Dynamics of inter- and intra-growth-island exciton localization in GaAs single quantum wells

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A distinct two-exponential decay of excitonic transitions in coherent GaAs single quantum wells with atomically flat terraces is observed by time-resolved photoluminescence under direct excitation. The measured two-component time behavior is a direct consequence of the in-plane exciton localization due to the inter- and intra-growth-island exciton transfer. These interesting time behaviors are found to be strongly dependent on the emission energy at low temperatures, but the dependence disappears when excitons are delocalized and become mobile by thermal activation.

I. INTRODUCTION

Excitons in semiconductor quantum wells (QW's) exhibit a number of intriguing optical properties which result from the quasi-two-dimensional (2D) confinement of the electrons and holes.[1] Exciton states in the usual $GaAs/Al_xGa_{1-x}As$ quantum wells are, however, significantly broadened (inhomogeneous linewidth broadening of 3–10 meV) by a perturbed potential field for quantum confinement.[2–4] This broadening originates from microscopic, statistical variations of the fluctuating well width as well as of the alloy composition in the barrier. Such inhomogeneity seriously affects the dynamical behavior of exciton emissions when the spectral spreading exceeds the exciton thermal energy. Hegarty, Goldner, and Sturge[3] reported that the interface roughness results in an energy separation of localized and mobile excitons by a "mobility edge." However, it is generally difficult to quantitatively evaluate the influence of such inhomogeneity on the exciton recombination dynamics because of the random and complex distributions of the QW layer thickness.[5] Therefore complicated time behaviors such as nonexponential decays have been previously reported by several groups[6–11] but no coherent explanation has yet been found.

Owing to the recent progress in improving the heterointerfaces,[12,13] on the other hand, it is now possible to prepare QW growth islands with atomically flat terraces of area comparable to, or greater than, the exciton Bohr radius. This provides a unique chance to study the exciton-emission dynamics. However, only a few reports have been published so far and there seems to be a controversy among the previous studies.[9,11] Therefore an exploration of the exciton recombination kinetics and their relevance to the localization and relaxation processes is still an open issue even in such spatially coherent QW islands.

In this paper the dynamics of radiative recombinations of the lowest heavy-hole (1HH) exciton are studied in spatially coherent GaAs single QW's (SQW's) by time-resolved photoluminescence (PL) measurements. In order to avoid possible ambiguities arising from interwell-thickness fluctuations, we investigate the SQW sample with the highest quality attainable by modern growth techniques.[13] The increased spatial coherence of exciton states in the present sample allows us to observe distinct exciton transitions, well separated in energy, which correspond to the different SQW growth islands. Under direct excitation conditions where the excitons are uniformly generated only in the wells, we have observed distinct and systematic emission-time behaviors with two distinct exponential decay rates. This allows us to easily analyze the data of the recombination kinetics. Moreover, we have studied the effect of lattice temperature on the dynamics to examine the decay behaviors, taking both the exciton trapping and detrapping between the islands into account. We are thus able to obtain a consistent picture for the dynamical behavior of the exciton emissions in the picosecond time domain. It is important to note that our model differs from the very recent mechanism proposed by Jiang and co-workers[10] to explain the very slow decay behavior of the excitonic transitions in the nanosecond time domain.

II. EXPERIMENT

The GaAs SQW sample of this study was grown by growth-interruption molecular-beam epitaxy (MBE).[13,14] The high-quality SQW's are confined by GaAs/AlAs short-period superlattice barriers in order to obtain the atomically smooth heterointerfaces and the high radiative recombination efficiency.[15] The L_z values of the different SQW growth islands range from 19 to 22 monolayers (ML's). Hereafter we name these 22-, 21-, 20-, and

19-ML SQW islands *A*, *B*, *C*, and *D*, respectively, as in the previous paper.[14] Details of the growth procedures and the sample characterization have been described elsewhere.[13,14]

The time-resolved PL experiments were performed by using a pyridine-2 dye laser synchronously pumped by a mode-locked Ar^+ laser. For direct excitation, the dye laser with about 10 ps pulse width was tuned to 1.676 eV, at which the superlattice barrier is transparent. The carrier density in the well was of the order of 10^{10} cm^{-2}. The PL signals were dispersed by a 0.32-m monochromator (Jobin-Yvon HR320), and the time evolution of the emission intensity at different energies was measured at 15–60 K by a Synchroscan streak-camera system (Hamamatsu C1587). The measurements of low-intensity cw PL and PL excitation (PLE) spectra were performed at 6 K using the monochromized radiation from a halogen lamp for excitation and a photon-counting system for detection.[9] When focused to a few-mm-diameter spot size, the excitation power density was less than 1 $\mu W/cm^2$, which corresponds to an areal carrier density of the order of 10^5 cm^{-2}.

III. RESULTS AND DISCUSSION

The dashed curve in Fig. 1 shows the PL spectrum of the SQW sample. Owing to the increased coherence of the exciton states, well-separated sharp exciton peaks (linewidth <3 meV) are observed at energies of 1.600, 1.605, and 1.611 eV which correspond to growth islands *A*, *B*, and *C*, respectively, in agreement with the previous investigations.[14] The emission peak from island *D* is not seen in the spectrum because of the weak excitation intensity employed. The proportion of the peak intensities is also in agreement with the previous results[14] in spite of the difference of the excitation spot size. This result exemplifies uniformity of the distribution of the SQW islands.

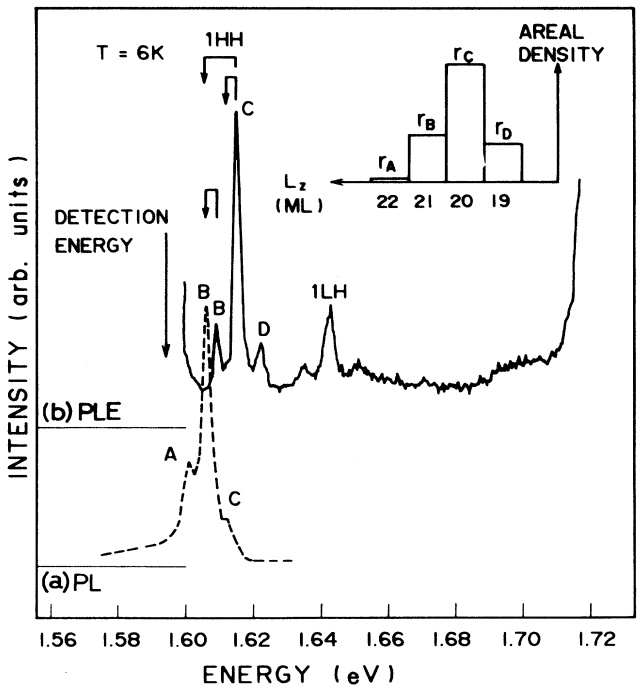

FIG. 1. PL [dashed curves (a)] and PLE [solid curve (b)] spectra of the 19–22-ML GaAs SQW. The excitation energy is 2.066 eV for PL. The detection energy for PLE is indicated by a downward arrow. The inset schematically shows areal density distributions of the four SQW growth islands.

When we detected at the lower-energy shoulder of peak *A* and the energy of the excitation light was scanned, the PLE spectrum shown by the solid curve in Fig. 1 was obtained. Distinct resonances for the lowest 1HH and light-hole (1LH) excitons associated with each of the islands are clearly separated. In the PLE spectrum the areal density of the island *C* is the highest. The leading 1HH emission peak *B* is red shifted from the main 1HH resonance peak *C* by 9 meV (Stokes shift). This finding is consistent with the previously observed dominance of peak *C* in the PL spectra taken at temperatures higher than 60 K due to the thermal population.[14] Therefore these results already show strong evidence for the in-plane exciton localization at low temperatures from island *C* to island *B* within the SQW's (inter-growth-island exciton localization). In addition, we observe that the 1HH emission peak is red shifted by 2–4 meV from the corresponding 1HH resonance line within each island. This result demonstrates that the excitons are localized even within the individual SQW island at locally wider well regions (intra-growth-island exciton localization), also in agreement with the previous studies of the temperature dependence of the PL peak energy.[14]

Figures 2(a)–2(d) show decay curves (in log scale) measured at 15 K of the PL intensity at the discrete exciton emission bands *A*, *B*, *C*, and *D* under direct excitation. Obviously, the decays are strongly dependent on the detected emission energy of the exciton bands. We should note the delayed rise of the exciton emission at band *A* compared to band *D*, which reflects the exciton transfer from the high-energy bands to the low-energy ones. In curves 2(a)–2(d), the time behavior does not exhibit a single exponential decay but it rather reveals distinct two-exponential decays with fast and slow components. At early times, the high-energy exciton bands are characterized by a fast decay. This fast decay slows down at the lower-energy exciton band. The fast decay component merges into the slow component at the lowest possible detection energy. Therefore it is straightforward to conclude that the fast decay rates are determined not by the radiative recombination lifetime but by the inter-growth-island exciton transfer due to the exciton localization towards the wider SQW islands. This conclusion is in excellent agreement with the Stokes shift ($C \rightarrow B$) observed in the cw PL and PLE experiments.

Conversely, we observe the slow decay component at later times. Its decay rate is approximated by a single-exponential decay with a time constant of about 420 ps, regardless of the detection energy. We measured similar energy-dependent time behaviors also under indirect excitation. As shown in Fig. 2(e) for the exciton band *A*, the decay rate at later times is well fitted by the same single-

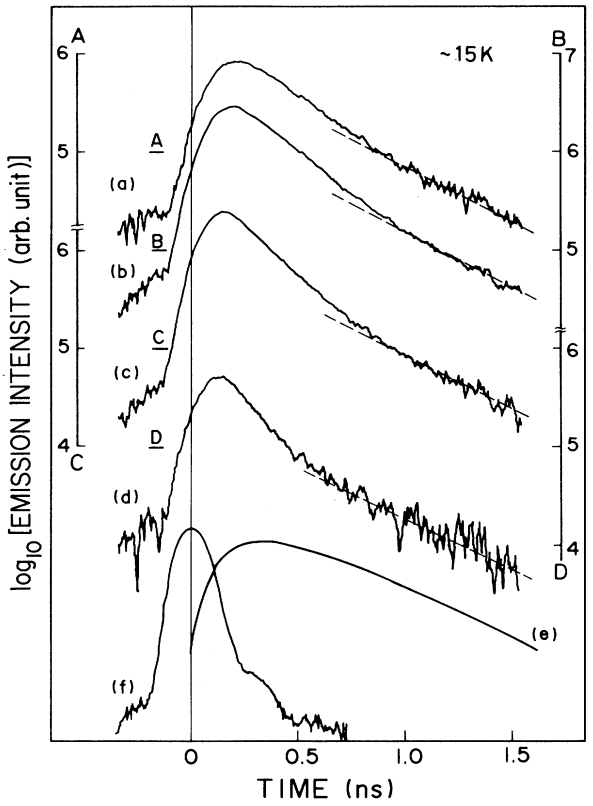

FIG. 2. Semilog plot of the time decay of the 1HH exciton emissions in the 19–22-ML GaAs SQW. Decay curves at the emission energy of the exciton band: (a) A; (b) B; (c) C; (d) D under direct excitation (excitation energy is 1.676 eV). Decay curve (e) is measured at the exciton band A under indirect excitation (excitation energy is 1.771 eV). Curve (f) is the instrumental response function for curves (a)–(e). Curves are vertically shifted for clarity.

exponential time constant as the slow component observed under direct excitation. Therefore we attribute this slow decay rate to the actual exciton radiative recombination lifetime, including nonradiative lifetimes, which is assumed to be constant for the four SQW islands. We note that the rise of the emission intensity under indirect excitation is significantly delayed. This delay is easily explained by the slower perpendicular transport of electron-hole pairs or excitons in the superlattice through tunneling.[8]

In what follows, we will quantitatively analyze the time-resolved PL results on the basis of a rate-equation model. For our experimental conditions, the 1.676-eV laser pulse uniformly creates electron-hole pairs at energy well above the 1HH exciton resonances. Therefore they rapidly relax in the same well where generated within 1 ps, emitting longitudinal-optical (LO) phonons.[16] Here we consider only the kinetics of exciton population, assuming that excitons are formed in a time much less than the transfer time. This does not matter since the same argument is valid for the electron-hole pairs. The number of excitons generated in the ith island is assumed to be proportional to the areal density r_i ($i=A$, B, C, and D) which is determined from the PLE spectrum. Because the energy separation of the exciton bands between the islands is greater than the lattice thermal energy (~ 1 meV), the uniformly distributed hot excitons can thermalize and then diffuse to the wider-well regions. In the meantime, they radiatively recombine within the radiative recombination lifetime τ_r ($=420$ ps). It is noteworthy from Fig. 2 that, at very early times (<100 ps), the emission intensity from band C is instantaneously higher than that from band B. When the exciton trapping time τ_{ij} ($i,j=A,B,C,D$) of the wider-well island j from the narrower-well island i is much faster than the radiative time τ_r, the excitons mostly recombine at the wider-well island. It should be noted that detrapping of the excitons by the narrower well is difficult in this case because of the large potential step. In addition, the excitons localize at spatially wider-well regions even within the island (intra-growth-island exciton localization).

Taking the exciton trapping time τ_{ti} within the ith island into account, we numerically solve four-component rate equations for the main exciton bands B and C, which are given by

$$\frac{dN_C}{dt}=r_C G(t)-\frac{N_C}{\tau_r}-\frac{N_C}{\tau_{tC}}-\frac{N_C}{\tau_{CB}}\ , \tag{1}$$

$$\frac{dn_C}{dt}=\frac{N_C}{\tau_{tC}}-\frac{n_C}{\tau_r}\ , \tag{2}$$

$$\frac{dN_B}{dt}=r_B G(t)-\frac{N_B}{\tau_r}-\frac{N_B}{\tau_{tB}}+\frac{N_C}{\tau_{CB}}-\frac{N_B}{\tau_{BA}}\ , \tag{3}$$

$$\frac{dn_B}{dt}=\frac{N_B}{\tau_{tB}}-\frac{n_B}{\tau_r}\ , \tag{4}$$

where N_i and n_i ($i=B$ and C) are exciton densities at the ith island terrace and the intrinsic defect sites within the ith island, respectively. $G(t)$ is the generation of excitons by exciting pulses. The time evolution of the emission intensity I_i is given by $(N_i+n_i)/\tau_r$ for the ith exciton band. In the calculations, we also consider the exciton transfer from the island terrace B to island A, but we neglect the exciton supply from island D because the contribution from band D is smaller by almost one order of magnitude. Numerical solutions for the emission intensity are fitted to the observed decay curve using the trapping times and the relative intensities of the exciton bands as adjustable parameters. The best fitted curves to the experimental data are obtained with the values $\tau_{CB}=140$ ps, $\tau_{BA}=400$ ps, $\tau_{tC}=1$ ns, and $\tau_{tB}=8$ ns and are shown by dashed curves in Fig. 3.

The above analysis is based on the model illustrated in the inset of Fig. 3. This model shows that the fast decay component is related to the exciton radiative recombinations at the island terrace. Therefore the decay rates as fast as 140 ps are strongly influenced by the inter-growth-island exciton transfer, and they depend on the energy of the exciton bands. The transfer time increases (decreases) when the available states at the lower-energy

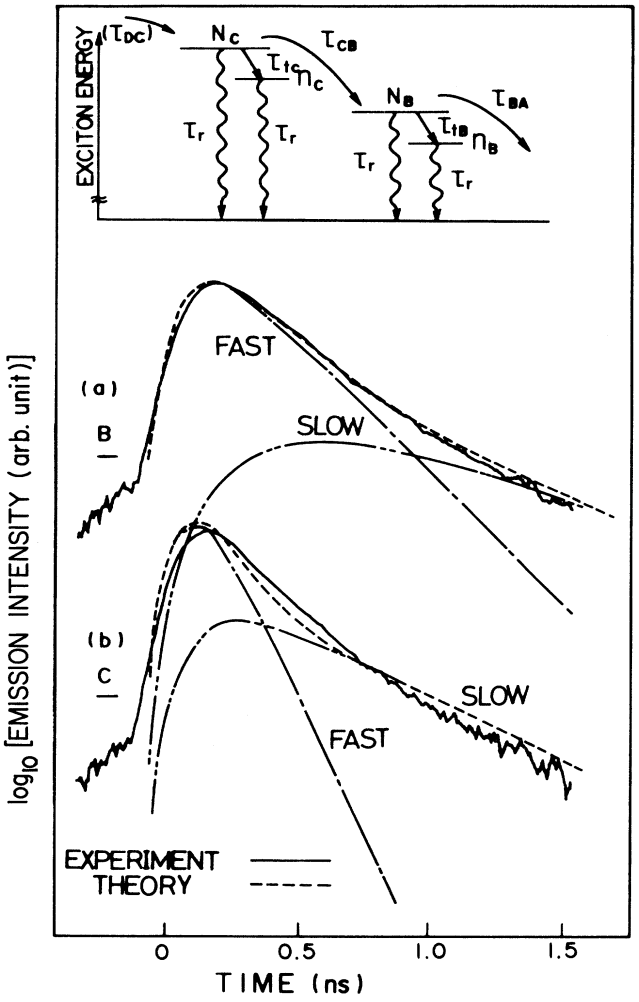

FIG. 3. Experimental (solid) and calculated (dashed) decay curves for the time evolution of the exciton-emission bands *B* and *C*. The fast (slow) decay components are shown by dot-dashed (double-dot-dashed) curves. The inset shows a schematic model for the exciton energy distributions which are used in the rate-equation analysis of the inter- and intra-growth-island exciton transfer.

exciton band decrease (increase). The transfer time is found to change also with the generated exciton density relative to the absolute number of the available sites. These results indicate that the exciton transfer dynamics depend on the geometric structures of the islands and the exciton-exciton interactions, which are beyond the scope of the present study. However, it is important to note that the results are consistent with the band-filling effects observed in the previous cw PL experiments under high-intensity excitation conditions.[13] The observed slow rate of the intra-growth-island exciton transfer is thus interpreted as a consequence of the reduced density of available defect sites.

The slow decay component is attributed to the exciton radiative recombinations at the intrinsic defect sites localized within the islands. During the thermalization process, the excitons are partly captured by such defect sites. Once the excitons are trapped by such sites, they only recombine there since they cannot move out of the defect sites because of the potential step. This mechanism provides the reason why the decay rate at later times can be fitted by the single-exponential time constant which is equal to the radiative recombination lifetime.

Finally, it is important to note that this dramatic change of the energy-dependent time behaviors disappears at higher temperatures. The fast decay component merges to the slow decay component at lattice temperatures above 60 K where the excitons thermally jump out of the potential steps. This result is easily understood by the exciton detrapping processes operative among the SQW growth islands and is in excellent agreement with previous studies of the temperature-dependent exciton population.[14]

IV. CONCLUSION

The recombination dynamics of the heavy-hole exciton have been investigated in spatially coherent GaAs single quantum wells, in which distinct excitonic transitions corresponding to different atomically flat terraces are observed. Under direct excitation, we observe distinct time behaviors with two-exponential decay rates, fast and slow, which are strongly dependent on the emission energy of the discrete exciton bands. Our experimental results indicate that the origin of the two-component time behaviors is a direct consequence of the in-plane exciton localization due to the inter- and intra-growth-island exciton transfer. Based on a simplified rate-equation analysis, a consistent model is obtained to explain the observed time behaviors. These results are in excellent agreement with those derived from the observed Stokes shifts in the cw photoluminescence and excitation spectra.

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