Energy Transfer of Excitons between Quantum Wells

Separated by a Wide Barrier

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We present a microscopic theory of the excitonic Stokes and anti-Stokes energy transfer mechanisms between two widely separated unequal quantum wells with a large energy mismatch (Δ) at low temperatures (T). Exciton transfer through dipolar coupling, photon-exchange coupling and over-barrier ionization of the excitons through exciton-exciton Auger processes are examined. The energy transfer rate is calculated as a function of T and the center-to-center distance d between the two wells. The rates depend sensitively on T for plane-wave excitons. For localized excitons, the rates depend on T only through the T -dependence of the localization radius.

For Stokes energy transfer, dominant energy transfer occurs through photon-exchange interaction. The rate has a slow dependence on d , yielding reasonable agreement with recent data from GaAs/Al_xGa_{1-x}As quantum wells. The dipolar rate is about an order of magnitude smaller for large d (e.g., $d = 175$ Å) with a stronger range dependence $\propto d^{-4}$. However, the latter can be comparable to the radiative rate for small d (e.g., $d \leq 80$ Å).

For anti-Stokes transfer through exchange-type (e.g., dipolar and photon-exchange) interactions, we show that thermal activation $\propto \exp(-\Delta/k_B T)$ is essential for the transfer contradicting a recent nonactivated result based on the Förster-Dexter theory.

Phonon-assisted transfer yields a negligibly small rate for $\Delta \gg k_B T$. On the other hand, energy transfer through over-barrier ionization of excitons via Auger processes yields a

significantly larger nonactivated rate which is independent of d . This rate is large enough (when the exciton density is not too small) to explain recent data.

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I. INTRODUCTION

Energy transfer of excitons between deep semiconductor quantum wells and quantum dots separated by thick barriers is not only an academically interesting phenomenon but plays a fundamental role in optoelectronic devices based on artificially structured semiconductors such as quantum-well (QW) and quantum-dot lasers and light-emitting diodes. While energy transfer inside a single QW has been extensively studied in the past [1, 2], inter-well energy transfer has received considerably less attention. Recently, surprisingly large low temperature (T) Stokes and anti-Stokes energy transfer rates have been observed by Tomita, Shah and Knox (TSK) between two widely separated GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ QW's using time-resolved photoluminescence excitation (PLE) and PL spectroscopy. [3] For Stokes transfer, the observed transfer rate from a 50-Å QW to a 100-Å QW with an energy mismatch $\Delta = 60$ meV was in the range 3×10^8 - 1×10^9 sec^{-1} for samples with a center-to-center distance d of 175 - 375 Å and at $T = 4$ - 80 K. Anti-Stokes transfer from a wide to a narrow QW proceeded at a much slower rate (10^4 - 10^6 sec^{-1}). [3] These rates (W) were too large and showed too little dependence on d to be explained by a standard carrier tunneling model with $W \propto \exp(-\alpha d)$ (α is a constant) [4] or dipolar coupling with $W \propto d^{-4}$. [3] Surprisingly, samples with larger d showed faster anti-Stokes transfer rates than those with smaller d . [3] For Stokes transfer, however, the observed rates were nearly independent of d . In this paper, we present a theoretical explanation for the principal characteristics of these intriguing data.

The anomalously large energy transfer rates were also observed by Kim et al. and explained by carrier tunneling by assuming large trans-barrier GaAs clusters in the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barrier. [4] However, the statistical rate based on such a model is expected to

decay rapidly with d . In this paper, we present several important intrinsic mechanisms of energy transfer without assuming GaAs clustering in the barrier region. Dipolar coupling, real and virtual photon-exchange coupling and over-barrier ionization of the excitons through exciton-exciton Auger processes are examined.

The dominant Stokes energy transfer occurs through the decay of excitons in the higher-energy QW into free electron-hole pairs in the lower-energy QW, as proposed by TSK. [3] However, we find that photon-exchange interaction yields much faster transfer rates than the dipolar rate investigated by TSK. The radiative rate depends very slowly on d , decaying logarithmically at large d . For plane-wave excitons, this rate decreases rapidly with T . For localized excitons, the rate depends sensitively on the localization radius ξ . It depends on T only through ξ and is independent of T if ξ is insensitive to T at low temperatures. The predicted rates are in reasonable agreement with TSK's data. Photon exchange is recognized as a viable energy transfer mechanism for paramagnetic impurities in insulators. [5] Dipolar coupling yields rates smaller at least by an order of magnitude for $d \geq 175 \text{ \AA}$ and with a stronger d^{-4} -dependence. However, the dipolar rate can be comparable to the radiative rate at a short distance (e.e.g, $d < 80 \text{ \AA}$). The calculated rate vanishes at $T = 0$, reaches a maximum and then decays at higher T . This T -dependence of the dipolar Stokes energy transfer rate for plane-wave excitons conflicts with the recent result of TSK's dipolar rate, which is finite at 0 K and decreases monotonically as a function of T . The physical significance of this discrepancy will be discussed later.

For anti-Stokes transfer through exchange-type (e.g., dipolar and photon-exchange) coupling, we show rigorously that thermal activation $\propto \exp(-\Delta/k_B T)$ is necessary, in general, to overcome the energy mismatch $\epsilon_{2K_{\parallel}} - \epsilon_{1K_{\parallel}} = \Delta$ between the initial (QW1) and final

(QW2) QW's contradicting the TSK's nonactivated rates based on the Förster-Dexter theory. [6, 7] Here $\mathbf{K}_{\parallel}$ is the wave vector for the center-of-mass motion. The phonon-assisted rate is calculated for photon-exchange and dipolar transfer, yielding a negligibly small rate and an activated T -dependence $\propto \exp(-\Delta/k_B T)$. On the other hand, energy transfer through over-barrier ionization of the excitons via Auger processes is shown to yield a significantly larger nonactivated rate which is independent of d . This rate is large enough (when the exciton density is not too small) to explain the recent data. The rate decreases rapidly with increasing exciton localization radius. Larger rates observed by TSK for a sample with larger $d = 375 \text{ \AA}$ as compared to one with $d = 275 \text{ \AA}$ can be explained if one assumes a shorter exciton localization radius (e.g., rougher interfaces) or a larger exciton density for the 375- $\text{\AA}$ sample.

This paper is organized as follows. In Sec. II, we present a basic formalism and give the wave functions of the plane-wave and localized excitons and free electron-hole pairs. The Stokes energy transfer rate is calculated in Sec. III using dipolar and photon-exchange (i.e., radiative) interactions. In Sec. IV, we establish a useful field-theoretic formalism for exciton transfer by expressing the rate in terms of a correlation function, which is then evaluated using a standard diagram expansion technique. We show here that anti-Stokes transfer through a trans-barrier exchange mechanism such as dipole-dipole or photon-exchange coupling is always activated for large Δ ($\gg$ level widths). Anti-Stokes transfer rate is studied in Sec. V in terms of over-barrier ionization of the excitons through exciton-exciton Auger processes and also using dipolar and radiative phonon-assisted processes. The paper is summarized in Sec. VI with brief discussions.

II. BASIC FORMALISM

In this section, a basic exciton formalism is presented. Partially employing the notations of Takagahara [1], the quasi-two-dimensional ground (1s) exciton state with a wave vector $\mathbf{K}_{\parallel}$ for the in-plane center-of-mass motion in the j -th QW is represented as

$$|j, \mathbf{K}_{\parallel}\rangle = \frac{\nu_0}{L} \sum_{\mathbf{r}_e, \mathbf{r}_h} e^{i\mathbf{K}_{\parallel} \cdot \mathbf{R}_{\parallel}} F_j(\mathbf{r}_{e\parallel}, \mathbf{r}_{h\parallel}, z_e - z_j, z_h - z_j) a_{c\mathbf{r}_e}^{\dagger} a_{v\mathbf{r}_h} |0\rangle, \quad (2.1)$$

where $\Omega = L^3 = N\nu_0$ is the sample volume, ν_0 is the unit cell volume, N is the total number of the unit cells, $|0\rangle$ signifies the ground state with an empty conduction band (c) and a filled valence band (v), and z_j is the z -coordinate at the center of the QW. The envelop function F_j depends on the depths and the widths of the QW. The creation and destruction operators $a_{c\mathbf{r}_e}^{\dagger}$ and $a_{c\mathbf{r}_e}$ ($a_{v\mathbf{r}_h}^{\dagger}$ and $a_{v\mathbf{r}_h}$) creates and destroys an electron in the conduction (valence) band at the position $\mathbf{r}_e$ ($\mathbf{r}_h$) in the Wannier representation. The quantity $\mathbf{R}$ is the position vector of the center of mass defined by

$$\mathbf{R} = \alpha_e \mathbf{r}_e + \alpha_h \mathbf{r}_h; \quad \alpha_e = m_e / M, \quad \alpha_h = m_h / M, \quad (2.2)$$

where $M = m_e + m_h$ is the sum of the electron (m_e) and hole (m_h) mass. In the following, a vector $\mathbf{q} = (q_{\parallel}, q_z)$ is decomposed into components parallel ($q_{\parallel}$) and perpendicular (q_z) to the QW plane. We define $q = |\mathbf{q}|$ and $q_{\parallel} = |\mathbf{q}_{\parallel}|$ as the absolute magnitudes of the vectors.

Converting the $\mathbf{r}_{\sigma}$ -summation into integration

$$\sum_{\mathbf{r}_{\sigma}} \rightarrow \frac{1}{\nu_0} \int d^3 r_{\sigma}, \quad \sigma = e, h, \quad (2.3)$$

we obtain the normalization condition

$$\langle j', \mathbf{K}_{||} | j, \mathbf{K}_{||} \rangle = \delta_{j,j'} \delta_{\mathbf{K}_{||}, \mathbf{K}_{||}'} \quad (2.4a)$$

$$\int d^2 r_{||} \int dz_e \int dz_h |F_j(\mathbf{r}_{||}, z_e, z_h)|^2 = 1, \quad (2.4b)$$

where $\mathbf{r}_{||} = \mathbf{r}_{e||} - \mathbf{r}_{h||}$ and the overlap of the confinement wave functions between the two QW's is neglected.

A localized exciton at site $\mathbf{R}_a$ in the j -th QW is represented by

$$|j, \mathbf{R}_a\rangle = \nu_0 \sum_{\mathbf{r}_e, \mathbf{r}_h} G(\mathbf{R}_{||} - \mathbf{R}_a) F_j(\mathbf{r}_{e||} - \mathbf{r}_{h||}, z_e - z_j, z_h - z_j) a_{c\mathbf{r}_e}^\dagger a_{v\mathbf{r}_h} |0\rangle, \quad (2.5)$$

where the normalized center-of-mass wave function $G(\mathbf{R}_{||})$ is approximated by a Gaussian function with a localization radius ξ :

$$G(\mathbf{R}_{||}) = \frac{1}{\xi \sqrt{\pi}} \exp(-R_{||}^2 / \xi^2). \quad (2.6)$$

The normalization condition in Eq. (2.4b) is still valid. Eqs. (2.1), (2.5) and the normalization conditions yield:

$$\langle j, \mathbf{K}_{||} | j', \mathbf{R}_a \rangle = \frac{2\sqrt{\pi}\xi}{L} \exp(-i\mathbf{K}_{||} \cdot \mathbf{R}_a - \frac{1}{2}\xi^2 K_{||}^2) \delta_{j,j'}. \quad (2.7)$$

To obtain a numerical estimate of the transfer rate, we use, for the envelop function $F_j(\mathbf{r}_{||}, z_e, z_h)$, a product of two-dimensional exciton radial wave function $F(\mathbf{r}_{||})$ and quasi-two-dimensional confinement function $F_j(z_e, z_h)$:

$$F_j(\mathbf{r}_{||}, z_e, z_h) = F(\mathbf{r}_{||}) F_j(z_e, z_h), \quad (2.8a)$$

where

$$F(r_{\parallel}) = \frac{2\sqrt{2}}{\sqrt{\pi}a_B} e^{-2r_{\parallel}/a_B}, \quad (2.8b)$$

$$F_j(z_e, z_h) = \phi_{ej}(z_e)\phi_{hj}(z_h), \quad (2.8c)$$

and $a_B = \kappa\hbar^2/(\mu e^2)$ is the bulk exciton Bohr radius. Here μ is the reduced mass and κ is the average dielectric constant. The functions $F(r_{\parallel})$, $\phi_{ej}(z)$ and $\phi_{hj}(z)$ are normalized. The approximation in Eq. (2.8b) is employed for an order of magnitude estimate of the rates. This approximation is adequate for narrow QW's. For wide QW's, the result can be improved by employing variational expressions for $F(r_{\parallel}, z_e, z_h)$. [8, 1, 9]

A free electron-hole pair moving with wave vectors $\mathbf{k}_{e\parallel}$, $\mathbf{k}_{h\parallel}$ with the center-of-mass motion wave vector $\mathbf{K}_{\parallel}$ and the relative wave vector $\mathbf{k}_{\parallel}$ is represented as

$$|j, \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}\rangle = \frac{v_0}{L^2} \sum_{\mathbf{r}_e, \mathbf{r}_h} e^{i\mathbf{K}_{\parallel} \cdot \mathbf{R}_{\parallel}} e^{-i\mathbf{k}_{\parallel} \cdot (\mathbf{r}_{e\parallel} - \mathbf{r}_{h\parallel})} F_j(z_e - z_j, z_h - z_j) a_{c\mathbf{r}_e}^{\dagger} a_{\mathbf{r}_h} |0\rangle, \quad (2.9)$$

where $F_j(z_e, z_h)$ is given in Eq. (2.8c), $\mathbf{k}_{e\parallel} = \alpha_e \mathbf{K}_{\parallel} - \mathbf{k}_{\parallel}$ and $\mathbf{k}_{h\parallel} = \alpha_h \mathbf{K}_{\parallel} + \mathbf{k}_{\parallel}$. The total kinetic energy is the same in either representation: The wave vectors satisfy $(\hbar^2/2)(k_{e\parallel}^2/m_e + k_{h\parallel}^2/m_h) = (\hbar^2/2)(K_{\parallel}^2/M + k_{\parallel}^2/\mu)$. Eqs. (2.1) and (2.9) yield

$$\langle j, \mathbf{K}_{\parallel} | j', \mathbf{K}'_{\parallel}, \mathbf{k}'_{\parallel} \rangle = \frac{1}{L} \delta_{\mathbf{K}_{\parallel}, \mathbf{K}'_{\parallel}} \delta_{j, j'} \int d^2 r_{\parallel} \int dz_e \int dz_h e^{-i\mathbf{k}'_{\parallel} \cdot \mathbf{r}_{\parallel}} F_j^*(r_{\parallel}, z_e, z_h) F_j(z_e, z_h). \quad (2.10)$$

In the quasi-two-dimensional approximation in Eq. (2.8a), the overlap in Eq. (2.10)

reduces to

$$\langle j, \mathbf{K}_{\parallel} | j', \mathbf{K}'_{\parallel}, \mathbf{k}_{\parallel} \rangle = \frac{1}{L} \delta_{\mathbf{K}_{\parallel}, \mathbf{K}'_{\parallel}} \delta_{j, j'} \int d^2 r_{\parallel} e^{-i \mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel}} F^*(\mathbf{r}_{\parallel}) = \frac{8\sqrt{2\pi} a_B \delta_{j, j'}}{L(k_{\parallel}^2 a_B^2 + 4)^{3/2}} \delta_{\mathbf{K}_{\parallel}, \mathbf{K}'_{\parallel}}. \quad (2.11)$$

The second equality is obtained by turning the angular integration into the zeroth-order Bessel function $J_0(k_{\parallel}' r_{\parallel})$.

III. STOKES ENERGY TRANSFER

In this section, we study the Stokes energy transfer rate through dipolar and photon - exchange coupling and compare their relative importance and also with the observed data of TSK. The dependence of the rate on the well separation, temperature and the localization radius is studied. In this case, the energy transfer occurs from narrow QW1 to wide QW2 with the ground sublevel of QW2 lying below that of QW1 by an amount Δ . Experimentally, observed exciton transfer rates are in the range $10^8 - 10^9 \text{ sec}^{-1}$ for GaAs/Al_{0.3}Ga_{0.7}As QW's separated by $d = 175 - 375 \text{ \AA}$, with $\Delta = 60 \text{ meV}$, $b_1 = 50$ and $b_2 = 100 \text{ \AA}$ as mentioned earlier. [3]

A. Dipolar Energy Transfer

A1. Dipolar coupling between the wells

Dipolar coupling arises from the electron-electron interaction Hamiltonian

$$H_{ee} = \frac{1}{2} \int d^3 r \int d^3 r' \psi^\dagger(\mathbf{r}) \psi^\dagger(\mathbf{r}') \frac{e^2}{\kappa |\mathbf{r}' - \mathbf{r}|} \psi(\mathbf{r}') \psi(\mathbf{r}), \quad (3.1)$$

where $\psi(\mathbf{r})$ is the field operator defined by

$$\psi(\mathbf{r}) = \sum_{j, \mathbf{r}_e} \phi_{j\mathbf{r}_e}(\mathbf{r}) a_{c\mathbf{r}_e} + \sum_{j, \mathbf{r}_h} \phi_{j\mathbf{r}_h}(\mathbf{r}) a_{v\mathbf{r}_h}, \quad (3.2)$$

and $\phi_{j\mathbf{r}_\gamma}(\mathbf{r})$ ($\gamma = e, h$) is the Wannier function at the cell $\mathbf{r}_\gamma$ in the j -th QW. Suppressing the ground-state label $|j', 0\rangle$ from $|j, \mathbf{K}_\parallel; j', 0\rangle = |j, \mathbf{K}_\parallel\rangle$, employing Eqs. (2.1), (3.1) and (3.2), the matrix element of H_{ee} between the exciton states in QW1 and QW2 is derived extending Takagahara's result [1]:

$$\langle 2, \mathbf{K}_\parallel' | H_{ee} | 1, \mathbf{K}_\parallel \rangle = \delta_{\mathbf{K}_\parallel, \mathbf{K}_\parallel'} \int dz \int dz' C(\mathbf{K}_\parallel, R_z) F_2^*(0, z', z') F_1(0, z, z), \quad (3.3a)$$

$$C(\mathbf{K}_\parallel, R_z) = \int d^2 R_\parallel e^{i\mathbf{K}_\parallel \cdot \mathbf{R}_\parallel} C(\mathbf{R}_\parallel, R_z), \quad (3.3b)$$

where $R_z = z - z' - d$ with $|z|, |z'| \ll d$ and

$$C(\mathbf{R}_\parallel, R_z) = \int d^3 r \int d^3 r' \frac{e^2}{\kappa |\mathbf{r} - \mathbf{r}' + \mathbf{R}|} \phi_{1v}^*(\mathbf{r}) \phi_{1c}(\mathbf{r}) \phi_{2c}^*(\mathbf{r}') \phi_{2v}(\mathbf{r}'). \quad (3.4)$$

Here, the position vectors $\mathbf{r}$ and $\mathbf{r}'$ are with respect to the centers of the cell at $\mathbf{r}_e \equiv \mathbf{r}_h \equiv 0$ and $\mathbf{R} = (\mathbf{R}_\parallel, R_z)$. A two-dimensional version of Eq. (3.4) was also studied by TSK. [3]

Expanding the integrand in Eq. (3.4) in $\mathbf{r}$ and $\mathbf{r}'$, we obtain

$$C(\mathbf{R}_\parallel, R_z) = \frac{e^2}{\kappa R^3} [\mathbf{D}_1 \cdot \mathbf{D}_2 - 3(\hat{\mathbf{R}} \cdot \mathbf{D}_1)(\hat{\mathbf{R}} \cdot \mathbf{D}_2)], \quad (3.5)$$

where $\hat{\mathbf{R}} = \mathbf{R} / R$ and

$$\mathbf{D}_j = \int d^3 r \phi_{jv}^*(\mathbf{r}) \mathbf{r} \phi_{jc}(\mathbf{r}). \quad (3.6)$$

The Hamiltonian in Eq. (3.3a) allows an exciton in QW1 to be annihilated, exciting an exciton in QW2. A free electron-hole pair state $|2, \mathbf{K}_\parallel', \mathbf{k}_\parallel'\rangle$ can be created in QW2 instead of an exciton. The Hamiltonian for this process is obtained by replacing $F_2(\mathbf{r}_\parallel, z_e, z_h) \rightarrow$

$L^{-1} \exp(-i\mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel}) F_2(z_e, z_h)$ in Eq. (3.3a), yielding

$$\langle 2, \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel} | H_{ee} | 1, \mathbf{K}_{\parallel} \rangle = L^{-1} \delta_{\mathbf{K}_{\parallel}, \mathbf{K}_{\parallel}'} \int dz \int dz' C(\mathbf{K}_{\parallel}, R_z) F_2^*(z', z') F_1(0, z, z). \quad (3.7)$$

The quantity $C(\mathbf{K}_{\parallel}, R_z)$ is the $\mathbf{K}_{\parallel}$ -component of the dipolar energy of a single dipole $\mathbf{D}_1$ in QW1 interacting with a sheet of uniform continuum dipoles $\mathbf{D}_2$ in QW2 at a perpendicular distance R_z and equals

$$C(\mathbf{K}_{\parallel}, R_z) = (\pi D_1 D_2 e^2 / \kappa) K_{\parallel} e^{-K_{\parallel} |R_z|} [\cos(2\phi_{\mathbf{K}_{\parallel}} - \phi_D) + \cos \phi_D], \quad (3.8)$$

where $\phi_{\mathbf{K}_{\parallel}}$ is the angle between $\mathbf{K}_{\parallel}$ and $\mathbf{D}_1$ and ϕ_D is the angle between the two vectors $\mathbf{D}_1$ and $\mathbf{D}_2$ in the QW plane. Note that the expression in Eq. (3.8) vanishes in the limit $K_{\parallel} \rightarrow 0$ contradicting the result of TSK. The vanishing of $C(\mathbf{K}_{\parallel}, R_z)$ for $K_{\parallel} = 0$ means that the interaction energy of a single dipole with a sheet (or line) of uniform dipoles vanishes as well known. [10] The derivation of Eq. (3.8) is given in Appendix A. [11]

Employing the quasi-two-dimensional approximation introduced in Eq. (2.8a) and inserting Eq. (3.8) in Eqs. (3.3a) and (3.7), we obtain

$$\langle 2, \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel} | H_{ee} | 1, \mathbf{K}_{\parallel} \rangle = \frac{8D_1 D_2 e^2}{\kappa a_B^2} [\cos(2\phi_{\mathbf{K}_{\parallel}} - \phi_D) + \cos \phi_D] \delta_{\mathbf{K}_{\parallel}, \mathbf{K}_{\parallel}'} K_{\parallel} \langle e^{-K_{\parallel} |R_z|} \rangle, \quad (3.9a)$$

$$\langle e^{-K_{\parallel} |R_z|} \rangle = \int dz \int dz' F_2^*(z', z') F_1(z, z) e^{-K_{\parallel} |z - z' - d|}, \quad (3.9b)$$

and

$$\langle 2, \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel} | H_{ee} | 1, \mathbf{K}_{\parallel} \rangle = \frac{2\sqrt{2\pi} D_1 D_2 e^2}{a_B \kappa L} [\cos(2\phi_{\mathbf{K}_{\parallel}} - \phi_D) + \cos \phi_D] \delta_{\mathbf{K}_{\parallel}, \mathbf{K}_{\parallel}'} K_{\parallel} \langle e^{-K_{\parallel} |R_z|} \rangle. \quad (3.10)$$

The quantity $\langle \exp(-K_{\parallel} |R_z|) \rangle$ defined in Eq. (3.9b) is the average over the electron and hole

distribution in the QW's. For an order of magnitude estimate, a rectangular distribution over the well widths b_1 and b_2 is employed, yielding

$$\langle e^{-K_{\parallel}|R_z|} \rangle = e^{-t} S(t), \quad S(t) = \frac{\sinh(tb_1/2d)}{tb_1/2d} \frac{\sinh(tb_2/2d)}{tb_2/2d}, \quad (3.11)$$

where $t = K_{\parallel}d$.

The angular factors in Eqs. (3.9a) and (3.10) are to be squared and averaged over $\phi_{\mathbf{K}_{\parallel}}$ for the transfer rate, yielding

$$\frac{1}{2\pi} \int_0^{2\pi} (\cos(2\phi_{\mathbf{K}_{\parallel}} - \phi_D) + \cos\phi_D)^2 d\phi_{\mathbf{K}_{\parallel}} + (\phi_D \rightarrow \phi_D + \frac{\pi}{2}) = 2, \quad (3.12)$$

where the second term is the contribution from the perpendicular polarization of $\mathbf{D}_2$. [3] In the following, we calculate the dipolar exciton energy transfer rate.

A2. Direct energy transfer from localized to plane-wave exciton states

A direct transition from $|1, \mathbf{K}_{\parallel}\rangle$ to $|2, \mathbf{K}_{\parallel}\rangle$ is impossible because the momentum ($\mathbf{K}_{\parallel}' = \mathbf{K}_{\parallel}$) and the energy cannot be conserved simultaneously (i.e., $E_{1\mathbf{K}_{\parallel}} = E_{2\mathbf{K}_{\parallel}} + \Delta$). However, momentum conservation can be relaxed for localized initial exciton states, yielding

$$W = \frac{2\pi}{\hbar} \sum_{\mathbf{K}_{\parallel}'} |\langle 2, \mathbf{K}_{\parallel}' | H_{ee} | 1, \mathbf{R}_a \rangle|^2 \delta(E_{\mathbf{K}_{\parallel}'} - \Delta), \quad (3.13a)$$

where $E_{\mathbf{K}_{\parallel}} = \hbar^2 K_{\parallel}^2 / 2M^2$. Here we ignore the energy distribution of the localized excitons. The energy of localized excitons with the localization radius ξ is of the order of $\hbar^2 / 2\xi^2 M^2 \approx 1.8 \text{ meV} \ll \Delta$ for $\xi = 100 \text{ \AA}$ and is neglected. This rate is evaluated using Eqs. (2.7) and

(3.9) and (3.12) and equals

$$W = (K_{\Delta}\xi)^2 W_0 e^{-(K_{\Delta}\xi)^2} \langle e^{-K_{\Delta}|R_z|} \rangle^2, \quad (3.13b)$$

where $W_0 = 512\pi M(D_1 D_2 e^2 / \kappa a_B^2)^2 / \hbar^3 = 2.14 \times 10^{11} \text{ sec}^{-1}$ for $m_e = 0.067$, $m_h = 0.14$, $M = 0.207$ (in units of free electron mass m_0), $\kappa = 12.4$, $a_B = 144.5 \text{ \AA}$ and $D_1 = D_2 = 5.5 \text{ \AA}$ [3]. These parameters are summarized in Table I. In Eq. (3.13b), $(\hbar K_{\Delta})^2 / 2M = \Delta$, yielding $K_{\Delta} = 1/17.5 \text{ \AA}$ for $\Delta = 60 \text{ meV}$. Therefore, the quantity $\langle \exp(-K_{\Delta}|R_z|) \rangle^2$ is of the order of $\exp(-2dK_{\Delta}) = e^{-20} = 2.06 \times 10^{-9}$ for $d = 175 \text{ \AA}$. Therefore, the rate in Eq. (3.13b) is negligibly small for all ξ for large d and Δ .

A3. Energy transfer from plane-wave excitons to free electron-hole pairs

We now calculate the rate where an exciton $|1, \mathbf{K}_{\parallel}\rangle$ is annihilated in QW1 creating a free electron hole pair $|2, \mathbf{K}_{\parallel}'\rangle$ in QW2. This rate equals

$$W = \frac{2\pi}{\hbar} \beta \int dE_{\mathbf{K}_{\parallel}} e^{-\beta E_{\mathbf{K}_{\parallel}}} \langle \sum_{\mathbf{k}_{\parallel}, \mathbf{K}_{\parallel}'} |\langle 2, \mathbf{K}_{\parallel}', \mathbf{k}_{\parallel} | H_{ee} | 1, \mathbf{K}_{\parallel} \rangle|^2 \delta(\epsilon_{\mathbf{k}_{\parallel}'} - \Delta') \rangle_{\mathbf{K}_{\parallel}}, \quad (3.14)$$

where $\epsilon_{\mathbf{k}_{\parallel}} = \hbar^2 k_{\parallel}^2 / 2\mu$, $\beta = 1/k_B T$, $\Delta' = \Delta - E_B (> 0)$, E_B is the exciton binding energy and $\beta \int dE e^{-\beta E}$ is the Boltzmann average over the initial states. The angular brackets denote the angular average over the direction of $\mathbf{K}_{\parallel}$. In this process, the center-of-mass energy is conserved. The extra energy Δ' is dissipated into the relative motion of the electron and hole.

Inserting Eqs. (3.10) - (3.12) in Eq. (3.14), carrying out the $\mathbf{k}_{\parallel}$ '-summation, we find

$$W = W_{o,dip}(\xi_T) g\left(\frac{d}{\xi_T}\right), \quad g(t) = \int_0^{\infty} x^3 e^{-x^2} e^{-2tx} S(tx)^2 dx, \quad (3.15a)$$

$$W_{o,dip}(\xi_T) = \frac{32\pi\mu}{\hbar^3} \left(\frac{e^2 D_1 D_2}{\kappa a_B \xi_T} \right)^2, \quad (3.15b)$$

$$\xi_T = \sqrt{\frac{\hbar^2}{2Mk_B T}} = \frac{462}{\sqrt{T}} \text{ \AA}, \quad (M = 0.207m_o) \quad (3.15c)$$

where $S(x)$ is defined in Eq. (3.11) and T is in kelvin for the exciton thermal length ξ_T .

Using the parameters in Table I, we estimate $W_{o,dip} = 2.87 \times 10^8 T \text{ sec}^{-1}$, where T is again in kelvin. The transfer rate is plotted in Fig. 1 as a function of the temperature for $d = 175 \text{ \AA}$ and $d = 375 \text{ \AA}$ and for the well widths $b_1 = 50 \text{ \AA}$ and $b_2 = 100 \text{ \AA}$ as well as for the two-dimensional limit $b_1 = b_2 = 0$. It is seen there that the effect of the well width is small. Note that the rate vanishes as T for decreasing T due to the fact that $C(\mathbf{K}_{\parallel}, R_z) \propto K_{\parallel}$ for $K_{\parallel} \rightarrow 0$ in contrast to TSK's result which is finite at $T = 0$. The calculated rates for $d = 175 \text{ \AA}$ ($375 \text{ \AA}$) displayed in Fig. 1 are about one order (two orders) of magnitude smaller than TSK's data, which are insensitive to d and T . However, the rate for a much shorter distance is much larger as shown by the dash-dotted curve on the right axis for $d = 80 \text{ \AA}$.

A4. Energy transfer from localized excitons to free electron-hole pairs

The theoretical results displayed in Fig. 1 show a rapid T -dependence in contrast to TSK's data. We now show that the T -dependence is absent for localized excitons. The rate for localized excitons is given by

$$W = \frac{2\pi}{\hbar} \sum_{\mathbf{k}_{\parallel}, \mathbf{K}_{\parallel}} |\langle 2, \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel} | H_{ee} | 1, \mathbf{R}_a \rangle|^2 \delta(\epsilon_{\mathbf{k}_{\parallel}} - \Delta'), \quad (3.16)$$

where we ignore the distribution of the localization lengths of the excitons. This expression can be evaluated using Eqs. (2.7) and (3.10) - (3.12) for $\Delta' > 0$, yielding

$$W = W_{o,dip}(\xi)g\left(\frac{d}{\xi}\right), \quad (3.17)$$

which has the same form as in Eq. (3.15) except that ξ_T is replaced by ξ . The rate is plotted as a function of ξ in Fig. 2 for $d = 175 \text{ \AA}$ and $d = 375 \text{ \AA}$ and for the well widths $b_1 = 50 \text{ \AA}$ and $b_2 = 100 \text{ \AA}$ and for the two-dimensional limits $b_1 = b_2 = 0$. The rate becomes maximum at about $\xi = 140 \text{ \AA}$ for $d = 175 \text{ \AA}$. The maximum occurs at a larger ξ for $d = 375 \text{ \AA}$ and is not shown in Fig. 2. These rates are of the same order of magnitude as those shown in Fig. 1 and are too small to explain the data.

B. Photon-Exchange Energy Transfer

B1. Exciton-photon coupling

The electrons see the photon field through the Hamiltonian [12]

$$H_{phr} = -\frac{e}{m_0 c} \mathbf{A} \cdot \mathbf{p}, \quad (3.18a)$$

$$\mathbf{A} = i \sum_{\mathbf{k}, \lambda} \left[\frac{2\pi c^2 \hbar}{\Omega \epsilon \omega_{\lambda \mathbf{k}}} \right]^{1/2} [b_{\lambda \mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}} - b_{\lambda \mathbf{k}}^\dagger e^{-i\mathbf{k} \cdot \mathbf{r}}] \hat{\mathbf{e}}_{\lambda \mathbf{k}}, \quad (3.18b)$$

$$\mathbf{p} = \frac{m_0}{i\hbar} \int \psi^\dagger(\mathbf{r}) [\mathbf{r}, H_0] \psi(\mathbf{r}) d^3 r, \quad (3.18c)$$

where $b_{\lambda \mathbf{k}}^\dagger$ ($b_{\lambda \mathbf{k}}$) creates (destroys) a photon with mode λ , wave vector $\mathbf{k}$, frequency $\omega_{\lambda \mathbf{k}} = k\tilde{c} = kc/n$, polarization $\hat{\mathbf{e}}_{\lambda \mathbf{k}}$ and $\epsilon = n^2$ where n is the refractive constant. The quantity H_0 is the single particle Hamiltonian for the electrons in the conduction and valence bands.

The exciton-photon coupling is obtained inserting Eqs. (2.1), (3.2) into (3.18) and is

given by

$$\begin{aligned} \langle j, 0, N_{\lambda\mathbf{k}} + 1 | H_{ph} | j, \mathbf{K}_{\parallel}, N_{\lambda\mathbf{k}} \rangle = & \frac{eE_g L}{\hbar c} \left[\frac{2\pi c^2 \hbar (N_{\lambda\mathbf{k}} + 1)}{\Omega \epsilon \omega_{\lambda\mathbf{k}}} \right]^{1/2} \delta_{\mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}} \\ & \times e^{-ik_z z_j} \hat{\mathbf{e}}_{\lambda\mathbf{k}} \cdot \mathbf{D}_j \int e^{-ik_z z} F_j(0, z, z) dz, \end{aligned} \quad (3.19)$$

where $N_{\lambda\mathbf{k}}$ is the photon occupation number, E_g is the gap energy and “ $j, 0$ ” on the left-hand side indicates the ground state of the j -th QW. Creation of an exciton by one-photon absorption is given by the complex conjugate of Eq. (3.19).

Hamiltonian for the creation of a free electron-hole pair through absorption of a photon of mode $\lambda\mathbf{k}$ is similarly obtained by inserting Eqs. (2.9), (3.2) into (3.18) and equals

$$\begin{aligned} \langle j, \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}, N_{\lambda\mathbf{k}} | H_{ph} | j, 0, N_{\lambda\mathbf{k}} + 1 \rangle = & \frac{eE_g}{\hbar c} \left[\frac{2\pi c^2 \hbar (N_{\lambda\mathbf{k}} + 1)}{\Omega \epsilon \omega_{\lambda\mathbf{k}}} \right]^{1/2} \delta_{\mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}} \\ & \times e^{ik_z z_j} \hat{\mathbf{e}}_{\lambda\mathbf{k}} \cdot \mathbf{D}_j^* \int e^{ik_z z} F_j^*(z, z) dz. \end{aligned} \quad (3.20)$$

Note that $\mathbf{k}_{\parallel}'$ represents the in-plane wave vector for the relative motion of the electron and the hole, while $\mathbf{k}$ is the photon wave vector. The expression in Eq. (3.20) is independent of the electron-hole relative wave vector $\mathbf{k}_{\parallel}'$. The range dependence of the photon-exchange interaction is introduced through the phase factors $\exp(\pm ik_z z_j)$ in Eqs. (3.19) and (3.20) as will be seen later.

B2. Energy transfer from localized excitons to free electron-hole pairs

We calculate the transfer rate of an exciton localized in QW1 to a free electron-hole pair state QW2 through photon-exchange interaction. The rate is given by a two-step process,

namely

$$W = \frac{2\pi}{\hbar} \sum_{\mathbf{k}_{\parallel}, \mathbf{K}_{\parallel}} |T_{12,a}|^2 \delta(\varepsilon_{\mathbf{k}_{\parallel}} + E_{\mathbf{K}_{\parallel}} - \Delta'), \quad (3.21)$$

where the t-matrix is given by

$$T_{12,a} = \sum_{\lambda\mathbf{k}} \frac{\langle 2, \mathbf{K}_{\parallel}', \mathbf{k}_{\parallel}', N_{\lambda\mathbf{k}} | H_{ph} | 2, 0, N_{\lambda\mathbf{k}} + 1 \rangle \langle 1, 0, N_{\lambda\mathbf{k}} + 1 | H_{ph} | 1, R_a, N_{\lambda\mathbf{k}} \rangle}{E_g - \hbar\omega_{\lambda\mathbf{k}} - i\Gamma}. \quad (3.22)$$

Here, the equilibrium photon occupation is zero (i.e., $N_{\lambda\mathbf{k}} = 0$), Γ is the exciton damping and the confinement energy is neglected compared with the gap energy in the intermediate-state denominator of Eq. (3.22). The t-matrix in Eq. (3.22) describes a process where a photon is spontaneously emitted from an exciton in QW1 and reabsorbed in QW2, exciting a free electron-hole pair. In view of the fact that the $T_{12,a}$ is independent of $\mathbf{k}_{\parallel}'$, the $\mathbf{k}_{\parallel}'$ -summation in Eq. (3.21) can be carried out immediately, yielding

$$W = \frac{L^4 \mu}{4\pi^2 \hbar^3} \int d^2 K_{\parallel}' |T_{12,a}|^2 \theta(\Delta' - E_{\mathbf{K}_{\parallel}'}), \quad (3.23)$$

where $\theta(x)$ is the unit step function.

Inserting Eqs. (2.7), (3.19) and (3.20) in Eq. (3.22), we find

$$T_{12,a} = \frac{8\sqrt{2}\xi E_g e^2 D_1 D_2}{\hbar L^2 c n a_B} e^{-i\mathbf{K}_{\parallel}' \cdot \mathbf{R}_a} e^{-\xi^2 K_{\parallel}'^2 / 2} Q(\mathbf{K}_{\parallel}', d), \quad (3.24)$$

$$Q(\mathbf{K}_{\parallel}', d) = \frac{\sqrt{\pi} a_B E_g}{4\sqrt{2}} \int_{-\infty}^{\infty} dk_z e^{ik_z d} \frac{\int dz F_1(0, z, z) e^{-ik_z z} \int dz' F_2(z', z') e^{ik_z z'}}{(E_g - \hbar\tilde{c}\sqrt{K_{\parallel}'^2 + k_z^2} - i\Gamma)\sqrt{K_{\parallel}'^2 + k_z^2}} P(\mathbf{K}_{\parallel}', k_z), \quad (3.25)$$

where

$$P(\mathbf{K}'_{\parallel}, k_z) = \sum_{\lambda} (\hat{\mathbf{e}}_{\lambda}(\mathbf{K}'_{\parallel}, k_z) \cdot \hat{\mathbf{D}}_1) (\hat{\mathbf{e}}_{\lambda}(\mathbf{K}'_{\parallel}, k_z) \cdot \hat{\mathbf{D}}_2), \quad (3.26)$$

and $\hat{\mathbf{D}}_j = \mathbf{D}_j / D_j$ is a unit vector. We observe that the dominant contribution to the k_z -integration in Eq. (3.25) arises from the photons with energies comparable to or smaller than the gap, namely from small $k_z \lesssim \xi_g^{-1}$ where $\xi_g = \hbar\tilde{c} / E_g = 353 \text{ \AA}$ for $E_g = 1.52 \text{ eV}$ and $n = 3.68$. [13] On the other hand, the z, z' integrations in the numerator of Eq. (3.25) are confined within $|z| < b_1 \ll \xi_g$, $|z'| < b_2' \ll \xi_g$. As a result, we can approximate $k_z z = k_z z' = 0$ in the numerator of Eq. (3.25). The z' -integration then yields unity while the z -integration yields $\int F_1(0, z, z) dz$. Employing a quasi-two-dimensional approximation introduced in Eq. (2.8): $F_1(0, z, z) = 2(2/\pi)^{1/2} F_1(z, z)/a_B$, we simplify Eq. (3.25) as.

$$Q(\mathbf{K}'_{\parallel}, d) = \int_0^{\infty} dk_z \cos(k_z d) \left(\frac{1}{(E_g / \hbar\tilde{c} - \sqrt{K_{\parallel}'^2 + k_z^2} - i\Gamma / \hbar\tilde{c})} + \frac{1}{\sqrt{K_{\parallel}'^2 + k_z^2}} \right) P_{\mathbf{K}'_{\parallel}, k_z}. \quad (3.27)$$

In Appendix B, we show that the angular integration with respect to $\phi_{\mathbf{K}'_{\parallel}}$ in the $\mathbf{K}'_{\parallel}$ -integration in Eq. (3.23) yields the following equality, when summed over $\mathbf{D}_1 \parallel \mathbf{D}_2$ and $\mathbf{D}_1 \perp \mathbf{D}_2$,

$$\frac{1}{2\pi} \int_0^{2\pi} d\phi_{\mathbf{K}'_{\parallel}} \sum_{\parallel, \perp} P(\mathbf{K}'_{\parallel}, k_z) P(\mathbf{K}'_{\parallel}, k_z) = \frac{1}{2} \left[1 + \frac{k_z^2 k_z'^2}{K^2 K'^2} \right], \quad (3.28)$$

where $K^2 = K_{\parallel}'^2 + k_z^2$ and $K'^2 = K_{\parallel}'^2 + k_z'^2$.

Inserting Eqs. (3.24) - (3.28) in Eq. (3.23), we find

$$W = \frac{W_{o,rad}}{2\pi^2} \int_0^\infty dx \theta(\xi^2 / \xi_{\Delta}^2 - x) e^{-x} (|I_o(xd^2 / \xi^2)|^2 + |I_1(xd^2 / \xi^2)|^2), \quad (3.29)$$

$$I_n(x) = \int_0^\infty dz \frac{z^{2n} \cos z}{[x + z^2]^n} \left\{ \frac{1}{\sqrt{x + z^2}} + \frac{1}{\frac{d}{\xi_g} - \sqrt{x + z^2} - i\gamma} \right\}, \quad (3.30)$$

where $W_{o,rad} = 32\pi\mu(E_g e^2 D_1 D_2)^2 / (\hbar^5 c^2 \epsilon a_B^2)$, $\xi_{\Delta}^2 = \hbar^2 / (2M\Delta')$ and $\gamma = \Gamma d / \hbar c = (\Gamma / E_g)(d / \xi_g) \ll 1$. For GaAs, $E_B = 4.06$ meV, $M = 0.207$, yielding $\xi_{\Delta} = 18.1$ Å for $\Delta = 60$ meV. Using the parameters introduced earlier, we estimate $W_{o,rad} = 3.64 \times 10^8 \text{ sec}^{-1}$. Note that the radiative rate in Eqs. (3.29) and (3.30) is independent of the QW widths as long as the widths are much smaller than ξ_g .

The transfer rate in Eq. (3.29) is the sum of the nonresonant and resonant contributions, i.e. $W = W_{nrs} + W_{res}$ where W_{nrs} is the contribution from the principal part of the integral in Eq. (3.30) while W_{res} is from the imaginary part. The resonant rate is given by

$$W_{res} = \frac{W_{o,rad}}{2} \int_0^\infty dx \theta(\xi^2 / \xi_{\Delta}^2 - x) \theta(\xi^2 / \xi_g^2 - x) e^{-x} \cos^2\left(\frac{d}{\xi} \sqrt{(\xi / \xi_g)^2 - x}\right) \times \left\{ \frac{(\xi / \xi_g)^2}{(\xi / \xi_g)^2 - x + \gamma} + (\xi_g / \xi)^2 [(\xi / \xi_g)^2 - x] \right\}, \quad (3.31)$$

where γ is inserted to avoid a weak logarithmic divergence and accounts for the finite-width correction to the delta function. The latter arises from the imaginary part of the second term in Eq. (3.30) in the limit $\gamma \rightarrow 0$. The net result depends only logarithmically on γ . The resonant rate W_{res} reduces to $W_{res} = W_{o,rad} \cos^2(d / \xi_g)$ in the limit $\xi \gg \xi_g$, ξ_{Δ} , and to $W_{res} = W_{o,rad} (\ln[(\xi^2 / \xi_g^2 + \gamma) / \gamma] + 1/2) \xi^2 / (2\xi_g^2)$ in the limit $\xi_g \gg \xi$, ξ_{Δ} , d . The

real part of the integral in Eq. (3.30) is calculated excluding the region $|x - x_s| < \gamma$ around the singularity point x_s . We use $\Gamma = 0.01$ meV corresponding to the lifetime of about 100 picosecond [3] for the numerical analysis presented below. This value corresponds to $\gamma = 3.26 \times 10^{-6}$ for $d = 175$ Å. The rate increases (decreases) by about 25% when γ is reduced (increased) by an order of magnitude.

Figure 3(a) displays the resonant, nonresonant and total transfer rates as a function of ξ for $d = 175$ Å. The resonant and nonresonant rates are equally important and have maxima near $\xi \sim \xi_g$, where $\hbar\omega_k$ (with $k \sim \xi^1$) becomes nearly resonant with E_g . The total transfer rate is insensitive to d as shown in Fig. 3(b) as a function of ξ (lower axis) for $d = 175, 275$ and 375 Å also in Fig. 4. The rate is independent of T for T -independent ξ and is in the range of TSK's Stokes transfer rate data [3] shown as a function of T (upper axis) in Fig. 3(b). The calculated rate is also plotted as a function of d in Fig. 4 for $\xi = 150, 400, 1500$ Å. The rate has a very slow dependence on d in agreement with TSK's data. The resonant part (not shown in Fig. 4) of the total rate for large $\xi = 1500 \gg \xi_g = 353$ Å vanishes near $d = \pi\xi_g/2 = 554$ Å as predicted by $W_{\text{res}} = W_{\text{O,rad}} \cos^2(d/\xi_g)$ and creates a shallow maximum for the total rate near $d = 350$ Å as seen in Fig. 4. It is possible from Figs. 3 and 4 that samples with a larger d can have faster rates depending on the parameter ξ .

B3. Energy transfer from plane-wave excitons to free electron-hole pairs

The energy transfer rate from a plane-wave exciton state in QW1 to a free electron-hole pair in QW2 can be obtained from the formalism in the previous section B2. The rate is given by

$$W = \frac{2\pi}{\hbar} \beta \int dE_{\mathbf{K}_{\parallel}} e^{-\beta E_{\mathbf{K}_{\parallel}}} < \sum_{\mathbf{k}_{\parallel}, \mathbf{K}_{\parallel}} |U_{12}(\mathbf{K}_{\parallel}', \mathbf{K}_{\parallel})|^2 \delta(\varepsilon_{\mathbf{k}_{\parallel}} - \Delta') >_{\mathbf{K}_{\parallel}}, \quad (3.32)$$

where the $T_{12}(\mathbf{K}_{||}', \mathbf{K}_{||})$ is obtained by replacing $|1, \mathbf{R}_a, N_{\lambda\mathbf{k}}\rangle$ with $|1, \mathbf{K}_{||}, N_{\lambda\mathbf{k}}\rangle$ in $T_{12,a}$ in Eq. (3.22). The Boltzmann average is taken over the initial energy $E_{\mathbf{K}_{||}}$ in Eq. (3.32). The initial and final energies $E_{\mathbf{K}_{||}}$ and $E_{\mathbf{K}_{||}'}$ cancel in the delta function of Eq. (3.32) due to $\mathbf{K}_{||} = \mathbf{K}_{||}'$. This means that the unit step function $\theta(\xi^2/\xi_{\Delta}^2 - x)$ in Eqs. (3.29) - (3.31) is not necessary and should be replaced by unity. Otherwise, the rate in Eq. (3.32) reduces to Eqs. (3.29) - (3.31) except that ξ is replaced with ξ_T defined in Eq. (3.15c), namely

$$W = \frac{W_{o,rad}}{2\pi^2} \int_0^{\infty} dx e^{-x} (|I_o(xd^2/\xi_T^2)|^2 + |I_1(xd^2/\xi_T^2)|^2), \quad (3.33)$$

where $W_{o,rad}$ and $I_n(x)$ were defined following Eq. (3.30). For the numerical results shown in Figs. 3 and 4, the cutoff factor $\theta(\xi^2/\xi_{\Delta}^2 - x)$ in Eq. (3.29) has a negligible effect because ξ^2/ξ_{Δ}^2 is very large. Therefore, the rates displayed Figs. 3 - 4 can be directly translated into T -dependent rates by equating $\xi = \xi_T = 462/T^{1/2}$ Å, where T is in kelvin. The energy transfer rate is shown as a function of T in Fig. 5 for $d = 175$ and 375 Å. The rapid decay with rising T is consistent with the steep decay with decreasing ξ below the maximum in Fig. 3 in view of $\xi = \xi_T = 462/T^{1/2}$ Å. We can also deduce from Fig. 3 that the rate has a maximum near $\xi_T = 390$ Å, namely near 1.4 K in Fig. 5. The rapid T -dependent decay at high T is due to the fact that the photon energy in the denominators of Eq. (3.27) becomes large. Figure 6 displays the d -dependence of the rate at $T = 4, 10$ and 50 K.

The radiative energy transfer rate in Fig. 4 and 6 decays very slowly for extremely large d . We show in Appendix C that the asymptotic behavior depends on the sample shape. The rate saturates to an asymptotic value independent of d in a long sample where d is much larger than the radii of the QW's. In the opposite limit, the rate slows down logarithmi-

cally. However, the rate is limited by the photon mean free path if there are other optical absorbing centers present in the system.

IV. FIELD-THEORETIC FORMALISM FOR EXCITON TRANSFER

A. Formalism

In this section, we express the transfer rate in terms of a correlation function, which allows a rigorous and systematic calculation of the rate by using a standard diagrammatic technique. The total Hamiltonian is the sum of H_t and H where

$$H = \sum_{jk} (\varepsilon_{jk} - \mu_j) \hat{n}_{jk} + H', \quad (4.1a)$$

$$H_t = \hat{t} + \hat{t}^\dagger. \quad (4.1b)$$

Here the first term of H is the energy of the exciton which is either at $j = 1$ or at $j = 2$ with the occupation number $\hat{n}_{jk} = 0$ or 1. The chemical potential μ_j at site j is introduced to project out the initial unphysical occupation of site 2 later. The second term H' in Eq. (4.1a) describes the rest of the system (e.g., phonons) and its interactions with the exciton. The operator $\hat{t}^\dagger$ transfers an exciton from QW1 to QW2 and $\hat{t}$ from QW2 to QW1.

The transition rate is given to the lowest order in $\hat{t}$ by

$$W = \frac{2\pi}{\hbar Z} \sum_{nm} [e^{-\beta E_n} |\langle m | \hat{t}^\dagger | n \rangle|^2 - e^{-\beta E_m} |\langle n | \hat{t} | m \rangle|^2] \delta(E_n - E_m + \Omega), \quad (4.2)$$

where $H|n\rangle = E_n|n\rangle$, $\Omega = \mu_1 - \mu_2$ and Z is the distribution function for H . The first term in Eq. (4.2) describes the transfer rate from QW1 to QW2 and the second term the back transfer. To ensure that QW1 is occupied and QW2 is empty initially, we let $\Omega \rightarrow \infty$. The

expression in Eq. (4.2) can be rewritten as (see Appendix D)

$$W = \frac{2}{\hbar} \text{Im} \mathcal{F}(\omega_r \rightarrow \Omega + i0), \mathcal{F}(\omega_r) = \int_0^\beta du e^{\omega_r u} \langle e^{uH} \hat{t} e^{-uH} \hat{t}^\dagger \rangle, \quad (4.3)$$

where $\omega_r = 2\pi i r / \beta$ is analytically continued to slightly above the real axis. Here r is an integer. The angular brackets in Eq. (4.3) signify the thermodynamic average. The current correlation function in Eq. (4.3) can be evaluated systematically applying a standard temperature ordered diagram expansion technique. [14, 15]

B. Application to Anti-Stokes Transfer of Plane-wave Excitons

The above result is useful for studying the role of damping and scattering for anti-Stokes transfer. Since there is only one exciton present in the system, we can employ a Fermion representation and write $\hat{n}_{jk} = c_{jk}^\dagger c_{jk}$ where $k = \mathbf{K}_\parallel$, $c_{jk}^\dagger$, c_{jk} are creation and destruction operators and $\hat{t}^\dagger = \sum_k t_{\mathbf{K}_\parallel} c_{2k}^\dagger c_{1k}$. Here $t_{\mathbf{K}_\parallel} = \langle 2, \mathbf{K}_\parallel | H_t | 1, \mathbf{K}_\parallel \rangle$ represents dipolar and photon-exchange coupling studied in the previous section. We assume that the exciton is interacting with the phonons and other static scattering centers (e.g., surface roughness, impurities).

The rate in Eq. (4.3) is given by a bubble diagram and a one-rung diagram shown in Fig. 7, where the solid lines are dressed exciton propagators and the wave line is a phonon propagator or a single-impurity line. The bubble diagram equals [14, 15]

$$\mathcal{F}(\omega_r) = -\frac{1}{\beta} \sum_{\mathbf{K}_\parallel \ell} |t_{\mathbf{K}_\parallel}|^2 \frac{1}{\zeta_\ell + \mu_1 - \varepsilon_{1\mathbf{K}_\parallel} - S_{1\mathbf{K}_\parallel}(\zeta_\ell)} \frac{1}{\zeta_\ell + \mu_2 + \omega_r - \varepsilon_{2\mathbf{K}_\parallel} - S_{2\mathbf{K}_\parallel}(\zeta_\ell + \omega_r)}, \quad (4.4a)$$

where $S_{j\mathbf{K}_\parallel}(\zeta_\ell)$ is the self-energy part, $\zeta_\ell = (2\ell + 1)\pi i \beta^{-1}$ and ℓ is an integer. Performing

the ℓ -summation and inserting Eq. (4.4a) in Eq. (4.3), we find [14, 15, 16]

$$W = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} dz [f_1(z) - f_2(z)] \sum_{\mathbf{K}_{\parallel}} |\mathbf{t}_{\mathbf{K}_{\parallel}}|^2 \rho_{1\mathbf{K}_{\parallel}}(z) \rho_{2\mathbf{K}_{\parallel}}(z), \quad (4.4b)$$

$$\rho_{j\mathbf{K}_{\parallel}}(z) = \frac{1}{\pi} \frac{\Gamma_{j\mathbf{K}_{\parallel}}(z)}{(z - E_{j\mathbf{K}_{\parallel}})^2 + \Gamma_{j\mathbf{K}_{\parallel}}(z)^2}, \quad (4.4c)$$

where $f_j(z) = \exp(-\beta(z - \mu_j))$ is the occupation function (i.e., the Fermi function in the non-degenerate limit) and $\Gamma_{j\mathbf{K}_{\parallel}}(z)$ is the imaginary part of the self-energy. The quantity $\Gamma_{j\mathbf{K}_{\parallel}}(z)$ represents damping due to exciton interactions and will be given later. The exciton shift (i.e., the real part of the self-energy) is absorbed into $E_{j\mathbf{K}_{\parallel}}$ in the spectral function in Eq. (4.4c). The second term $\propto f_2(z)$ in Eq. (4.4b) is zero in the limit $\Omega \rightarrow \infty$ and will be dropped because QW2 is empty initially.

In the limit $\Delta \gg \Gamma$, dominant contributions arise from the two non-overlapping resonances at $z = E_{1\mathbf{K}_{\parallel}}$ and $z = E_{2\mathbf{K}_{\parallel}}$ in Eq. (4.4b), yielding

$$W = \frac{2}{\hbar Z_1 \Delta^2} \sum_{\mathbf{K}_{\parallel}} |\mathbf{t}_{\mathbf{K}_{\parallel}}|^2 [e^{-\beta E_{1\mathbf{K}_{\parallel}}} \Gamma_{2\mathbf{K}_{\parallel}}(E_{1\mathbf{K}_{\parallel}}) + e^{-\beta E_{2\mathbf{K}_{\parallel}}} \Gamma_{1\mathbf{K}_{\parallel}}(E_{2\mathbf{K}_{\parallel}})], \quad (4.5)$$

where $\exp(-\beta\mu_1) = \sum_{\mathbf{K}_{\parallel}} \exp(-\beta E_{1\mathbf{K}_{\parallel}}) \equiv Z_1$ for a single-particle occupancy (i.e., $\sum_{\mathbf{K}_{\parallel}} f_1(E_{1\mathbf{K}_{\parallel}}) = 1$) in QW1. The physical origin of the expression in Eq. (4.5) will be discussed later in this section.

The nonactivated result of TSK is obtained from Eq. (4.4b) by replacing the occupation function by $f_1(z) = f_1(E_{1\mathbf{K}_{\parallel}})$ at the lower-energy resonance $z = E_{1\mathbf{K}_{\parallel}}$:

$$W_1 = \frac{2\pi}{\hbar Z_1} \sum_{\mathbf{K}_{\parallel}} |\mathbf{t}_{\mathbf{K}_{\parallel}}|^2 e^{-\beta E_{1\mathbf{K}_{\parallel}}} \int_{-\infty}^{\infty} dz \rho_1(z) \rho_2(z), \quad (4.6)$$

and using a Lorentzian function for $\rho_1(z)$ centered at $E_{1K_{\parallel}}$ and a sharply-peaked Gaussian function centered at $E_{2K_{\parallel}}$. Next step is to take the Gaussian resonance at $z = E_{2K_{\parallel}}$ and replace the integral by $\rho_1(E_{2K_{\parallel}})$, yielding a nonactivated result. However, this step is incorrect because the occupation function should also reflect the same Gaussian resonance $f_1(z) = f_1(E_{2K_{\parallel}}) \propto \exp(-\beta E_{2K_{\parallel}}) \propto e^{-\beta \Delta}$ but not $f_1(z) = f_1(E_{1K_{\parallel}})$ as in Eq. (4.6). Therefore, the correct rate is activated as shown by the second term in Eq. (4.5). In the following, we show that the first term in Eq. (4.5) is also activated by $e^{-\beta \Delta}$. For this purpose, it is necessary to examine the expression for $\Gamma_{jK_{\parallel}}(z)$. The basic argument relies on the fact that 1) $\Gamma_{2K_{\parallel}}(E_{1K_{\parallel}}) = 0$ unless $K_{\parallel}$ is large enough to satisfy $E_{1K_{\parallel}} \geq \Delta$ for elastic scattering and 2) $\Gamma_{2K_{\parallel}}(E_{1K_{\parallel}}) \propto e^{-\beta \Delta}$ for $E_{1K_{\parallel}} \sim k_B T$ for phonon scattering.

Damping from static scattering is given by the well-known golden-rule expression

$$\Gamma_{jK_{\parallel}}(z) = \pi \sum_{K'_{\parallel}} \langle |U_j(K'_{\parallel}, K_{\parallel})|^2 \rangle \delta(E_{jK'_{\parallel}} - z), \quad (4.7)$$

where $\langle |U_j(K'_{\parallel}, K_{\parallel})|^2 \rangle$ is the ensemble average of the absolute square of the scattering matrix in the j -th QW. Choosing the origin of the energy as $E_{1K_{\parallel}} = E_{K_{\parallel}}$ and $E_{2K_{\parallel}} = E_{K_{\parallel}} + \Delta$, it is clear that $\Gamma_{2K_{\parallel}}(E_{1K_{\parallel}}) = 0$ in Eq. (4.7) unless $E_{1K_{\parallel}} = E_{2K_{\parallel}}$ ($\geq \Delta$) in the delta function, meaning that the first term in Eq. (4.5) is also activated by $e^{-\beta \Delta}$.

For the electron-phonon scattering, $\Gamma_{jK_{\parallel}}(z)$ is given by [15]

$$\Gamma_{jK_{\parallel}}(z) = \pi \sum_{K_{\parallel}sq \pm} |V_{jsq}|^2 \left(n_{sq} + \frac{1}{2} \pm \frac{1}{2}\right) \delta_{K_{\parallel}, K_{\parallel} \pm q_{\parallel}} \delta(E_{jK_{\parallel}} \pm \hbar \omega_{sq} - z), \quad (4.8)$$

where the electron-phonon matrix element is defined for one-phonon absorption (-) and emission (+) by

$$\langle j, K'_{\parallel}, n_{sq} + \frac{1}{2} \pm \frac{1}{2} | H | j, K_{\parallel}, n_{sq} \rangle = V_{j,sq} e^{iq_z z_j} \left(n_{sq} + \frac{1}{2} \pm \frac{1}{2}\right)^{1/2} \delta_{K_{\parallel}, K_{\parallel} \pm q_{\parallel}}, \quad (4.9)$$

and n_{sq} is the boson function for the phonon mode s with a wave vector q and energy $\hbar\omega_{sq}$. A full expression for the exciton-phonon interaction $V_{j,sq}$ will be given in the next section. For one-phonon emission, $\Gamma_{2K_{\parallel}}(E_{1K_{\parallel}}) = 0$ in Eq. (4.8) unless $E_{1K_{\parallel}} = E_{2K_{\parallel}'} + \hbar\omega_{sq} (\geq \Delta)$ in the delta function, meaning that the first term in Eq. (4.5) is again activated by $e^{-\beta\Delta}$. For one-phonon absorption, we have $\hbar\omega_{sq} = E_{2K_{\parallel}'} - E_{1K_{\parallel}} \geq \Delta$, $n_{sq} \propto e^{-\beta\Delta}$ and therefore $\Gamma_{2K_{\parallel}}(E_{1K_{\parallel}}) = e^{-\beta\Delta}$ in Eq. (4.8), again yielding an activated result for the first term in Eq. (4.5).

The physical origin of the expression given in Eq. (4.5) can be understood by rederiving the full phonon-assisted rate in terms of an alternative standard perturbation method. The resulting rate will be used in the next section. For this purpose, we consider two possible perturbation channels which connect the initial state $|1, K_{\parallel}, n_{sq}\rangle$ to the final state $|2, K_{\parallel}', n_{sq}+1/2 \pm 1/2\rangle$ through intermediate states via emission (+) and absorption (-) of a phonon. The thermally averaged rate equals

$$W = \frac{2\pi}{\hbar Z_1} \sum_{K_{\parallel}} e^{-\beta E_{1K_{\parallel}}} \sum_{K_{\parallel}', sq \pm} |t_{12, \pm}|^2 \delta(E_{2K_{\parallel}'} \pm \hbar\omega_{sq} - E_{1K_{\parallel}}), \quad (4.10)$$

where the t-matrix is given by [17]

$$t_{12, \pm} = \left(\frac{t_{K_{\parallel}} e^{iq_z z_2} V_{2, sq}}{-\Delta} + \frac{t_{K_{\parallel}'} e^{iq_z z_1} V_{1, sq}}{\Delta} \right) \left(n_{sq} + \frac{1}{2} \pm \frac{1}{2} \right)^{1/2} \delta_{K_{\parallel}, K_{\parallel}' \pm q_{\parallel}}. \quad (4.11)$$

The first term of Eq. (4.11) describes a process where the exciton crosses the barrier through dipolar and photon-exchange interactions (i.e., via $t_{K_{\parallel}}$) to an intermediate virtual state $|2, K_{\parallel}, n_{sq}\rangle$ and is then scattered to the final state by $V_{2, sq}$ emitting and absorbing a phonon. For the second term, the exciton is scattered to an intermediate state $|1, K_{\parallel}', n_{sq}+1/2 \pm 1/2\rangle$ inside QW1 by $V_{1, sq}$ emitting and absorbing a phonon and then crosses the

barrier into the final state via $t_{\mathbf{K}_{||}}$. The first term of Eq. (4.5) is obtained immediately by inserting the first term of Eq. (4.11) in Eq. (4.10) and utilizing the expression in Eq. (4.8). In Appendix E, we show that the second term of Eq. (4.5) follows from the second term of Eq. (4.11). The cross terms of the above t-matrix yield the contribution shown by the one-rung diagram in Fig. 7:

$$W = -\frac{2\pi}{\hbar Z_1 \Delta^2} \sum_{\mathbf{K}_{||}} e^{-\beta E_{1\mathbf{K}_{||}}} \sum_{\mathbf{K}'_{||}, s\mathbf{q}\pm} [t_{\mathbf{K}_{||}} e^{iq_z(z_2-z_1)} V_{2,s\mathbf{q}}(t_{\mathbf{K}'_{||}} V_{1,s\mathbf{q}})^* + c.c.](n_{s\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}) \quad (4.12)$$

$$\times \delta_{\mathbf{K}_{||}, \mathbf{K}'_{||} \pm \mathbf{q}_{||}} \delta(E_{2\mathbf{K}'_{||}} \pm \hbar \omega_{s\mathbf{q}} - E_{1\mathbf{K}_{||}}),$$

More detailed study of the total one-phonon-assisted rate will be carried out in the next section. An analogous analysis can be applied to static scattering.

V. ANTI-STOKES ENERGY TRANSFER

In this section, we study exciton transfer from wide QW1 to narrow QW2 (e.g., $b_1 = 100$ and $b_2 = 50$ Å). The ground sublevel of QW2 lies Δ ($= 60$ meV) above that of QW1. We have shown in the previous section that dipolar or photon-exchange anti-Stokes energy transfer is activated. It turns out that phonon-assisted transfer rate $\propto e^{-\beta\Delta}$ is negligibly small at low temperatures. Therefore, we first consider exciton transfer through Auger-like ionization of the excitons over the barrier, which is a dominant anti-Stokes energy transfer mechanism if the exciton density is not too low. This mechanism yields sufficiently large rates to explain recent data.

A. Over-barrier Ionization of Plane-wave Excitons

In this process, an exciton in $|\mathbf{K}_{1||}\rangle$ is annihilated giving its energy to ionize another exciton $|\mathbf{K}_{2||}\rangle$ over the barrier into a free electron-hole state $|\mathbf{K}'_{||}, \mathbf{k}'_{||}, k'_{ze}, k'_{zh}\rangle$. Once

excited over the barrier, the carriers fall into QW2 immediately in the time scale of a pico-second which is much shorter than the radiative lifetime. Phonon-assisted carrier capture is faster in the narrower QW. The QW index j will be suppressed here because this process occurs only in QW1 while QW2 is empty. The final unconfined free electron-hole state is described by

$$|K_{\parallel}', k_{\parallel}', k_{ze}', k_{zh}'\rangle = \frac{v_0}{L^2} \sum_{r_e, r_h} e^{iK_{\parallel}' \cdot R_{\parallel}} e^{-ik_{\parallel}' \cdot (r_e - r_h)} F(k_{ze}', z_e; k_{zh}', z_h) a_{cr_e}^{\dagger} a_{cr_h} |0\rangle, \quad (5.1a)$$

where

$$F(k_{ze}', z_e; k_{zh}', z_h) = \phi_{e, k_{ze}'}(z_e) \phi_{h, k_{zh}'}(z_h) \quad (5.1b)$$

is the product of the normalized electron and hole wave functions. These functions are propagating above the barrier in the z -direction.

Following the same procedure employed in Eqs. (3.1) - (3.6), we find

$$\begin{aligned} \langle K_{\parallel}', k_{\parallel}', k_{ze}', k_{zh}' | H_{ee} | K_{1\parallel}, K_{2\parallel} \rangle &= \frac{1}{L^2} \delta_{K_{1\parallel} + K_{2\parallel}, K_{\parallel}'} \int dZ \int dZ' C_{D_1}(K_{1\parallel}, R_z) F(0, Z, Z') \int d^2 r_{\parallel} \\ &\times \int dz' F(r_{\parallel}, Z, z') [F^*(k_{ze}', Z; k_{zh}', z') e^{i(k_{\parallel}' + \alpha_h K_{1\parallel}) \cdot r_{\parallel}} - F^*(k_{ze}', z'; k_{zh}', Z) e^{i(k_{\parallel}' - \alpha_e K_{1\parallel}) \cdot r_{\parallel}}] \\ &+ (1 \rightarrow 2), \end{aligned} \quad (5.2a)$$

where $R_z = Z' - Z$ and

$$C_{D_1}(K_{\parallel}, R_z) = \int d^2 R_{\parallel} e^{iK_{\parallel} \cdot R_{\parallel}} C_{D_1}(R) = -\frac{2\pi i e^2}{\kappa} \hat{K}_{\parallel} \cdot D_1 e^{-K_{\parallel} |R_z|}. \quad (5.2b)$$

Here $C_{D_1}(\mathbf{R})$ is the monopole-dipole interaction

$$C_{D_1}(\mathbf{R}_{\parallel}, R_z) = -\frac{e^2}{\kappa} \frac{\mathbf{R} \cdot \mathbf{D}_1}{R^3} \quad (5.2c)$$

and $\mathbf{R} = (\mathbf{R}_{\parallel}, R_z)$. In Eq. (5.2), the transition dipole $\mathbf{D}_j$ is that of the initial confined state $|\mathbf{K}_{j\parallel}\rangle$.

The Z, Z' integrations are sharply localized in QW1. We approximate $C_{D_1}(\mathbf{K}_{1\parallel}, R_z)$ by its average over the probability distribution of Z and Z'

$$C_{D_1}(\mathbf{K}_{\parallel}) = -\frac{2\pi i e^2}{\kappa} \hat{\mathbf{K}}_{\parallel} \cdot \mathbf{D}_1 B(K_{\parallel}); \quad B(K_{\parallel}) = \langle e^{-K_{\parallel}|Z-Z'|} \rangle, \quad (5.3)$$

where $B(K_{\parallel}) = 2(x - 1 + e^{-x})/x^2$ with $x = bK_{\parallel}$ for a rectangular distribution of the electron and hole densities and b is the width of QW1. At this point, we employ the quasi-two-dimensional approximations in Eq. (2.8) for the exciton wave function in Eq. (5.2a).

Carrying out the $\mathbf{r}_{\parallel}$ -integration for the matrix element in Eq. (5.2a), we find

$$\begin{aligned} \langle \mathbf{K}'_{\parallel}, \mathbf{k}'_{\parallel}, k'_{ze}, k'_{zh} | H_{ee} | \mathbf{K}_{1\parallel}, \mathbf{K}_{2\parallel} \rangle &= \frac{32}{L^2} \delta_{\mathbf{K}_{1\parallel} + \mathbf{K}_{2\parallel}, \mathbf{K}'_{\parallel}} C_{D_1}(\mathbf{K}_{1\parallel}) \int dZ \int dz' F^*(k'_{ze}, Z; k'_{zh}, z') \\ &\times F_g(Z, z') \left[\frac{1}{[(\mathbf{k}'_{\parallel} + \alpha_h \mathbf{K}_{1\parallel})^2 a_B^2 + 4]^{3/2}} - \frac{1}{[(\mathbf{k}'_{\parallel} - \alpha_e \mathbf{K}_{1\parallel})^2 a_B^2 + 4]^{3/2}} \right] + (1 \rightarrow 2), \end{aligned} \quad (5.4)$$

where a subscript "g" is introduced to distinguish the ground-state confinement function $F_g(Z, z')$ from the over-barrier wave function, which is assumed to be symmetric in the electron and the hole coordinates.

Thermally averaged total exciton-exciton ionization rate is given by

$$W = \frac{2\pi}{\hbar Z_1} \sum_{\mathbf{K}_{1\parallel}, \mathbf{K}_{2\parallel}} e^{-\beta E_{\mathbf{K}_{1\parallel}}} f_{\mathbf{K}_{2\parallel}} \sum_{\mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}, k_{ze}', k_{zh}'} |\langle \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}, k_{ze}', k_{zh}' | H_{ee} | \mathbf{K}_{1\parallel}, \mathbf{K}_{2\parallel} \rangle|^2 \quad (5.5)$$

$$\times \delta(E_{\mathbf{K}_{1\parallel}} + E_{\mathbf{K}_{2\parallel}} - 2E_B + E_g - [E_{\mathbf{K}_{\parallel}} + \varepsilon_{\mathbf{k}_{\parallel}} + \varepsilon_e(k_{ze}') + \varepsilon_h(k_{zh}')]),$$

where $f_{\mathbf{K}_{2\parallel}} = 2\pi\hbar^2 N_{ex} \beta \exp(-\beta E_{\mathbf{K}_{2\parallel}}) / M$ is the exciton occupancy, N_{ex} is the two-dimensional exciton density, $\varepsilon_e(k_{ze}')$, $\varepsilon_h(k_{zh}')$ are the electron, hole energies in the z -direction. Because $F_g(Z, z')$ is confined inside the QW in Eq. (5.4), the Z, z' integrations yield negligible contributions for large $|k_{ze}'|, |k_{zh}'| > \pi/b$. Therefore, we approximate $\varepsilon_e(k_{ze}') + \varepsilon_h(k_{zh}')$ = V_0 in the energy delta function in Eq. (5.5) for small $|k_{ze}'|, |k_{zh}'| < \pi/b$, where V_0 is the sum of the well depths in the conduction and valence bands. Also, the exciton energies are of the order of the thermal energy and are neglected. The energy delta function then yields $\varepsilon_{\mathbf{k}_{\parallel}} = E_g - V_0 - 2E_B \equiv E_g^*$.

The wave number $k_{\parallel}'$ for the electron-hole relative motion in the denominators of Eq. (5.4) is much larger than $K_{1\parallel}$ in view of the fact that $\varepsilon_{\mathbf{k}_{\parallel}} \gg E_{1\mathbf{K}_{\parallel}}$. This allows us to expand the two terms in the brackets of Eq. (5.4) to the first order in $\mathbf{K}_{1\parallel}$, yielding

$$\langle \mathbf{K}_{\parallel}, \mathbf{k}_{\parallel}, k_{ze}', k_{zh}' | H_{ee} | \mathbf{K}_{1\parallel}, \mathbf{K}_{2\parallel} \rangle = -\frac{96}{L^2} \delta_{\mathbf{K}_{1\parallel} + \mathbf{K}_{2\parallel}, \mathbf{K}_{\parallel}} \int dZ \int dz' F_g(Z, z') \quad (5.6)$$

$$\times F^*(k_{ze}', Z; k_{zh}', z') \frac{C_{D1}(\mathbf{K}_{1\parallel})(\mathbf{k}_{\parallel} \cdot \mathbf{K}_{1\parallel}) + C_{D2}(\mathbf{K}_{2\parallel})(\mathbf{k}_{\parallel} \cdot \mathbf{K}_{2\parallel})}{k_{\parallel}^5 a_B^3}.$$

Here $k_{\parallel}'$ is fixed by the condition $\varepsilon_{\mathbf{k}_{\parallel}} = E_g^*$.

Inserting Eq. (5.6) in Eq. (5.5), using $\langle (\mathbf{k}_{\parallel}' \cdot \mathbf{K}_{1\parallel})^2 \rangle = (k_{\parallel}' K_{1\parallel})^2 / 2$, $\langle (\mathbf{k}_{\parallel}' \cdot \mathbf{K}_{1\parallel})(\mathbf{k}_{\parallel}' \cdot \mathbf{K}_{2\parallel}) \rangle = \mathbf{K}_{1\parallel} \cdot \mathbf{K}_{2\parallel} k_{\parallel}'^2 / 2$ for the angular averages over $\mathbf{k}_{\parallel}'$ and summing over the variables $\mathbf{K}_{1\parallel}, \mathbf{k}_{\parallel}'$,

k_{ze}' , k_{zh}' , we simplify the rate in Eq. (5.5) as

$$W = \frac{96^2 \mu \pi \beta N_{ex}}{\hbar L^2 k_{\parallel}^8 a_B^6 M Z} \left(\frac{2\pi e^2}{\kappa} \right)^2 \sum_{\mathbf{K}_{1\parallel}, \mathbf{K}_{2\parallel}} e^{-\beta E_{\mathbf{K}_{1\parallel}}} e^{-\beta E_{\mathbf{K}_{2\parallel}}} [|\hat{\mathbf{K}}_{1\parallel} \cdot \mathbf{D}_1 B(K_{1\parallel})|^2 K_{1\parallel}^2 + |\hat{\mathbf{K}}_{2\parallel} \cdot \mathbf{D}_2 B(K_{2\parallel})|^2 K_{2\parallel}^2 + 2(\hat{\mathbf{K}}_{1\parallel} \cdot \mathbf{D}_1) B(K_{1\parallel})(\hat{\mathbf{K}}_{2\parallel} \cdot \mathbf{D}_2) B(K_{2\parallel})(\mathbf{K}_{1\parallel} \cdot \mathbf{K}_{2\parallel})]. \quad (5.7)$$

The k_{ze}' , k_{zh}' summations over the product of the square of the absolute value of the product of the Z' and z' integrations in Eq. (5.6) yields unity.

The expression in Eq. (5.7) is further simplified upon carrying out the angular averages over $\mathbf{K}_{1\parallel}$, $\mathbf{K}_{2\parallel}$ and yields

$$W = \frac{96^2 \pi^2 2 \mu N_{ex}}{\hbar^3 k_{\parallel}^8 a_B^6} \left(\frac{e^2 D_1}{\kappa} \right)^2 \int_0^\infty d(\xi_T K_{1\parallel})^2 e^{-(\xi_T K_{1\parallel})^2} \int_0^\infty d(\xi_T K_{2\parallel})^2 e^{-(\xi_T K_{2\parallel})^2} \times [B(K_{1\parallel})^2 K_{1\parallel}^2 + \frac{1}{2} B(K_{1\parallel}) B(K_{2\parallel}) K_{1\parallel} K_{2\parallel} \hat{\mathbf{D}}_1 \cdot \hat{\mathbf{D}}_2], \quad (5.8)$$

where $D_1 = D_2$. Averaging over $\mathbf{D}_1 \parallel \mathbf{D}_2$, $\mathbf{D}_1 \perp \mathbf{D}_2$ and performing the $K_{j\parallel}^2$ -integrations, the expression in Eq. (5.8) is further simplified as

$$W = W_{o,ion}(\xi_T) \left[\int_0^\infty x e^{-x} B(\sqrt{x} / \xi_T)^2 dx + \frac{1}{4} \left(\int_0^\infty \sqrt{x} e^{-x} B(\sqrt{x} / \xi_T) dx \right)^2 \right], \quad (5.9)$$

where $W_{o,ion}(\xi_T) = (96\pi)^2 N_{ex} (e^2 D_1 / \kappa)^2 E_B^3 / \hbar \xi_T^2 E_g^{*4}$. For the parameters in Table I, $N_{ex} = 5 \times 10^{10} \text{ cm}^{-2}$ and $E_g^* = 1.09 \text{ eV}$ corresponding to GaAs/Al_{0.3}Ga_{0.7}As, we estimate

$W_{o,ion}(\xi_T) = 1.25T \times 10^4 \text{ sec}^{-1}$ where T is in kelvin. The temperature dependence of the exciton ionization rate is plotted in Fig. 8 for $b = 100$ and $50 \text{ \AA}$. The theoretical rates are in the range of TSK's observed data.

B. Over-barrier Ionization of Localized Excitons

In this process, the initial exciton states $|K_{1\parallel}\rangle$ and $|K_{2\parallel}\rangle$ are replaced by localized states $|R_1\rangle$ and $|R_2\rangle$. Final free electron-hole states $|K_{\parallel}', k_{ze}', k_{zh}'\rangle$ are the same. The rate in Eq. (5.5) is replaced with

$$W = \frac{2\pi}{\hbar} \sum_{R_2} \sum_{K_{\parallel}', k_{ze}', k_{zh}'} | \langle K_{\parallel}', k_{\parallel}', k_{ze}', k_{zh}' | H_{ee} | R_1, R_2 \rangle |^2 \quad (5.10)$$

$$\times \delta(E_g - 2E_B - [E_{K_{\parallel}'} + \epsilon_{k_{\parallel}'} + \epsilon_e(k_{ze}') + \epsilon_h(k_{zh}')]).$$

The matrix element in Eq. (5.10) can be evaluated using Eqs. (2.7) and (5.6) and yields

$$\begin{aligned} \langle K_{\parallel}', k_{\parallel}', k_{ze}', k_{zh}' | H_{ee} | R_1, R_2 \rangle = & -\frac{384\pi\xi^2}{L^4 k_{\parallel}'^5 a_B^3} e^{-iK_{\parallel}'(R_1+R_2)/2} \sum_{Q_{\parallel}} e^{iQ_{\parallel}R} e^{-\xi^2(Q_{\parallel}^2 + K_{\parallel}'^2/4)} \\ & \times \int dZ \int dz' F_g(Z, z') F^*(k_{ze}', Z; k_{zh}', z') [C_{D1} (\frac{1}{2}K_{\parallel}' - Q_{\parallel}) k_{\parallel}' \cdot (\frac{1}{2}K_{\parallel}' - Q_{\parallel}) \\ & + C_{D2} (\frac{1}{2}K_{\parallel}' + Q_{\parallel}) k_{\parallel}' \cdot (\frac{1}{2}K_{\parallel}' + Q_{\parallel})], \end{aligned} \quad (5.11)$$

where $\mathbf{R} = \mathbf{R}_1 - \mathbf{R}_2$, $K_{1\parallel} = K_{\parallel}'/2 - Q_{\parallel}$ and $K_{2\parallel} = K_{\parallel}'/2 + Q_{\parallel}$. The factor $\exp(-\xi^2 K_{\parallel}'^2/4)$ in Eq. (5.11) restricts $K_{\parallel}'$ to $K_{\parallel}' < 2/\xi$. Since ξ is of the order of b or larger, we can neglect $E_{K_{\parallel}'}$ in the energy delta function in Eq. (5.10). We also approximate $\epsilon_e(k_{ze}') + \epsilon_h(k_{zh}') = V_0$ as before. The energy delta function then yields $\epsilon_{k_{\parallel}'} = E_g - V_0 - 2E_B \equiv E_g^*$.

Inserting Eq. (5.11) in Eq. (5.10), using $\sum_{\mathbf{R}} \exp(i(Q_{\parallel} \pm Q_{\parallel}') \cdot \mathbf{R}) = N_{ex} L^2 \delta_{Q_{\parallel}, \mp Q_{\parallel}'}$ and car-

rying out the angular average over $\mathbf{k}_{||}'$, we find

$$W = \frac{2\mu N_{ex}}{\hbar^3} \left(\frac{384\pi\xi^2}{L^2 k_{||}^4 a_B^3} \right)^2 \sum_{\mathbf{Q}_{||}, \mathbf{Q}_{||}'} e^{-2\xi^2(Q_{||}^2 + Q_{||}'^2)} [|\mathbf{Q}_{||}' - \mathbf{Q}_{||}|^2 (|C_{D_1}(\mathbf{Q}_{||}' - \mathbf{Q}_{||})|^2 + |C_{D_2}(\mathbf{Q}_{||}' - \mathbf{Q}_{||})|^2) + 2C_{D_1}(\mathbf{Q}_{||}' - \mathbf{Q}_{||})C_{D_2}(\mathbf{Q}_{||}' + \mathbf{Q}_{||})^* (\mathbf{Q}_{||}' - \mathbf{Q}_{||}) \cdot (\mathbf{Q}_{||}' + \mathbf{Q}_{||})], \quad (5.12)$$

where a new variable $\mathbf{Q}_{||}' = \mathbf{K}_{||}'/2$ is introduced and $\Sigma_{\mathbf{K}_{||}'}(..) = 4\Sigma_{\mathbf{Q}_{||}'}(..)$. We now rotate the coordinate by yet another transformation $\mathbf{K}_{||} = (\mathbf{Q}_{||}' - \mathbf{Q}_{||})/2^{1/2}$, $\mathbf{K}_{||}' = (\mathbf{Q}_{||}' + \mathbf{Q}_{||})/2^{1/2}$ and rescale $2^{1/2}\mathbf{K}_{||} \rightarrow \mathbf{K}_{||}$ and $2^{1/2}\mathbf{K}_{||}' \rightarrow \mathbf{K}_{||}'$.

Averaging over the two independent angular directions of $\mathbf{K}_{||}$ and $\mathbf{K}_{||}'$, we obtain

$$W = \frac{96^2 \pi^2 2\mu N_{ex}}{\hbar^3 k_{||}^8 a_B^6} \left(\frac{e^2 D_1}{\kappa} \right)^2 \int_0^\infty d(\xi K_{||})^2 e^{-(\xi K_{||})^2} \int_0^\infty d(\xi K_{||}')^2 e^{-(\xi K_{||}')^2} \times [K_{||}^2 B(K_{||})^2 + \frac{1}{2} K_{||} K_{||}' B(K_{||}) B(K_{||}') \hat{\mathbf{D}}_1 \cdot \hat{\mathbf{D}}_2]. \quad (5.13)$$

This result is identical to Eq. (5.8) if ξ is replaced by ξ_T . When averaged over $\mathbf{D}_1 \parallel \mathbf{D}_2$, $\mathbf{D}_1 \perp \mathbf{D}_2$, it yields

$$W = W_{o,ion}(\xi) \left[\int_0^\infty x e^{-x} B(\sqrt{x}/\xi)^2 dx + \frac{1}{4} \left(\int_0^\infty \sqrt{x} e^{-x} B(\sqrt{x}/\xi) dx \right)^2 \right], \quad (5.14)$$

where $W_{o,ion}(\xi) = (96\pi)^2 N_{ex} (e^2 D_1 / \kappa)^2 E_B^3 / \hbar \xi^2 E_g^{*4}$. For the parameters in Table I and for $N_{ex} = 5 \times 10^{10} \text{ cm}^{-2}$ and $E_g^* = 1.09 \text{ eV}$ corresponding to GaAs/Al_{0.3}Ga_{0.7}As, we estimate $W_{o,ion}(\xi) = 2.67 \times 10^9 \xi^{-2} \text{ sec}^{-1}$ where ξ is in Å. The calculated rate in Eq. (5.14) is plotted

in Fig. 9 as a function of ξ (lower axis) for a two-dimensional exciton density $N_{ex} = 5 \times 10^{10} \text{ cm}^{-2}$ and is independent of the temperature. The rate is in the range of TSK's data [3] which are shown as a function of T in the upper axis.

C. Phonon-assisted Exciton Transfer

In this section, we present a detailed analysis of the one-phonon-assisted Stokes and anti-Stokes rate through dipolar and exchange interactions. The anti-Stokes rate equals $\exp(-\beta\Delta)$ times the Stokes rate. We restrict the analysis only to the case where both the initial and final states are plane-wave exciton states for simplicity. The electron-phonon interaction is given in the j -th QW by [1]

$$\langle j, \mathbf{K}'_{\parallel} | H_{e-phn} | j, \mathbf{K}_{\parallel} \rangle = V_{j,sq} e^{iq_z z_j} \delta_{\mathbf{K}'_{\parallel} - \mathbf{K}_{\parallel}, \mathbf{q}_{\parallel}} (b_{sq} + b_{s,-q}^{\dagger}), \quad (5.15a)$$

$$V_{j,sq} = \Xi_{cq} H_j(\alpha_h \mathbf{q}_{\parallel}, q_z) - \Xi_{vq} H_j(-\alpha_e \mathbf{q}_{\parallel}, q_z), \quad (5.15b)$$

$$H_j(\mathbf{Q}_{\parallel}, Q_z) = \int d^2 r_{\parallel} \int dz_e \int dz_h | F_j(\mathbf{r}_{\parallel}, z_e, z_h) |^2 e^{i(Q_z z_e + \mathbf{Q}_{\parallel} \cdot \mathbf{r}_{\parallel})}, \quad (5.15c)$$

where $V_{j,sq}$ was introduced already in Eq. (4.9) and Ξ_{cq}, Ξ_{vq} are the electron-phonon coupling constants in the conduction and valence bands, respectively. Because the activation energy Δ is already larger than the L. O. phonon energy $\hbar\omega_0 = 36.2 \text{ meV}$, we expect L. O. phonons to play a dominant role. Acoustic phonons with weaker interaction with the electrons give subsidiary contributions. Therefore, we take $\Xi_{cq} = \Xi_{vq} = \Xi_{LOq}$, where

$$\Xi_{LOq} = \frac{\Xi_0}{q} = \frac{1}{q} \left(\frac{2\pi\hbar\omega_0 e^2}{\Omega\epsilon} \right)^{1/2}, \quad \frac{1}{\epsilon} = \frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_s}. \quad (5.16)$$

In the quasi-two-dimensional approximation of Eq. (2.8), we find

$$H_j(\mathbf{Q}_{\parallel}, Q_z) = \frac{64\rho_j(Q_z)}{(Q_{\parallel}^2 a_B^2 + 16)^{3/2}}, \quad \rho_j(Q_z) = \int dz \phi_j(z)^2 e^{iQ_z z}. \quad (5.17)$$

Here, symmetric QW's will be assumed with real $\rho_j(Q_z)$ for simplicity. Inserting Eqs.

(5.16) and (5.17) in Eq. (5.15b), we find

$$V_{j,sq} = V_{\parallel}(q_{\parallel})\rho_j(q_z)/q; \quad V_{\parallel}(q_{\parallel}) = 64\Xi_o\Lambda(q_{\parallel}), \quad (5.18)$$

$$\Lambda(q_{\parallel}) = \left[\frac{1}{(\alpha_h^2 q_{\parallel}^2 a_B^2 + 16)^{3/2}} - \frac{1}{(\alpha_e^2 q_{\parallel}^2 a_B^2 + 16)^{3/2}} \right],$$

where $V_{\parallel}(q_{\parallel})$ is a function of $q_{\parallel}$ only.

The one-phonon-assisted anti-Stokes rate is given after inserting Eqs. (5.18) and (4.11) in Eq. (4.10) by

$$W = \frac{2\pi}{\hbar Z_1 \Delta^2} \sum_{\mathbf{K}_{\parallel}} e^{-\beta E_{\mathbf{K}_{\parallel}}} \sum_{\mathbf{K}_{\parallel}, \mathbf{q} \pm} [|t_{\mathbf{K}_{\parallel}}|^2 \rho_1(q_z)^2 + |t_{\mathbf{K}_{\parallel}}|^2 \rho_2(q_z)^2 - \rho_1(q_z)\rho_2(q_z)] \quad (5.19)$$

$$\times (t_{\mathbf{K}_{\parallel}}^* t_{\mathbf{K}_{\parallel}} e^{iq_z d} + c.c.)] (V_{\parallel}(q_{\parallel})/q)^2 (n_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}) \delta_{\mathbf{K}_{\parallel} - \mathbf{K}_{\parallel}, \mathbf{q}_{\parallel}} \delta(E_{\mathbf{K}_{\parallel}} + \Delta \pm \hbar\omega_o - E_{\mathbf{K}_{\parallel}}),$$

where $\mathbf{q}_{\parallel}$ is replaced by $-\mathbf{q}_{\parallel}$ for the emission process.

At this point, we approximate

$$\rho_j(q_z)^2 = \frac{\delta_j^2}{q_z^2 + \delta_j^2}, \quad (5.20)$$

which has a correct limit $\rho_j(0)^2 = 1$ and a width δ_j of the order of π/b_j . [18] We also

employ a similar Lorentzian form for $\rho_1(q_z)\rho_2(q_z)$ with a width δ' . We then find

$$\int_{-\infty}^{\infty} dq_z \frac{e^{\pm i q_z d}}{q_z^2 + q_{\parallel}^2} \frac{\delta_j^2}{q_z^2 + \delta_j^2} = \frac{\pi \delta_j^2}{\delta_j^2 - q_{\parallel}^2} \left(\frac{e^{-q_{\parallel} d}}{q_{\parallel}} - \frac{e^{-\delta_j d}}{\delta_j} \right) \rightarrow \frac{\pi}{q_{\parallel}} \eta_j(q_{\parallel}) e^{-q_{\parallel} \eta_j(q_{\parallel}) d}, \quad (5.21)$$

where the arrow indicates the following asymptotic behavior: $\eta_j(q_{\parallel}) = \delta_j / q_{\parallel}$ for $\delta_j \ll q_{\parallel}$ and $\eta_j(q_{\parallel}) = 1$ for $\delta_j \gg q_{\parallel}$. A similar asymptotic relationship holds for the $\rho_1(q_z)\rho_2(q_z)$ integral with $\eta'(q_{\parallel}) = \delta' / q_{\parallel}$ for $\delta' \ll q_{\parallel}$ and $\eta'(q_{\parallel}) = 1$ for $\delta' \gg q_{\parallel}$. Note that $d = 0$ in Eq. (5.21) for the first two terms in Eq. (5.19). It turns out that the third term ($\propto \rho_1(q_z)\rho_2(q_z)$) is negligibly small in the limit $d \gg b_j$.

The dominant rate arises from the absorption process (corresponding to the lower sign) in Eq. (5.19). Approximating $n_{\mathbf{q}} = e^{-\beta \hbar \omega_{\mathbf{q}}}$, carrying out the $\mathbf{K}_{\parallel}$ and q_z -summations, we obtain

$$W = \frac{e^{-\beta \Delta} \Omega M}{\hbar^3 Z_1 \Delta^2} < \sum_{\mathbf{K}_{\parallel}} \frac{1}{2 q_{\parallel}} e^{-\beta E_{\mathbf{K}_{\parallel}}} [|t_{\mathbf{K}_{\parallel}}|^2 \eta_1(q_{\parallel}) + |t_{\mathbf{K}_{\parallel}}|^2 \eta_2(q_{\parallel})] \quad (5.22)$$

$$-(t_{\mathbf{K}_{\parallel}}^* t_{\mathbf{K}_{\parallel}} + c.c.) \eta'(q_{\parallel}) e^{-q_{\parallel} \eta'(q_{\parallel}) d} V_{\parallel}(q_{\parallel})^2 >_{\mathbf{K}_{\parallel}},$$

where $q_{\parallel} = \mathbf{K}_{\parallel}' - \mathbf{K}_{\parallel}$. The energy conservation condition $E_{\mathbf{K}_{\parallel}} + \hbar \omega_0 = \Delta + E_{\mathbf{K}_{\parallel}}'$ indicates that this process is available only to those high-energy excitons in QW1 which can reach the QW2 exciton band with one-phonon absorption. The analysis has been general so far. For large Δ , we ignore $E_{\mathbf{K}_{\parallel}}' \sim k_B T \ll \Delta - \hbar \omega_0$ at low temperatures and approximate $E_{\mathbf{K}_{\parallel}} + \hbar \omega_0 = \Delta + E_{\mathbf{K}_{\parallel}}' \approx \Delta$, $q_{\parallel} \approx K_{\parallel} \approx K_{\parallel}^* \equiv [2M(\Delta - \hbar \omega_0)/\hbar^2]^{1/2} = 1/27.8 \text{ \AA}$ in Eq. (5.22).

C1. Dipolar phonon-assisted exciton transfer

Employing the expression for the dipolar exchange interaction in Eq. (3.9a) for $t_{\mathbf{K}_{\parallel}}$, per-

forming angular averages such as given in Eq. (3.12) and adding $\mathbf{D}_1 \parallel \mathbf{D}_2$ and $\mathbf{D}_1 \perp \mathbf{D}_2$, we rewrite Eq. (5.22) as

$$W = \frac{W_{LO}^{dip} e^{-\beta\Delta}}{K_{\parallel}^{*2} Z_1} \sum_{\mathbf{K}_{\parallel}'} e^{-\beta E_{\mathbf{K}_{\parallel}'}} [K_{\parallel}'^2 \eta_1(K_{\parallel}^*) \langle e^{-K_{\parallel}' |R_z|} \rangle^2 + K_{\parallel}^{*2} \langle e^{-K_{\parallel}^* |R_z|} \rangle^2 \eta_2(K_{\parallel}^*) \quad (5.23a)$$

$$- K_{\parallel}' K_{\parallel}^* \eta'(K_{\parallel}^*) \langle e^{-K_{\parallel}' |R_z|} \rangle \langle e^{-K_{\parallel}^* |R_z|} \rangle e^{-K_{\parallel}' \eta'(K_{\parallel}^*) d}],$$

$$W_{LO}^{dip} = \frac{2\pi 64^3 M \omega_o K_{\parallel}^*}{\hbar^2 \Delta^2 \epsilon'} \left(\frac{e^3 D_1 D_2}{\kappa a_B^2} \right)^2 \Lambda(K_{\parallel}^*)^2. \quad (5.23b)$$

Using $\epsilon^{-1} = 0.012$ for GaAs [19] and the parameters listed in Table I, we estimate $W_{LO}^{dip} = 3.87 \times 10^8 \text{ sec}^{-1}$. The second and third terms in Eq. (5.23) decay exponentially as $\propto \exp(-2K_{\parallel}' d) = \exp(-d/13.9 \text{ \AA})$ and are negligibly small for large d as confirmed by our numerical analysis. The dominance of the first term means that the excitons prefer to cross the barrier with a small momentum $K_{\parallel}'$ to maximize $|t_{\mathbf{K}_{\parallel}'}|^2 \propto \exp(-2K_{\parallel}' d)$. This is achieved by being scattered inside the initial QW from the initial high-momentum state $\mathbf{K}_{\parallel} = \mathbf{K}_{\parallel}^*$ to a low-momentum intermediate state $\mathbf{K}_{\parallel}'$ before crossing the barrier. Using the same argument for the back (i.e., Stokes) transfer from QW2 to QW1, the exciton-phonon scattering occurs in the lower-energy QW1 after crossing the barrier with a small initial momentum $K_{\parallel}'$.

Retaining only the first term in Eq. (5.23a), carrying out the $\mathbf{K}_{\parallel}'$ -summation and employing the approximation given in Eq. (3.11), we find

$$W = W_{LO}^{dip} e^{-\beta\Delta} \frac{2k_B T}{E_{K_{\parallel}^*}} \eta_1(K_{\parallel}^*) g(d / \xi_T), \quad (5.24)$$

where $E_{K_{\parallel}^*} = \Delta - \hbar\omega_0$ and the function $g(t)$ was defined in Eq. (3.15a). The one-phonon - assisted Stokes rate due to phonon emission can be derived in a similar way and has the same expression as that in Eqs. (5.23) and (5.24) without the activation factor $e^{-\beta\Delta}$. Comparing with Eq. (3.15), the phonon-assisted Stokes rate $e^{\beta\Delta}W$ has exactly the same temperature dependence as the dipolar Stokes energy transfer rate from plane-wave exciton states to free electron-hole pairs. The factor $\eta_1(K_{\parallel}^*)$ indicates 1) that phonon absorption occurs in the initial QW1 and 2) that the rate is faster rate for a smaller well width as well known. [18] For numerical estimate we take $\eta_1(K_{\parallel}^*) = 1$ corresponding to the narrow QW limit. Figure 10 displays the temperature dependence of $e^{\beta\Delta}W$ (i.e., the Stokes rate) for $d = 175$ and $375 \text{ \AA}$ for $b_1 = 100 \text{ \AA}$ and $b_2 = 50 \text{ \AA}$.

C2. Radiative phonon-assisted exciton transfer

Photon-exchange coupling between $|1, K_{\parallel}\rangle$ and $|2, K_{\parallel}'\rangle$ is given by

$$t_{K_{\parallel}} = \sum_{\lambda k} \frac{\langle 2, K_{\parallel}, N_{\lambda k} | H_{ph} | 2, 0, N_{\lambda k} + 1 \rangle \langle 1, 0, N_{\lambda k} + 1 | H_{ph} | 1, K_{\parallel}, N_{\lambda k} \rangle}{E_g - \hbar\omega_{\lambda k} - i\Gamma}, \quad (5.25)$$

yielding

$$t_{K_{\parallel}} = J_{rad} Q(K_{\parallel}, d); \quad J_{rad} = \frac{16E_g e^2 D_1 D_2}{\hbar \pi c n a_B^2}. \quad (5.26)$$

The quantity $Q(K_{\parallel}, d)$ is the same as given in Eq. (3.27) but is different from the expression in Eq. (3.25) where the final state is free electron-hole state.

Using Eq. (3.28) and summing over $\mathbf{D}_1 \parallel \mathbf{D}_2$ and $\mathbf{D}_1 \perp \mathbf{D}_2$, we can show the following

relationship for $Q(\mathbf{K}_{\parallel}, d)$:

$$\sum_{\parallel+1} \langle Q(\mathbf{K}_{\parallel}, d)^2 \rangle_{\mathbf{K}_{\parallel}} = \frac{1}{2} (|I_0(x)|^2 + |I_1(x)|^2), \quad (5.27a)$$

$$\sum_{\parallel+1} \langle Q(\mathbf{K}_{\parallel}, d) \rangle_{\mathbf{K}_{\parallel}} = \frac{1}{2} (I_0(x) + I_1(x)), \quad (5.27b)$$

where $x = (K_{\parallel}d)^2$ and $I_n(x)$ was defined in Eq. (3.30). The quantities on the left hand sides of Eq. (5.27) appears in Eq. (5.22) when $t_{\mathbf{K}_{\parallel}}$ is replaced by $= J_{rad} Q(\mathbf{K}_{\parallel}, d)$.

The second and third terms in Eq. (5.22) contain terms proportional to $I_n(x^*)^2$ and $I_n(x^*)$, respectively, where $x^* = (K_{\parallel}^* d)^2$. We now show that these quantities are negligibly small. For this purpose, we note that $K_{\parallel}^* \xi_g = 12.7 \gg 1$ for the present parameters, allowing us to expand the second term in Eq. (3.30) in $1/K_{\parallel}^* \xi_g$, yielding

$$I_n(x^*) = -\frac{d}{\xi_g} \int_0^{\infty} \frac{z^{2n} \cos z}{[x^* + z^2]^{n+1}} dz. \quad (5.28)$$

We find $I_0(x^*) = -\pi \exp(-K_{\parallel}^* d) / (2K_{\parallel}^* \xi_g)$ and $I_1(x^*) = \pi d (1 - 1/K_{\parallel}^* d) \exp(-K_{\parallel}^* d) / (4\xi_g)$.

Both of these quantities are exponentially small in view of $K_{\parallel}^* d = d / 27.8 \text{ \AA} \gg 1$. This means that photon-exchange interaction is small for a large momentum transfer. We therefore retain only the first term in Eq. (5.22).

Using Eq. (5.27) and following the same procedure employed for dipolar transition, we find

$$W = \frac{W_{LO}^{rad} e^{-\beta \Delta}}{2\pi^2} \int_0^{\infty} dx e^{-x} (|I_0(xd^2 / \xi_T^2)|^2 + |I_1(xd^2 / \xi_T^2)|^2), \quad (5.29)$$

$$W_{LO}^{rad} = \frac{64^2 \pi^3 J_{rad}^2 M \omega_o e^2}{\hbar^2 \Delta^2 K_{\parallel}^* e'} \Lambda(K_{\parallel}^*).$$

For the parameters in Table I, we estimate $W_{LO}^{rad} = 3.99 \times 10^6 \text{ sec}^{-1}$. The expression $e^{\beta\Delta}W$ in Eq. (5.29) has exactly the same form as that in Eq. (3.33) except for the rate constant W_{LO}^{rad} . The one-phonon-assisted radiative Stokes transfer rate is displayed in Fig. 11 as a function of T . The rate has the same T and d dependences as the photon-exchange Stokes rate for excitons to transfer into free electron-hole pairs displayed in Figs. 5 and 6 except that it is smaller by a factor $W_{LO}^{rad} / W_0 = 1.10 \times 10^{-2}$. Nevertheless, it is about one order of magnitude faster than the phonon-assisted dipolar Stokes rate plotted in Fig. 10. The anti-Stokes rate is further smaller by a factor $e^{-\beta\Delta}$.

VI. CONCLUSIONS AND REMARKS

We have studied Stokes and anti-Stokes exciton energy transfer between two QW's separated by a wide barrier and with a large energy mismatch $\Delta \gg k_B T, E_B$. Several important intrinsic energy transfer mechanisms such as dipolar coupling, real and virtual photon-exchange and over-barrier ionization of the excitons through exciton-exciton Auger processes have been examined. Phonon-assisted transfer rates were found to be too small to explain the data.

For Stokes energy transfer, dominant energy transfer occurs through the decay of excitons in the higher-energy QW into free electron-hole pairs in the lower-energy QW. We found that energy transfer through photon-exchange interaction is much faster than the dipolar transfer rate except at a very short distance d . The radiative rate has a very slow dependence on d , decaying logarithmically at large d in contrast with the rapid d^4 dependence of the dipolar rate. For plane-wave excitons, the rapid temperature dependence of the radiative rate displayed in Fig. 5 is inconsistent with TSK's data. For localized excitons, the rate (shown in Fig. 3) is independent of T if the localization radius is independent

of T , yielding better agreement with the data. The excitons may indeed be localized in the initial narrow QW, where the wave functions can be localized even by small layer fluctuations.

For anti-Stokes transfer through dipolar as well as photon-exchange coupling, we showed that thermal activation is essential for the energy transfer to occur.

Phonon-assisted rate was calculated for this case using photon-exchange and dipolar coupling, yielding a negligibly small rate and an activated T -dependence $\propto \exp(-\Delta/k_B T)$. On the other hand, energy transfer through over-barrier ionization via Auger processes yielded a significantly larger nonactivated rate which is independent of d . This rate is also independent of T for localized excitons and is large enough to explain TSK's data [3] as shown in Figs. 8 and 9 if the exciton densities are assumed to be in the range $10^{10} - 10^{11} \text{ cm}^{-2}$. The temperature dependence of the rate predicted by the plane-wave exciton model seems to be consistent with the data, suggesting that the excitons in the initial wide QW may be delocalized.

The theoretical predictions of this paper can be checked by measuring the transfer rate between two QW's separated by an extremely large distance. The radiative Stokes transfer and anti-Stokes transfer through over-barrier exciton ionization are effective over a long range. The ionization rate is proportional to the exciton density.

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Appendix A

In this appendix, we derive Eq. (3.8), namely we evaluate

$$C(\mathbf{K}_{\parallel}, R_z) = \frac{e^2}{\kappa} \int d^2 R_{\parallel} e^{i\mathbf{K}_{\parallel} \cdot \mathbf{R}_{\parallel}} \frac{1}{(R_{\parallel}^2 + R_z^2)^{3/2}} [\mathbf{D}_1 \cdot \mathbf{D}_2 - 3(\hat{\mathbf{R}}_{\parallel} \cdot \mathbf{D}_1)(\hat{\mathbf{R}}_{\parallel} \cdot \mathbf{D}_2)]. \quad (\text{A.1})$$

Assuming $\mathbf{D}_1 \parallel \mathbf{x}$, defining $\phi_{\mathbf{K}_{\parallel}}$ (ϕ_D) as the angle between $\mathbf{K}_{\parallel}$ ($\mathbf{D}_2$) and the x-axis, and θ as the angle between $\mathbf{K}_{\parallel}$ and $\mathbf{R}_{\parallel}$, we can rewrite Eq. (A.1) as

$$C(\mathbf{K}_{\parallel}, R_z) = \frac{e^2}{\kappa} \int_0^{\infty} R_{\parallel} dR_{\parallel} \int_{-\pi}^{\pi} d\theta e^{iK_{\parallel} R_{\parallel} \cos \theta} \frac{D_1 D_2}{(R_{\parallel}^2 + R_z^2)^{3/2}} [\cos \phi_D - 3\psi], \quad (\text{A.2})$$

where $\psi = \cos(\theta - \phi_{\mathbf{K}_{\parallel}}) \cos(\theta - \phi_{\mathbf{K}_{\parallel}} + \phi_D)$. Rewriting $\psi = [\cos(2\theta - 2\phi_{\mathbf{K}_{\parallel}} + \phi_D) + \cos \phi_D]/2 = [\cos(2\theta) \cos(2\phi_{\mathbf{K}_{\parallel}} - \phi_D) + \sin(2\theta) \sin(2\phi_{\mathbf{K}_{\parallel}} - \phi_D) + \cos \phi_D]/2$, dropping the $\sin(2\theta)$ term and transforming the θ integrals into Bessel functions, we find

$$C(\mathbf{K}_{\parallel}, R_z) = \pi D_1 D_2 \frac{e^2}{\kappa} \int_0^{\infty} r dr \left[\frac{2J_0(K_{\parallel} r) \cos \phi_D}{(r^2 + R_z^2)^{3/2}} - \frac{3r^2 J_0(K_{\parallel} r) \cos \phi_D}{(r^2 + R_z^2)^{5/2}} + \frac{3r^2 J_2(K_{\parallel} r) \cos(2\phi_{\mathbf{K}_{\parallel}} - \phi_D)}{(r^2 + R_z^2)^{5/2}} \right]. \quad (\text{A.3})$$

To proceed further, it is useful to employ the following identity: [11]

$$\int_0^{\infty} \frac{x^{n+1} J_n(bx)}{(x^2 + a^2)^{n+1/2}} dx = \frac{\sqrt{\pi} b^{n-1} e^{-ab}}{2^n \Gamma(n+1/2)}, \quad (\text{A.4})$$

where $\Gamma(x)$ is the gamma function. The first term of Eq. (A.3) can be evaluated by setting n

= 0 in Eq. (A.4) and taking a derivative with respect to a on both sides, yielding

$$\int_0^{\infty} r dr \left[\frac{J_0(K_{\parallel} r)}{(r^2 + R_z^2)^{3/2}} \right] = e^{-K_{\parallel} R_z} / R_z, \quad (\text{A.5a})$$

where $R_z = |R_z|$. The second term of Eq. (A.3) is evaluated by rewriting $r^2 = (r^2 + R_z^2) - R_z^2$, using Eq. (A.5a) and the identity obtained by taking a first derivative of both sides of Eq. (A.5a) with respect to R_z , yielding

$$\int_0^{\infty} r dr \frac{r^2 J_0(K_{\parallel} r)}{(r^2 + R_z^2)^{5/2}} = (2 - K_{\parallel} R_z) e^{-K_{\parallel} R_z} / 3 R_z. \quad (\text{A.5b})$$

Finally, the third term of Eq. (A.3) is evaluated by using $n = 2$ in Eq. (A.4), yielding

$$\int_0^{\infty} r dr \frac{r^2 J_2(K_{\parallel} r)}{(r^2 + R_z^2)^{5/2}} = K_{\parallel} e^{-K_{\parallel} R_z} / 3. \quad (\text{A.5c})$$

Eq. (3.8) is then obtained by inserting Eqs. (A.5) in Eq. (A.3).

Appendix B

In this appendix, we derive Eq. (3.28). Introducing the polar angles θ, θ' for $\mathbf{K} = (\mathbf{K}_{\parallel}', k_z)$ and $\mathbf{K}' = (\mathbf{K}_{\parallel}', k_z')$ and the azimuthal angle ϕ for $\mathbf{K}_{\parallel}'$, we find $\hat{\mathbf{K}} = \sin\theta(\cos\phi, \sin\phi, \cot\theta)$ and choose the two perpendicular polarization vectors $\hat{\mathbf{e}}_{1\mathbf{K}} = (\sin\phi, -\cos\phi, 0)$, $\hat{\mathbf{e}}_{2\mathbf{K}} = \sin\theta(-\cot\theta\cos\phi, -\cot\theta\sin\phi, 1)$ and let $\hat{\mathbf{D}}_1 = (1, 0, 0)$. For the case $\hat{\mathbf{D}}_1 = \hat{\mathbf{D}}_2$, we find $P(\mathbf{K}_{\parallel}', k_z) = \sin^2\phi + \cos^2\phi\cos^2\theta$. Similarly, $P(\mathbf{K}_{\parallel}', k_z') = \sin^2\phi + \cos^2\phi\cos^2\theta'$. The average over ϕ

yields

$$\frac{1}{2\pi} \int_0^{2\pi} d\phi_{\mathbf{K}_\perp} P(\mathbf{K}'_{\parallel}, k'_z) P(\mathbf{K}'_{\parallel}, k_z) = \frac{1}{8} (3 + \cos^2 \theta + \cos^2 \theta' + 3 \cos^2 \theta \cos^2 \theta'). \quad (\text{B.1a})$$

For the case $\hat{\mathbf{D}}_2 = (0, 1, 0)$, we find $P(\mathbf{K}'_{\parallel}, k_z) = -\sin^2 \theta \sin \phi \cos \phi$. The average over ϕ

yields

$$\frac{1}{2\pi} \int_0^{2\pi} d\phi_{\mathbf{K}_\perp} P(\mathbf{K}'_{\parallel}, k'_z) P(\mathbf{K}'_{\parallel}, k_z) = \frac{1}{8} \sin^2 \theta \sin^2 \theta'. \quad (\text{B.1b})$$

Adding Eqs. (B.1a) and (B.1b), we find

$$\frac{1}{2\pi} \int_0^{2\pi} d\phi_{\mathbf{K}_{\parallel+\perp}} \sum P(\mathbf{K}'_{\parallel}, k'_z) P(\mathbf{K}'_{\parallel}, k_z) = \frac{1}{2} [1 + \cos^2 \theta \cos^2 \theta'] = \frac{1}{2} [1 + \frac{k_z^2 k_z'^2}{K^2 K'^2}], \quad (\text{B.2})$$

which is the relationship given in Eq. (3.28).

Appendix C

In this appendix, we study the asymptotic behavior of the radiative rate for $d \rightarrow \infty$.

Inserting $x = (K_{\parallel} \xi)^2$, we rewrite Eqs. (3.29) and (3.30) as

$$W = \frac{\xi^2 W_{o,rad}}{\pi^2} \int_0^{\infty} dK_{\parallel} K_{\parallel} e^{-(K_{\parallel} \xi)^2} \theta(1/\xi_{\Delta}^2 - K_{\parallel}^2) (|I_0(K_{\parallel}^2 d^2)|^2 + |I_1(K_{\parallel}^2 d^2)|^2), \quad (\text{C. 1})$$

$$I_n(K_{\parallel}^2 d^2) = \frac{1}{2} \int_{-\infty}^{\infty} dk_z e^{ik_z d} \left(\frac{k_z}{k}\right)^{2n} \left\{ \frac{1}{k} + \frac{1}{\xi_g^{-1} - k - i\gamma/d} \right\}, \quad (\text{C. 2})$$

for localized excitons where $k = (k_{\parallel}^2 + k_z^2)^{1/2}$ and $k_{\parallel} = K_{\parallel}$. For plane-wave excitons, $\xi = \xi_T$

and the step function θ is to be replaced by unity in Eq. (C.1). Introducing the identity

$$\frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} d^2 r_{\parallel} e^{i\mathbf{r}_{\parallel} \cdot (\mathbf{k}_{\parallel} - \mathbf{K}_{\parallel})} = \delta(\mathbf{K}_{\parallel} - \mathbf{k}_{\parallel}), \quad (\text{C.3})$$

Eq. (C.2) is then recast into

$$I_n(K_{\parallel}^2 d^2) = \frac{1}{8\pi^2} \int d^2 r_{\parallel} e^{-i\mathbf{r}_{\parallel} \cdot \mathbf{K}_{\parallel}} \int_{-\infty}^{\infty} d^3 k e^{i\mathbf{k} \cdot \mathbf{r}} \left(\frac{k_z}{k}\right)^{2n} \left\{ \frac{1}{k} + \frac{1}{\xi_g^{-1} - k - i\gamma/d} \right\}, \quad (\text{C.4})$$

where $\mathbf{r} = (\mathbf{r}_{\parallel}, d)$. Carrying out the $\mathbf{k}$ -integration in the polar coordinate and also the $\mathbf{r}_{\parallel}$ -integration in the cylindrical coordinate, we find

$$I_n(K_{\parallel}^2 d^2) = \frac{1}{\xi_g} \int_0^{\infty} dr_{\parallel} J_0(K_{\parallel} r_{\parallel}) \frac{r_{\parallel}}{r} \left(\frac{d}{r}\right)^{2n} \int_0^{\infty} dx \frac{\sin x}{r \xi_g^{-1} - x - i\eta}, \quad (\text{C.5})$$

where $\eta = \gamma r/d = r/(\tau c)$, $\tau = \hbar/\Gamma$ and the factor $(d/r)^{2n}$ arises from $(k_z/k)^{2n}$ in the limit $d \rightarrow \infty$. The upper limit of the quantity η is the ratio of the sample size to the distance traversed by light during the lifetime and is assumed to be very small (i.e., $\eta \ll 1$). The x -integral in Eq. (C.5) can be expressed in terms of $\text{ci}(k_g r)$ and $\text{si}(k_g r)$ functions in the limit $\eta \rightarrow 0$, yielding $-\pi e^{-k_g r}$ for $k_g r \rightarrow \infty$, where $k_g = \xi_g^{-1}$. [5] Therefore, Eq. (C.5) can be rewritten in these limits as

$$I_n(K_{\parallel}^2 d^2) = -\pi k_g \int_0^{\infty} dr_{\parallel} J_0(K_{\parallel} r_{\parallel}) \frac{r_{\parallel}}{r} \left(\frac{d}{r}\right)^{2n} e^{-ik_g r}. \quad (\text{C.6})$$

When d is much larger than the QW radius, we expand $r = d + r_{\parallel}^2/2d$. Inserting this result in the exponent of Eq. (C.6) and replacing $r = d$ elsewhere, we find [11]

$$I_n(K_{\parallel}^2 d^2) = i\pi e^{-ik_g d} e^{iK_{\parallel}^2 d/2k_g}. \quad (\text{C.7})$$

Finally, a d -independent asymptotic rate $W = W_{\text{orad}}$ is obtained when Eq. (C.7) is inserted

in Eq. (C.1). Here, the condition $\xi > \xi_\Delta$ is assumed for localized excitons.

In other cases, we change the variable from $r_\parallel$ to r in Eq. (C.6) obtaining

$$I_n(K_\parallel^2 d^2) = -\pi k_g \int_0^\infty dr J_n(K_\parallel \sqrt{r^2 - d^2}) \left(\frac{d}{r}\right)^{2n} e^{-ik_g r}. \quad (C.8)$$

Hereafter, we study only the contribution from $|I_0|^2$ and show that the rate does not vanish for $d \rightarrow \infty$ but approaches a lower bound slowly. Note also that $d/r \leq 1$ in Eq. (C.8) for I_1 and the contribution from $|I_1|^2$ does not alter this conclusion. The integration in Eq. (C.8) is given by [11]

$$\begin{aligned} I_0(K_\parallel^2 d^2) &= \pi i k_g e^{-id\sqrt{k_g^2 - K_\parallel^2}} / \sqrt{k_g^2 - K_\parallel^2}, \quad k_g > K_\parallel \\ &= -\pi k_g e^{-d\sqrt{K_\parallel^2 - k_g^2}} / \sqrt{K_\parallel^2 - k_g^2}, \quad k_g < K_\parallel \end{aligned} \quad (C.9)$$

Inserting this result in Eq. (C.1) and going back to the original variable $x = (K_\parallel \xi)^2$ employed in Eqs. (3.29) and (3.30), we find

$$\begin{aligned} W &= \frac{1}{2} \xi^2 k_g^2 W_{o,rad} \left[\int_0^{(\xi k_g)^2} dx \frac{e^{-x} \theta(\xi^2 / \xi_\Delta^2 - x)}{\xi^2 k_g^2 - x + \gamma} \right. \\ &\quad \left. + \int_{(\xi k_g)^2}^\infty dx \frac{e^{-x} e^{-(2d/\xi)\sqrt{x - \xi^2 k_g^2}} \theta(\xi^2 / \xi_\Delta^2 - x)}{x - \xi^2 k_g^2 + \gamma} \right]. \end{aligned} \quad (C.10)$$

Here, γ accounts for the Lorentzian width and prevents a weak logarithmic divergence at the singularity point $x = (k_g \xi)^2$ as in Eq. (3.29). The second term in Eq. (C.10) vanishes as $(\xi/d)^2/\gamma$ for $d \rightarrow \infty$ and is dropped. The first term can be rewritten in the limit $\gamma \rightarrow 0$ as

$$W = \frac{1}{2} (\xi k_g)^2 W_{o,rad} e^{-(\xi k_g)^2} \left[\int_0^{(\xi k_g)^2} \frac{e^t - 1}{t} dt + \ln \frac{(\xi k_g)^2}{\gamma} \right]. \quad (C.11)$$

Here $\xi > \xi_\Delta$. In view of the fact that $\gamma = dl/(\tau \tilde{c})$ ($\ll 1$), the second term decreases logarithmically.

mically as a function of d while the first term is independent of d .

Appendix D

In this appendix, we show the equivalence of Eq. (4.3) and Eq. (4.2). Inserting the eigenstates and the eigenvalues defined by $E_m H|m\rangle = E_m|m\rangle$ in Eq. (4.3) and performing the u -integration, we find

$$\mathcal{F}(\omega_r) = \frac{1}{Z} \sum_{mn} |\langle n|\hat{t}|m\rangle|^2 \frac{e^{-\beta E_m} - e^{-\beta E_n}}{E_n - E_m + \omega_r}, \quad (\text{D.1})$$

where Z is the distribution function and use is made of $\exp(\beta\omega_r) = 1$ for $\omega_r = 2\pi ir/\beta$. Here r is an integer. Analytically continuing ω_r to $\Omega + i0$, we obtain

$$W = \frac{2}{\hbar} \text{Im} \mathcal{F}(\Omega + i0) = \frac{2\pi}{\hbar Z} \sum_{mn} [e^{-\beta E_n} |\langle m|\hat{t}^\dagger|n\rangle|^2 - e^{-\beta E_m} |\langle n|\hat{t}|m\rangle|^2] \times \delta(E_n - E_m + \Omega), \quad (\text{D.2})$$

which is identical to Eq. (4.2).

Appendix E

In this appendix, we show that the second term of Eq. (4.11) yields the expression in the second term of Eq. (4.5). Inserting the second term of Eq. (4.11) in Eq. (4.10), we find

$$W = \frac{2\pi}{\hbar Z_1} \sum_{\mathbf{K}_\parallel} e^{-\beta E_{1\mathbf{K}_\parallel}} \sum_{\mathbf{K}'_\parallel, s\mathbf{q}\pm} \left| \frac{t_{\mathbf{K}_\parallel} V_{1,s\mathbf{q}}}{\Delta} \right|^2 (n_{s\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}) \delta_{\mathbf{K}_\parallel, \mathbf{K}'_\pm \mathbf{q}_\parallel} \delta(E_{2\mathbf{K}'_\pm} \pm \hbar\omega_{s\mathbf{q}} - E_{1\mathbf{K}_\parallel}). \quad (\text{E.1})$$

We now replace $E_{1\mathbf{K}_\parallel}$ in the exponent by $E_{1\mathbf{K}_\parallel} = E_{2\mathbf{K}'_\pm} \pm \hbar\omega_{s\mathbf{q}}$ and interchange $\mathbf{K}_\parallel \leftrightarrow \mathbf{K}'_\pm$. Using the identity $\exp(\mp \beta \hbar \omega_{s\mathbf{q}})(n_{s\mathbf{q}} + 1/2 \pm 1/2) = n_{s\mathbf{q}} + 1/2 \mp 1/2$ and Eq. (4.8), W can

be rewritten as

$$W = \frac{2}{\hbar \Delta^2 Z_{1\mathbf{K}_{\parallel}}} \sum |t_{\mathbf{K}_{\parallel}}|^2 e^{-\beta E_{2\mathbf{K}_{\parallel}}} \Gamma_{1\mathbf{K}_{\parallel}}(E_{2\mathbf{K}_{\parallel}}), \quad (\text{E.2})$$

which is identical to the second term of Eq. (4.5).

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Table 1 Sample parameters for the GaAs/Al_{0.3}Ga_{0.7}As quantum wells employed in the text.

electron mass m_e	$0.067m_0$	dielectric constant κ	12.4
hole mass m_h	$0.14m_0$	exciton binding energy E_B	4.06 meV
total mass M	$0.207m_0$	dipole moment $D_1 = D_2$	5.5 Å
band gap E_g	1.52 eV	energy mismatch Δ	60.0 meV
refractive index n	3.68	L.O. phonon energy	36.2 meV

Figure Captions

- Fig. 1 Dipolar Stokes transfer rate for plane-wave excitons from a 50-Å QW to a 100-Å QW separated by $d = 175$ and 375 Å with a 60-meV energy mismatch. The dotted curve is for the 2D limit.
- Fig. 2 Dipolar Stokes transfer rate for localized excitons from a 50-Å QW to a 100-Å QW separated by $d = 175$ and 375 Å with a 60-meV energy mismatch. The dotted curve is for the 2D limit.
- Fig. 3 Photon-exchange Stokes transfer rate for localized excitons from a narrow to a wide QW separated by (a) $d = 175$ and (b) $d = 175 - 375$ Å with a 60-meV energy mismatch. The dotted curve is for the 2D limit. Contributions from the resonant and nonresonant processes are shown in (a). Temperature-dependent data (symbols) of TSK [3] are shown in (b) in the upper axis.
- Fig. 4 Photon-exchange Stokes transfer rate as a function of the well-to-well separation d for localized excitons from a narrow to a wide QW with a 60-meV energy mismatch for the exciton localization radius $\xi = 150, 400$ and 1500 Å.
- Fig. 5 Photon-exchange Stokes transfer rate as a function of the temperature for plane-wave excitons from a narrow to a wide QW separated by $d = 175$ and 375 Å with a 60-meV energy mismatch. The symbols indicate the data from TSK. [3]
- Fig. 6 Photon-exchange Stokes transfer rate as a function of the well-to-well separation d for plane-wave excitons from a narrow to a wide QW with a 60-meV energy mismatch for $T = 4, 10$ and 50 K.

Fig. 7 Basic diagrams for the correlation function defined in Eq. (4.3) for the exciton transfer rate. The solid lines are dressed exciton propagators and the wave line a phonon propagator. The arrows indicate the direction of momentum and energy flow. The bubble diagram (a) yields the result in Eq. (4.5) and the one-rung diagram (b) the interference term in Eq. (4.12).

Fig. 8 Over-barrier ionization rate of plane-wave excitons through Auger-like two-exciton processes from a QW with $b = 50$ (dotted curve) and $100 \text{ \AA}$ (solid curve).

Symbols represent TSK's anti-Stokes rate data from samples with $b = 100 \text{ \AA}$. [3]

Fig. 9 Temperature-independent over-barrier ionization rate of localized excitons through Auger-like two-exciton processes from $50\text{-\AA}$ (dotted curve) and $100\text{-\AA}$ (solid curve) QW's. Symbols represent TSK's T -dependent anti-Stokes rate data (upper axis). [3]

Fig. 10 L.O.-phonon-assisted dipolar Stokes transfer rate for plane-wave excitons from a $50\text{-\AA}$ QW to a $100\text{-\AA}$ QW separated by $d = 175$ and $375 \text{ \AA}$ with $\Delta = 60\text{-meV}$ energy mismatch. The dotted curve is for the 2D limit. The anti-Stokes rate is obtained by multiplying the Stokes rate by $\exp(-\beta\Delta)$.

Fig. 11 L.O.-phonon-assisted photon-exchange Stokes transfer rate for plane-wave excitons from a $50\text{-\AA}$ QW to a $100\text{-\AA}$ QW separated by $d = 175$ and $375 \text{ \AA}$ with $\Delta = 60\text{-meV}$ energy mismatch. The dotted curve is for the 2D limit. The anti-Stokes rate is obtained by multiplying the Stokes rate by $\exp(-\beta\Delta)$.

Fig. 1

Fig. 2

Photon-Exchange Stokes Transfer Rate

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

Fig. 11