Resonant Raman study of low-temperature exciton localization in GaAs quantum wells

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We have observed an unexpected temperature dependence in the intensity of Raman scattering resonant with the ground-state exciton in GaAs-$Al_xGa_{1-x}As$ quantum-well heterostructures. We show that the temperature dependence is related to the homogeneous linewidth for quantum-well excitons and yields new insight into low-temperature exciton localization.

The optical properties of semiconductor quantum-well heterostructures are dominated by a series of sharp quasi-two-dimensional excitons. The inhomogeneous broadening of the ground-state $n=1$ heavy-hole exciton has been extensively probed by luminescence spectroscopy.[1] While the inhomogeneous linewidth represents the random distribution of environments occupied by excitons, the processes which limit the lifetime of an individual exciton eigenstate are reflected in the homogeneous linewidth. The homogeneous broadening contains contributions from many elastic and inelastic dephasing mechanisms, including exciton-phonon interactions and scattering by ionized impurities and by fluctuations in quantum-well thickness.[2,3] The radiative contribution to the exciton linewidth is much smaller than any of these. Investigations of the homogeneous exciton linewidth in GaAs-$Al_zGa_{1-x}As$ quantum-well heterostructures have been performed by means of resonant Rayleigh scattering and spectral hole burning within the ground-state exciton line.[4-6]

In this paper we present a new experimental approach to the investigation of the processes which contribute to the homogeneous exciton linewidth in quantum wells. We report the discovery of an unexpectedly strong temperature dependence in Raman scattering resonant with quasi-two-dimensional quantum-well excitons. We establish a direct relation between the Raman intensity and the homogeneous exciton linewidth and show that the resonant Raman method is a particularly effective probe of the linewidth at low temperatures, $T<10$ K, where the homogeneous linewidth is most influenced by exciton localization. For the $n=1$ exciton, we find that the Raman intensity displays a large increase in efficiency with decreasing temperature. By contrast, the Raman intensity resonant with the $n=2$ exciton is much less sensitive to variations in temperature. We show that this difference in temperature dependence can be interpreted in terms of the different processes which limit the relaxation times of ground- and excited-state excitons. In this aspect, the resonant Raman method is unique: It can be used to study the homogeneous linewidths of the high-lying quantum-well exciton states.

Resonant Raman measurements were made on several [001]-oriented molecular-beam-epitaxially-grown GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well heterostructures with well widths ranging from 63 to 222 Å. In order to characterize exciton trapping in our samples, we have also measured the temperature dependence of optical emission. We present here data for a representative sample consisting of 65 periods of 96 Å GaAs—98 Å $Al_{0.28}Ga_{0.72}As$. Samples were cooled by immersion in superfluid liquid helium or by contact with He gas. Temperature was monitored with a calibrated carbon-glass resistor. A tunable LD 700 dye laser operating cw between 700 and 820 nm was the source of excitation. The laser intensity was kept well below 1 W/cm^2.

Figure 1(a) shows the temperature dependence of the resonant Raman efficiency. The phonons involved in the Raman scattering are optical vibrations at the longitudinal optic frequency $\hbar\omega_{LO}$ of GaAs. Circles show the scattering efficiency when the incident laser photon energy $\hbar\omega_L=1.5515$ eV is in strong resonance with the $n=1$ heavy-hole exciton; squares are for $\hbar\omega_L=1.6530$ eV, resonant with the $n=2$ heavy-hole exciton. The temperature dependence for the two excitons is surprisingly dissimilar. Figure 1(b) displays the temperature dependence of the peak luminescence efficiency resulting from optical recombination of the $n=1$ ground-state exciton. Comparison of the luminescence and Raman efficiencies for the $n=1$ exciton shows that both decline sharply with increasing temperature. However, the functional dependences are clearly different. While the luminescence in-

tensity saturates at temperatures below ~ 7 K, the Raman efficiency continues to rise as the temperature is decreased. As shown below, the behavior of the luminescence is not unexpected and can be explained in terms of a conventional theory of exciton trapping. However, the sharp variation of the Raman intensity over this limited temperature range is most remarkable and is the main subject of this paper.

The decrease of luminescence intensity with increasing temperature has long been a signature of bound exciton recombination. At low temperatures, excitons trapped on defects possess an enhanced oscillator strength[7] and high luminescence efficiency. With increasing temperature the excitons become free or dissociate into free electron-hole pairs, with a corresponding decrease in radiative efficiency. We interpret the luminescence data shown in Fig. 1(b) by assuming that the intensity is proportional to the number of excitons bound to defects. For simplicity we assume that the defects are characterized by a single binding energy. In the case when the number of photoexcited excitons is much less than the number of trapping sites, the fraction of trapped excitons is given by[8]

$$N_t = \frac{1}{1+CTe^{-E_b/kT}} , \tag{1}$$

where $C=2\pi Mk/(N_s h^2)$, N_s is the number of defect sites per unit area, E_b is the binding energy of a free exciton to an interface defect, and M is the mass of the exciton.

We have performed a fit of the luminescence data to expression (1). The curve is normalized to the low-temperature saturated value of the luminescence intensity which reflects the situation when nearly all excitons are trapped. We obtain the best fit, shown as the solid line in Fig. 1(b), with $E_b=25$ K$=2.1$ meV and $C=0.8$ K^{-1}. This value of C provides an estimate of the defect density per quantum well. For in-plane electron and hole effective masses $m_e=0.067m_0$ and $m_h=0.2m_0$, we obtain $N_s=6\times 10^9$ cm^{-2} per quantum well. The energy $E_b=2.1$ meV which results from the fit of the luminescence is comparable to the dissociation energy for a quantum-well exciton bound to impurities[9-12] and is also consistent with the binding energy associated with interface roughness.[13]

The above analysis establishes the effects of exciton trapping on optical emission in our samples. We now show that the temperature dependence of the Raman intensity reflects the rate of exciton transfer between trapping sites. When the incident laser frequency $\hbar\omega_L$ is close to resonance with a quantum-well exciton transition energy E, the dominant term in the Raman intensity is proportional to[14]

$$I_R(\hbar\omega_L) \sim \left| \frac{1}{[\hbar\omega_L - E - iG(E)][\hbar\omega_L - \hbar\omega_{\rm LO} - E' - iG(E')]} \right|^2 , \tag{2}$$

where E' is an intermediate exciton state coupled to the exciton E by the exciton–optical-phonon interaction. $G(E)$ and $G(E')$ are the corresponding homogeneous exciton linewidths. For the incoming resonance (i.e., $|\hbar\omega_L - E| \ll |\hbar\omega_L - \hbar\omega_{\rm LO} - E'|$) we have

$$I_R(\hbar\omega_L) \cong \text{const} \times \frac{1}{(\hbar\omega_L - E)^2 + [G(E)]^2} . \tag{3}$$

Since the quantum-well exciton is inhomogeneously broadened, we must add up the independent contributions of each homogeneous line to the Raman intensity. Taking a distribution of exciton energies $\rho(E)$ which is peaked at E_X, we have

$$I_R(\hbar\omega_L) \sim \int dE \frac{\rho(E)}{(\hbar\omega_L - E)^2 + [G(E)]^2} . \tag{4}$$

We have performed measurements with incident photon energies $\hbar\omega_L$ at different positions within the inhomogeneously broadened exciton line $\rho(E)$ and find essentially the same temperature behavior. This indicates that $G(E)$ is slowly varying over the inhomogeneous linewidth.[15] Therefore, we can approximate $G(E)$ by a constant G. In the case when the distribution $\rho(E)$ is much broader than the homogeneous linewidth G,

$$I_R(\hbar\omega_L) \sim \frac{\rho(\hbar\omega_L - E_X)}{G} . \tag{5}$$

Thus close to resonance the Raman intensity is inversely proportional to the homogeneous linewidth.

Equation (5) suggests that the temperature variation of the Raman intensity shown in Fig. 1(a) is due to a strongly temperature-dependent homogeneous linewidth for the ground-state exciton. If the exciton linewidth were governed by a process which is activated with temperature, one would expect linear behavior on a plot of $\ln G \sim \ln(1/I_R)$ as a function of $1/T$. Such a plot is shown in Fig. 2. This figure displays two distinct temperature regimes. For temperatures above ~ 7 K the data can be described by a straight line corresponding to activation, with an energy consistent with the luminescence data. We interpret this behavior in terms of localized exciton states which above ~ 7 K are activated to delocalized excitons, free to move in the plane of the quantum-well layer. At lower temperatures, the departure from this linear dependence indicates a changeover to a different exciton relaxation mechanism.

It has been speculated that the low-temperature transfer of localized excitons occurs via variable range hopping[6] and/or phonon-assisted tunneling.[2] The temperature dependence for variable range hopping is of the form $\ln G = -K/T^m$ where both K and m depend on the dimensionality of the system.[16] On the other hand, phonon-assisted tunneling of quasi-two-dimensional excitons in quantum-well heterostructures has been predicted[2]

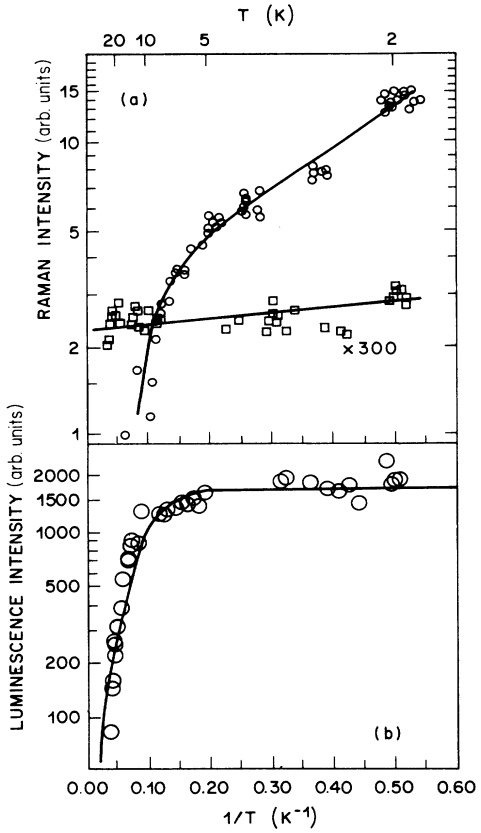

FIG. 1. (a) Temperature dependence of the Raman intensity resonant with the $n=1$ (○) and $n=2$ (□) heavy-hole excitons. (b) Temperature dependence for the luminescence efficiency of the $n=1$ heavy-hole exciton. The line through the data is a fit to Eq. (1).

to yield a dependence $\ln G \sim B/T^{\alpha}$, where α is estimated to be between -1.6 and -1.7. In order to compare our data with both of these models, we show in Fig. 3(a) $\ln(1/I_R)$ plotted as a function of $1/T^{1/3}$, corresponding to two-dimensional variable range hopping, and in Fig. 3(b) $\ln(1/I_R)$ as a function of $T^{1.6}$. The data for temperatures below 7 K are in reasonable agreement with both models. However, the plot in Fig. 3(b) corresponding to phonon-assisted tunneling seems to suggest the existence of a third temperature regime below 2 K. Additional measurements at lower temperatures are required in order to distinguish between the different exciton transfer mechanisms in the localized regime.

In contrast to the case of the $n=1$ exciton, Fig. 1(a) shows that Raman scattering resonant with the $n=2$ heavy-hole exciton is only weakly temperature dependent. In addition, the Raman intensity is reduced from that of the $n=1$ by a factor of ~ 300. By Eq. (5), this suggests a much larger homogeneous linewidth and hence a shorter lifetime. Such behavior indicates that the $n=2$ exciton is not localized and that its relaxation is determined by processes which occur on a faster time scale and which are largely temperature independent. The $n=2$ exciton is composed of higher-lying electron and hole quantum-well subbands and overlaps in energy with the continuum of states associated with the $n=1$ heavy- and light-hole ex-

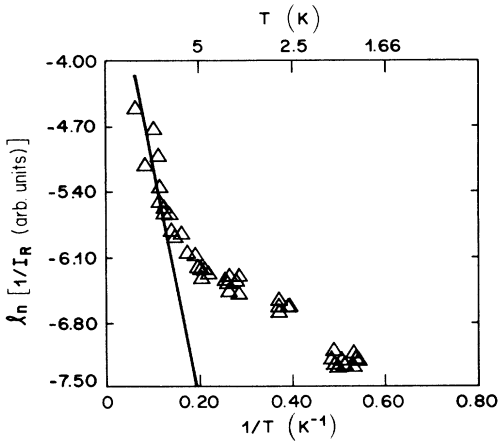

FIG. 2. Dependence of homogeneous linewidths ($\propto 1/I_R$) of the $n=1$ exciton on $1/T$. Solid line corresponds to an activation energy of 2.1 meV.

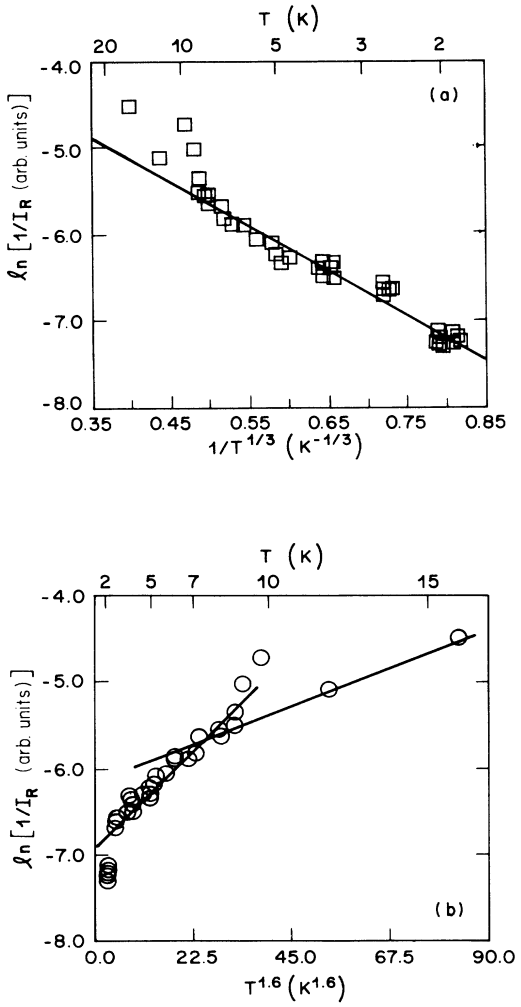

FIG. 3. Dependence of homogeneous linewidths ($\propto 1/I_R$) of the $n=1$ exciton on (a) $1/T^{1/3}$; (b) $T^{1.6}$.

citon transitions. Thus it is likely that relaxation is dominated by decay to lower-lying states.

In conclusion, the intensity of Raman scattering resonant with quasi-two-dimensional excitons in semiconductor quantum wells displays a surprising temperature dependence. We demonstrate that this effect represents a new experimental probe of the homogeneous linewidth. We find that quasi-two-dimensional exciton dynamics exhibits two distinct regimes for temperatures below 20 K. Above ~ 7 K, our luminescence and Raman data indicate the presence of activated detrapping processes, in agreement with previous results.[6] By means of resonant Raman scattering we have been able to extend investigations of the homogeneous linewidth to $T = 1.6$ K. Here we observe contrasting behavior between the $n = 1$ and $n = 2$ excitons which shows that relaxation of these species at low temperatures occurs by very different mechanisms. In the case of the $n = 1$ exciton, our data indicate that dephasing of the state is determined by localization phenomena. Definitive assignment of the physical processes which drive these effects is the subject of future investigations at temperatures below 2 K.

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