Review of intrinsic and extrinsic optical properties of GaAs quantum wells

VIVEK SRINIVAS*
Department of Electrical Communication Engineering, Indian Institute of Science, Bangalore 560 012, India.

Abstract

The intrinsic optical properties of excitons in quantum wells include rapid radiative recombination, fast electron spin relaxation and high reflectivity modulation. We show that these properties are altered in a fundamental way due to phonon, exciton–exciton defect, and impurity scattering.

Keywords: Excitons, GaAs, qantum wells, polaritons, reflectivity, photoluminescence.

It is of fundamental importance to be able to separate the intrinsic optical properties from the extrinsic effects[1–9]. Recently, it has been possible to grow quantum well samples that have extremely low defect concentration[1–4]. In this work we show that scattering by defects influences the photoluminescence linewidth, the electron spin relaxation rate, the radiative lifetime and the reflectivity of GaAs quantum wells.

In the presence of lattice defects or impurities, the free exciton can become localized which lowers its energy. We have observed that bound excitons can also broaden the homogeneous linewidth. The net low-temperature linewidth, taking into account the intrinsic effects as well as the extrinsic ionized impurity and bound exciton scattering, is given by[4]

$$\Gamma = \Gamma_0 + \gamma T + \Gamma_+ \, e^{-E_h/k_b T} + \Gamma_{BX}\left(1 + \frac{\rho_{2D} \, k_b T}{N_T} e^{-E_{B\backslash}/k_h T}\right)^{-1} + \cdots, \tag{1}$$

where Γ_0 is the intrinsic linewidth due to radiative decay and spin relation of holes, and ρ_{2D}, the two-dimensional density of free exciton states. E_{BX} is the energy difference between the bound exciton state and the lowest free exciton state, γ, the longitudinal acoustic phonon broadening term, Γ_-, the ionized impurity term, and Γ_{BX}, the bound exciton scattering term. The typical broadening for the extrinsic and intrinsic processes is illustrated in Fig. 1.

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
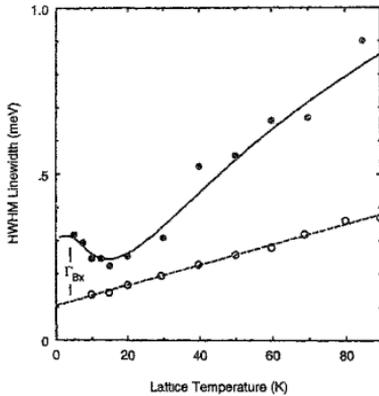
FIG. 1. Photoluminescence linewidths as a function of the lattice temperature for the 32.5 nm quantum well. The dashed line is a linear fit to the data (open circles) for the intrinsic quantum well. The solid line is the theoretical fit, discussed in the text, to the data (filled circles) for the extrinsic case.

the wave vector along the growth direction that causes the exciton polarization to couple strongly to the radiative modes[8,9]. The decay rate and hence the intrinsic homogeneous radiative linewidth is proportional to the oscillator strength per unit area.

Thermalization causes the excitons to occupy a Maxwell–Boltzmann distribution in energy. Scattering by defects localizes the exciton polarization with a localization length $2\Lambda_0$, leading to a spread of the in-plane momentum q, *i.e.*, the in-plane momentum for a given in-plane kinetic energy is no longer a delta function in momentum space but is instead given by a point spread function. Assuming Gaussian statistics for the localization, we may derive the net exciton momentum distribution function by convolving the above independent distributions to be

$$P(q) = \frac{1}{k_i^2} \exp\left[-\frac{q^2}{2k_i^2} \right] \text{ where } k_i^2 \equiv \frac{m_\perp k_B T}{\hbar^2} + \left(\frac{2\pi}{\Lambda_0} \right)^2. \tag{2}$$

At finite temperatures, the heavy and light hole exciton states are partially ionized into free carriers. Taking into account the thermodynamic equilibrium between the exciton and the free carrier states in a 2-D system, we may derive the exciton population in terms of the absorbed photon density N, as

$$r_x \equiv \frac{N_x}{N} = 1 - \frac{K_e}{2N} \left(\sqrt{1 + \frac{4N}{K_e}} - 1 \right) \tag{3}$$

where $K_e = N_e N_h / N_g = \dfrac{m_e m_h}{m_x} \dfrac{k_b T}{\pi \hbar^2} \exp\left[\dfrac{-E_b}{k\,T} \right]$,

N_x = exciton density; N_e = electron density; N_h = hole population; and

m_e, m_h, and m_λ are effective mass of electron, hole and exciton, respectively.

The excitonic population factor, r_X, is determined by the lattice temperature, the exciton-binding energy E_h, and the excitation intensity N. By assuming that the rate equation is dominated by the exciton recombination, the net radiative lifetime can be derived to be:

$$\tau^{-1} = r_X \int_0^{k_0} \left\langle \tau_q^{-1} \right\rangle P(|q|) \, dq \approx \frac{3}{4} \tau_0^{-1} \left(\frac{2 m_\tau}{\hbar^2 k_0^2} k_B T + \frac{2\lambda_0^2}{\Lambda_0^2} \right)^{-1} r_X; \text{ for } k_c \gg k_0 \qquad (4)$$

where $\left\langle \tau_q^{-1} \right\rangle$ is the radiative recombination rate averaged over the L and T modes[10] and the lifetime of the exciton with zero in-plane momentum. It is evident that the lifetime near $T = 0$ K is determined by the coherence length $2\Lambda_0$.

The exciton spin relaxation rate is determined by the spin relaxation processes of the electrons and holes, as long as these relaxation times are short compared to the exciton lifetime[5,6]. Our experimental results suggest that the intrinsic electron spin relaxation is fast (a few picoseconds) for quantum wells. However, the electron spin relaxation process can be readily influenced by scattering processes involving phonons, impurities and other excitons. The scattering effects are clearly evident in our intensity and temperature-dependent linewidth and polarization data. Thus the fast intrinsic electron/exciton spin relaxation process was observed only in our best samples of *single quantum wells with wide well widths, narrow* (homogeneously broadened) *intrinsic linewidth*, and *very small impurity content*.

The electron spin relaxation rate due to the DP (D'yakanov and Perel) process in quantum wells can be derived to be[6-8]

$$\tau_{DP}^{-1} = \alpha \tau_p \Omega_{DP}^2 = \alpha \tau_p \frac{a_{42}^2}{\hbar^2} k_z^4 k_\parallel^2 \qquad |k_z| \gg |k_\parallel|, \qquad (5)$$

where Ω_{DP} is the precession frequency due to the conduction band spin splitting, τ_p, the momentum relaxation time, a_{42}, the band-structure splitting parameter which is taken to be 21 eV Å^3 and α, a numerical coefficient that depends on the momentum relaxation mechanism. The momenta parallel and perpendicular to the quantum well plane are given by $k_\parallel$ nd k_z, respectively. *The DP mechanism in a two-dimensional system is greatly enhanced due to the presence of the k_z^4 term.* For a 15-nm quantum well at 7 K, Ω_{DP}^{-1} is around 25 ps. The other electron spin relaxation mechanism to be considered is the spin exchange interaction in excitons. The electron spin relaxation rate due to exchange interaction, as a function of the hole spin lifetime and exchange energy, can be calculated in terms of an average precession frequency that is proportional to the exchange energy. For the case of a short hole spin lifetime, *the spin relaxation time of the electron due to the exchange process is given by*

$$\tau_{ex}^{-1} \approx \tau_h \overline{\Omega}_{exch}^2; \qquad \tau_h < \overline{\Omega}_{exch}^{-1} \qquad (6)$$

where τ_h is the hole spin lifetime, $\overline{\Omega}_{exh} = \overline{\Delta}_{exch}/2\hbar \approx \Delta_{exch}/4\hbar$, the average precession frequency, and $\overline{\Delta}_{exch}$, the exchange splitting between the exciton states $J = 1$ and $J = 2$. By combining the DP and exchange processes, the spin relaxation time of the electron can be expressed as

$$\tau_s^{-1} = \tau_{ex}^{-1} + \alpha \, \tau_p \, \frac{a_{42}^2}{\hbar^2} k_z^4 \, k_{\parallel}^2. \tag{7}$$

We have observed that *the intrinsic electron spin relaxation time is very short at low temperatures*. As the intensity was raised, we clearly observed a polarized signal in the time-resolved photoluminescence (PL). Our time-resolved PL measurements clearly exhibit all the well-understood selection rules for the formation of the heavy hole (Hh) excitons. On exciting the light hole (Lh) by σ^+ light, $-1/2$ Lh and $+1/2$ electronic states are created. The $-1/2$ Lh relaxes to $\pm 3/2$ Hh states. The observation of a fast rise in Γ signal implied that the $-1/2$ Lh exciton is very quickly converted to the $+3/2$ Hh exciton by emission of even parity phonons. The *hole* spin population attains equilibrium distribution in a few momentum relaxation times. The electron spin, however, may not be entirely randomized even after the thermal distribution of energy states has been achieved. The observation of a slow decay of the Hh polarization factor is attributed to be entirely due to electron spin relaxation. By assuming that the hole is totally randomized at $t = 0$, the polarization factor, $P(t) = \left(I^- - I^+\right)/\left(I^- + I^+\right)$ can be expressed as $P(t) = P_0 \exp(-t/\tau_s)$, where P_0 is determined by the generation rates of the spin-up and spin-down electrons. We were able to extend the dynamic range of our relaxation time measurements beyond the temporal resolution of our system by using the time integral of $P(t)$, which equals $P_0 \tau_s$ in our analysis.

Figure 2(a) shows the time integral of $P(t)$ as a function of the excitation intensity. We also plot the integrated PL linewidth for the same excitation level in Fig. 2(b). The homogeneous exciton linewidth can be expressed as $\Gamma = \Gamma_0 + \gamma_{LA}T + \Gamma_{MB} + \Gamma_{imp}$, where Γ_0 is the intrinsic linewidth due to radiative decay and spin relaxation of holes, $\gamma_{LA} \approx 1.7 \, \mu eV / K$ is the longitudinal acoustic phonon scattering term, Γ_{MB}, the many-body interaction term of carriers and excitons, and Γ_{imp}, the impurity broadening term. The dotted line in Fig. 2(b) is a fit to the linearized theory $\left(\Gamma = \Gamma_0 + \gamma E_b a_b^2 N\right)$. Our data indicate that there is a sublinear carrier density dependence. The solid line in Fig. 2(b) is an empirical fit of the linewidth to the function $\Gamma = \Gamma_0 + \beta\sqrt{N}$ where $\beta = 6.5 \times 10^{-7}$ meV-cm and $\Gamma_0 = 0.145$ meV. The solid line in Fig. 2(a) is a fit of the spin relaxation time to the formula in eqn (7), using the measured linewidth for $\tau_p = \hbar/(\Gamma - \Gamma_0)$. The two fitting parameters used are: τ_{ex}, the average spin relaxation time due to the exchange mechanism, and α, the coefficient that is associated with the exciton–exciton scattering process. We deduce the value of P_0 from the high intensity data to be 0.5 ($\pm$ 0.2). The value of τ_{ex} is estimated to be 0.9 ns and α to be 0.4. The lower limit of the
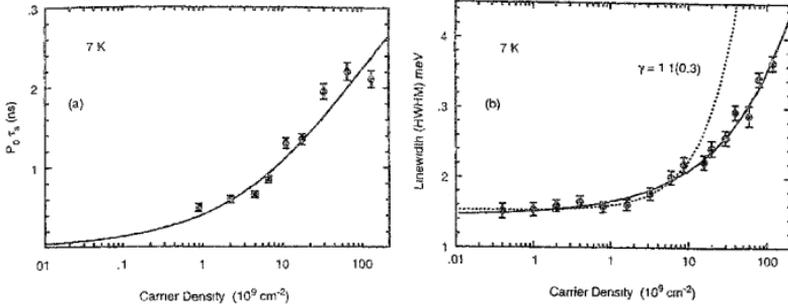
FIG. 2. (a) Average polarization, obtained from the time-resolved photoluminescence measurements *vs* carrier density, and (b) integrated photoluminescence *vs* carrier density at 7 K. The quantum well width is 15 nm and the excitation source is tuned to the light-hole transition.

spin relaxation time of the holes, τ_h, may be obtained from the temperature-dependent PL linewidth measurement of Γ_0 to be (6 ± 2) ps. From these values of τ_h and τ_{ex}, we can estimate the spin exchange energy, using eqn (6), to be between 20 and 50 μeV.

One can indirectly measure the oscillator strength by measuring the radiative life-time. A more elegant method of measuring the oscillator strength is through reflectivity measurements. The reflectivity of single quantum wells is shown to exhibit enhanced modulation of around 70% at resonance. We show that the magnitude of the reflectivity modulation is a very sensitive measure of the extrinsic linewidth broadening mechanisms. On the basis of the magnitude and width of the reflectivity peak, we deduce the intrinsic radiative linewidth that is in reasonable agreement with the value of the oscillator strength obtained from photoluminescence lifetime measurements.

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

$$r_{qw} = \tfrac{1}{2}\left(\alpha' - 1\right)\exp(i k_0 L), \tag{8a}$$

$$\alpha' = \frac{\omega - \varpi - i\Gamma_0 - i\Gamma_s}{\omega - \varpi + i\Gamma_0 - i\Gamma_s}, \tag{8b}$$

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

$$\Gamma_0 = \frac{e^2}{4\pi\varepsilon_0} \; F\frac{\pi}{n m_0 c} \quad \text{and} \quad \beta = \frac{\displaystyle\int_{-L/2}^{+L/2} dz \int_{-L/2}^{+L/2} dz' \, \rho(z)\,\rho(z')\sin\!\left(k_0|z - z'|\right)}{\left|\displaystyle\int_{-L/2}^{+L/2} dz \, \rho(z)\cos(k_0 z)\right|^2}, \tag{9}$$

where F is the oscillator strength per unit area, n, the refractive index in the barrier, m_0, the free electron mass, $k_0 \equiv n\omega/c$ is the photon wavevector, and $\rho(z) \equiv f_e(z_e) f_h(z_h)$ is the product of the confinement functions for the electrons and holes in the well. The peak magnitude of r_{qw} is given by $\Gamma_0/(\Gamma_0 + \Gamma_s)$. We note that the reflectivity resonance $\bar{\omega}$ is red shifted 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

$$R = \left| \frac{r_{12} + r_{qw}(\omega) \exp[\phi]}{1 + r_{12}\ r_{qw}(\omega) \exp[\phi]} \right|^2 , \tag{10}$$

where $\phi = k_0 d$ is the phase change due to propagation in the cap layer of thickness d.

The scattering rate or the extrinsic linewidth, $\Gamma_s = \Gamma_{spin} + \gamma_{LA} T + \Gamma_{LO} + \Gamma_{imp}$ 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 per cent. 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 per cent. According to theoretical expectations, it may be, in principle, possible to observe close to 100% reflectivity modulation as η $\left(\text{this is equal to} \left(\dfrac{\Gamma}{\Gamma + \Gamma_s}\right)\right)$ 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 eqn (3) we obtain $\eta \approx 0.51$ 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 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. 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.

Figure 3 shows the reflectivity at two temperatures. The red shift seen in the peaks is due to the reduction in band gap with increase in the lattice temperature. Even at a lattice temperature of 150 K, we can observe reflectivity modulation of a few per cent. Significant reduction may be seen at lattice temperatures exceeding 120 K when LO phonons begin to contribute substantially to the extrinsic linewidth.

In conclusion, we have identified the intrinsic and extrinsic processes in the photoluminescence linewidth, lifetime, polarization and reflectivity measurements. The effects of impurity and phonon scattering on the optical properties of GaAs quantum wells have been quantitatively established.

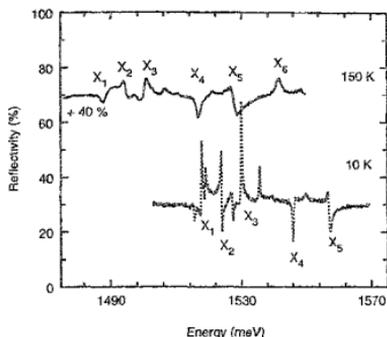This paper is a review of the work reported by Srinivas *et al*[1-4].

FIG 3 Reflectance data at two temperatures show that the large reflectivity modulation is quenched by the onset of LO phonon vibrations at lattice temperatures ≥ 120 K.

References

1. SRINIVAS, V., HRYNIEWICZ, J., CHEN, Y. J. AND WOOD, C. E. C.

Intrinsic linewidths and radiative lifetimes of free excitons in GaAs quantum wells, *Phys Rev B*, 1992, 46, 10193–10196 .

2. SRINIVAS, V., CHEN, Y. J. AND WOOD, C. E. C.

Spin relaxation of two-dimensional electrons in GaAs quantum wells, *Phys Rev B*, 1993, 47, 10907–10910

3 SRINIVAS, V , CHEN, Y J. AND WOOD, C. E. C.

Reflectivity of two-dimensional polaritons in GaAs quantum wells, *Phys Rev B*, 1993, 48, 12300–12303

4 SRINIVAS, V., CHEN, Y J AND WOOD, C. E. C.

Bound exciton induced photoluminescence broadening in GaAs quantum wells, *Solid St. Commun.*, 1994, 89, 611–614.

5. UENOYAMA, T. AND SHAM, L. J

Carrier relaxation and luminescence polarization in quantum wells, *Phys Rev B*, 1990, 42, 7114–7123.

6. DAMEN, T C., VIÑA, L., CUNNINGHAM, J E., SHAH, J. AND SHAM, L. J.

Subpicosecond spin relaxation dynamics of excitons and free carriers in GaAs quantum wells, *Phys. Rev Lett.*, 1991, 67, 3432–3435

7. D'YAKONOV, M. I. AND PEREL', V I.

Spin orientation of electrons associated with the interband absorption of light in semiconductors, *Sov Phys , JETP*, 1971, 33, 1053–1059.

8. ANDREANI, L. C. AND BASSANI, F.

Exchange interaction and polariton effects in quantum-well excitons, *Phys Rev B*, 1990, 41, 7536–7544.

9. TASSONE, F., BASSANI, F AND ANDREANI, L. C.

Quantum well reflectivity and exciton-polariton dispersion, *Phys Rev B*, 1990, 45, 6023–6030.

10 SCHMITT-RINK, S., MILLER, D. A B. AND CHEMLA, D S

Theory of linear and non-linear optical properties, *Phys. Rev B*, 1987, 35, 8113–8115.