

Exciton localization in corrugated GaAs/AlAs superlattices grown on (311) GaAs substrates

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Using temperature-dependent photoluminescence we have studied the exciton-localization effect in GaAs/AlAs corrugated superlattices (CSL's) grown on (311) GaAs substrates. In this structure, the creation of deeper localized states below the ground states by the perturbation of the corrugated interfaces is suggested. This effect is supposed to lead to the redshift of the exciton emission relative to that in the (100) reference sample at low temperature. In type-II CSL's the local potential minima in the Γ point of GaAs serve as the initial states of the Γ transition through the resonant injection of electrons from the X point of AlAs to the localized states in the Γ point of GaAs, resulting in the observed strong X -band emission up to room temperature, but with a Γ -like behavior in the temperature dependences of the linewidth and the exciton emission energy.

The growth of GaAs/(Al,Ga)As quantum wells (QW's) on non-(100)-oriented GaAs substrates has attracted significant interest because of its potential in the precise fabrication of nanostructured semiconductors.¹⁻⁵ Recently, Nötzel and co-workers¹⁻³ developed a method to directly synthesize GaAs/AlAs quantum wires by molecular-beam epitaxy (MBE) on GaAs (311) surfaces. Various one-dimensional (1D) properties, such as a pronounced optical anisotropy¹ and an increased exciton-phonon interaction^{1,6} have been demonstrated in these structures and attributed to the additional lateral confinement of carriers. The observed redshift of the luminescence was correlated with the height of the surface corrugation.¹ On the other hand, the structure is actually an interface corrugated superlattice (CSL) with macrosteps, which are realized due to the breaking up of a flat surface with high surface energy into facets corresponding to planes with lower surface energy. Such faceting process may lead to the formation of undesirable macroscopic surface defects and microroughness of the interfaces in the growth,^{5,7} and consequently to a perturbation of the periodic corrugation of the interfaces. This perturbation is anticipated to create local potentials for the excitons as the interface roughness does in the conventional SL and QW's.^{8,9} Therefore the emission of this structure at low temperature can be dominated by the localized excitons bound to the growth-induced defects of the corrugated structures.

In this paper, we demonstrate the exciton localization effect in GaAs/AlAs CSL grown on (311) GaAs substrates through the analysis of temperature-dependent photoluminescence (PL) data. We have found that the large deviation of the exciton emission from the expected free-exciton energy in the low-temperature range faded at high temperatures and have associated it to a thermal detrapping process of the localized excitons. In the meanwhile the strong X -band emission persisted up to

room temperature in the type-II CSL samples, and no thermal transition from type-II to type-I emission was observed. These behaviors are believed to be due to an effective transfer of the X -point electrons in AlAs into the localized states in the Γ point of GaAs.

The samples used in this study were grown by MBE simultaneously on GaAs substrates with the (100) and (311) orientations. In the case of (311) surface the reflection high-energy electron diffraction pattern indicated the splitting of the (311) planar surface into an ordered ensemble of falling and rising steps oriented along the [011] direction. The total structure is thus a periodic ensemble of wide and narrow GaAs and AlAs regions with their thickness ranging from 20 to 70 Å. The layer thickness of the (100) sample and also the average layer thickness of the (311) samples were measured to be equal for both orientations. The structural perfection is comparable for both samples.⁷

In the PL measurements the samples were fixed on the cold finger of a closed-cycle cryostat with variable temperature (10–300 K). The 514.5-nm line from an argon laser was used for sample excitation. The luminescence was collected by a double-grating monochromator and detected with a cooled GaAs photomultiplier and photon-counting electronics. In the whole range of temperatures, strong luminescence was recorded. This fact allow us to locate unambiguously the emission peak of the excitons.

In Fig. 1 we have plotted the peak energy of excitons as a function of temperature from 10 up to 300 K for both 43/47-Å GaAs/AlAs CSL and (100) reference samples. In these samples the emissions were identified, both experimentally and theoretically, as type-I direct (Γ) transitions.¹⁰ It is noted that at 10 K the peak energy from the CSL sample is redshifted by 16 meV relative to that of the (100) sample. However, as temperature increases the separation of the two curves in Fig. 1 is gradually re-

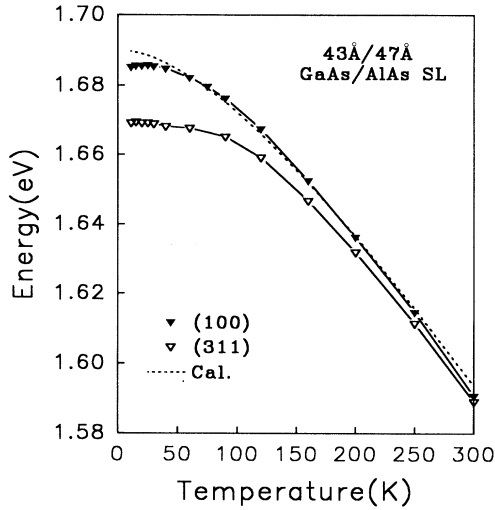


FIG. 1. Luminescence peak energies of excitons as a function of temperature for a 43/47-Å GaAs/AlAs CSL and (100) a reference sample. The temperature dependence of the calculated free exciton energy is also shown by a dotted line.

duced, and at 300 K they almost coincide.

In order to understand the physical origin of the large redshift observed at low temperature, a calculated temperature dependence of the free-exciton energy (E_g) is shown in the same figure. The calculation was based on the effective-mass approximation with the exciton binding energy of ~ 10 meV. Here emphasis was paid on the temperature variation of the exciton energies, rather than on their absolute values. It is well accepted that E_g is expected to follow the band-gap variation of bulk GaAs with temperature,^{8,11} and the curves in Fig. 1 should remain parallel to one another at all temperatures. It is the case for the (100) sample, where the exciton energies present the same variation with temperature as E_g , except for that at very low temperature (< 40 K). For the CSL sample, however, its energy position does not follow the temperature dependence of E_g up to 120 K, and then agrees with the E_g variation. This behavior can be explained in terms of exciton localization. At low temperature the excitons are strongly localized at interface defects and fluctuations, and the luminescence is governed by the localized-exciton emissions. When the temperature increases a thermal delocalization of the localized excitons occurs, and finally the luminescence is dominated by the free-exciton emission. Therefore, the low-temperature deviation of PL peak energy from the expected free-exciton energy can be regarded as a measure of the exciton localization on interface defects. In our 43/47-Å CSL sample the measured deviation amounts to 20 meV at 10 K, whereas that of the (100) sample is only 4 meV (Fig. 1). This result indicates that a strong-exciton localization exists in the CSL structure, and deeper localized states have been introduced. This is in accordance with the observed large redshift of the emission, although

the enhanced heavy-hole mass along [311] also will contribute to the redshift.³ It is noted that the thermal delocalization process was also known from bulk materials,¹² where the localization of the excited carriers originates from the compositional disordering.

Figure 2 gives another example from a pair of 29/29-Å GaAs/AlAs samples, which were identified as type-II superlattices.¹⁰ At low temperature, the emissions of both samples are indirect (denotes as X in the figure), involving X electrons in AlAs and Γ holes in GaAs. When the temperature is raised, a thermal transition from type-I to type-II emission was observed in the (100) sample, *but not* in the CSL sample. In the figure, all observed PL peak positions are plotted. For the sake of comparison the calculated temperature variations of the relevant free-exciton energies are indicated as dotted and dashed curves for the (311) and (100) samples, respectively. In the calculation, the different temperature coefficients of X band Γ energy gaps were taken into account. It can be seen that at 10 K a large deviation of 23 meV is observed for the CSL sample compared to 10 meV for the (100) sample, reflecting the existence of deeper localized states in the CSL structure.

Figures 3 and 4 display the measured PL spectra at various temperatures for both 29/29-Å samples. In the (100) reference sample the low-temperature PL was dominated by the indirect (X) transition. When the temperature is raised, a high-energy peak separated from X transition by 42 meV appears. This peak was identified as a direct (Γ) transition involving electrons and holes both confined in GaAs. The identification resulted from the time-resolved luminescence measurements¹³ and the theoretical calculation as well. From Fig. 3, we find that the emission intensity of the Γ transition relative to that of X point is drastically enhanced with increasing temper-

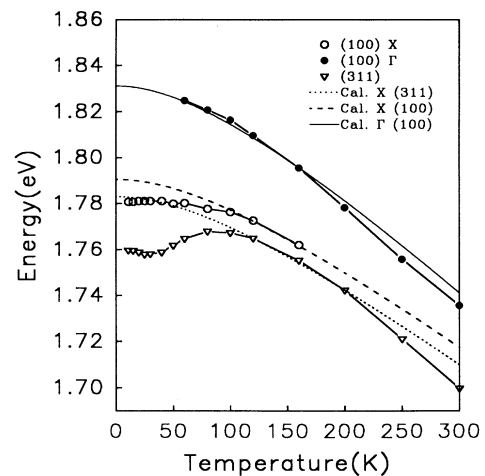


FIG. 2. Luminescence peak energies of excitons as a function of temperature for the 29/29-Å samples. The calculated temperature variations of the relevant free-exciton energies are shown as dotted and dashed curves for (311) and (100) samples, respectively.

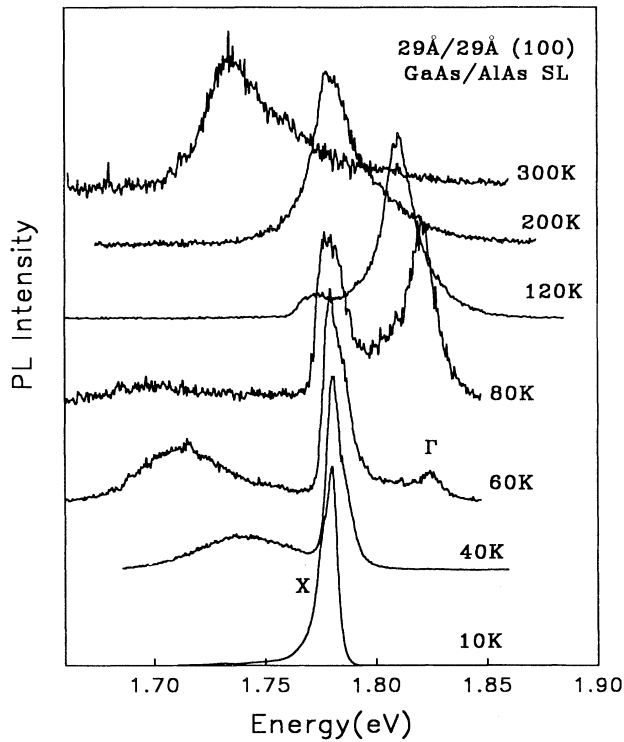


FIG. 3. Photoluminescence spectra of the 29/29-Å GaAs/AlAs superlattices grown on (100) GaAs substrate measured at various temperatures. Note the emergence of Γ transitions at higher temperatures.

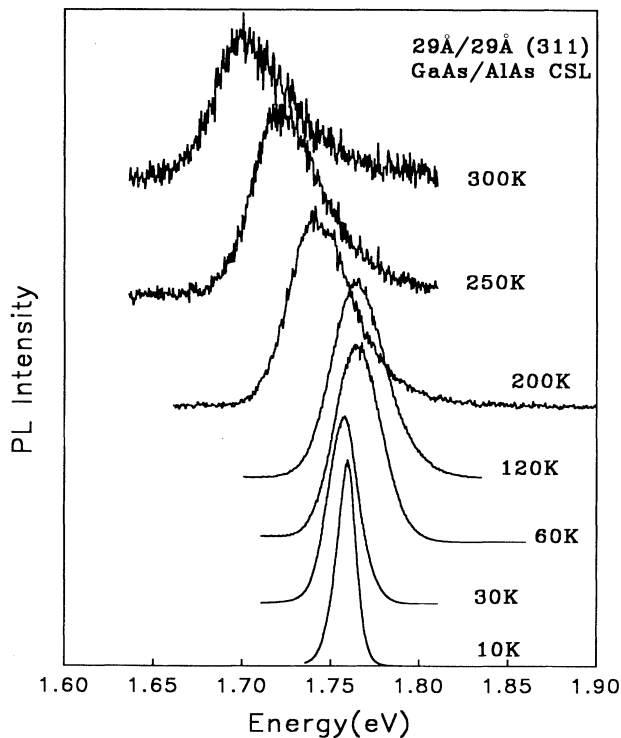


FIG. 4. Photoluminescence spectra of the 29/29-Å GaAs/AlAs corrugated superlattices grown on (311) GaAs substrate measured at different temperatures.

ature. This enhancement is interpreted by taking into account both the thermal distribution of electrons at Γ states and the high direct recombination probability. Above 100 K the Γ transition becomes dominant. To our surprise, however, the above described behavior does not duplicate in the type-II CSL sample. In the whole temperature range of 10–300 K, the PL spectra are characterized by a strong single luminescence peak with the high-energy tail developing with temperature, as indicated in Fig. 4. The luminescence peak was affirmed, by PL decay-time measurements and the energy position as well, to evolve from the indirect (X) transition at low temperature. In other words, no direct interband transition was observed in this CSL sample although it is expected to be 57 meV (Ref. 14) above the X transition. The reason for the absence of the direct transition at high temperature will be discussed later.

In another set of our experiments, the temperature dependence of the exciton linewidth was investigated in the temperature range of 10–300 K. In general, the measured PL linewidths of the CSL samples are found to be always broader than those of the (100) samples. For example, at 10 K the linewidth from 43/47-Å GaAs/AlAs CSL sample is 15 meV, while that of the reference sample is only 4 meV. We believe that it is an inhomogeneous broadening effect, giving further evidence of the existence of large potential fluctuations in CSL structures. In our type-I superlattices, no matter which substrate was used, the exciton linewidth increases with temperature (not shown). Such a temperature dependence has been reported frequently,^{11,15} and is usually interpreted using models which involve various types of broadening mechanisms, including acoustic- and optical-phonon scattering and impurity scattering.¹⁶ In type-II superlattices, however, the variation of the linewidth with temperature shows substantially different properties and, to our knowledge, was not much reported. In Fig. 5 we present the PL linewidth [full width at half maximum (FWHM)] of the exciton emissions as a function of temperature from the 29/29-Å type-II GaAs/AlAs superlattices. It can be seen as follows that (i) in the low-temperature range below 60 K the linewidths of X transitions for both samples show a very steep linear dependence on the temperature.¹⁷ The linear dependence can be attributed to the acoustic-phonon scattering which dominates at low temperature. In the 2D system the scattering rate is proportional to the effective mass of carriers.¹⁶ Since the effective mass of X electrons is much greater than that of Γ electrons, the slope of the linewidth for the X transition is much higher than that of the Γ transition. From Fig. 5 the slope of the X transition in the (100) sample is estimated to be $1.8k_B$, which is approximately 1 order of magnitude higher than that of the Γ transition. It is interesting that the slope of the CSL sample comes up to $5k_B$, much higher than that for the (100) sample. The reason for this finding is not clear at present, but a detailed study is in progress. (ii) In the (100) sample, the variation of linewidth with temperature becomes complicated due to the different transitions dominating at different temperatures. It is seen that the slope of FWHM is higher for the X transition than for the Γ transition, as illustrated in the

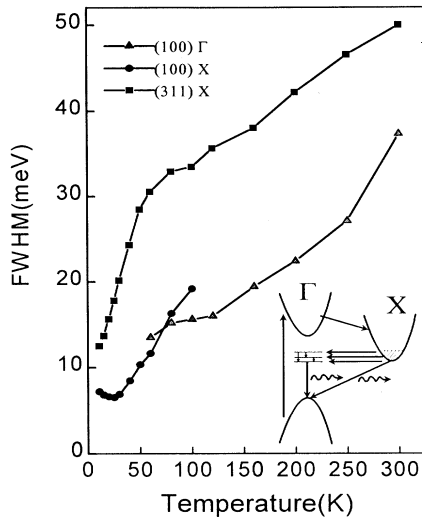


FIG. 5. Photoluminescence linewidths (FWHM) of the exciton emissions for the 29/29-Å samples as a function of temperature from 10 up to 300 K. The electron injection process from the X point into the localized states at Γ point and the relevant recombination processes are shown in the inset.

lower part of Fig. 5. Nevertheless, the temperature dependence of the linewidth for the Γ transition can still be described using the existing models.^{11,16} (iii) For the CSL sample the exciton linewidth increases very fast in the temperature range below 60 K and, then, increases with a lower rate close to that of the Γ transition in the reference sample. Furthermore, the turnover temperature around 60 K in the upper curve of Fig. 5 is almost the same as the onset temperature of Γ transition in the reference sample (see lower part of Fig. 5). From all these results we conclude that the X transition of the CSL sample exhibits a similar behavior as the Γ transition concerning the temperature dependence of the linewidth. In the following, we will correlate the above finding to the strong-exciton localization in the CSL structure.

In a recent work, Zrenner *et al.*¹⁸ proposed a model to describe the quantum-dot formation by interface fluctuations in AlAs/GaAs quantum wells. In this model a variety of shallow and deep localized states are assumed to be created below the ground-state exciton at the Γ point of GaAs by interface fluctuations. The authors suggested that the electrons in the X band can inject resonantly from the X point into local-potential minima of the direct GaAs at the Γ point due to the resonant Γ - X mixing.¹⁸ We apply this model to explain the experimental results of our CSL structures, where the perturbed corrugation of interfaces is assumed to introduce deeper

localized states for in the Γ -point states of GaAs, as demonstrated in the above discussion. We assume these localized states to be energetically in the vicinity of the X minimum. At low temperature the luminescence is dominated by the indirect transition involving the localized electrons at X point of AlAs and holes at Γ point of GaAs. These localized states are expected to be shallow due to the large effective mass. Therefore they are easily to be thermal detrapped, which will cause the luminescence peak blueshifted, as shown in Fig. 4 below 120 K. When the temperature is further raised, the X -point states will energetically align with the localized states at the Γ point of GaAs via both the thermal population and the relative shift of Γ and X minima, which is caused by the different temperature coefficients of Γ and X band gaps. In this sense the X -point minimum acts as a charge reservoir which can be used to inject electrons from the X point into the localized states at the Γ point of the direct GaAs, as sketched in the inset of Fig. 5. As a consequence, the Γ -point transitions between the localized states and valence-band states are expected in a superposition with the X transition in energy. Experimentally, it leads to the observation of the strong X transition up to room temperature and the absence of the interband excitonic Γ transition (see Fig. 4). Furthermore, one would expect that the observed X transition at higher temperature will show in some respect the behavior of the Γ transition. This has been proved in our experiments, indeed. As mentioned before, the temperature dependence of the linewidth of X transition presents a similar slope as the Γ transition. In addition, the temperature variation of the X -transition energy for the CSL sample follows that of the Γ transition, rather than that expected for the X transition, as revealed in Fig. 2.

In conclusion, we have demonstrated the strong exciton localization effect in GaAs/AlAs corrugated superlattices grown in (311)-oriented GaAs substrates. The localization is supposed to be caused by the perturbation of the corrugated interfaces. It leads to the redshift of the exciton emission at low temperature, and the Γ -like behaviors of the temperature dependencies of the linewidth and the exciton emission energy of the X transition in type-II superlattices. We believe that the problems discussed here are of universal significance in the study of the optical properties of corrugated superlattices grown on other non-(100)-orientated substrates. For a full understanding of the exciton localization in nanostructures it will be important to evaluate the combined effect of the exciton localization and of lateral confinement.

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