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Temperature dependence of the exciton population in emission spectra of GaAs single quantum wells with enlarged monolayer-flat growth islands

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The temperature dependence of the exciton population is investigated in GaAs single quantum wells (SQW) 19–22 monolayers wide with enlarged monolayer-flat growth islands grown by growth interrupted molecular-beam epitaxy. A comparative study with in-plane photocurrent spectroscopy shows that free-excitonic recombinations within the growth islands dominate the emission spectra even at room temperature. Detailed studies of the lowest heavy-hole exciton spectral characteristics as a function of the lattice temperature allow us to directly find the in-plane exciton localization and exciton detrapping processes between the SQW growth islands. A simple model analysis was made to explain observed temperature-dependent variations of the population at the exciton bands corresponding to the four different SQW growth islands, assuming Gaussian line-shape functions.

I. INTRODUCTION

Heterointerface properties are crucial for the spectral characteristics of the excitonic transitions in quantum-well (QW) heterostructures, which are widely applied for photonic devices, such as those based on the quantum-confined Stark effects (QCSE).[1,2] Exciton states in the usual quantum well are significantly broadened (inhomogeneous broadening[3–6]) by a perturbed potential field for quantum confinement, which results from unavoidable microscopic, statistical variations of the fluctuating well width as well as of the alloy composition in the barrier. These microscopic variations strongly affect the line shape of excitons in both emission and absorption spectra. Recent investigations[7–13] to improve the heterointerfaces in layered semiconductor structures revealed that QW growth islands can be prepared by growth-interruption techniques with atomically flat terraces larger in area compared to the exciton Bohr radius. Since the excitons have different confinement potentials, separated excitonic transitions with narrower linewidths are observed as a result of increased spatial coherence of the excitonic states. This provides us a unique opportunity to study the exciton population and the transfer of excitons to the different QW growth islands. Recently the exciton-trapping dynamics at low temperatures in the GaAs single QW (SQW) was studied by time-resolved photoluminescence experiments[14] and by optical investigations[15] under resonant excitation conditions to selectively create excitons in the different growth islands. Previously, the effect of lattice temperature on exciton trapping on intrinsic interface defects caused by well-width fluctuations was investigated by Delalande *et al.*[16,17] in the GaAs SQW but without enlarged terraces. In this paper, we present detailed investigations of the temperature dependence of the exciton population in a 19–22-monolayer-wide GaAs SQW with enlarged monolayer-flat growth islands by photoluminescence (PL) and in-plane photocurrent (PC) measurements. The high quality of our GaAs SQW sample grown by molecular-beam epitaxy (MBE) allows us to unambiguously study temperature-dependent exciton trapping and detrapping processes between the different SQW growth islands by observing the well-separated excitonic transitions by virtue of enlarged terraces. Firm evidence for the in-plane exciton localization is presented. It is shown that excitons are preferentially trapped at intrinsic defect sites spatially localized in the wider wells at low temperatures. Variation of the exciton population as a function of the lattice temperature is explained by a simple model calculation which takes into account thermal population of the individual exciton bands.

II. EXPERIMENT

A high-quality undoped GaAs SQW sample was grown at a nominal substrate temperature of 670 °C on a semi-insulating GaAs(100) substrate by MBE in a fully computer-controlled Varian Gen II system. The SQW sample consists of a GaAs well confined by GaAs/AlAs short-period superlattices (SPS) with 40 periods below and above.[18,19] Growth was made with 2 min growth interruption under arsenic fluxes at each SQW heterointerface to enhance Ga and Al migration, which is expected to enlarge areas of monolayer-flat SQW terraces. The well widths (L_z) of the different SQW growth islands are ranged from 19 to 22 monolayers (ML). This was confirmed by a calibration curve to relate the SQW transition energy and the L_z value, as discussed later. The well width of the SPS layers is 3.4 nm (~12 ML) and the barrier thickness (L_B) is 1.2 nm (~4.2 ML). Our SQW heterostructure was further sandwiched between two $Al_{0.3}Ga_{0.7}As$ layers to avoid carrier leakage. Photoluminescence measurements were performed in a standard lock-in detection based system equipped with a variable temperature cryostat (10.5–300 K). A Kr-ion laser at 647.1 nm was used as an excitation source. The carrier density is estimated to be 10^{17} cm^{-3} at the excitation power of 30 W/cm^2 assuming a 100-μm spot size. In-plane photocurrent spectra[20] were measured at 77 K and at room temperature (293 K) to extract information concerning the exciton-absorption spectral features.

III. RESULTS AND DISCUSSION

A. Free-excitonic transitions

Figure 1 shows PC (solid curve) and PL (dotted-dashed curve) spectra of the SPS-confined SQW sample at (a) 77 K and (b) 293 K. In the PC spectrum (a), two clear peaks due to exciton resonance absorption by the SQW layer are seen at 1.602 eV (774 nm) and 1.632 eV (759.5 nm). Resonance enhancement effects observed on the absorption spectral features allow us to assign them as the $n=1$ electron to heavy-hole (1 hh) and to light-hole (1lh) free-excitonic transitions of the SQW.[20] Additional intense PC structures seen at the higher-energy sides are attributed to the GaAs/AlAs SPS layers. The assignment of these SPS peaks was argued in other publications[19] and will not be discussed here. In the PL spectrum, on the other hand, fine structures (peak and shoulders: *B*, *C*, and *D*) were observed for the SQW layer, as in Fig. 1(a), at energies of $B=1.598$ eV (776 nm), $C=1.603$ eV (773.5 nm), and $D=1.611$ eV (769.5 nm) at 77 K. The energy of the leading peak *C* coincides within 1 meV with that of the 1hh free-exciton resonance energy. Because of the closeness of the leading peak energies in the PC and PL spectra, it is concluded that the luminescence is dominated by the 1hh free-excitonic transitions.[20] The same argument is also applied even at room temperature, as is demonstrated in Fig. 1(b). We also attribute emission peaks *B* and *D* in Fig. 1(a) to free-excitonic transitions belonging to the different growth islands whose well widths differ by plus or minus one monolayer, as discussed later. A lack of the corresponding fine structures in the PC spectrum might arise from a large diameter of the probe light (3 mm ϕ) because of some averaging effects on the size distributions of terraces.

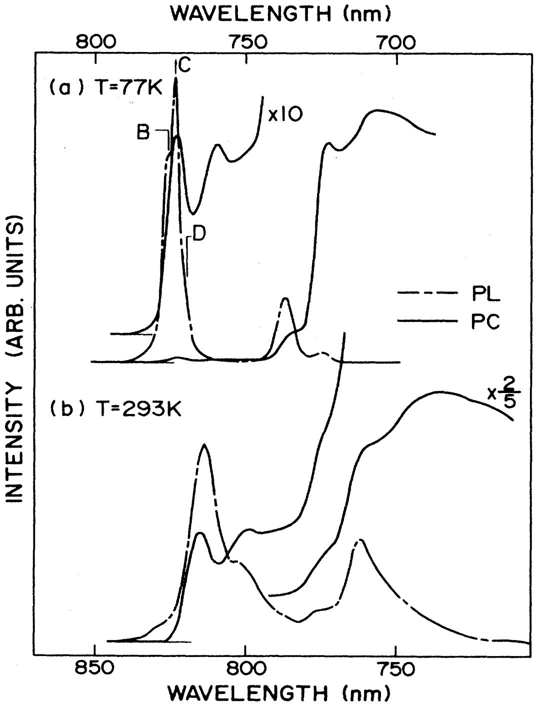

FIG. 1. In-plane photocurrent (PC) and photoluminescence (PL) spectra of the SPS-confined SQW sample at (a) 77 K and (b) 293 K.

B. Temperature dependence of PL spectra

Figure 2 shows experimental results of the temperature dependence of PL spectra for the SQW sample in the range between 200 and 11 K. At temperatures above 100 K, the peak *C* is predominant, and the spectral line shape does not change significantly except for the emission intensity due to the 1lh exciton transition. When the lattice temperature (*T*) was decreased below 100 K, however, substantial changes were observed in the relative intensities of the SQW peaks associated with the different growth islands (*B*, *C*, and *D*). With decreasing temperature, peak *B* gradually increased its intensity and dominated the spectrum at 11 K, while the intensities of peak *C* and shoulder *D* relative to peak *B* monotonously decreased. At the lower-energy side, a new structure related to terrace *A* appeared at lower temperatures. As shown in the inset of Fig. 2, well-separated triplet peaks (*A*, *B*, and *C*) were more clearly observed when the exci-

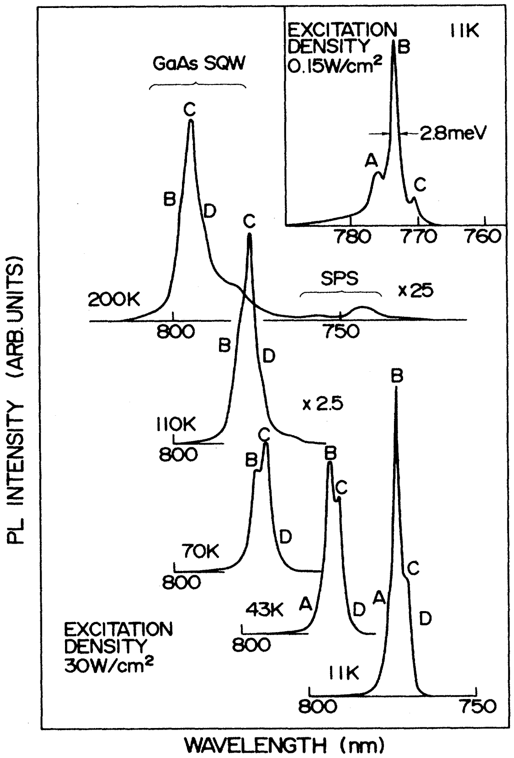

FIG. 2. Temperature dependence of photoluminescence spectra of the SPS-confined SQW. The excitation power density (carrier density) is 30 W/cm² (10^{17} cm^{-3}). The inset shows the photoluminescence spectrum at 11 K under the excitation power density of 0.15 W/cm².

tation power was decreased to 0.15 W/cm^2. The increased spectral weight of peak C relative to peaks A and B under high excitation is attributed to the band-filling effects.[21] The energy positions for peaks A, B, and C at 11 K are located at 1.598 eV (775.8 nm), 1.603 eV (773.4 nm), and 1.610 eV (770.3 nm), respectively. The linewidth of the leading peak B is found to be 2.8 meV. The observed splitting (5–8 meV) of the free-excitonic emissions indicates that the size of the SQW growth islands is, as expected, far larger than the exciton Bohr radius (15 nm). It is noteworthy that the PL linewidth for our similar SQW sample prepared for comparison without growth interruption is typically 5 meV under the same growth and measurement conditions.[21] Therefore, the narrow linewidth for the present SQW sample is taken as evidence for the Ga and Al ordering along the heterointerfaces and the spatial coherence of exciton states. In order to get a calibration curve to quantitatively relate the SQW 1hh transition energy and the L_z value, we prepared four other similar SPS-confined SQW samples but with different SQW well widths ranging from 7.0 to 4.1 nm.[22] Results of the SQW 1hh transition energies, as well as the calculated 1hh subband spacing[23] based on the Krönig-Penney model, are shown in Fig. 3 together with the temperature evolution of the transition energies for terraces A, B, C, and D. We find that the transition energy is strongly dependent on the well width (L_z) and that the measured values at low temperatures are precisely on the empirically determined calibration curve if peaks A, B, C, and D are associated with the SQW terraces with widths of 22, 21, 20, and 19 ML, respectively. This result provides us with firm evidence that the excitonic emission peaks (A, B, C, and D) are in fact arising from the monolayer-flat SQW growth islands. It should be noted, however, in Fig. 3 that the 1hh emission peaks still exhibited Stokes shifts of ~4 meV at low temperatures if deviations from a dashed curve [vertically shifted temperature dependence of the bulk GaAs direct band gap $E_g(T)$] are attributed to the in-plane exciton localization. Such deviations may be caused by the remaining disorder at the bottom (GaAs on AlAs) heterointerface, as discussed by Tanaka and Sakaki.[13]

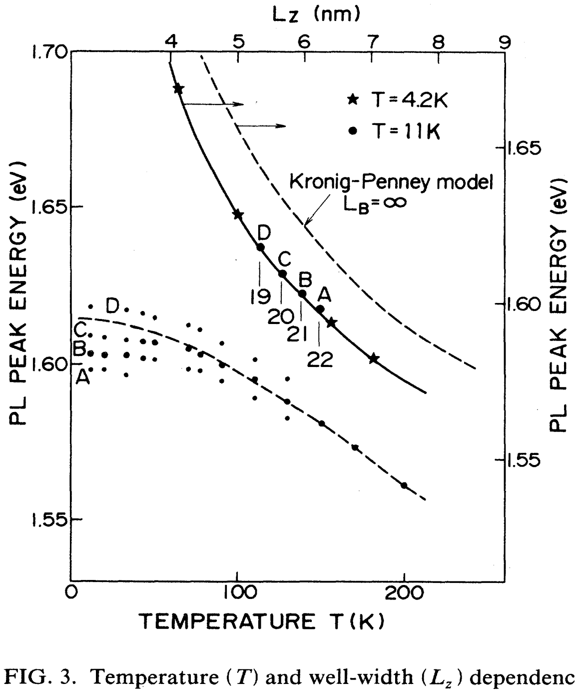

FIG. 3. Temperature (T) and well-width (L_z) dependences of the 1hh exciton transition energies for SQW growth islands A, B, C, and D. The lower dashed curve is obtained by fitting the temperature dependence of the bulk GaAs band gap to the 1hh exciton peak energy at 200 K. The solid curve in the upper figure is the guide to the eye.

The above temperature-dependent results of the SQW emission spectra are qualitatively interpreted as follows (see Fig. 4). When electron-hole pairs are introduced into the SQW by photoexcitation, we assume that excitons are quickly formed[14] and that they can diffuse towards the local potential minimum in the wider wells. At lower temperatures, this energy-relaxation process (exciton cooling) can quickly occur via acoustic-phonon emissions[24] before they are radiatively recombined. Detrapping of the exciton by the narrower well is difficult when the exciton thermal energy ($T < 50$ K) is less than the potential step (~5 meV) between the SQW growth islands. Note in Fig. 2 that peak B has the highest intensity at 11 K, while peak C is dominant above 70 K. Thus, we deduce that the portion of the exciton band associated with terrace C is the highest, while those of A, B, and D are smaller. The dominance of exciton peak B at 11 K is therefore attributed to the in-plane exciton localization, i.e., the exciton transfer from the narrower wells to the wider wells ($D,C \rightarrow B,A$). In this case, the relative emission intensities are basically determined by the competition between the radiative recombination lifetime within the terrace and the exciton transfer to the neighboring island regions with the wider well widths. The spatially localized wider SQW wells (B and A) form intrinsic defect states, which

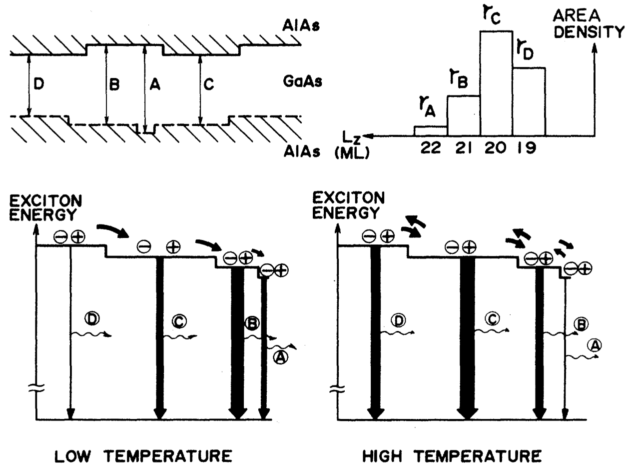

FIG. 4. Schematic model to sketch the behavior of excitons in wells with enlarged monolayer-flat growth islands at low and high temperatures. The upper left part represents the spatial configuration of the well with some disorder at the bottom heterointerface (boundary with the dashed lines). The upper right part represents the density distributions of the four SQW growth islands.

is observable as separate peaks by virtue of enlarged terrace widths. Our experimental results in Fig. 2 suggest that the exciton transfer time from terrace C to terrace B is much faster than the radiative lifetime of 0.3 ns (Ref. 25) for $L_z \sim 6$ nm, in general agreement with Deveaud *et al.*[14] At higher temperatures, on the other hand, the excitons thermally occupy the higher-energy states, and the exciton detrapping process becomes important, as schematically explained in Fig. 4. When the thermal energy of the exciton exceeds the potential step, excitons can go back to the narrower well regions. In such high-temperature regimes, the relative intensities of the SQW emissions are proportional to the area ratios of the growth islands, since the excitons are thermalized and the occupation number depends mainly on the state density of the specific exciton band.

TABLE I. Summary of the SQW well width (L_z), the area ratios r_i, and the energy center E_i of the exciton band associated with the ith growth island. The energy of E_A is taken to be zero.

Growth island	A	B	C	D
L_z (ML)	22	21	20	19
r_i (%)	1	14	54	30
E_i (meV)	0	5	11.5	19.5

C. Theoretical consideration

In this section, we would like to quantitatively explain the temperature-dependent variations of the emission intensities due to the exciton detrapping process between the four SQW growth islands. For the calculation, we first have to estimate the area ratios r_i of the ith terraces where $i = A, B, C$, and D. These values may be estimated from the emission peak intensity ratios I_i/I_j at 77 K by the equation

$$I_i/I_j=(r_i/r_j)\exp[-(E_i-E_j)/kT] , \tag{1}$$

assuming a Boltzmann factor $\exp(-E_i/kT)$ for the exciton occupation, where E_i is the energy center of the ith exciton band and k is the Boltzmann constant. Values of r_i estimated are listed in Table I. We further assume that the line-shape function for the excitonic recombinations can be approximated by a normalized Gaussian function:

$$f_i(E)=(1/\sigma\sqrt{2\pi})\exp\{-[(E-E_i-\sigma)/\sigma]^2/2\} . \tag{2}$$

The standard energy deviation σ is given by half of the linewidth of the PL peak, which we take to be a constant value of 1.4 meV. Then, the emission intensity $I_i(T)$ from the ith island at T is approximated by the following equation:

$$I_i(T)=\eta_i r_i \int_0^\infty f_i(E)\exp(-E/kT)dE \Big/ \sum_i r_i \int_0^\infty f_i(E)\exp(-E/kT)dE . \tag{3}$$

Our approximation is based on the following assumptions. (1) We assume that the emission intensity is simply proportional to the number of excitons occupied at the ith exciton band. (2) The radiative recombination lifetimes are assumed to be the same for the four SQW growth islands. (3) Temperature dependence of the internal quantum efficiency η_i may be approximated by an empirical equation of the form given by Zucker *et al.*,[26]

$$\eta_i=1/[1+CT\exp(-E_a/kT)] , \tag{4}$$

where E_a is an activation energy for the nonradiative recombination, and C is a fitting parameter. The values of $E_a=30$ meV and $C=0.1$ K^{-1} are taken to fit the experimental data. By Eq. (4) we would like to describe all effects of the nonradiative recombination channels against the 1hh free-excitonic recombination including recombinations at higher-energy states such as the continuum states (exciton dissociation) and the 1lh exciton. The above model is only a rough description of the exciton detrapping process, which limits the accuracy of our calculation. Experimental and calculated results of the relative emission intensity are shown in Fig. 5 against the lattice temperature. In spite of the crudeness of our model, agreement between theory and experiment is quite satisfactory. It should be mentioned, however, that agreement is poor at temperatures higher than 150 K. This is because we do not attempt to optimize the parameters to calculate η_i under the situation where not enough information is available at present to discuss temperature dependence of the internal quantum efficiency for the two-dimensional exciton in QW's. Disagreement for the

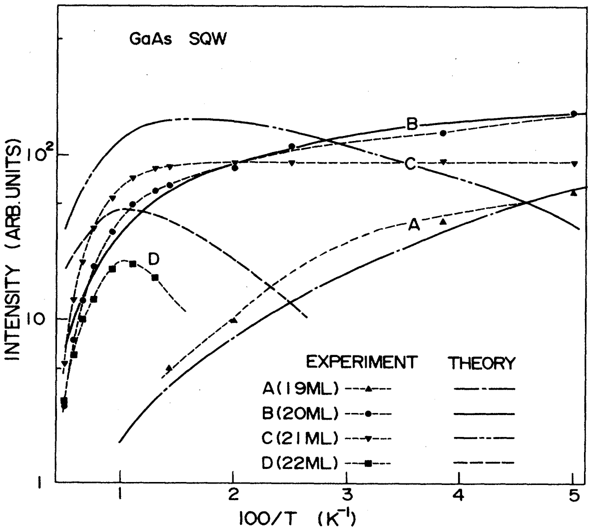

FIG. 5. Temperature-dependent variations of the SQW emission intensity associated with islands A, B, C, and D. The excitation power density is 30 W/cm^2.

emission intensity of island *C* below 40 K is partly due to the band-filling effects under the intense excitation, which is neglected in our calculations but is experimentally observed in Fig. 2. In spite of the fact that our model is simple, the temperature-dependent variations of the emission intensity between the four SQW growth islands are well explained by including the exciton population due to exciton detrapping at the energetically higher exciton band. This is clearly illustrated in our experiments by virtue of the increased coherency of the excitonic states associated with the enlarged terraces.

IV. CONCLUSION

Temperature dependence of the exciton population is investigated in 19–22-monolayer-wide GaAs single quantum wells with enlarged monolayer-flat growth islands by photoluminescence and in-plane photocurrent measurements. The high quality of our GaAs SQW sample allows us to provide firm evidence for the in-plane exciton localization by observing temperature dependence of the well-separated lowest heavy-hole free-excitonic transitions in emission spectra by virtue of enlarged terraces. Clear variations of the exciton population due to the exciton detrapping process between the SQW growth islands are experimentally demonstrated and theoretically explained as a function of the lattice temperature by a simple model which takes into account thermal population of the individual exciton bands associated with the enlarged terraces.

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