

Neutral donor-acceptor-pair recombination under a uniform electric field

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The kinetics of the recombination of electrons trapped by donors, with holes trapped by acceptors, in semiconductor materials within a uniform electric field has been investigated. A simple form has been derived which shows that the decay rate of neutral donor-acceptor-pair (DAP) recombination increases under the presence of the electric field. This analysis can also be applied to the processes of neutral DAP recombination in quantum wells and superlattices where electric fields are always present in the interface regions.

I. INTRODUCTION

Neutral donor-acceptor-pair (DAP) recombination in semiconductor materials is an important mechanism in photoluminescence.¹⁻³ Neutral DAP's are formed when ionized donors D^+ (acceptor A^-) capture electrons (holes) which were created by incident photons. There is a certain probability that electrons trapped by donors recombine with the holes trapped by acceptors, which means that the neutral DAP is an unstable system. When a neutral DAP recombines, a photon is emitted and a $D^+ - A^-$ pair left behind. The energy of emitted photons depends on the distance between the donor and acceptor.¹ Photoluminescence corresponding to DAP recombination is therefore called bound-to-bound transition or low-energy series (LES) in contrast with free-to-bound transition or high-energy series (HES).⁴ The kinetics of DAP recombination has been studied by Thomas, Hopfield, and Augustyniak,⁵ and Colbow⁶ both theoretically and experimentally.

Recently, photoluminescence techniques have been widely used to study the optical properties of quantum wells (QW's) and superlattices⁷⁻¹⁰ (SL's). Unlike the DAP recombination inside the bulk of semiconductors, the processes of DAP recombination in QW and SL structures are much more complicated. There are induced electric fields in the interface regions, so the decay rate and energy of the emitted photons of DAP recombination in QW and SL structures depend not only on the distance between donors and acceptors, but also depend strongly on the locations of the neutral donors and acceptors. Because of the surface effects, the distribution of neutral donors and acceptors is not homogeneous. To date, the influence of these surface fields on the DAP recombination is still unknown and has not been previously investigated. The distribution of D^0 and A^0 , and surface fields at the interfaces of these quantum structures are very complicated and unknown. The recombination of neutral DAP's in semiconductors under a uniform electric field has been treated theoretically in this paper. Our results provide some information about the

DAP recombination in QW's and SL's under the effect of the surface field.

The influence of a uniform electric field on the neutral DAP recombination has been studied by Colbow.⁶ The electric field effect was taken into account by considering the action of the field on the impurity wave functions, using a WKB approximation. He assumed that an applied electric field has two fundamentally different effects. The first concerns the ground state of the system; that is, the acceptor-donor pair after the electron-hole recombination. The second effect involves a distortion of the hole and electron wave functions by the applied field, and thus a change in the hole-electron recombination rate. The first effect has been considered only to increase the bandwidth of the pair emission. In Colbow's consideration, the changing of the decay rate of neutral DAP comes only from the changing of the wave functions of donors and acceptors. By comparing the decay rates of DAP recombinations in the presence and absence of an electric field, the dipole moment between $D^+ - A^-$ (after neutral DAP recombination) will also affect the decay rate, because of the interaction between the dipole moment and electric field. This effect has not been considered previously. In fact, this is the most important effect when compared to others.

In this paper, we will calculate the decay rate of neutral DAP recombination in the presence of a uniform electric field by a different method. The effects of distortion of the hole and electron wave functions, the changing of binding energies of donors and acceptors, and the interaction between the dipole moment of $D^+ - A^-$ and the electric field have all been considered. The first two effects were calculated by a variational method which is valid under a weak electric field. The decay rate of DAP recombination in the presence of a uniform electric field increases in a different fashion compared with Colbow's results.⁶ The amount of increase in our calculations of the decay rate is higher than that in Ref. 6. In addition, because of the interaction between the dipole moment and the electric field, there is an upper limit on the angle between the line connecting the donor-acceptor pair and

the axis parallel to the electric field, beyond which the DAP recombination cannot produce photons with particular energies.

II. CALCULATIONS

In the effective-mass approximation, the Hamiltonian of a neutral donor-acceptor pair in the presence of an electric field is

$$H = (-\hbar^2/2m_e)\nabla_e^2 + (-\hbar^2/2m_h)\nabla_h^2 + \frac{1}{2} \sum_{\substack{i,j=1 \\ (i \neq j)}}^4 (q_i q_j / \epsilon_i | \mathbf{r}_i - \mathbf{r}_j |) + \sum_{i=1}^4 q_i \mathbf{r}_i \cdot \mathbf{E} . \quad (1)$$

The first two terms in Eq. (1) are the kinetic energies of the electron and hole with respect to a fixed origin of a coordinate system. The second term is the potential of interactions between particles and the third term is the potential of interactions between particles and electric field \mathbf{E} . In Eq. (1), $q_i = e$ for $i = 1, 4$ and $q_i = -e$ for $i = 2, 3$. $\epsilon_i = \epsilon$ for $i = 1, 3$ and $\epsilon_i = 1$ for $i = 2, 4$. $i = 1, 2, 3$, and 4 correspond to the ionized donor, electron, ionized acceptor, and hole, respectively. With $\mathbf{E} = 0$, the third term equals zero, and Eq. (1) reduces to the Hamiltonian of a neutral donor-acceptor pair in the absence of an electric field. When the separation between the donor and acceptor, $R = |\mathbf{r}_1 - \mathbf{r}_3|$, is much larger than the distances between the electron and donor, $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$, and hole and acceptor, $r_{34} = |\mathbf{r}_3 - \mathbf{r}_4|$ ($R \gg r_{12}, r_{34}$), then we can write Eq. (1) as

$$H = H_1 + H_2 , \quad (2)$$

where H_1 (H_2) is the Hamiltonian for the neutral donor (acceptor) in the presence of an electric field;

$$H_1 = (-\hbar^2/2m_e)\nabla_e^2 - (e^2/\epsilon r_{12}) + e \mathbf{r}_{12} \cdot \mathbf{E} , \quad (3a)$$

$$H_2 = (-\hbar^2/2m_h)\nabla_h^2 - (e^2/\epsilon r_{34}) + e \mathbf{r}_{43} \cdot \mathbf{E} . \quad (3b)$$

In the presence of a uniform electric field, the following three factors affect the neutral DAP recombination: (i) The changing of the binding energies of donors and acceptors, (ii) distortion of the wave functions of electrons and holes which changes the wave function overlapping between electrons and holes, and (iii) the dipole moment of $D^+ - A^-$ after $D^0 - A^0$ recombination that interacts with the electric field. The first effect is not as important as the other two. The third effect is the most important. Here we have neglected the field effect on the distribution of neutral donors and acceptors, i.e., we assume that D^0 and A^0 distribution is uniform in the presence of the electric field. In most cases, the electrons and holes are at the ground state, so we only calculate the DAP decay for both at the ground state. For the first two effects, we used a variational method to solve the binding energies and wave functions of the ground states of D^0 and A^0 under a uniform electric field. Only the calculation for D^0 has been shown here. The calculation for A^0 is exactly the same except that we have to replace the effective Bohr radius, a_D and binding energy, E_D of the donor by a_A and E_A of the acceptor. For simplicity, we also as-

sume that the acceptor level is much deeper than that of donor ($E_A \gg E_D$), so considerably stronger fields are needed to perturb the acceptors. In this paper, we only consider the case where one type of neutral impurity is in excess, say the acceptor impurity. So one donor atom is then regarded as being surrounded by a random array of acceptors and being independent of other donors.

For an isolated donor in an electric field F pointing along the z direction, from Eq. (3), the Hamiltonian in the effective-mass approximation is

$$H = -(\hbar^2/2m)\nabla^2 - e^2/\epsilon r + eFz , \quad (4)$$

where r is the distance between the electron and donor, m is the effective mass of the electron, ϵ is the dielectric constant, and F is the strength of electric field. Using electron Bohr radius a_D as the unit of distance and donor binding energy $E_D (= e^2/2\epsilon a_D)$ as the unit of energy and $e\epsilon^{-1/2}$ as the unit of charge, Eq. (4) becomes

$$H = -\nabla^2 - 2/r + \xi z , \quad (5)$$

where $\xi = eFa_D/E_D$ is a dimensionless parameter representing the electric field strength.

The variational method is to calculate

$$\langle E \rangle = \langle \psi | H | \psi \rangle / \langle \psi | \psi \rangle , \quad (6)$$

and then minimize $\langle E \rangle$, where ψ is a trial wave function.

The trial wave function should become the exact solution for an electron bound to D^+ for $F=0$. Choosing one of the donor centers D^+ as the origin of the coordinate, then the trial wave function should be symmetric about the z axis which is parallel to the direction of the electric field and passes through the origin. Also the wave function and its derivative should be continuous across the $z=0$ plane. With the above restrictions taken into consideration, in the cylindrical coordinate, we choose the following as our trial wave function:

$$\psi = \begin{cases} Ne^{-(\rho^2 + \beta_1 z^2)^{1/2}} , & z > 0 \\ Ne^{-(\rho^2 + \beta_2 z^2)^{1/2}} , & z \leq 0, \end{cases} \quad (7)$$

where N is the normalization constant, and β_1 and β_2 are two variational parameters. For $F=0$, we should have $\beta_1 = \beta_2 = 1$, and Eq. (7) represents the exact solution of the neutral donor. We only give the results here, the detailed calculations can be found in Ref. 11. (The integrations can only be done in the spherical coordinate system.) We have

$$\langle H(\beta_1, \beta_2) \rangle = 2 \left[\sum_{i=1}^2 \beta_i^{-1/2} \right]^{-1} f(\beta) . \quad (8)$$

Taking $\partial H / \partial \beta_i = 0$ ($i = 1, 2$), we obtain

$$\beta_1^{3/2} \frac{\partial f}{\partial \beta_1} = \beta_2^{3/2} \frac{\partial f}{\partial \beta_2} , \quad (9)$$

$$\beta_1^{-1/2} + \beta_2^{-1/2} = -\frac{1}{2} \beta_1^{-3/2} f(\beta) \left[\frac{\partial f}{\partial \beta_1} \right]^{-1} ,$$

where

$$f(\beta) = \sum_{i=1}^2 \left(\frac{1}{6}(2+\beta_i)\beta_i^{-1/2} - \Theta(\beta_i-1)(\beta_i-1)^{-1/2} \tan^{-1}[(\beta_i-1)^{1/2}] \right. \\ \left. - \Theta(1-\beta_i)(1-\beta_i)^{-1/2} \ln\{[1+(1-\beta_i)^{1/2}]\beta_i^{-1/2}\} + \frac{3}{8}\xi(-1)^i\beta_i \right), \\ \frac{\partial f}{\partial \beta_i} = \frac{1}{6}\beta_i^{-3/2}(-1+\beta_i/2) + \frac{3}{8}\xi(-1)^i\beta_i^{-2} \\ + \frac{1}{2}\Theta(\beta_i-1)((\beta_i-1)^{-3/2} \tan^{-1}[(\beta_i-1)^{1/2}] - \beta_i^{-1}(\beta_i-1)^{-1}) \\ - \frac{1}{2}\Theta(1-\beta_i)((1-\beta_i)^{-3/2} \ln\{[1+(1-\beta_i)^{1/2}]\beta_i^{-1/2}\} - \beta_i^{-1}(1-\beta_i)^{-1})$$

($i=1,2$), and $\Theta(x)$ is a step function which is unity for x larger than 0 and 0 otherwise. The variational parameters β_1 and β_2 can be determined from the solutions of Eq. (9). For $F=0$ and $\xi=0$, we have $\beta_1=\beta_2=1$, as they should be.

We have solved Eq. (9) by numerical calculation. Before we present the numerical results, the solution for the weak field (small ξ) will be discussed. For a weak field, both β_1 and β_2 are very close to 1, so we can write $\beta_1=1+b_1$ and $\beta_2=1+b_2$, where b_1 and b_2 are two small values. From Eq. (9), if we only keep to the first order of b_1 and b_2 , we obtain

$$\beta_1 = 1 + 45\xi/22, \\ \beta_2 = 1 - 45\xi/22. \quad (10)$$

Figure 1 depicts the plot of β_1 and β_2 as a function of electric field strength ξ , which is obtained by solving Eq. (9) numerically. Both curves end at $\xi=0.275$. This is because of the fact that there are no solutions satisfying Eq. (9) above this point. Since we have used a bound state wave function as a trial wave function, no bound states exist if the strength of the applied electric field is higher than a certain value. This also tells us that the variational method used here is valid only to a certain degree and it is most suitable for the case of weak electric fields.

We notice from Fig. 1 that for the region of $\xi < 0.15$, β_1 (β_2) linearly increases (decreases) as ξ increases with a slope of 2.05. This agrees with Eq. (10). From Eq. (8), the dependence of the total energy as a function of ξ can be approximately written as $E = -1 - 0.9\xi^2$. Here we point out that the energy shift obtained by this calculation is smaller than the results of the perturbation method. The important results we obtained are the wave functions of an electron trapped by a donor in the presence of the electric field, which is essential for calculating the donor-acceptor-pair recombination rate. The wave functions of electrons and holes obtained here are better than those obtained by Colbow⁶ by using the WKB method. For the case of the weak field, our wave functions are guaranteed by the quantitative agreement of our calculated total energy values with those obtained by Silverstone¹² and Hehenberger *et al.*¹³ However, the total energy values obtained by the WKB method were far from the real values. Now we consider the third effect,

the interaction between the dipole moment of D^+A^- and the electric field. Here we compare the decay rate of neutral DAP recombination with emitted photons at the same energy, $h\nu$, for both in the presence and absence of the electric field. Because of the energy conservation of DAP's before and after recombination, we have the photon energy emitted by the DAP recombination for the cases with and without the electric fields, respectively,

$$h\nu = \begin{cases} E_0 + e^2/\epsilon R, & F=0 \\ E'_0 + e^2/\epsilon R^* - (e\epsilon^{-1/2})FR^* \cos\theta, & F \neq 0, \end{cases} \quad (11)$$

where $E_0 = E_g - (E_D + E_A)$, $E'_0 = E_g - (E_D^* + E_A^*)$, E_g is the energy gap, and E_D^* (E_A^*) is the donor (acceptor) binding energy under the electric field. Here we have assumed that no phonons are involved. R^* is the distance between D^0 and A^0 . The last term in the $F \neq 0$ case of

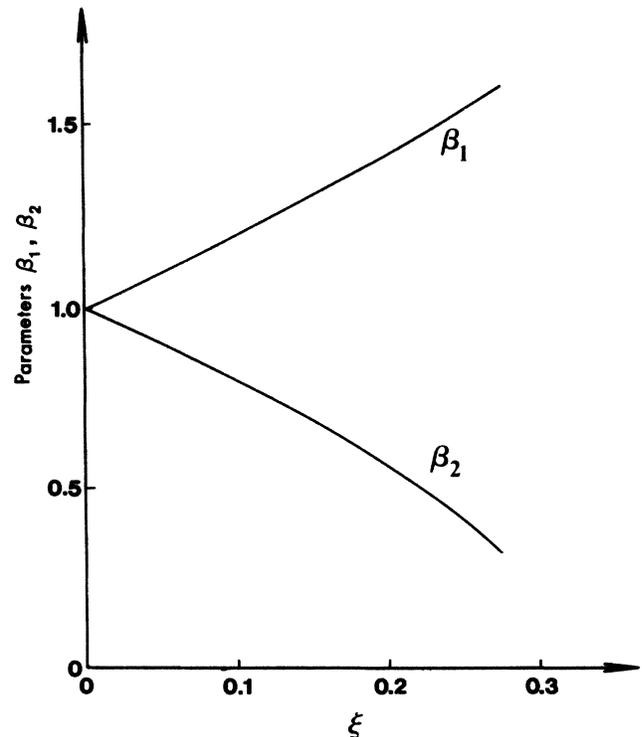


FIG. 1. The variational parameters β_1 and β_2 as functions of electric field ξ .

Eq. (11) represents the energy interaction between the D^+-A^- dipole moment (after neutral DAP recombination) and the electric field. The major difference for the recombination rates of a neutral DAP between a zero field and a nonzero field comes from this term. For $F=0$, A^{0s} at a spherical surface recombine with a D^0 at the origin to emit photons of the same energy. The presence of the dipole moment term causes this equal energy surface to have an angular dependence; i.e., for $F=0$, the surface is spherical, and for $F \neq 0$, it becomes a surface on which the distance to the center is R^* depending on the angle θ .

From Eq. (11), comparing the DAP recombination with and without an electric field of the same emitted photon energy, we obtain (in units of a_D and E_D)

$$1/R^* + \xi' R^* \cos\theta = \Delta E + 1/R, \quad (12)$$

where $\Delta E = \Delta E_D + \Delta E_A$, $\Delta E_D = \frac{1}{2}(E_D^* - E_D)$, $\Delta E_A = \frac{1}{2}(E_A^* - E_A)$, and $\xi' = (\xi/2\epsilon^{1/2})$. E_A^* can also be obtained by using the same $H(\beta_1, \beta_2)$ of Eq. (8), but with

$$I = \int \psi_e \psi_h d\mathbf{r} = \begin{cases} N \exp\{ -[(\rho - R^* \sin\theta)^2 + \beta_1(z - R^* \cos\theta)^2]^{1/2} \}, & 0 \leq \theta < \pi/2 \\ N \exp\{ -[(\rho - R^* \sin\theta)^2 + \beta_2(z - R^* \cos\theta)^2]^{1/2} \}, & (\pi/2 \leq \theta \leq \pi). \end{cases} \quad (14)$$

Noting that ρ and $z \ll R^*$, we have

$$I = \begin{cases} N \exp\{ -R^*[1 + (\beta_1 - 1) \cos^2\theta]^{1/2} \}, & 0 \leq \theta < \pi/2 \\ N \exp\{ -R^*[1 + (\beta_2 - 1) \cos^2\theta]^{1/2} \}, & \pi/2 \leq \theta \leq \pi \end{cases} \quad (15)$$

Averaging for θ , we obtain the following expression for the neutral DAP recombination decay rate under a uniform electric field

$$\langle W_F \rangle = (N^2/4\pi) \int_0^{2\pi} \int_0^\pi |I|^2 \sin\theta d\theta d\phi. \quad (16)$$

III. RESULTS AND DISCUSSION

Let us use the above results to calculate the neutral DAP recombination decay rate for CdS semiconductors at liquid helium temperatures. We have for CdS (Refs. 2, 4, and 14) $E_D = 31$ meV, $E_A = 169$ meV, $E_g = 2.582$ eV, and $\epsilon = 8.33$. The LES zero-phonon peak position is at $h\nu = 518$ nm or $E = 2.3935$ eV. From these parameters, we have $e^2/\epsilon R = 0.0115$ eV so that $R = 5.4a_D$, and $\xi' = 0.173\xi$.

For a weak field, the condition of $R\xi \ll 1$ holds, and $R\Delta E = 0.45R\xi^2$ can be neglected. From Eq. (13), we obtain

$$R^* = R(1 - \xi' R^2 \cos\theta).$$

From Eq. (16), we have

$$\langle W_F \rangle = W_0 Q(\xi), \quad (17)$$

where W_0 is the decay rate of neutral DAP recombination with emitted photon energy $h\nu = 518$ nm (corre-

sponding to A^{0s} at a distance $R = 5.4a_D$ away from D^0) in the absence of an electric field.

$$R^* = 2R(1 + R\Delta E)^{-1} \{ 1 + [1 + 4\xi' R^2 (\cos\theta) \times (1 + R\Delta E)^{-2}]^{1/2} \}^{-1}. \quad (13)$$

Now we can calculate the neutral DAP recombination under a uniform electric field. The probability of recombination between a hole bound to an acceptor at the position $(\rho, z) = (R^* \sin\theta, R^* \cos\theta)$, and an electron bound to a donor at the origin is proportional to the square of the overlap of the wave functions.⁶ If we define W_F as the recombination rate under a uniform electric field, then $W_F = |I|^2$, and

sponding to A^{0s} at a distance $R = 5.4a_D$ away from D^0) in the absence of an electric field.

$$Q(\xi) = \int_0^1 \cosh(2\xi' R^3 x) dx = (1/2\xi' R^3) \sinh(2\xi' R^3) \quad (18)$$

is the ratio of the decay rate with an electric field to the decay rate without the electric field. For CdS in a weak field, we have

$$Q(\xi) = (\sinh\eta)/\eta, \quad (19)$$

where $\eta = 54.5\xi$. In Fig. 2, we plot $Q(\xi)$ as a function of ξ in the region of 0 to 0.05. The corresponding strength of the electric field for CdS is also displayed on the top scale of Fig. 2. For the case of $F=0$ and $\xi'=0$, we have $Q=1$ and $\langle W \rangle = W_0$, as they should be.

The evidence to this increase in decay rate of DAP recombination has been observed in time-resolved excitation spectroscopy of CdS at liquid-helium temperatures.^{11,15} The decay rate is about 50% larger for short excitation wavelength photons (478 nm) compared with those of long-wavelength photons (490 nm). One of the differences between these two excitations is that the absorption coefficient of 490 nm is about 3 orders smaller than that of 478 nm for CdS at liquid-helium temperatures.¹⁶ When the sample is illuminated by photons with

a wavelength of 478 nm, most photons will be absorbed at the surface region. On the other hand, photons of excitation wavelength 490 nm generate most D^0 and A^0 in the interior. Because of the surface field, we expect an increase in the decay rate for short excitation wavelengths according to the above calculations. This is exactly what we observed,¹⁵ and the average surface field is about 10^4 V/cm for the ultra-high-purity CdS with etched surfaces.

By inspection of Eq. (13), we have a restriction on R^* , which can be expressed as

$$1 + 4\xi'R^2(\cos\theta)(1 + R\Delta E)^{-2} \geq 0$$

or

$$\cos\theta \geq -(1 + R\Delta E)^2 / 4\xi'R^2 = -(1 + 2.5\xi^2) / 20\xi ; \tag{20}$$

if $(1 + 2.5\xi^2) / 20\xi < 1$, then there is a restriction on θ . Only within the angular region of 0 to θ_0 does the recombination of the neutral DAP produce photons whose energy corresponds to the energy $h\nu$ of a D^0 - A^0 pair (distance R apart) in the absence of the electric field. Beyond this region, the DAP recombination of the neutral DAP will not contribute to the luminescence signal detected at an energy $h\nu$. For $\xi < 0.05$, there is no restriction on θ . For $\xi \geq 0.05$, the critical angle θ_0 can be calculated from Eq. (20), and we have

$$\theta_0 = \cos^{-1} [-(1 + 2.5\xi^2) / 20\xi] . \tag{21}$$

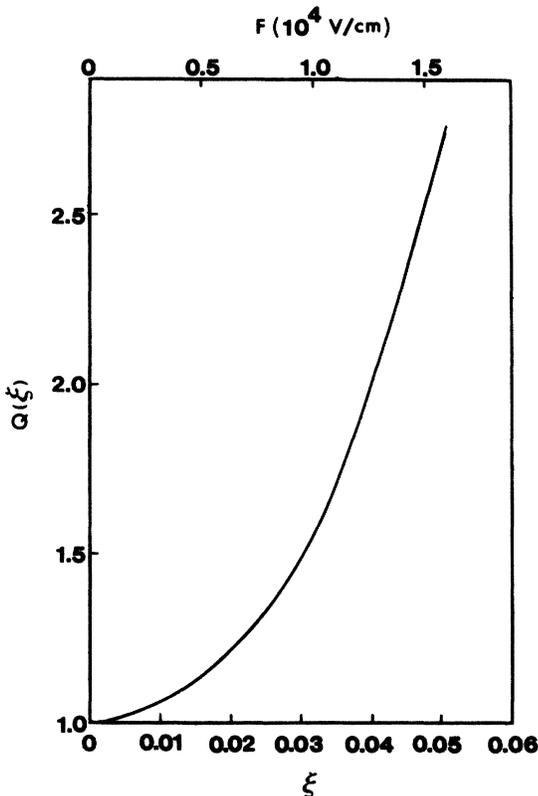


FIG. 2. Increase in the decay rate of neutral DAP recombination as a function of electric field ξ . Upper scale is the electric field strength for CdS.

We have plotted the critical angle θ_0 for $h\nu=518$ nm as a function of ξ in Fig. 3. If ξ increases from 0.05 to 0.1, θ_0 drops dramatically from 180° to 120° , then decreases slowly to 106° at $\xi=0.2$.

Besides the effects of the increase on the decay rate of DAP recombination, the electric field also changes the line shape. It will cause a linewidth broadening. The main effect is caused by the angular dependence of R^* [see Eq. (13)], with $R^* \leq R$ for $0 \leq \theta \leq \pi/2$ and $R^* > R$ for $\pi/2 < \theta \leq \pi$. The line shape for $F=0$ depends on the probability of a neutral donor recombined with neutral acceptors at a spherical surface with a distance R from the donor. For $F \neq 0$, this probability also depends on the angle θ . The increase of the probability in the region $0 \leq \theta \leq \pi/2$ is larger than the decrease of the probability in the region $\pi/2 < \theta \leq \pi$. After averaging over θ from 0 to π , the total effects to the line shape by the electric field are that the DAP recombination peak shifts to higher energy and broadens preferentially on the higher energy tail. These effects have been observed experimentally.^{6,7} As has been discussed by Colbow,⁶ under an electric field F , one may expect the linewidth from DAP emission to increase by an amount of the order of

$$\langle |\delta E| \rangle = \frac{1}{2} eFR , \tag{22}$$

provided the radiation is mainly caused by the pairs whose reaction rate is increased by the field. Another effect of linewidth broadening is caused by the broadening of the shallow donor levels under an electric field.^{18,19}

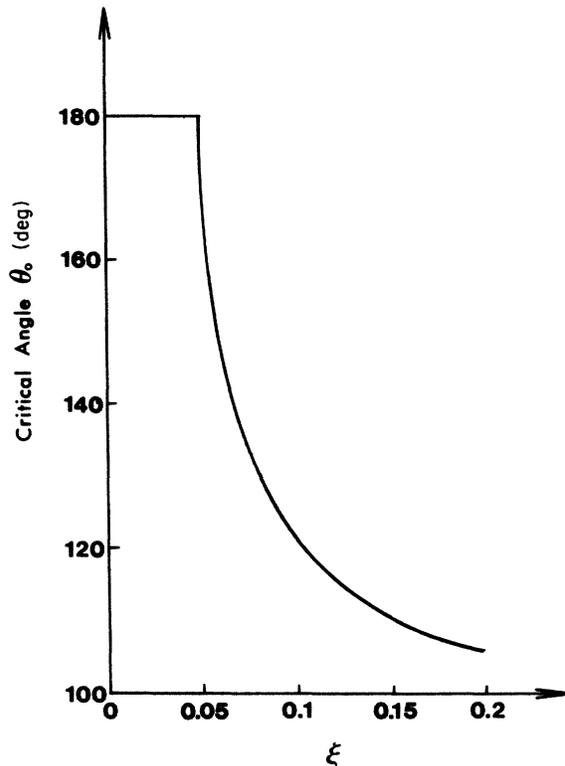


FIG. 3. The critical angle θ_0 for $h\nu=518$ nm as a function of electric field ξ .

In conclusion, we present a calculation of neutral DAP recombination in the presence of a uniform electric fields, and we showed that the decay rate of DAP recombination will increase. This calculation also gives us some hints about the neutral DAP recombination at the semiconductor surfaces and interface regions. If we know the exact distribution of the surface fields, the kinetics of the neutral DAP recombination can be treated in the same

way. Work is currently in process on the neutral DAP recombinations at the semiconductor surfaces.

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