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DEPENDENCE OF THE EXCITON-POLARITON PHOTOLUMINESCENCE LINESHAPE IN GaAs ON EPITAXIAL LAYER THICKNESS

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The lineshape of the exciton-polariton photoluminescence spectra in thin undoped GaAs layers is observed to be a function of the layer thickness. A doublet peak (or alternatively a single peak with a "reabsorption" dip at the maximum) in thick samples is transformed into a "single" peak as the layer thickness is reduced by etching. This behavior is in agreement with a model based on enhanced scattering near the exciton energy. This scattering is a function of the concentration of scattering centers which is fixed in these samples and of the average scattering length; (i.e., the sample thickness).

A CHARACTERISTIC LOW TEMPERATURE photoluminescence feature of high purity gallium arsenide, associated with the fundamental bandgap, is a two-peaked structure near the free exciton energy. It was originally suggested that this lineshape arose in a fundamental way from the coupling of photons and free excitons (exciton-polaritons) [1] and was reflective of the quasi-thermal mode distributions of the upper and lower branches near the exciton energy [2]. However, later theoretical work showed that the intrinsic luminescence spectrum arising from the free exciton-polariton mode in GaAs should be a single peak (or a doublet with a much smaller separation than observed experimentally) [3]. In other materials [4], theoretical models incorporating strong reabsorption effects have been used to explain the presence of a dip at the free exciton energy. A similar mechanism has been used to explain resonant fluorescence results in GaAs [5].

We have recently demonstrated that enhanced scattering of exciton-polaritons at the exciton energy is clearly responsible for the exciton-polariton lineshape in pure GaAs [6]. The magnitude of the dip generated in the exciton-polariton peak is a function of the density of scattering centers [6]. In this paper we will demonstrate that, for a fixed density of scattering centers, the depth of the dip depends on the scattering distance, i.e., the distance an exciton-polariton in the bulk must travel through the sample before emerging at the surface. This scattering distance is controlled by varying the thickness of a given sample by etching.

EXPERIMENTAL

Layers of nominally undoped GaAs were grown by molecular beam epitaxy on (1 0 0) oriented, undoped, semi-insulating GaAs substrates. Details of the growth technique are described elsewhere [7]. Hall measurements at room temperature and 77 K indicated the samples were *p*-type with a carrier density of approximately 10^{15} cm^{-3} and with mobilities of 400 cm^2 V^{-1}-sec and 8000 cm^2 V^{-1}-sec respectively, but surface depletion effects precluded an accurate determination of the net carrier concentration. Low resolution photoluminescence spectra revealed an appreciable strength for the (e, A°) transition which is due to free to bound transitions involving residual carbon acceptor impurities. The luminescence features associated with donors were unusually weak [6].

After the epilayer was grown, each wafer was cleaved into several small rectangles (1 cm x 0.5 cm). Half of each rectangle was etched [8] so as to reduce the epilayer thickness uniformly. By varying the etching time, a set of samples was produced having a fixed impurity concentration but with varying epitaxial layer thicknesses. When the optical experiments were completed, the samples were cleaved and the cross-sections stained and examined to determine the epitaxial layer thickness by comparing the etched and unetched portions. Thickness accuracy was estimated to be ± 0.2 μm.

Photoluminescence was excited with about 0.1 W cm^{-2} of incident power density from a He–Ne laser (632.8 nm). The spectra were measured with a double grating monochromator and a cooled GaAs photocathode photomultiplier. The samples were cooled in an exchange gas cryostat to 5 K. The photoluminescence of the unetched portion of each sample was compared with that from the etched region. In order to take into account slight differences in optical collection efficiency, the spectra from these unetched portions were normalized to the exciton-bound-to-a-neutral-acceptor, (A°, X),

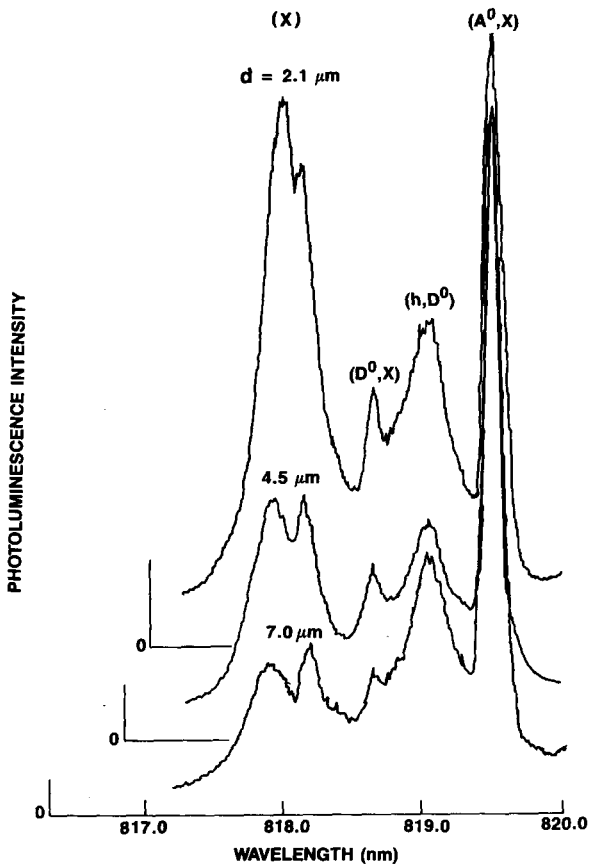

Fig. 1. Low temperature (5 K) photoluminescence (632.8 nm excitation, 0.1 W cm^{-1} incident power density) of undoped epitaxial GaAs with different thicknesses, d. The vertical scale has been displaced for clarity of comparison.

luminescence. This procedure resulted in a slight scaling of the spectra from the corresponding etched regions. This is reflected in a representative set of spectra shown in Fig. 1.

Looking at the exciton-polariton luminescence peaks in Fig. 1, labelled (X), we note two striking facts. As the sample was *thinned* from 7.0 to 2.1 μm, the intensity *increased*. Also the doublet in the thickest sample tended toward a "single" peak in the thinnest sample as the layer thickness was decreased. Other luminescence peaks, associated with excitons bound to neutral donors (D°, X), free-to-bound transitions involving donors (h, D°) and excitons bound to neutral acceptors (A°, X) were nearly independent of sample thickness, indicating uniform carrier concentration.

DISCUSSION

The model we use to interpret the exciton-polariton lineshape is similar to the "reabsorption" concept [4] in which a dip occurs at a photoluminescence maximum due to strong absorption processes. However, in the exciton-polariton formalism, the "absorption" is, in reality, due to the scattering of exciton-polaritons from one energy-momentum state to another [9]. This may occur with little or no change in energy, as in impurity scattering, but ultimately the modes will be scattered into states with different energies and the exciton-polariton mode distribution will be renormalized. This "reabsorption" process is incorporated into the model by assuming that an exciton-polariton at position, x, is "reabsorbed" (i.e. inelastically scattered in the sense described above) with probability $\exp(-\alpha(E)x)$ (where $\alpha(E)$ is an energy dependent "absorption" function) before it reaches the surface. If we assume, as previous authors [4, 9], that the spatial steady state distribution of exciton-polariton modes can be described by $\exp(-x/L_d)$ as measured from the sample surface (where L_d is the steady state diffusion length), then the final result for the luminescence lineshape can be expressed as [9]:

$$I(E) = \frac{P(E)}{1 + \alpha(E)L_d} \tag{1}$$

where $P(E)$ is the energy dependent intrinsic photoluminescence lineshape in the absence of enhanced scattering ("reabsorption") effects. $\alpha(E)$ is defined as $\Gamma(E)/v(E)$, where $\Gamma(E)$ is the scattering rate and $v(E)$ is the exciton-polariton group velocity. $v(E)$ slows by four orders of magnitude at the inflection point of the lower exciton-polariton branch [10] near the photoluminescence intensity maximum. Thus, even if $\Gamma(E)$ is weakly energy dependent, the variation in $v(E)$ strongly enhances $\alpha(E)$ within a narrow range near the exciton energy.

$P(E)$ is a complicated function involving the group velocity, the density of states, and the distribution function of the exciton-polariton, as well as the transmissivity of the exciton-polariton at the sample/vacuum interface [9, 11]. A recent calculation of $P(E)$ has been made by Askary and Yu [3] who extended the work of Sumi [11] to include both the upper and lower exciton-polariton branches. It predicted a single structure with a small splitting which was a consequence of the small value of the difference of the longitudinal and transverse exciton energies, $E_L - E_T = 0.074$ meV [12]. The single nature of the structure was due to the fact that the model ignored the spatial dependence of the exciton-polariton distribution function and impurity scattering.

We have chosen to use equation (1) directly with phenomenological expressions for $P(E)$ and $\alpha(E)$. $P(E)$ was chosen as a Lorentzian with a halfwidth of 0.35 meV and a peak height which is slightly larger than that observed for our thinnest sample, $d = 2.1$ μm. These values produced a lineshape which was in good agreement

with the spectrum for $d = 2.1\,\mu m$, except that the theoretical curve had a maximum at the position of the experimentally observed dip. The form of $\alpha(E)$ was also chosen as a Lorentzian with a maximum defined as $1/L_s$, where L_s is some scattering length. We found that a halfwidth of 0.15 meV for $\alpha(E)$ gave the best fit to our experimental spectra. The peak positions of $\alpha(E)$ and $P(E)$ were chosen to be at the same energy, E_{max}. The thicknesses of the epitaxial layers, d, were comparable to the diffusion lengths, L_d, in undoped GaAs and so we have chosen to set $L_d = d$.

In Fig. 2 a set of lineshapes is shown which were calculated using a value of $L_s = 2.8\,\mu m$, or $\alpha(E_{max}) = 3.6 \times 10^3\ cm^{-1}$; L_d was taken to be 2.1 μm, 4.5 μm or 7.0 μm. This set of calculated lineshapes exhibits the behavior observed in the experimental spectra of Fig. 1. Further, when $L_d/L_s \to 0$, $I(E) \to P(E)$, a single Lorenzian peak. This can occur for fixed L_s, when $L_d \to 0$, as in Fig. 1, or equivalently when $L_s \to \infty$ ($\alpha \to 0$), which occurs when the scattering center density is decreased [6]. As L_d/L_s increases, a dip develops and the intensity decreases due to increased scattering.

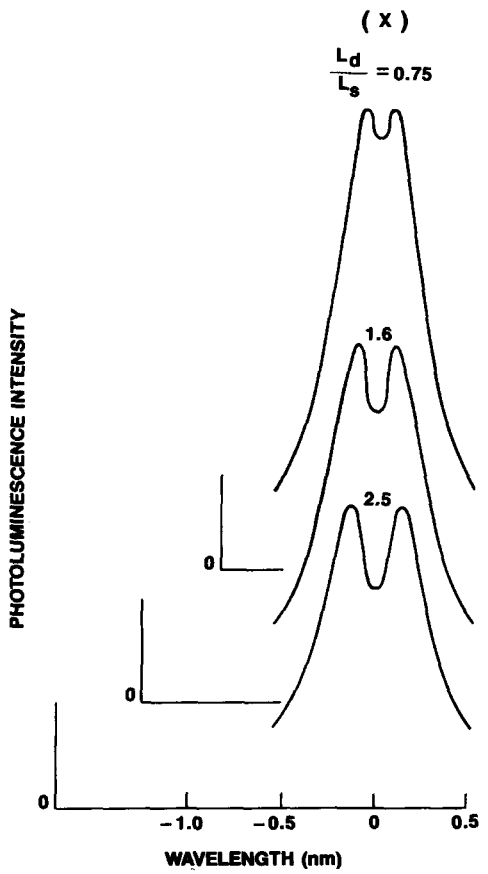

Fig. 2. Free exciton-polariton photoluminescence lineshapes calculated using the model described in the text. The curves are parameterized by the ratio of diffusion length, L_d, to scattering length, L_s. They have been displaced vertically for clarity.

This simple model does not account for the relative sizes of the split photoluminescence peaks or the precise shape of the dip itself. A better description would take into account the probability that the photoluminescence and scattering enhancement maxima would occur at slightly different energies, due to thermalization effects, and that a more realistic expression for $\alpha(E)$ would include the cusplike behavior of $v(E)$ [10].

In conclusion, we have observed that for thin undoped GaAs samples with a fixed concentration of scattering centers, the lineshape of the free exciton-polariton photoluminescence peak is dependent upon the thickness of the sample. This is a consequence of the fact that in thicker samples, exciton-polariton modes have, on the average, a larger scattering distance to transverse on their journey to the sample surface and external detection. In thinner samples the modes are, on the average, closer to the surface and have a greater likelihood of leaving the sample before undergoing many renormalizing scattering events.

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