Two-dimensional excitonic emission in InAs submonolayers

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Photoluminescence (PL) and time-resolved photoluminescence (TRPL) were used to study optical emissions of ultrathin InAs layers with average layer thickness ranging from $\frac{1}{12}$ to 1 ML grown on GaAs substrates. We have found that the inhomogeneous broadening of the PL from InAs layers can be well described by the quantum-well model with InAs islands coupling to each other and being regarded as a quasiwell. From the temperature dependence of the exciton linewidth, the exciton–LO-phonon scattering coefficient was found to be comparable to that in conventional two-dimensional quantum wells. In the TRPL measurements, the PL decay time increases linearly with temperature, which is a typical characteristic of free excitons in quantum wells. All these results indicate that the excitons localized in InAs exhibit two-dimensional properties of quantum wells, despite the topographical islandlike structure. [S0163-1829(96)01647-5]

I. INTRODUCTION

Recently much interest has focused on the growth of highly strained In(Ga)As-GaAs heterostructures on GaAs substrates[1–10] because such a strained epitaxy may provide a direct means to form quantum wires or dots without any patterning process. It has been reported that the growth of highly strained InAs layers onto GaAs substrates could lead to spontaneous formations of clusters, so-called self-organized quantum dots.[3]

More recently, Wang *et al.*[7] found that a uniform array of InAs wires elongated along the $[01\bar{1}]$ direction is formed in the case of submonolayer (SML) InAs deposition on GaAs substrates from an optical anisotropy study. Scanning-tunneling-microscopy studies have revealed that the growth of InAs SML structure on (100) or vicinal GaAs surfaces results in a spontaneous formation of coherent InAs ML-high wirelike islands with a width of ~4 nm and an elongation along the $[01\bar{1}]$ direction.[8] Transmission-electron-microscopy (TEM) studies also displayed the coherent islands with a typical lateral size of 5 nm in an InAs SML deposited on (100) GaAs.[9] Many optical studies on an InAs SML structure demonstrated its high luminescence efficiency,[7] large exciton oscillator strength,[10] and huge binding energy.[11] The above phenomena were interpreted in terms of the lateral quantum confinement. On the other hand, Brandt, Lage, and Ploog[12] found that excitons localized in an ultrathin (0.8-ML) InAs layer still show two-dimensional properties in optical response experiments. Clearly, the optical properties of such a system still remain poorly understood. Does an exciton in such structures exhibit zero-dimensional (0D) properties? Does an exciton feel the confinement of the isolated islands when their lateral size (approximately 50 Å) (Ref. 9) is smaller than the exciton diameter (250 Å)?

In this paper, we used temperature-dependent continuous-wave (cw) and time-resolved photoluminescence (PL) to study the optical properties of SML InAs structures embedded in a GaAs matrix. Based on the temperature-dependent PL linewidth analysis, the exciton–LO-phonon scattering coefficient was estimated to be 12 meV, which is comparable to that of typical quantum wells. In time-resolved photoluminescence (TRPL) measurements, the measured PL decay time is a strong function of the average InAs thickness, from 2.1 ns for $\frac{1}{12}$ ML to 0.35 ns for 1 ML. This result is similar to that obtained in very thin quantum wells (QW's), which was attributed to the penetration of the envelope wave function of electrons into the barriers. In addition, the PL decay time increases linearly with temperature, showing a typical characteristic of free excitons in quantum wells. From our experiments we conclude that the exciton emission in InAs SML structures is of two-dimensional (2D) nature. The structure can be treated as a very thin quantum well, and the InAs island distribution as interface roughness.

II. EXPERIMENTAL DETAILS

The structures were grown by elemental source molecular-beam epitaxy on (100) GaAs semi-insulating substrates. After oxide desorption a 0.3-μm-thick GaAs buffer

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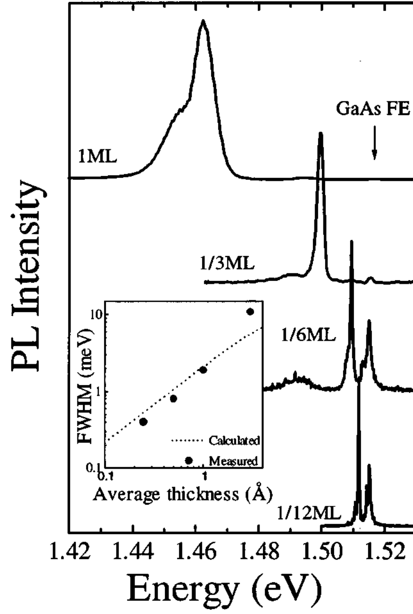

FIG. 1. PL spectra of four samples at 10 K. The excitation power was selected at a very low level of 0.1 mW. The inset depicts the calculated (dashed line) and measured exciton linewidth dependencies on the average InAs layer thickness.

layer was grown at 600 °C. This layer was followed by a 40-period 25-Å GaAs/25-Å $Al_{0.4}Ga_{0.6}As$ superlattice (SL) to trap impurities and to prevent nonequilibrium carrier spreading into the semi-insulating substrate. The SL was followed by 2000-Å GaAs, then the substrate temperature was lowered to 450 °C, and the arsenic beam equivalent pressure was increased to 10^{-5} Torr. Subsequently, a 100-Å GaAs layer was deposited, which was followed by a single InAs layer, and 20 Å of GaAs was grown on top. The substrate temperature was then increased to 600 °C, with the arsenic pressure reduced to 3×10^{-6} Torr. Under this condition, a 2000-Å-thick GaAs layer and a 40-period 25-Å GaAs/25-Å $Al_{0.4}Ga_{0.6}As$. Superlattices were grown to prevent surface recombination and to avoid surface-related electric fields. Finally, a 100-Å GaAs cap layer was grown to protect the surface. The details of the growth were published elsewhere.[7]

For TRPL measurements, the excitation of the samples was carried out with a dicyanomethylene (DCM) dye laser synchronously pumped by a mode-locked Ar^+ laser with a repetition rate of 82 MH_z. The PL signal was dispersed by a small monochromator and detected, by a 2D synchroscan streak camera with a total time resolution of 30 ps. For cw PL measurements, the dye laser worked in a continuous-wave mode, and the signal from the samples was dispersed by a SPEX 1403 (0.85 m) spectrometer with an energy resolution of 0.1 meV, and detected by a cooled GaAs photomultiplier and a photon counting system.

III. RESULTS AND DISCUSSIONS

A. cw PL measurements

Figure 1 shows low-temperature PL spectra of structures containing InAs layers with different average InAs layer thickness of $\frac{1}{12}$, $\frac{1}{6}$, $\frac{1}{3}$, and 1 ML (1 ML=3.00 Å), respectively. Both InAs-related exciton transitions and GaAs free-exciton (FE) emissions (1.515 eV) are observed in all four samples. Three remarkable features in the spectra are that (i) the full width at half maximum (FWHM) of the InAs heavy-hole (hh) exciton line decreases with decreasing InAs thickness, from 9.5 meV for 1 ML to 0.4 meV for $\frac{1}{12}$ ML; (ii) the InAs-related emission energy increases with the decrease of the average thickness of the InAs layer; (iii) the PL intensity ratio between the InAs-related emission energy and that from GaAs emissions decreases with decreasing InAs thickness. In Table I we list sample parameters, and the exciton energies and linewidths obtained from a PL measurement at 10 K.

TABLE I. Average thickness of SML InAs samples, and the exciton energy and FWHM obtained from PL spectra at 10 K.

Sample	Average L_Z (ML)	E_{hh} (eV)	FWHM (meV)
A	1	1.4635	9.5
B	1/3	1.5000	1.9
C	1/6	1.5097	0.8
D	1/12	1.5118	0.4

The exciton line broadening in semiconductor quantum wells is related to various scattering processes, and to an inhomogeneous fluctuation of the well thickness. At low temperatures the exciton linewidth is expressed as

$$\Gamma=\Gamma_0+\Gamma_{in}, \quad (1)$$

where Γ_0 is the homogeneous linewidth of the exciton line, mainly due to acoustical-phonon interaction and spin-flip scattering,[13] and Γ_{in} is the inhomogeneous spectral broadening mainly originated from interface roughness. According to the virtual-crystal approximation (VCA) model proposed by Singh and co-workers,[14] the exciton cannot ''feel'' the individual single growth islands when the lateral size of such islands is smaller than the exciton Bohr radius (a_B). The island distribution only contributes to the spectral broadening Γ_{in} of the exciton emission, which can be written as[15]

$$\Gamma_{in}=\left.\frac{\partial E_g^{2D}}{\partial L_Z}\right|_{L_Z=\langle L_Z\rangle}\delta_s, \quad (2)$$

$$\delta_s=\delta\sqrt{p(1-p)}\,\frac{d_I}{2a_B}, \quad (2')$$

where d_I is the average lateral extension of interface islands of height δ, a_B the Bohr radius of the exciton, $p(0<p<1)$ the average island coverage of the heterointerface, and $\langle L_Z\rangle$ the mean QW thickness, while δ_s represents the standard deviation.

Using this quantum-well model we fitted the exciton linewidth dependence on the InAs layer thickness L_Z under the assumptions that (i) the InAs layers are treated as very thin quantum wells, and (ii) the InAs island structure is regarded as interface roughness which inhomogeneously broadens the exciton linewidth. In the inset of Fig. 1 we show the best fit to the exciton linewidth dependence on the average InAs layer thickness, together with the experimental data (solid circles). In the calculation the effective-mass approximation

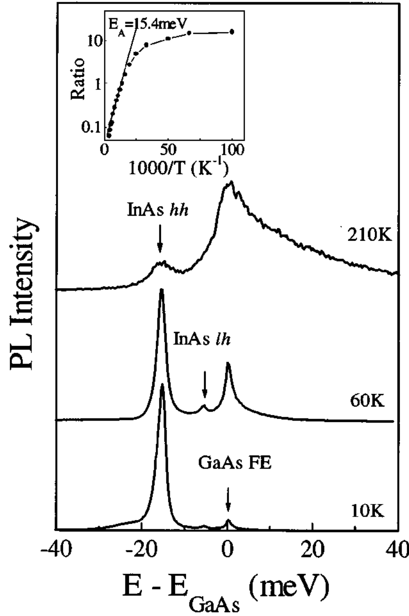

FIG. 2. PL spectra measured at several temperatures for sample B under the excitation power of 1 mW. The inset shows the Arrhenius plot of the PL intensity ratio between the InAs hh and GaAs FE.

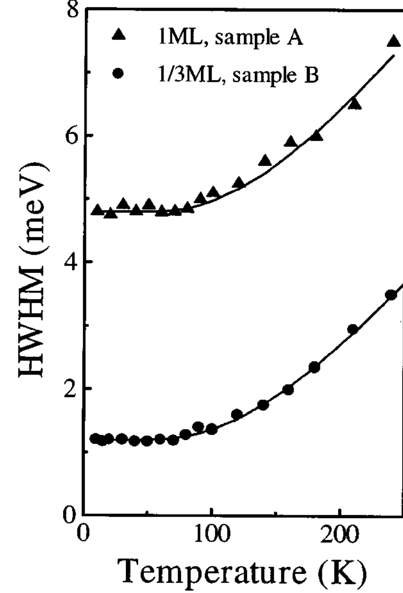

FIG. 3. Temperature dependencies of the FWHM for samples A and B. The solid lines are the best fit to Eq. (3).

(EMA) was used, and the well width was replaced by the average InAs thickness.[16] For simplicity the same standard deviations δ_s were used for all samples.[17] It can be seen from the inset of Fig. 1 that the calculated FWHM's are in good agreement with the measured ones, indicating that the quantum-well model is applicable to the InAs SML structures. In other words the InAs SML can indeed be treated as a conventional quantum well with its interface roughness determined by the average InAs coverage. From the line broadening, we may estimate the lateral size of the growth islands. Assuming the exciton Bohr radius to be 150 Å, the value often used in 2D quantum wells, for $\langle L_Z \rangle = \frac{1}{3}$ ML ($p = \frac{1}{3}$) the estimated lateral size d_I of the 1-ML-high (δ=1 ML) islands[7–9] is about 35 Å, which is in good agreement with the TEM results observed by Alonso *et al.*[9]

PL spectra measured at several temperatures are shown in Fig. 2 for sample B. The InAs hh exciton dominates the entire luminescence processes at low temperatures. As temperature increases, the intensity of the InAs luminescence becomes weaker. At high temperatures, the GaAs free-exciton recombinations gradually dominate the luminescence processes. This is caused by the thermal activation of InAs excitons back to GaAs, where they recombine through radiative and nonradiative channels. The inset shows the Arrhenious plot of the intensity ratio between the InAs- and GaAs-integrated luminescence intensities. From the curve we can obtain the activation energy E_A. It is very interesting that E_A exactly equals the confinement energy. This indicates the high quality of the investigated samples.[18] For a 1-ML sample, similar results are obtained (not shown here). Another remarkable feature in Fig. 2 is that, as temperature increases, the luminescence line is broadened, especially in the high-temperature regime. Figure 3 shows the InAs hh exciton linewidth as a function of temperature. [Note that the linewidth represents the half-width at half maximum (HWHM) here. At high temperatures we determined the HWHM from the low-energy side of the spectra to avoid the influence of thermal population effect of high-energy levels and the interference with the GaAs luminescence.]

In the following we further explore the two-dimensional nature of excitons in SML InAs through the temperature dependence of the exciton linewidth, which reflects the exciton-phonon scattering coefficients.[19–23] As the acoustic-phonon broadening is very small compared with the LO-phonon broadening, a typical expression for the temperature-dependent linewidth $\Gamma(T)$ can be written[19,20]

$$\Gamma(T) = \Gamma(0) + \Gamma_{LO}/[\exp(E_{LO}/k_B T) - 1], \tag{3}$$

where $\Gamma(0)$ is the inhomogeneous broadening at zero temperature, and E_{LO} is the LO-phonon energy. Γ_{LO} is the LO-phonon-scattering coefficient, proportional to the sum of possibilities of an exciton scattered from the ground state to all possible states by LO phonons. Apparently, the second term in Eq. (3) represents broadening due to LO-phonon anti-Stokes scattering.

Equation (3) is valid for all dimensionalities. The difference among different dimensionalities lies in the value of the scattering coefficient. When a system's dimensionality approaches zero, the electron density of states is no longer continuous. Difficulty should then appear in the exciton–LO-phonon-scattering processes because of the requirement of energy conservation in scattering processes.[24,25] Therefore, in the zero-dimensional system, no significant thermal broadening effect would be expected.[4]

The data in Fig. 3 can be well fitted using Eq. (3) with E_{LO}=36 meV of the GaAs LO-phonon energy. Note that here the LO phonons are lattice vibrations of the GaAs matrix, indicating that InAs embedded in GaAs is similar to impurities in GaAs. The scattering coefficient Γ_{LO} was found to be $\sim 12 \pm 1$ meV for both samples. Such a value is much smaller compared with that of GaAs bulk material [20 meV

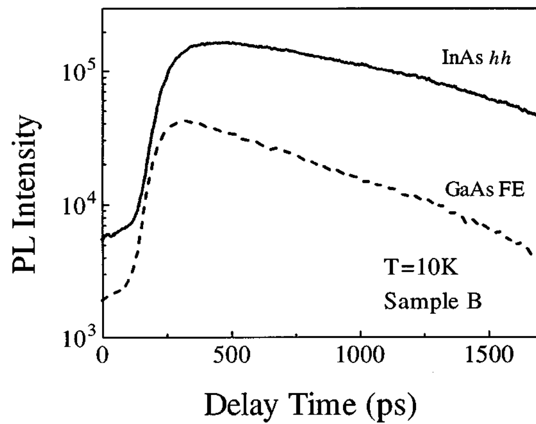

FIG. 4. Temporal evolution of GaAs and InAs exciton luminescence for SML sample B.

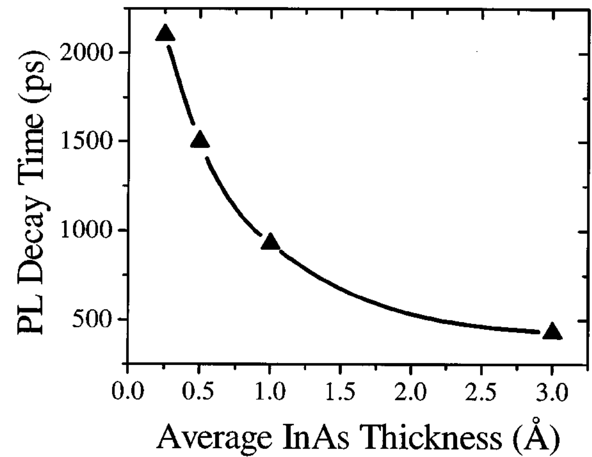

FIG. 5. Well-width dependence of the PL decay time.

(Ref. 20)], indicating confinement in the growth direction. Also, it is very close to that found in $GaAs/Al_xGa_{1-x}As$ (Refs. 21 and 22) and $In_xGa_{1-x}As/GaAs$ (Ref. 23) quantum wells, demonstrating that scatterings between carriers and LO phonons are still efficient and that carriers in InAs SML structures have two-dimensional properties.[16] In other words, the LO-phonon-scattering rate is not reduced, though InAs has an island distribution in the GaAs matrix,[7–9] i.e., there is no phonon bottleneck effect. This high LO-phonon-scattering rate accounts for high luminescence efficiencies in SML structures. The fast scattering rate is further confirmed in the study of hot-carrier relaxation in InAs SML structures.[26]

B. Time-resolved PL measurements

The decay process of exciton emission in an InAs SML has been studied using time-resolved PL spectroscopy. Figure 4 shows the typical temporal profiles of GaAs and InAs luminescence at 10 K for the $\frac{1}{3}$-ML sample. The intensity of the InAs luminescence decays monoexponentially after an approximate 250-ps rising time. The relative delay (~80 ps) of the InAs luminescence with respect to the GaAs luminescence is believed to be associated with the carrier trapping process from the GaAs barrier to the InAs islands. The high efficiency of the luminescence indicates efficient trapping and relaxation processes regardless of the islandlike structure. Moreover, the measured 250-ps rise time is comparable to that found in typical 2D quantum wells,[27] suggesting again that the phonon bottleneck effect is not dominant in our InAs structure. In addition, the high efficiency of the luminescence, the narrow linewidth, and the monoexponential decay indicate that the emission processes are dominated by excitonic recombinations.[16] The exciton radiative lifetime could thus be directly obtained from the exponential decay curves. In Fig. 5 we plotted the PL decay time τ of the InAs hh exciton emission as a function of the InAs average thickness at 10 K. An increasing decay time with decreasing InAs thickness was observed, and a very long PL decay time (2.1 ns) was measured from the sample of $\frac{1}{12}$ ML.

It is generally known that the exciton lifetime is inversely proportional to its coherence volume over which the coherent polarization of the exciton persists.[28] The lifetime in quantum-dot or quantum-wire systems is expected to be longer than that in QW's because of the reduction of the coherence volume.[29] However, this is only valid for the coherent volume being larger than the exciton diameter,[30] and is not effective to describe the exciton dynamics in InAs SML structures where the lateral extension of the growth islands is much smaller than the exciton diameter. We found that the recombination process in our InAs SML can be understood in the framework of thin quantum wells. As reported for very narrow QW's,[31] the increasing PL decay time with decreasing InAs thickness can be attributed to penetration of the electron envelope wave function into the GaAs barriers. In the limit of thickness approaching zero, the quasi-2D excitons become quasi-3D excitons of the barrier material.[31] As shown in Fig. 5, the measured decay times are 2.1, 1.5, 0.8, and 0.3 ns for InAs average thicknesses of $\frac{1}{12}$, $\frac{1}{6}$, $\frac{1}{3}$, and 1 ML, respectively. It is very clear that the exciton lifetime approaches the bulk GaAs value[32] (10 ns at 10 K) as the average InAs thickness decreases. In addition, the increasing lifetime in the SML with thinner InAs average thickness is responsible for the decreasing relative PL intensity of InAs hh exciton emission, as seen in Fig. 1.

To explore the carrier dynamics in InAs SML structures further, we also investigated the temperature dependence of the excitonic lifetime (Fig. 6). At low temperatures both curves exhibit a flat temperature dependence. This is believed to be associated with the exciton localization effect at low temperatures.[33] At higher temperatures however, the PL decay times present a linear temperature dependence for both samples, that is a typical characteristic of free excitons in quantum wells.[28] In QW's, only excitons with small momentum ($\mathbf{k}\approx\mathbf{0}$) can recombine radiatively because of the requirement of momentum conservation. When temperature increases, an increased number of excitons will populate states with large $\mathbf{k}$ which cannot recombine radiatively, leading to an increase of the exciton lifetime. Due to the properties of the 2D density of states in QW's, it is expected that the lifetime of excitons increases linearly with temperature.[28,34] In our cases, localized excitons dominate the luminescence processes at low temperatures. As the temperature increases, localized excitons are thermally-activated to form free excitons (extended states) which have a dispersion in $\mathbf{k}$ space. So the excitonic lifetime increases linearly with temperature in

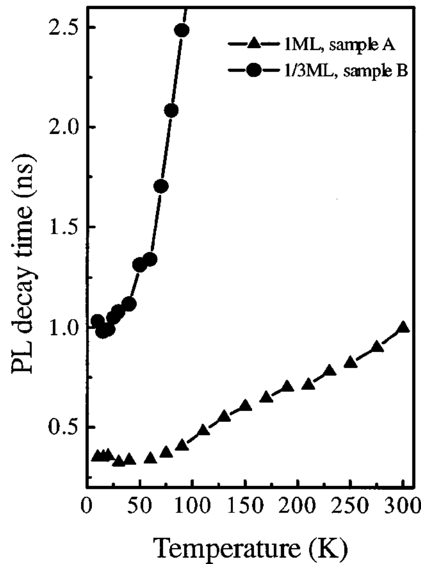

FIG. 6. Temperature dependencies of the PL decay time for samples A and B.

the high-temperature range. We therefore conclude that the InAs SML structure exhibits 2D properties in its exciton dynamics. On the other hand, in the case of a 0D system, there is no dispersion in **k** space, and no **k**-conservation requirement for excitonic radiative recombination. The exciton lifetime in this case would depend only on its coherent length, and have no dependence on temperature, contrary to our observation. We may then safely suggest the InAs insertion in GaAs only changes the center-of-mass motion of the exciton in the growth direction, but does not confine the center-of-mass motion of the exciton along the plane at high temperatures. The SML plane acts just like a QW with great interface roughness, and the 2D free-exciton recombination dominates the recombination processes at high temperatures.

IV. CONCLUSION

In this paper, we examined the optical behavior of SML InAs structures embedded in a GaAs matrix by using PL and TRPL experiments. It was found that the PL linewidth decreases with the average InAs thickness, and can be described in the framework of the quantum-well model. The temperature-dependent linewidth analysis shows that carriers confined in a SML InAs insertion have a large exciton–LO-phonon-scattering coefficient. TRPL measurements revealed that the exciton lifetime increases linearly with temperature. All these results indicate that excitons confined in SML InAs have two-dimensional properties, and the distribution of islandlike InAs in the InAs plane does not prohibit exciton center-of-mass motion in lateral direction.

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