Intercenter carrier transitions in partly disordered silicon: calculations

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A nonadiabatic approximation is used in a calculation of the probabilities of transitions involving one- and two-electron states. An analysis is made of the field dependences of the frequency factor and of the activation energy of intercenter transitions. Necessary calculations are made of the parameters of a model of generation–recombination transitions in disordered silicon.

Difficulties are frequently encountered in unambiguous interpretation of the characteristics of electron transitions occurring in generation–recombination processes in semiconductors or in semiconductor structures with a complex spectrum of localized states in the band gap. This applies also to real surfaces of semiconductors, amorphous and polycrystalline semiconductors, semiconductors wity a high dislocation density, interfaces between a metal and a semiconductor and at a junction, etc. An analysis of the recent experimental data on generation–recombination processes shows that the main recombination channel in these materials usually includes intercenter electron transitions.

Information on the spin states of carriers has extended greatly the scope of investigations of characteristics of electron transitions in such semiconductors. There are several models[1-3] that account for the dependence of the rate of recombination of nonequilibrium carriers on the spin states of free and localized electrons. The Kaplan–Solomon–Mott model[3] allows for the recombination of carriers via pair states and has been confirmed by direct experiments.[3-5] Figure 1 shows schematically the generation–recombination transitions in this model. It was reported in Ref. 6 that each of the centers in a pair in most of the investigated objects can be in one or two-electron states.

There is much which is not yet clear about the origin and physics of two-electron states and little is known about the pair states themselves, such as the radius of the electron wave functions of the states, the distance between the partners in a pair, and the methods suitable for the determination of the parameters of such states.

We shall try to tackle most of the problems mentioned above in the present and following papers. The present paper reports calculations of transitions involving one-and two-electron states, gives an analysis of the field dependences of the frequency factor and the activation energy of intercenter transitions, and derives relationships necessary for the determination of the parameters of a model of generation–recombination transitions in disordered silicon. In the following paper we shall give the experimental data for partly disordered silicon and discuss them using the results given below: we shall estimate the radii of electron wave functions (4-7 Å) and the separation between the pair partners (25-40 Å), and we shall also account for the field dependences of the frequency factor and the activation energy.

1. ELECTRON STATES LOCALIZED AT STRUCTURE DEFECTS AND THE POLARON EFFECT

We shall consider a structure defect in an ideal periodic lattice, for example, a vacancy in silicon. Among the various types of local vibrations of atoms associated with this defect we shall select the "softest" mode and we shall label its con-
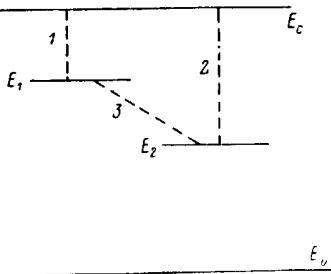
FIG. 1. Schematic representation of the investigated electron transitions.

figurational coordinate X. Following Holstein [7],[8] and Anderson,[9] we shall assume that the distrotion of the atomic system of a defect (which results, for example, because of the capture of an electron) is due to this softest mode. We shall try to obtain the simplest description of such a defect which has captured $n = 0$, 1, or 2 electrons and, following tradition,[7-11] we shall confine our treatment to the harmonic approximation for the energy of the atomic subsystem $kX^2/2$ and to the linear approximation for the energy of its interaction with captured electrons $-nQX$, where Q is the deformation potential. The total energy of the system

$$W(X, n) = kX^2/2 - nQX + n\varepsilon + U_c\delta_{n2} \qquad (1)$$

includes also the one-electron energy $n\varepsilon$ and the energy of the Coulomb repulsion U_C, which is "activated" only if $n = 2$.

Minimization of Eq. (1) with respect to X allows us to find the equilibrium energies of the system $E(n)$ and the equilibrium displacements X^0 (n)

$$E(n) = n\varepsilon - n^2W + U_c\delta_{n2}, \quad X^0(n) = nX^0, \quad X^0 = Q/k, \qquad (2)$$

as well as the polaron shift W

$$W = Q^2/2k. \qquad (3)$$

If the polaron shift is sufficiently large, so that the Hubbard energy

$$U = E(2) - 2E(1) = -2W + U_c \qquad (4)$$

becomes negative, then two electrons are more likely to occupy one center than two different centers.

The strong polaron effect in disordered semiconductors is due to anomalously low rigidity k of stretched bonds. For our purpose it is important that stretched bonds appear at dislocations and, consequently, U centers with a large polaron effect may concentrate around dislocations. Therefore, even in the case of relatively weakly deformed semiconductors in which the total number of such centers is small, the probability of their close distribution in the coordinate space may be high. This in turn may induce transitions between the U centers and consequently give rise to spin-dependent effects in the generation and recombination of carriers.

2. NONADIABATIC ELECTRON TRANSITIONS BETWEEN U CENTERS

In the range of parameters of interest to us the activation energy (or the barrier height) for a

transition of an electron between two U centers is much greater than the energy T. Therefore, these transitions are of multiphonon nature. The excitation of the atomic subsystem can then be considered classically using the configurational coordinates of the centers X_1 and X_2 of interest to us. We can say that mechanical vibrations of frequencies not exceeding the Debye value are induced by the influence of temperature and the electron states in the defects adjust themselves adiabatically to the current states of the configurational coordinates $X_1(t)$ and $X_2(t)$. For certain displacements corresponding to crossing of the electron terms we can expect a transition of an electron between centers and the frequency of such transitions is $\gamma(X_1, X_2)$. The value of γ can be found simply using quantum-mechanical perturbation theory [12]

$$\hbar\gamma(X_1, X_2) = 2\pi |\mathcal{J}|^2 \delta [W_-(X_1, X_2) - W_+(X_1, X_2)]. \qquad (5)$$

Here, $W_+(X_1, X_2)$ and $W_-(X_1, X_2)$ are the energies of two centers before and after the transfer of an electron from the first to the second center at fixed values of the configurational coordinates X_1 and X_2; the energy interval of the overlap $\mathcal{J}$ is the matrix element in the Hamiltonian of the transition

$$\mathcal{H} = \mathcal{J}(\hat{a}_1^+\hat{a}_2 + \hat{a}_2^+\hat{a}_1). \qquad (6)$$

If the probability of a transition during the time of one passage through a $W_+ = W_-$ resonance is low (nonadiabatic conditions), the interference between different passages through the resonance can be ignored and the total transition frequency is found by averaging $\gamma(X_1, X_2)$ over different configurations of the system of two centers X_1 and X_2 with a Boltzmann probability function [13]

$$f(X_1, X_2) = Z^{-1}\exp[(W_-(X_1^0, X_2^0) - W_-(X_1, X_2))/T]. \qquad (7)$$

Here, X_1^0 and X_2^0 are the equilibrium displacements of the centers 1 and 2, respectively; Z is the partition function governed by the probability normalization condition. We therefore have

$$\gamma = \int f(X_1, X_2)\gamma(X_1, X_2) dX_1 dX_2. \qquad (8)$$

3. DEPENDENCE OF THE OVERLAP INTEGRAL ON THE CONFIGURATIONAL COORDINATES

In calculation of the transition frequency γ it is usual to ignore the dependence of $\mathcal{J}$ on X_1 and X_2 (see, for example, Ref. 14). We shall show that this approximation may be incorrect.

We shall consider a defect with a characteristic size a of the order of several lattice constants a_0. At a distance $r > a$ from the center of a defect the field of the defect vanishes so that the electron function Ψ falls on increase in r in a universal manner independent of the nature of the defect. In the effective mass approximation when an electron "sees" a crystal as a continuous medium, the function Ψ in the range $r > a$ satisfies the free Schrodinger equation. In the spherically symmetric case, we have [12]

$$\Psi \propto r^{-1}\exp(-\varkappa r). \qquad (9)$$

The localization radius $r_0 = 1/\varkappa$ of an electron depends only on the electron energy ε measured from the bottom of the conduction band:

$$\hbar^2\varkappa^2/2m = -\varepsilon, \quad \varepsilon < 0. \qquad (10)$$

Calculations of the matrix element of the kinetic energy operator of an electron using the functions $\Psi(r)$ and $\Psi(R-r)$ of Eq. (9) gives the familiar asymptotic (for large values of R) dependence of the energy of an overlap integral on the distance between the centers R:

$$\mathcal{J}(R) = \mathcal{J}_0 \exp(-\varkappa R). \qquad (11)$$

Here, $\mathcal{J}_0 \simeq \varepsilon/2$; in the case of deep centers in silicon we can assume that $\mathcal{J}_0 \simeq$ 0.1–0.2 eV.

We shall need this estimate later in an analysis of the experimental data. It is worth noting that $\varkappa$ in Eq. (11) depends on ε in accordance with Eq. (10), and that ε represents the energy of a transition between the centers 1 and 2 in a state which is adiabatically adjusted to fit the current values of the configurational coordinates X_1 and X_2. In the case of the $|0, 2> \rightarrow |1, 1>$ transition (with two electrons at the first center and none at the other in the initial state and with one electron per center in the final state), we have

$$\varepsilon = \varepsilon_2 - Q_2 X_2 + U_{c2} = \varepsilon_1 - Q_1 X_1. \qquad (12)$$

We can see that the energy overlap integral depends exponentially (strongly) on X_1 and X_2. Therefore, when integration with respect to X_1 and X_2 in Eq. (8) is carried out, this dependence can generally be ignored.

4. FREQUENCY OF $|0, 2> \rightarrow |1, 1>$ TRANSITIONS IN NONADIABATIC CONDITIONS

We shall use the above framework to find the frequency of the $\gamma(0, 2 \rightarrow 1, 1)$ transitions when in the initial state there are two electrons at the second center and the first is empty and in the final state each center has one electron. The nature of these centers may be different so that we shall distinguish the deformation potential Q and other phenomenological parameters of the centers by labeling them with the indices 1 and 2. It follows from Eqs. (1) and (2) that

$$W_-(X_1, X_2) = (k_1 X_1^2 + k_2 X_2^2)/2 - 2Q_2 X_2 + 2\varepsilon_2 + U_{c2}, \qquad (13a)$$
$$W_-^0 \equiv W_-(X_1^0, X_2^0) = 2\varepsilon_2 - 4W_2 + U_{c2}, \qquad (13b)$$
$$W_+(X_1, X_2) = (k_1 X_1^2 + k_2 X_2^2)/2 - Q_1 X_1 - Q_2 X_2 + \varepsilon_1 + \varepsilon_2. \qquad (13c)$$

According to Eqs. (7) and (13) the probability function is Gaussian

$$f = Z^{-1} \exp[-(k_2(X_2 - 2X_2^0)^2 + k_1 X_1^2)/2T] \qquad (14)$$

where

$$Z = 2\pi T/(k_1 k_2)^{1/2}, \quad X_2^0 = Q_2/k_2.$$

Substituting Eqs. (11) and (13) into Eq. (5) and also Eqs. (14) and (5) into Eq. (8), we find the initial expression for the frequency of the $|0, 2> \rightarrow |1, 1>$ transitions:

$$\gamma = (\mathcal{J}_0^2 \sqrt{k_1 k_2}/\hbar^2 T)$$
$$\times \int \exp(-\Phi(X_1, X_2)) \delta(Q_1 X_1 - Q_2 X_2 - \varepsilon_1 + \varepsilon_2 + U_{c2}) dX_1 dX_2,$$

where

$$\Phi = 2\varkappa(\varepsilon)R + (W_-(X_1, X_2) - W_-^0)/T.$$

We can find the integral with respect to X_2 with the aid of the δ function, and that with respect

to X_1 by the saddle point method. The saddle point X_1, X_2 is defined by the condition for the minimum of Φ:

$$k_1 X_1 + k_2 X_2 Q_1/Q_2 = Q_1 [2 - \varkappa(\varepsilon) RT/|\varepsilon|], \qquad (16a)$$
$$\varepsilon_1 - Q_1 X_1 = -Q_2 X_2 + \varepsilon_2 + U_{c2} = \varepsilon(X_1, X_2) \equiv \varepsilon. \qquad (16b)$$

We shall assume that, in accordance with Eq. (10), we have $\kappa(\varepsilon) \propto \sqrt{-\varepsilon}$ and that ε is given by Eq. (12). We shall calculate Φ'' at the saddle point:

$$\Phi'' = d^2\Phi/dX_1^2 = k_1 [(W_1 + W_2)/TW_2 - \varkappa R W_2/\varepsilon^2].$$

Our calculations are valid if the separation between the centers is not too high, so that $\Phi'' > 0$. This gives the condition

$$\varkappa R < \varepsilon^2 (W_1 + W_2). \qquad (17)$$

If we introduce a characteristic temperature

$$T^* = T\varepsilon^2 (W_1 + W_2)/[\varepsilon^2(W_1 + W_2) - \varkappa(\varepsilon)RTW_2^2], \qquad (18)$$

then the condition (17) can be rewritten in the form $T^* > 0$. Assuming that this condition is satisfied, we find from Eqs. (15)–(18) that the frequency of the activated $|0, 2> \rightarrow |1, 1>$ transitions is

$$\gamma(0, 2 \rightarrow 1, 1) = (\pi T^*)^{1/2} \mathcal{J}^2(\varepsilon) \exp(-\Delta E_g/T)/\hbar T(W_1 + W_2)^{1/2}. \qquad (19)$$

Here, $\mathcal{J}(\varepsilon)$ is the energy overlap integral of Eqs. (11) and (10) at the saddle value of ε of the tunneling electron energy of Eq. (12):

$$\varepsilon = \frac{1}{W_1 + W_2} [W_1 \varepsilon_2 + W_2(\varepsilon_1 + U_{c2} - 4W_1) + 2W_1 W_2 T\varkappa R/|\varepsilon|], \qquad (20)$$

where ΔE_g is the activation energy of the investigated $|0, 2> \rightarrow |1, 1>$ transitions. This energy is

$$\Delta E_g = \Delta E - \Delta E_r, \qquad (21)$$

where

$$\Delta E = \varepsilon_1 - \varepsilon_2 - U_{c2} + 3W_2 - W_1 \qquad (22)$$

is the difference between the equilibrium (in respect of the configurational coordinates X_1 and X_2) energies of Eq. (2) of two electrons after a transition (in the $|1, 1>$ state) and before a transition (in the $|0, 2>$ state); $\Delta E_r = \Delta E_{r1} + \Delta E_{r2}$ is the energy of a configurational barrier. This energy consists of the already known term ΔE_{r1} of Ref. 15 and an additional term ΔE_{r2}, which appears because of the energy dependence of the overlap integral

$$\Delta E_{r1} = [\varepsilon_1 - \varepsilon_2 - U_{c2} - 2W_1 + 2W_2]^2/4(W_1 + W_2), \qquad (23a)$$

$$\Delta E_{r2} = (\varkappa R)^2 W_2^2 W_1 T^2/\varepsilon^4 (W_1 + W_2)^3. \qquad (23b)$$

This energy occurs in the exponential temperature dependence of the frequency of the reverse (recombination) transition ($|1, 1> \rightarrow |0, 2>$)

$$\gamma(1, 1 \rightarrow 2, 0) = \sqrt{\pi T^*} \mathcal{J}^2(\varepsilon) \exp(-\Delta E_r/T)/\hbar T(W_1 + W_2)^{1/2}. \qquad (24)$$

The dependence $\mathcal{J}(\varepsilon)$ gives rise to several effects. Firstly, in the numerator of the preexponential factor we find that temperature is modified to $T \rightarrow T^* > T$. Secondly, it follows from Eq. (20) that the energy of a tunneling electron in-

creases and so does the overlap integral $\mathcal{J}(\varepsilon)$. Finally, the configurational barrier described by the system (23) becomes higher. If we use γ_0 to denote the frequency of a transition ignoring the dependence $\mathcal{J}(\varepsilon)$, but only allowing for this factor, we obtain

$$\gamma_0 \to \gamma = \gamma_0 \sqrt{\frac{T^*}{T}} \exp\left[(\varkappa R)^2 \frac{W_1 W_2 T}{\varepsilon^2(W_1+W_2)}\left(2 - \frac{W_3^2}{\varepsilon^2(W_1+W_2)}\right)\right]. \quad (25)$$

This relationship is valid in the case of generation and recombination transitions. In its derivation we have assumed that the relative change in the electron energy $\delta\varepsilon$ is small and we have used the expansion $\varkappa(\bar{\varepsilon} + \delta\bar{\varepsilon}) = \varkappa(\bar{\varepsilon})[1 - \delta\bar{\varepsilon}/2\bar{\varepsilon}]$.

These calculations are valid if the separation between the centers is not too large so that $T^* > 0$. In the opposite case the energy ε does not correspond to a minimum, but to a maximum of the argument of the exponential function Φ in Eq. (15). This means that in the case of large distances between the centers the intercenter transitions are characterized by these large fluctuations X_1 and X_2 when $\bar{\varepsilon} \to 0$, i.e., that the energy of a tunneling electron approaches the bottom of the conduction band and the electron localization radius at a center $r_0 \propto 1/\chi \propto 1/\sqrt{|\bar{\varepsilon}|}$ becomes greater than the separation between the centers. In this case the exponential smallness of the overlap integral $\mathcal{J} \exp(-\varkappa R)$ disappears from the expressions for the transition frequency and this is compensated by an additional smallness in the temperature exponent.

5. NONADIABATIC INTERCENTER TRANSITIONS IN AN ELECTRIC FIELD

a) Mechanisms responsible for the dependence of the transition frequency on the applied electric field

An electric field $\mathcal{E}$ homogeneous in space and constant in time should affect the frequency of intercenter transitions for a number of reasons.

1. First of all, the energy of an electron ε_n localized at a center with a coordinate r_n acquires a correction $\delta\varepsilon_n$. It follows from Eqs. (20)-(22) that this alters the energy of the configurational barrier ΔE_r and the activation energy ΔE_g:

$$\Delta E_r \to \Delta E_r + \delta E_r, \quad (26a)$$

$$\Delta E_g \to \Delta E_g + \delta E_g, \quad (26b)$$

$$\delta E_r = \delta\varepsilon \left[\delta\varepsilon + 4W_2 - 4W_1 - 2U_{c2}\right]/4(W_1+W_2), \quad (26c)$$

$$\delta E_g = \delta E_r + \delta\varepsilon, \quad (26d)$$

$$\delta\varepsilon = e\mathcal{E}\mathbf{R}, \quad (26e)$$

where

$$\mathbf{R} = \mathbf{r}_1 - \mathbf{r}_2.$$

In order to estimate the value of $\delta\varepsilon$, which can be done quite easily in experiments, we shall assume that $\mathcal{E}$ has the usual value in a reverse-biased p-n junction in Si: $|\mathcal{E}| \simeq 10^5$ V/cm; we shall also assume that the distance between the centers is $|R| \simeq 100$ Å. Then, the change in the activation barrier height alters the transition frequency at room temperature by a factor $\exp(\delta\varepsilon/T)$, i.e., by three-four orders of magnitude.

2. Moreover, the applied electric field alters the state of an electron at a center and, consequently, its energy ε_n measured from the bottom of the conduction band. If the field is $\mathcal{E}$, the expression for the energy given by Eq. (1) must be supplemented by $-\beta ne|\mathcal{E}|X$, where the numerical coefficient β is of the order of unity. The actual value of β depends on the configurational coordinate X. Therefore, in the expression (1) for the electric field we now have $Q \to Q + \beta e \mathcal{E}$. In the case of a vacancy in Si (Ref. 16) we find that $Q \simeq 1$ eV/Å. In a field $|\mathcal{E}| \simeq 10^5$ V/cm, we obtain $e|\mathcal{E}| = 1.3\cdot10^{-3}$ eV/Å, i.e., $e|\mathcal{E}|/Q \simeq 10^{-2}$-$10^{-3}$. It follows from Eq. (3) that the change in the polaron shift in the field is $\delta W = 2W\beta e|\mathcal{E}|/Q$. The value of W is of the order of 0.2-0.3 eV, so that we have $\delta W \simeq 10^{-3}$ eV. The difference between the values of W between the two centers gives rise to an additiona [compared with Eq. (26b)] change in the activation energy, which should be compared with the energy T. The estimates made above show that, with the exception of the temperature range $T < 10$ K, we can ignore the influence of the electric field on W.

3. Generally speaking, a field $\mathcal{E}$ alters the position of the center of gravity of an electron cloud at a center and, consequently, the effective distance between the centers. However, this effect is negligible: $\Delta R \simeq e|\mathcal{E}|/k$ for $|\mathcal{E}| \simeq 10^6$ V/cm and $k \simeq 10$ eV/Å. Even when $1/\varkappa \simeq a_0 \simeq 5$ Å the relative change in the overlap integral is less than 10^{-2}.

4. Finally, we must bear in mind that the electric field has a direct influence on the overlap integral when the distance between the centers is fixed. This important effect will be discussed in the next subsection.

b) Dependence of the overlap integral on the applied electric field

The application of an electric field influences the wave function $\Psi(\mathbf{r})$ of an electron tunneling between the investigated centers. In the space between the centers the function $\Psi(\mathbf{r})$ satisfies the following Schrodinger equation (z is the electric field direction):

$$-\hbar^2\Delta\Gamma/2m = (E - e|\mathcal{E}|z)\Psi. \quad (27)$$

We shall seek the solution $\Psi(\mathbf{r})$ in the semi-classical approximation

$$\Psi(\mathbf{r}) = \exp(-S(\mathbf{r})), \quad S = \hbar S_0 + \hbar^2 S_1. \quad (28)$$

Hence, in the axially-symmetric case we find that S_0 is described by

$$(\partial S_0/\partial z)^2 + (\partial S_0/\partial\rho)^2 = (1 - 2e|\mathcal{E}|mz)\varkappa^2 \equiv \varkappa^2(z). \quad (29)$$

Here, ρ is the transverse coordinate and $1/\varkappa$ is the localization radius of an electron in the absence of the field (10). Equation (29) is the Hamiltonian—Jacobi equation for the time-independent part of the Euclidean action S_0 of a classical particle moving (in imaginary time) under the influence of a constant force. The path of such motion can be found in a trivial manner by integrating the Lagrange function with respect to time and along a classical path, which gives the action, and then express S_0 in terms of the coordinates of the initial $z = 0$, $\rho = 0$,

and final z, ρ points of motion. This gives

$$S_0(\rho, z) = (3e|\mathscr{E}|m)^{-1}[(\varkappa^2(z/2) + e|\mathscr{E}|mr)^{3/2} - (\varkappa^2(z/2) - e|\mathscr{E}|mr)^{3/2}],$$

(30)

where

$$\varkappa^2(z/2) = \varkappa^2 - e|\mathscr{E}|mz, \quad r^2 = z^2 + \rho^2.$$

The validity of this solution can readily be confirmed by direct substitution in Eq. (29). We shall analyze the resultatnt expression (30) for S_0 expanding it as a series in r:

$$S_0 = \varkappa(z/2)r[1 - r^2/24z_0^2 - \ldots], \quad z_0 = \varkappa^2(z/2)/e|\mathscr{E}|m.$$

(31)

Here, z_0 is the distance along the z axis from the point with the coordinate $z/2$ to the turning point, where $\varkappa^2(z) = 0$. It should be noted that the expansion of Eq. (31) converges well: even at the limit of validity at $r = z_0$ the two terms in Eq. (31) ensure that the precision is $1.5 \cdot 10^{-2}$; however, if $r \le 2z_0$, then to within 10^{-2} we need to consider only the first term of the expansion, i.e., we can assume that $S_0 = \varkappa^2(z/2)r$. In this approximation the influence of the applied electric field on the argument of the exponential function reduces to renormalization of the localization radius $\varkappa \to \varkappa(z/2) = (\varkappa^2 - e|\mathscr{E}|mz)^{1/2}$. This result can be interpreted as follows: we can calculate the semiclassical shift of the phase of S_0 at a point at a certain coordinate z_1, $r_1 = \sqrt{\rho_1^2 + z_1^2}$ by replacing in Eq. (29) $\varkappa^2(z)$ with the average value of this quantity along a classical path, i.e., by replacing it with $\varkappa^2(Z_1/2)$. The resultant equation can be solved in a trivial manner. In the spherical case we have $S_0 = \varkappa^2(z_1/2) r$, which agrees with the first term of the expansion in Eq. (31).

In the next order of the classical expansion (28) we have to calculate S_1, which determines the preexponential factor in the expression for the wave function. Since in $\mathscr{E} = 0$ this factor is independent of $\varkappa$, the influence of the field on the preexponential factor cannot be great; even if we do not approach too closely the turning point, this influence can be ignored compared with the field dependence of the argument of the exponential function for $\varkappa$. In this way we obtain the following approximation for $\Psi(\mathbf{r})$, which replaces Eq. (9):

$$\Psi(r) \simeq \exp[-S_0(\rho, z)]/r.$$

(32)

The saddle-point method is used to calculate the overlap integral containing the functions (32). The classical path is called the saddle path when it joins the points with the coordinates $\mathbf{r}_1$ and $\mathbf{r}_2$ at which the centers are located. The argument of the exponential function in the integrand $\propto \Psi(r-r_1)\Psi^*(r-r_2)$ contains a sum of the Euclidean actions and this sum is independent of the position of the point $\mathbf{r}$ on the classical path and is given by $S_0(z_-, \rho_-)$ of Eq. (30) if z_- and ρ_- are the components of the vector $R = \mathbf{r}_2 - \mathbf{r}_1$: $z = z_2 - z_1$, $\overline{\rho} = \rho_1 - \rho_2$. This gives the argument of the exponential function in the overlap integral: $\mathscr{J} \propto \exp[-S_0(z, \rho)]$. If R is less than the distance from the point $(\mathbf{r}_1+\mathbf{r}_2)/2$ to the turning point, then — as demonstrated above — we can limit the expansion (31) to the first term: $S_0(z-\rho_-) = \overline{\varkappa}R$, $\overline{\varkappa} \equiv \varkappa(z_-/2)$. If we recall that Eqs. (30) and (31) are obtained on the assumption that $r_1 = 0$, it becomes clear that $z_-/2 = (z_1 + z_2)/2$. In calculation of the preexponential factor in $\mathscr{J}$ we ignore the dependence $\varkappa(z)$ and take $\varkappa(z)$ at this

point: $\varkappa = \overline{\varkappa}$. Consequently, $\mathscr{J}$ is described by an expression similar to Eq. (11):

$$\mathscr{J}(R) \simeq \mathscr{J}_0(\varkappa) \exp(-\varkappa R),$$

(33)

where $\mathscr{J}_0(\overline{\varkappa}) \simeq \hbar^2 \varkappa^2/4m$. Therefore, in our approximation the influence of an electric field on the overlap integral reduces to the dependence $\varkappa(\varepsilon)$; χ should be calculated from Eq. (10) in terms of the tunneling electron energy $\overline{\varepsilon}$ of Eq. (20) measured from the bottom of the conduction band at the point with the coordinate $\mathbf{r} = (\mathbf{r}_1 + \mathbf{r}_2)/2$. Using Eqs. (10), (20), and (2), we obtain an expression which describes the change in the energy ε in an electric field $\mathscr{E}$:

$$\overline{\varepsilon} \to \overline{\varepsilon} + T^*\delta\varepsilon/2T, \quad \delta\varepsilon = e\mathscr{E}R.$$

(34)

This change in $\overline{\varepsilon}$ results, in accordance with Eq. (23), in an additional [compared with Eq. (26)] change in the height of the configurational barrier:

$$\Delta E_r \to \Delta E_r + \delta E_r + \delta_1 E_r,$$

(35)

where

$$\delta_1 E_r = \frac{3W_1W_2T^*T}{2(W_1+W_2)}\left[\frac{\varkappa R W_2^{\frac{1}{2}}}{\bar{\varepsilon}^2(W_1+W_2)}\right]^2 \delta\varepsilon.$$

c) Influence of the applied electric field on the frequency of intercenter transitions

We shall estimate the general change in the frequency of the carrier-generation transition $|0, 2\rangle \to |1, 1\rangle$ due to the total change in the argument of the exponential function in an electric field. The change is due to the field dependence of the activation energy and of the configurational barrier [Eqs. (26) and (34)] and of the overlap integral [Eqs. (33) and (34)]:

$$\gamma(\mathscr{E})/\gamma(0) = \exp[\delta\varepsilon(A + B/T + C) + (\delta\varepsilon)^2(D + F/T)].$$

(36)

The term A is due to the change in the overlap integral in a field and is obtained by expanding $\varkappa(\varepsilon)$ as a series using Eqs. (34) and (18):

$$A = \bar{\varepsilon}(W_1 + W_2)\varkappa R/[2\bar{\varepsilon}^2(W_1 + W_2) - TW_2^{\frac{1}{2}}\varkappa R].$$

(37)

The term B is due to the field dependence of the activation energy ΔE_g of Eq. (26):

$$B = (-2W_2 + U_{e2})/(W_1 + W_2) = U_2/(W_1 + W_2).$$

(38)

Here, U_2 is the Hubbard energy. The term C is related to the additional change in the barrier ΔE_r represented by Eq. (35):

$$C = \frac{-3W_2^{\frac{1}{2}}TW(\varkappa R)^2}{2[\bar{\varepsilon}^2(W_1+W_2) - T(\varkappa R)W_2^{\frac{1}{2}}]\bar{\varepsilon}^2(W_1+W_2)^2}.$$

(39)

The term D is due to the change in the overlap integral and F caused by the change in the configurational barrier [Eq. (26)]:

$$D = \varkappa R\bar{\varepsilon}(W_1+W_2)/16[\bar{\varepsilon}^2(W_1+W_2) - TW_2^{\frac{1}{2}}\varkappa R],$$

(40)

$$F = -1/4(W_1 + W_2).$$

(41)

If we limit the treatment to the effects which are linear functions of the distance between the

centers, then

$$A = \hbar R/2\varepsilon, \quad B = U_2/(W_1 + W_2),$$
$$C = 0, \quad D = \hbar R/16\varepsilon^2, \quad F = -1/T\,(W_1 + W_2). \tag{42}$$

The main qualitative conclusion that follows from Eqs. (36) and (42) is that the application of an electric field has different effects on the temperature-dependent part of the exponential function (terms B and F) and on the temperature-independent part which is a linear function of R (terms A and D $\propto \kappa R$). If $\delta\varepsilon < 0$, the energy barrier height decreases and the probability of thermal fluctuations necessary for the transition increases. However, the tunneling probability falls. If the distance between the centers is not too large, the former effect predominates and the transition frequency increases on application of the field. It is easy to separate experimentally these two effects by determination of the temperature dependences of the transition frequency in different electric fields.

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