# Luminescence linewidths of excitons in GaAs quantum wells below 150 K

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We present detailed experimental studies and a theoretical model of the linewidth as a function of temperature ($<150$ K) for both heavy-hole and light-hole excitons in a GaAs-$Al_xGa_{1-x}As$ quantum well. The contributions to the linewidth of the exciton luminescence include interactions with polar optical phonons, with acoustic phonons (via deformation and piezoelectric potentials), with ionized impurities, and with fluctuations in the thickness of the well. We find that the linewidth increases sublinearly with temperature in the range 0 K to about 70 K but increases more sharply as the temperature is raised still higher.

## I. INTRODUCTION

The optical properties of a quasi-two-dimensional (Q2D) system exhibit many unique characteristics which differ from those of bulk semiconductors. A most important and interesting feature is that the excitonic nonlinear absorption persists and is observed[1-4] even at room temperature. Not only is the underlying physics interesting, but also the prospects for new optical devices are promising.[4-6] In a GaAs/AlGaAs multiple quantum well, the half-width at half maximum (HWHM) of the heavy-hole (HH) exciton line shape as a function of temperature ($T$) has been studied using absorption by Chemla *et al.*[6] ($4<T<300$ K) and by Iwamura *et al.*[7] ($100<T<500$ K), respectively. The main contributions to the linewidth of excitons is attributed to lattice interactions via polar optical phonons and to inhomogeneous fluctuations in the thickness of the well.[6,7] Our experience in the study of the electron transport in a heterojunction structure[8] would suggest that these contributions are dominant only at high temperatures. In the low-temperature range, acoustic phonon scattering should be the dominant mechanism. The reason is that, at low temperatures, the linewidth of excitons is proportional to the phonon population which goes linearly with $T$ for acoustic phonons but is negligible for polar optical phonons.

The main purpose of this paper is to examine, both theoretically and experimentally, the linewidth of excitons in quantum wells as a function of temperature due to scattering by acoustic and polar optical phonons, by ionized donor impurities, and by well thickness fluctuations, and to discover the dominant mechanisms. We focus our discussion here on the linewidth in the low-temperature range ($T<150$ K) since for higher temperatures the line shape is broadened to such an extent that HH and light-hole (LH) excitons overlap and the experimental accuracy of HWHM is poor. In Sec. II we describe our experimental technique and results for the linewidth of the exciton as a function of temperature. In Sec. III a theoretical model is presented for calculating the linewidth due to various scattering mechanisms. Finally, in Sec. IV a discussion of our numerical results is given.

## II. EXPERIMENTAL RESULTS

Temperature-dependent photoluminescent spectra were measured on an undoped, 20 period GaAs/AlGaAs multiple-quantum-well structure grown by molecular-beam epitaxy. The nominal thickness of the quantum wells was 20 nm, and the linewidth at low temperatures (see Fig. 1) is consistent with an average fluctuation in the thickness of the wells of about $\pm 1$ monolayer. The low-temperature spectrum indicates the presence of impurities, probably donors, in the wells. Since this would distort the full-width at half maximum of the luminescence line shapes of the heavy-hole exciton (and since the light-hole exciton peak lies on the high-energy tail of the heavy-hole exciton line shape and is also distorted), the HWHM were determined by visual inspection as indicated in Fig. 1. The linewidths determined in this way in our sample are consistent with linewidths measured in excitation spectra. Also, the peak positions of the heavy excitons were identical, within experimental error, for photoluminescence and excitation spectra. Thus the exciton linewidths in our sample are homogeneously broadened.

The sample was excited with about 0.1 W/cm$^2$ from a He-Ne laser and the spectra were measured with a double grating spectrometer coupled to a cooled GaAs photomultiplier tube which was monitored with a photon counting system. Spectra were measured at temperatures from 5.4 to about 150 K at which point the linewidths of the luminescence for the heavy- and light-hole excitons were comparable to the splitting between them. As the temperature was raised, the populations of the light- and heavy-hole exciton were increased in such a way that the luminescence intensity from light-hole exciton relative to that of heavy-hole exciton rose from 0.35% at 5.4 K to about 20% at 105 K (see Fig. 1).

The experimental HWHM are given as a function of temperature in Fig. 2 for both the heavy- and light-hole excitons. For temperatures above 150 K, the line shapes for the light- and heavy-hole excitons have broadened to such an extent that the line shape of the light-hole exciton is barely distinguishable as a high-energy shoulder on the heavy-hole exciton. Due to this effect, HWHM measurements above 150 K for the light-hole exciton and the heavy-hole exciton were not attempted. Thinner wells would separate the peaks of the light- and heavy-hole excitons further, but would make photoluminescence measurements on the light-hole exciton very difficult at low temperatures due to thermal population effects.

In Fig. 2 we see that (1) for the heavy-hole exciton, HWHM grows sublinearly as the temperature is increased from 5.4 to 70 K but rises more sharply as temperature is raised higher than 70 K; and (2) for the light-hole exciton, HWHM as a function of temperature behaves similarly to that of the heavy-hole exciton but with a smaller differential increase with temperature.

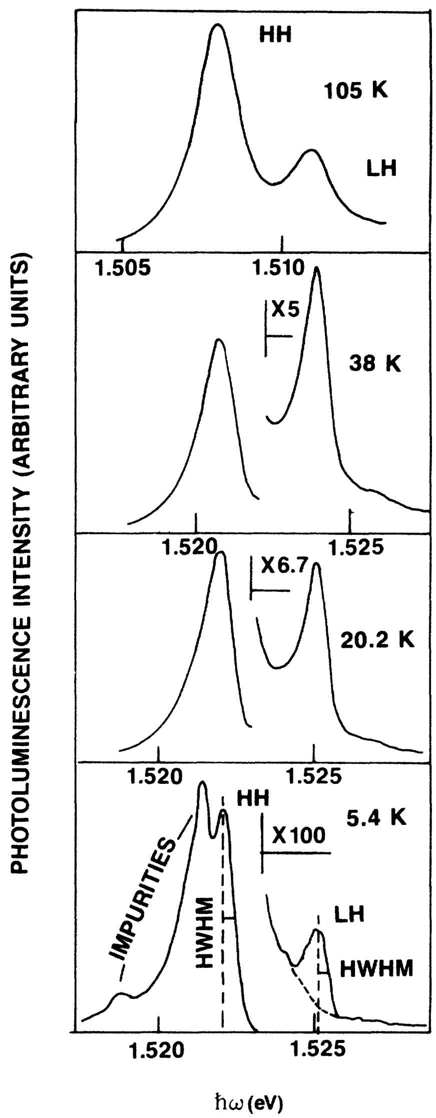


FIG. 1. Temperature-dependent photoluminescent spectra of the MBE grown, 20 period, GaAs/AlGaAs multiple-quantum-well structure.

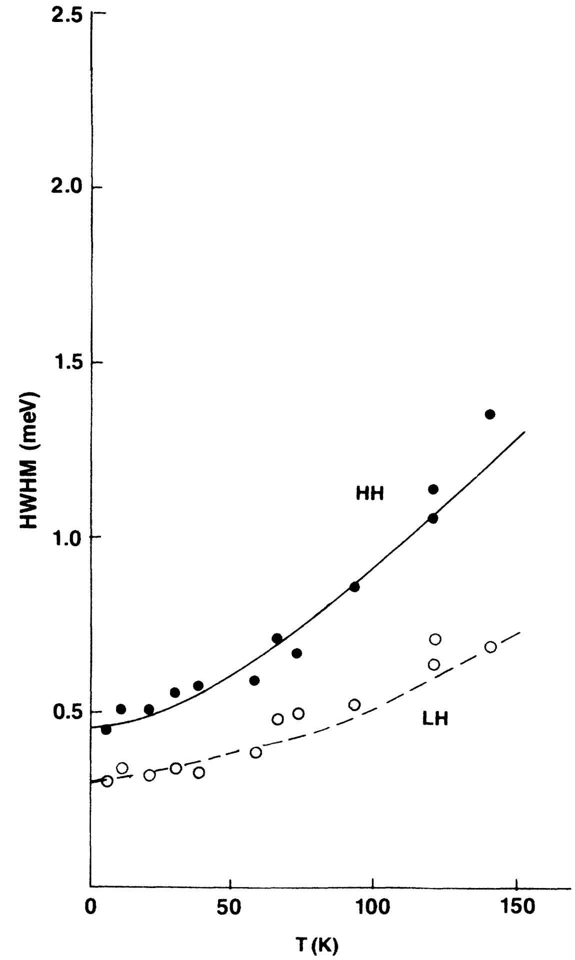


FIG. 2. Experimental and theoretical HWHM of excitons as a function of temperature for both the heavy (HH—solid line) and light (LH—dashed line) excitons.

## III. THEORETICAL CONSIDERATION

In order to explain the experimental results shown in Fig. 2, we calculated the linewidth broadening due to various scattering mechanisms. Because the barrier width of the multiple quantum wells is so thick, we may consider the active regions as isolated wells. By using the well-known Fermi golden rule, the transition rate $W^{(j)}$ for various scattering mechanisms can be calculated if the unperturbed wave function of the quasiparticle is specified. The linewidth (HWHM) is then determined by $\Gamma_j = \hbar W^{(j)}/2$ for various scattering mechanisms. We assume that electron-hole pairs which are generated by light are completely confined in a GaAs active layer of thickness $L$ in the $z$ direction, and are free to move along the GaAs/AlGaAs interface, the $\rho$ plane. In this case, the wave function of the exciton in the ground state may be approximated by[9]

$$\Psi(\rho,z_e,z_h,\mathbf{R}) = A e^{i\mathbf{K}\cdot\mathbf{R}} e^{-\beta\rho/2} \cos(\pi z_e/L) \cos(\pi z_h/L) , \quad (1)$$

where $A$ is a normalization constant, $\mathbf{K}$ and $\mathbf{R}$ are the wave vector and the polar vector of the center of mass of the exciton in the $\hat{\rho}$ plane, $\rho$ is the relative distance between electron and hole, $\beta$ is a variational parameter, and $z_e$ and $z_h$ are the respective projections on the $z$ axis of the electron and hole positions. This wave function has been employed to evaluate the binding energy of the exciton in quantum wells by several authors.[9,10]

### A. Phonons

The transition rate $W^{(j)}$ due to phonon-exciton interaction is given by Tait and Weiher[11] as

$$W^{(j)}=2\pi/\hbar \sum_{\mathbf{Q}K_f} |C^{(j)}(\mathbf{Q})|^2[N_f(E_f)+1]N_i(E_i) \times[N_{\mathbf{Q}}\delta(E_f-E_i-\Delta E_{\mathbf{Q}})\delta_{\mathbf{k}_f-\mathbf{k}_i,\mathbf{q}}+(N_{\mathbf{Q}}+1)\delta(E_f-E_i+\Delta E_{\mathbf{Q}})\delta_{\mathbf{k}_f-\mathbf{k}_i,-\mathbf{q}} , \quad (2)$$

where $j$ refers to longitudinal acoustic phonon or polar optical phonon, $Q=(\mathbf{q}^2+\mathbf{q}_z^2)^{1/2}$ is the phonon wave vector with the $\mathbf{q}$ component parallel to the $\hat{\rho}$ plane, $N_i$ $(E_i)$ and $N_f$ $(E_f)$ are the exciton population with initial (final) energy $E_i$ $(E_f)$, $N_Q$ is the phonon population, and $C^{(j)}(Q)$ is the matrix element for scattering. The Dirac and Kronecker $\delta$ functions ensure the conservation of energy and momentum for absorption and emission of phonons. Here, we assume that the polariton (exciton plus photon) behaves excitonlike, thus the dispersion relation is simplified to

$$E_{i(f)}=\frac{\hbar^2K_{i(f)}^2}{2M}+\frac{\hbar^2\pi^2}{2\mu L^2}+E_g-E_{\text{ex}}(L) , \quad (3)$$

where $m_e$ and $m_h$ are the electron and hole masses, $M=m_e+m_h$ is the exciton mass, $\mu=(m_e^{-1}+m_h^{-1})^{-1}$ is the reduced mass, $K_{i(f)}$ is the initial (final) wave vector of the center of mass of the exciton, $E_g$ is the energy gap of the bulk semiconductor in the active layer, and $E_{\text{ex}}(L)$ is the binding energy of the exciton. Furthermore, we assume that when the crystal is excited by a reasonably intense source of excitation, all excitons are populated at $K_i \simeq 0$, thus $N_i(E_i)[N_f(E_f)+1]\simeq 1$. We calculate $W^{(j)}$ for acoustic and polar optical phonon scattering from Eqs. (1)–(3) and consider all phonon mechanisms to be inelastic.

#### *1. Acoustic phonon scattering*

There are two types of scattering potentials: one involving piezoelectric and the other deformation mechanisms. We treat them separately, and present only the main results below. For piezoelectric scattering, we obtain

$$W_{\pm}^{(\text{pz})}(K_i)=\frac{1}{\hbar(2\pi)^2}\left[\frac{4\pi e}{\epsilon}\right]^2\left[\frac{k_BT}{2\rho u^2}\right]\left[\frac{4M\beta^6}{L^2\hbar^3}\right] \times\int_0^{2\pi}d\theta\, I[k=(\pi/L),q]\left[\frac{h^{(e)}}{[\beta^2+(q\, m_h/M)^2]^{3/2}}-\frac{h^{(h)}}{[\beta^2+(q\, m_e/M)^2]^{3/2}}\right]^2 , \quad (4)$$

where

$$I(k,q)=\frac{\pi}{2q^2(k^2+q)^2}\left[\frac{(2k^2+3q^2)\pi}{k}-\frac{k^4(1-e^{-2\pi q/k})}{q(k^2+q^2)}\right] . \quad (5)$$

In the above, $\pm q=\mathbf{K}_f-\mathbf{K}_i$, $\hbar^2K_f^2/2M=\hbar^2K_i^2/2M\pm\hbar uq$, $\theta$ is the angle between $\mathbf{K}_i$ and $\mathbf{K}_f$, $\rho$ is the mass density, $u$ is the acoustic velocity, $h^{(e)}$ $(h^{(h)})$ is the piezoelectric constant for the electron (hole), $\epsilon$ is the dielectric constant, and $\pm$ refers to phonon absorption and phonon emission, respectively. For the case of a very narrow quantum well, i.e., $k \gg q$ and $\mathbf{K}_i \simeq 0$, Eqs. (4) and (5) may be evaluated to give

$$W_{+}^{(\text{pz})}(0)=\frac{k_BTe^{-2MuL/\hbar}}{24\rho u^3\hbar^2}\left[\frac{4\pi e}{\epsilon}\right]^2(h^{(e)}-h^{(h)})^2 . \quad (6)$$

Note that $W_{+}^{(\text{pz})}(0)$ is independent of the variational parameter, $\beta$, and that $W_{-}^{(\text{pz})}(0)$ has been discarded because the phonon emission is not allowed at $K_i=0$. For deformation potential scattering, we obtain

$$W_{\pm}^{(\text{df})}(K_i)=\left[\frac{3\beta^6k_BT}{4\pi\hbar\rho u^2L}\right]\left[\frac{M}{2\hbar^2}\right]\int_0^{2\pi}d\theta\left[\frac{E_c}{[\beta^2+(q\, m_h/M)^2]^{3/2}}-\frac{E_v}{[\beta^2+(q\, m_e/M)^2]^{3/2}}\right]^2 \quad (7)$$

or

$$W_{+}^{(\mathrm{df})}(0)=\frac{3k_BTM}{4\hbar^3\rho u^2L}(E_c-E_v)^2\ , \tag{8}$$

where $E_c$ and $E_v$ are the deformation potentials of the conduction and valence bands, respectively. Note that $W_{+}^{(\mathrm{df})}(0)$ is independent of $\beta$.

### 2. *Polar optical phonon scattering*

Because the optical phonon energy, $\hbar\omega_0$, is larger than the binding energy of an exciton $E_{\mathrm{ex}}(L)$, after a collision of an exciton with an optical phonon, the exciton is either totally ionized or the optical phonon energy is transformed into kinetic energy of the center of mass with elevation of the exciton to an excited state. In the former case, we obtain

$$W_{+}^{(\mathrm{pop})}(0)=\frac{32e^2\omega_0\beta^4m_eN_Q}{\pi\hbar^2L^2}\left[\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon}\right]\int_0^{K_{\max}}dk_hk_h\int_0^{2\pi}d\theta\, I(k,q)[(4k_h^2+\beta^2)^{-3/2}-(4k_0^2+\beta^2)^{-3/2}]^2 \tag{9}$$

with

$$k_0=\left[\frac{2m_e}{\hbar^2}\right]^{1/2}\left[-E_{\mathrm{ex}}+\hbar\omega_0-\frac{\hbar^2k_h^2}{2m_h}\right]^{1/2}\ , \tag{10}$$

$$K_{\max}=\left[\frac{2m_h}{\hbar^2}\right]^{1/2}(-E_{\mathrm{ex}}+\hbar\omega_0)^{1/2}\ , \tag{11}$$

$$q^2=k_h^2+k_0^2+2k_0k_h\cos\theta\ , \tag{12}$$

where $N_Q$ is optical phonon population, $\epsilon_\infty(\epsilon)$ is the high-frequency (static) dielectric constant, $k_h$ is the wave vector for the free hole, and $I(k,q)$ has been defined in Eq. (5). At $K_i=0$, only phonon absorption is allowed. In the latter case, the exciton is in an excited state and the center of mass is moving after the phonon energy has been absorbed. Thus, the linewidth calculation is tedious and lengthy and will be presented elsewhere.[12] However, numerical results are included in the next section.

### B. Ionized impurities

As indicated in Fig. 1, the sample under study contains a certain number of donors in the quantum wells. The calculation of the scattering cross section of free excitons from impurities (neutral and ionized) in a Q2D system is extremely complicated. First, for low energies ($K_i\simeq 0$), the Born approximation is not applicable.[13] Second, because of the quantum confinement of free carriers, the binding energy of an impurity depends not only on the thickness of the active layer but also on the location of the impurity.[14] In view of these difficulties, particularly the first one, we invoke a phenomenological formula. We know that the transition rate, $W^{(\mathrm{imp})}$, is proportional to the number $N^+$ of scattering centers $N$ that are ionized, thus we may write

$$W^{(\mathrm{imp})}(0)=CN^+=W_0e^{-\langle E_b\rangle/k_BT}\ , \tag{13}$$

where $\langle E_b\rangle$ is the binding energy averaged over all possible locations of the impurities. Here $W_0=CN$ is a parameter to be determined from fits of the experimental data.

The following numerical values for GaAs were used in the evaluation of HWHM ($\Gamma_j=\hbar W^{(j)}/2$): $m_e=0.0665m_0$, $m_{\mathrm{lh}}=0.094m_0$ for light hole, $m_{\mathrm{hh}}=0.34m_0$ for heavy hole, $\epsilon=12.9$, $\epsilon_\infty=11.6$, $\hbar\omega_0=36$ meV, $\rho=5.36$ g/cm$^3$, $u=5.22\times10^5$ cm/sec, $|h^{(e)}-h^{(h)}|=0.15$ C/m$^2$, $|E_c-E_v|=7$ eV, $\langle E_b\rangle\simeq10$ meV for donor impurities, $L=200$ Å. The values chosen for hole masses should only be considered representative inasmuch as the issues concerning the energy diagram for the valence band are still controversial. Since these masses enter the linewidths via $M$, the heavy-hole contributions will be sensitive to variations in the masses, although no more so than other assumptions of the theory. By combining Eqs. (6), (8), (9), and (13), we obtain the total HWHM in meV to be

$$\Gamma_{\mathrm{tot}}^{+}=\Gamma_0^{+}+(1.47\times10^{-3})T+\frac{4}{e^{\hbar\omega_0/k_BT}-1}+\Gamma_{\mathrm{imp}}^{+}e^{-\langle E_b\rangle/k_BT} \tag{14}$$

for the heavy-hole exciton, and

$$\Gamma_{\mathrm{tot}}=\Gamma_0^{-}+(1.19\times10^{-3})T+\frac{2.45}{e^{\hbar\omega_0/k_BT}-1}+\Gamma_{\mathrm{imp}}^{-}e^{-\langle E_b\rangle/k_BT} \tag{15}$$

for the light-hole exciton, respectively. Here, $\Gamma_0^{\pm}$ is the linewidth due to the inhomogeneous fluctuations of well thickness and $\Gamma_{\mathrm{imp}}^{\pm}$ is the linewidth due to the fully ionized impurity scattering. They will be determined from experimental data. In Eqs. (14) and (15), the second terms represent the linewidths due to acoustic phonon scattering via deformation and piezoelectric potentials while the third terms are due to optical phonon scattering. To obtain the third terms, we take the final states of the excitons to be $1s$, $2s$, $2p$, and the free electron-hole pair with $\beta$ value from Ref. 9. The theoretical values are approximated to be 4 meV for a heavy-hole exciton in a 200-Å well [third term in Eq. (14)] and 5 meV for a 100-Å well. The latter is consistent with the value of 5.5 meV reported by Chemla *et al.*[6] and Iwamura *et al.*[7] The difference between the theoretical and experimental results for the 100-Å well case may be attributed to the fact that the trial wave function used in Eq. (1) is oversimplified since the finite depth of the potential well and the effective mass mismatch between GaAs and GaAlAs are ignored.[15] A more realistic trial function would provide for penetration of the wave function into the AlGaAs barrier with matching conditions at the interfaces that reflect the mass variation from layer to layer. This would yield an improved ground-state energy, but would have a minimal effect on the temperature dependence of the scattering rates, and thus would not change the overall conclusion of this work.

## IV. DISCUSSION

To fit the experimental data shown in Fig. 2, we choose $\Gamma_0^+=0.45$ meV, $\Gamma_{\rm imp}^+=0.75$ meV for the heavy-hole exciton (solid line), and $\Gamma_0^-=0.3$ meV, $\Gamma_{\rm imp}^-=0.2$ meV for the light-hole exciton (dashed line), respectively. We notice that for both heavy- and light-hole excitons, the theoretical predictions fall slightly below the experimental HWHM. However, our theoretical model does predict that HWHM increases sublinearly in the low-temperature range for both heavy and light excitons. In Fig. 3 we show the contributions to the HWHM from interactions between heavy-hole excitons (solid lines) and light-hole excitons (dashed line) and polar optical phonons and ionized impurities. For the heavy- (light-) hole exciton, we see that (i) at low temperatures acoustic phonon scattering is the dominant mechanism, (ii) when $T$ is larger than 20 K (25 K), ionized impurity scattering begins to make a significant contribution to the HWHM, and (iii) when $T$ is higher than 200 K, polar optical phonon scattering becomes the dominant mechanism. Also, we notice that ionized impurity scattering is stronger for the heavy-hole exciton than for the light-hole exciton. This explains why, in the sublinear region, the HWHM of the heavy-hole exciton increases faster than that of the light-hole exciton, as shown in Fig. 2.

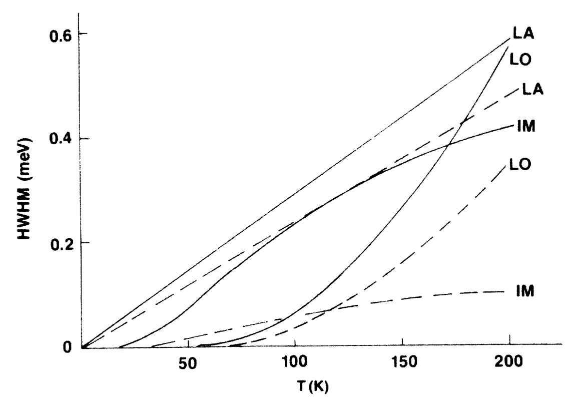


FIG. 3. Contributions to the HWHM from interactions between light-hole (dashed line) and heavy-hole (solid line) excitons and acoustic phonons (LA), polar optical phonons (LO), and impurities (IM).


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