Dynamical Stokes shift due to interface nanoroughness in growth islands of GaAs single quantum wells

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Time-resolved photoluminescence (PL) spectra have been measured in a GaAs single quantum well prepared by growth-interrupted molecular beam epitaxy (MBE). The measurements were performed at 16 K by using a streak-camera-based system for detection and a pyridine 2 dye laser synchronously pumped by a mode-locked Ar⁺ laser for excitation. The narrow GaAs SQW sample was mounted in a temperature variable cryostat and directly excited by 10-ps optical pulses with a wavelength of 765 nm. The quantum wells are separated by 36 nm AlₓGa₁₋ₓAs (x = 0.17) barriers. In order to avoid surface recombination a thick (~0.2 μm) AlₓGa₁₋ₓAs (x = 0.3) cladding layer is added. In this paper we will focus on the central SQW with a 5.5-nm width. A detailed comparison with the other SQW’s will be presented elsewhere. Time and spectrally resolved PL experiments were performed at 16 K by using a streak-camera-based system for detection and a pyridine 2 dye laser synchronously pumped by a mode-locked Ar⁺ laser for excitation. The narrow GaAs SQW sample was mounted in a temperature variable cryostat and directly excited by 10-ps optical pulses with a wavelength of 765 nm (1.621 eV) for which the barriers are transparent. This excitation energy was selected in order to avoid resonant excitation of any particular island and, at the same time, to ensure rapid exciton formation by exciting more than the energy of one longitudinal optical phonon above the main exciton level. The average excitation power was 34 mW, which corresponds to an excitation power density of a few W/cm². The low-intensity cw PL and PL excitation (PLE) measurements were made at 6 K in a different system using monochromatized radiation from a halogen lamp for excitation and a photon counting system for detection.

The PL and PLE spectra of the Lₓ = 5.5 nm GaAs SQW are shown in Fig. 1. The PL spectrum (dashed curve) was measured using an excitation wavelength of 600 nm, while the PLE (solid curve) was detected at 788 nm. Three sharp PL lines (associated with terraces A, B, and C) are observed with a narrow but finite linewidth of about 2 meV (full width at half maximum). Additional shoulders or tails are also resolved on the lower-energy side. The energy separations be-
Fig. 1. cw PL and PLE spectra of 5.5-nm GaAs single quantum well. The PL spectrum is shown by the dotted line and the PLE by the solid line. The four PL peaks are due to ML islands differing in thickness by approximately 1 ML, A (20 ML), B (19 ML), and C (18 ML), while the first three PLE peaks are the 1s free heavy-hole exciton resonances associated with terraces B (19 ML), C (18 ML), and D (17 ML).

The most important point we would like to stress here is, however, that the PL emission lines in the transient spectra exhibit an additional, much smaller redshift in energy, while the intensities of the different peaks evolve due to changes in the exciton population of the terraces. In order to show the time evolution and to compare it with the cw PL and PLE results, the measured PL peak energies are plotted in Fig. 3 for terraces A, B, and C as a function of time after the excitation pulse. The PL and PLE peak energies of the cw spectra are also shown in Fig. 3 although peak A cannot be resolved for PLE. From the data of Figs. 2 and 3 the following important points are derived. First, the PL peak energy...
within each terrace decreases continuously before all excitons have disappeared by radiative recombination. Second, a dynamical Stokes shift occurs between the free and localized excitons within each terrace. Third, even at longer times the PL line has a finite width with low-energy tails. Furthermore, we note that the free exciton resonance itself has a finite linewidth of about 2 meV, which also reflects the confinement potential fluctuations. Thus, the continuous change of the dynamical Stokes shift and the linewidth of the localized excitons rule out impurity related effects as the origin of exciton localization in our case. It is inferred that the localized excitons are bound by isoelectric traps due to small-scale confinement potential fluctuations caused by nanometer-scale interface roughness.

The finite linewidth observed for the free and localized excitons is naturally explained by the same interface nanoroughness, whose lateral length scale is less than the exciton Bohr radius. In previous, similar TRPL studies on the narrower $L_z = 3.5$ nm SQW, the localized exciton shows a distinct PL peak instead of the continuously varying peak energy with time as in the present case. The observation of doublets with shifting weights corresponding to the free and localized exciton was also confirmed for the narrower $L_z = 3.5$ nm SQW using the present TRPL experimental system. However, this difference between the two SQW’s can be understood if the expected change of the exciton Bohr radius with reducing $L_z$ is taken into account. Here we use excitons as a probe to examine the interface potential fluctuations. Hence, the reduction of the probe size by decreasing the well width makes it possible to visualize miniature-size terraces by a discrete exciton level, i.e., isoelectric trap sites for localized excitons. When the exciton lateral size increases, small-scale potential fluctuations due to interface nanoroughness are averaged out. As a result, we expect to observe continuous changes of the exciton population in spatially incoherent exciton bands. Finally we note that applying Vegard’s law, we implicitly assume that the averaged $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloy barrier has actually atomic fluctuations due to statistically distributed Ga and Al atoms in the group III sublattice. The experimental results presented here directly show the existence of nanoroughness within the macroscopically flat island terraces, which supports the bimodal interface roughness model for the atomic-scale structures of the GaAs/$\text{Al}_x\text{Ga}_{1-x}\text{As}$ quantum well heterointerface.

In summary, heterointerfaces of narrow GaAs/$\text{Al}_x\text{Ga}_{1-x}$As single quantum wells with growth islands have been investigated by time-resolved photoluminescence. The transient PL spectra reveal two types of dynamical Stokes shifts. One is due to the exciton transfer between the island terraces, the other one due to transfer within the islands. The continuous change of the PL peak energy with increasing time within each island is evidence for the existence of a small-scale nanoroughness within the macroscopically flat quantum well island terraces.

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12. The impurity level of the sample used for the present study is much less than the estimated exciton density of about $10^{16}$ cm$^{-3}$ in the TR PL experiments. This conclusion is drawn from the fact that thick undoped GaAs epilayers grown in the same MBE system regularly contain hole densities of less than $10^{15}$ cm$^{-3}$ according to Hall measurements.