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Pressure behaviour of photoluminescence from InAs submonolayer in GaAs matrix

G H Li, H X Han, K Ding and Z P Wang

National Laboratory for Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, PO Box 912, Beijing 100083, People's Republic of China

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Abstract. We have investigated the dependence on hydrostatic pressure of the photoluminescence of an InAs submonolayer embedded in a GaAs matrix at 15 K and for pressure up to 8 GPa. Strong InAs-related emissions are observed in all three samples at ambient pressure. The temperature dependence of the emission intensity for these peaks can be well characterized by the thermal activation of excitons from the InAs layer to the GaAs matrix. With increasing pressure, the InAs-related peaks shift to higher energies. The pressure coefficients of these peaks are very close to that of the free exciton in bulk GaAs. Some weak peaks observed at pressures above 4.2 GPa are attributed to indirect transitions involving X states in the InAs layer. These results are similar to the pressure behaviour observed in the InAs/GaAs monolayer structures. A group of new lines has been observed in the spectra when pressure is increased beyond 2.5 GPa, which is attributed to the N isoelectronic traps in the GaAs matrix.

1. Introduction

InAs/GaAs heterostructures have recently attracted much attention due to their unusual electronic and optical properties [1, 2]. Epitaxial isomorphic growth of InAs on GaAs can be achieved only up to a few monolayers (ML) due to the very large mismatch in lattice constant of about 6.8% [3]. On the other hand, there is a marked contrast in the electronic structure of the two materials. Thus even a single monolayer of InAs has a strong influence on the optical properties in these heterostructures. A strong InAs-related luminescence has been observed in the InAs/GaAs monolayer structures [4–6] and explained either by a thin quantum well (QW) model [4] or by an isoelectronic impurity model [5, 6]. Recently there have been several reports of highly efficient luminescence from InAs submonolayers deposited in a GaAs matrix [7–10]. Many of them explain their results within the thin QW picture [7–10]. The isoelectronic impurity model has also been used in some cases [7].

The pressure investigation of InAs/GaAs monolayer structures [11] showed that the pressure behaviour of InAs-related emissions is similar to that of GaAs matrix. It implies that the thin QW model is more suitable to the InAs monolayer structure. It is valuable to extend this investigation to the submonolayer regime in order to know whether the thin QW model is still valid when the In coverage is further reduced.

In this work we report the dependence on pressure of the low-temperature luminescence measured in three InAs/GaAs samples with average InAs thickness of 1/3, 1/2 and 1 ML. The measured pressure coefficients of the InAs-related emissions are similar to that of the GaAs matrix in all samples. This implies that even at the submonolayer case, the pressure

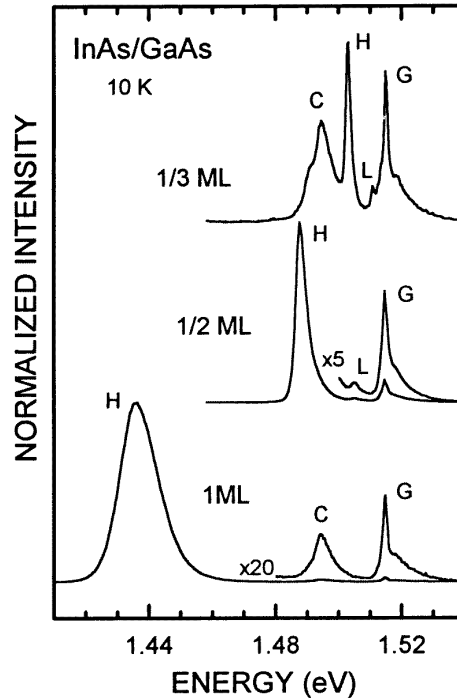


Figure 1. Photoluminescence spectra of InAs/GaAs submonolayer structures with different InAs layer thickness at 10 K and ambient pressure.

behaviour of InAs-related emissions can still be understood within the QW model. A group of new peaks has been observed in these samples when pressure is increased beyond 2.5 GPa. The pressure behaviour of these peaks is like that of a deep level. But further investigation demonstrates that these emissions do not come from the InAs layer. They are related to the N isoelectronic traps in the GaAs matrix.

2. Experimental details

The samples were grown by MBE on semi-insulating (100) GaAs substrates, The InAs submonolayer was grown with a rate of 0.2 ML s^{-1} at 400°C . The thickness of the InAs layer was measured using the RHEED oscillation and calibrated by high-resolution x-ray double crystal diffraction. The details of sample growth have been published elsewhere [12]. The photoluminescence at ambient pressure was performed at 10–300 K by using an APD closed-cycle cryogenic refrigeration system. For the pressure experiments the samples were mechanically thinned to total thickness of about $20 \mu\text{m}$ and then cut into pieces of about $100 \times 100 \mu\text{m}^2$ in size. Photoluminescence under pressure was performed at 15 K using a diamond-anvil cell (DAC) mounted in the cryogenic refrigerator. Condensed argon was used as the pressure-transmitting medium. Pressure was calibrated using the ruby fluorescence method [13].

The luminescence was excited by the 488.0 nm line of an Ar^+ laser with a power density of about $10^2\text{--}10^3 \text{ W cm}^{-2}$. The excited luminescence was analysed by a JY-HRD1 double-grating monochromator equipped with a GaAs photomultiplier operating in the photo-counting mode.

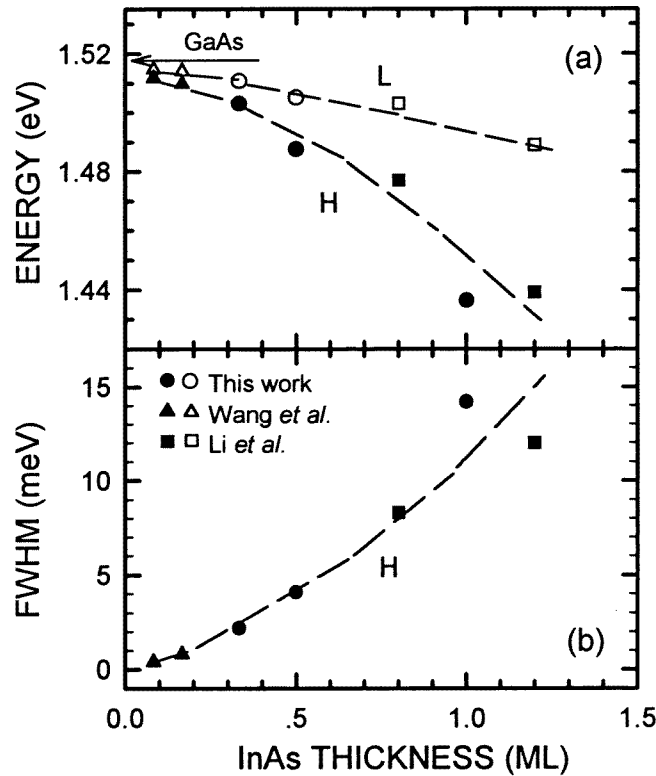


Figure 2. Dependence of (a) peak energies of heavy- (H) and light- (L) hole exciton emissions and (b) full width at half maximum (FWHM) of H emission on InAs layer thickness. The dashed lines are guides to the eye. The arrow indicates the energy position of the free-exciton transition in bulk GaAs.

3. Results and discussion

3.1. Photoluminescence at ambient pressure

Figure 1 shows the 10 K photoluminescence (PL) spectra measured at ambient pressure for three samples with InAs thickness of 1/3, 1/2 and 1 ML. The peaks labelled as G at 1.516 eV are the free-exciton emission lines of bulk GaAs. The peaks at 1.497 eV in the PL spectra of 1/3 and 1 ML samples (labelled as C) are the band-to-carbon-acceptor transition in bulk GaAs. In addition, two InAs-related peaks were observed on the low-energy side of peak G. The dominant peaks (H) are attributed to the radiative recombination of the heavy-hole exciton in the InAs layer. The weak structures (L) in the spectra of 1/3 and 1/2 ML samples correspond to the light-hole exciton transition. The peak C in the 1/2 ML sample was covered by the strong peak H.

In figure 2 we plot the measured peak energy for peaks H and L and the full width at half maximum (FWHM) for peak H as a function of the average thickness of the InAs layer. The results of Wang *et al* [7] and Li *et al* [11] are also shown in the figure. The dashed lines in the figure are guides to the eye. It can be seen that both peaks H and L shift to higher energy and tend to the energy position of the free-exciton transition in bulk GaAs when the InAs thickness is reduced. The FWHM of peak H decreases from 14 meV

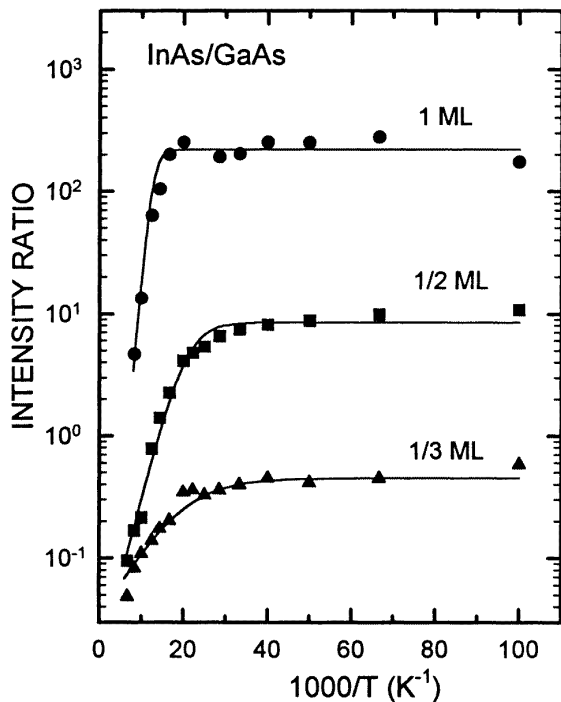


Figure 3. Arrhenius plot of the intensity ratio between InAs (H) and GaAs (G) integrated luminescence intensities for 1/3, 1/2 and 1 ML samples. The solid lines are the calculation results (see text).

for the 1 ML sample to 2.2 meV for the 1/3 ML sample. Yuan *et al* [9] and Tran *et al* [10] have discussed the dependence on InAs thickness of energy position and line width of InAs-related peaks. Both the increase of the peak energies and the decrease of the line widths can be well explained within a two-dimensional quantum well picture. Our results are nearly the same as theirs.

The Arrhenius plot of the intensity ratio between InAs (H) and GaAs (G) integrated luminescence intensities is shown in figure 3. The intensity ratio at 10 K is 0.58, 11 and 170 for 1/3, 1/2 and 1 ML samples, respectively. It has been demonstrated that the large intensity ratio for the InAs/GaAs monolayer structure is due to the strong capture of photogenerated carriers by the InAs layer [11]. The intensity ratio is smaller in the submonolayer samples. This implies that the carrier capture is less effective in the submonolayer structure. As the temperature increases, the intensity ratio decreases exponentially for each sample. It is caused by the thermal activation of excitons in the InAs layer back to the GaAs matrix. The solid lines in the figure are the calculated results using

$$I = \frac{A}{1 + B \exp(-E_B/kT)} \quad (1)$$

where A and B are fitting parameters. The activation energy E_B is taken from the difference of the peak energies of G and H. The agreement between experimental and calculation results is very good.

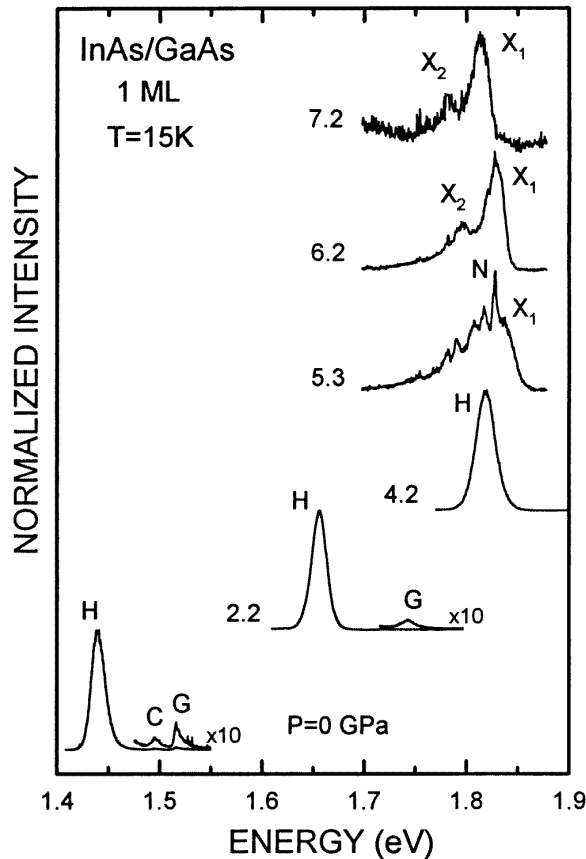


Figure 4. Low-temperature PL spectra of 1 ML InAs/GaAs samples at several pressures.

3.2. Pressure dependence

The PL spectra of the 1 ML sample under several pressures are shown in figure 4. In the pressure range below about 4.2 GPa, all the features in the PL spectra shift to higher energies with increasing pressure. Above 4.2 GPa the intensity of peak H drops dramatically and two new emission lines (labelled as X_1 and X_2) appear. They shift to lower energies with increasing pressure. The situation is very similar to that of the InAs/GaAs monolayer structures in [11]. Similarly, we also attribute these peaks to the indirect optical transitions involving X states in the InAs layer. A group of lines labelled as N in the spectra at 5.3 GPa will be discussed later.

Figure 5 shows the PL spectra of 1/3 and 1/2 ML samples at different pressures. The spectra for $P = 0$ GPa are nearly the same as shown in figure 1, except for the lack of peak L due to the lower signal-to-noise ratio in the DAC. The peaks shift to higher energies with a similar rate as in the case of the 1 ML sample. A group of new peaks (labelled as N) appears when pressure is increased beyond 2.5 GPa. They also have a blue shift but at lower rate. We attribute these peaks to the radiative recombination of excitons bound to the nitrogen isoelectronic traps in the GaAs matrix. The reason will be discussed later. Above about 4.2 GPa the intensity of peaks G and H decreases dramatically and the N

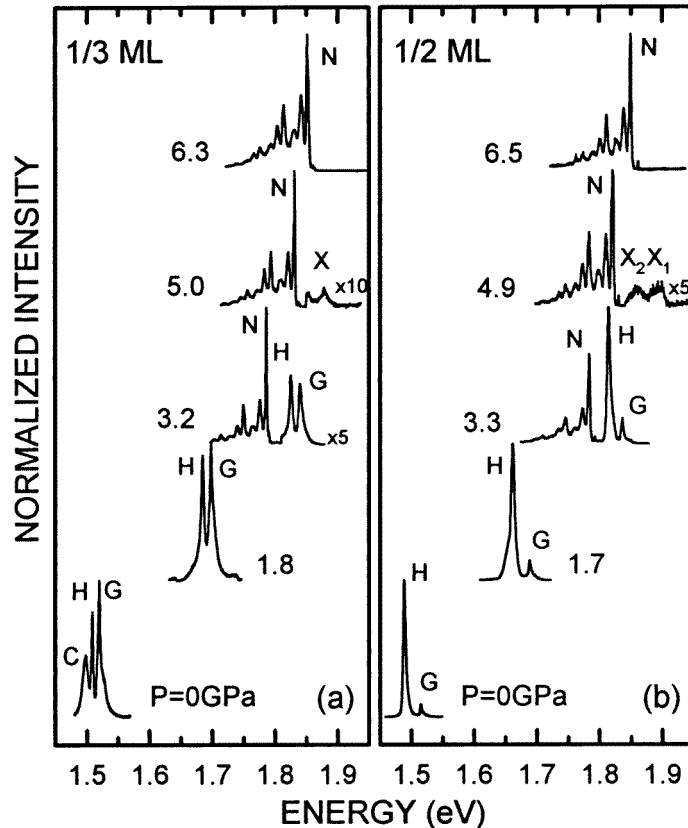


Figure 5. Low-temperature PL spectra of (a) 1/3 ML and (b) 1/2 ML InAs/GaAs submonolayer structures under several pressures.

lines become the dominant structure. The weak X-related peaks (X for the 1/3 ML sample and X_1 , X_2 for the 1/2 ML sample) are also observed but only in the pressure range of 4.2–5.5 GPa. Then they are covered by the stronger N lines.

In figure 6 we summarize the results for the pressure dependence of the PL peak energies for 1/3, 1/2 and 1 ML samples. The solid lines represent the results of least-squares fits to the experimental data using quadratic or linear relations. The corresponding first- and second-order pressure coefficients for peaks G and H are listed in table 1. The dotted line in figure 6 is the pressure dependence of the indirect transition between the X valley and the valence band in bulk GaAs [11].

It can be seen from figure 6 and table 1 that the pressure coefficients of the InAs-related emissions (peaks H) are similar to that of the free-exciton emission line in the GaAs matrix (peak G). Moreover, the pressure coefficient of peaks H is closer to that of peak G when the thickness of the InAs layer is decreased. These results can be explained within the thin QW model. In a QW structure, the pressure coefficient of the emission from the QW is between that of the well and barrier materials and tends to that of the barrier material when the well width is reduced [14].

The pressure investigation of the InAs/GaAs monolayer [11] has indicated that the strain in the InAs layer splits the X valleys of InAs into X_Z and X_{XY} states with X_Z higher than

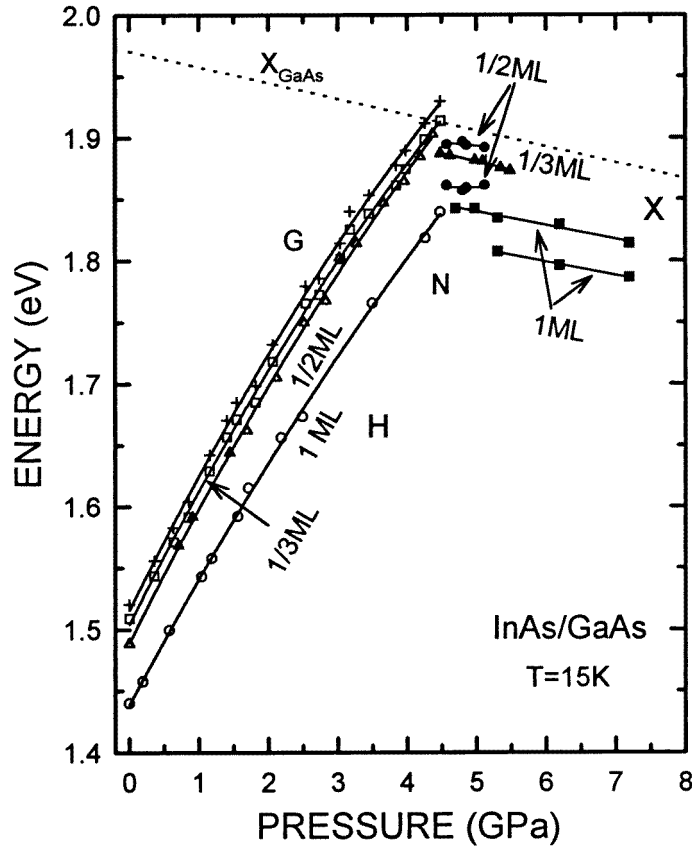


Figure 6. Pressure dependence of the energy position of several PL peaks for 1/3, 1/2 and 1 ML samples. The solid lines represent the results of least-squares fits to the experimental data. The dotted line is the pressure dependence of the indirect optical transition from the X valley to the valence band in bulk GaAs [11].

Table 1. Pressure coefficients of several PL peaks of the InAs/GaAs submonolayer structures obtained from least-square fits to the experimental data using $E(P) = E(0) + a_1P + a_2P^2$.

| Sample | Peak | $E(0)$ (eV) | a_1 (meV GPa ⁻¹) | a_2 (meV GPa ⁻²) |
|-------------|------|----------------|-----------------------------------|-----------------------------------|
| 1 ML | H | 1.439(7) | 106(5) | -3.8(7) |
| 1/2 ML | H | 1.489(7) | 113(5) | -4.3(7) |
| 1/3 ML | H | 1.504(7) | 113(5) | -4.9(7) |
| GaAs matrix | G | 1.516(7) | 114(5) | -5.0(7) |

X_{XY} , and the X_{XY} level falls below the energy of the X minimum in GaAs. Our results for the 1 ML sample are similar to those of [11]. Then we also attribute the peak X_1 to the type-II transition from X states in GaAs to InAs heavy-hole states, and X_2 to the type-I transition between conduction-band X_{XY} and heavy-hole states of the InAs monolayer. In the submonolayer case, the energy position for X-related peaks is higher than that of the 1 ML sample. In the 1/2 ML sample, we still observe two X-related peaks. But only

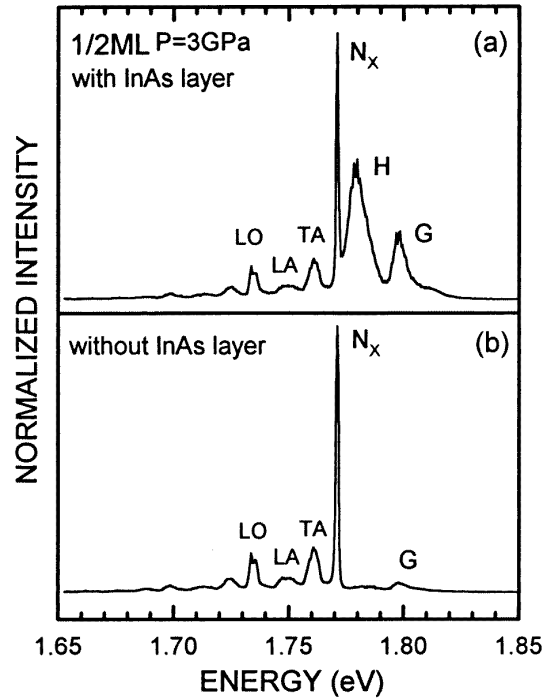


Figure 7. PL spectra measured on the areas (a) with InAs layer and (b) without InAs layer of a piece of the 1/2 ML sample in the pressure cell at 3 GPa and 15 K.

one X-related peak is observed in the 1/3 ML sample. Perhaps the peak corresponding to the type-II transition is missing in this sample due to the interface roughness. Since the X-related peaks are very weak in the submonolayer structure and only be observed in a smaller pressure range, we do not discuss these peaks in more detail.

3.3. Nitrogen-related lines

A group of new emission lines (N) appears in the PL spectra of 1/2 and 1/3 ML samples when pressure is increased beyond about 2.5 GPa. These peaks become the dominant structure in the spectra at pressures above 4.2 GPa. Similar lines are also observed in the spectra of the 1 ML sample but only at a few pressures since the luminescence intensity of the InAs-related emission is much stronger. The spectrum at 5.3 GPa in figure 4 is an example for observed N lines in the 1 ML sample. The structure of N lines is similar to the N_X lines in the N-doped GaAs [15], consisting of a main zero-phonon peak (N_X) and various phonon replicas as shown in figure 7. Its pressure behaviour is also similar to that of N isoelectronic traps reported in [15]. In figure 8 we summarize the measured peak energies of N_X for three samples as a function of pressure. The pressure dependence of nitrogen isoelectronic traps in GaAs [15] is also plotted in the figure as a dashed line. It can be seen that the agreement between our experimental data and the results of [15] is very good. This suggests that the N emission lines observed in our samples do not come from the InAs layer but from the N isoelectronic traps in the GaAs matrix. To confirm this conclusion we have prepared a piece of sample in which a part of the InAs layer has been

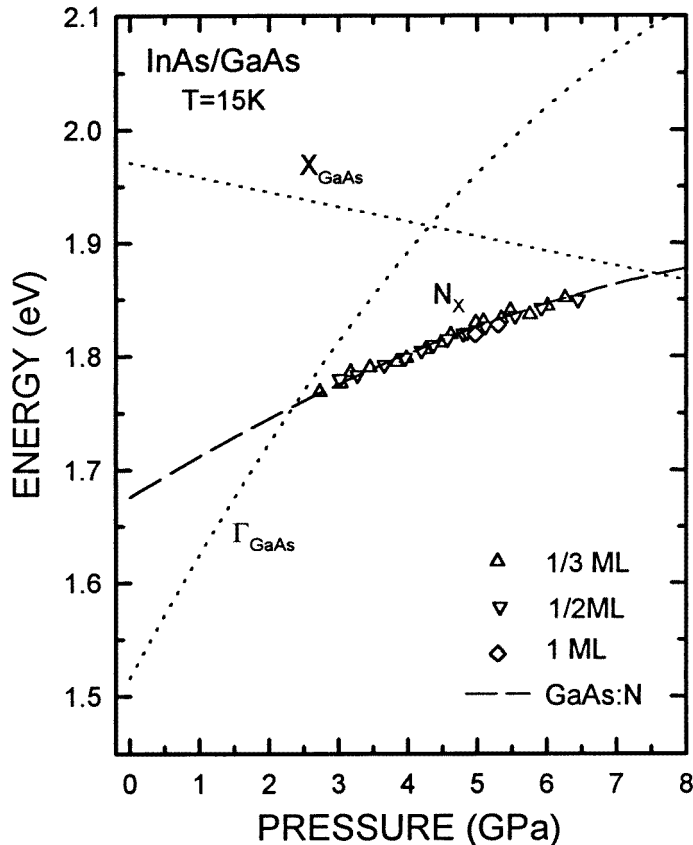


Figure 8. Pressure dependence of the energy position of peak N_X for 1/3, 1/2 and 1 ML samples. The dotted lines are the pressure dependence of Γ and X band gaps of bulk GaAs. The dashed line is taken from [15].

removed. The spectra measured on the areas with and without an InAs layer are presented in figure 7(a) and (b) respectively. Only peak H does not appear in the spectra measured on the area without the InAs layer. This demonstrates that only the H line comes from the InAs layer; both G and N lines come from the GaAs matrix. Leroux *et al* [16] have also observed similar N line structures in their MBE-grown GaAs samples. They proposed that the weak decomposition of boron nitride crucibles at high temperature may be the reason for the nitrogen doping in GaAs. It may be a similar situation in our samples.

4. Summary

We have investigated the dependence on pressure up to 8 GPa of the low-temperature photoluminescence emitted from InAs/GaAs submonolayer structures. The strong InAs-related emissions are observed in all three samples at ambient pressure. When the average thickness of the InAs layer is reduced, these peaks shift to higher energy. Meanwhile, their line width and relative intensity decrease. As the temperature increases, the intensity ratio between InAs- and GaAs-related emissions decreases exponentially due to the thermal

activation of excitons from the InAs layer to the GaAs matrix. Under pressure, the InAs-related peaks shift to higher energies with a rate similar to that of the free-exciton transition in bulk GaAs. Moreover, the pressure coefficients are closer to that of bulk GaAs for submonolayer samples. This can be well explained within the thin QW model. When pressure is increased beyond 2.5 GPa, a group of new lines appears in the spectra. We attribute these lines to the radiative recombination of excitons bound to the N isoelectronic traps in the GaAs matrix according to their line structure and pressure behaviour. Above 4.2 GPa, the intensity of Γ -related emissions drops dramatically and some weak X-related emissions appear, indicating that the structures undergo a transition from a direct to an indirect band material.

Acknowledgments

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