Localization and energy transfer of quasi-two-dimensional excitons in GaAs-AlAs quantum-well heterostructures

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A theory of energy transfer of the quasi-two-dimensional excitons in GaAs-AlAs quantum-well heterostructures is developed, and the recently observed slow and nonexponential energy relaxation of excitons is explained quantitatively in terms of the one-phonon-assisted transfer of localized excitons among islandlike structures within a quantum well. The nonexponential behavior of energy relaxation is clarified as a general feature to be observed in the low-energy tail of the density of states. The dependence of the energy relaxation rate on the quantum-well thickness is discussed along with the same dependence of the absorption bandwidth. The correlation between the energy relaxation rate and the absorption bandwidth is explained qualitatively on the basis of the scaling property of the rate equation for the exciton distribution function.

I. INTRODUCTION

Recently, semiconductor quantum-well (QW) heterostructures have been extensively investigated because of interest in their fundamental physical properties as well as in their potential device applications. Optical techniques such as time-resolved photoluminescence[1-3] and resonant Raman[4,5] and Rayleigh scattering,[6] are quite promising to elucidate the salient features of the quasi-two-dimensional excitons in QW structures. Recently, Masumoto *et al.*[3] studied the time-resolved photoluminescence of $1s$ excitons ($n=1$, e-hh) in GaAs-AlAs multiple-quantum-well structures and found the anomalously slow relaxation of the average energy of luminescence, which shows a nonexponential decay for about several hundred picoseconds after photoexcitation. In addition, the decreasing rate of the average energy of luminescence is too small to be accounted for in terms of the kinetic-energy relaxation on the dispersion curve of the quasi-two-dimensional exciton accompanied by emission of acoustic phonons. In the experiments of Masumoto *et al.* the GaAs layers are selectively excited by choosing the laser energy suitably and ensuring that the AlAs barrier layers are sufficiently thick to rule out the possibility of interlayer migration of excitons. On the other hand, it is well known that the topological disorder of the interface produces sizable optical effects. From the linewidth analysis of the luminescence and excitation spectra, and from transmission electron microscopy, an islandlike structure of the QW interface measuring one monolayer high and about 300 Å laterally was proposed.[7-9] On the basis of this model, a theory is developed to explain the experimental results of photoluminescence in terms of the intralayer migration of localized excitons among islandlike structures with different well thicknesses.

After photoexcitation, the generated electron-hole pairs quickly lose their energy and form excitons with emission of a number of phonons. At the next stage, the kinetic-energy relaxation on the dispersion curve of the quasi-two-dimensional exciton takes place with a relaxation rate that is 1 order of magnitude faster than the observed rate. After these processes are completed, the anomalously slow energy relaxation begins showing nonexponential behavior. In this stage the lowest $1s$ exciton in the GaAs layer can be considered to be localized at some islandlike structure since the Bohr radius of the quasi-two-dimensional exciton in the sample of Ref. 3 is estimated to be about 100 Å according to recent theories[10-12] and is less than the average lateral size of the islandlike structures in a QW. Furthermore, the fluctuation of the well thickness of one monolayer produces the fluctuation of the exciton energy of about several meV.[3] This amount of energy fluctuation is sufficient to localize the excitons at the energetically local minimum sites. The localized excitons will then migrate among the local minimum sites in search of the lower-energy sites with emission of acoustic phonons. This intralayer migration of localized excitons is the key idea for explaining the anomalously slow energy relaxation. In fact, our theory explains the salient features of the experimental results quantitatively or qualitatively.

The paper is organized as follows. The two-dimensional aspects of the localized excitons and their interaction with acoustic phonons are essentially new and have not yet been investigated fully. In Sec. II the interaction Hamiltonian of the quasi-two-dimensional exciton with the acoustic phonons is derived microscopically for the first time. In Sec. III the kinetic-energy relaxation rate of the quasi-two-dimensional excitons is estimated on the basis of the result obtained in Sec. II and is shown to be 1 order of magnitude larger than the observed value. Thus the kinetic-energy relaxation is ruled out as the candidate which can explain the slow energy relaxation. In Sec. IV the one-phonon-assisted exciton transfer between localized sites is formulated from the microscopic point of view, making use of the perturbation theory with respect to the exciton-phonon interaction Hamiltonian and the intersite transfer Hamiltonian. In Sec. V the matrix element of the intersite transfer Hamiltonian of the localized excitons is calculated microscopically. In Sec. VI the exciton

citon transfer rate is estimated explicitly for both the deformation-potential coupling and the piezoelectric coupling, and for various cases of the localization form and the type of intersite transfer. In Sec. VII the rate equation for the distribution function of localized excitons is derived, neglecting the interlayer transfer of excitons and thus reducing the problem to that of a single QW. By integrating the rate equation numerically, the average energy of luminescence is calculated and compared with the experimental data. From the comparison between theory and experiment the constant of the exciton transfer integral is determined and found to be in good agreement with the theoretically estimated value. The experimentally observed nonexponential behavior of energy relaxation is clarified theoretically from a general point of view. Finally, in Sec. VIII the dependence of the energy relaxation rate on the QW thickness is discussed on the basis of the scaling property of the rate equation, and the observed correlation between the energy relaxation rate and the absorption bandwidth is explained in a qualitative way.

II. INTERACTION OF QUASI-TWO-DIMENSIONAL EXCITONS WITH ACOUSTIC PHONONS

In this section the interaction Hamiltonian of the quasi-two-dimensional exciton with acoustic phonons is derived for both the deformation-potential and piezoelectric coupling. In the case of GaAs-AlAs QW structures, the electron and hole of the excitons are considered to be well confined within a QW since the band-gap discontinuity is quite large. On the other hand, the lattice properties of GaAs and AlAs, for example, the lattice constant and elastic moduli, are in close proximity.[13] Thus the acoustic phonons which interact with the quasi-two-dimensional exciton in a GaAs layer can be considered to have three-dimensional character. One can derive the interaction Hamiltonian of the quasi-two-dimensional exciton with acoustic phonons starting from the three-dimensional exciton-phonon interaction Hamiltonian.

Let us now consider the quasi-two-dimensional exciton state with a total wave vector $\mathbf{K}_{||}$ and represent it as

$$|\lambda,K_{||}\rangle=\frac{v_0}{L}\sum_{\mathbf{r}_e,\mathbf{r}_h} e^{i\mathbf{K}_{||}\cdot\mathbf{R}_{||}}F_\lambda(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h)a^\dagger_{c\mathbf{r}_e}a_{v\mathbf{r}_h}|0\rangle\ , \tag{2.1}$$

where v_0 and L are, respectively, the volume of the unit cell and the linear dimension of the quantization volume, $a^\dagger_{\alpha\mathbf{r}}$ ($a_{\alpha\mathbf{r}}$) the creation (annihilation) operator of the αth-(conduction- or valence-) band electron in the Wannier representation, $|0\rangle$ the crystal ground state, and $\mathbf{R}$ the coordinate of the exciton center of mass defined by

$$\mathbf{R}=(m_e\mathbf{r}_e+m_h\mathbf{r}_h)/(m_e+m_h)\ , \tag{2.2}$$

with the electron (hole) effective mass m_e (m_h). The envelope function for the λth electron-hole internal motion is denoted by F_λ. In the following any position vector or wave vector will be decomposed into components parallel and perpendicular to the QW interface as $\mathbf{r}=(\mathbf{r}_{||},z)$ or $\mathbf{k}=(\mathbf{k}_{||},k_z)$. Then, rewriting the operators in the Wannier representation with those in the Bloch representation by a well-known relation

$$a_{vn}=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_n}a_{v\mathbf{k}}\ , \tag{2.3}$$

where N is the number of unit cells related to L by $Nv_0=L^3$, and transforming the discrete sum over the lattice sites into a spatial integral by

$$\sum_{\mathbf{r}_e}\rightarrow\frac{1}{v_0}\int d^3r_e\ , \tag{2.4}$$

one obtains

$$|\lambda,\mathbf{K}_{||}\rangle=\sum_{\mathbf{k},\mathbf{k}'} f_\lambda(\mathbf{k},\mathbf{k}';\mathbf{K}_{||})\delta_{\mathbf{k}_{||}-\mathbf{k}'_{||},\mathbf{K}_{||}}a^\dagger_{c\mathbf{k}}a_{v\mathbf{k}'}|0\rangle\ , \tag{2.5}$$

with

$$f_\lambda(\mathbf{k},\mathbf{k}';\mathbf{K}_{||})=\frac{1}{L^2}\int d^2r_{||}\int dz_e\int dz_h\,F_\lambda(\mathbf{r}_{||},z_e,z_h)\exp[i(\alpha_e\mathbf{K}_{||}-\mathbf{k}_{||})\cdot\mathbf{r}_{||}-ik_zz_e+ik'_zz_h]\ , \tag{2.6}$$

where α_e and α_h are defined by

$$\alpha_e=m_e/(m_e+m_h)\ \text{ and }\ \alpha_h=m_h/(m_e+m_h)\ . \tag{2.7}$$

The three-dimensional electron-phonon (e-ph) interaction for the deformation-potential (DF) coupling is written as[14]

$$H^{\mathrm{DF}}_{e\text{-ph}}=\sum_{\mathbf{k},\mathbf{q}}\left[\frac{\hbar|\mathbf{q}|}{2\rho uV}\right]^{1/2}(D_ca^\dagger_{c,\mathbf{k}+\mathbf{q}}a_{c\mathbf{k}}+D_va^\dagger_{v,\mathbf{k}+\mathbf{q}}a_{v\mathbf{k}})(b_{\mathbf{q}}+b^\dagger_{-\mathbf{q}}) \tag{2.8}$$

$$=\sum_{\mathbf{k},\mathbf{q}}[\Xi_c(\mathbf{q})a^\dagger_{c,\mathbf{k}+\mathbf{q}}a_{c\mathbf{k}}+\Xi_v(\mathbf{q})a^\dagger_{v,\mathbf{k}+\mathbf{q}}a_{v\mathbf{k}}](b_{\mathbf{q}}+b^\dagger_{-\mathbf{q}})\ , \tag{2.9}$$

where D_c (D_v), ρ, and u are the deformation potential for the conduction (valence) band, the mass density, and the sound velocity of the longitudinal-acoustic (LA-) phonon mode, respectively, and the coupling functions Ξ_c and Ξ_v are introduced by (2.9) for later use. The interaction Hamiltonian of the quasi-two-dimensional exciton with acoustic phonons for the deformation-potential coupling is obtained by calculating the matrix element of $H^{\mathrm{DF}}_{e\text{-ph}}$ between two exciton states $|\lambda,\mathbf{K}_{||}\rangle$ and $|\lambda,\mathbf{K}'_{||}\rangle$, where for simplicity, the change of the electron-hole internal motion is not taken into account. The result is given as

$$\langle\lambda,\mathbf{K}'_{||}\,|\,H^{\rm DF}_{e\text{-ph}}\,|\,\lambda,\mathbf{K}_{||}\rangle=\sum_{\mathbf{k},\mathbf{k}'}\sum_{l,l'}\Xi_c(l-\mathbf{k})f^*_\lambda(l,l';\mathbf{K}'_{||})f_\lambda(\mathbf{k},\mathbf{k}';\mathbf{K}_{||})\delta_{l_{||}-l'_{||},\mathbf{K}'_{||}}\delta_{\mathbf{k}_{||}-\mathbf{k}'_{||},\mathbf{K}_{||}}\delta_{\mathbf{k}',l'}$$
$$-\sum_{\mathbf{k},\mathbf{k}'}\sum_{l,l'}\Xi_v(\mathbf{k}'-l')f^*_\lambda(l,l';\mathbf{K}'_{||})f_\lambda(\mathbf{k},\mathbf{k}';\mathbf{K}_{||})\delta_{l_{||}-l'_{||},\mathbf{K}'_{||}}\delta_{\mathbf{k}_{||}-\mathbf{k}'_{||},\mathbf{K}_{||}}\delta_{\mathbf{k},l}\ . \tag{2.10}$$

Substituting expression (2.6) and converting the discrete sum over $\mathbf{k}$ into an integral by

$$\sum_{\mathbf{k}}\rightarrow\left[\frac{L}{2\pi}\right]^3\int d^3k\ , \tag{2.11}$$

one can reduce the first term as

$$\sum_{q_z}\Xi_c(\mathbf{K}'_{||}-\mathbf{K}_{||},q_z)\int d^2r'_{||}\int dz'_e\int dz'_h\int d^2r_{||}\int dz_e\int dz_h\,F^*_\lambda(\mathbf{r}'_{||},z'_e,z'_h)F_\lambda(\mathbf{r}_{||},z_e,z_h)$$
$$\times\delta^{(2)}(\mathbf{r}_{||}-\mathbf{r}'_{||})\delta(z_h-z'_h)\delta(z_e-z'_e)\exp[iq_zz_e-i\alpha_h(\mathbf{K}_{||}-K'_{||})\cdot\mathbf{r}_{||}]$$
$$=\sum_{q_z}\Xi_c(\mathbf{K}'_{||}-\mathbf{K}_{||},q_z)\int d^2r_{||}\int dz_e\int dz_h\,|\,F_\lambda(\mathbf{r}_{||},z_e,z_h)\,|^2\exp[iq_zz_e-i\alpha_h(\mathbf{K}_{||}-\mathbf{K}'_{||})\cdot\mathbf{r}_{||}]\ , \tag{2.12}$$

where the arguments of Ξ_c are written explicitly; the first argument is the component of the phonon wave vector parallel to the QW interface and the second is the perpendicular component. Similarly the second term in (2.10) can be reduced as

$$-\sum_{q_z}\Xi_v(\mathbf{K}'_{||}-\mathbf{K}_{||},q_z)\int d^2r_{||}\int dz_e\int dz_h\,|\,F_\lambda(\mathbf{r}_{||},z_e,z_h)\,|^2\exp[iq_zz_h+i\alpha_e(\mathbf{K}_{||}-\mathbf{K}'_{||})\cdot\mathbf{r}_{||}]\ . \tag{2.13}$$

Thus the quasi-two-dimensional exciton-phonon interaction Hamiltonian $H^{\rm DF\,(Q2D)}_{\rm ex\text{-}ph}$ for the deformation-potential coupling is given by

$$H^{\rm DF\,(Q2D)}_{\rm ex\text{-}ph}=\sum_{\mathbf{K}_{||},\mathbf{K}'_{||},q_z}[\Xi_c(\mathbf{K}'_{||}-\mathbf{K}_{||},q_z)H_\lambda(-\alpha_h(\mathbf{K}_{||}-\mathbf{K}'_{||}),q_z)$$
$$-\Xi_v(\mathbf{K}'_{||}-\mathbf{K}_{||},q_z)H_\lambda(\alpha_e(\mathbf{K}_{||}-\mathbf{K}'_{||}),q_z)]\,|\,\lambda,\mathbf{K}'_{||}\rangle\langle\lambda,\mathbf{K}_{||}\,|\,(b_{\mathbf{K}'_{||}-\mathbf{K}_{||},q_z}+b^\dagger_{\mathbf{K}_{||}-\mathbf{K}'_{||},-q_z})\ , \tag{2.14}$$

with

$$H_\lambda(\mathbf{Q}_{||},Q_z)=\int d^2r_{||}\int dz_e\int dz_h\,|\,F_\lambda(\mathbf{r}_{||},z_e,z_h)\,|^2\exp(iQ_zz_e+i\mathbf{Q}_{||}\cdot\mathbf{r}_{||})\ , \tag{2.15}$$

where the symmetry $F_\lambda(\mathbf{r}_{||},z_e,z_h)=F_\lambda(\mathbf{r}_{||},z_h,z_e)$ is supposed to hold. This is a quite general expression for the deformation-potential coupling. To obtain a more explicit expression, the envelope function $F_\lambda(\mathbf{r}_{||},z_e,z_h)$ must be specified. For the lowest $(1s)$ exciton state, a variational envelope function was assumed as[11]

$$F_{1s}(\mathbf{r}_{||},z_e,z_h)=N_n\exp\{-[\alpha^2r^2_{||}+\beta^2(z_e-z_h)^2]^{1/2}\}\cos\left[\frac{\pi z_e}{L_z}\right]\cos\left[\frac{\pi z_h}{L_z}\right]\ , \tag{2.16}$$

where N_n is the normalization constant and α and β are variational parameters to minimize the energy. Here the infinite band-gap discontinuity is assumed and the envelope function is zero outside the region of $|\,z_e\,|\le L_z/2$ and $|\,z_h\,|\le L_z/2$. With use of this envelope function, the function $H_{1s}(\mathbf{Q}_{||},Q_z)$ is calculated as

$$H_{1s}(\mathbf{Q}_{||},Q_z)=\int d^2r_{||}\int_{-L_z/2}^{L_z/2}dz_e\int_{-L_z/2}^{L_z/2}dz_h\exp(iQ_zz_e+i\mathbf{Q}_{||}\cdot\mathbf{r}_{||})\,|\,F_{1s}(\mathbf{r}_{||},z_e,z_h)\,|^2$$
$$=N_n^2\int d^2r_{||}\int_{-L_z/2}^{L_z/2}dz_e\int_{-L_z/2}^{L_z/2}dz_h\exp\{iQ_zz_e+i\mathbf{Q}_{||}\cdot\mathbf{r}_{||}-2[\alpha^2r^2_{||}+\beta^2(z_e-z_h)^2]^{1/2}\}\cos^2\left[\frac{\pi z_e}{L_z}\right]\cos^2\left[\frac{\pi z_h}{L_z}\right]\ . \tag{2.17}$$

By making the variables nondimensional, this can be reduced to

$$\frac{N_n^2L_z^2}{\alpha^2}\int d^2r_{||}\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h\exp\{iQ_zL_zz_e+i\mathbf{Q}_{||}\cdot\mathbf{r}_{||}/\alpha-2[r^2_{||}+\beta^2L_z^2(z_e-z_h)^2]^{1/2}\}\cos^2(\pi z_e)\cos^2(\pi z_h)\ . \tag{2.18}$$

Introducing the two-dimensional polar coordinate for $\mathbf{r}_{||}$ and making use of the decomposition formula[15]

$$e^{iz\cos\theta}=\sum_{n=-\infty}^{\infty}J_n(z)\exp[in(\theta+\pi/2)]\ , \tag{2.19}$$

one can further reduce (2.18) to

$$\frac{2\pi N_n^2 L_z^2}{\alpha^2}\int_0^\infty dr\, rJ_0(br)\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h \exp\{iQ_zL_zz_e-2[r^2+\beta^2L_z^2(z_e-z_h)^2]^{1/2}\}\cos^2(\pi z_e)\cos^2(\pi z_h)\ , \tag{2.20}$$

where $b=|\mathbf{Q}_{||}|/\alpha$ and J_0 is the zeroth-order Bessel function. By use of the formula[15]

$$\int_0^\infty dx\, xJ_0(bx)\exp[-a(x^2+y^2)^{1/2}]=\frac{a[1+(a^2+b^2)^{1/2}y]}{(a^2+b^2)^{3/2}}\exp[-y(a^2+b^2)^{1/2}]\ , \tag{2.21}$$

the integral over r in (2.20) can be performed as

$$\frac{4\pi N_n^2L_z^2}{\alpha^2(4+b^2)^{3/2}}\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h\exp[iQ_zL_zz_e-(4+b^2)^{1/2}\beta L_z|z_e-z_h|]$$

$$\times[1+(4+b^2)^{1/2}\beta L_z|z_e-z_h|]\cos^2(\pi z_e)\cos^2(\pi z_h)$$

$$=\frac{4\pi N_n^2L_z^2}{\alpha^2(4+b^2)^{3/2}}G((4+b^2)^{1/2}\beta L_z,Q_zL_z)\ , \tag{2.22}$$

where the function $G(\gamma,\delta)$ is defined by

$$G(\gamma,\delta)=\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h\exp(i\delta z_e-\gamma|z_e-z_h|)(1+\gamma|z_e-z_h|)\cos^2(\pi z_e)\cos^2(\pi z_h)\ , \tag{2.23}$$

and the explicit expression of G is given in the Appendix. Consequently, one obtains

$$H_{1s}(\mathbf{Q}_{||},Q_z)=\frac{4\pi N_n^2L_z^2}{\alpha^2(4+b^2)^{3/2}}G((4+b^2)^{1/2}\beta L_z,Q_zL_z)\ , \tag{2.24}$$

with $b=|\mathbf{Q}_{||}|/\alpha$.

Let us now determine the normalization constant N_n in (2.16). The quasi-two-dimensional exciton state given by (2.1) is normalized as

$$1=\langle\lambda,\mathbf{K}_{||}|\lambda,\mathbf{K}_{||}\rangle=\frac{v_0^2}{L^2}\sum_{\mathbf{r}_e,\mathbf{r}_h}|F_\lambda(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h)|^2$$

$$=\frac{1}{L^2}\int d^3r_e\int d^3r_h\,|F_\lambda(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h)|^2\ , \tag{2.25}$$

where the discrete sum is converted into an integral by (2.4). Substituting the variational envelope function (2.16), it is calculated as

$$1=N_n^2\int d^2r_{||}\int_{-L_z/2}^{L_z/2}dz_e\int_{-L_z/2}^{L_z/2}dz_h\exp\{-2[\alpha^2r_{||}^2+\beta^2(z_e-z_h)^2]^{1/2}\}\cos^2\left[\frac{\pi z_e}{L_z}\right]\cos^2\left[\frac{\pi z_h}{L_z}\right]$$

$$=2\pi N_n^2\int_0^\infty dr\,r\int_{-L_z/2}^{L_z/2}dz_e\int_{-L_z/2}^{L_z/2}dz_h\exp\{-2[\alpha^2r^2+\beta^2(z_e-z_h)^2]^{1/2}\}\cos^2\left[\frac{\pi z_e}{L_z}\right]\cos^2\left[\frac{\pi z_h}{L_z}\right]\ . \tag{2.26}$$

By use of the partial integration, the r integration can be performed as

$$\int_0^\infty dr\,r\exp[-2(\alpha^2r^2+\Delta^2)^{1/2}]=e^{-2|\Delta|}(1+2|\Delta|)/4\alpha^2\ , \tag{2.27}$$

where Δ is independent of r. Then the normalization condition is reduced to

$$1=\frac{\pi N_n^2L_z^2}{2\alpha^2}\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h(1+2\beta L_z|z_e-z_h|)\exp(-2\beta L_z|z_e-z_h|)\cos^2(\pi z_e)\cos^2(\pi z_h)\ , \tag{2.28}$$

where the variables are made nondimensional. Let us introduce an integral $I(\gamma)$ defined by

$$I(\gamma)=\tfrac{1}{2}\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h(1+\gamma|z_e-z_h|)e^{-\gamma|z_e-z_h|}\cos^2(\pi z_e)\cos^2(\pi z_h)\ , \tag{2.29}$$

whose explicit expression is given in the Appendix. Finally, the normalization condition becomes

$$1=\pi N_n^2L_z^2I(2\beta L_z)/\alpha^2\ , \tag{2.30}$$

and the normalization constant N_n is determined as

$$N_n=\frac{\alpha}{L_z[\pi I(2\beta L_z)]^{1/2}} . \tag{2.31}$$

Summarizing all the results, the quasi-two-dimensional exciton-phonon interaction for the deformation-potential coupling is obtained as

$$H_{\text{ex-ph}}^{\text{DF (Q2D)}}=\tfrac{1}{2}\sum_{\mathbf{K}_{||},\mathbf{K}'_{||},q_z}\left[\frac{\hbar[(\mathbf{K}_{||}-\mathbf{K}'_{||})^2+q_z^2]^{1/2}}{2\rho uV}\right]^{1/2} \times\left[\frac{D_c G((4+b_h^2)^{1/2}\beta L_z,q_z L_z)}{[1+(b_h/2)^2]^{3/2}}-\frac{D_v G((4+b_e^2)^{1/2}\beta L_z,q_z L_z)}{[1+(b_e/2)^2]^{3/2}}\right]\Big/ I(2\beta L_z) \times|1s,\mathbf{K}'_{||}\rangle\langle 1s,\mathbf{K}_{||}|(b_{\mathbf{K}'_{||}-\mathbf{K}_{||},q_z}+b^{\dagger}_{\mathbf{K}_{||}-\mathbf{K}'_{||},-q_z}) , \tag{2.32}$$

with

$$b_h=\alpha_h|\mathbf{K}_{||}-\mathbf{K}'_{||}|/\alpha,\quad b_e=\alpha_e|\mathbf{K}_{||}-\mathbf{K}'_{||}|/\alpha ,$$

where α_e and α_h are defined in (2.7) and α and β are the variational parameters in (2.16). This expression is rather complicated, and thus it is instructive to look into the expression under a few limiting situations. When the QW thickness L_z is zero, namely in the extremely two-dimensional case, $H_{\text{ex-ph}}^{\text{DF (Q2D)}}$ may be simplified to

$$H_{\text{ex-ph}}^{\text{DF (2D)}}=\lim_{L_z\to 0}H_{\text{ex-ph}}^{\text{DF (Q2D)}}=\sum_{\mathbf{K}_{||},\mathbf{K}'_{||}}\left[\frac{\hbar|\mathbf{K}_{||}-\mathbf{K}'_{||}|}{2\rho uS}\right]^{1/2}\left[\frac{D_c}{[1+(b_h/2)^2]^{3/2}}-\frac{D_v}{[1+(b_e/2)^2]^{3/2}}\right] \times|1s,\mathbf{K}'_{||}\rangle\langle 1s,\mathbf{K}_{||}|(b_{\mathbf{K}'_{||}-\mathbf{K}_{||}}+b^{\dagger}_{\mathbf{K}_{||}-\mathbf{K}'_{||}}) , \tag{2.33}$$

where the component q_z of the phonon wave vector is set equal to zero, the quantization volume V is replaced by the quantization area S, and use is made of the relation

$$G(0,0)=2I(0)=\tfrac{1}{4} . \tag{2.34}$$

Secondly, let us consider the three-dimensional limit where L_z becomes infinite. Since in this case there is no preferential spatial direction, q_z is set equal to zero and the subscript $||$ on $\mathbf{K}$ is dropped. It is seen that

$$G(\gamma,0)=2I(\gamma) , \tag{2.35}$$

and the limiting form of (2.29) is given as

$$\lim_{\gamma\to\infty}I(\gamma)=3/4\gamma . \tag{2.36}$$

Then, when the QW thickness L_z is infinite, i.e., in the three-dimensional case, $H_{\text{ex-ph}}^{\text{DF (Q2D)}}$ may be simplified to

$$H_{\text{ex-ph}}^{\text{DF (3D)}}=\lim_{L_z\to\infty}H_{\text{ex-ph}}^{\text{DF (Q2D)}}=\sum_{\mathbf{K},\mathbf{K}'}\left[\frac{\hbar|\mathbf{K}-\mathbf{K}'|}{2\rho uV}\right]^{1/2} \times\left[\frac{D_c}{[1+(b_h/2)^2]^2}-\frac{D_v}{[1+(b_e/2)^2]^2}\right]|1s,\mathbf{K}'\rangle\langle 1s,\mathbf{K}|(b_{\mathbf{K}'-\mathbf{K}}+b^{\dagger}_{\mathbf{K}-\mathbf{K}'}) . \tag{2.37}$$

This is exactly the well-known result for the three-dimensional exciton.[14] Comparing (2.33) and (2.37), it is found that the most striking difference between the two- and three-dimensional cases consists in the power law in the second set of large parentheses. For the long-wavelength phonons, this difference may not be significant since the quantities in the second sets of large parentheses of (2.33) and (2.37) are both essentially D_c-D_v. On the other hand, when the short-wavelength phonons are concerned, the difference in the power law will lead to a significant difference in the optical and transport properties.

So far, the deformation-potential coupling has been discussed exclusively. However, it is known that the contribution from the piezoelectric coupling is not negligible in GaAs. The piezoelectric coupling arises from the longitudinal electric field induced by the strain field associated with acoustic-phonon modes. The piezoelectric electron-phonon interaction Hamiltonian for the zinc-blende type crystal with T_d symmetry is given by[16]

$$H_{e\text{-ph}}^{\text{PZ}}=\sum_{\mathbf{k},\mathbf{q},\sigma}\frac{8\pi e e_{14}}{\epsilon_0 q^2}\left[\frac{\hbar}{2\rho\omega_\sigma(\mathbf{q})V}\right]^{1/2}(\xi_x q_y q_z+\xi_y q_x q_z+\xi_z q_x q_y)(a_{c,\mathbf{k}+\mathbf{q}}^{\dagger}a_{c\mathbf{k}}+a_{v,\mathbf{k}+\mathbf{q}}^{\dagger}a_{v\mathbf{k}})(b_{\mathbf{q}\sigma}+b_{-\mathbf{q}\sigma}^{\dagger}) , \tag{2.38}$$

where e_{14}, ϵ_0, and ξ_α are the piezoelectric constant, the longitudinal dielectric constant without the piezoelectric contribution, and the αth component of the phonon polarization vector, respectively, and the subscript σ specifies the longitudinal-acoustic (LA) or transverse-acoustic (TA) phonon mode. The Cartesian components in (2.38) are referred to with respect to the cubic crystallographic axes of the zinc-blende type crystal. The derivation of the piezoelectric exciton-phonon interaction for the quasi-two-dimensional exciton is straightforward. Repeating the same procedure as in (2.10)—(2.15), one obtains

$$\begin{aligned}H_{\text{ex-ph}}^{\text{PZ (Q2D)}}=&\frac{4\pi e e_{14}}{\epsilon_0}\sum_{\mathbf{K}_{||},\mathbf{K}'_{||},q_z,\sigma}\left[\frac{\hbar}{2\rho u_\sigma V(|\mathbf{K}_{||}-\mathbf{K}'_{||}|^2+q_z^2)^{5/2}}\right]^{1/2}\\&\times[\xi_x(\mathbf{K}'_{||}-\mathbf{K}_{||})_y q_z+\xi_y(\mathbf{K}'_{||}-\mathbf{K}_{||})_x q_z+\xi_z(\mathbf{K}'_{||}-\mathbf{K}_{||})_x(\mathbf{K}'_{||}-\mathbf{K}_{||})_y]\\&\times\left[\frac{G((4+b_h^2)^{1/2}\beta L_z,q_z L_z)}{[1+(b_h/2)^2]^{3/2}}-\frac{G((4+b_e^2)^{1/2}\beta L_z,q_z L_z)}{[1+(b_e/2)^2]^{3/2}}\right]\Big/I(2\beta L_z)\\&\times|1s,\mathbf{K}'_{||}\rangle\langle 1s,\mathbf{K}_{||}|(b_{\mathbf{K}'_{||}-\mathbf{K}_{||},q_z,\sigma}+b_{\mathbf{K}_{||}-\mathbf{K}'_{||},-q_z,\sigma}^{\dagger}) ,\end{aligned} \tag{2.39}$$

where the functions G and I are the same as in (2.32), and b_e and b_h are given under (2.32). In this case the factor within the small square brackets of (2.39) gives rise to the anisotropic effect. If the extremely two-dimensional case is considered and q_z is set equal to zero, the anisotropic factor becomes $\xi_z(\mathbf{K}'_{||}-\mathbf{K}_{||})_x(\mathbf{K}'_{||}-\mathbf{K}_{||})_y$, which simply implies that the piezoelectric coupling is possible only with the TA-phonon mode having the polarization vector in the z direction. On the other hand, in the limit $L_z=\infty$ expression (2.39) exactly reproduces the well-known result[16] for the three-dimensional exciton as

$$\begin{aligned}H_{\text{ex-ph}}^{\text{PZ (3D)}}=\lim_{L_z\to\infty}H_{\text{ex-ph}}^{\text{PZ (Q2D)}}=&\sum_{\mathbf{K},\mathbf{q},\sigma}\frac{8\pi e e_{14}}{\epsilon_0 q^2}\left[\frac{\hbar}{2\rho\omega_\sigma(\mathbf{q})V}\right]^{1/2}(\xi_x q_y q_z+\xi_y q_x q_z+\xi_z q_x q_y)\\&\times\left[\frac{1}{[1+(\alpha_h|\mathbf{q}|/2\alpha)^2]^2}-\frac{1}{[1+(\alpha_e|\mathbf{q}|/2\alpha)^2]^2}\right]|1s,\mathbf{K}+\mathbf{q}\rangle\langle 1s,\mathbf{K}|(b_{\mathbf{q}\sigma}+b_{-\mathbf{q}\sigma}^{\dagger}) .\end{aligned} \tag{2.40}$$

Thus, Eq. (2.32) for $H_{\text{ex-ph}}^{\text{DF (Q2D)}}$ and Eq. (2.39) for $H_{\text{ex-ph}}^{\text{PZ (Q2D)}}$ give quite general expressions of the exciton-phonon interaction for the quasi-two-dimensional exciton that reduce smoothly (as $L_z\to\infty$) to those for the three-dimensional exciton.

III. KINETIC-ENERGY RELAXATION OF QUASI-TWO-DIMENSIONAL EXCITONS

We now discuss the kinetic-energy relaxation on the dispersion curve of the quasi-two-dimensional exciton, adapting Conwell's argument,[17] which was originally developed for the three-dimensional case. It will be shown that the kinetic-energy relaxation is too fast to explain the experimentally observed energy relaxation rate. The quasi-two-dimensional exciton state with a total wave vector $\mathbf{K}_{||}$ is denoted by $|\mathbf{K}_{||}\rangle$, assuming the lowest $1s$ state for the electron-hole internal motion. The matrix element of the exciton-phonon interaction derived in Sec. II will be denoted by $H_{\text{ex-ph}}(\mathbf{Q}_{||},Q_z)$ for the phonon wave vector $\mathbf{Q}$ defined by $\mathbf{Q}=(\mathbf{Q}_{||},Q_z)$. The increasing rate of the number of phonons with wave vector $(\mathbf{Q}_{||},Q_z)$ due to phonon emission by excitons is given by

$$\begin{aligned}&\frac{2\pi}{\hbar}\sum_{\mathbf{K}_{||}}|H_{\text{ex-ph}}(\mathbf{Q}_{||},Q_z)|^2(1+n_{\mathbf{Q}})f(\mathbf{K}_{||}+\mathbf{Q}_{||})\\&\quad\times\delta(E(\mathbf{K}_{||}+\mathbf{Q}_{||})-E(\mathbf{K}_{||})-\hbar\omega_{\mathbf{Q}}) ,\end{aligned} \tag{3.1}$$

where n and f are the phonon occupation number and the exciton distribution function, respectively, $E(\mathbf{K}_{||})$ the parabolic two-dimensional exciton energy, and $\hbar\omega_{\mathbf{Q}}$ the energy of acoustic phonon with wave vector $\mathbf{Q}$. Similarly, the decreasing rate of the number of phonons with wave vector $(\mathbf{Q}_{||},Q_z)$ is given by

$$\frac{2\pi}{\hbar}\sum_{\mathbf{K}_{\parallel}}|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2 n_Q f(\mathbf{K}_{\parallel}) \times\delta(E(\mathbf{K}_{\parallel})-E(\mathbf{K}_{\parallel}+\mathbf{Q}_{\parallel})+\hbar\omega_{\mathbf{Q}}) . \tag{3.2}$$

The common argument of the δ functions in (3.1) and (3.2) can be written as

$$E(\mathbf{K}_{\parallel}+\mathbf{Q}_{\parallel})-E(\mathbf{K}_{\parallel})-\hbar\omega_{\mathbf{Q}} =\hbar^2(2\mathbf{K}_{\parallel}\cdot\mathbf{Q}_{\parallel}+Q_{\parallel}^2)/2M-\hbar\omega_{\mathbf{Q}} , \tag{3.3}$$

$$\frac{dN_{\mathbf{Q}}}{dt}=\frac{4\pi}{\hbar}|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2\frac{L^2}{(2\pi)^2}\int_{K_Q}^{\infty}dK_{\parallel}K_{\parallel}\int_0^{\pi}d\theta\frac{M\delta(\theta-\theta_0)}{\hbar^2|\mathbf{K}_{\parallel}\cdot\mathbf{Q}_{\parallel}\sin\theta|}[(1+n_Q)f(\mathbf{K}_{\parallel}+\mathbf{Q}_{\parallel})-n_Q f(\mathbf{K}_{\parallel})] , \tag{3.5}$$

where θ_0 is the angle between $\mathbf{K}_{\parallel}$ and $\mathbf{Q}_{\parallel}$ satisfying the energy-conservation condition and L is the linear dimension of quantization volume defined in Sec. II. In the following let us assume that

$$Mu/\hbar\ll|\mathbf{Q}_{\parallel}|/2 , \tag{3.6}$$

where u is the sound velocity of acoustic-phonon modes and rewrite the integral over $\mathbf{K}_{\parallel}$ in (3.5) with that over the energy defined by $E=\hbar^2K_{\parallel}^2/2M$. Then it is calculated as

$$\frac{dN_{\mathbf{Q}}}{dt}=\frac{4\pi}{\hbar}\frac{L^2}{(2\pi)^2}\left[\frac{M}{\hbar^2}\right]^2\frac{|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2}{|\mathbf{Q}_{\parallel}|}\frac{\hbar}{\sqrt{2M}} \times\int_{E_Q}^{\infty}dE[(1+n_Q)f(E+\hbar\omega_{\mathbf{Q}}) -n_Q f(E)]/\sqrt{E}\,|\sin\theta_0| , \tag{3.7}$$

with

$$E_Q=\hbar^2K_{\mathbf{Q}}^2/2M\cong\hbar^2Q_{\parallel}^2/8M .$$

By the inequality (3.6), one may approximate as

$$\cos\theta_0=(2M\omega_{\mathbf{Q}}/\hbar-Q_{\parallel}^2)/2|\mathbf{K}_{\parallel}\cdot\mathbf{Q}_{\parallel}|\cong-|\mathbf{Q}_{\parallel}|/2|\mathbf{K}_{\parallel}| , \tag{3.8}$$

and one has

$$|\sin\theta_0|\cong(1-E_Q/E_K)^{1/2} , \tag{3.9}$$

where E_K is defined by $\hbar^2K_{\parallel}^2/2M$. Equation (3.7) is the general expression for the increasing rate of phonon numbers. In the following, the exciton distribution function is assumed to follow the Boltzmann statistics, namely

$$f(E)=f_0\exp(-\beta_e E) , \tag{3.10}$$

with

$$\beta_e=1/k_B T_e ,$$

where T_e is the effective temperature of excitons and f_0 is some constant. In fact, the experimentally determined exciton distribution function can be described by the Boltzmann statistics fairly well, as will be shown in Sec. VII. This situation may be attained by frequent collisions among excitons. Then, calculating the integral

where M is the exciton translational mass. The energy-conservation condition leads to the condition that the magnitude of exciton wave vector $\mathbf{K}_{\parallel}$ must satisfy

$$|\mathbf{K}_{\parallel}|\geq|2M\omega_{\mathbf{Q}}/\hbar-Q_{\parallel}^2|/2|\mathbf{Q}_{\parallel}|=K_{\mathbf{Q}} , \tag{3.4}$$

where $K_{\mathbf{Q}}$ is defined by the right-hand side. Then the total increasing rate of the number of phonons with wave vector $(\mathbf{Q}_{\parallel},Q_z)$ is calculated as

$$\int_{E_Q}^{\infty}dE\frac{e^{-\beta_e E}}{\sqrt{E}\,(1-E_Q/E)^{1/2}}=\left[\frac{\pi}{\beta_e}\right]^{1/2}e^{-\beta_e E_Q} , \tag{3.11}$$

one obtains

$$\frac{dN_{\mathbf{Q}}}{dt}=\frac{4\pi}{\hbar}\frac{L^2}{(2\pi)^2}\left[\frac{M}{\hbar^2}\right]^2\frac{\hbar\sqrt{\pi}}{(2M\beta_e)^{1/2}}\frac{|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2}{|\mathbf{Q}_{\parallel}|} \times f_0 e^{-\beta_e E_Q}[(1+n_Q)\exp(-\beta_e\hbar\omega_{\mathbf{Q}})-n_Q] . \tag{3.12}$$

The average energy relaxation rate of excitons is given by

$$\left\langle\frac{dE}{dt}\right\rangle=-\frac{1}{N}\sum_{\mathbf{Q}}\hbar\omega_{\mathbf{Q}}\frac{dN_{\mathbf{Q}}}{dt} , \tag{3.13}$$

where N is the total number of excitons defined by

$$N=\sum_{\mathbf{K}_{\parallel}}f(E(\mathbf{K}_{\parallel}))=\frac{f_0ML^2}{2\pi\beta_e\hbar^2} . \tag{3.14}$$

The explicit expression of (3.13) is written as

$$\left\langle\frac{dE}{dt}\right\rangle=-\frac{(2\pi M\beta_e)^{1/2}}{\hbar^2} \times\sum_{\mathbf{Q}}\frac{|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2}{|\mathbf{Q}_{\parallel}|}\hbar\omega_{\mathbf{Q}} \times e^{-\beta_e E_Q}[(1+n_Q)\exp(-\beta_e\hbar\omega_{\mathbf{Q}})-n_Q] . \tag{3.15}$$

To obtain a more explicit result, the deformation-potential coupling derived in Sec. II will be substituted for the exciton-phonon interaction $H_{\text{ex-ph}}$. Since the function G in (2.32) is a slowly varying function with respect to $|\mathbf{Q}_{\parallel}|$ and Q_z for the physically important range of parameters, one may safely set $Q_z=0$, i.e., $|\mathbf{Q}_{\parallel}|=|\mathbf{Q}|$ in G to obtain

$$H_{\text{ex-ph}}^{\text{DF}}(\mathbf{Q}_{\|},Q_z)\cong\left[\frac{\hbar|\mathbf{Q}|}{2\rho uV}\right]^{1/2}\left[\frac{D_c}{[1+(\alpha_h|\mathbf{Q}|/2\alpha)^2]^{3/2}}-\frac{D_v}{[1+(\alpha_e|\mathbf{Q}|/2\alpha)^2]^{3/2}}\right]$$

$$\cong\left[\frac{\hbar|\mathbf{Q}|}{2\rho uV}\right]^{1/2}(D_c-D_v)\ , \tag{3.16}$$

where to simplify the algebraic manipulations, it is supposed that b_h and b_e in (2.32) are small, and that $(4+b_h^2)^{1/2}$ and $(4+b_e^2)^{1/2}$ are nearly equal to 2. Substituting this expression into (3.15), one obtains

$$\left\langle\frac{dE}{dt}\right\rangle=-(2\pi M\beta_e)^{1/2}\frac{(D_c-D_v)^2}{4\pi^2\rho}\times\int_0^\infty dQ\,Q^3e^{-\beta_eE_Q}\times[(1+n_{\mathbf{Q}})\exp(-\beta_e\hbar\omega_{\mathbf{Q}})-n_{\mathbf{Q}}]\ , \tag{3.17}$$

with

$$E_Q=\hbar^2Q^2/8M\ .$$

By using the experimental data

$$T_e\cong 20\text{ K}\quad\text{and}\quad T_L=4.2\text{ K}\ ,$$

where T_L is the lattice temperature and, choosing the material parameters of GaAs given in detail in Sec. VII, the energy relaxation rate is calculated as

$$\left\langle\frac{dE}{dt}\right\rangle\cong 20\times10^6\text{ eV/s}\ . \tag{3.18}$$

This value is 1 order of magnitude larger than the observed value of $(2-3)\times10^6$ eV/s. Furthermore, it can be shown that the effective temperature of excitons decreases from 20 to 4.2 K within several tens of picoseconds. Since there is an additional relaxation mechanism due to the piezoelectric coupling, the theoretical value of the energy relaxation rate becomes larger and the discrepancy increases. Thus it is concluded that the observed slow energy relaxation cannot be explained by the kinetic-energy relaxation on the dispersion curve of the quasi-two-dimensional exciton. When the excitons relax on the two-dimensional dispersion curve and accumulate on the low-energy portion of the density of states, the excitons become more and more immobile and can be considered as distributed among the energetically local minimum sites which are induced by the well-thickness fluctuation in the lateral direction of a QW. Under such a situation the energy relaxation occurs through the exciton migration in search of the lower-energy sites, and the exciton system eventually approaches the energetically global minimum state. This process of energy relaxation is considered slow compared to the kinetic-energy relaxation and, in fact, explains quantitatively the experimental energy relaxation rate.

IV. ONE-PHONON-ASSISTED TRANSFER OF LOCALIZED QUASI-TWO-DIMENSIONAL EXCITONS

Let us now consider the quasi-two-dimensional exciton transfer among localized sites, such as the islandlike structures in a QW that are induced by the well-thickness fluctuation in the lateral direction. A general theory will be developed without recourse to the details of the localized sites, namely the microscopic structure of the disorder. In the process of exciton transfer the energy mismatch of excitons is compensated for by acoustic phonons. At low temperatures only the one-phonon-assisted process needs to be taken into account, since the relevant energy mismatch is less than 1 meV and rather small. The exciton state localized at site $\mathbf{R}_a$ will be denoted by $|\mathbf{R}_a\rangle$, assuming the lowest $1s$ state for the electron-hole internal motion. The relevant Hamiltonians for the phonon-assisted exciton transfer are the exciton-phonon interaction Hamiltonian denoted by $H_{\text{ex-ph}}$ and the intersite transfer Hamiltonian denoted by H_{ss}. Then there are three possibilities for the exciton transfer from site $\mathbf{R}_a$ to site $\mathbf{R}_b$, namely

$$\text{(a)}\quad|\mathbf{R}_a;n_{\mathbf{Q}}\rangle\xrightarrow[H_{\text{ex-ph}}]{}|\mathbf{R}_b;n_{\mathbf{Q}}\pm1\rangle\ , \tag{4.1}$$

$$\text{(b)}\quad|\mathbf{R}_a;n_{\mathbf{Q}}\rangle\xrightarrow[H_{\text{ex-ph}}]{}|\mathbf{R}_a;n_{\mathbf{Q}}\pm1\rangle\xrightarrow[H_{ss}]{}|\mathbf{R}_b;n_{\mathbf{Q}}\pm1\rangle\ , \tag{4.2}$$

$$\text{(c)}\quad|\mathbf{R}_a;n_{\mathbf{Q}}\rangle\xrightarrow[H_{ss}]{}|\mathbf{R}_b;n_{\mathbf{Q}}\rangle\xrightarrow[H_{\text{ex-ph}}]{}|\mathbf{R}_b;n_{\mathbf{Q}}\pm1\rangle\ , \tag{4.3}$$

where $n_{\mathbf{Q}}$ represents the occupation number of phonons relevant to the exciton transfer. Term (a) arises from the first-order perturbation process with respect to $H_{\text{ex-ph}}$, whereas terms (b) and (c) are the contributions from the second-order perturbation process using both $H_{\text{ex-ph}}$ and H_{ss} once for each.[18] As will be seen later, term (a) is possible through the overlap of exciton wave functions and is short ranged in nature, while terms (b) and (c) are effective over a long range, in general. The transition amplitude of the exciton transfer for each process in (4.1)—(4.3) is given as follows:

$$\text{(a)}\quad\langle\mathbf{R}_b;n_{\mathbf{Q}}\pm1|H_{\text{ex-ph}}|\mathbf{R}_a;n_{\mathbf{Q}}\rangle\ , \tag{4.4}$$

$$\text{(b)}\quad\frac{\langle\mathbf{R}_b;n_{\mathbf{Q}}\pm1|H_{ss}|\mathbf{R}_a;n_{\mathbf{Q}}\pm1\rangle\langle\mathbf{R}_a;n_{\mathbf{Q}}\pm1|H_{\text{ex-ph}}|\mathbf{R}_a;n_{\mathbf{Q}}\rangle}{\mp\hbar\omega_{\mathbf{Q}}}\ , \tag{4.5}$$

$$\text{(c)}\quad\frac{\langle\mathbf{R}_b;n_{\mathbf{Q}}\pm1|H_{\text{ex-ph}}|\mathbf{R}_b;n_{\mathbf{Q}}\rangle\langle\mathbf{R}_b;n_{\mathbf{Q}}|H_{ss}|\mathbf{R}_a;n_{\mathbf{Q}}\rangle}{\pm\hbar\omega_{\mathbf{Q}}}\ , \tag{4.6}$$

where $\omega_{\mathbf{Q}}$ is the phonon frequency with wave vector $\mathbf{Q}$. The intersite exciton transfer Hamiltonian H_{ss} arises from the electron-electron interaction Hamiltonian, and the matrix element is independent of the phonon state. Thus it can be written as

$$J(|\mathbf{R}_a-\mathbf{R}_b|)=\langle \mathbf{R}_b | H_{ss} | \mathbf{R}_a \rangle . \tag{4.7}$$

The explicit calculation of $J(\mathbf{R})$ will be given in Sec. V. To calculate the matrix element of $H_{\text{ex-ph}}$, the localized exciton state must be specified more explicitly.

The localized quasi-two-dimensional exciton state can be represented as

$$|\mathbf{R}_a\rangle = v_0 \sum_{\mathbf{r}_e,\mathbf{r}_h} G(\mathbf{R}_{||}-\mathbf{R}_a) F_{1s}(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h) \times a^{\dagger}_{c\mathbf{r}_e} a_{v\mathbf{r}_h} |0\rangle , \tag{4.8}$$

where the $1s$ exciton envelope function F_{1s} is given in (2.16) and the function G describes the two-dimensional localization of the exciton center of mass $\mathbf{R}$ defined by (2.2). The notations v_0, $a_{c\mathbf{r}_e}$, and $a_{v\mathbf{r}_h}$ are given in Sec. II. The function G is normalized as

$$\int d^2R_{||} \, |G(\mathbf{R}_{||}-\mathbf{R}_a)|^2 = 1 , \tag{4.9}$$

and thus the localized exciton state $|\mathbf{R}_a\rangle$ is normalized correctly

$$\langle \mathbf{R}_a | \mathbf{R}_a \rangle = 1 . \tag{4.10}$$

The functional form of G depends on the details of microscopic configuration of the localized state. Until now, there has been no systematic study of this subject. However, the dynamical properties of the system, such as the energy relaxation, are not expected to be very sensitive to the microscopic details of localization, but may be characterized by only a few parameters, such as the localization length. In this paper two typical cases of exciton localization will be examined, namely (1) the Gaussian case,

$$G(\mathbf{R}_{||}) = \frac{1}{\sqrt{\pi}\xi} \exp(-R_{||}^2/2\xi^2) , \tag{4.11}$$

and (2) the exponential case,

$$G(\mathbf{R}_{||}) = \frac{1}{\sqrt{2\pi}\xi} \exp(-|\mathbf{R}_{||}|/2\xi) , \tag{4.12}$$

where ξ is the characteristic localization length. Then, introducing the two-dimensional Fourier transform of the localization function by

$$G(\mathbf{R}_{||}-\mathbf{R}_a) = \int d^2K_{||} \, e^{i\mathbf{K}_{||}\cdot\mathbf{R}_{||}} g(\mathbf{K}_{||},\mathbf{R}_a) , \tag{4.13}$$

one obtains

$$\begin{aligned} |\mathbf{R}_a\rangle &= v_0 \sum_{\mathbf{r}_e,\mathbf{r}_h} G(\mathbf{R}_{||}-\mathbf{R}_a) F_{1s}(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h) a^{\dagger}_{c\mathbf{r}_e} a_{v\mathbf{r}_h} |0\rangle \\ &= \int d^2K_{||} \, g(\mathbf{K}_{||},\mathbf{R}_a) v_0 \sum_{\mathbf{r}_e,\mathbf{r}_h} e^{i\mathbf{K}_{||}\cdot\mathbf{R}_{||}} F_{1s}(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h) a^{\dagger}_{c\mathbf{r}_e} a_{v\mathbf{r}_h} |0\rangle \\ &= L \int d^2K_{||} \, g(\mathbf{K}_{||},\mathbf{R}_a) |\mathbf{K}_{||}\rangle . \end{aligned} \tag{4.14}$$

where use is made of (2.1). Thus, evidently, the localized exciton state consists of the superposition of extended exciton states. The explicit expression of g is given as follows: (1) Gaussian case,

$$g(\mathbf{K}_{||},\mathbf{R}_a) = \frac{\xi}{2\pi\sqrt{\pi}} \exp(-i\mathbf{K}_{||}\cdot\mathbf{R}_a - \xi^2 K_{||}^2/2) , \tag{4.15}$$

and (2) exponential case,

$$\begin{aligned} g(\mathbf{K}_{||},\mathbf{R}_a) &= \frac{e^{-i\mathbf{K}_{||}\cdot\mathbf{R}_a}}{2\pi\sqrt{2\pi}\xi} \int_0^{\infty} dR \, R J_0(K_{||}R) \exp(-R/2\xi) \\ &= \frac{e^{-i\mathbf{K}_{||}\cdot\mathbf{R}_a}}{\pi\sqrt{2\pi}} \frac{2\xi}{[1+(2\xi K_{||})^2]^{3/2}} , \end{aligned} \tag{4.16}$$

where ($K_{||}=|\mathbf{K}_{||}|$, $J_0(x)$ is the zeroth-order Bessel function, and the following formula[15] is used:

$$\int_0^{\infty} dx \, x \, e^{-ax} J_0(bx) = \frac{a}{(a^2+b^2)^{3/2}} . \tag{4.17}$$

The matrix element of the exciton-phonon interaction can be calculated by using the expression (4.14) as

$$\langle \mathbf{R}_b;n_{\mathbf{Q}}\pm 1 | H_{\text{ex-ph}} | \mathbf{R}_a;n_{\mathbf{Q}}\rangle = L^2 \int d^2K'_{||} \int d^2K_{||} \, g^*(\mathbf{K}'_{||},\mathbf{R}_b) g(\mathbf{K}_{||},\mathbf{R}_a) \langle \mathbf{K}'_{||};n_{\mathbf{Q}}\pm 1 | H_{\text{ex-ph}} | \mathbf{K}_{||};n_{\mathbf{Q}}\rangle . \tag{4.18}$$

The matrix element on the right-hand side is taken between the two-dimensionally extended exciton states and can be written as

$$\langle \mathbf{K}'_{||};n_{\mathbf{Q}}\pm 1 | H_{\text{ex-ph}} | \mathbf{K}_{||};n_{\mathbf{Q}}\rangle = \delta_{\mathbf{K}'_{||}\pm\mathbf{Q}_{||},\mathbf{K}_{||}} \langle \mathbf{K}'_{||} | H_{\text{ex-ph}} | \mathbf{K}_{||}\rangle_{\mathbf{Q}} = \frac{(2\pi)^2}{L^2} \delta^{(2)}(\mathbf{K}'_{||}\pm\mathbf{Q}_{||}-\mathbf{K}_{||}) H_{\text{ex-ph}}(\mathbf{Q}_{||},Q_z) , \tag{4.19}$$

where in the second equality the Kronecker δ function is changed to the Dirac δ function and a factor related to the phonon absorption or emission is not written explicitly by incorporating the factor in $H_{\text{ex-ph}}(\mathbf{Q}_{||},Q_z)$. As shown in Sec. II, the matrix element $\langle \mathbf{K}'_{||} | H_{\text{ex-ph}} | \mathbf{K}_{||}\rangle_{\mathbf{Q}}$ depends only on the phonon momentum $\mathbf{Q}$ and thus is written as $H_{\text{ex-ph}}(\mathbf{Q}_{||},Q_z)$. Then, in the case of Gaussian localization one obtains

$$\langle \mathbf{R}_b;n_{\mathbf{Q}}\pm1 \mid H_{\text{ex-ph}} \mid \mathbf{R}_a;n_{\mathbf{Q}}\rangle = \exp\left[-i\frac{\mathbf{Q}_{\parallel}\cdot(\mathbf{R}_a+\mathbf{R}_b)}{2}-\frac{\xi^2Q_{\parallel}^2}{4}-\frac{(\mathbf{R}_a-\mathbf{R}_b)^2}{4\xi^2}\right]H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)\ , \tag{4.20}$$

while, in the case of exponential localization, the result is given as

$$\langle \mathbf{R}_b;n_{\mathbf{Q}}\pm1 \mid H_{\text{ex-ph}} \mid \mathbf{R}_a;n_{\mathbf{Q}}\rangle = \frac{8\xi^2}{\pi}\exp[-i\mathbf{Q}_{\parallel}\cdot(\mathbf{R}_a+\mathbf{R}_b)/2] \times\int d^2K_{\parallel}\frac{\exp[i\mathbf{K}_{\parallel}\cdot(\mathbf{R}_b-\mathbf{R}_a)]}{[1+(2\xi\mid\mathbf{K}_{\parallel}-\mathbf{Q}_{\parallel}/2\mid)^2]^{3/2}[1+(2\xi\mid\mathbf{K}_{\parallel}+\mathbf{Q}_{\parallel}/2\mid)^2]^{3/2}}H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)\ . \tag{4.21}$$

Now, the transition amplitude of the exciton transfer from site $\mathbf{R}_a$ to site $\mathbf{R}_b$ for the case of Gaussian localization is calculated as

$$\langle \mathbf{R}_b \mid T \mid \mathbf{R}_a\rangle_{\mathbf{Q}} = e^{-i\mathbf{Q}_{\parallel}\cdot(\mathbf{R}_a+\mathbf{R}_b)/2}\exp\left[-\frac{\xi^2Q_{\parallel}^2}{4}-\frac{(\mathbf{R}_a-\mathbf{R}_b)^2}{4\xi^2}\right]H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z) + \frac{J(\mid\mathbf{R}_a-\mathbf{R}_b\mid)}{E_a-E_b}(e^{-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_b}-e^{-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_a})\exp\left[-\frac{\xi^2Q_{\parallel}^2}{4}\right]H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)\ . \tag{4.22}$$

The first term on the right-hand side is the contribution from process (a) in (4.1) and the second term combines the contribution from processes (b) and (c) in (4.2) and (4.3). The first term contains the Gaussian factor $\exp[-(\mathbf{R}_a-\mathbf{R}_b)^2/4\xi^2]$ arising from the overlap integral between two localized exciton states and has a short-range character. On the other hand, the second term in (4.22) depends on the distance $\mid\mathbf{R}_a-\mathbf{R}_b\mid$ through the function $J(\mid\mathbf{R}_a-\mathbf{R}_b\mid)$ and the coherence factor

$$\exp(-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_b)-\exp(-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_a)\ ,$$

which are generally effective over a long range. The common factor $\exp(-\xi^2Q_{\parallel}^2/4)$ implies that the magnitude of the wave vector of phonons which can interact with the localized exciton is limited within a few times the inverse localization length. In the case of exponential localization, the transition amplitude shows similar features, although its expression is more complicated. When the transition amplitude is obtained, the exciton transfer rate can be calculated by

$$T(\mid E_a-E_b\mid,\mid\mathbf{R}_a-\mathbf{R}_b\mid)=\frac{2\pi}{\hbar}\sum_{\mathbf{Q}}\mid\langle\mathbf{R}_b\mid T\mid\mathbf{R}_a\rangle_{\mathbf{Q}}\mid^2 \times\delta(E_a-E_b\pm\hbar\omega_{\mathbf{Q}})\ . \tag{4.23}$$

In the absolute square of the transition amplitude, there appears the interference between two terms in (4.22). However, this interference term will be neglected since the relevant spatial range of the two terms is quite different and the contribution from the interference term may be small. To calculate the exciton transfer rate more explicitly, the matrix element of the intersite transfer Hamiltonian H_{ss} must be calculated. This matrix element, $J(R)$, will be studied in the next section.

V. INTERSITE TRANSFER MATRIX ELEMENT FOR LOCALIZED QUASI-TWO-DIMENSIONAL EXCITONS

In this section the matrix element $J(R)$ of the intersite transfer Hamiltonian H_{ss} is calculated, and it is shown that $J(R)$ behaves like the dipole-dipole interaction at a distance much longer than the exciton Bohr radius and the localization length. As given in (4.8), the localized quasi-two-dimensional exciton state can be represented as

$$\mid\mathbf{R}_a\rangle = v_0\sum_{\mathbf{r}_e,\mathbf{r}_h}\tilde{F}(\mathbf{r}_e,\mathbf{r}_h;\mathbf{R}_a)a_{c\mathbf{r}_e}^{\dagger}a_{v\mathbf{r}_h}\mid0\rangle = v_0\sum_{\mathbf{r}_e,\mathbf{r}_h}G(\mathbf{R}_{\parallel}-\mathbf{R}_a)F_{1s}(\mathbf{r}_{e\parallel}-\mathbf{r}_{h\parallel},z_e,z_h)a_{c\mathbf{r}_e}^{\dagger}a_{v\mathbf{r}_h}\mid0\rangle\ . \tag{5.1}$$

The intersite transfer Hamiltonian H_{ss} is given by the electron-electron interaction Hamiltonian, namely

$$H_{ss}=\tfrac{1}{2}\int d^3r\int d^3r'\,\psi^{\dagger}(\mathbf{r})\psi^{\dagger}(\mathbf{r}')\frac{e^2}{\epsilon_0\mid\mathbf{r}-\mathbf{r}'\mid}\psi(\mathbf{r}')\psi(\mathbf{r})\ , \tag{5.2}$$

where ϵ_0 and $\psi(\mathbf{r})$ are the dielectric constant and the electron-field operator, respectively. Then the matrix element is calculated as

$$\begin{aligned}J(\mid\mathbf{R}_a-\mathbf{R}_b\mid)&=\langle\mathbf{R}_b\mid H_{ss}\mid\mathbf{R}_a\rangle\\&=v_0^2\sum_{\mathbf{r}_e',\mathbf{r}_h'}\sum_{\mathbf{r}_e,\mathbf{r}_h}\tilde{F}^*(\mathbf{r}_e',\mathbf{r}_h';\mathbf{R}_b)\tilde{F}(\mathbf{r}_e,\mathbf{r}_h;\mathbf{R}_a)\langle0\mid a_{v\mathbf{r}_h'}^{\dagger}a_{c\mathbf{r}_e'}H_{ss}a_{c\mathbf{r}_e}^{\dagger}a_{v\mathbf{r}_h}\mid0\rangle\\&=v_0^2\sum_{\mathbf{r}_e',\mathbf{r}_h'}\sum_{\mathbf{r}_e,\mathbf{r}_h}\tilde{F}^*(\mathbf{r}_e',\mathbf{r}_h';\mathbf{R}_b)\tilde{F}(\mathbf{r}_e,\mathbf{r}_h;\mathbf{R}_a)[V(c\mathbf{r}_e',v\mathbf{r}_h;c\mathbf{r}_e,v\mathbf{r}_h')-V(c\mathbf{r}_e',v\mathbf{r}_h;v\mathbf{r}_h',c\mathbf{r}_e)]\ ,\end{aligned} \tag{5.3}$$

with

$$V(\alpha_1 n_1,\alpha_2 n_2;\alpha_3 n_3,\alpha_4 n_4)=\tfrac{1}{2}\int d^3r\int d^3r'\,\phi^*_{\alpha_1 n_1}(\mathbf{r})\phi^*_{\alpha_2 n_2}(\mathbf{r}')\frac{e^2}{\epsilon_0|\mathbf{r}-\mathbf{r}'|}\phi_{\alpha_3 n_3}(\mathbf{r}')\phi_{\alpha_4 n_4}(\mathbf{r})\ , \tag{5.4}$$

where $\phi_{\alpha n}(\mathbf{r})$ is the Wannier function of the αth band at site n. When the intersite distance is longer than the localization length ξ and the two-dimensional exciton Bohr radius a_B^{2D}, i.e.,

$$|\mathbf{R}_a-\mathbf{R}_b|\gg\xi,a_B^{2D}\ , \tag{5.5}$$

it turns out that the exchange term, namely the first term in square brackets on the right-hand side of (5.3), is dominant. Then, by using the usual multipole expansion of the Coulomb interaction,[19] one obtains

$$J(|\mathbf{R}_a-\mathbf{R}_b|)=\frac{v_0^2}{2|\mathbf{R}_a-\mathbf{R}_b|^3}\boldsymbol{\mu}^\dagger(1-3\mathbf{n}\cdot\mathbf{n})\boldsymbol{\mu}\sum_{\mathbf{r}_e,\mathbf{r}_h}\sum_{\mathbf{r}'_e,\mathbf{r}'_h}\delta_{\mathbf{r}_e,\mathbf{r}_h}\delta_{\mathbf{r}'_e,\mathbf{r}'_h}\tilde{F}^*(\mathbf{r}'_e,\mathbf{r}'_h;\mathbf{R}_b)\tilde{F}(\mathbf{r}_e,\mathbf{r}_h;\mathbf{R}_a)\ , \tag{5.6}$$

with

$$\boldsymbol{\mu}=e\int d^3r\,\phi^*_{v\mathbf{r}_e}(\mathbf{r})(\mathbf{r}-\mathbf{r}_e)\phi_{c\mathbf{r}_e}(\mathbf{r})\ \text{ and }\ \mathbf{n}=(\mathbf{R}_a-\mathbf{R}_b)/|\mathbf{R}_a-\mathbf{R}_b|\ , \tag{5.7}$$

where the Wannier functions are assumed to be well localized at each site. To calculate (5.6) more explicitly, the Gaussian localization in (4.11) will be employed as

$$\tilde{F}(\mathbf{r}_e,\mathbf{r}_h;\mathbf{R}_a)=\frac{1}{\sqrt{\pi}\xi}\exp(-|\mathbf{R}_{||}-\mathbf{R}_a|^2/2\xi^2)F_{1s}(\mathbf{r}_{e||}-\mathbf{r}_{h||},z_e,z_h)\ . \tag{5.8}$$

Using the normalization factor N_n in (2.31), one obtains

$$\begin{aligned}v_0^2\sum_{\mathbf{r}_e,\mathbf{r}_h}\sum_{\mathbf{r}'_e,\mathbf{r}'_h}&\tilde{F}^*(\mathbf{r}'_e,\mathbf{r}'_h;\mathbf{R}_b)\tilde{F}(\mathbf{r}_e,\mathbf{r}_h;\mathbf{R}_a)\delta_{\mathbf{r}_e,\mathbf{r}_h}\delta_{\mathbf{r}'_e,\mathbf{r}'_h}\\&=\left[\frac{1}{\sqrt{\pi}\xi}\int d^2r_{||}\exp(-|\mathbf{r}_{||}-\mathbf{R}|^2/2\xi^2)\right]^2\int_{-L_z/2}^{L_z/2}dz'_e\int_{-L_z/2}^{L_z/2}dz_e\,F^*_{1s}(\mathbf{0},z'_e,z'_e)F_{1s}(\mathbf{0},z_e,z_e)\\&=N_n^2\pi\xi^2L_z^2=(\alpha\xi)^2/I(2\beta L_z)\ ,\end{aligned} \tag{5.9}$$

where the function $I(x)$ is defined in (2.29). Then the matrix element in (5.6) becomes

$$J(|\mathbf{R}_a-\mathbf{R}_b|)=\frac{(\alpha\xi)^2}{2I(2\beta L_z)}\frac{\boldsymbol{\mu}^\dagger(1-3\mathbf{n}\cdot\mathbf{n})\boldsymbol{\mu}}{|\mathbf{R}_a-\mathbf{R}_b|^3}\ . \tag{5.10}$$

This is a typical form of the dipole-dipole interaction. In the following the angular dependence in (5.10) is dropped and the isotropic form will be assumed as

$$J(|\mathbf{R}_a-\mathbf{R}_b|)=\frac{\mu^2(\alpha\xi)^2A}{2I(2\beta L_z)}\frac{1}{|\mathbf{R}_a-\mathbf{R}_b|^3}\ , \tag{5.11}$$

with

$$A=[\langle(1-3\mathbf{n}\cdot\mathbf{n})^2\rangle]^{1/2}=(\tfrac{11}{8})^{1/2}\ , \tag{5.12}$$

where the angular brackets denote the angular average. As is well known, the longitudinal-transverse (LT) splitting of excitons at the zone center is given by[19]

$$\Delta_{\mathrm{LT}}(\mathbf{k}=\mathbf{0})=\frac{4\mu^2}{a_B^3}\ , \tag{5.13}$$

where a_B is the Bohr radius of the three-dimensional exciton. Finally, one arrives at the expression

$$J(|\mathbf{R}_a-\mathbf{R}_b|)=\frac{\Delta_{\mathrm{LT}}a_B^3(\alpha\xi)^2A}{8I(2\beta L_z)}\frac{1}{|\mathbf{R}_a-\mathbf{R}_b|^3}=\frac{\tilde{J}}{|\mathbf{R}_a-\mathbf{R}_b|^3}\ . \tag{5.14}$$

By choosing the values $\Delta_{\mathrm{LT}}=0.08$ meV,[20] $a_B=136$ Å,[21] $\xi=150$ Å, $\alpha^{-1}=100$ Å, and $\beta L_z=0.37$,[11] it is calculated as

$$\tilde{J}(\text{theor})=5.3\times10^2\ \text{eV Å}^3\ . \tag{5.15}$$

This value will be compared with that which gives the best fit of theory to experiment for the energy relaxation in Sec. VII.

So far, the behavior of $J(R)$ has been studied at long distances, i.e., under the condition of (5.5). In the intermediate range, in which the intersite distance is comparable to the exciton Bohr radius and/or the localization length, the calculation of $J(R)$ is a rather involved problem. In this range, an exponential-type tunneling transfer is usually assumed without a rigorous theoretical basis.[22] Some interpolation between tunneling-type transfer and dipole-dipole type transfer may be appropriate to simulate the true behavior of $J(R)$. However, the details of the distance dependence of the intersite transfer do not affect sensitively the energy relaxation of localized excitons, since the energy relaxation rate is determined by the spatial integral of $J^2(R)$ multiplied by other functions. In this paper both cases, dipole-dipole type transfer and tunneling-type transfer, will be examined.

VI. ESTIMATION OF EXCITON TRANSFER RATE

Now that the matrix elements of the quasi-two-dimensional exciton-phonon interaction and the intersite transfer Hamiltonian have been determined, the exciton transfer rate can be estimated explicitly. The transition amplitude is given by (4.22) for the case of Gaussian localization. As mentioned there, the interference between the two terms in (4.22) will be neglected. Then one has

$$|\langle \mathbf{R}_b|T|\mathbf{R}_a\rangle_{\mathbf{Q}}|^2 \cong \exp\left[-\frac{\xi^2 Q_{\parallel}^2}{2}-\frac{|\mathbf{R}_a-\mathbf{R}_b|^2}{2\xi^2}\right]|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2 + \frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}|e^{-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_b}-e^{-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_a}|^2\exp\left[-\frac{\xi^2 Q_{\parallel}^2}{2}\right]|H_{\text{ex-ph}}(\mathbf{Q}_{\parallel},Q_z)|^2 , \tag{6.1}$$

where the exciton-phonon matrix element is abbreviated as $H_{\text{ex-ph}}$, and the arguments $\mathbf{Q}_{\parallel}$ and $\mathbf{Q}_z$ are the components of the phonon wave vector parallel and perpendicular to the QW interface, respectively. In the second term, there appears the coherence factor $|\exp(-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_b)-\exp(-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_a)|^2$ due to the interference between phonon emission or absorption at different sites. The matrix element of the quasi-two-dimensional exciton-phonon interaction for the deformation-potential coupling is given by (2.32) as

$$H_{\text{ex-ph}}^{\text{DF (Q2D)}}(\mathbf{Q}_{\parallel},Q_z)=\frac{1}{2}\left[\frac{\hbar(Q_{\parallel}^2+Q_z^2)^{1/2}}{2\rho u V}\right]^{1/2}\left[D_c\frac{G((4+b_h^2)^{1/2}\beta L_z,Q_zL_z)}{[1+(b_h/2)^2]^{3/2}}-D_v\frac{G((4+b_e^2)^{1/2}\beta L_z,Q_zL_z)}{[1+(b_e/2)^2]^{3/2}}\right]\Big/ I(2\beta L_z) \tag{6.2}$$

$$=\Xi_D(|\mathbf{Q}_{\parallel}|,Q_z)/\sqrt{V} , \tag{6.3}$$

with

$$b_h=\alpha_h|\mathbf{Q}_{\parallel}|/\alpha \quad \text{and} \quad b_e=\alpha_e|\mathbf{Q}_{\parallel}|/\alpha ,$$

where a factor related to the phonon absorption or emission is omitted and Ξ_D is defined by (6.3). In the summation over $\mathbf{Q}=(\mathbf{Q}_{\parallel},Q_z)$ in (4.23), the magnitude $|Q|=(Q_{\parallel}^2+Q_z^2)^{1/2}$ is fixed by the energy-conservation factor and only the angular integration remains. The integration of the coherence factor over the polar angle ϕ results in

$$\int_0^{2\pi}d\phi\,|e^{-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_b}-e^{-i\mathbf{Q}_{\parallel}\cdot\mathbf{R}_a}|^2=4\pi[1-J_0(|\mathbf{Q}_{\parallel}||\mathbf{R}_a-\mathbf{R}_b|)] , \tag{6.4}$$

where J_0 is the zeroth-order Bessel function. Then the transition probability for the deformation-potential (DF) coupling is calculated as

$$T_{\text{DF}}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)=\frac{2\pi}{\hbar}\sum_{\mathbf{Q}}|\langle\mathbf{R}_b|T|\mathbf{R}_a\rangle_{\mathbf{Q}}|^2\delta(E_a-E_b\pm\hbar\omega_{\mathbf{Q}})$$
$$=\frac{Q^2}{2\pi\hbar^2 u}\exp\left[-\frac{|\mathbf{R}_a-\mathbf{R}_b|^2}{2\xi^2}\right]\int_0^{\pi}d\theta\sin\theta\exp\left[-\frac{\xi^2Q^2\sin^2\theta}{2}\right]\Xi_D^2(Q\sin\theta,Q\cos\theta)$$
$$+\frac{Q^2}{\pi\hbar^2 u}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\int_0^{\pi}d\theta\sin\theta\exp\left[-\frac{\xi^2Q^2\sin^2\theta}{2}\right]$$
$$\times[1-J_0(Q|\mathbf{R}_a-\mathbf{R}_b|\sin\theta)]\Xi_D^2(Q\sin\theta,Q\cos\theta) , \tag{6.5}$$

where the magnitude of phonon wave vector $\mathbf{Q}$ is denoted simply by Q and is given by $|E_a-E_b|/\hbar u$ with the sound velocity u of the longitudinal-acoustic phonon mode. Similarly, in the case of exponential localization, one has

$$T_{\text{DF}}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)=\frac{Q^2}{(2\pi)^2\hbar^2 u}\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta K^2(\mathbf{Q}_{\parallel},\mathbf{R}_a-\mathbf{R}_b)\Xi_D^2(Q\sin\theta,Q\cos\theta)$$
$$+\frac{Q^2}{\pi\hbar^2 u}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\int_0^{\pi}d\theta\sin\theta I^2(Q\sin\theta)[1-J_0(Q|\mathbf{R}_a-\mathbf{R}_b|\sin\theta)]$$
$$\times\Xi_D^2(Q\sin\theta,Q\cos\theta) , \tag{6.6}$$

where

$$I(|\mathbf{Q}_{\parallel}|)=\frac{8\xi^2}{\pi}\int d^2K_{\parallel}\frac{1}{[1+(2\xi|\mathbf{K}_{\parallel}-\mathbf{Q}_{\parallel}/2|)^2]^{3/2}[1+(2\xi|\mathbf{K}_{\parallel}+\mathbf{Q}_{\parallel}/2|)^2]^{3/2}} , \tag{6.7}$$

$$K(\mathbf{Q}_{||},\mathbf{R}_a-\mathbf{R}_b)=\frac{8\xi^2}{\pi}\int d^2K_{||}\frac{\exp[i\mathbf{K}_{||}\cdot(\mathbf{R}_b-\mathbf{R}_a)]}{[1+(2\xi\,|\,\mathbf{K}_{||}-\mathbf{Q}_{||}/2\,|\,)^2]^{3/2}[1+(2\xi\,|\,\mathbf{K}_{||}+\mathbf{Q}_{||}/2\,|\,)^2]^{3/2}}\,. \tag{6.8}$$

Here, I is a function of only the magnitude of $\mathbf{Q}_{||}$, while the function K is dependent also on the angle between $\mathbf{Q}_{||}$ and $\mathbf{R}_a-\mathbf{R}_b$. These are the complete expressions of the exciton transfer rate for the case of deformation-potential coupling. In practice, however, the calculation of the angular integrals in (6.5) and (6.6) is rather cumbersome, and it is desirable to simplify these expressions by introducing a reasonable approximation. It is confirmed numerically that the function $\Xi_D(|\mathbf{Q}_{||}|,Q_z)$ is a slowly varying function with respect to $|\mathbf{Q}_{||}|$ and Q_z for a physically important range of parameters, where $|\mathbf{Q}_{||}|, |Q_z| \leq 10^6\ \mathrm{cm}^{-1}$ and $L_z \leq 100$ Å. Thus one may safely set $\theta=\pi/2$ in $\Xi_D(Q\sin\theta,Q\cos\theta)$ and put it outside the integral and further replace the integral over θ by π times the arithmetic mean of the values of the integrand at $\theta=0$ and $\theta=\pi/2$. Then the transition probability in (6.5) can be approximated as

$$T_{\mathrm{DF}}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)\cong\frac{Q^2}{4\hbar^2u}\exp\left[-\frac{|\mathbf{R}_a-\mathbf{R}_b|^2}{2\xi^2}-\frac{\xi^2Q^2}{2}\right]\Xi_D^2(Q,0)$$
$$+\frac{Q^2}{2\hbar^2u}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\exp\left[-\frac{\xi^2Q^2}{2}\right][1-J_0(Q\,|\,\mathbf{R}_a-\mathbf{R}_b\,|\,)]\Xi_D^2(Q,0)\,. \tag{6.9}$$

With use of relation (2.35), $\Xi_D(Q,0)$ can be written as

$$\Xi_D(Q,0)=\left[\frac{\hbar Q}{2\rho u}\right]^{1/2}\left[\frac{D_cI((4+b_h^2)^{1/2}\beta L_z)}{[1+(b_h/2)^2]^{3/2}}-\frac{D_vI((4+b_e^2)^{1/2}\beta L_z)}{[1+(b_e/2)^2]^{3/2}}\right]\Big/I(2\beta L_z)\,. \tag{6.10}$$

Noting that $b_e\cong b_h\leq 1$ for the wave vector $|\mathbf{Q}|\leq 10^6\ \mathrm{cm}^{-1}$, it can be approximated as

$$\Xi_D(Q,0)\cong\left[\frac{\hbar Q}{2\rho u}\right]^{1/2}\left[\frac{D_c}{[1+(b_h/2)^2]^{3/2}}-\frac{D_v}{[1+(b_e/2)^2]^{3/2}}\right]. \tag{6.11}$$

In a similar way, one can simplify (6.6) for the case of exponential localization. As seen from (6.8), the function $K^2(\mathbf{Q}_{||},\mathbf{R})$ is sensitively dependent on the angle between $\mathbf{Q}_{||}$ and $\mathbf{R}$. However, when integrated spatially over $\mathbf{R}$, the angular dependence vanishes and the result depends weakly on $|\mathbf{Q}_{||}|$. In the rate equation for the exciton distribution function, as will be shown later, the spatial integral of K^2 is physically relevant. Thus, in anticipation of their later use in the rate equation, the θ and ϕ integrations in (6.6) can be simplified as

$$T_{\mathrm{DF}}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)\cong\frac{Q^2}{4\hbar^2u}K^2(\mathbf{Q},\mathbf{R}_a-\mathbf{R}_b)\Xi_D^2(Q,0)$$
$$+\frac{Q^2}{2\hbar^2u}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}I^2(Q)[1-J_0(Q\,|\,\mathbf{R}_a-\mathbf{R}_b\,|\,)]\Xi_D^2(Q,0)\,. \tag{6.12}$$

Equations (6.9) and (6.12) with (6.11) are the basic expressions of the exciton transfer rate via the deformation-potential coupling that will be used in the rate equation.

Next, the probability of exciton transfer via the piezoelectric coupling will be calculated. In this case the coupling is highly anisotropic. Let us introduce a piezoelectric coupling function $\Xi_P^{\sigma}(|\mathbf{Q}_{||}|,Q_z)$ defined by

$$\Xi_P^{\sigma}(|\mathbf{Q}_{||}|,Q_z)=\frac{4\pi ee_{14}}{\epsilon_0}\left[\frac{\hbar}{2\rho u_{\sigma}(Q_{||}^2+Q_z^2)^{1/2}}\right]^{1/2}\left[\frac{G((4+b_h^2)^{1/2}\beta L_z,Q_zL_z)}{[1+(b_h/2)^2]^{3/2}}-\frac{G((4+b_e^2)^{1/2}\beta L_z,Q_zL_z)}{[1+(b_e/2)^2]^{3/2}}\right]\Big/I(2\beta L_z)\,, \tag{6.13}$$

where the functions G and I and the variables b_h and b_e are the same as in (2.32) and the suffix σ specifies the longitudinal-acoustic (LA-) or transverse-acoustic (TA-) phonon mode. Then, for the case of Gaussian localization the exciton transfer rate via the piezoelectric coupling is calculated as

$$T_{\rm PZ}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)$$
$$=\frac{2\pi}{\hbar}\sum_{\mathbf{Q}}|\langle\mathbf{R}_b|T|\mathbf{R}_a\rangle_{\mathbf{Q}}|^2\delta(E_a-E_b\pm\hbar\omega_{\mathbf{Q}})$$
$$=\frac{1}{(2\pi\hbar)^2}\exp\left[-\frac{|\mathbf{R}_a-\mathbf{R}_b|^2}{2\xi^2}\right]\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta\exp\left[-\frac{\xi^2Q_\sigma^2\sin^2\theta}{2}\right]$$
$$\times[\Xi_P^\sigma(Q_\sigma\sin\theta,Q_\sigma\cos\theta)]^2A_\sigma^2(\theta,\phi)$$
$$+\frac{1}{(2\pi\hbar)^2}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta\exp\left[-\frac{\xi^2Q_\sigma^2\sin^2\theta}{2}\right]$$
$$\times|e^{-i\mathbf{Q}_\sigma\cdot\mathbf{R}_b}-e^{-i\mathbf{Q}_\sigma\cdot\mathbf{R}_a}|^2[\Xi_P^\sigma(Q_\sigma\sin\theta,Q_\sigma\cos\theta)]^2A_\sigma^2(\theta,\phi)\ , \quad (6.14)$$

where the magnitude of phonon wave vector $\mathbf{Q}_\sigma$ is determined by $|E_a-E_b|/\hbar u_\sigma$ with the sound velocity u_σ for σ (LA or TA) phonon mode, and the anisotropy factor $A_\sigma(\theta,\phi)$ is given as

$$A_{\rm LA}(\theta,\phi)=\tfrac{3}{2}\sin^2\theta\cos\theta\sin2\phi\ , \quad (6.15)$$

$$A_{\rm TA}(\theta,\phi)=\tfrac{1}{8}(\sin\theta-3\sin3\theta)\sin2\phi\ \text{ or }\ \tfrac{1}{2}\sin2\theta\cos2\phi\ , \quad (6.16)$$

where the results are shown for two possibilities of the polarization direction of the TA mode. Formula (6.14) is quite general, but its calculation is rather complicated. Thus it is desirable to simplify the expression by introducing the same kind of approximation as to reduce (6.5) to (6.9). As noted before, the function G in (6.13) is a slowly varying function with respect to $|\mathbf{Q}_{||}|$ and Q_z for the physically important range of parameters, and thus $\Xi_P^\sigma(|\mathbf{Q}_{||}|,Q_z)$ is also such a function. Furthermore, since the main contribution to the transfer rate comes from the range $\xi Q\le 1$, the localization factor $\exp(-\xi^2Q^2\sin^2\theta/2)$ may be regarded as slowly varying with respect to θ. Thus one may suppose that $\theta=\pi/2$ in both Ξ_P^σ and the localization factor and put these factors outside the integral. The piezoelectric coupling function is reduced to

$$\Xi_P^\sigma(Q,0)=\frac{8\pi ee_{14}}{\epsilon_0}\left[\frac{\hbar}{2\rho u_\sigma Q}\right]^{1/2}\left[\frac{I((4+b_h^2)^{1/2}\beta L_z)}{[1+(b_h/2)^2]^{3/2}}-\frac{I((4+b_e^2)^{1/2}\beta L_z)}{[1+(b_e/2)^2]^{3/2}}\right]\Big/I(2\beta L_z)\ , \quad (6.17)$$

with

$$b_h=\alpha_hQ/\alpha\ \text{ and }\ b_e=\alpha_eQ/\alpha\ .$$

By noting that $b_h\cong b_e\le 1$, the piezoelectric coupling function is further simplified to

$$\Xi_P^\sigma(Q,0)\cong\frac{8\pi ee_{14}}{\epsilon_0}\left[\frac{\hbar}{2\rho u_\sigma Q}\right]^{1/2}\left[\frac{1}{[1+(b_h/2)^2]^{3/2}}-\frac{1}{[1+(b_e/2)^2]^{3/2}}\right]\ . \quad (6.18)$$

Then performing the angular integration of the anisotropic factor and the coherence factor, one obtains

$$T_{\rm PZ}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)\cong\frac{1}{(2\pi\hbar)^2}\exp\left[-\frac{|\mathbf{R}_a-\mathbf{R}_b|^2}{2\xi^2}\right]\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}\exp\left[-\frac{\xi^2Q_\sigma^2}{2}\right][\Xi_P^\sigma(Q_\sigma,0)]^2B_\sigma$$
$$+\frac{1}{(2\pi\hbar)^2}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}\exp\left[-\frac{\xi^2Q_\sigma^2}{2}\right][\Xi_P^\sigma(Q_\sigma,0)]^2f_\sigma\ , \quad (6.19)$$

with

$$B_{\rm LA}=12\pi/35,\ \ B_{\rm TA}=16\pi/35\ ,$$
$$f_{\rm LA}=2B_{\rm LA}-\frac{9\pi}{2}\int_0^{\pi}d\theta\sin^5\theta\cos^2\theta[J_0(Q_{\rm LA}R_{ab}\sin\theta)-J_4(Q_{\rm LA}R_{ab}\sin\theta)\cos(4\phi_{ab})]\ , \quad (6.20)$$
$$f_{\rm TA}=2B_{\rm TA}-\frac{\pi}{2}\int_0^{\pi}d\theta\sin^3\theta(\sin^2\theta-2\cos^2\theta)^2[J_0(Q_{\rm TA}R_{ab}\sin\theta)-J_4(Q_{\rm TA}R_{ab}\sin\theta)\cos(4\phi_{ab})]$$
$$-\frac{\pi}{2}\int_0^{\pi}d\theta\sin\theta\sin^22\theta[J_0(Q_{\rm TA}R_{ab}\sin\theta)+J_4(Q_{\rm TA}R_{ab}\sin\theta)\cos(4\phi_{ab})]\ ,$$

where J_0 and J_4 are the zeroth- and fourth-order Bessel function, respectively, and $R_{ab}=|\mathbf{R}_a-\mathbf{R}_b|$ and ϕ_{ab} is the angle between $\mathbf{R}_a-\mathbf{R}_b$ and one of the crystallographic axes in the QW interface plane. These formulas are sufficiently simple to be useful in practical calculation of the exciton transfer rate.

For the sake of completeness, the results for the case of exponential localization will be given. The general formula is given as

$$\begin{aligned}T_{\mathrm{PZ}}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)=&\frac{1}{(2\pi\hbar)^2}\sum_\sigma\frac{Q_\sigma^2}{u_0}\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta K^2(\mathbf{Q}_\sigma,\mathbf{R}_a-\mathbf{R}_b)[\Xi_P^\sigma(Q_\sigma\sin\theta,Q_\sigma\cos\theta)]^2A_\sigma^2(\theta,\phi)\\&+\frac{1}{(2\pi\hbar)^2}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\\&\times\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta I^2(Q_\sigma\sin\theta)\\&\times|e^{-i\mathbf{Q}_\sigma\cdot\mathbf{R}_b}-e^{-i\mathbf{Q}_\sigma\cdot\mathbf{R}_a}|^2[\Xi_P^\sigma(Q_\sigma\sin\theta,Q_\sigma\cos\theta)]^2A_\sigma^2(\theta,\phi)\,.\end{aligned}\tag{6.21}$$

Introducing the same kind of approximation as used in reducing (6.14) to (6.19), one can simplify (6.21) to

$$\begin{aligned}T_{\mathrm{PZ}}(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)\cong&\frac{1}{(2\pi\hbar)^2}\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}K^2(\mathbf{Q}_\sigma,\mathbf{R}_a-\mathbf{R}_b)[\Xi_P^\sigma(\mathbf{Q}_\sigma,0)]^2B_\sigma\\&+\frac{1}{(2\pi\hbar)^2}\frac{J^2(|\mathbf{R}_a-\mathbf{R}_b|)}{|E_a-E_b|^2}\sum_\sigma\frac{Q_\sigma^2}{u_\sigma}I^2(Q_\sigma)[\Xi_P^\sigma(Q_\sigma,0)]^2f_\sigma\,,\end{aligned}\tag{6.22}$$

where B_σ and f_σ are the same as defined in (6.20), and the meaning of $K^2(\mathbf{Q},\mathbf{R})$ is given above (6.12).

In the rate equation for the exciton distribution function, there appears the two-dimensional spatial integration of the transfer probability $T(|E_a-E_b|,|\mathbf{R}_a-\mathbf{R}_b|)$. Let us now calculate the spatial integral assuming a suitable form for the inter-site matrix element $J(R)$. In Sec. V, it is shown that $J(R)$ behaves as

$$J(R)=\tilde{J}/R^3\,,\tag{6.23}$$

like the dipole-dipole interaction in the region where R is larger than both the localization length and the Bohr radius of the two-dimensional exciton. However, it is quite difficult to derive, theoretically, the behavior of $J(R)$ over the entire range of R. Thus a dipole-dipole type transfer will be assumed for $J(R)$, with a lower cutoff at the localization length ξ. As another choice, the tunneling-type transfer given by

$$J(R)=\tilde{J}\exp(-\delta R/2)\tag{6.24}$$

will be examined, where δ^{-1} is the order of the two-dimensional exciton Bohr radius and $\tilde{J}$ is a phenomenological constant. In this case it is not necessary to introduce a cutoff. Since there appear a few types of spatial dependence on $|\mathbf{R}_a-\mathbf{R}_b|$ in the exciton transfer rate, it is sufficient to give the results for typical terms. In the integration the spatial distribution function $F(R)$ of nearest-neighbor sites at a distance R must be included. In the two-dimensional case, $F(R)$ is given by

$$F(R)=\exp(-\pi R^2/\sigma_0)\,,\tag{6.25}$$

where σ_0^{-1} is the areal number density of islandlike structures. Since σ_0 is of the order of 10^{-11} cm^2, $F(R)$ can often be neglected, in comparison with other more rapidly decaying functions. Then the typical spatial integrals are calculated as

$$\int d^2R\exp(-R^2/2\xi^2)=2\pi\xi^2\,,\tag{6.26}$$

$$\int d^2R\,K^2(\mathbf{Q}_{||},\mathbf{R})=256\xi^4\int d^2K_{||}\frac{1}{[1+(2\xi|\mathbf{K}_{||}-\mathbf{Q}_{||}/2|)^2]^3[1+(2\xi|\mathbf{K}_{||}+\mathbf{Q}_{||}/2|)^2]^3}\,.\tag{6.27}$$

For the dipole-dipole type transfer (6.23), one obtains

$$\begin{aligned}\int_\xi^\infty dR\,R\int_0^{2\pi}d\phi J^2(R)F(R)[1-J_0(QR)]=&\int_\xi^\infty dR\,R\int_0^{2\pi}d\phi J^2(R)F(R)[1-J_0(QR)+\cos(4\phi)J_4(QR)]\\=&\frac{2\pi\tilde{J}^2}{\xi^4}\left[\frac{\pi^2\xi^4}{2\sigma_0^2}\Gamma(-2,\pi\xi^2/\sigma_0)-H(Q\xi)\right],\end{aligned}\tag{6.28}$$

with

$$H(x)=x^4\int_x^\infty dz\,J_0(z)\exp(-\pi z^2/\sigma_0Q^2)/z^5\,,\tag{6.29}$$

where $\Gamma(z,p)$ is the incomplete Γ function defined by

$$\Gamma(z,p)=\int_p^\infty dt\, e^{-t}t^{z-1}\ . \tag{6.30}$$

Here, the nearest-neighbor site distribution function $F(R)$ is included explicitly, because both the dipole-dipole type transfer $J(R)$ and the zeroth-order Bessel function $J_0(QR)$ show rather slow spatial decay. Similarly, for the tunneling-type transfer (6.24) the above quantity is calculated as

$$\int d^2R\, J^2(R)[1-J_0(QR)]=\frac{2\pi\tilde{J}^2}{\delta^2}\left[1-\frac{1}{[1+(Q/\delta)^2]^{3/2}}\right], \tag{6.31}$$

where formula (4.17) is used. By combining all of the results, the exciton transfer rate and its spatial integration can be calculated for both the deformation-potential and piezoelectric coupling, for both cases of the Gaussian and exponential localization, and for both cases of the dipole-dipole type and tunneling-type transfer.

Finally, for illustrative purposes, let us calculate the spatially integrated transfer rate for the simplest case of Gaussian localization and tunneling-type transfer. For the deformation-potential coupling, making use of (6.9), (6.11), and (6.31), one obtains

$$\int d^2R\, T_{\rm DF}(E,R)=\frac{\pi Q^3}{2\hbar\rho u_{\rm LA}^2}\left[\frac{\xi^2}{2}+\frac{\tilde{J}^2}{\delta^2E^2}\left[1-\frac{1}{[1+(Q/\delta)^2]^{3/2}}\right]\right]\exp\left[-\frac{\xi^2Q^2}{2}\right]$$
$$\times\left[\frac{D_c}{[1+(\alpha_hQ/2\alpha)^2]^{3/2}}-\frac{D_v}{[1+(\alpha_eQ/2\alpha)^2]^{3/2}}\right]^2. \tag{6.32}$$

Similarly, for the case of piezoelectric coupling, it is calculated as

$$\int d^2R\, T_{\rm PZ}(E,R)=\frac{1}{4\pi\hbar\rho}\left[\frac{8\pi ee_{14}}{\epsilon_0}\right]^2\sum_\sigma\left[\xi^2B_\sigma+\frac{\tilde{J}^2\bar{f}_\sigma}{\delta^2E^2}\right]\frac{Q_\sigma}{u_\sigma^2}\exp\left[-\frac{\xi^2Q_\sigma^2}{2}\right]$$
$$\times\left[\frac{1}{[1+(\alpha_hQ_\sigma/2\alpha)^2]^{3/2}}-\frac{1}{[1+(\alpha_eQ_\sigma/2\alpha)^2]^{3/2}}\right]^2, \tag{6.33}$$

with

$$B_{\rm LA}=12\pi/35,\quad B_{\rm TA}=16\pi/35\ ,$$
$$\bar{f}_{\rm LA}=\frac{9\pi}{2}\int_0^\pi d\theta\sin^5\theta\cos^2\theta\left[1-\frac{1}{[1+(Q_{\rm LA}\sin\theta/\delta)^2]^{3/2}}\right], \tag{6.34}$$
$$\bar{f}_{\rm TA}=\frac{\pi}{2}\int_0^\pi d\theta\sin^3\theta(9\sin^4\theta-16\sin^2\theta+8)\left[1-\frac{1}{[1+(Q_{\rm TA}\sin\theta/\delta)^2]^{3/2}}\right],$$

where the suffix σ indicates the LA- or TA-phonon mode. The material parameters chosen in the numerical calculation are given in Sec. VII. The results are shown in Fig. 1. It is found that the contribution from the piezoelectric coupling is smaller than that from the deformation-potential coupling, but it is not negligible. The energy dependence is similar for both cases of the deformation-potential and piezoelectric coupling. Roughly speaking, the peak position is determined by the localization factor $\exp(-\xi^2Q_\sigma^2/2)$, namely $E\cong\hbar u_\sigma\xi^{-1}$. In fact, this estimate gives the right order of 0.3–0.4 meV. The overall features in Fig. 1 are preserved for other cases of combination of the Gaussian and exponential localization, and the tunneling- and dipole-dipole type transfers.

VII. RATE EQUATION FOR EXCITON DISTRIBUTION FUNCTION AND NONEXPONENTIAL BEHAVIOR OF ENERGY RELAXATION

Now that the transition probability of the exciton transfer is derived, one can set up the rate equation for the exciton distribution function. In the following the interlayer transfer of excitons through the AlAs barrier layer is neglected and only the exciton transfer within a QW will be taken into account. Thus the problem is reduced to that of a single QW. As noted in the Introduction, due to the fluctuation of the well thickness in the lateral direction of a QW, the quasi-two-dimensional excitons can be localized at such islandlike structures. The energy relaxation occurs through the exciton migration in search of lower-energy sites. Now for simplicity an assumption will be introduced that the line broadening is microscopic,[22] i.e., that there is no correlation between the energy of the localized exciton and its position in space. Thus the energy distribution of the localized exciton at any particular site depends only on the overall density of states but not on the nearby configuration of the localized site. In addition, the density of states of localized excitons is assumed to be proportional to the absorption spectrum at low temperatures. This assumption is reasonable because in the low-energy tail of the density of states the contribution from the localized excitons is dominant. Under these assumptions the rate equation for the distribution function $f(E,t)$ of the localized exciton with energy E is derived as

$$\frac{d}{dt}f(E,t)=-\gamma_0 f(E,t)-\sigma_0^{-1}\int d^2R\,F(R)\int dE'\,D(E')f(E,t)\{n(E'-E)\Theta(E'-E)$$
$$+[1+n(E-E')]\Theta(E-E')\}T(|E-E'|,R)$$
$$+\sigma_0^{-1}\int d^2R\,F(R)\int dE'\,D(E')f(E',t)\{n(E-E')\Theta(E-E')$$
$$+[1+n(E'-E)]\Theta(E'-E)\}T(|E-E'|,R)\,, \tag{7.1}$$

where γ_0, σ_0^{-1}, n, T, and Θ are the generally energy-dependent decay rate including both the radiative and nonradiative contributions, the areal number density of islandlike structures, the phonon occupation number, the exciton transfer rate calculated in Sec. VI, and the Heaviside step function, respectively; $F(R)$ is the distribution function of nearest-neighbor sites given by (6.25) and $D(E)$ is the density of states of localized excitons normalized as

$$\int dE\,D(E)=1\,. \tag{7.2}$$

The second and third terms in (7.1) represent the probability leaving and coming into the exciton state with energy E, respectively. For the exciton transfer rate $T(E,R)$, the contributions from both the deformation-potential and piezoelectric coupling are taken into account, namely

$$T(E,R)=T_{\rm DF}(E,R)+T_{\rm PZ}(E,R)\,. \tag{7.3}$$

In the numerical integration of the rate equation (7.1), the following parameters are chosen:

$$\rho=5.3\ \text{g/cm}^3\ \text{(Ref. 23)},\quad u_{\rm LA}=4.81\times10^5\ \text{cm/s (Ref. 20)},\quad u_{\rm TA}=3.34\times10^5\ \text{cm/s (Ref. 13)}\,,$$
$$L_z=76\ \text{Å (Ref. 3)},\quad a_B=136\ \text{Å (Ref. 21)},\quad \xi=150\ \text{Å},\quad \sigma_0=10^{-11}\ \text{cm}^2\,,$$
$$\alpha=10^6\ \text{cm}^{-1}\ \text{(Ref. 11)},\quad D_v=3.1\ \text{eV (Ref. 24)},\quad D_c=-6.5\ \text{eV (Ref. 25)},\quad m_e=0.067m_0\ \text{(Ref. 26)}\,, \tag{7.4}$$
$$m_h=0.45m_0\ \text{(Ref. 26)},\quad e_{14}=1.6\times10^{-5}\ \text{C/cm}^2\ \text{(Ref. 27)},\quad \epsilon_0=12.56\ \text{(Ref. 28)},\quad \gamma_0^{-1}=480\ \text{ps (Ref. 29)}\,,$$

where m_0 is the free-electron mass and γ_0 is determined from the experimental decay curve of the spectrally integrated luminescence intensity[29] and is assumed to be energy independent, for simplicity. The transfer-integral constant $\tilde{J}$ in (6.23) or (6.24) is left as an adjustable parameter. The density of states $D(E)$ is approximated by a Gaussian with a peak at 1.6225 eV and a half-width of 7.5 meV. The initial distribution function is taken from the experimental data of the time-resolved luminescence spectrum. In Fig. 2 the time-resolved luminescence spectrum divided by the absorption spectrum is plotted on the logarithmic scale.[29] This figure can be considered as representing the exciton distribution function $f(E,t)$. Surprisingly enough, the experimental $f(E,t)$ can be described by the Boltzmann distribution fairly well, especially in the higher-energy region $E\geq1.614$ eV. The Boltzmann statistics may be maintained by the frequent collisions among excitons. As for the initial distribution function in the numerical integration of the rate equation, the effective temperature of excitons is assumed to be 20 K above 1.614 eV and -10 K below that energy, respectively, to simulate the experimental distribution function at $t=100$ ps. The calculated average energy of luminescence shows a nonexponential behavior, i.e., decay with an

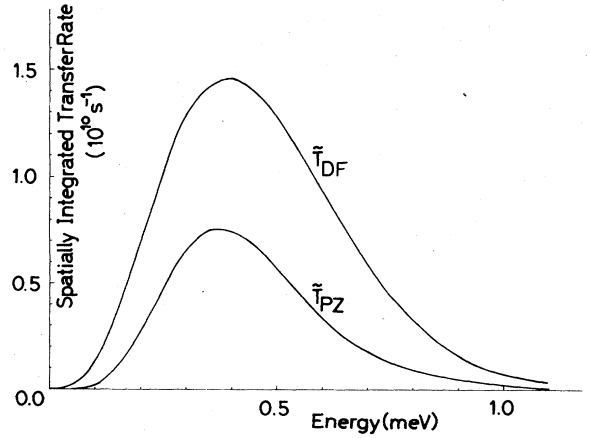

FIG. 1. Energy dependence of the spatially integrated one-phonon-assisted transfer rate of excitons for deformation-potential coupling (DF) and piezoelectric coupling (PZ).

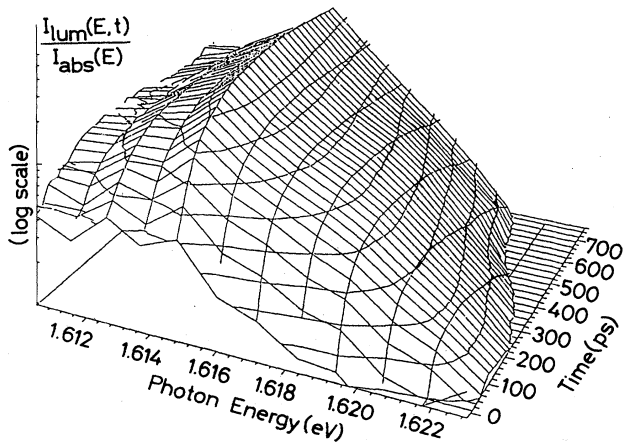

FIG. 2. Experimental data of the time-resolved luminescence spectrum divided by the absorption spectrum which can be considered as representing the exciton distribution function.

almost constant rate. At first, let us assume for $J(R)$ the dipole-dipole type transfer in (6.23). In Fig. 3 the theoretical result for the case of Gaussian localization is compared with the experimental data of Ref. 30. The best fit is obtained by adjusting $\tilde{J}$ as

$$\tilde{J}(\text{Gaussian})=11.7\times10^2\ \text{eV}\,\text{Å}^3\ . \tag{7.5}$$

A similar result is obtained in the case of exponential localization by taking

$$\tilde{J}(\text{exponential})=10.2\times10^2\ \text{eV}\,\text{Å}^3\ . \tag{7.6}$$

In the latter case the magnitude of the wave vector of phonons participating in the exciton transfer, or, equivalently, the possible energy mismatch of excitons, is larger than that in the former case, as seen from (4.15) and (4.16). This leads to the faster energy relaxation in the case of exponential localization than in the case of Gaussian localization for the same localization length ξ, and thus the smaller value of $\tilde{J}$ is obtained in the former case. For the tunneling-type transfer in (6.24), the parameter δ is assumed to be $10^6\ \text{cm}^{-1}$, which is on the order of the inverse Bohr radius of the quasi-two-dimensional exciton. From the fitting to the experimental curve, the phenomenological constant $\tilde{J}$ is determined as 0.44 and 0.31 meV for the case of Gaussian localization and exponential localization, respectively. These values are on the same order as those of the dipole-dipole type transfer in (6.23) estimated at $R\cong\xi$. For the case of dipole-dipole—type transfer, the agreement within a factor of 2 or 3 between the theoretical value in (5.15) and the values estimated from the experiment in (7.5) and (7.6) is quite satisfactory in view of ambiguities in the material parameters. This confirms the adequacy of both our model for the localized excitons in QW heterostructures and our theory of the energy transfer.

Let us now explain the observed nonexponential behavior of energy relaxation. Experimentally, the average energy of luminescence showed decay with an almost constant rate. The average energy of luminescence at time t is defined by

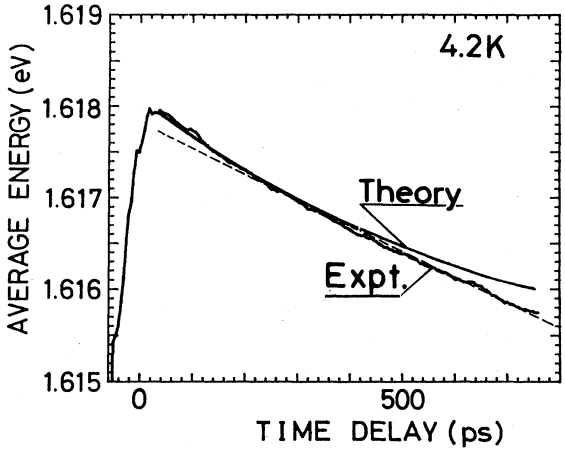

FIG. 3. Comparison of the theoretical calculation of the average energy of luminescence with the experimental data cited from Ref. 30.

$$\langle E\rangle(t)=\int dE\,ED(E)f(E,t)\Big/\int dE\,D(E)f(E,t)\ , \tag{7.7}$$

where $D(E)f(E,t)$ is proportional to the intensity of luminescence with energy E at time t. The energy relaxation rate is then calculated by $d\langle E\rangle/dt$. On the basis of the analytical expression of $d\langle E\rangle/dt$, the nonexponential behavior of energy relaxation will be clarified. To simplify the rate equation (7.1), the spatial integral of the exciton transfer rate will be denoted by $\tilde{T}$, namely

$$\tilde{T}(|E-E'|)=\sigma_0^{-1}\int d^2R\,T(|E-E'|,R)F(R)\ . \tag{7.8}$$

Furthermore, the time dependence due to the radiative and nonradiative decay processes will be separated out by setting

$$f(E,t)=e^{-\gamma_0 t}\tilde{f}(E,t)\ . \tag{7.9}$$

Then the rate equation for $\tilde{f}(E,t)$ can be written as

$$\begin{aligned}\frac{d}{dt}\tilde{f}(E,t)=&-\int dE'\,D(E')\tilde{f}(E,t)\tilde{T}(|E-E'|)\{n(E'-E)\Theta(E'-E)+[1+n(E-E')]\Theta(E-E')\}\\&+\int dE'\,D(E')\tilde{f}(E',t)\tilde{T}(|E-E'|)\{n(E-E')\Theta(E-E')+[1+n(E'-E)]\Theta(E'-E)\}\ .\end{aligned} \tag{7.10}$$

It is easily found that

$$\frac{d}{dt}\int dE\,D(E)\tilde{f}(E,t)=0\ , \tag{7.11}$$

and, correspondingly,

$$\int dE\,D(E)\tilde{f}(E,t)=D_0\ , \tag{7.12}$$

where D_0 is the initial value of the total exciton population. This relation implies simply that the total luminescence intensity or the total exciton population decreases with the decay rate γ_0. Next, by multiplying $ED(E)$ on both sides of (7.10) and by integrating over E, one obtains

$$\frac{d}{dt}\left[\int dE\, E D(E)\tilde{f}(E,t)\right]=\int dE \int dE' D(E)D(E')\tilde{f}(E',t)(E-E')\tilde{T}(|E-E'|) \times\{n(E-E')\Theta(E-E')+[1+n(E'-E)]\Theta(E'-E)\} . \tag{7.13}$$

As shown in Fig. 1, the function $\tilde{T}(E)$ is a rapidly decreasing function compared to the density of states $D(E)$ and the phonon occupation number $n(E)$. This is simply due to the exciton localization factors in (6.5), (6.6), (6.14), and (6.21). Thus it is allowed to expand as

$$D(E)=D(E')+\frac{dD(E')}{dE'}(E-E')+\cdots . \tag{7.14}$$

Substituting this expansion into the integrand on the right-hand side of (7.13) and changing the integration variables from (E,E') to $(E',E-E')$, one finds

$$\frac{d}{dt}\left[\int dE\, E D(E)\tilde{f}(E,t)\right]=-\int dE' D^2(E')\tilde{f}(E',t)\int_0^\infty dE\, E\tilde{T}(|E|) + \int dE' D(E')\frac{dD(E')}{dE'}\tilde{f}(E',t)\int_0^\infty dE\, E^2\tilde{T}(|E|)[1+2n(E)]+\cdots . \tag{7.15}$$

The average energy of luminescence defined by (7.7) can be rewritten as

$$\langle E\rangle(t)=\int dE\, E D(E)\tilde{f}(E,t)\Big/\int dE\, D(E)\tilde{f}(E,t) =D_0^{-1}\int dE\, E D(E)\tilde{f}(E,t) , \tag{7.16}$$

where the equation (7.12) is used. Then the energy relaxation rate is calculated as

$$-\frac{d\langle E\rangle}{dt}=-D_0^{-1}\frac{d}{dt}\left[\int dE\, E D(E)\tilde{f}(E,t)\right] \cong D_0^{-1}\int_0^\infty dE\, E\tilde{T}(|E|)\int dE' D^2(E')\tilde{f}(E',t) , \tag{7.17}$$

where only the first term on the right-hand side of (7.15) is retained. As shown in (7.12), the integral $\int dE\, D(E)\tilde{f}(E,t)$ is exactly time independent. Thus it is expected that the right-hand side of (7.17) is almost time independent, if the functional form of $D^2(E)$ is not very different from that of $D(E)$. This may be the case because at the later stage, after photoexcitation, the excitons have accumulated on the low-energy tail of the density of states. In this energy region, $D(E)$ is a smooth function and one may approximate (7.17) as

$$-\frac{d\langle E\rangle}{dt}=W\int dE' D^2(E')\tilde{f}(E',t) \cong W\bar{D}\int dE' D(E')\tilde{f}(E',t) =\bar{D}\int_0^\infty dE\, E\tilde{T}(E) , \tag{7.18}$$

with

$$W=D_0^{-1}\int_0^\infty dE\, E\tilde{T}(E) ,$$

where $\bar{D}$ is a representative value of $D(E)$ at the peak region of $\tilde{f}(E,t)$. The final expression is exactly time independent. This explains qualitatively the observed nonexponential behavior of energy relaxation. In fact, expression (7.18) gives the right order of the observed energy relaxation rate. The generality of the above argument suggests that the nonexponential behavior of energy relaxation is a universal feature to be observed in the tail region of optical transitions which is induced by any kind of inhomogeneity or disorder.

VIII. DEPENDENCE OF ENERGY RELAXATION RATE ON THE QUANTUM-WELL THICKNESS

Here the dependence of energy relaxation rate on the QW thickness L_z is discussed. Experimentally, the dependence of L_z^{-2} is observed along with the same dependence of the absorption bandwidth, as shown in Fig. 4.[30] This experimental result suggests that there is some kind of correlation between the energy relaxation rate and the absorption bandwidth. In fact, according to (7.18), the L_z dependence of the energy relaxation rate arises from $\bar{D}$

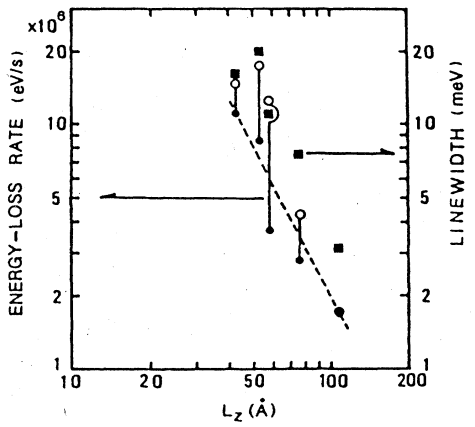

FIG. 4. Observed dependence on the quantum-well thickness L_z of the energy relaxation rate and the absorption bandwidth of the $1s$ exciton. The dashed line shows the L_z^{-2} dependence. The open circles represent the energy relaxation rates at the initial stage after photoexcitation, while the solid circles are those at the later stage.

since the integral factor $\int dE\, E\tilde{T}(E)$ is only weakly dependent on L_z. When the width of the density of states is increased, the value $\bar{D}$ at the low-energy tail becomes larger, since the density of states is normalized by (7.2). Thus the above correlation can be understood qualitatively. To investigate in more detail the relationship between the energy relaxation rate and the absorption bandwidth, one can make full use of the scaling property of the rate equation.

The characteristic width of the density of states of excitons will be denoted by σ, which is identified here with the absorption bandwidth. Then, by expressing the σ dependence explicitly, the rate equation (7.10) can be written as

$$\frac{d}{dt}\tilde{f}(E,\sigma,t)=-\int dE'\,D(E',\sigma)\tilde{f}(E,\sigma,t)\tilde{T}(|E-E'|)\{n(E'-E)\Theta(E'-E)+[1+n(E-E')]\Theta(E-E')\}$$
$$+\int dE'\,D(E',\sigma)\tilde{f}(E',\sigma,t)\tilde{T}(|E-E'|)\{n(E-E')\Theta(E-E')+[1+n(E'-E)]\Theta(E'-E)\}\ , \tag{8.1}$$

where $\tilde{T}(E)$ is defined by (7.8) and is not dependent on σ. Since the density of states $D(E,\sigma)$ is normalized as

$$\int dE\,D(E,\sigma)=1\ , \tag{8.2}$$

the following scaling relation can be postulated,

$$\sigma D(\sigma E,\sigma)=\tilde{D}(E)\ , \tag{8.3}$$

where $\tilde{D}$ is a universal function independent of σ. By scaling the variables (E,E') as $(\sigma E,\sigma E')$ and using (8.3), one finds

$$\frac{d}{dt}\tilde{f}(\sigma E,\sigma,t)=-\int dE'\,\tilde{D}(E')\tilde{f}(\sigma E,\sigma,t)\tilde{T}(\sigma|E-E'|)\{n(\sigma(E'-E))\Theta(E'-E)+[1+n(\sigma(E-E'))]\Theta(E-E')\}$$
$$+\int dE'\,\tilde{D}(E')\tilde{f}(\sigma E',\sigma,t)\tilde{T}(\sigma|E-E'|)\{n(\sigma(E-E'))\Theta(E-E')+[1+n(\sigma(E'-E))]\Theta(E'-E)\}\ . \tag{8.4}$$

Furthermore, the transfer rate $\tilde{T}$ is supposed to be scaled as

$$\tilde{T}(\sigma|E-E'|)=\sigma^{\alpha}U(|E-E'|)\ , \tag{8.5}$$

where U is a universal function that is independent of, or weakly dependent on, the scaling parameter σ. Consequently, (8.4) can be rewritten as

$$\frac{d}{dt}\tilde{f}(\sigma E,\sigma,t)=-\sigma^{\alpha}\left[\int dE'\,K(E,E')\tilde{f}(\sigma E,\sigma,t)-\int dE'\,L(E,E')\tilde{f}(\sigma E',\sigma,t)\right]\ . \tag{8.6}$$

Here, K and L are the integral kernels corresponding to each term on the right-hand side of (8.4) which can be regarded as universal functions since the parameter σ included in the phonon occupation number n gives rise to only a weak dependence on σ. Scaling the time variable as

$$t\rightarrow t/\sigma^{\alpha}\ , \tag{8.7}$$

one finally arrives at the universal rate equation,

$$\frac{d}{dt}\tilde{f}(\sigma E,\sigma,t/\sigma^{\alpha})=-\int dE'\,K(E,E')\tilde{f}(\sigma E,\sigma,t/\sigma^{\alpha})+\int dE'\,L(E,E')\tilde{f}(\sigma E',\sigma,t/\sigma^{\alpha})\ . \tag{8.8}$$

Since the integral kernels K and L are universal functions, the solution of the rate equation (8.8) can also be regarded as universal, namely

$$\tilde{f}(\sigma E,\sigma,t/\sigma^{\alpha})=G(E,t)\ , \tag{8.9}$$

where G is a universal function independent of σ. This is an important consequence of the scaling property of the rate equation.

Now the average energy of luminescence is calculated by

$$\langle E\rangle_{\sigma}=\frac{\int dE\,ED(E,\sigma)\tilde{f}(E,\sigma,t)}{\int dE\,D(E,\sigma)\tilde{f}(E,\sigma,t)}\ . \tag{8.10}$$

Scaling the integration variable E as σE and using the relation (8.3), it is rewritten as

$$\langle E\rangle_{\sigma}=\frac{\sigma\int dE\,E\tilde{D}(E)\tilde{f}(\sigma E,\sigma,t)}{\int dE\,\tilde{D}(E)\tilde{f}(\sigma E,\sigma,t)}=\frac{\sigma\int dE\,E\tilde{D}(E)G(E,\sigma^{\alpha}t)}{\int dE\,\tilde{D}(E)G(E,\sigma^{\alpha}t)}\ , \tag{8.11}$$

where the scaling property (8.9) is used. Then the energy relaxation rate is calculated as

$$\frac{d}{dt}\langle E\rangle_{\sigma}=\sigma^{1+\alpha}\frac{d}{du}\left[\frac{\int dE\,E\tilde{D}(E)G(E,u)}{\int dE\,\tilde{D}(E)G(E,u)}\right]\Bigg|_{u=\sigma^{\alpha}t}\ . \tag{8.12}$$

Here the derivative term with respect to u is a universal function since $\tilde{D}$ and G are universal functions. It can be

expected that the derivative term is a smooth function with respect to u at the large argument, and that it takes an almost constant value independent of σ at large t. Thus the energy relaxation rate can be scaled as

$$\frac{d}{dt}\langle E\rangle_\sigma \propto \sigma^{1+\alpha} , \tag{8.13}$$

in terms of the absorption bandwidth σ, where the exponent α is determined by the scaling property of the transfer rate as shown in (8.5). The exponent α can be estimated, in principle, from the dependence of the transfer rate on the energy or the phonon wave vector. However, as investigated in Sec. VI, the expression of the transfer rate contains these variables in a rather complicated way and the determination of the exponent α is not straightforward. The exponent α should be determined in the energy region in which $\tilde{T}(E)$ takes a significant value and from which the contribution to the exciton transfer process is dominant. For example, in Fig. 1, where $\tilde{T}_{\rm DF}(E)$ and $\tilde{T}_{\rm PZ}(E)$ are shown for the case of Gaussian localization and for tunneling-type transfer, the relevant energy region is considered to be $0.2 \le E \le 0.8$ meV. In this region the exponent α can be considered to be 0 since there is no definite power-law dependence on energy. The same situation holds for other cases of combinations of the Gaussian and exponential localization and the tunneling-type and dipole-dipole—type transfer. Consequently, it can be concluded that $\alpha=0$ and that

$$\frac{d}{dt}\langle E\rangle_\sigma \propto \sigma . \tag{8.14}$$

The scaling argument successfully explains the observed correlation between the energy relaxation rate and the absorption bandwidth, though in a qualitative way. On the other hand, the observed dependence of the absorption bandwidth on the QW thickness L_z is different from the expected one. Since the kinetic energy of the electron and hole confined in a QW is approximately proportional to L_z^{-2},[31] the fluctuation of the exciton energy is proportional to L_z^{-3} if the amount of well-thickness fluctuation is independent of L_z. In reality, however, the bandwidth of excitons is determined by the details of atomic-scale disorder of the QW interface and a naive theory fails to predict the observed dependence on L_z. A detailed theory based on the microscopic morphology of the QW interface will be necessary to explain the L_z^{-2} dependence of the absorption bandwidth.

IX. SUMMARY

The slow energy relaxation of the quasi-two-dimensional excitons in GaAs-AlAs QW heterostructures is explained quantitatively in terms of the intralayer migration of excitons localized at the energetically local minimum sites induced by the fluctuation of the well thickness along the QW interface. The nonexponential behavior of energy relaxation is clarified theoretically as a general feature to be observed in the low-energy tail of the density of states. The dependence of the energy relaxation rate on the QW thickness is discussed along with the same dependence of the absorption bandwidth. The correlation between the energy relaxation rate and the absorption bandwidth is explained qualitatively on the basis of the scaling property of the rate equation.

Finally, let us briefly comment on the energy relaxation of excitons in three dimensions, such as the excitons in mixed semiconductors. The underlying physics is analogous to that in the quasi-two-dimensional case discussed in this paper, although the origin of the inhomogeneity or disorder is different in two cases. In fact, the slow and nonexponential energy relaxation of excitons in the band-edge region was observed recently in mixed semiconductors.[32] This can be regarded as one of the experimental verifications of the general feature of the nonexponential relaxation clarified in Sec. VII.

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APPENDIX

The explicit expressions of $I(\gamma)$ defined by (2.29) and $G(\gamma,\delta)$ defined in (2.23) are given in the following. The argument δ of G is changed to 2δ for simplicity of expression:

$$I(\gamma)=\frac{1}{4\gamma^2(\gamma^2+4\pi^2)^3}[3\gamma^7+28\pi^2\gamma^5+96\pi^4\gamma^3-112\pi^4\gamma^2+128\pi^6\gamma-192\pi^6+16\pi^4 e^{-\gamma}(\gamma^3+7\gamma^2+4\pi^2\gamma+12\pi^2)] , \tag{A1}$$

$$G(\gamma,2\delta)=\int_{-1/2}^{1/2}dz_e\int_{-1/2}^{1/2}dz_h \exp(i\,2\delta z_e-\gamma\,|\,z_e-z_h\,|\,)(1+\gamma\,|\,z_e-z_h\,|\,)\cos^2(\pi z_e)\cos^2(\pi z_h) \tag{A2}$$

$$=\frac{\pi^2}{2\delta}\left[\frac{P(\gamma,\delta;0)\sin\delta+Q(\gamma,\delta;0)\cos\delta}{\pi^2-\delta^2}+\frac{P(\gamma,\delta;\pi)\sin\delta+Q(\gamma,\delta;\pi)\cos\delta}{(\pi+\delta)(4\pi+2\delta)}\right.$$

$$\left.+\frac{P(\gamma,\delta;-\pi)\sin\delta+Q(\gamma,\delta;-\pi)\cos\delta}{(\pi-\delta)(4\pi-2\delta)}\right] , \tag{A3}$$

with

$$P(\gamma,\delta;x)=\frac{1}{(\gamma^2+4x^2)^2[\gamma^2+4(x+\delta)^2]^2}(2\gamma^7+8[x^2+(x+\delta)^2]\gamma^5+16[x^4+(x+\delta)^4]\gamma^3$$
$$-2\delta(2x+\delta)e^{-\gamma}\{\gamma^6+4\gamma^5+4[x^2+(x+\delta)^2]\gamma^4+8[\delta^2+2x(x+\delta)]\gamma^3$$
$$+16x^2(x+\delta)^2\gamma^2\}) , \tag{A4}$$

$$Q(\gamma,\delta;x)=\frac{1}{(\gamma^2+4x^2)^2[\gamma^2+4(x+\delta)^2]^2}$$
$$\times(-3(2x+\delta)\gamma^6-4[(x+\delta)^3+6x(x+\delta)^2+6x^2(x+\delta)+x^3]\gamma^4$$
$$-16[3x(x+\delta)^4+2x^2(x+\delta)^3+2x^3(x+\delta)^2+3x^4(x+\delta)]\gamma^2$$
$$-64x^3(x+\delta)^4-64x^4(x+\delta)^3+(2x+\delta)e^{-\gamma}\{\gamma^7+3\gamma^6+4[x^2+(x+\delta)^2+x(x+\delta)]\gamma^5$$
$$+4[x^2+(x+\delta)^2+5x(x+\delta)]\gamma^4$$
$$+16x(x+\delta)[x^2+(x+\delta)^2+x(x+\delta)]\gamma^3$$
$$+16x(x+\delta)[x^2+(x+\delta)^2+3x(x+\delta)+2\delta^2]\gamma^2$$
$$+64x^3(x+\delta)^3\gamma+64x^3(x+\delta)^3\}) . \tag{A5}$$

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