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Relaxation of excitons in corrugated GaAs/AlAs superlattices

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We report time-resolved spectroscopy on exciton luminescence in low-dimensional systems. The reduction of dimensionality from two in quantum wells to one in corrugated superlattices can completely change the initial relaxation process. In the one-dimensional system exciton-phonon scattering is, even at high exciton densities, faster than exciton-exciton scattering since the latter is reduced whereas the former is enhanced if the final state falls into the singularity in the density of one-dimensional states.

Fabrication of one-dimensional (1D) semiconductor quantum-wire structures is intriguing since peculiar physical properties, as, e.g., strong optical nonlinearity,[1] high carrier mobility,[2,3] large exciton binding energy,[4] density of states with a strong singularity,[5] and reduced carrier-carrier scattering rate[6,7] are expected in these low-dimensional systems. Some of these theoretically predicted properties have been experimentally verified: for example, Nagamune *et al.* demonstrated an anisotropic shrinkage of excitons[8] and Oestreich *et al.* demonstrated that the exciton-exciton scattering rate in a quantum wire is reduced.[9] Other properties of 1D systems still need to be experimentally demonstrated.

Investigation of this one-dimensional physics depends critically on the successful fabrication of quantum-wire structures. The main problem is to fabricate such small structures that the spacing between quantized states is large enough that effects of reduced dimensionality can be clearly observed. Recently, an approach toward the synthesization of quasi-1D structures has been achieved by growing corrugated superlattices (CSL).[10] The basic concept is the *in situ* formation of an array of macrosteps during the molecular-beam epitaxy growth on high-index-oriented GaAs substrates. The structures have small wire dimensions with width and thickness of typically 5 nm. Various 1D effects like, e.g., an increased exciton binding energy and a pronounced optical anisotropy, have been demonstrated in these CSL structures.[10] The structures have clear advantages with respect to wires fabricated in conventional ways: The wires of CSL's are very thin (4.3 nm), closely packed (with a distance of about 3.2 nm), and they have no free surfaces. Therefore, large quantization energies, large optical signals, and negligible nonradiative surface recombination are obtained with CSL's.

Using these CSL structures we demonstrate in this paper how the initial relaxation process of hot excitons created by photons of a short laser pulse changes completely if the dimension of the system is reduced from 2D to 1D: In two dimensions, exciton-exciton or exciton–free-carrier scattering is at a moderate excitation density of 10^{10} cm^{-2}, by far the fastest initial process. In particular, it is faster than the optical phonon emission rate: no phonon replicas are observable in the emission of free excitons in GaAs quantum wells. In one dimension, the situation is different for two reasons: (i) exciton-exciton scattering is strongly reduced in a 1D dispersion[9] and (ii) the singularities in the density of 1D states lead to a strong variation of phonon scattering rate: the rate is very fast, if the final scattering states are within these singularities, and the rate decreases, if this is not the case.

The GaAs/AlAs CSL structures were grown on (311) substrates. Therefore, the wire direction is $(\bar{2}33)$. The sample, from which we show the results here, has a period length of 4.3 nm (GaAs)/4.7 nm (AlAs). Other period lengths show similar results. Reference quantum wells with similar thicknesses have been grown on (100)-oriented substrates. Details of the growth can be found in Ref. 10.

First we report on cw experiments. Photoluminescence (PL) is measured using a Kr^+ laser as excitation source. The light from a halogen lamp dispersed by a 0.5-m double-grating monochromator is used as excitation source for photoluminescence excitation (PLE) spectra. All experiments are performed at 10 K. Figure 1 depicts the PL and PLE spectra. The PL peak has a full width at half maximum of 10 meV and an almost symmetrical line shape, indicating a good sample quality. The line shape of PLE depends strongly on the detection wavelength: curve 1 is detected at a photon energy on the low-energy side of the luminescence maximum. Two peaks are observed and assigned to the heavy-hole and light-hole excitonic transitions, respectively, similar to the case of a 6.6-nm-thick CSL reported previously.[10] However, when the detection wavelength is at the high-energy side of the PL peak, the PLE spectrum shows very sharp, pronounced structures (curve 2). Peaks appear at distances of 36 and 48 meV (and combinations of these values) from the detection photon energy. These values correspond exactly to the GaAs and AlAs LO phonons, indicating that relaxation processes strongly influence the shape of the PLE spectra.

More detailed information about the relaxation mechanism is obtained by time-resolved spectroscopy. A synchronously pumped pyridine dye laser, with a repetition frequency of 82 MHz and an optical pulse width of about 5 ps, is used for excitation. The luminescence is dispersed

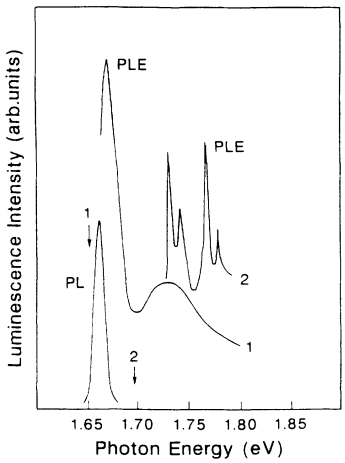

FIG. 1. cw PL and PLE spectra. The line shape of PLE spectra (curves 1 and 2) is dependent on the PL detection energy. The detection photon energy are marked by arrows. The LO-phonon-related structures are observed only in PLE curve 2.

by a spectrometer and time-resolved by a Hamamatsu streak camera with a time resolution of 15 ps. The excitation density is 10^{10} cm^{-2}. The luminescence maximum at the band edge increases within the first 100 ps and then decays slowly to $1/e$ intensity in 500 ps demonstrating that nonradiative recombination is negligible.

Here we concentrate on the evolution of the spectra at short delay times. Figure 2 depicts a temporal evolution of spectra excited by laser pulses at 1.7273 eV, with a polarization parallel to the wire orientation. The spectra are recorded at time delays of 0, 13, 24, and 36 ps, respectively. Two pronounced LO-phonon replicas, 36 and 48 meV below the excitation laser energy, are clearly observed at short delay times. They correspond to LO_{GaAs} and LO_{AlAs} phonons, respectively. They disappear after about 30 ps. Note that these replicas are not detectable in time-integrated spectra, since there the long-lived band edge luminescence dominates. The relative intensity of the replicas decrease with increasing excitation density. We do not observe such LO-phonon replicas in the reference quantum wells even at the lowest excitation densities. Therefore, we conclude that exciton thermalization in the quasi-one-dimensional quantum system differs significantly from that in the quasi-two-dimensional quantum wells. Note that this is true even for such high excess energies as several tenths of meV since we still excite exclusively the lowest subband. In contrast, quantum wires with a wire width of 60 nm and correspondingly closer spacing of the subbands show a much faster thermalization for such high excess energies of the excitons.[9]

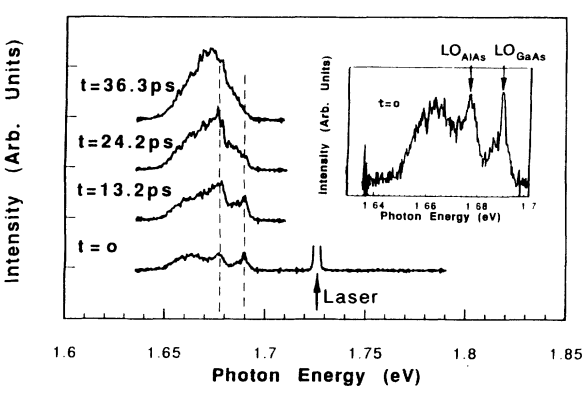

FIG. 2. Temporal evolution of PL spectra excited by the optical pulse at 1.7273 eV. The excitation pulse is polarized parallel to the wire direction of the CSL. The inset is an amplification of the spectrum recorded at $t=0$ ps. Two arrows indicate the LO-phonon-related peaks.

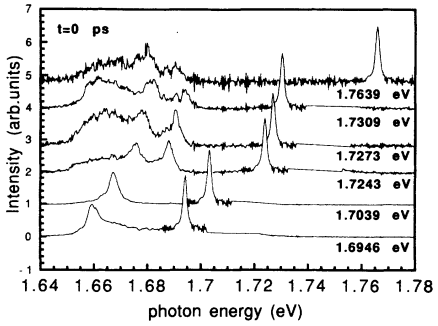

FIG. 3. Luminescence spectra at $t=0$ ps dependent on excitation phonon energy. The excitation densities remain at 10^{10} cm^{-2}.

We depict in Fig. 3 the dependence of the phonon replicas on the excess energy of the laser photons recorded with a time delay of $t=0$ ps. Strong replicas are only observed if their energetic position is between 1.66 and 1.7 eV, i.e., the regime where excitons couple to photons in the exciton-polariton picture. This regime coincides with the singularity in the density of states. The position of this singularity is indicated by the heavy-hole maximum in the PLE curve 1 in Fig. 1. Two-phonon replicas are also observed, and they resemble closely the one-phonon replicas as shown more clearly in Fig. 4. Replicas with participation of AlAs phonons, i.e., the barrier phonons, are also strong if they are close to the 1D singularity of the density of states. The reason for this resonance

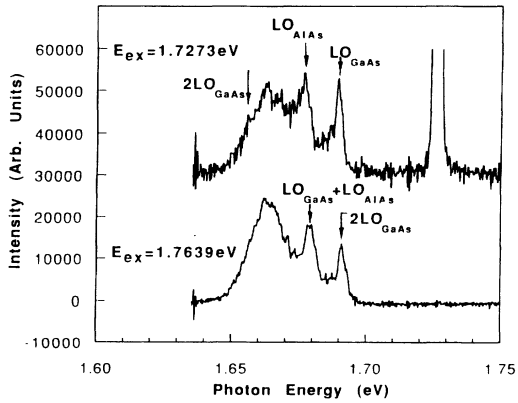

FIG. 4. PL spectra of the CSL sample excited at two different laser energies. The signals are recorded at $t=0$ ps and integrated in a time interval of 13 ps. LO-phonon-related peaks are indicated by arrows. The excitation density is by a factor of 20 higher than that for the case of Fig. 1 to Fig. 3.

behavior may be twofold: (i) scattering into these states of strongly enhanced density is very efficient[11] and (ii) excitons which are scattered into these states have a negligible quasimomentum in wire direction and therefore are able to leave the wire structure as photons (polariton effect). The first effect is in particular specific to the 1D quantization whereas the second effect is equally important in 2D systems. It is therefore probable that the first effect dominates in our experimental observation.

Note that the phonon replicas are strongest if they appear on the high-energy side of the band-edge luminescence proving that they are an intrinsic effect of the quantum-wire structure. We therefore exclude effects like localization of excitons as possible origin for the observation of phonon replicas.[12] The PL maximum in the CSL sample shifts with time to higher energies. This also demonstrates that localized states have at least only low density. The shift to higher energies, as shown in Fig. 2, is probably due to band filling.

Let us now discuss the detailed structure of the phonon replicas. The phonon features in Fig. 2 are most pronounced at zero time delay, as shown in the inset. The LO_{GaAs} peak can be clearly decomposed into two parts: (1) A sharp peak at slightly higher energy which has the same half-width at half maximum as the excitation light pulse and disappears as quickly as the laser excitation pulse does; (2) a broader peak which is asymmetric, with the lower-energy side broader than the higher-energy side. At $t=13$ and 24 ps, the sharp phonon peak attenuates significantly, while the underlying peak becomes broader, more asymmetric, and moves slightly to lower energies. At $t=36$ ps, the LO-phonon peaks are very weakly observed on a background luminescence band, manifesting the thermalization of hot excitons on this time scale. The spectral form of LO-phonon replicas can be attributed to two possible mechanisms. (1) A photon can couple to a photonlike exciton near the band edge via emission of a LO phonon. The generated excitons could remain in phase, because the exciton dephasing time in this sample is of the order of 10 ps,[13] much longer than the LO-phonon emission time. The coherent emission of excitons leads to the same spectral width and the same temporal width of the sharp LO-phonon peak as the laser pulse. This process is equivalent to a Raman process. (2) A photon can transform into an excitonlike exciton-polariton via emission of an acoustic phonon to match momentum and the energy conservation rule. The generated hot exciton then emits a LO phonon to reach the photonlike $k=0$ state on the exciton-polariton dispersion curve. In this process acoustic phonons with various energies can be involved, leading to an asymmetric spectral broadening at the low-energy side of the phonon replica. Since coherence is lost, this process is equivalent to hot exciton luminescence. The formed excitons could also undergo density-dependent exciton-exciton scattering before reaching the band edge. These scattering processes additionally broaden the replica. Increasing the excitation density by a factor of 20 we observe essentially the same spectral feature of hot exciton luminescence, as shown in Fig. 4. The insensitivity on density indicates that in the excitation range of 10^{10} cm^{-2} the exciton-exciton scattering rate is too slow to compete with the exciton–LO-phonon scattering during the relaxation. With further increase of excitation density, the LO-phonon replica becomes more symmetrical and weaker compared to the background luminescence, consistent with an enhanced exciton-exciton interaction with increasing density. We conclude that exciton-exciton scattering in the quasi-1D system is strongly reduced with respect to the 2D system since in the reference quantum well no phonon replicas are observed even at a much lower density.

The participation of both LO_{GaAs} and LO_{AlAs} phonons in an exciton relaxation process but not in a free electron process is further confirmed by the values of the energy shift, which are equal to multiples of the LO-phonon energies in GaAs and AlAs. Otherwise the energy shift of phonon peaks should be dependent on the electron and hole effective mass ratio m_e^*/m_h^* and would be larger than the phonon energy by a factor of $(1+m_e^*/m_h^*)$.[14] The participation of LO_{AlAs} in the relaxation process simply implies that the wave function of the excitons strongly penetrates into the AlAs barrier. Similar observations are made in ultrashort period GaAs/AlAs superlattices.[15]

In conclusion, we have measured time-resolved hot exciton luminescence and demonstrated that the initial relaxation dynamics of hot excitons in quantum wires differs from that in quantum wells. Reduced exciton-exciton scattering rate and enhanced exciton–LO-phonon interactions near the band edge (if the final state falls into the singularity of the density of states) lead to a long exciton thermalization time and a clear time-resolved observation of LO-phonon replicas. Both effects contribute to this observation; however on the basis of the present experimental data, we are not able to distinguish which one is the dominant mechanism.

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