Observation of Polaritons in GaAs: A New Interpretation of the Free-Exciton Reflectance and Luminescence

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The reflectance and luminescence of high-purity epitaxial layers of GaAs have been studied at low temperatures. Strong polariton and spatial dispersion effects are seen for the $n=1$ free exciton. The longitudinal-exciton energy is found to be 1.5151 ± 0.0001 eV (at 2 K) and the transverse-exciton energy is quite near 1.5149 eV. Two luminescence structures at 1.5154 and 1.5148 eV are identified with radiative decay from the upper and lower polariton branches, respectively.

We report the first detailed measurements of the free-exciton reflectance of GaAs and present a new interpretation of the free-exciton luminescence. The reflectance data (for exceedingly pure epitaxial layers at 2 K) clearly demonstrate polariton behavior and the effects of spatial dispersion. Two features in the luminescence data are identified with the free exciton corresponding to decay of polaritons from the upper and lower branches of the polariton dispersion curve. A structure near 1.5154 eV previously identified by Gilleo, Bailey, and Hill[1] with the $n=1$ free exciton is shown to arise from radiative decay from the upper polariton branch (UPB). A second structure at 1.5148 eV previously assumed[1,2] to be an impurity structure is identified with radiative decay from the lower polariton branch (LPB).

Our data were obtained on epitaxial samples of GaAs (20 to 40 μm thick) grown on Cr-doped semi-insulating substrates using the $AsCl_3|Ga|H_2$ synthesis system. Growth details are presented elsewhere.[3] The samples were n type with typical net donor concentrations of approximately 4×10^{13} cm^{-3}, total impurity concentrations less than 2×10^{14} cm^{-3}, and mobilities of approximately 200 000 cm^2/V sec at 77 K.

Reflectance data[4] for two samples are shown as the dashed curves in Fig. 1. These two spectral shapes are seen on numerous samples with as-grown (100) surfaces. Both curves exhibit distinct effects of exciton spatial dispersion (finite exciton mass, which for GaAs is $m^* = m_c^* + m_0/\gamma_1 = 0.21 m_0$). Hopfield and Thomas[5] have shown that the "spike" and the "nonclassical" spectral shape arise from spatial dispersion. We have used their theory to obtain the solid curves in Fig. 1. Except for the surface boundary-layer thickness l, the parameters for the theory are known ($m^* = 0.21 m_0$,[6] $\epsilon_0 = 12.53$,[7] and $4\pi\alpha_0 = 1.6 \times 10^{-3}$[8]). By adjusting only this one parameter, l, we obtain a good fit to the data. It is important to note that only the surface-layer thickness was changed to obtain the two rather different theoretical curves ($l = 435$ and 290 Å, respectively, for curves a and b in Fig. 1). The fact that a different l is needed for different samples indicates that the actual surface conditions (i.e., occupancy of surface states and band bending) vary from sample to sample.

Hopfield[5,9] has shown that the spike occurs at the cutoff energy for the UPB, which must coincide with the longitudinal-exciton energy at $k=0$. Thus we conclude that the longitudinal-exciton energy is 1.5151 ± 0.0001 eV. This spike can be exceedingly sharp. We have measured widths (full width at half-maximum) of less than 0.1 meV which are still limited by spectrometer resolution.

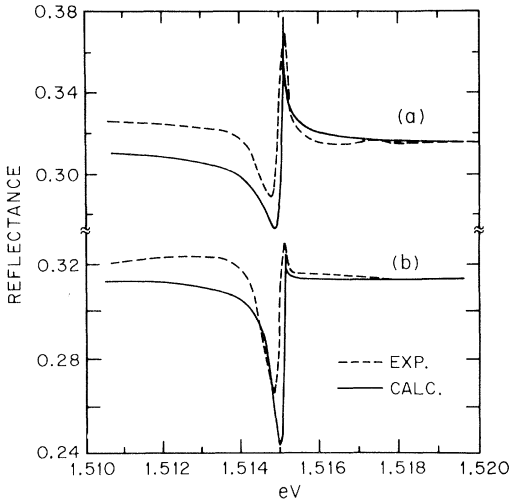

FIG. 1. Reflectance data (dashed curve) and calculated reflectance (solid curve) for two samples at 2 K. The absolute amplitudes of the data have been scaled to obtain a reflectance of 0.314 (the background value due to ϵ_0) at 1.519 eV. The spectrophotometer resolution was approximately 0.08 meV. The reflectance curve b and the luminescence data presented in Figs. 2 and 3 were obtained on the same sample.

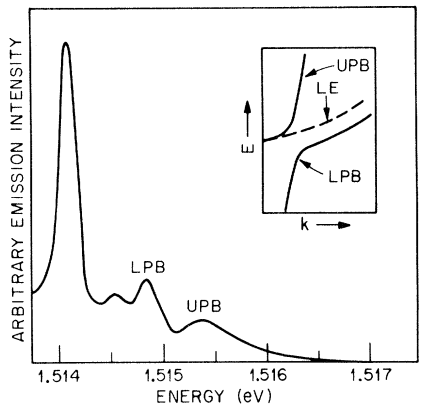

FIG. 2. Luniescence at 4.2 K for low pump power at 6328 Å. The structures labeled UPB and LPB occur at 1.5154 and 1.5148 eV, respectively. The inset illustrates the upper (UPB) and lower polariton branches (LPB) and the longitudinal-exciton (LE) dispersion curves.

The transverse-exciton energy is not as easily determined. The polarizability ($4\pi\alpha_0 = 1.6 \times 10^{-3}$), which accounts for the measured absorption strength and gives reasonable agreement with the measured reflectance amplitude, predicts a longitudinal-transverse exciton splitting of 0.1 meV. The difficulty, however, is that the transverse exciton does not necessarily fall at an extremum of the reflectance curve. In the theoretical curves in Fig. 1 the transverse energy falls quite near the negative peak. The data then seem to indicate a longitudinal-transverse splitting as large as 0.25 meV. Thus we conclude that the longitudinal exciton and UPB (at $k = 0$) are clearly at 1.5151 eV, and that the transverse exciton (or "knee" of the LPB) is quite near 1.5149 eV. A full account of the temperature and stress dependence, spatial dispersion and surface effects, and the weaker subsidiary structure in the reflectance will be presented elsewhere.

The luminescence spectrum in the spectral region near the free exciton illustrated in Fig. 2 is typical of that seen in these high-purity layers for low pump power. (Essentially the same spectrum has been seen in pure p-type layers.) Compared with the spectra observed in earlier work,[1,2] the structure in Fig. 2 is narrower, and fewer impurity-induced lines are observed. The structure which peaks at 1.5154 eV in Fig. 2 was identified by Gilleo, Bailey, and Hill[1] as free-exciton luminescence. In agreement with the work of Leite, Shah, and Gordon,[10] we find that the peak energy of this structure shifts to higher energy as the pump intensity increases. The important feature not previously recognized is that the low-energy "edge" of this structure (see Fig. 1 of Ref. 10) does not shift with pump power. It remains very near the longitudinal exciton energy at 1.5151 eV. We identify this structure with radiative decay from the upper polariton branch (UPB) as indicated in the inset in Fig. 2. Similarly, the structure centered near 1.5148 eV in Fig. 2 is essentially at the transverse-exciton energy. We identify it with decay from the lower polariton branch (LPB).

We have confirmed these interpretations by studying the temperature dependence of the luminescence and the stress behavior of both the reflectance and the luminescence. The dominant 1.5140-eV feature in Fig. 2 (which is associated with a bound exciton decaying at an isolated neutral donor site[11]) quenches rapidly as a function of increasing temperature. In contrast, the intensities of the UPB and LPB features are essentially constant in the temperature range 4–30 K, demonstrating that *both* features quench with a substantially greater activation energy than does the 1.5140-eV line. This behavior is consistent with the polariton interpretation presented here, but is inconsistent with an identification of the 1.5154-eV feature with the "free-exciton" and the 1.5148-eV feature with a bound exciton-donor complex.

The reflectance was studied as a function of compressive uniaxial stress along the [001] and [110] crystallographic directions. The stress splits the structure into a σ-polarized ($\vec{E} \perp$ stress) upper-energy component and nearly degenerate σ- and π-polarized ($\vec{E} \parallel$ stress) lower-energy components. The line shapes of the lower (π and σ) structures are essentially the same as shown in Fig. 1; the upper (σ) component is somewhat broadened. The observed splitting is consistent with the piezoelectroreflectance results of Pollak and Cardona.[12] This confirms that the structure arises from the free exciton (as shown below, the bound-exciton structure has a much weaker stress dependence). The exchange splitting of the lower π and σ structure is observed to be less than 0.1 meV.

The stress dependence of the two polariton luminescence structures is presented in Fig. 3. Significantly, the overall stress behavior of the UPB and LPB is the same as that observed for the reflectance structure (and hence the same as that observed in Ref. 12). In contrast to this, the bound-exciton luminescence at 1.5140 eV exhibits a considerably smaller splitting for a given level

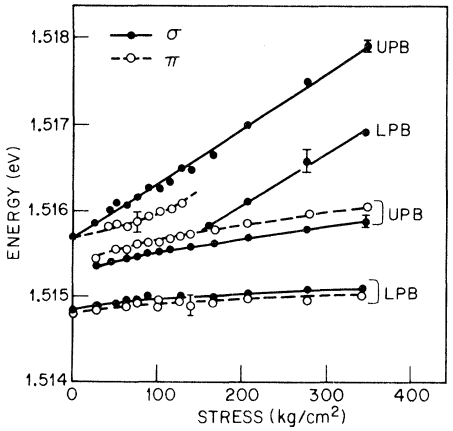

FIG. 3. Stress results at 8 K for compressive uniaxial stress along [001] and k along [100]. Essentially identical results were obtained for stress along [011] and k along [100]. π and σ polarizations are defined as $\vec{E} \parallel$ stress and $\vec{E} \perp$ stress, respectively, where $\vec{E}$ is the electric vector of the light.

of stress. Gilleo, Bailey, and Hill[13] have studied the stress behavior of the UPB portion of this structure. (They did not report the LPB structure. As the impurity concentration and pump power increase, the bound-exciton structure begins to mask the LPB structure.) Except for the anticrossing of the π line discussed below, we agree with their results for the UPB.

In the low-stress region an apparent anticrossing is seen in the π spectrum. This could result from an interaction of the π-polarized UPB either with the longitudinal exciton which accompanies the upper σ-polarized UPB or with the (otherwise) optically inactive component of the Γ_4 exciton (spin triplet) which is in the same energy region. Such coupling could arise from linear-k terms[14,15] or from the valence-band anisotropy.[16] Since the appropriate coupling constants are not known, we are not able to give a detailed identification of the coupling mechanism.

The question arises whether the 1.5154-eV structure is really associated with the UPB, or whether it should be associated with longitudinal excitons which could couple to the light at a rough surface. In the stressed crystal, there is only one π-polarized free-exciton mode (for $n=1$). This mode couples with the photons (except for $k \parallel$ stress when it is a pure longitudinal mode) to form π-polarized upper and lower polariton branches. In the stressed crystal there is no longitudinal mode degenerate with the π-polarized UPB at $k=0$. Thus, the fact that we observe two π lines indicates clearly that the luminescence arises from the upper and lower polariton branches.

Luminescence from the upper polariton branch has been observed previously in CdS.[17] In that case, the UPB and LPB structures were not well resolved and were only seen for certain geometries in which the mixed longitudinal-transverse excitons are weakly coupled to the photons. In contrast, for GaAs, the upper- and lower-branch structures are well resolved for the ordinary transverse-exciton modes. The results for these two materials suggest that such a polariton point of view may be important in the interpretation of luminescence results for other materials.

Generally, theoretical treatments, such as that by Toyozawa,[18] have concentrated upon the luminescence from the LPB (near the "knee") and have disregarded the UPB. A simple calculation indicates that, for the energies at which luminescence is seen in GaAs, the group velocity and density of states in the UPB are, respectively, a factor of 2 and 0.2 times the values in the LPB. Thus it is quite reasonable to expect luminescence from the upper branch. We are hopeful that the results presented here will encourage new interest in the theory of polariton luminescence.

Our main result is that we have demonstrated the existence and importance of polariton (and spatial-dispersion) effects in the reflectance and luminescence spectra of GaAs. This point of view removes some of the difficulties encountered in earlier work which associated the 1.5154-eV structure with the "free exciton." For example, the photoreflectance at 2 K [see Fig. 2(a) of Ref. 19] was associated with bound excitons because the structure was 0.5 meV below the "free-exciton" energy. From our work we see that this structure should rather be interpreted as a photo-induced change in the intrinsic polariton reflectance.

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Indirect–Band-Gap Super-Radiant Laser in GaP Containing Isoelectronic Traps

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We report observations which can be interpreted as super-radiant laser emission in the indirect–band-gap semiconductor GaP. Optical gain, as high as 10^4 cm^{-1} at 2°K, has been measured over a range of temperatures from 2 to 300°K. The gain arises from a new process not important at low excitation intensities. It is demonstrated that isoelectronic traps are an important part of this new process.

We report observations which can be interpreted as stimulated emission in optically pumped GaP doped with either nitrogen or bismuth isoelectronic traps. For GaP(N) the stimulated emission occurs in the green over a wavelength range extending from ~5400 to ~5700 Å, and is observed at temperatures from 2°K to above room temperature. For a pump excitation intensity of 2×10^7 W/cm^2, the peak optical gain[1] associated with the stimulated emission exceeds 10^4 cm^{-1} at 2°K and is greater than 200 cm^{-1} at room temperature. For GaP(Bi) the optical gain is spectrally shifted, occurring in the yellow from 5550 Å to beyond 6000 Å. The gain, until now not measured in any indirect–band-gap semiconductor,[2,3] is quite large for both GaP(N) and GaP(Bi), and it is comparable in magnitude to that measured[1,4,5] in direct gap materials at these temperatures and excitation conditions. The measured gain spectra cannot be simply related to the oscillator strengths measured in this spectral region for low excitation intensities,[6-8] thus implying that the stimulated emission is due to a new process which does not contribute at low intensities. It is shown that isoelectronic traps (either N or Bi) are an important part of this new process.

The observations reported in this paper were made using an experimental technique in which emission spectra are studied as a function of the optical excitation length. Various modifications of this technique have been used extensively for optical-gain measurements in gases,[9] liquids,[10] and direct–band-gap semiconductors.[1,4,5] Shaklee and Leheny[1] have given the details of the experimental technique as applied to semiconductors, and no further description of the technique will be given here. In the present work, this technique is utilized to establish the presence of gain in the indirect-gap material GaP.

In Fig. 1, we show with dashed lines the GaP stimulated emission spectra for two different lengths of excitation. It is seen that doubling the excitation length leads to an increase in emission intensity of a factor of 10. These spectra were