Enhanced exciton-phonon scattering in $In_xGa_{1-x}As/GaAs$ quantum wires

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We have investigated the dephasing of excitons due to scattering by acoustic phonons in quantum wires by means of temperature-dependent time-integrated four-wave mixing. Free-standing $In_{0.135}Ga_{0.865}As/GaAs$ quantum wires with lateral sizes between 29 and 85 nm have been studied. By measuring the phase relaxation time of the excitonic resonance at low carrier densities, we have determined the homogeneous linewidth. From the temperature dependence of the homogeneous linewidth at temperatures between 5 and 30 K, we have evaluated the temperature coefficient γ_{ac}, which is a measure for the exciton-acoustic phonon scattering strength. γ_{ac} is found to increase with decreasing wire width. It is shown that this dependence on the wire width is consistent with a microscopic theory of exciton-acoustic phonon coupling. [S0163-1829(97)07243-3]

Low-dimensional semiconductor heterostructures such as quantum wires and quantum dots have attracted great interest due to their promising application potential in electronics and optoelectronics.[1–3] Still many aspects of their dynamical properties, including exciton dynamics, remain open. In intrinsic semiconductor systems, exciton-phonon scattering gives the dominant temperature-dependent contribution to the exciton lifetime and linewidth at nonzero temperature and low carrier densities.[4] There has been considerable interest in recent years in the effects of confinement and of reduced dimensionality on carrier and exciton scattering rates.

To date, experimental studies of the exciton linewidths in confined geometries have concentrated mainly on the higher-temperature regime ($\geqslant 80$ K) in quantum wells.[5–7] Less work has been done on the regime at lower temperatures, where the homogeneous linewidth can be written as $\Gamma_h = \Gamma_h(T=0\text{ K}) + \gamma_{ac}T$. The coefficient γ_{ac} is related to the scattering rate of excitons by acoustic phonons. Further, most work to date on exciton linewidths has involved photoluminescence or reflectance studies, which then require the determination of the homogeneous linewidth from the inhomogeneously broadened lines.

A well-known technique to measure homogeneous linewidths directly is the investigation of the phase relaxation time T_2 in four-wave mixing experiments.[8,9] With this technique, experiments have been made on quantum wells, mainly in the $GaAs/Al_xGa_{1-x}As$ material system.[8–11] On quantum wires, there are only few experimental results. Mayer *et al.*[12] studied exciton-acoustic phonon scattering in 150-nm-wide $GaAs/Al_xGa_{1-x}As$ quantum wires. To date, however, there are no systematic studies on the wire width dependence of this scattering process. Moreover, phase relaxation processes in quantum wires of other material systems have not been investigated up to now.

In the present work, we report on such a systematic study of the wire width dependence of the exciton linewidth due to acoustic phonon scattering. Free-standing $In_{0.135}Ga_{0.865}As/GaAs$ quantum wires with lateral sizes between 29 and 85 nm were investigated. For the smallest wires, the extension of the exciton wave function (double Bohr radius $2a_B \approx 26$ nm) becomes comparable to the wire width and pronounced confinement effects are expected to occur. In the present structures, the Coulomb correlation effects are already significantly enhanced due to the one-dimensional confinement as shown, e.g., by the increase of the exciton binding energy.[13] We have used time-integrated degenerate four-wave mixing to determine the homogeneous linewidth of the excitonic resonance at varying temperatures between 5 and 30 K. In this temperature regime, the homogeneous linewidth is found to depend linearly on the temperature for all quantum wires, thus allowing us to determine the temperature coefficient γ_{ac}. This coefficient is found to have a value of 10.4 μeV/K for the 85-nm-wide, quasi-two-dimensional quantum wires and to increase by 15% for the smallest quantum wires investigated here. This rise is consistent with theoretical calculations for the wire width dependence of the exciton-phonon scattering process.

The quantum wires were fabricated starting from an $In_{0.135}Ga_{0.865}As/GaAs$ multiple quantum well consisting of 20 $In_xGa_{1-x}As$ wells with a width of 3 nm and an In-content x of 13.5%. The quantum wells are separated by 60-nm-wide GaAs barriers. The wire structures were defined by high-resolution electron-beam lithography and etched by an electron-cyclotron-resonance-enhanced reactive ion etching process using Cl_2/Ar as etching gases.[14] By this method, high-quality quantum wires with a wide range of lateral sizes were obtained. The etch depth was 1.4 μm, in order to assure that all 20 $In_{0.135}Ga_{0.865}As$ quantum wells are etched through. The wire width was determined directly from scanning electron micrographs. For comparison, we also investigated a quasi-two-dimensional reference.

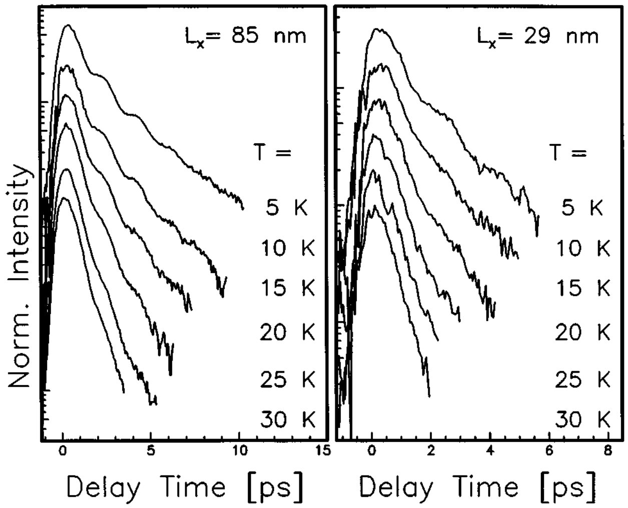

FIG. 1. Evolution of the four-wave-mixing signal with the delay time for 85 and 29 nm wide quantum wires for varying temperatures.

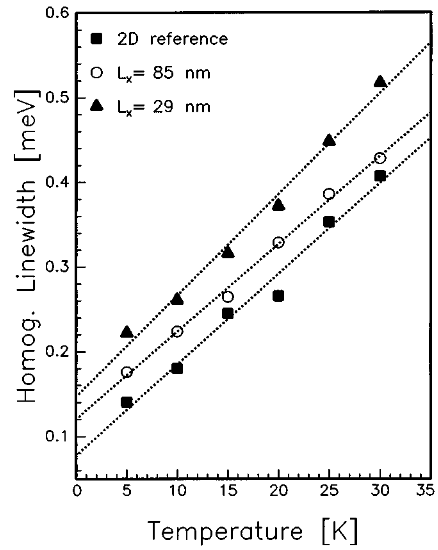

FIG. 2. Temperature dependence of the homogeneous linewidth for three typical structures. The dotted lines are linear fits.

To study the scattering of excitons by acoustic phonons, we have measured the temperature dependence of the homogeneous linewidth. We have used the method of time-integrated, degenerate four-wave mixing in the two-pulse self-diffraction configuration.[9] In this technique, a short laser pulse is split up in two pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, which are both focused upon the sample. The second pulse is delayed for a variable time by an optical delay line. The first pulse induces a macroscopic coherent polarization of the photoexcited excitons. With the coherent part of this polarization left after the delay time, the second pulse builds up a grating and a part of it is diffracted by this grating into the direction $2\mathbf{k}_2-\mathbf{k}_1$. The intensity of the diffracted light scales with the coherent polarization left from the first pulse after the delay time. The coherence is destroyed by phase-breaking mechanisms such as scattering of the excitons by phonons. Therefore, the decay of the four-wave-mixing signal gives a direct measure for the phase relaxation time T_2.[15] The phase relaxation time is connected with the homogeneous linewidth Γ_h by $\Gamma_h=2\hbar/T_2$.[9]

For an excitation source, we used a fs titanium-sapphire laser, pumped by an Ar-ion laser. The titanium-sapphire laser yields pulses of 65 fs length with a spectral width of about 35 meV. The photon energy of the laser was tuned to the middle of the absorption line of the heavy-hole exciton for each wire width individually. To avoid the excitation of higher states, we compressed the pulses to a spectral width of 2.3 meV, using a grating and a slit. The spectral linewidth of the laser was somewhat smaller than the width of the absorption line of the heavy-hole exciton, assuring that only the ground state of the heavy-hole exciton was excited. In this way it was possible to measure the homogeneous linewidth of the excitonic resonance. The time-averaged excitation power was about 15 W/cm^2, leading to a carrier density of 4×10^9 cm^{-2} for the two-dimensional reference. The sample was held in a variable-temperature He gas-flow cryostat. The diffracted laser light was detected by a photomultiplier using standard lock-in technique.

Figure 1 shows the evolution of the four-wave-mixing signal with delay time for two different structures, namely, for 85- and 29-nm-wide quantum wires. The temperature varies from 5 K (upper curves) to 30 K (lower curves) and is indicated at each trace. For clarity, the curves for different temperatures are shifted vertically against each other. From the semilogarithmic representation of the decay curves, one can see that for longer delay times the signal decreases exponentially. Superimposed on this exponential decay is a periodic modulation, which we ascribe to polarization interferences between the donor-bound and the free exciton.[16] As a result of wire width fluctuations on the order of a few nanometers, the luminescence of the quantum wires is mainly inhomogeneously broadened. From the decay time τ of the four-wave-mixing signal we, therefore, obtain the dephasing time T_2 by $T_2=4\times\tau$.[15]

At low temperatures, the dephasing arises due to scattering of the excitons by impurities, compositional fluctuations, interface roughness, or wire width fluctuations. As can be seen in Fig. 1, the dephasing times of the 29-nm-wide wires at the lowest temperatures are shortened by almost a factor of 2 as compared to the 85-nm-wide ones. With increasing temperature the dephasing times of the 85- and the 29-nm-wide quantum wires become considerably shorter, which is a consequence of the growing influence of the scattering of the excitons by acoustic phonons.

This can be seen more systematically in Fig. 2, where the temperature dependence of the homogeneous linewidth is shown for three typical structures, the two-dimensional reference, the 85 and the 29-nm-wide quantum wires. As expected theoretically, the homogeneous linewidth depends linearly on temperature for all structures studied here. Therefore, we use linear fits to estimate the homogeneous linewidths at zero temperature $\Gamma_h(T=0\text{ K})$ and the temperature coefficients γ_{ac}.

Figure 2 shows that the temperature coefficient γ_{ac} for the 85-nm-wide wires is comparable to that of the two-dimensional reference, while the coefficient for the 29-nm-wide wires is considerably greater. As mentioned above, the dephasing times for the 29 nm wires at low temperature are shortened by almost a factor of 2 compared to the 85-nm-

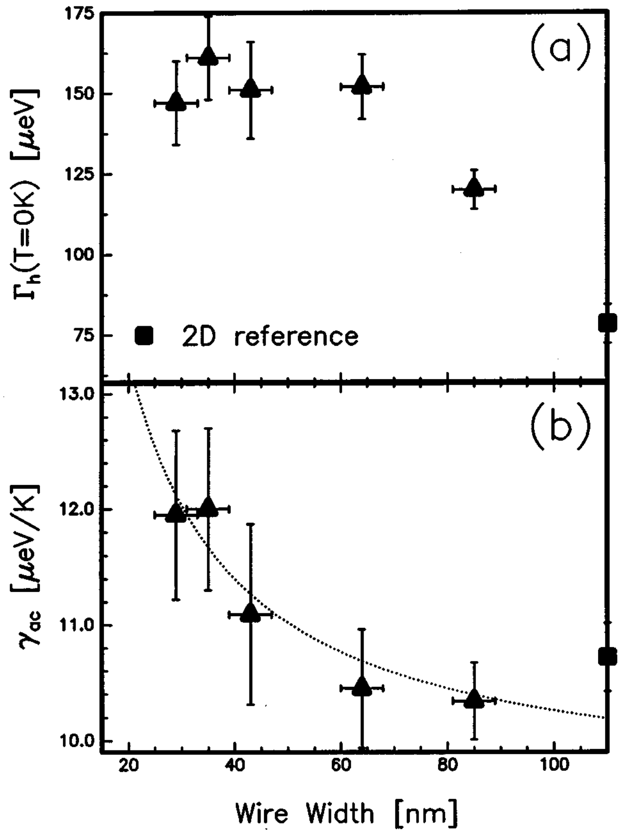

FIG. 3. Wire width dependence of the homogeneous linewidth at zero temperature $\Gamma_h(T=0\ \mathrm{K})$ (a) and of the temperature coefficient γ_{ac} (b). The solid squares denote the values of the two-dimensional reference. The dotted line in (b) shows the result of a theoretical calculation of the wire width dependence of γ_{ac} as described in the text.

wide quantum wires. The systematic dependence of $\Gamma_h(T=0\ \mathrm{K})$ on the wire width is shown in Fig. 3(a). The solid square denotes the value of the two-dimensional reference. $\Gamma_h(T=0\ \mathrm{K})$ rises from 0.078 meV for the two-dimensional reference up to 0.152 meV for the 64-nm-wide quantum wires. Below a wire width of about 50 nm it stays nearly constant.

We suppose that the rise of the homogeneous linewidth at zero temperature with decreasing wire width is associated with the enhanced influence of the scattering of the excitons by wire width fluctuations. Because the surface to volume ratio increases with decreasing wire width, this scattering mechanism will be of growing influence for smaller wires. On the other hand, $\Gamma_h(T=0\ \mathrm{K})$ is nearly constant for wire widths smaller than about 50 nm. This is also the width for which quantization effects due to the lateral confinement of the exciton wave function become important; for example, the exciton binding energy starts to rise here.[13] For such wire widths, the localization of the excitons begins to play an important role. This may limit a further growth of $\Gamma_h(T=0\ \mathrm{K})$.[17]

The coefficient γ_{ac} is a measure for the scattering of the excitons by acoustic phonons. For the two-dimensional reference we determine γ_{ac} to be 10.7 μeV/K. There have been many investigations of exciton-acoustic phonon scattering in GaAs/$Al_xGa_{1-x}As$ quantum wells.[5,9–11,18,19] Most previous publications report smaller values, around 2 or 3 μeV/K, than those obtained here for the temperature coefficient. However, these investigations address quantum wells wider than 10 nm, and it is expected that the scattering rate increases with decreasing quantum well thickness.[11] Ruf *et al.*[6] reported a γ_{ac} of 6.5 μeV/K for a quantum well with a width comparable to the width of the quantum well studied here. Furthermore, we investigate $In_xGa_{1-x}As$/GaAs quantum wells. The strain and the alloy fluctuations in the structures might cause a significant increase of γ_{ac}.

We find that the temperature coefficient γ_{ac} also depends systematically on the wire width and, therefore, gives important information on the effects of confinement for the exciton-acoustic phonon scattering. Figure 3(b) displays the dependence of γ_{ac} on the wire width. The two-dimensional reference, which has a value of 10.7 μeV/K, is denoted by the solid square. For our wide wires, the temperature coefficient is similar to the two-dimensional reference. In the 85-nm-wide wires γ_{ac} is 10.4 μeV/K. This reduction of γ_{ac} might be due to the partial strain release in large, quasi-two-dimensional wires.

However, below about 50 nm, γ_{ac} is seen to increase monotonically and reaches a value of nearly 12 μeV/K for 29-nm-wide wires. This is an increase of about 15% as compared to the large wires, which is well outside of the experimental error.

In order to understand this wire width dependence, we consider the contribution to the homogeneous linewidth given by exciton-acoustic phonon scattering. The homogeneous exciton linewidth in the wires is obtained in perturbation theory in a manner similar to that given previously for bulk[4] and for quantum wells.[20] For wire widths for which kT is less than or of the order of the energy of the highest laterally confined state, we obtain a simple expression for the exciton linewidth:

$$\Gamma_{\mathrm{phon}}=\frac{1}{\hbar(2\pi)^2}\sum_{\lambda}\int_{-\infty}^{\infty}dq_x\int_{-\infty}^{\infty}dq_z N(\mathbf{q}')|V^{(0,\lambda)}(\mathbf{q}')|^2.$$

Here the wire axis is in the y direction, $V^{(0,\lambda)}(\mathbf{q})$ is the matrix element for exciton scattering from the lowest to the λth laterally confined state, and q' is such that $\Delta E_\lambda-\hbar^2 q_y'^2/2M=\hbar\nu_s q'$, where ν_s is the velocity of sound, M is equal to m_e+m_h, and ΔE_λ is the lateral subband spacing to the λth state. From an examination of the dependence of Γ_{phon} on the wire width L_x, we find that $\Gamma_{\mathrm{phon}}\propto\gamma_{ac}T\propto L_x^{-1}$. The L_x dependence of the temperature coefficient γ_{ac} arises from the effects of lateral confinement on the exciton wave function. The dotted line in Fig. 3(b) is a L_x^{-1} fit to the data. This curve fits the data well, indicating that the acoustic phonon scattering gives a good explanation of the observed wire width dependence.

In summary, we have measured the temperature dependence of the homogeneous linewidth of the excitonic resonance in $In_{0.135}Ga_{0.865}As$/GaAs quantum wires with widely varying lateral sizes by means of time-integrated degenerate four-wave mixing. We have seen that the homogeneous linewidth at zero temperature is rising due to the enhanced scattering of the excitons by the wire width fluctuations. The rise saturates for wire widths smaller than 50 nm most likely due to the localization of the exciton wave function. The temperature coefficient γ_{ac}, which is a measure for the scattering of the excitons by acoustic phonons, increases monotonically with decreasing wire width. We show that this dependence is consistent with the results of microscopic theoretical calculations for exciton-phonon scattering in quantum wires.

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