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

Kenzo Fujiwara
*Kyushu Institute of Technology, Tobata-ku, Kita-Kyushu 804, Japan*

Holger T. Grahn and Klaus H. Ploog
*Paul-Drude-Institut für Festkörperelektronik, D-10117 Berlin, Germany*
(Received 14 August 1996)

Time-resolved photoluminescence (PL) spectra have been measured in a GaAs single quantum well prepared by growth-interrupted molecular beam epitaxy. In addition to the usual Stokes shift caused by the exciton transfer from narrower to wider quantum well islands with macroscopically flat terraces, a dynamical Stokes shift is observed in the transient PL spectra for each excitonic line from the different monolayer islands. The continuous change of the PL peak energy with time reflects the localization of excitons within the islands. This observation directly proves the existence of interface nanoroughness with a lateral length scale smaller than the exciton Bohr radius. [S0163-1829(97)00728-5]

The accurate control of heterointerfaces is crucial for any type of device that utilizes heterostructures. Therefore, the formation of interfaces in heterostructures and its understanding have been of great interest in determining how precisely the shape of the interface can actually be controlled on an atomic scale, in particular in GaAs/$Al_xGa_{1-x}As$ system.[1–4] In quantum well (QW) heterostructures prepared by growth-interrupted molecular beam epitaxy (MBE), growth islands are formed with atomically flat terraces comparable to in lateral size or even larger than the exciton Bohr radius.[5–8] In such spatially coherent QW's, the exciton emission is split into peaks originating from different terraces, whose well widths differ by one or several monolayers (ML). The dynamics of exciton transfer between the terraces and the spatial (lateral) island distribution have been previously studied by time-resolved photoluminescence and cathodoluminescence.[2,4,8–11] However, the basic question about the preparation of smooth terraces is still an open one.

In this paper, atomic-scale heterointerfaces have been investigated in coherent GaAs single quantum well (SQW) growth islands by time-resolved photoluminescence (TRPL). The measurements of transient PL spectra in the picosecond time domain reveal several sharp PL lines due to different terraces. The time evolution of the relative intensities can be understood in terms of exciton transfer between the island terraces (Stokes shift). In addition, each PL line exhibits a much smaller dynamical Stokes shift with energies corresponding to a fractional monolayer. A comparison with cw PL and PL excitation spectra indicates that the dynamical Stokes shift occurs between the 1*s* free exciton resonance and the exciton localized within the terrace. These spectroscopic results indicate that the heterointerfaces consist of two types of potential fluctuations, macroscopic and microscopic, with laterally probed length scales larger and smaller, respectively, than the exciton Bohr radius. The dynamical Stokes shift observed between and within the growth islands in conjunction with a finite linewidth of both free and localized excitons strongly suggests that the origins of the exciton localization are isoelectric traps rather than impurities. These isoelectric traps are formed by atomic-scale nanoroughness, which exists in addition to the macroscopically coherent QW island terraces.

A GaAs SQW sample was grown by MBE with growth interruption for 2 min under arsenic exposure at each interface. The sample contains three different GaAs SQW's with nominal well widths $L_z$ of 3.5, 5.5, and 7.8 nm. The quantum wells are separated by 36 nm $Al_xGa_{1-x}As$ ($x=0.17$) barriers. In order to avoid surface recombination a thick (~0.2 $\mu$m) $Al_xGa_{1-x}As$ ($x=0.3$) 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/cm$^2$. 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 $L_z=5.5$ 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-

 

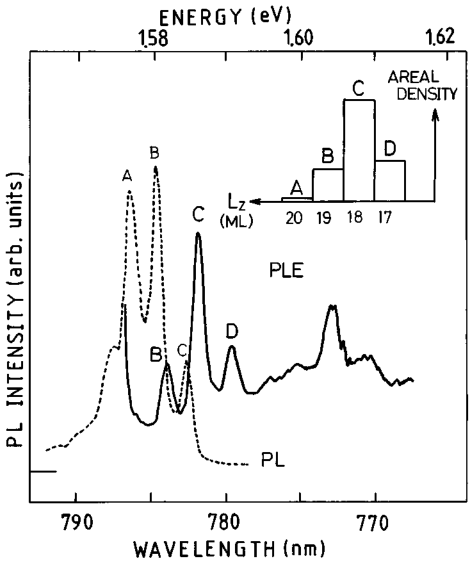


FIG. 1. cw PL and PLE spectra of 5.5-nm GaAs single quantum well. The PL spectrum is shown by the dotted line and the PLE by the solid line. The four PL peaks are due to ML islands differing in thickness by approximately 1 ML, $A$ (20 ML), $B$ (19 ML), and $C$ (18 ML), while the first three PLE peaks are the 1$s$ free heavy-hole exciton resonances associated with terraces $B$ (19 ML), $C$ (18 ML), and $D$ (17 ML).

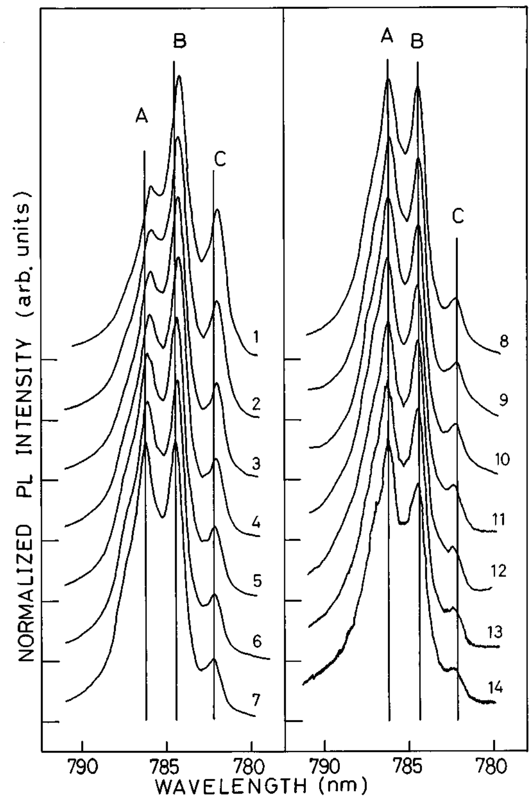


FIG. 2. Normalized PL spectra of 5.5-nm GaAs single quantum well as a function of time after pulsed excitation. The transient spectra No. 1–14 are detected at 100 ps (No. 1), 175 ps (No. 2), 225 ps (No. 3), through 1075 ps (No. 14) averaged over 75 ps. The peak positions of the time-integrated PL spectrum are indicated by vertical lines.

tween the PL peaks are 3.6 and 4.0 meV. These values are slightly smaller than the calculated energy gap changes due to well-width fluctuations by 1 ML. In comparison with the PLE spectrum, which reveals three distinct resonances associated with the 1$s$ heavy-hole excitons in the terraces $B$, $C$, and $D$ (as well as the light-hole excitons), the strongest PL peak $B$ is shifted by 5.7 meV from the main exciton resonance $C$ to lower energies (usual Stokes shift). This PL redshift is due to the exciton center-of-mass motion (exciton transfer) from the terrace $C$ to the terrace $B$ during the radiative recombination processes. The PL emission from terrace $D$ can only be observed in the spectra by plotting the intensity on a logarithmic scale. The association of each PL and PLE peak to the terraces can be confirmed by temperature-dependent PL experiments since the exciton localization effect is reduced at higher temperatures.[8] For example, the emission intensity from the largest density terrace $C$ dominates the PL spectrum at 90 K. It should be noted that the energy position of each PL peak is redshifted by about 1.5 meV from the corresponding exciton resonance line associated with the island terrace. This energy shift corresponds to a change of the well width by 0.35 ML. It is further noticeable that the resonance line has a finite linewidth of 2 meV. Both findings already indicate that the PL characteristics reflect the existence of microscopic, statistical variations of the confinement potential fluctuations at the heterointerfaces.

Figure 2 shows the time evolution of the transient PL spectra for the 5.5-nm GaAs SQW. The PL spectra No. 1,2,3, . . . ,14 were measured at 100,175,250, . . . ,1075 ps, respectively, after the excitation pulse averaged over 75 ps each. The PL intensity is vertically shifted for clear comparison and normalized by the highest peak intensity in each spectrum. In Fig. 2 the peak positions of the time-integrated PL spectrum are also indicated by vertical lines. In spectrum No. 1 measured 100 ps after the excitation pulse, the PL spectra are already dominated by terrace $B$. With increasing time delay, the intensity of peaks $B$ and $C$ decreases, while the intensity of peak $A$ increases with respect to peaks $B$ and $C$. For example, in spectrum No. 8 after 625 ps, the strongest emission band has already shifted to the lowest energy peak $A$. For spectrum No. 14, the PL line shape has become quite asymmetric and very different from the cw PL spectrum shown in Fig. 1. The dynamical changes of the PL intensity in Fig. 2 indeed reflect the fact that during radiative recombination the excitons are moving from terraces $B$ and $C$ to terraces $A$ and $B$, respectively. These results are consistent with previous observations of the intergrowth island exciton transfer.[9]

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

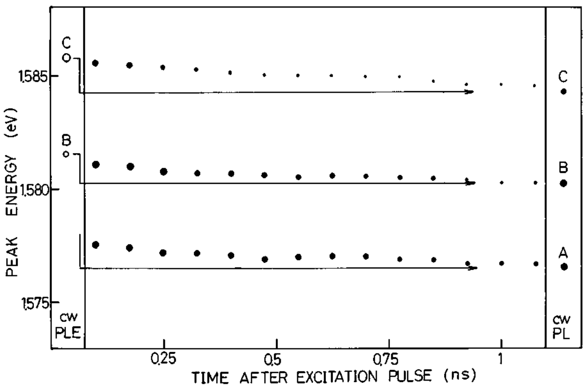


FIG. 3. PL peak energy vs time after pulsed excitation obtained from the transient PL spectra. The peak energies of the cw PL and PLE spectra are also shown at the right and left, respectively.

within each terrace decreases continuously before all excitons have disappeared by radiative recombination. Second, a dynamical Stokes shift occurs between the free and localized exciton 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. The Stokes shifts do not depend on the excitation intensity over two orders of magnitude.[12] 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,[11] 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 $Al_xGa_{1-x}As$ 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[3] for the atomic-scale structures of the $GaAs/Al_xGa_{1-x}As$ quantum well heterointerface.

In summary, heterointerfaces of narrow $GaAs/Al_xGa_{1-x}As$ 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.

The authors would like to thank Roberto Cingolani (presently at University of Lecce, Lecce, Italy) for PLE measurements and Hisashi Katahama for time-resolved PL experiments at ATR Optical and Radio Communications Research Laboratories.

[1] J. Behrend, M. Wassermeier, W. Braun, P. Krispin, and K. H. Ploog, Phys. Rev. B **53**, 9907 (1996).
[2] U. Jahn, K. Fujiwara, R. Hey, J. Kastrup, H. T. Grahn, and J. Menniger, J. Cryst. Growth **150**, 43 (1995).
[3] D. Gammon, B. V. Shanabrook, and D. S. Katzer, Phys. Rev. Lett. **67**, 1547 (1991).
[4] C. A. Warwick and R. F. Kopf, Appl. Phys. Lett. **60**, 386 (1992); M. A. Herman, D. Bimberg, and J. Christen, J. Appl. Phys. **70**, R1 (1991).
[5] C. Weisbuch, R. Dingle, A. C. Gossard, and W. Wiegmann, Solid State Commun. **38**, 709 (1981).
[6] H. Sakaki, M. Tanaka, and J. Yoshino, Jpn. J. Appl. Phys. 2, Lett. **24**, L417 (1985).
[7] T. Fukunaga, KL. I. Kobayashi, and H. Nakajima, Jpn. J. Appl. Phys. 2, Lett. **24**, L510 (1985).
[8] K. Fujiwara *et al.*, J. Appl. Phys. **66**, 148 (1989); Phys. Rev. B **40**, 9698 (1989).
[9] K. Fujiwara, H. Katahama, K. Kanamoto, R. Cingolani, and K. Ploog, Phys. Rev. B **43**, 13 978 (1991).
[10] B. Deveaud, T. C. Damen, and J. Shah, Appl. Phys. Lett. **51**, 828 (1987).
[11] R. Klann, H. T. Grahn, and K. Fujiwara, Phys. Rev. B **51**, 10 232 (1995).
[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 $10^{14}$ cm$^{-3}$ according to Hall measurements.