Intrinsic linewidths and radiative lifetimes of free excitons in GaAs quantum wells

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(Received 8 July 1992)

The giant oscillator strength for radiative transitions of excitons in quantum wells is largely due to the macroscopic polarization of a two-dimensional system. At low temperatures the large oscillator strength leads to a short radiative lifetime of free excitons. We investigate the photoluminescence linewidths and lifetimes of free excitons in a series of extremely high-quality GaAs quantum wells as a function of lattice temperature, excitation intensity, and quantum-well width. With only negligible defect states in our quantum-well sample, we are able to correlate the time-resolved data with temperature-dependent linewidth measurements on the series of quantum wells to estimate the homogeneous linewidth and acoustic-phonon scattering rate of free excitons. Our studies show that thermalization of the excitonic states, ionization into free carriers, and a reduction in the coherence volume of the exciton polarization due to defect scattering, lead to a decrease in the net radiative recombination rate.

The exciton dynamics of quantum wells (QW's) has been an extensively studied subject for many years. Previous work in cw photoluminescence (PL) and photoluminescence excitation (PLE) presented a good understanding of the exciton transition energies of QW's and many of the temperature-dependent effects in PL linewidths.[1-4] Recent time-resolved studies of various scattering processes of QW excitons have also resolved some of the fundamental issues involved in the lifetime of the two-dimensional (2D) exciton.[5-14] However, so far there is not a single experiment that can directly connect the scattering processes, measured by the time-resolved experiments, to the measured intrinsic linewidth of a 2D exciton. This is mainly due to limitations of the sample quality: various defects in typical QW samples broaden the linewidth of the PL and PLE spectrum, which makes it difficult to determine experimentally the intrinsic exciton linewidth. In this paper we present PL and PLE linewidth and lifetime results of free excitons in a series of extremely high-quality GaAs QW's as a function of lattice temperature, excitation intensity, and QW width. With only negligible defect states in the QW's, we are able to correlate the time-resolved lifetime data with temperature-dependent linewidth measurements to establish the homogeneous linewidth of 2D free excitons. We also measure the acoustic-phonon scattering rate of the free excitons directly from the PL linewidth measurements. Our studies show that thermalization of the excitonic states, ionization into free carriers, and a reduction in the coherence volume of the exciton polarization due to defect scattering all contribute to a decrease in the net radiative recombination rate. We conclude that exciton ionization and scattering by defects and acoustic phonons must be taken into account to obtain a qualitative understanding of the temperature dependence of radiative lifetime.

The free-exciton linewidth Γ of a QW can be expressed as follows:[3,4]

$$\Gamma=\left|\frac{\partial E_n}{\partial L_z}\langle\delta L_z\rangle\right|+\Gamma_I+\gamma T+\frac{\Gamma_{\mathrm{LO}}}{\exp(\hbar\omega_{\mathrm{LO}}/K_BT)-1}+\Gamma_0. \tag{1}$$

The first two terms in Eq. (1) are related to the imperfections in a QW: the first term, an inhomogeneous linewidth broadening, arises from the QW width fluctuation δL_z, and the alloy fluctuation (which is ignored in our discussion). The second term, Γ_I, which is induced by impurities, can be derived from the ionized impurity scattering[3] and the bound-exciton scattering effects.[4] The third term is due to acoustic-phonon scattering. Optical-phonon scattering (mainly from the longitudinal-optical phonon) leads to the fourth term in Eq. (1). The last term is the intrinsic linewidth of the 2D exciton. The intrinsic linewidth Γ_0 is determined by a number of scattering processes: radiative recombination, spin-flip scattering, exciton-exciton scattering, exciton-carrier scattering, etc. Recently[12] it has been shown that the spin-flip scattering time of the heavy hole is around 5 ps (for an 8-nm QW). For a free exciton, assuming that the electron relaxes with the hole, the spin-flip scattering is the fastest (and thus the dominant) process, which leads to an intrinsic linewidth of ≈ 125 μeV.

Following Andreani, Tassone, and Bassani,[14] we derive, from Fermi's "golden rule," the intrinsic radiative lifetime of the exciton as

$$\tau_q^{-1}=\frac{e^2}{4\pi\epsilon_0}F\frac{\pi}{nm_0c}\times\begin{cases}k_0/(k_0^2-q^2)^{1/2}, & T \text{ mode}, \quad |q|\le k_0\\ (k_0^2-q^2)^{1/2}k_0, & L \text{ mode}, \quad |q|\le k_0\\ 0, & |q|>k_0,\end{cases} \tag{2}$$

where $F \equiv |\phi(0)|^2 2|\rho_{cv}|^2/m_0\hbar\omega_0$ is the exciton oscillator strength per unit area, $\hbar\omega_0$ is the transition energy, n the refractive index of the semiconductor, m_0 the free-electron mass, q the exciton in-plane momentum, and $k_0 \equiv n\omega/c$ the photon wave vector in the semiconductor. We note that both F and $\hbar\omega_0$ are well-width dependent.

The rate equation for the carrier density N_c can be written as

$$\frac{dN_c}{dt} = -\sum_q \frac{N_{x,q}}{\tau_q} - \sum_{q_e,q_h} N_{e,q_e} N_{h,q_h} B_{q_e,q_h} , \quad (3)$$

where τ_q is defined in Eq. (2), N_x, N_e, and N_h are the number of excitons, electrons, and holes in momentum states q, q_e, and q_h, respectively, and B_{q_e,q_h} is the bimolecular band-to-band recombination rate. The nonradiative processes are neglected in Eq. (3).

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 δ 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

$$P(q) = \frac{1}{k_c^2} \exp\left[-\frac{q^2}{2k_c^2}\right]$$

$$\text{where } k_c^2 \equiv \frac{m_x k_B T}{\hbar^2} + \left[\frac{2\pi}{\Lambda_0}\right]^2 . \quad (4)$$

At finite temperatures, the heavy- and light-hole exciton states are partially ionized into free carriers. The thermodynamic equilibrium between the exciton and free-carrier states in a 2D system can be written, using the Saha equation (see, for example, Ref. 15) as

$$\frac{N_e N_h}{N_x} = K_e = \frac{m_e m_h}{m_x} \frac{kT}{\pi\hbar^2} \exp\left[-\frac{E_b}{kT}\right] , \quad (5)$$

where N_x is the exciton density, N_e is the electron density, N_h is the hole population, and m_e, m_h, and m_x are the effective masses of the electron, hole, and exciton, respectively. By particle conservation and charge neutrality we have $N_e = N_h = (N - N_x)$, where N is the number of photons absorbed per unit area. The exciton population in terms of the absorbed photon density N is

$$r_x \equiv \frac{N_x}{N} = 1 - \frac{K_e}{2N}\left[\left[1 + \frac{4N}{K_e}\right]^{1/2} - 1\right] . \quad (6)$$

The excitonic population factor r_x is determined by the lattice temperature, the exciton binding energy E_b, 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

$$\tau^{-1} = r_x \int_0^{k_0} \langle \tau_q^{-1} \rangle P(|q|) dq$$

$$\approx \tfrac{3}{4}\tau_0^{-1}\left[\frac{2m_x}{\hbar^2 k_0^2} k_B T + \frac{2\lambda_0^2}{\Lambda_0^2}\right]^{-1} r_x \quad \text{for } k_c \gg k_0 , \quad (7)$$

where $\langle \tau_q^{-1} \rangle$ is the radiative recombination rate averaged over the L and T modes,[16] and $\tau_0 = [(e^2/4\pi\epsilon_0)F(\pi/nm_0c)]^{-1}$ is 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 sample used in our experiments was 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 layer of 100 nm. The $Al_xGa_{1-x}As$ barrier ($x = 0.3$) between each of the quantum wells and the cap layer on the superlattice was 22.5 nm. To avoid absorption in 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 low-temperature PL and PLE experiments were carried out in a liquid-helium cold-finger dewar, using a triple monochromator and computer-controlled photon-counting system. The sample was mounted on a sapphire disk to avoid strain at low temperatures. Negligible Stokes shifts were observed in the PL and PLE spectra. The PL data exhibit high luminescence efficiencies, narrow free-exciton linewidths, and very weak bound-exciton luminescence, indicating that the sample is of exceptionally high quality.

Figure 1 shows the linewidth versus QW width L_Z, excited with a 780-nm cw source (0.2 W cm^2) at 8 K. The solid curve is an inhomogeneous linewidth calculation[17] based on the first term in Eq. (1), using $\delta L_Z = 0.03$ nm.

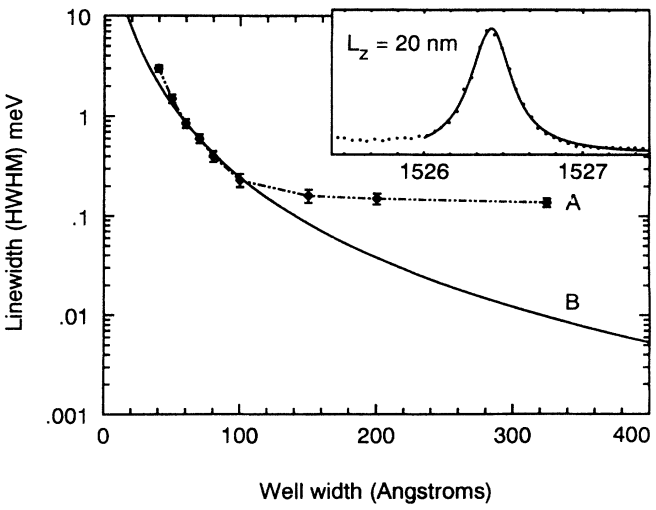

FIG. 1. Photoluminescence linewidth vs quantum well width at 8 K of the quantum wells in the sample. The solid line is the calculation for a well width fluctuation of 0.03 nm. The inset shows the free-exciton PL of the 20-nm QW and a Lorenztian line fit.

The curve fits the narrow-well data quite well, indicating that the inhomogeneous broadening dominates the narrow-well cases. A typical free-exciton PL spectrum is shown in the inset. We note that these are some of the narrowest excitonic linewidths that have ever been measured. For L_Z greater than 15 nm, where the well-width fluctuation effect is relatively small, the measured linewidth saturates to around 125 μeV. By taking into account the spectrometer resolution (better than 25 μeV) and the small inhomogeneous contribution, we estimate an intrinsic homogeneous linewidth between 80 and 120 μeV for the wider QW's. Assuming the dominant component of the intrinsic linewidth is due to the spin-flip scattering of the excitons, we place a lower limit of around 8–10 ps on the spin scattering time in the wider ($L_Z > 10$ nm) QW's.

The linewidths versus temperature of the 32.5- and 15-nm wells are shown in Fig. 2. In the temperature range of 8–70 K, the acoustic scattering process dominates. Therefore, the data provide a direct measurement of the acoustic scattering coefficient γ. The slopes of $3(\pm 0.5)$ μeV/K and $1.7(\pm 0.5)$ μeV/K for the 32.5- and 15-nm wells, respectively, are slightly less than the values of 5 μeV/K for a 27.7-nm QW and 2.5 μeV/K for a 13.5-nm QW reported by Schultheis *et al.*[9] The smaller values are consistent with the theory proposed by Damen *et al.*,[10] which showed that the rate of scattering of excitons out of the radiative volume is roughly half the dephasing rate measured by the four-wave-mixing technique.

Time-resolved luminescence measurements were made with a microchannel plate photomultiplier tube using the single-photon time-correlation technique. A picosecond cavity-dumped dye (STY-8) laser at 4-MHz repetition rate was used as the excitation source. The spectral resolution of the monochromator was approximately 100 μeV half width at half maximum (HWHM) and the temporal resolution of the system approximately 75 ps full width at half maximum (FWHM). The time-resolved photoluminescence data showed the characteristic features of increase in lifetime with temperature and well widths. We also observe a slow rise of exciton luminescence and a long spin relaxation time of excitons, similar to the observations reported by other groups.[10–12] Due to the sensitivity of our single-photon correlation technique and the high-quality sample, we were able to carry out time-resolved measurements over a much larger excitation range (in particular at low excitation intensities) than previously reported. We have also observed excitation intensity dependence in linewidth and spin relaxation. These results will be reported in another paper.

The photoluminescence decay time was measured at temperatures between 8 and 80 K. In these measurements the excitation was linearly polarized at a wavelength of 735 nm (above the light-hole states) and the average intensity was between 0.01 and 2 W cm^{-2}. Figure 3 shows the lifetime-versus-temperature data and calculations [using Eq. (7)] for two of the quantum wells. The data for all the wells showed intensity-dependent photoluminescence lifetimes. The lifetimes of the 32.5-nm quantum well are calculated with the following parameters: $E_b = 5.5$ meV, $m_x = 0.25 m_0$, $F = 16 \times 10^{-3}$ nm^{-2}, and $\alpha = 2 \times 10^4$ cm^{-1}. The parameters used for

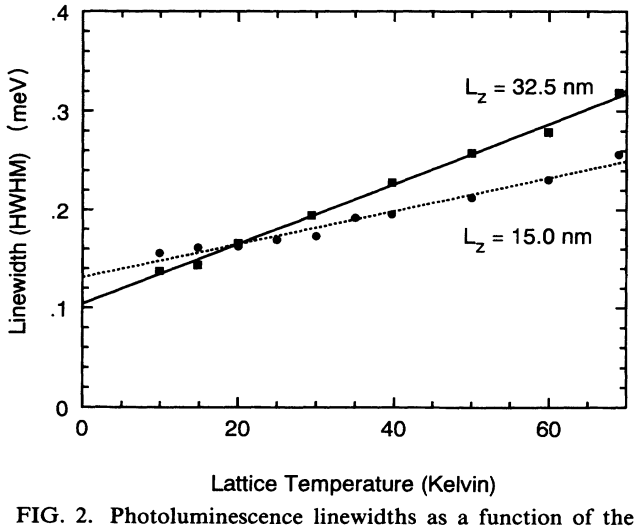

FIG. 2. Photoluminescence linewidths as a function of the lattice temperature for the 32.5- and 15-nm quantum wells. The pump source is a cw laser at excitation intensity of 0.1 W cm^{-2} and wavelength of 750 nm.

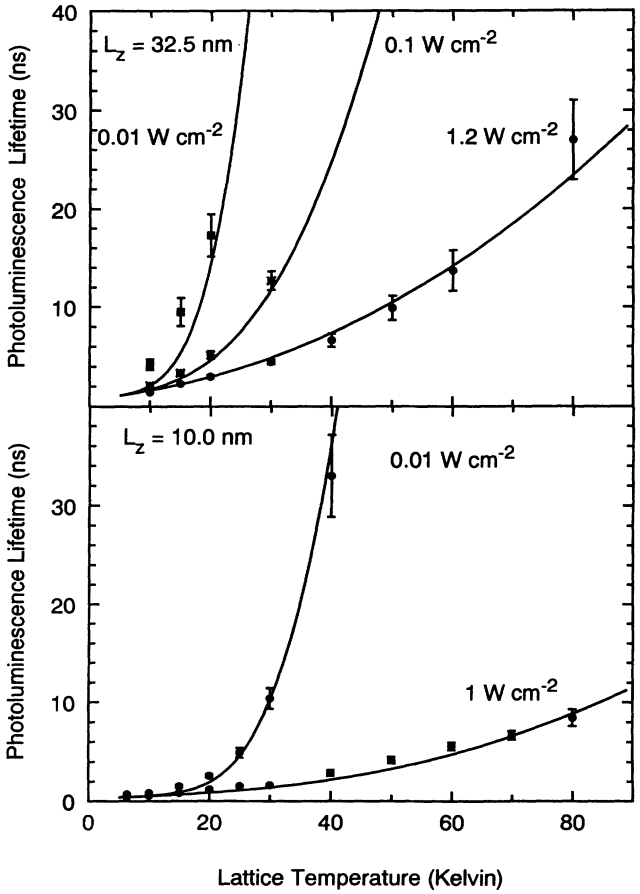

FIG. 3. Radiative lifetime vs lattice temperature for two quantum wells at different excitation intensities. The solid lines are the theoretical fits discussed in the text.

the 10-nm quantum-well calculations are the following: $E_b = 9.2$ meV, $m_x = 0.25 m_0$, $F = 55 \times 10^{-3}$ nm^{-2}, and $\alpha = 2 \times 10^4$ cm^{-1}. The coherence length of the exciton is estimated from the extrapolated $T = 0$ lifetime to be approximately 120 nm.

The radiative recombination rate as a function of intensity for a 15-nm quantum well at 25 K is plotted in Fig. 4. The parameters used in the calculation are $F = 30 \times 10^{-3}$ nm^2, $E_b = 7.75$ meV, and an absorption coefficient of 5×10^4 cm^{-1}. We observe that the lifetime decreases with intensity, saturating to the intrinsic exciton lifetime only when $N \gg K_e$. For the parameters listed above $K_e = 1.6 \times 10^9$ cm^{-2}. At higher intensities, a larger fraction of the photogenerated carriers exist as excitons, resulting in an enhancement of the radiative-recombination rate.

The values for the oscillator strengths and binding energies used in our calculations are generally available in the literature.[18] The only adjustable parameter in our calculations is the absorbed photon density, which serves as a scaling factor and was estimated from the incident power density and the band-to-band absorption coefficient in the quantum wells. The values used for the absorption coefficient (between 2 and 5×10^4 cm^{-1}) agree very well with those reported by other groups.[19]

In summary, we have demonstrated a quantitative understanding of the physics of the exciton dynamics in quantum wells in the regime of low excitation intensity. The results are of general validity and hold for GaAs/$Al_xGa_{1-x}As$ as well as other semiconductor systems. The homogeneous linewidth of free excitons has been directly estimated from PL and PLE measurements. The temperature-dependent acoustic-phonon scattering of excitons was clearly demonstrated and the scattering coefficient measured by photoluminescence experiments. We have shown that the thermodynamic equilibrium between free carriers and excitons plays an important role in the exciton dynamics. We have also shown, both theoretically and experimentally, that the thermalization of the excitonic states (temperature), the scattering and ionization effect of defects (coherence length), and carrier density (excitation intensity) all play an important role in determining the radiative recombination rate of 2D excitons.

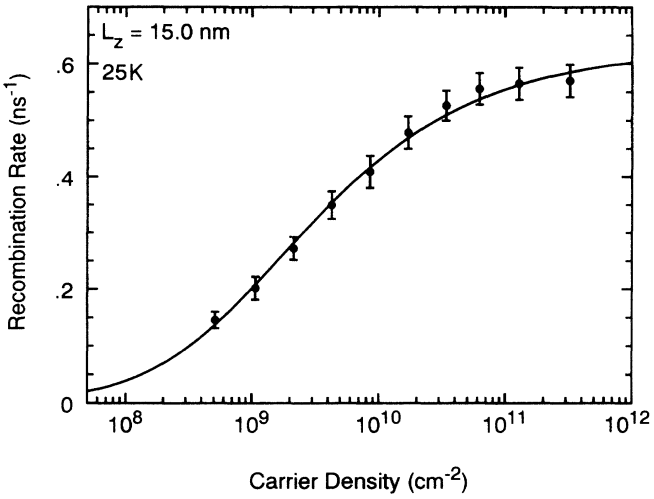

FIG. 4. Radiative recombination rate vs excitation intensity at a fixed lattice temperature of the 15-nm quantum well. The solid line is the theoretical calculation as discussed in the text.

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