}^R\nu_{21}] . \tag{8}$$

Similarly, the formation rate of recognizable *F* centers is

$$R_F=\nu_{2F}n_2=A^{-1}[\xi_1\nu_{2F}\nu_{12}+\xi_2\nu_{2F}(\nu_{1g}+\nu_{12})] . \tag{9}$$

III. RESULTS AND DISCUSSION

A. Group-I alkali halides

We first inquire what the necessary conditions are for anticorrelation to occur; that is, for the sum R_L+R_F to be independent of temperature. Under the conditions

$$\nu_{1g}=\nu_{1g}^R \text{ and } \nu_{2g}\ll\nu_{2F} \tag{10}$$

we obtain

$$R_L = B^{-1}[\xi_1(\nu_{21}+\nu_{2F})+\xi_2\nu_{21}]\ , \tag{11}$$

$$R_F = B^{-1}[\xi_1\nu_{12}\nu_{2F}/\nu_{1g}^R+\xi_2(\nu_{2F}+\nu_{12}\nu_{2F}/\nu_{1g}^R)]\ , \tag{12}$$

where

$$B = \nu_{21}+\nu_{2F}+\nu_{12}\nu_{2F}/\nu_{1g}^R\ . \tag{13}$$

It can be seen that

$$R_L+R_F = \xi_1+\xi_2\ . \tag{14}$$

The conditions in Eq. (10) amount to closing off all channels from the STE/*F*-*H* adiabatic potential surface to the electronic ground state other than luminescence. The conditions in Eq. (10) are sufficient for anticorrelation, defined by the relation $R_L+R_F=$ const, to occur. However, they are not sufficient to reduce Eq. (11) to a form that corresponds to the observed temperature dependence of luminescence quenching. It is observed that in group-I alkali halides the temperature dependences of the STE luminescence and of the *F*-center yield have the form

$$I_L \propto R_L = \frac{K_1}{1+K_2\exp(-E/kT)} \tag{15}$$

and

$$R_F = \frac{K_1}{1+K_2^{-1}\exp(E/kT)}\ , \tag{16}$$

where E is an activation energy.

With the further conditions

$$\nu_{21} \gg \nu_{2F} \quad \text{and} \quad \nu_{12} \gg \nu_{1g}^R \tag{17}$$

applied to Eqs. (11)–(13), we recover equations of the observed form (15) and (16), where

$$K_1 = \xi_1+\xi_2 \tag{18}$$

and

$$K_2\exp(-E/kT) = \frac{\nu_{12}\nu_{2F}}{\nu_{21}\nu_{1g}^R}\ . \tag{19}$$

The correspondence of the exponential factors on both sides of Eq. (19) with reference to the individual barriers defined in Fig. 2 gives $E=E_F$.

Whereas the conditions in Eq. (10) prescribed isolation of the STE/*F*-*H* potential surface from all decay channels except radiative decay, the conditions in Eq. (17) prescribe that states 1 and 2 communicate with each other more rapidly than with any of the other available configurations. That is, self-trapped excitons and nearest-neighbor *F*-*H* pairs transform *reversibly* from one to the other many times while the processes of radiative decay and conversion to more stable *F*-*H* pairs (the third minimum and beyond in Fig. 2) are occurring. Note, furthermore, that in group-I alkali halides, the activation barrier E_F which governs quenching of self-trapped exciton luminescence has nothing to do with the barrier E_{1g} for crossing nonradiatively to the electronic ground state. We have in fact had to assume that the latter channel is nonexistent or has a very high activation barrier in order to obtain anticorrelation of R_L and R_F for group-I alkali halides.

B. Group-II alkali halides

In group-II alkali halides, as illustrated in Fig. 1(b), there is thermally activated quenching of STE luminescence and there is a thermally activated increase of *F*-center formation at some higher temperature, but the two processes are not correlated. Furthermore, there is typically a finite temperature-independent yield of *F*-center formation at low temperature.

We return to Eqs. (8) and (9). In the general case,

$$\nu_{1g} = \nu_{1g}^R+\nu_{1g}^N(T)$$

since anticorrelation is not observed in group II.

Again making the assumptions (for sake of simplicity) that

$$\nu_{2g} \ll \nu_{2F},\ \ \nu_{21} \gg \nu_{2F},\ \ \text{and}\ \ \nu_{12} \gg \nu_{1g}^R \tag{20}$$

we obtain

$$R_L = \nu_{1g}^R n_1 = \frac{\xi_1+\xi_2}{\nu_{1g}/\nu_{1g}^R+\nu_{12}\nu_{2F}/(\nu_{1g}^R\nu_{21})}\ , \tag{21}$$

$$R_F = \nu_{2F} n_2 = \frac{\xi_1+\xi_2}{1+\nu_{1g}\nu_{21}/(\nu_{12}\nu_{2F})}\ . \tag{22}$$

The equations for R_L and R_F no longer have the symmetry that was evident in Eqs. (15) and (16). There are two competing processes tending to quench the luminescence rate R_L: activation over barrier E_{1g} (rate ν_{1g}) and activation over barrier E_F (rate $\nu_{12}\nu_{2F}/\nu_{21}$). The smaller barrier will govern the quenching of luminescence. In KCl, $E_{1g}=7$ meV (Ref. 23) and $E_F=75$ meV.[5,24] From all available evidence, $E_F > E_{1g}$ in group-II alkali halides.

The rate R_F in Eq. (22) is governed at temperatures above the onset of luminescence quenching by the single effective barrier E_F-E_{1g}, replacing the barrier E_F in Eq. (17) for group-I crystals. Actually, the attribution $E_F=75$ meV for KCl in the paragraph immediately above is not quite correct. An activation energy of 75 meV was observed for the *F*-center yield at high temperature.[5,24] Therefore, $E_F-E_{1g}=75$ meV, or $E_F=82$ meV. Values of the activation energy for *F*-center formation at high temperature in other group-II alkali halides can be interpreted similarly. Note that as long as $E_F > E_{1g}$ (where $E_F=E_{12}+E_{2F}-E_{21}$ from Fig. 2), R_F increases with increasing temperature, as observed.

Our discussion of Eq. (22) has been specifically for the case of moderately high temperatures where thermally activated *F*-center formation in group-II crystals comes into play. On further consideration of Eq. (22), it appears that in order for finite *F*-center yield to be observed at low temperature (e.g., in the range of liquid-helium temperature), the relative barrier heights would have to satisfy the relation

$$E_{21}+E_{1g}-E_{12}-E_{2F} > 0\ . \tag{23}$$

Therefore (see Fig. 2)

$$E_{1g}-E_F > 0\ . \tag{24}$$

Equation (24) is not consistent with the experimental observation that the luminescence quenching occurs at lower temperature than the increase in the F-center yield. The finite F-center yield at low temperature is therefore not describable by the present model, which assumed thermal equilibrium and steady-state conditions for the solution of Eqs. (1) and (2). Furthermore, F-center formation occurring in thermal equilibrium at such low temperature could not account for the ejection of halogen atoms from the surface with ~ 1 eV of kinetic energy, as observed in group-II alkali halides at low temperature.[25]

Therefore, we must conclude that there remains the need for a mechanism which can couple electronic energy of STE excited states into athermal kinetic energy of a halogen atom in group-II alkali halides at low temperature. Excited-state mechanisms discussed by Kabler,[6] Itoh and Saidoh,[7] Toyozawa,[8] and Itoh, Stoneham, and Harker[11] address this mechanism. Most recently, consideration of the off-center configuration of the self-trapped exciton[18] has suggested a mechanism by which the shape of the lower STE/F-H adiabatic potential surface in the neighborhood of $R_{FH}=0$ may effectively channel up to 1 eV of the energy of excited states into translational energy of the H center.[19,20]

IV. SUMMARY

Whereas the excited-state mechanisms are properly concerned with defect formation at low temperature in group-II crystals, we have shown in this paper that thermally activated F-center formation in both group-I and group-II crystals can be satisfactorily described in terms of a single model for the motion of an H center relative to its initial site (the F center or former STE), while it is on the STE/F-H adiabatic potential surface. In particular, the thermally activated process of F-center formation appears insensitive to how the excited-state population arrives on the STE/F-H potential surface, i.e., whether state 1 or state 2 is occupied first (see Fig. 2). Indeed, according to the conditions in Eq. (17) it is immaterial which state is populated first because they must rapidly communicate with each other if the anticorrelation behavior is to be observed. It has been shown that in a consistent treatment of the anticorrelation of STE luminescence yield and lifetime (13) with F-center yield, the STE quenching process cannot be attributed to nonradiative crossing to the electronic ground state. This is in contrast to the most widely held view of the quenching mechanism.[3,23] The competitive channel responsible for quenching is F-H pair separation along the adiabatic potential surface connecting STE and F-H pairs.

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