# Confined excitons in corrugated GaAs/AlAs superlattices

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A strong modification of excitons is observed in corrugated GaAs/AlAs superlattice structures directly grown by molecular-beam epitaxy on (311)*A* GaAs substrates. The increased exciton continuum energies and the phonon sidebands of the exciton luminescence point out the length scale of lateral localization. The observation of hot-exciton luminescence up to high temperatures and excitation intensities reveals the high stability of the confined excitons. The pronounced degree of optical anisotropy and the anisotropic optical alignment of the excitons is in agreement with a lateral potential introduced by the periodically corrugated interfaces.

## I. INTRODUCTION

Advanced crystal-growth techniques such as molecular-beam epitaxy (MBE) and metal organic vapor-phase epitaxy (MOVPE) have made it possible to precisely fabricate heterojunctions and superlattices on an atomic scale.[1] Compared to bulk semiconductors, enhanced exciton binding energies and oscillator strengths are obtained in quantum wells and superlattices,[2] and strongly increased mobilities in modulation-doped heterojunctions.[3] These properties have been essential for the development of improved high-speed electron and optoelectronic devices. To study the fundamental electronic properties and to develop novel device concepts it is now important to further reduce the dimensionality of semiconductor structures to one and zero dimensions. This has been tried commonly by subsequent lateral patterning of two-dimensional heterostructures.[4] This approach, however, often introduces uncontrolled defects. Hence, the lateral dimensions of the fabricated quantum-wire and quantum-dot structures are fairly wide and the confinement energies are low. The direct synthesis by epitaxy should reduce the defect concentration considerably and thus allow the fabrication of quantum-wire structures for device applications. The most established method at present is the growth of fractional layer superlattices on vicinal substrates,[5-7] which, however, suffers from the unintentional fluctuations in growth rate and local misorientation.

We have recently developed a method to directly synthesize superlattice structures with periodic corrugation of the interfaces using the unique property of GaAs high-index surfaces to lower their surface energy by forming periodic arrays of macrosteps or facets composed of low-index planes.[8] This ordered surface structure is then transferred to the heterointerface during conventional MBE. As revealed by the optical properties, the corrugated heterointerfaces give rise to a lateral potential in GaAs/AlAs multilayer structures. Reflection high-energy electron diffraction (RHEED) directly shows the breaking up of the flat (311)*A* surface into a periodic array of upward and downward steps oriented along [$\bar{2}$33] [Fig. 1(a)]. In this azimuth, the RHEED pattern shows streaks alternatingly split into sharp satellites, giving the lateral periodicity of 32 Å, and along their length, giving the step height of 10 Å.[8,9] RHEED intensity dynamics reveals a pronounced oscillation during the deposition of the first monolayers of GaAs on AlAs and vice versa due to a phase change of the surface corrugation during the heterogeneous growth on the facets. This phase change generates a periodic array of alternating narrow and wide regions of GaAs and AlAs, respectively, thus forming the corrugated superlattice (CSL) structure shown in Fig. 1(b).[8] High-resolution transmission-electron microscopy (HREM) has confirmed the presence of alternating thicker and thinner GaAs and AlAs channels along [$\bar{2}$33] with average distances of 32 and 10 Å height of the interface corrugation. The structural perfection of GaAs/AlAs multilayer structures grown simul-

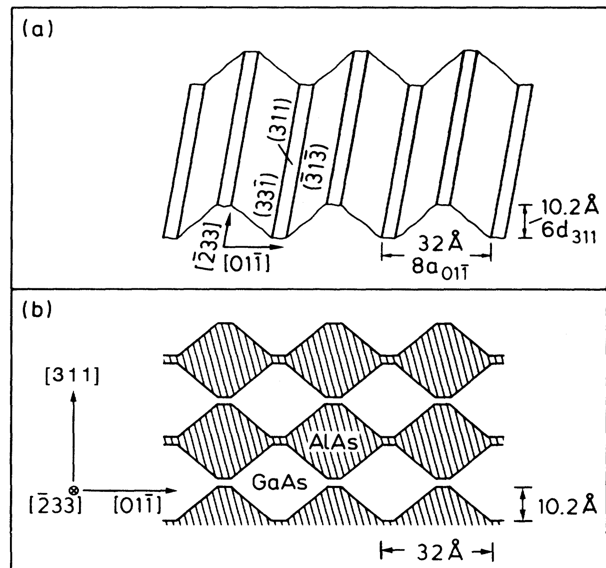


FIG. 1. (a) Schematic of the stepped (311)*A* GaAs surface. (b) Schematic ($\bar{2}$33) cross section of the GaAs/AlAs corrugated superlattice structure.

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taneously on (311)*A* and (100) GaAs substrates as reference is comparable as determined by high-resolution x-ray diffraction.[8]

In this paper we report on the strong modifications of the excitonic properties of CSL structures directly grown on corrugated (311)*A* GaAs substrates. The exciton continuum energies determined from photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy drastically exceed those in the reference (100) superlattices (SL's), which is attributed to a strong localization of the excitons in the GaAs channel regions. The appearance of pronounced LO-phonon sidebands of the exciton luminescence is related to the length scale of lateral exciton localization. PLE measurements reveal a series of LO- and TA-phonon-related lines, which are attributed to hot-exciton relaxation. The observation of hot-exciton luminescence up to high temperatures and excitation densities manifests the high stability of the excitons. The pronounced degree of optical anisotropy and the anisotropic optical alignment of excitons is in agreement with the lateral potential introduced by the periodically corrugated interfaces.

The paper is organized as follows. In Sec. II we describe the sample preparation and experimental setups. The results of the spectroscopic measurements are discussed in Sec. III. Finally, in Sec. IV, we summarize our results on confined excitons in corrugated GaAs superlattice structures.

## II. SAMPLE PREPARATION AND EXPERIMENTAL SETUPS

For the present investigations, a series of samples has been grown by MBE simultaneously on (100)- and (311)-oriented GaAs substrates comprising GaAs/AlAs multilayer structures of 0.8 $\mu$m total thickness. The GaAs (AlAs) layer thicknesses on (100) substrates have been adjusted by the growth parameters to 66 Å (61 Å), 56 Å (50 Å), and 29 Å (29 Å), respectively, and determined independently by double crystal x-ray diffraction. The growth was monitored by RHEED using a 30-keV electron beam at 1° glancing angle. The growth temperature was 600 °C and the growth rate was 1 $\mu$m/h for GaAs and 0.5 $\mu$m/h for AlAs. The $As_4$/Ga flux ratio was 5. The low-temperature PL and PLE measurements were performed with the samples mounted in an optical He-flow cryostat. Light from a broadband 600-W halogen lamp dispersed by a 0.5-m double-grating monochromator and the red line (1.916 eV) of a $Kr^+$ laser served as excitation sources. The detection system consisted of a 1-m single-grating monochromator and a cooled photomultiplier operating in the photon-counting mode.

## III. RESULTS AND DISCUSSION

The PL and PLE spectra of a 66-Å GaAs CSL structure and of the reference (100) SL are presented in Fig. 2. The spectra of the CSL structure are redshifted compared to those of the reference SL. The redshift of the electron-heavy-hole (*e*-HH) transition increases from 4 meV for the 66-Å GaAs sample to 24 meV for the 43-Å

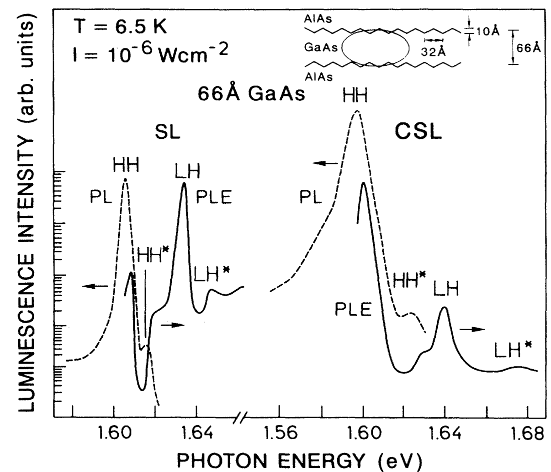


FIG. 2. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra of 66-Å GaAs superlattice (SL) and corrugated superlattice (CSL) structures. The excitonic transitions are indicated. The inset shows a scale figure of the CSL with the estimated exciton size; for details see text.

GaAs/41-Å AlAs multilayer structure discussed in Ref. 8. This value strongly exceeds that expected from the valence-band anisotropy assuming uncorrugated interfaces. The enhanced HH mass along [311], $m_{HH[311]}$, accounts for only a redshift of 7 meV for the 43-Å GaAs sample, obtained from a finite quantum-well model with $m_{HH[311]}$ interpolated between $m_{HH[100]}$ and $m_{HH[111]}$ taken from Ref. 10 ($m_{HH[311]}=0.49m_0$ for GaAs and $0.86m_0$ for AlAs). Hence, the observed redshift is attributed to the presence of the thicker GaAs channel regions, which reduces the optical transition energies. For the 66-Å GaAs sample, however, the redshift of the e-HH transition arising from the valence-band anisotropy is hardly distinguishable from the observed one. This behavior indicates the increased influence of the interface corrugation on thinner GaAs layers where the registration of the thicker and thinner regions is well established.

Although the structural perfection of the GaAs/AlAs interfaces is comparable for (100) and (311) multilayer structures as derived from x-ray diffraction[8] and Raman measurements,[11] the full width at half maximum of the PL line of the (311) sample amounts to 6 meV whereby that of the (100) sample is only 2 meV. This behavior is attributed to the lower quantum-mechanical connectivity of neighboring (channel) regions in the CSL structures compared to SL's due to inhomogeneities, which are correspondingly more significant for the electronic properties of CSL structures, and the localization of free carriers due to disorder (monolayer fluctuations) is more efficient.[12]

The additional features observed in PLE at energies above the HH and light-hole (LH) exciton resonances are attributed to the corresponding HH and LH continuum energies, marked by HH* and LH*. Note that the HH continuum energy is also resolved in the PL spectra. The two-dimensional (2D) continuum energies for the refer-

ence (100) SL amount to HH* = 12 meV and LH* = 13 meV, in agreement with previous investigations.[13] The CSL structure, however, exhibits exciton continuum energies of HH* = 28 meV and LH* = 29 meV, which strongly exceed those of the reference SL. For uncorrugated structures almost no dependence on the orientation is predicted for HH* and only a 10% anisotropy for LH*.[14] Hence, the observed continuum energies are attributed to the strong localization of the excitons in the thicker GaAs channel regions.[15] The lateral extension of the HH exciton in the CSL structure can be estimated from Ref. 16, where exciton binding energies in square-shaped quantum wires are calculated. For a vertical extension of 66 Å and 29 meV for HH* a lateral extension of about 80 Å is obtained from Fig. 1 of Ref. 16. This evaluation does not take into account monolayer fluctuations which might influence the exciton binding energy due to strong localization parallel to the channels. The lateral extension of the exciton exceeds the geometrical width of the channel region of 32 Å determined from RHEED. Hence, in the present CSL structure where the vertical potential is much stronger than the lateral one, the channel regions are laterally coupled and the excitons can be described to be formed from states of a perturbed two-dimensional system. This is reflected in the smooth low-energy onset of the HH and LH continuum observed in the PLE spectra of the CSL structure which, in this description, originates from a transition between 1D and 2D behavior: Regarding the present CSL structure as a strongly coupled quantum-wire array, the lateral coupling has the consequence that the peaked 1D density of states becomes blurred, whereby the corresponding maximum of the HH and LH continuum in PLE is attributed to the energy position of the unperturbed 1D density-of-states peak. From the present experiments, however, we cannot distinguish between the modification of the density of states due to the lateral potential introduced by the interface corrugation and that due to monolayer fluctuations, which will additionally smooth the onset of the continuum.

The intensity of LO-phonon sidebands of the exciton luminescence in the CSL structures is drastically enhanced compared to the reference SL. We show this effect in the PL spectra of Fig. 3 obtained from 56-Å GaAs CSL and SL structures. The intensity ratio of the 1 LO-phonon sideband and the exciton line remains below $10^{-4}$ in the reference SL, whereas for the CSL structure it amounts to $5\times10^{-3}$. This intensity ratio is related to the length scale of lateral exciton localization,[17] which is determined solely by interface fluctuations in the reference SL. In the CSL structure, the length scale of exciton localization is referred to the interface corrugation with additional inhomogeneities. Based on the results of Ref. 18, where a correlation between the length scale of lateral exciton localization by disorder and the relative intensity of phonon sidebands is given, we estimate the lateral localization length to about 50–70 Å for the CSL structure, and to exceed 100 Å for the reference SL. This value is in good agreement with the lateral extension of the exciton wave function determined from the measured exciton continuum energies in the preceding paragraph. Again, monolayer fluctuations will influence the localization length, especially along the GaAs channels. However, the strong lateral localization of excitons in the CSL structures also explains the observed high luminescence intensity[8] up to 400 K to be due to the free exciton motion possible only along the channels which reduces the nonradiative interface recombination.

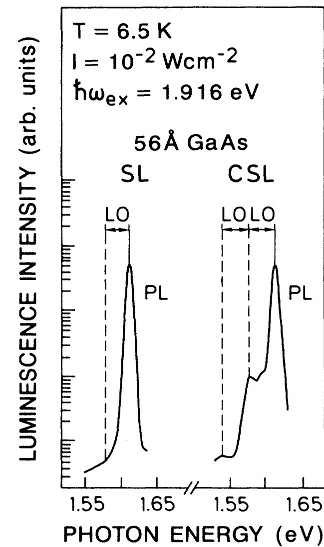


FIG. 3. Photoluminescence spectra of 56-Å GaAs superlattice (SL) and corrugated superlattice (CSL) structures.

When the detection wavelength is set on the high-energy side of the PL line we observe pronounced oscillatory structures in the PLE spectra of the CSL structure (see Fig. 4 for the 56-Å GaAs CSL structure). The lines are separated by 36 and 11.5 meV, corresponding to the LO- and TA($X$)-phonon energies in GaAs. The lines move in energy with the detection wavelength. Whether these lines have to be described in the framework of hot-exciton luminescence (HL) or resonant Raman scattering (RRS) is a well-known controversy.[19] HL is an incoherent process within the exciton lifetime including the indirect creation of excitons with large **k** vector and subsequent relaxation through LO-phonon emission. On the contrary, RRS is a coherent process within the exciton dephasing time involving virtual intermediate electronic states. A distinction between HL and RRS becomes meaningless when the two time scales are similar.[20] Most accurately, the whole problem is accounted for by polariton-mediated RRS,[21] where HL and RRS are described on the same basis. Polaritons with high energies are photogenerated well above the fundamental band gap and relax via LO-phonon emission down to the low-energy part of the polariton dispersion curve. With high probability the relaxation takes place along the exciton-like branch. However, with a weaker probability relaxation is also allowed along the photonlike branch, giving rise to RRS lines. A huge enhancement of RRS or HL results when the LO-phonon cascade ends in the polariton bottleneck region. The bottleneck polariton states re-

veal the longest step in the whole process. In this region acoustical phonon or impurity scattering becomes important, giving rise to loss in coherence.

The following experiments (Fig. 4) show that in our CSL structures the coherence between ingoing and outgoing photons (initial and final states) is lost. We therefore believe that the observed oscillatory structure is best described within the HL framework.[22] The 1LO-phonon line which is in resonance with the LH-exciton state in the 56-Å GaAs CSL structure exhibits a pronounced degree of negative circular polarization of $P=-0.42$ ($P=[I_{++}-I_{+-}]/[I_{++}+I_{+-}]$) [Fig. 4(a)]. The degree of linear polarization is negligible. This behavior shows the LH exciton to act as intermediate state for the hot-exciton creation, which is followed by the incoherent relaxation to the HH-exciton state in agreement with the selection rules for LH- and HH-exciton transitions.[13] The oscillatory structure is repeated in the PLE spectra with a period corresponding to the LO-phonon energy, thus revealing the indirect exciton creation via LO-phonon emission, whereby LO and TA phonons are involved in the relaxation process. The degree of circular polarization of the 1LO-phonon line drastically drops when the detection wavelength is shifted towards lower energies [$P=-0.15$ in Fig. 4(b)], and is accompanied by a broadening and finally a disappearance of the oscillatory structure. When the low-energy side of the PL line (localized centers) is monitored, solely HH- and LH-exciton resonances are resolved [Fig. 4(c)] and the degree of circular polarization is negligible. This behavior reflects the intrinsic nature of the oscillatory structure in the present structure, which is blurred due to the acoustical phonon or impurity scattering during the exciton (bottleneck polariton) migration within the localized exciton band. These incoherent processes become more and more dominant with increasing strength of localization, i.e., increased exciton lifetime in the localized exciton band, corresponding to the lower-energy parts of the PL line.

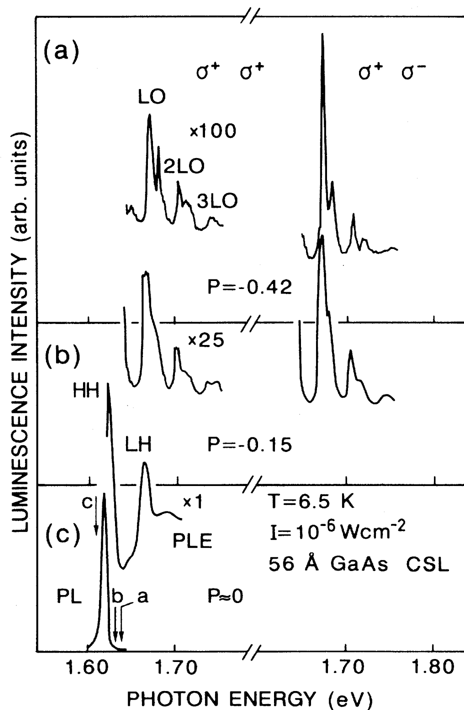


FIG. 4. (a)–(c) Photoluminescence (PL) and photoluminescence excitation (PLE) spectra of a 56-Å GaAs corrugated superlattice (CSL) structure. The excitonic transitions, as well as the detection energies for PLE (vertical arrows $a,b,c$), and the polarizer geometries are indicated.

So far, HL has been observed in II-VI semiconductors where a strong LO-phonon coupling is combined with a high exciton binding energy.[22] Hence, the observation of HL in these CSL structures demonstrates the high stability of excitons created well above the fundamental band gap, which is attributed to the strong localization resulting from the interface corrugation. The energy of exciton localization in the CSL structure is limited by the energy corresponding to HH- and LH-exciton transitions in the narrow GaAs regions. In these regions the photogenerated carriers relax separately as usual in SL's without the formation of hot excitons. Hence, the damping characteristics of the oscillatory structure observed in PLE provides an estimate of the localization potential. For the present CSL structure the well-resolved oscillatory structure up to the 2LO-phonon line yields an energy of lateral localization of about 90 meV.

In the 29-Å GaAs CSL structure where the interface corrugation imposes the strongest lateral localization, we observe HL on the high-energy side of the PL spectrum. In Fig. 5 we show that pronounced HL lines are resolved when the energy of the exciting light is separated by one or several LO GaAs phonon energies. In experiments using the 1.916 line of a $Kr^+$ laser as excitation source, additional sharp LO RRS lines are resolved which are superimposed on the spectra but with a negligible integral intensity compared to the HL lines.[19] The width of the

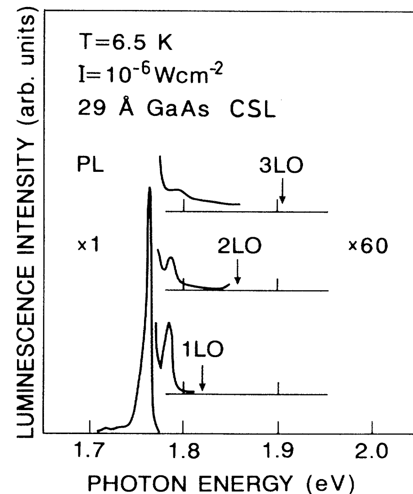


FIG. 5. Photoluminescence (PL) spectra of a 29-Å GaAs corrugated superlattice (CSL) structure. The respective energies of the exciting light are indicated by the vertical arrows.

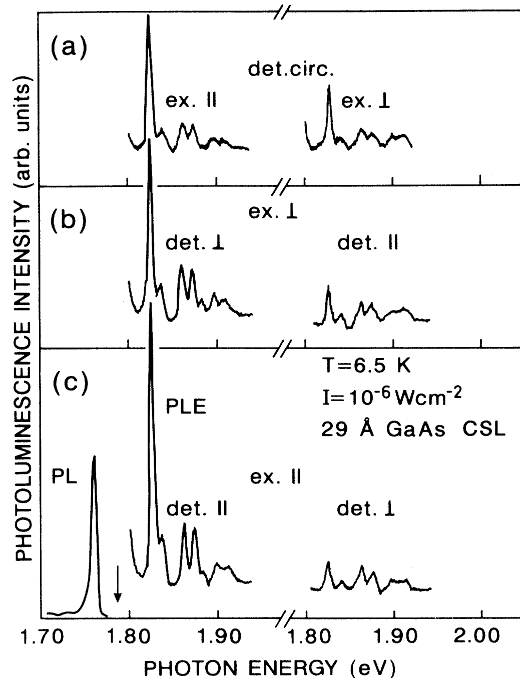


FIG. 6. (a)–(c) Photoluminescence (PL) and photoluminescence excitation (PLE) spectra of a 29-Å GaAs corrugated superlattice (CSL) structure. The detection energy and polarizer geometries with respect to the corrugation ([$\bar{2}$33] direction) are indicated.

HL lines increases with the number of emitted phonons due to the spectral diffusion during the LO-phonon cascade (Fig. 5). The width of the HL lines also increases with increasing excitation density and temperature. However, up to 100 W $cm^{-2}$ excitation density and temperatures higher than 150 K, the HL lines are still distinguishable. This behavior shows the high stability of the hot excitons with respect to screening and temperature rise.

In the PLE spectra of the 29-Å CSL structure we observe a distinct oscillatory structure [Fig. 6(a)] which reveals a pronounced optical anisotropy of the hot-exciton creation in the CSL structure. When detecting with a circular polarizer in front of the receiving monochromator, the intensity of the 1LO-phonon line is found to be two times higher for excitation with light linear polarized parallel to the wire axis than for the perpendicular polarization. The degree of optical anisotropy amounts to $P=0.51$, whereby $P$ is defined in analogy to the degree of circular polarization discussed above. This optical anisotropy is in agreement with a lateral potential introduced by the interface corrugation[23] and is further observed in the optical anisotropy of the HH and LH exciton resonances in PL and PLE (not shown here). In this 29-Å GaAs CSL structure the 1LO-phonon line is in resonance with the HH-exciton state and exhibits a pronounced degree of positive circular polarization ($P=0.60$) in agreement with the selection rules.[13] Due to the fast relaxation process through the LO-phonon emission, a high degree of optical alignment of the excitons is observed in PLE for excitation with linear-polarized light [Figs. 6(b) and 6(c)].[24] The intensity of the 1LO-phonon line is several times stronger when the linear polarizer in front of the receiving monochromator is collinear with the polarization of the exciting light than for the crossed polarizer geometry. However, the degree of optical alignment detected for the 1LO-phonon line amounts to only $P=0.60$ when the exciting light is polarized perpendicular to the channel axis [Fig. 6(b)] whereby it amounts to $P=0.84$ for polarization of the exciting light parallel to the channel axis [Fig. 6(c)]. This behavior indicates stronger depolarizing mechanisms for excitons aligned perpendicular to the channels. A way to understand this phenomenon is to assume the exciton to exist in an elliptic polarization state after the absorption of linear polarized light which is oriented along the direction of polarization. This elliptic state is easily perturbed, i.e., rotated or deformed by the interface corrugation when aligned perpendicular to the channel axis.

## IV. CONCLUSION

In summary, we have observed strong modifications of the excitonic properties which are attributed to a lateral potential in corrugated GaAs/AlAs superlattice structures directly grown by MBE on periodically corrugated GaAs (311)$A$ substrates. Enhanced exciton continuum energies and phonon sidebands of the exciton luminescence compared to reference (100) superlattices provide an estimate of the length scale of lateral localization. The observation of hot-exciton luminescence in PL and PLE spectroscopy up to high excitation intensities and temperatures reveals the high stability of the confined excitons in these corrugated superlattice structures. The pronounced degree of optical anisotropy and the anisotropic optical alignment is in agreement with a lateral potential introduced by the periodically corrugated interfaces.

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