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Localized and delocalized two-dimensional excitons in GaAs-AlGaAs multiple-quantum-well structures

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We have measured the homogeneous linewidth Γ_h and the diffusion constant D of resonantly excited excitons confined to GaAs layers 50 to 200 Å wide. We find that Γ_h and D increase sharply as the exciton energy increases through the center of the inhomogeneously broadened absorption line. Below the center Γ_h is thermally activated. We conclude that the excitons are effectively localized below the line center and delocalized above it, as predicted by classical percolation theory.

A particle in a random potential is either delocalized or localized. If delocalized, its wave function is a propagating wave scattered by the potential fluctuations, while if it is localized the wave function is centered on some fixed point and decays exponentially at large distances. Mott[1] has argued that the energy spectra of localized and delocalized particles are separated by a sharp boundary called the mobility edge. This edge has proved remarkably elusive experimentally. Current literature primarily concerns itself with the conductivity near a metal-insulator transition, in which the Coulomb interaction (neglected in Mott's treatment) plays a fundamental role, and in which energy spectra are usually not measured.

Inhomogeneously broadened excitons have an easily measured spectrum and at low densities do not interact with each other. On the other hand, there is no simple handle, such as conductivity, on their motion, and the distinction between "localized" and "delocalized" is blurred by the finite lifetime. In this paper we take "localized" to mean not moving within the lifetime. We report the results of several different types of measurement aimed at detecting the motion of two-dimensional (2D) excitons in GaAs quantum wells, formed by sandwiching thin layers (about 100 Å) of GaAs between layers of the wider band-gap semiconductor AlGaAs. Excitons are confined to the GaAs (the AlGaAs being sufficiently thick to prevent tunneling through the adjacent GaAs layers)[2] but are free to move within the layer. Since each layer fluctuates somewhat in thickness L_z from point to point and the exciton energy depends strongly on L_z, the excitons move in a random potential. We find that the exciton is localized (on the time scale of its lifetime, about 0.5 nsec) at low energies and is delocalized at high. The transition region (the "effective mobility edge") is as abrupt as the quality of our samples permits. It is found to be close to the center of the exciton absorption band, which is the position predicted by classical percolation theory.

Our measurements were of three types: (1) resonant Rayleigh scattering (RS), which has been shown[3] to provide relative values of homogeneous linewidth Γ_h down to very low exciton densities; (2) hole burning, which gives absolute values of Γ_h, but only at rather high exciton densities, and only when Γ_h exceeds the laser linewidth;[4] (3) transient grating measurements, which can measure the exciton diffusion constant D directly, but only at high exciton densities.[5,6] At low temperatures and densities, a localized exciton should have $D=0$ and $\Gamma_h=\tau_R^{-1}\sim 10^9\ \mathrm{s}^{-1}$, where τ_R is the luminescent decay rate, since there is no mechanism other than recombination which can dephase a localized exciton. A delocalized exciton, on the other hand, will have $D>\hbar/m\sim 1\ \mathrm{cm}^2\mathrm{s}^{-1}$ (the value given by Mott's minimum metallic conductivity criterion,[1] m being the exciton mass) and Γ_h given by the scattering rate, which is $\gg 10^9\ \mathrm{s}^{-1}$.

Our samples were grown by molecular beam epitaxy. They consist of about 100 identical layers of GaAs separated by 200-Å layers of $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As}$. The thickness of the GaAs layer in the samples to be discussed here was 51, 102, or 205 Å. (Similar but less detailed results were obtained on several other samples.) The observed (predominantly inhomogeneous) linewidth Γ_x increases with decreasing L_z in agreement with the prediction of a simple model of monatomic width fluctuations.[7]

The RS technique for measuring Γ_h has been described elsewhere.[3] It was shown that if the strength of the RS at laser frequency ω, after correction for the background due to dirt and defects, is $I_s(\omega)$, the homogeneous linewidth is given approximately by

$$\Gamma_h=\left[\frac{I_s(\omega)}{K[1-T^2(\omega)]}+\Gamma_x^{-1}\right]^{-1}-\Gamma_0\ . \tag{1}$$

Here, Γ_x is the observed linewidth, $T(\omega)$ the transmission of the sample, and $\Gamma_0=L_z\omega_{LT}/\xi$, where ω_{LT} is the polariton splitting ($\sim 1\ \mathrm{cm}^{-1}$), ξ the correlation length (~ 300 Å) of the width fluctuations, and L_z the layer thickness. The constant K is difficult to calculate but can be obtained if the absolute value of Γ_h is known at one point. Equation (1) is accurate when $\Gamma_0\ll\Gamma_h\ll\Gamma_x$; outside this range it depends on the doubtful assumption that the broadening mechanisms are additive, i.e., Lorentzian. We obtain Γ_0 from the limiting value of I_s at low ω.

Figure 1 shows $\Gamma_h(\omega)$ obtained from RS at an exciton density $10^5\ \mathrm{cm}^{-2}$/layer (full line) and $\alpha(\omega)$ (dashed line), the absorption coefficient, for two of our samples at low temperature. For $L_z=205$ Å, Γ_x is too small for this method to be useful. Note the sharp change in Γ_h at the center of the line.

We measured hole burning in the same two samples with

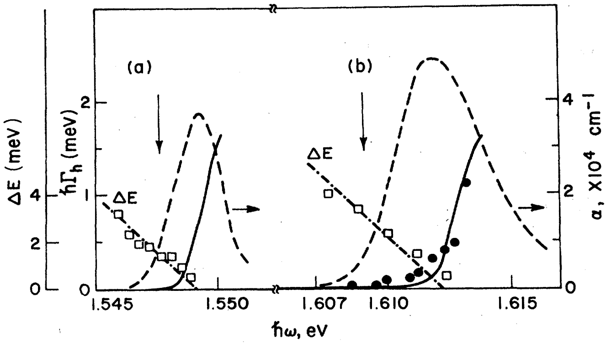

FIG. 1. Absorption coefficient α (dashed line), homogeneous linewidth Γ_h (full line from Rayleigh scattering, circles from hole burning), and activation energies ΔE (squares) as functions of exciton energy $\hbar\omega$, for GaAs quantum wells. The vertical arrows indicate the position of the luminescence peak. (a) $L_z = 102$ Å, $T = 1.5$ K; (b) $L_z = 51$ Å, $T = 5$ K. The dot-dashed line has slope -1. The absolute scale of Γ_h is only approximate.

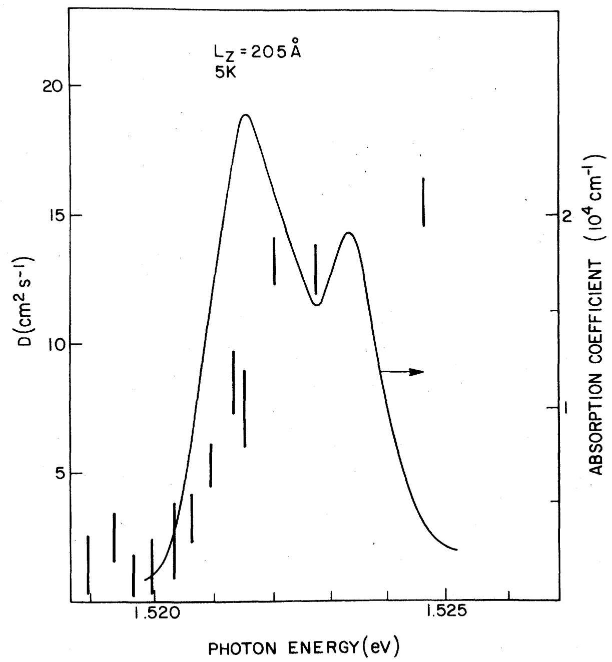

FIG. 2. Exciton diffusion constant D, obtained from Eq. (3), as a function of exciton energy ω, for GaAs quantum wells with $L_z = 205$ Å, at 5 K. Exciton density $\sim 10^{10}$ cm^{-2}/layer. The full line is the absorption spectrum. Note that the light and heavy exciton bands are incompletely resolved due to strain in the sample.

the substrate selectively etched away, using incipient saturation of absorption by a single laser pulse. The exciton density was $\sim 10^9$ cm^{-2}/layer. The absorption coefficient is given as a function of intensity I by

$$\alpha(I) = \alpha(0)/(1 + I/I_{\text{sat}}) \ , \tag{2}$$

where $I_{\text{sat}} \sim 10^{10}$ cm^{-2}/layer. If we again make the assumption of additive broadening mechanisms, we expect $I_{\text{sat}} \propto \Gamma_l + \Gamma_h$, where Γ_l is the transform-limited linewidth of the laser pulse. The values of Γ_h so obtained are shown by the circles in Fig. 1 for $L_z = 51$ Å. Again we see the sharp change in Γ_h at the center of the line. The absolute value of Γ_h is larger because of the contribution of exciton-exciton scattering at these relatively high intensities. Similar results were obtained from two laser pump-probe and pulse propagation measurements.[4]

In the transient grating experiments,[5,6] a spatially periodic distribution of exciton density is created in the layer by the interference of two picosecond pulses entering at different angles, and the decay of this "grating" is monitored by the diffraction of a third pulse. There is always a fast initial decay, whose rate depends on intensity but not on grating spacing d, followed by a slower exponential decay whose rate τ^{-1} depends on d according to[5]

$$\tau^{-1} = 2\tau_R^{-1} + \frac{8\pi^2 D}{d^2} \ , \tag{3}$$

where D is the diffusion coefficient. Since the measured luminescence decay rate[8] $\tau_R^{-1} \sim 2\times10^9$ s^{-1}, and the smallest practicable value of d is 2×10^{-4} cm, the minimum value of D we can measure is ~ 1 cm^2/s. For the 51-Å sample, the rapid initial decay dominates and D could not be measured.

The values of D so obtained for the 205-Å sample are shown in Fig. 2. Note that D is close to zero on the low-energy side of the line, while it has a high value on the high-energy side. A similar change is found in the 102-Å sample, for which we have data only at two exciton energies: one just below and one just above the center of the line. In both samples the change in D occurs in a region of high absorption, showing that intrinsic excitons are involved. Allowing for the high exciton density ($\sim 10^{10}$ cm^2/layer) the results are in qualitative agreement with the other measurements. It might be objected that the increase in D is simply a consequence of increased exciton temperature[9] and has nothing to do with localization. This is suggested by the Einstein relation $D = \mu kT$, where μ is the mobility. However, for neutral particle scattering, μ falls as E^{-1} or faster,[10] so that D would be expected to decrease rather than increase with exciton energy as observed.

The interpretation of the frequency dependence of Γ_h and D in terms of an effective mobility edge at the center of the line is confirmed by the temperature dependence of Γ_h. As the temperature is raised, the resonant scattering drops off, implying a rapid increase in Γ_h. We have made detailed measurements of $\Gamma_h(T)$ in two samples, with $L_z = 51$ and 102 Å, respectively. Arrhenius plots of Γ_h at different ω for the 51-Å sample are shown in Fig. 3. Similar data are obtained for the 102-Å sample. Both sets of data show the same trend. For excitons below the line center, there is a region of temperature, roughly 7–20 K, where the Arrhenius plot is linear, implying that Γ_h is thermally activated. The activation energies ΔE obtained from the slopes of the lines in this region are plotted against the exciton energy in Fig. 1 (squares). For both samples $\Delta E(\omega)$ falls close to a line of slope -1 intercepting the $\Delta E = 0$ axis at the center of the absorption line. Thus, it appears that in this temperature range the predominant contribution of Γ_h is thermal activation to the effective mobility edge. The data in Figs. 1 and 3 are not strictly quantitative since the range of Γ_h is not large, and any possible temperature dependence of the preexponential factor has been ignored in the analysis. In particular, the values of ΔE may be overestimated when

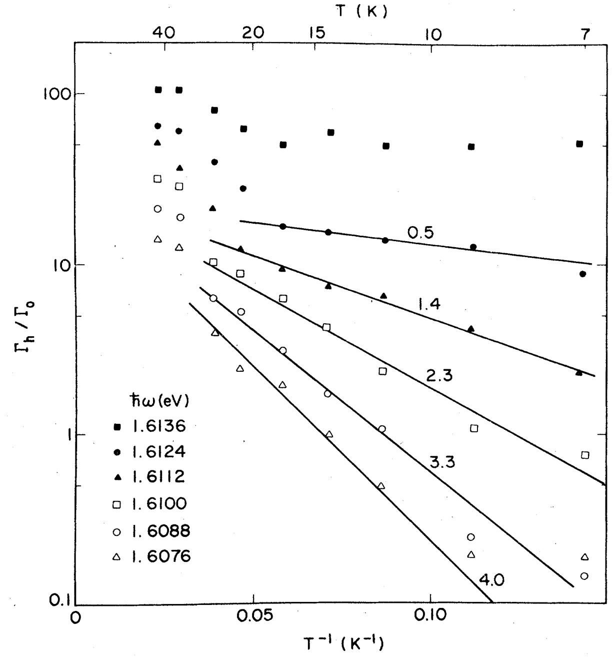

FIG. 3. Arrhenius plot of $\Gamma_h(T)$ at different exciton energies $\hbar\omega$ for the 51-Å sample. The value of $\hbar\omega$ is adjusted at each temperature to allow for the small shift of the exciton peak. Above 20 K the temperature dependence of Γ_h is dominated by exciton dissociation.

small. Nevertheless, the qualitative trend is clear; the lower the energy of the exciton, the smaller Γ_h and the stronger its temperature dependence.

Below 7 K $\Gamma_h(T)$ varies more slowly than expected for a thermally activated process. The data between 7 and 1.5 K can be fitted approximately to the relation $\ln\Gamma_h = (T_0/T)^{-1/n}$, $n = 3 \pm 1$. This suggests that, rather than activation to a well-defined mobility edge, we have here broadening by thermally activated variable-range hopping in the random potential. The exponent $\frac{1}{3}$ is predicted for a uniform density of states in a two-dimensional system.[1]

The localized-delocalized transition is about 1 meV wide in all our samples. This could be due to macroscopic broadening, i.e., inhomogeneous broadening on a scale much larger than the exciton diffusion length.[11] However, it is known that Mott's concept of a minimum metallic conductivity is not applicable at the metal-insulator transition.[12] It may be that the true Γ_h and D do in fact go smoothly to zero at the mobility edge, rather than changing abruptly as predicted by Mott.[1]

We conclude that excitons with energies below the line center are localized, and their motion is thermally activated. Above the line center they are delocalized, being free to move even at our lowest temperature (1.5 K in the Rayleigh scattering experiment). Classical percolation theory, in which tunneling is not taken into account, predicts that the percolation edge should be at the center of a symmetrically broadened line.[13] Tunneling would be expected to move the edge to lower energy, but this is not apparently important in this case. Classical percolation theory has been shown to give a good account of the localized-delocalized transition of triplet Frenkel excitons in organic crystals such as naphthalene.[14] Apparently, in spite of the enormous difference in spatial scale (the Wannier excitons in these GaAs systems are of order 200 Å across) the situation is similar here.

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[2] We use the words "delocalization" and "localization" to refer to motion or lack of it in the plane of the layer. Our excitons are always localized with respect to motion normal to the layer.

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[8] The luminescence decay is not exponential and the rate depends on the sample, and on temperature and intensity. This figure of 2×10^9 s^{-1} is typical for the initial decay rate under the conditions of the grating experiment.

[9] The exciting power is too low to produce an appreciable rise in *lattice* temperature.

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