Phonon broadening of excitons in GaAs/$Al_xGa_{1-x}As$ quantum wells

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We have made transmission and reflectivity measurements of the temperature-dependent exciton linewidths for high-quality GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells with well widths ranging between 28 and 340 Å and also for bulk GaAs. The LO-phonon contributions to the linewidths are found to depend only weakly on the well width. Detailed microscopic calculations of the well-width dependence of these linewidths have been made and are shown to be in quantitative agreement with experiment. These calculations provide a physical explanation of the weak dependence of the linewidth on the well width.

The temperature-dependent exciton linewidth arises from exciton scattering by phonons, and is a fundamental property of semiconductor systems. In the last two decades the effects of confinement on carriers, phonons, and carrier-phonon interactions in semiconductor quantum wells and superlattices have attracted considerable attention. The effects of confinement on the electron-phonon-scattering rates, which are important in a number of applications, have been studied especially intensely.[1] Scattering by longitudinal-optical (LO) phonons is of particular interest because it gives rapid carrier-phonon relaxation, and it is the subject of the present study. To date, it has been difficult to make detailed comparison between experiment and theory for carrier-phonon-scattering rates because of such effects as nonequilibrium phonon occupation and screening in the experiments.[1] In contrast, the temperature-dependent exciton linewidth studied here permits a relatively direct comparison between experiment and theory.

A comprehensive picture of the effects of confinement on the temperature-dependent exciton linewidth can be obtained from a study of the linewidth as a function of well width. Such a study provides a systematic picture of exciton-phonon scattering during the transformation between a quasi-two-dimensional system and a three-dimensional system. The most direct method of measuring the exciton linewidth is through transmission (or reflectance) spectroscopy. Its strength lies in its simplicity and in the fact that it can be done at extremely low power densities. However, because it measures the total linewidth, the homogeneous linewidth must be separated from inhomogeneous contributions. Although there are a number of measurements of the exciton linewidths in GaAs quantum wells,[2–7] the differences between these results are as large as an order of magnitude for the same well widths. In addition, Qiang *et al.*[6] reported a substantial monotonic increase of the linewidth with increasing well width due to optical-phonon scattering, and a reduction in the linewidth in narrow quantum wells by a factor of 20 as compared with the bulk.

Inhomogeneous broadening has been ignored in many of these previous studies.[2,3,6] This gives little error when the thermal broadening is much larger than the inhomogeneous broadening; however, when this is not true the errors can be quite large. We believe that the neglect of this issue, and to a smaller extent the neglect of acoustic-phonon broadening, accounts for most of the differences between previous experimental reports of optical-phonon broadening in quantum wells and also for the strong well-width dependence reported by Qiang *et al.*[6] Here, from a study of a number of high-quality samples, we report qualitatively different results for the well-width dependence of the linewidth as compared to Qiang *et al.*[6] Moreover, we show that our results are in agreement with microscopic calculations. A crucial difference between our work and previous studies is that we employ samples with smaller inhomogeneous linewidths. We have obtained data in which the thermal homogeneous linewidth becomes larger than the low-temperature (inhomogeneous) linewidth by as much as a factor of 20 for quantum-well samples with well widths greater than 100 Å. For smaller well widths the inhomogeneous linewidth is larger, and then a deconvolution is necessary to obtain accurate results for the homogeneous linewidths.[4,8]

In this paper we present experimental and theoretical results for the well-width dependence of the LO-phonon contribution to the linewidth of the lowest-energy exciton. In this way we are able to study the systematic transformation of exciton-phonon scattering in going from a quantized quasi-two-dimensional system to a bulk three-dimensional system. We find that the temperature-dependent exciton linewidth depends only weakly on the well width through this range. From microscopic calculations we show that the physical origin of this dependence arises from the increasing number of electron and hole subbands that become energetically accessible as final states as the system goes over to the bulk. The scattering to these higher-lying subbands makes up for the decrease in scattering to the states associated with the lowest-lying electron and hole subbands. It is worth noting that results obtained by early investigators suggested that confinement in GaAs quantum wells did not significantly affect the exciton–LO-phonon broadening parameter.[2,9,10] This preliminary conclusion was not based on complete work but at least partly on an earlier study of the broadening of bulk GaAs which is often misinterpreted.[11] Since then, other studies have reported significantly different results. Nevertheless, as we show here, the LO-phonon broadening depends only weakly on confinement. We also note that a qualitatively different result has been found to exist in II-VI quantum wells,[12,13] where in contrast to GaAs quantum wells the exciton binding energy can become larger than the LO-phonon energy in narrow quantum wells. This reduces the density of states into which the exciton can scatter and leads to an appreciable reduction in LO-phonon broadening for narrow quantum wells.[12]

High-quality (001) $GaAs/Al_{0.3}Ga_{0.7}As$ multiple-quantum-well (MQW), $GaAs/Al_{0.3}Ga_{0.7}As$ single-quantum-well (SQW), and bulk GaAs samples grown by molecular-beam epitaxy (MBE) were studied here. The sample with the narrowest well width (28 Å) was grown with a 2-min growth interrupt at the interfaces to reduce inhomogeneous broadening.[14] The $Al_{0.3}Ga_{0.7}As$ barriers are 100 Å thick in the MQW's, and range between 70 and 400 Å in the SQW's. The SQW samples are capped with GaAs such that the SQW's are at least 400 Å beneath the surface. The MQW and bulk samples were studied in transmission after the substrates were removed by etching. The SQW's were studied through reflectivity at near-normal incidence. In both cases a tungsten lamp with a 0.85-m double monochromator was used as the source, and a Si photodiode with lock-in techniques was used as a detector. The SQW's were found to have extremely narrow linewidths with a half width at half maximum (HWHM) of as little as 110 μeV at $T = 7$ K for a 340-Å well width. (All linewidths here are in terms of HWHM.)

In Fig. 1 the absorption spectra from the 1-μm (bulk) sample and the MQW samples are shown for several temperatures. The linewidth of the lowest-energy exciton was determined by fitting the exciton to a temperature-dependent Lorentzian convoluted with a constant Gaussian which accounts for the inhomogeneous broadening. This convolution is necessary for samples with large inhomogeneous linewidths (here those with well widths of 28 and 50 Å). The results of this fitting procedure are shown in Fig. 2 for the 50-Å MQW. This sample provides the most challenging spectra to analyze because of the relatively large inhomogeneous linewidth and also because of the interference fringes on the low-energy side of the exciton line. Nevertheless, we obtain good fits, except in the low-energy tail of the exciton, where oscillator strengths for both the excitons and the continua remain approximately constant with increasing temperature. The resulting homogeneous linewidths are shown in the inset to Fig. 2 as open circles. For samples with small inhomogeneous linewidths a simpler and sufficiently accurate approximation[4] for such a convolution is to measure the total linewidth and determine the homogeneous linewidth from the relation $\Gamma_{tot}^2 = \Gamma_{inh}^2 + \Gamma_{hom}^2$, where we take the exciton at zero temperature to be totally inhomogeneously broadened ($\Gamma_{inh} = \Gamma_0$). We have done this for samples with well widths larger than 100 Å. This assumption has little affect on the high-temperature linewidths, which are most important in determining the optical-phonon contribution to the broadening. The exciton excited states and continua were fit to a broadened step function.

For the reflectance spectra the excitons were fitted to a linear combination of the real and imaginary parts of a

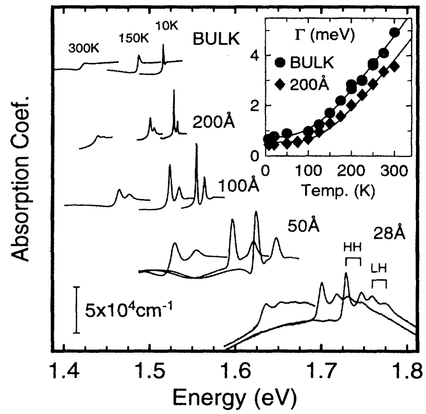

FIG. 1. Absorption spectra for the MQW and bulk samples. Spectra are shown for $T = 10$, 150, and 300 K for each sample except the 100-Å sample for which the intermediate spectra is at $T = 160$ K and the 50-Å sample, for which the high temperature spectrum is at $T = 330$ K. For the 28-Å sample the doublets for both the heavy- and light-hole excitons arise from the large monolayer-high islands which exist at the interfaces of this sample (Ref. 14). The inset shows the total linewidths resulting from the fits.

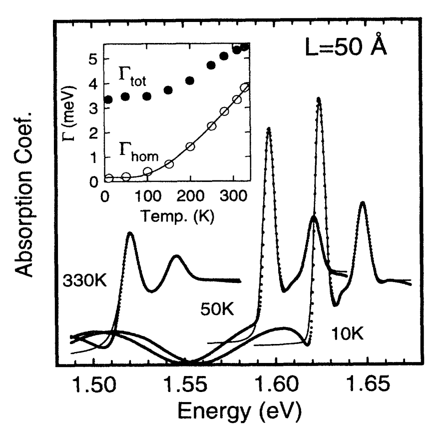

FIG. 2. The measured and fitted absorption spectra for the 50-Å MQW at several temperatures. The inset shows the temperature dependence of the total linewidth (solid circles) and also the homogeneous linewidth resulting from the full fitting procedure (open circles).

Lorentzian. An example of the resulting fits for the 170-Å SQW are shown in Fig. 3. The resulting linewidths (inset to Fig. 3) are very close in value to those measured with transmission on the MQW samples shown in the insets to Figs. 1 and 2. This analysis for the reflectance assumes that multiple reflections are negligible, which can be shown to be a good assumption as long as the linewidth is not too narrow. An important characteristic of both transmission and reflectance (as compared with, for example, luminescence) is that the integrated exciton oscillator strength remains approximately constant as a function of temperature. This provides additional confidence in the results. Moreover, we emphasize that our results are in large part independent of detailed line-shape analysis. When the thermal broadening is much larger than the inhomogeneous linewidth, as it is for most of our samples, we obtain the same result just by measuring the HWHM directly from the spectra. In fact, even if the thermal linewidth is comparable to the inhomogeneous linewidth, the HWHM still gives a sufficiently good measure if the low-temperature (inhomogeneous) linewidth is subtracted in squares to obtain the homogeneous linewidth. This can be shown for the 50-Å sample by using the HWHM (solid circles in the inset to Fig. 2) and comparing to the homogeneous linewidth resulting from the full fitting procedure (open circles).

In general the temperature-dependent homogeneous exciton linewidth is written as

$$\Gamma_{\rm hom}(T)=\Gamma_{\rm hom}(0)+\gamma_{AC}T+\gamma_{\rm LO}n(T) , \quad (1)$$

where the term linear in temperature is due to exciton scattering from acoustic phonons, and the term involving the Bose function $n(T)$ is due to interactions with LO phonons. $\Gamma_{\rm hom}(0)$, which may be due to impurities, is determined from the line-shape analysis and is quite small. We do not discuss temperature-dependent impurity scattering here.[3] An LO-phonon energy of 36.6 meV is used. As the temperature increases, the linewidth increases from the zero-temperature value first because of acoustic-phonon scattering. Above 60 K the optical-phonon contribution becomes important, and eventually it dominates the linewidth. From a fit to Eq. (1) we determine the values for $\gamma_{\rm LO}$ which are plotted in Fig. 4 as a function of well width. We note that the magnitude of $\gamma_{\rm LO}$ is approximately constant over the entire range of well widths. The experimental value of $\gamma_{\rm LO}$ of the bulk sample is shown by an arrow.

In order to obtain results for the optical-phonon contribution to the linewidth, the acoustic-phonon broadening has been taken into account. This contribution is determined by the temperature dependence of the linewidth below 60 K (inset to Fig. 3). Although $\Gamma_{\rm inh}$ is negligible compared to the high-temperature linewidths for most of our samples, it is a significant fraction of the linewidth up to 60 K even in the highest quality samples. For the SQW's we obtain good fits with values of $\gamma_{\rm AC}$ which are relatively constant ([1.9, 1.9, 2.6, and 2.2] μeV/K for the [130, 170, 250, and 340]-Å SQW's, respectively). These values are in generally good agreement with recent measurements of the acoustic-phonon broadening parameter for similar well widths by several groups.[5,7,15–17] Because the acoustic-phonon broadening is a small contribution to the total linewidth at the higher temperatures, the uncertainty in the resulting optical-phonon contribution is considerably less than the uncertainty in $\gamma_{\rm AC}$ for the SQW's. For example, by changing $\gamma_{\rm AC}$ by $\pm 50\%$, $\gamma_{\rm LO}$ changes by only $\pm 20\%$, and this results in considerably poorer fit.

The MQW and bulk samples have larger inhomogeneous linewidths than the SQW's because of either well-width fluctuations or strain fluctuations resulting from the removal of the substrates. For MQW's we have used the values of $\gamma_{\rm AC}$ found for the SQW's at the most similar well width. In the case of the 1-μm bulk sample, we take from the literature a value of $\gamma_{\rm AC}=7$ μeV/K determined from reflectance measurements on a 0.5-μm sample.[18] This value is considerably larger than those for quantum wells, but it is slightly smaller than the value of 8.5 μeV/K determined from four-wave mixing on a 0.2-μm sample.[15] We note that, for the bulk, in contrast to the quantum wells, the contribution to the exciton linewidth from acoustic phonons is comparable to that of LO pho-

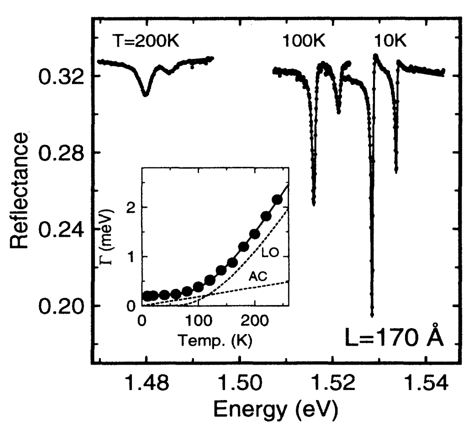

FIG. 3. The measured and fitted reflectance spectra of the 170-Å SQW at several temperatures. The inset shows the total linewidth as a function of temperature, and also the contribution to the fitted linewidth due to LO phonons and the acoustic phonon.

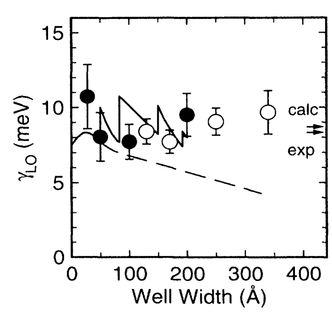

FIG. 4. The LO-phonon broadening parameter as a function of well width for MQW's (solid circles) and SQW's (open circles). The calculated values are shown with (solid line) and without (dashed line) exciton scattering into higher subbands. The measured and calculated bulk values are shown as arrows.

nons at room temperature.[19] Therefore, the 20% difference between previously reported measurements[15,18] of γ_{AC} would lead to approximately a 20% change in γ_{LO} for the bulk.

In previous theoretical work[20–22] we developed a full treatment of LO-phonon contributions to the temperature-dependent exciton linewidth. We performed evaluations of these linewidths in two cases: in bulk semiconductors,[21,22] and in narrow quantum wells[20] such that only one electron and one hole subband are energetically accessible as final states. Physically the linewidth arises from the scattering of the exciton by thermal phonons into higher-lying bound states and also scattering exciton states. We found that in order to obtain a quantitative understanding of the exciton linewidth it is important to include exciton transitions to all electron-hole continuum states as well as to bound exciton states. For bulk GaAs the value of the parameter γ_{LO} was found to be 8.7 meV,[22] and for narrow quantum wells γ_{LO} was found to be 7.5 meV. These results show the remarkable similarity of the values for two- and three-dimensional limits.

Exciton scattering to states arising from higher-lying subbands becomes important for larger well widths.[23] In the present work we have straightforwardly extended our treatment in Ref. 20 to address the full well-width dependence of the linewidth. This has been done by including scattering to the energetically accessible bound and continuum exciton states associated with higher-lying electron and hole subbands which become available as the well width becomes larger in a manner analogous to that done for the lowest subband in Ref. 20, and by including the coupling of the light- and heavy-hole subbands using the Lüttinger Hamiltonian.[24,25] These calculations were made using a variational treatment for the bound and scattering exciton states as used in Ref. 20. For calculational convenience the potential barriers were taken to be infinite, as was done in Ref. 20.

The results of these calculations are shown by the solid line in Fig. 4. In the calculations peaked features in γ_{LO} occur for intermediate well widths when scattering to exciton states associated with higher subbands becomes energetically accessible. These results show how the transformation of the exciton linewidth between the quasi-two-dimensional case and the three-dimensional case occurs as the excitonic spectrum changes. These microscopic results are in good overall agreement with experiment all the way from the narrow wells to bulk. Furthermore, these results show that the physical origin of the weak dependence of the linewidth on well width arises from the change in the effective dimensionality of the electron and hole states as more subbands become energetically accessible with increasing well widths. We note that the scattering to the lowest electron and hole subband,[20] which is shown by the dashed line in Fig. 4, gives a contribution that is linear in the well width L for small well widths, reaches a maximum, and then falls off as $1/L$ for large well widths as it must by normalization.[26] We have made estimates of the size of the features corresponding to exciton scattering into higher subbands in Γ_{LO} vs L, shown in Fig. 4 when realistic well-width fluctuations are included, and we find that these features are then expected to be quite weak. This suggests that they probably would not be observable in real materials systems.

In quantum-well systems like GaAs/AlAs the LO-phonon spectrum changes into modes that are confined to the GaAs or AlAs layers, plus interface modes. These effects of confinement modify the carrier scattering from individual phonon modes. We have included these effects of phonon confinement in our calculations of the exciton linewidth as we did in Ref. 20, and have compared these results with calculations using a bulk model for the phonons. We find that the effects of phonon confinement on the exciton linewidth are $<15\%$ for small well widths and are less for larger well widths. We attribute this result to the fact that the linewidth involves a sum over a complete set of phonon states, which is relatively insensitive to phonon confinement. This result is similar to results obtained recently for the effects of phonon confinement on the carrier relaxation rates in quantum wells and other low-dimensional structures.[1,27]

We now discuss in more detail the origin of the discrepancy between our experimental results and those of Qiang *et al.*[6] We differ in two major respects. Their value for the bulk γ_{LO} (20 meV) is more than twice as large as ours, and they find that γ_{LO} is reduced to as low as 1 meV for well widths below 100 Å. We suggest that the differences between their results and our own arise from two shortcomings in their analysis. First, for the bulk they do not account for the electron-hole continuum. Yet, since the binding energy of the exciton is only 4.2 meV for bulk GaAs, the exciton merges into the continuum above room temperature. Thus these authors are measuring primarily the scattering rate of the ionized electrons and holes, and not that of the exciton. Second, for the quantum wells the inhomogeneous linewidths in their work are as much as 80% of the total linewidth even at $T=500$ K. Yet they simply subtract the inhomogeneous linewidths to determine the thermal linewidths. This seriously underestimates the thermal broadening and also accounts for the strong well-width dependence of the linewidth parameter which they report. This point can be clearly appreciated from the inset to Fig. 2 for the 50-Å MWE by fitting Γ_{tot} instead of Γ_{hom} to Eq. (1).

In summary, we find both experimentally and theoretically that the LO-phonon contribution to the exciton linewidth in $GaAs/Al_{0.3}Ga_{0.7}As$ quantum wells has only a weak dependence on the well width. We have traced this behavior to the systematic changeover of the excitonic spectrum from quasi-two-dimensional to bulklike. We find quantitative agreement between experiment and theory for this behavior.

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[17]There is one exception. In Ref. 15 a value of 45 μeV/K for a 277-Å SQW is reported which is a factor of 2 larger than both our 250- and 340-Å results. We note that an extreme upper bound for γ_{AC} is obtained here by taking the total linewidth at 60 K to be due entirely to acoustic phonons. A lower bound is found by assuming that $\Gamma_{inh}=0$ and using Eq. (1) and adding Γ_0. For the 250-Å SQW this gives an upper bound of 3.9 μeV/K and a lower bound of 1.6 μeV/K; i.e., extreme upper and lower bounds with $\pm 50\%$ of our best value of 2.6 μeV/K.

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