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Localization and homogeneous dephasing relaxation of quasi-two-dimensional excitons in quantum-well heterostructures

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The mechanisms of dephasing relaxation (homogeneous linewidth) of quasi-two-dimensional excitons in quantum-well heterostructures are clarified for both localized and delocalized excitons. The recently observed energy and temperature dependences of the homogeneous linewidth Γ_h are explained quantitatively. Furthermore, a new exponent for the temperature dependence of Γ_h of the localized excitons at low temperatures, the energy dependence of Γ_h of the delocalized excitons, and the dependence of Γ_h on the quantum-well thickness are predicted.

Recently, the homogeneous linewidth and the diffusion constant of quasi-two-dimensional excitons in GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well structures have been measured by Hegarty, Goldner, and Sturge[1] with various techniques, such as resonant Rayleigh scattering, hole burning, and the transient grating method. Their measurements revealed the salient features of the energy and temperature dependence of the homogeneous linewidth of quasi-two-dimensional excitons. They found that the homogeneous linewidth increases sharply as the exciton energy increases through the center of the absorption line, and that below the line center the homogeneous linewidth is thermally activated. These experimental results suggest the existence of the mobility edge for the quasi-two-dimensional excitons in quantum-well heterostructures. From the linewidth analysis of the luminescence and its excitation spectra,[2] and independently from transmission electron microscopy,[3,4] it was suggested that the quantum-well interface has a kind of disorder, namely, islandlike structures having a height of one monolayer and a lateral size of about 300 Å. In the low-energy region the excitons can be localized at one of these islandlike structures since the exciton energy changes by several meV because of the one monolayer difference of the quantum-well thickness, and the Bohr radius of the quasi-two-dimensional exciton is about 100 Å for a typical well thickness of 100 Å.[5-7] The excitons localized at such islands of local minima in energy will migrate among the islands toward the lower-energy sites emitting acoustic phonons. The phonon-assisted migration of excitons among localized sites within a quantum-well layer was the key to understanding the behavior of energy relaxation in the low-energy region of photoluminescence spectra.[8,9]

In this paper the energy and temperature dependence of the homogeneous linewidth, which will be referred to as the dephasing relaxation constant hereafter, of the quasi-two-dimensional exciton and its absolute value are explained quantitatively on the basis of the same model. In the following calculation the dephasing relaxation rate is identified with the rate at which the exciton state changes some of its degrees of freedom, for example, energy, momentum, site, and polarization.[10]

First of all, the mechanisms of dephasing relaxation should be identified.[11] In the localized regime the excitons can tunnel to other localized sites accompanying absorption or emission of acoustic phonons in order to compensate for the energy mismatch. As another mechanism contributing to the homogeneous linewidth, one can consider the phonon-assisted transition to the extended (delocalized) exciton states. The latter mechanism is effective in the intermediate temperature range ($\gtrsim 10$ K) because the transition is associated with phonon absorption. Obviously this mechanism leads to the activation-type behavior of the temperature dependence of the homogeneous linewidth (Γ_h), which was observed experimentally.[1] On the other hand, the tunneling mechanism is working even at low temperatures (~ 1 K) and leads to the variable-range-hopping[12] behavior of Γ_h, which was also claimed to be observed.[1] As for the delocalized exciton state, dephasing relaxation is caused by acoustic phonon scattering on the two-dimensional dispersion curve. In fact, the phonon scattering rate is found to be enhanced by two orders of magnitude over that for the three-dimensional case because the phonon momentum perpendicular to the quantum-well interface can be arbitrary in the scattering. Another mechanism of dephasing relaxation of the delocalized exciton is elastic scattering by the potential fluctuation due to the layer-thickness fluctuation within a layer.

Now let us discuss the dephasing relaxation in the localized regime. The homogeneous linewidth of the localized exciton state with energy E due to phonon-assisted tunneling is calculated by

$$\Gamma_h^{\rm tl}(E)=\int dE'D(E')\tilde{T}(|E-E'|)\{n(E'-E)\Theta(E'-E)+[1+n(E-E')]\Theta(E-E')\}\ , \tag{1}$$

where D is the density of states of the localized exciton state, n is the phonon occupation number, and $\tilde{T}(|E-E'|)$ denotes the spatially integrated exciton transfer rate whose expression is given in Ref. 9. The dephasing relaxation rate due to the activation process is given by

$$\Gamma_h^{\rm ac}(E)=\frac{2\pi}{\hbar}\sum_{K_\parallel}\sum_{Q}|\langle K_\parallel|H_{\text{ex-ph}}|R_a\rangle_Q|^2 n_Q\delta(E_a-E_{K_\parallel}+\hbar\omega_Q)\ , \tag{2}$$

where $|K_\parallel\rangle$ is the delocalized exciton state with a wave vector $K_\parallel$ parallel to the quantum-well interface, $|R_a\rangle$ is the local-

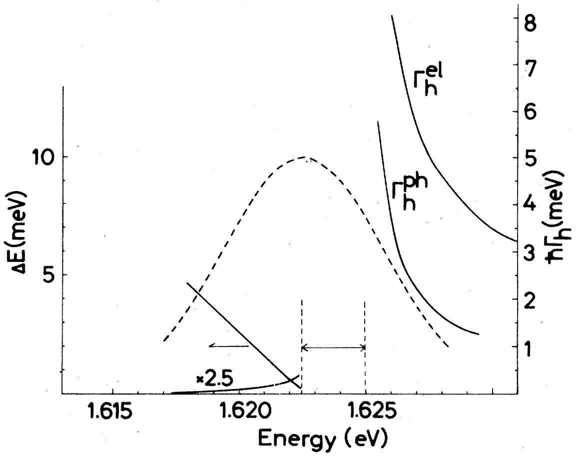

FIG. 1. The calculated homogeneous linewidth $\hbar\Gamma_h$ and activation energy ΔE as functions of exciton energy. The dashed line indicates the assumed exciton absorption spectrum. The region indicated by a double arrow is the supposed transition region between the localized and the delocalized regimes.

ized exciton state at site R_a, and $H_{\text{ex-ph}}$ denotes the quasi-two-dimensional exciton-phonon interaction derived in Ref. 9. As for the envelope function of the localized exciton, it is found that a Gaussian form is not adequate because the energy dependence of the calculated Γ_h^{ac} is too sharp to explain the experimental results. Instead, an exponential function is adopted for the localization envelope. Calculated dephasing relaxation constants are shown in Fig. 1. The density of states of the exciton is assumed to be proportional to the absorption spectrum and the mobility edge is set at the center of the absorption line. The absolute value of the exciton energy in the figure has no particular meaning. The quantum-well thickness is taken as 80 Å and the infinite-barrier model is adopted for the exciton state. The temperature is 5 K and other material parameters, e.g., the exciton-phonon coupling constants, are the same as chosen in Ref. 9. Below the center of the absorption line, the calculated Γ_h is of the order of 0.1 meV and increases with the exciton energy. This is in good agreement with the experimental results. The temperature dependence of Γ_h is examined in this energy region. The Arrhenius plots of Γ_h at various exciton energies are given in Fig. 2. The activation energies determined from the temperature dependence in Fig. 2(a) are plotted in Fig. 1 as ΔE on the left ordinate axis. As seen from Fig. 2(b), in the temperature region about 10 K there occurs a crossover in the dephasing mechanism from thermal activation to phonon-assisted tunneling because the latter mechanism is effective even at low temperatures (~ 1 K). Accordingly, the temperature dependence of Γ_h experiences a crossover between activation-type behavior and the behavior exhibited by Γ_h^{tl}. From the least-squares fit in the temperature range between 2 and 0.5 K, Γ_h^{tl} is found to obey the temperature dependence

$$\Gamma_h^{tl}(T)=\Gamma_0\exp[B/T^{\alpha}] , \qquad (3)$$

where B is positive and the exponent α is estimated to be

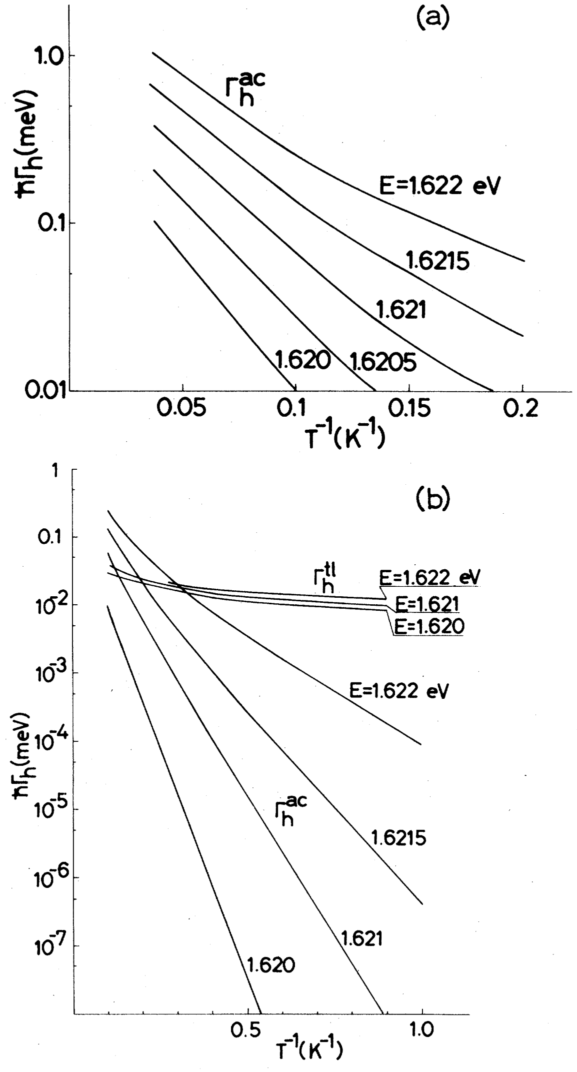

FIG. 2. Arrhenius plot of the homogeneous linewidth $\hbar\Gamma_h$ at various exciton energies in the localized regime; (a) for Γ_h^{ac} and (b) for both Γ_h^{ac} and Γ_h^{tl}.

about (-1.7)–(-1.6), depending weakly on the exciton energy. This exponent is different from that for variable range hopping in two dimensions.[12] This difference arises essentially from the difference in the quantity to be measured. The hopping conductivity is induced by the activation of electrons near the Fermi surface by phonon absorption, while both the phonon absorption and emission processes contribute to the dephasing relaxation rate. Thus it is not very surprising that we found a new exponent different from that for variable range hopping.

Next, the dephasing relaxation constant of the delocalized exciton state will be discussed. As mentioned before, dephasing relaxation is partly caused by acoustic phonon scattering on the two-dimensional dispersion curve and partly by elastic scattering by the potential fluctuation. The contribution from the phonon scattering is given by

$$\Gamma_h^{ph}(K_\parallel)=\frac{2\pi}{\hbar}\sum_{Q}|\langle K_\parallel \pm Q_\parallel|H_{\text{ex-ph}}|K_\parallel\rangle_Q|^2[n_Q\delta(E_{K_\parallel+Q_\parallel}-E_{K_\parallel}-\hbar\omega_Q)+(1+n_Q)\delta(E_{K_\parallel-Q_\parallel}-E_{K_\parallel}+\hbar\omega_Q)] \ . \qquad (4)$$

The calculated results are shown by Γ_h^{ph} in Fig. 1. The dispersion of the delocalized exciton state is assumed to begin from 2.5 meV above the mobility edge. This choice is rather arbitrary but does not seriously affect the qualitative features of the energy dependence of Γ_h. The dephasing relaxation rate due to elastic scattering by the potential fluctuation is calculated as follows. The fluctuation of the exciton energy is caused mainly by the fluctuation of the subband energy, since the binding energy of excitons is affected little by one monolayer change of the well thickness.[5–7] Within the effective-mass approximation the fluctuation of the exciton energy due to the fluctuation of the quantum-well thickness δL_z is given as

$$\delta E \cong \hbar^2 \pi^2 \delta L_z / \mu L_z^3 , \tag{5}$$

where μ is the reduced mass of the exciton. Assuming the scattering potential due to the exciton energy fluctuation to be a cylindrical one with radius ξ, the dephasing relaxation rate is calculated as

$$\Gamma_h^{\mathrm{el}}(K_{\parallel}) = \frac{8\pi M \xi^2 (\delta E)^2}{\hbar^3 \sigma_0} \int_0^{\pi/2} d\theta \left[\frac{J_1(2K_{\parallel}\xi\cos\theta)}{2K_{\parallel}\cos\theta} \right]^2 , \tag{6}$$

where M, δE, and σ_0^{-1} are the exciton total mass, the potential fluctuation, and the areal number density of the scattering potential, respectively, and J_1 denotes the first-order Bessel function. The calculated results are depicted as Γ_h^{el} in Fig. 1. The employed material parameters are again the same as in Ref. 9. The absolute values of Γ_h^{ph} and Γ_h^{el} are both of the order of meV in agreement with the experimental results, whereas they tend to decrease in the higher-energy region. This is because the magnitude of the wave vector of the participating phonons increases with the exciton energy in the case of Γ_h^{ph} and because of the $K_{\parallel}^{-2}$ behavior of (6) in the case of Γ_h^{el}, respectively. Experimentally the value of Γ_h in this energy region has not yet been studied in detail.

The dependence of the dephasing relaxation rate on the quantum-well thickness will now be discussed. In the localized regime both Γ_h^{ac} and Γ_h^{tl} depend on L_z through the matrix element of the exciton-phonon interaction. In the delocalized regime Γ_h^{el} in (6) is found to be inversely proportional to the sixth power of L_z, while Γ_h^{ph} in (4) depends on L_z through the exciton-phonon matrix element. Typical variations of Γ_h at various exciton energies at 5 K are plotted in Fig. 3. It is seen that the L_z dependence of Γ_h in the localized regime is rather sensitive to the exciton energy. On the other hand, Γ_h^{ph} in the delocalized regime is insensitive to the exciton energy and is dependent on L_z only weakly. These features will be useful to identify the mechanism of dephasing relaxation, and systematic experimental study on the L_z dependence is highly desired.

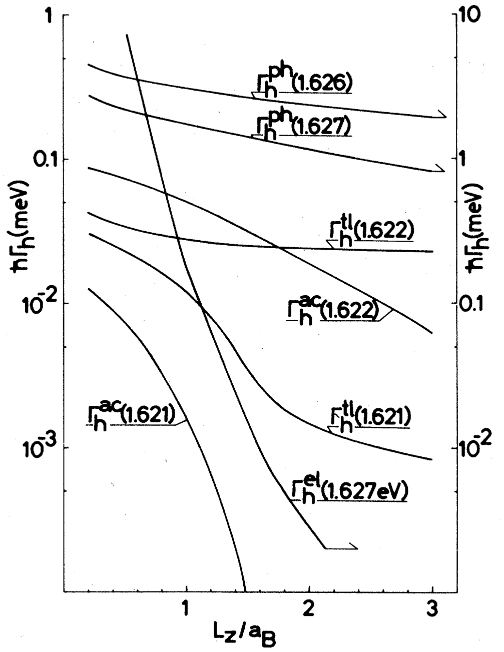

FIG. 3. Dependence of the homogeneous linewidth $\hbar\Gamma_h$ on the quantum-well thickness. The fixed energies of exciton are indicated in parentheses and a_B denotes the Bohr radius of the three-dimensional exciton.

In the energy region of transition between the localized and the delocalized regimes, it is difficult to describe the exciton state precisely and the calculation of Γ_h is left for future study.

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