Reflectivity of two-dimensional polaritons in GaAs quantum wells

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A very large absolute reflectivity (> 70%) near the exciton resonance is reported for single GaAs-$Al_xGa_{1-x}As$ quantum wells. The large reflectivity persists at lattice temperatures greater than 100 K. The reflectivity of single quantum wells is shown to be a function of the intrinsic radiative lifetime and the scattering rate of excitons with phonons and impurities. The macroscopic two-dimensional exciton polarization is well described by a microscopic model of nonlocal susceptibility. Based on the magnitude and width of the reflectivity peak, we deduce a value for the radiative linewidth of the free exciton which is in good agreement with the values obtained from radiative lifetime measurements.

The study of the reflectivity of quantum wells has assumed great technological importance in recent years.[1] By properly stacking quantum wells, one can obtain a very highly reflective mirror. However, the reflectivity of single quantum wells is not clearly understood. In fact, it is only recently that a theoretical description of the two-dimensional (2D) polarization has been given.[2,3] The optical properties of semiconductors, especially those of quantum-well heterostructures, have been the subject of intense research in recent years.[1–6] Much of this work has been in photoluminescence and absorption studies of these structures. While the various aspects of luminescence are theoretically understood, the experimental understanding of the intrinsic optical properties of semiconductors is still incomplete. It is of fundamental importance to be able to separate out the intrinsic effects from the extrinsic optical properties. Recently, the authors have published an account of the intrinsic linewidth and radiative lifetime measurements in GaAs quantum wells.[4] A quantitative understanding of the radiative lifetime of the free excitons was given in the above work. The theoretical model was based on the two-dimensional macroscopic polarization of the free exciton. The thermal distribution of exciton states, as well as the localization of the polarization due to defects, was shown to account for the experimentally observed temperature and intensity-dependent photoluminescence lifetimes. One of the key features of the above work was in deducing the intrinsic homogeneous linewidth of free excitons by systematically measuring the linewidth as a function of well width for given growth conditions. The linewidth versus temperature data exhibited a linear temperature dependence, indicating that LA-phonon scattering is the dominant *low*-temperature linewidth broadening mechanism. The measured linewidth at a temperature of about 10 K was greater than that expected due to the radiative linewidth and LA-phonon contributions alone. It was conjectured that the additional linewidth broadening seen was due to the spin relaxation of holes and consequently that of the excitons.[5] In Ref. 5 the authors showed that the spin dynamics of excitons was profoundly altered by exciton-exciton and exciton-impurity scattering. Time-resolved luminescence measurements were used to clearly demonstrate the rapid spin-relaxation rate of excitons in the small scattering regime. By varying the excitation intensity, and thereby the exciton-exciton scattering rate, the spin exchange interaction energy was estimated.

It is now a well established fact that the rapid radiative decay of excitons is due to the two-dimensional macroscopic polarization of excitons and the nonconservation of the wave vector along the growth direction which causes the exciton polarization to couple strongly to the radiative modes.[6,7] The decay rate and, hence, the intrinsic homogeneous radiative linewidth is proportional to the oscillator strength per unit area. One can indirectly measure the oscillator strength by measuring the radiative lifetime. However, the thermal distribution of exciton states and localization of the 2D polarization causes additional complications in this measurement of the oscillator strength. One of the more elegant methods of measuring the oscillator strength is through reflectivity measurements. The connection between reflectivity measurements and the intrinsic radiative lifetime was first pointed out by Andreani, Tassone, and Bassani.[7] They have proposed that one can, in principle, obtain the intrinsic radiative lifetime and hence the optical oscillator strength from reflectivity measurements alone.

The reflectivity of polaritons in bulk[8] GaAs, as well as in quantum wells,[9] has been studied extensively. It is widely accepted that the reflectivity line shape is dictated by the location of the polarization with respect to the surface. The finite mass of the exciton results in the spatial dispersion effect of bulk semiconductors. The spatial dispersion leads to two distinct polariton modes at a given frequency. The presence of two propagating modes requires additional boundary conditions to be invoked in the interpretation of reflectivity of bulk semiconductors. In the case of the bulk crystals it was suggested by Hopfield and Thomas[10] that the exciton is excluded from a small region just below the surface. In the experimental

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work by Sell *et al.*[8] the observed reflectivity spike was interpreted as evidence for spatial dispersion. The polarization free zone was shown to be governed by sample surface preparation. The surface properties of bulk crystals, cleaved in a vacuum at liquid-helium temperatures, have been studied by Schultheis and co-workers.[11] In this work it was shown that the electric field induced by a high density of surface states causes the appearance of sharp spikes in the reflectivity data.

The longitudinal-transverse splitting $\Delta E_{\rm LT}$ of the bulk exciton which is a measure of the oscillator strength has been placed between 0.08 and 0.25 meV by an analysis of the reflectivity data.[8,12] A more reliable estimate is $\Delta E_{\rm LT}=0.08$ meV which was obtained from resonant Brillouin scattering data of bulk GaAs.[13] In the case of quantum wells, it was shown by Andreani and Bassani[14] that the longitudinal-transverse splitting energy $\Delta E_{\rm LT}$, which corresponds to the long-range part of the exchange interaction, does not get enhanced by reduced dimensionality. However, the short-range exchange interaction does get enhanced as shown by other workers.[5,15] In fact, it was shown that $\Delta E_{\rm LT}^{\rm 2D}$ vanishes for small in-plane wave vectors (which would be the case in standard reflectivity measurements). It was demonstrated that the correct interpretation of the reflectivity data, in the absence of LT splitting, is given by a nonlocal model for the susceptibility. Under certain conditions[16] the nonlocal model may be replaced by the conventional local dielectric function (a Lorentzian line shape for the dielectric function) and one may obtain $\Delta E_{\rm LT}^{\rm 2D}$ as a fitting parameter. This method of obtaining $\Delta E_{\rm LT}^{\rm 2D}$ is purely phenomenological and has no physical relationship to $\Delta E_{\rm LT}^{\rm 2D}$, which vanishes for $k_\parallel L_z \gg 1$, where $k_\parallel$ is the in-plane exciton momentum and L_z is the quantum-well width.

In this work we continue our investigation of intrinsic processes in quantum-well structures by linking reflectivity measurements with measurements of the intrinsic radiative lifetime. Here, we present results that show clearly the validity of a microscopic model for the nonlocal macroscopic polarization. The values of the intrinsic radiative linewidth that we deduce from the reflectivity measurements agree very well with those used for the photoluminescence lifetime measurements.

For normal incidence the reflectivity amplitude of a single quantum well is given by[3]

$$r_{\rm QW}=\tfrac{1}{2}(\alpha-1)\exp(ik_0L)\ , \tag{1a}$$

$$\alpha=\frac{\omega-\varpi-i\Gamma_0+i\Gamma_s}{\omega-\varpi+i\Gamma_0+i\Gamma_s}\ , \tag{1b}$$

where Γ_0 is the intrinsic radiative linewidth, Γ_s is the linewidth broadening due to scattering by acoustic, optical phonons, and impurities, and $\varpi=\omega_0-\beta\Gamma_0$. We may show that

$$\Gamma_0=\frac{e^2}{4\pi\varepsilon_0}F\frac{\pi}{nm_0c}\ ,$$

$$\beta=\frac{\int_{-L/2}^{+L/2}dz\int_{-L/2}^{+L/2}dz'\rho(a)\rho(z')\sin(k_0|z-z'|)}{\left|\int_{-L/2}^{+L/2}dz\rho(z)\cos(k_0z)\right|^2}\ , \tag{2}$$

where F is the oscillator strength per unit area, n is the refractive index in the barrier, m_0 is the free-electron mass, $k_0\equiv n\omega/c$ is the photon wave vector, and $\rho(z)\equiv f_e(z_e)f_n(z_h)$ is the product of the confinement functions for the electrons and holes in the well. The peak magnitude of $r_{\rm QW}$ is given by $\Gamma_0/(\Gamma_0+\Gamma_s)$. We note that the reflectivity resonance ϖ is redshifted from the exciton resonance ω_0 by $\beta\Gamma_0$. Taking into account the reflectivity of the surface (r_{12}), we may write the net reflectivity of the single quantum well to be

$$R=\left|\frac{r_{12}+r_{\rm QW}(\omega)\exp[\phi]}{1+r_{12}r_{\rm QW}(\omega)\exp[\phi]}\right|^2\ , \tag{3}$$

where $\phi=k_0d$ is the phase change due to propagation in the cap layer of thickness d.

The sample, used in our experiments, has a series of isolated single quantum wells grown on a (100) semi-insulating substrate. A 36-period AlAs/GaAs superlattice smoothing layer was first grown on a nominally undoped GaAs buffer of 100 nm. The $Al_xGa_{1-x}As$ barrier ($x=0.3$) between each of the quantum wells was 22.5 nm. To avoid absorption in the thicker wells (smaller band gaps), 11 uncoupled quantum wells of nominal thickness 32.5, 20, 15, 10, 8, 7, 6, 5, 4, 3, and 2 nm were grown in this order from the substrate. The spacer between the surface and the top 32.5-nm quantum well is the 22.5-nm $Al_xGa_{1-x}As$ barrier. The low-temperature photoluminescence, photoluminescence excitation, and reflectivity experiments were carried out in a liquid-helium cold finger Dewar, using a triple monochromator and computer-controlled photon-counting system. Negligible Stokes shifts were observed in the luminescence and excitation spectra. The sample was mounted on a sapphire disk to avoid strain at low temperatures. The photoluminescence data exhibit a high luminescence efficiency, a narrow free-exciton linewidth of the order of 150 μeV half-width at half maximum (HWHM) for the wider wells, and very small bound exciton luminescence.[4,5] The reflectivity measurements were performed close to normal incidence by shining white light on the sample and collecting the reflected light into the monochromator which was scanned in wavelength. The incident optical power was kept to less than a few microwatts and the spectral width of the source limited by colored glass filters. The pump source for the luminescence measurements was a laser diode operating at 750 nm. The excitation intensity was less than 0.1 W/cm^2. The spectral resolution of the setup was around 25 μeV (HWHM). Ambiguities caused by spectrometer calibration are minimized by using the same instrument with identical settings for both the photoluminescence and reflectivity measurements. Furthermore, the measurements were performed in rapid succession on the same area of the sample.

Figure 1 shows the data of three quantum wells at a lattice temperature of 10 K. The absolute reflectivity was obtained by taking into account the responsivity of the source and detector combination, removing all linear artifacts, and scaling the background reflectivity amplitude to $r_{12}=0.56$. The reflectivity data were fit to the expression in Eq. (3) and the solid lines are the theoretical fits to the data. The three fitting parameters used are the following: the phase ϕ, the ratio $\eta=\Gamma_0/(\Gamma_0+\Gamma_s)$, and the scattering coefficient Γ_s. The measured photoluminescence and reflectivity linewidths are similar in magnitude. Figure 2 shows the reflectivity (solid line) and photoluminescence (dashed line) data of six quantum wells. The labels X_1 through X_6 denote the heavy-hole peaks of the 32.5-, 20-, 15-, 10-, 8-, and 7-nm wells in that order. The additional features seen are the light-hole resonances. The reflectivity line-shape change is attributed to the different distances of the quantum wells from the surface. The peak positions of the photoluminescence are slightly higher in energy than the reflectivity resonances. This is attributed to $\beta\Gamma_0$ defined in Eq. (2). From a series of scans of reflectivity and photoluminescence, we estimate the redshift of the reflectivity peak from the photoluminescence peak to be $125\pm(50)$ μeV for the 15-nm well. The reflectivity of the light hole is seen to be similar in shape but smaller in magnitude than the heavy-hole reflectivity.

The scattering rate[4] or the extrinsic linewidth $\Gamma_s=\Gamma_{\rm spin}+\gamma_{\rm LA}T+\Gamma_{\rm LO}+\Gamma_{\rm imp}+\cdots$ may be changed by elevating the sample temperature or by adding impurities. The reflectivity in samples with a significant impurity content was small, of the order of a few percent. In the case of very high quality and *nearly intrinsic* samples, the modulation of the reflectivity was observed to be of the order of tens of a percent. According to theoretical

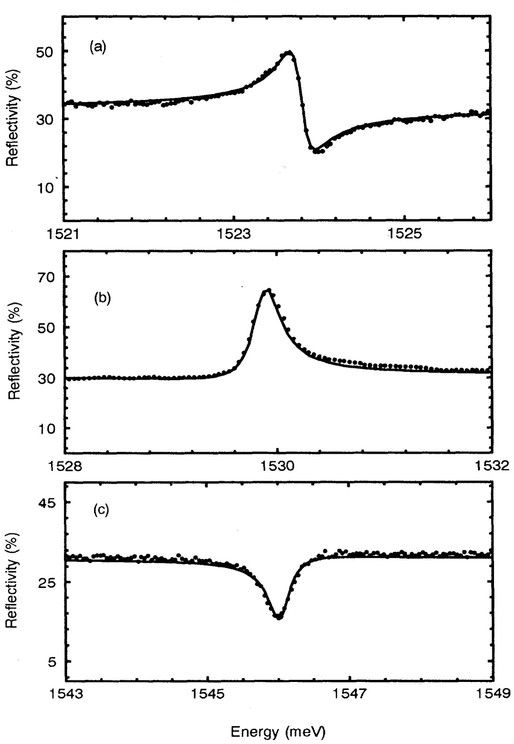

FIG. 1. Reflectivity at 10 K of three quantum wells whose widths are (a) 20 nm, (b) 15 nm, and (c) 10 nm. The solid line is the theoretical calculation discussed in the text. The data were taken close to normal incidence.

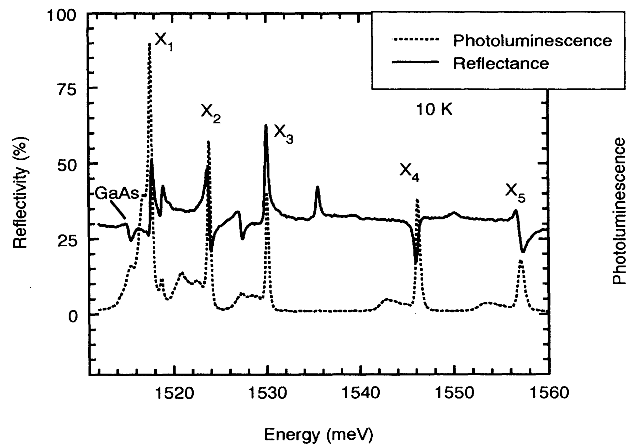

FIG. 2. Photoluminescence and reflectance data at 10 K. The labels X_1-X_5 correspond to the heavy-hole peaks of the 32.5-, 20-, 15-, 10-, and 8-nm wells, in that order. As discussed in the text, a small redshift of the reflectivity peaks (X_2-X_5) with respect to the luminescence peaks is observed.

expectations, it may be, in principle, possible to observe close to 100% reflectivity modulation as η approaches unity. However, a finite scattering linewidth Γ_s prevents η from reaching unity. We have observed a peak reflectivity of around 70% for a single 15-nm quantum well. From Eq. (3) we obtain $\eta\approx0.51$ which implies that the homogeneous linewidth broadening is of the same order of magnitude as the intrinsic radiative linewidth. The photoluminescence and reflectivity linewidths are around 150 μeV. From the measured values of the peak reflectivity and homogeneous linewidth, we use our theoretical model to *estimate* the intrinsic radiative linewidth Γ_0 of the heavy hole (hh) to be around $60(-15)$ μeV where the error bar is due to the spectrometer linewidth. The estimated value of Γ_0 is approximately twice the value obtained from photoluminescence lifetime measurements for the same quantum well.[4] This is attributed to the fact that, in a lifetime measurement, one usually measures the thermal average of the oscillator strengths of the optically active ($J=1$) and optically inactive ($J=2$) excitons.[5] Figure 3 shows the light-hole (lh) reflectivity data and fit (solid line) of the 15-nm well. The value of the phase constant used for the 15-nm light-hole data is 2.9 rad while the phase value used to fit the 15-nm heavy-hole data in Fig. 1 is 2.7 rad. The two independent estimates of the phase constant are identical (to within 0.2 rad) which further validates our model. The radiative linewidth of the light-hole resonance is estimated to be $25(-7)$ μeV. The weaker reflectivity modulation is attributed to the smaller oscillator strength, as well as a larger extrinsic linewidth of the light hole ($\hbar\Gamma_{s\text{-lh}}\approx150$ μeV; $\hbar\Gamma_{s\text{-hh}}\approx80$ μeV).

Figure 4 shows the reflectivity at two temperatures. The redshift seen in the peaks is due to the reduction in the band gap with an increase in the lattice temperature. Even at a lattice temperature of 150 K we can observe reflectivity modulation of a few percent. Significant reduction may be seen at lattice temperatures exceeding 120 K when LO phonons begin to contribute substantial-

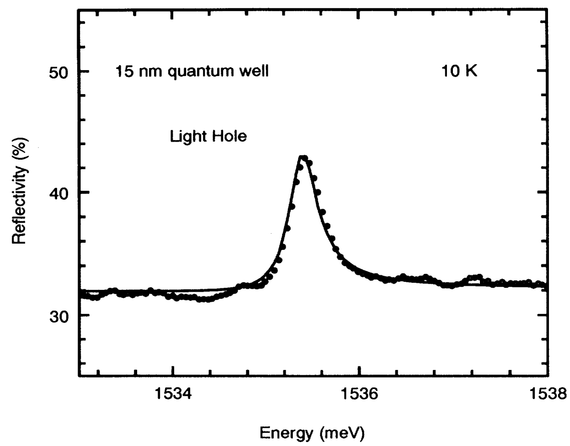

FIG. 3. Light-hole reflectivity of the 15-nm quantum well at 10 K. The solid line is the fit. The value of the phase constant used is nearly the same as that used to fit the heavy-hole data. The weaker reflectivity modulation is attributed to the smaller oscillator strength of the light-hole as compared to the heavy-hole oscillator strength.

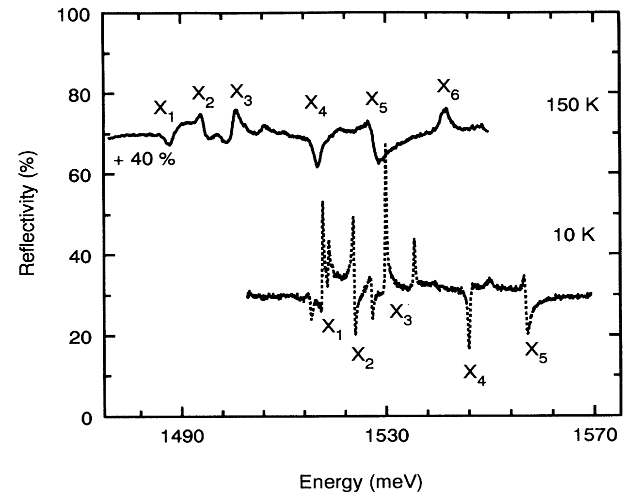

FIG. 4. Reflectance data at two temperatures show that the large reflectivity modulation is quenched by the onset of LO-phonon vibrations at lattice temperatures greater than about 120 K.

ly to the extrinsic linewidth.

In summary, we have shown that the microscopic model for the dielectric function accounts for the observed reflectivity of single quantum wells. We have also shown that the magnitude of the reflectivity modulation is a very sensitive measure of the extrinsic linewidth broadening mechanisms. Based on the magnitude and width of the reflectivity peak, we have deduced the intrinsic radiative linewidth which is in reasonable agreement with the value of the oscillator strength obtained from photoluminescence lifetime measurements. The reflectivity line shape was shown to be governed by the thickness of the cap layer. The large reflectivity modulation in the case of high-quality unintentionally doped samples was observed to persist at temperatures well over 100 K.

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[16]The dielectric function reduces to the local Lorentzian function when $\hbar\Gamma_s \gg \Delta E_{LT}$, where Γ_s is the linewidth broadening due to the extrinsic scattering process.