Photoluminescence from strained InAs monolayers in GaAs under pressure

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We have investigated the dependence on hydrostatic pressure of the photoluminescence of InAs monolayers embedded in bulklike GaAs at 10 K and for pressures up to 8 GPa. Below the pressure-induced Γ-X conduction-band crossover in GaAs (4.2 GPa) the optical emission is dominated by direct optical transitions between Γ-like excitonic states bound to the InAs monolayer. With increasing pressure this luminescence band shows a blueshift similar to the lowest direct band gap of bulk GaAs. At pressures above the band crossover two emission bands are observed. These bands, characterized by having negative pressure coefficients, are attributed to the type-I transition between conduction-band X_{xy} and heavy-hole states of the InAs monolayer and the type-II transition from X states in GaAs to InAs heavy-hole states. The results are interpreted in terms of tight-binding band-structure calculations for the strained InAs-monolayer–bulk-GaAs system.

I. INTRODUCTION

Highly strained InAs/GaAs heterostructures have recently attracted interest due to their unusual electronic and optical properties.[1–4] Epitaxial isomorphic growth of InAs on GaAs can be achieved only up to a small critical thickness of 2–3 monolayers (ML) due to the very large mismatch in lattice constant of about 6.8%.[5,6] There is a marked contrast in the electronic structure of the two materials: the lowest direct band gap of InAs (0.42 eV at 10 K) is about 1.1 eV smaller than that of GaAs.[7] Thus, in spite of its small thickness, even a single monolayer of InAs has a strong influence on the optical properties in these heterostructures. As established recently in a series of optical studies of such structures at ambient pressure,[2,4,8] the InAs monolayer acts as an attractive potential for electrons and holes. This gives rise to the formation of excitons localized at the two-dimensional sheet of In atoms, resulting in a redshift of the dominant photoluminescence (PL) band relative to that of bulk GaAs.

A well established change is induced in the electronic properties of the GaAs host material under hydrostatic pressure. The energy of the direct Γ point band gap increases with pressure while the energy of the X-like minimum is lowered, leading to a direct to indirect band gap crossover at about 4.2 GPa.[9] As in the case of bulk GaAs,[10] striking changes in the character and intensity of the optical emission are expected to occur under pressure in the GaAs/InAs monolayer system. An interesting aspect of technological importance is whether such structures show enhanced radiative recombination efficiency in the indirect case due to the breakdown of translational symmetry caused by the monolayer potential. A detailed understanding of the electronic structure at and above the crossover pressure can be obtained from a study of the excitonic luminescence for pressures covering the transition regime.

In this work we report the dependence on pressure of the low-temperature luminescence measured in two GaAs/InAs samples with different average InAs monolayer thicknesses. Below the Γ-X crossover pressure these samples show a strong PL band redshifted from the GaAs emission, which follows the change of the GaAs band gap under pressure. In the indirect regime, two emission bands are observed in the PL spectra, which are characteristic of the monolayer samples. Based on the pressure dependence of their energies and on the pressure and temperature dependence of their intensities in the crossover regime, we associate these emission bands with indirect optical transitions involving different X-like conduction-band states and the heavy-hole state of the InAs monolayer. In order to support the interpretation of the experimental results we have calculated the electronic structure over the entire Brillouin zone for different pressures using the sp^3s^* tight-binding method.[11] In this way we have obtained the energies and the wave functions of the band states which participate in the optical recombination processes, thus gaining further insight into the electronic properties of these monolayer structures.

II. EXPERIMENTAL DETAILS

The samples were grown by conventional molecular-beam epitaxy (MBE) on semi-insulating (100) GaAs substrates as described elsewhere.[8] Sample A consists of a

single InAs monolayer capped with 200 nm GaAs. Sample B consists of ten separate InAs monolayers sandwiched between 30-nm-thick GaAs layers. The average InAs layer thicknesses obtained from high-resolution double-crystal x-ray diffraction data are 3.9 Å and 2.6 Å (corresponding to 1.2 and 0.8 ML) for sample A and sample B, respectively.[8,12] For the pressure experiments the samples were mechanically thinned to total thicknesses of about 20 μm and then cut into pieces of about 100×100 μm^2 in size. Photoluminescence and transmission measurements were performed at 10 K using a diamond-anvil cell (DAC) to generate pressures up to 8 GPa. Condensed helium was used as the pressure-transmitting medium. Pressure was always changed at room temperature in order to ensure the best possible hydrostatic conditions. At low temperatures the pressure was measured *in situ* using the ruby fluorescence method[13] and temperature correction of the pressure calibration according to Ref. 14.

The luminescence was excited by the 514.5 nm line of an Ar^+ laser with a power density of about 20 W/cm^2. For transmission measurements in the spectral range below the GaAs band gap, white light from a tungsten lamp was focused onto the sample forming a spot of about 35 μm in diameter. Both the excited luminescence and the transmitted light were analyzed by a 0.6 m single-grating spectrometer equipped with a GaAs photomultiplier operating in the photon-counting mode. Transmission spectra were normalized using as a reference the intensity of light passing through the pressure cell next to the sample.

III. RESULTS AND DISCUSSION

A. Optical transition energies

Figure 1 shows the low-temperature PL spectra measured at ambient pressure for samples A (single InAs ML, 1.2 ML thickness) and B (10 ML of 0.8 ML thickness) together with the transmission spectrum of sample B. In the case of sample A the absorption was too weak to be measured. The dominant peak in the PL spectra of both samples (labeled as M) is attributed to the radiative recombination of the heavy-hole exciton in the InAs monolayer. These peaks are redshifted with respect to the free-exciton emission line (FE) of bulk GaAs at 1.516 eV. The redshift is larger for sample A which has a larger monolayer thickness. The transmission spectra of sample B display two weak absorption bands below the direct gap of GaAs. The one at lower energy and coincident with the M peak in emission corresponds to transitions between conduction-band and heavy-hole (HH) states in the InAs layer. The other absorption band is attributed to the light-hole (LH) counterpart. This assignment is discussed below in the framework of a tight-binding calculation.

The broad PL bands labeled as C and C' at about 20 meV below the FE and M peaks have been attributed[15] to band-to-carbon-acceptor transitions in bulk GaAs and to transitions involving carbon impurities near the InAs layer, respectively. The extrinsic origin of these bands

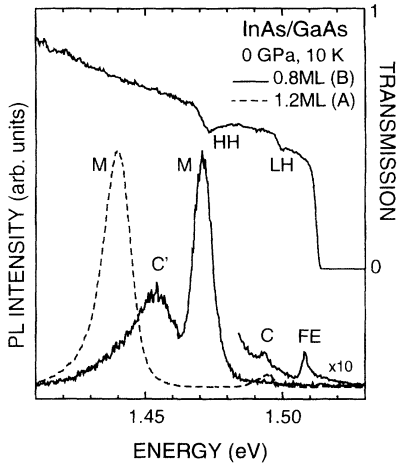

FIG. 1. Photoluminescence and transmission spectra (solid lines) of the 0.8 monolayer InAs/GaAs sample at 10 K and ambient pressure. The dashed line represents the PL spectrum for the 1.2 monolayer sample.

is revealed in their dependence on incident laser power. Both bands saturate in intensity at higher excitations levels. Also consistent with this interpretation is the fact that these transitions do not show up in transmission spectra because at low temperature most of the impurities are neutralized.

In the pressure range below the Γ-X crossover in GaAs, all the features in PL and transmission spectra shift to higher energies with increasing pressure, as shown for sample B in Fig. 2(a). Similar results were obtained for sample A. Above the crossover pressure two new emission lines (labeled M_1 and M_2) appear [see Fig 2(b)]. They shift to lower energies at a rate similar to that of the Γ-X indirect gap of GaAs. It is shown below that both emission lines involve states of the InAs monolayer.

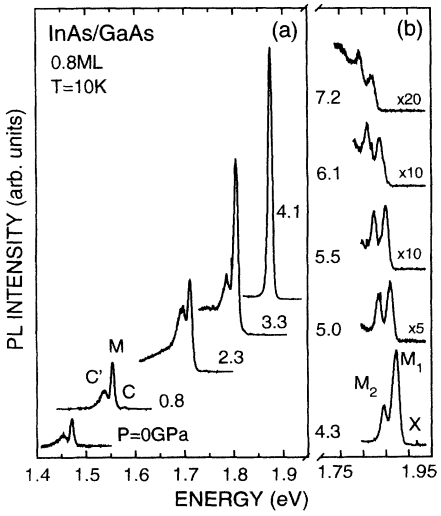

FIG. 2. PL spectra of the 0.8 monolayer sample for different pressures (a) below and (b) above the Γ-X conduction-band crossover pressure of 4.2 GPa.

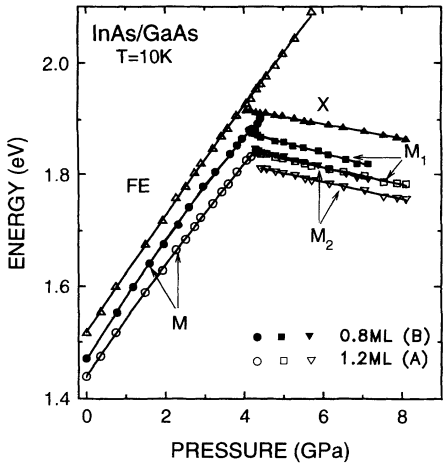

FIG. 3. Pressure dependence of the energy position of several PL peaks of the 0.8 and 1.2 monolayer samples.

In Fig. 3 we summarize the results for the pressure dependence of the PL-peak energies for both samples. The solid lines represent the results of least-squares fits to the experimental data using quadratic and linear relations. The corresponding first- and second-order pressure coefficients are listed in Table I. Within experimental uncertainty these coefficients agree with those measured for bulk GaAs,[9,10,16] with the exception of the M_1 and M_2 lines of sample B which display a somewhat stronger negative pressure shift compared to the indirect gap of GaAs.

Table II shows the energy separation between the various features observed in the PL spectra. By comparing the results of the two measured samples we notice a dependence on monolayer thickness of the InAs-related emission lines. In the 1.2 ML sample the redshift of the M peak with respect to the direct free-exciton line FE of GaAs is almost twice as large as for the 0.8 ML sample. In the indirect case a similar difference relative to the GaAs indirect transition is observed. On the other hand, the energy difference between the M_1 and M_2 transitions is nearly the same for both samples (28±2 meV for sample A and 27±2 meV for sample B) even though the absolute peak energies are significantly different.

TABLE I. Coefficients describing the dependence of the PL peaks of the InAs/GaAs monolayer structure on pressure obtained from least-square fits to the experimental data using $E(P) = E(0) + a_1 \times P + a_2 \times P^2$.

Peak		$E(0)$ (eV)	a_1 (meV/GPa)	a_2 (meV/GPa^2)
1.2 ML	M	1.439(2)	104(2)	-2.2(5)
0.8 ML	M	1.477(2)	104(2)	-1.4(5)
GaAs	FE	1.518(2)	108(2)	-1.7(5)
1.2 ML	M_1	1.906(2)	-15(1)	
1.2 ML	M_2	1.878(2)	-15(1)	
0.8 ML	M_1	1.948(2)	-18(1)	
0.8 ML	M_2	1.920(2)	-18(1)	
GaAs	X	1.971(2)	-13(1)	

TABLE II. Measured and calculated energy differences between optical transitions in the GaAs/InAs monolayer system at ambient pressure (direct gap case) and for pressures above 4.2 GPa (indirect gap case).

	Experiment 0.8 ML	Experiment 1.2 ML	Theory 1 ML
FE-M	43±1	77±1	93±8
LH-HH	26±1	44[a]	39±4
X-M_1	27±2	61±2	
M_1-M_2	27±2	28±2	
M_1-M_2-LO^X	2±2	3±2	8±3

[a] Reference 4.

B. Photoluminescence intensity

During the excitation of the PL, minority carriers are photogenerated in a spatial region defined by the surface and the penetration depth of light (~ 0.5 μm). In spite of the large volume ratio, the PL intensity of the FE line of GaAs is only a small fraction ($\approx 1/50$) of that of line M which corresponds to emission from the InAs monolayer region (see Fig. 1). This means that the characteristic radiative recombination lifetime $\tau_{\rm rec}$ of the carriers is much longer than the time $\tau_{\rm cap}$ needed by the carriers to reach the InAs monolayer and get captured there. Neglecting reabsorption of the emitted luminescence in the InAs we have for the intensity ratio

$$\frac{I_M(\text{InAs})}{I_{\rm FE}(\text{GaAs})} = \frac{\tau_{\rm rec}(\text{GaAs})}{\tau_{\rm cap}(\text{InAs})} . \quad (1)$$

Considering a typical recombination time of ~1 ns in very pure GaAs,[17] we estimate a capture time of about 20 ps. This yields a drift velocity of 2.5×10^6 cm/s for carriers in the GaAs layers of our samples, in good agreement with literature data.[18]

When pressure is increased beyond 3 GPa, the intensities of the GaAs-related peaks (FE, C) begin to decrease. At about 4 GPa a weak emission band related to indirect optical transitions from the X point in GaAs (labeled as X) appears, which is the indication of the Γ-X crossover[10] (see Fig. 3). The intensity of peak M, on the contrary, increases continuously up to the crossover pressure while the C' peak shows a similar decrease in intensity as peak C. This means that peak C' and peak M may have a different carrier supply. Brandt *et al.*[15] arrived at the same conclusion from results of their PL-excitation experiments.

Figure 4 shows the variation with pressure of the intensity of all InAs-related peaks. Above 4.2 GPa where the M_1 and M_2 peaks appear, the intensity of peak M drops dramatically by three orders of magnitude. At the crossing point peak M_1 is as strong as the M peak. Its intensity then decreases rapidly within about 0.3 GPa. Above 4.5 GPa both the M_1 and M_2 peaks have nearly the same intensities and the same decreasing tendency with further increase of the pressure. The continuous intensity transfer from line M to line M_1 in the pressure range of the crossover is an indication of mixing ef-

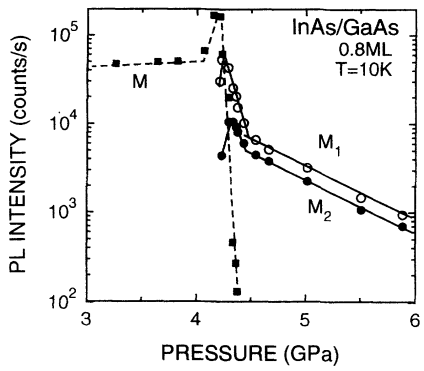

FIG. 4. Peak intensity of the InAs-related emission lines of the 0.8 monolayer sample as a function of pressure.

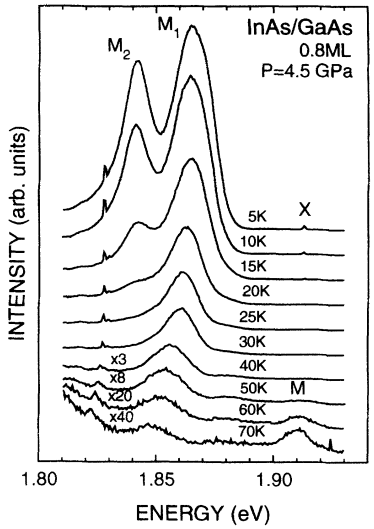

FIG. 5. Luminescence spectra of the 0.8 monolayer sample at a constant pressure of 4.5 GPa and different temperatures.

fects between X- and Γ-conduction-band states induced by the potential step at the monolayer. An anticrossing behavior of transition energies due to Γ-X mixing was previously reported for a GaAs/AlAs superlattice in an axial electric field.[19] In our case the anticrossing is difficult to observe because of the small interaction potential (estimated to be of the order of ~ 1 meV). This effect is enhanced by additional confinement of the wave functions such as in a system of InAs quantum dots which displays a well-resolved pressure-induced anticrossing behavior.[20]

Another possible effect of a Γ-X mixing is the enhancement of the oscillator strength for indirect optical transitions compared to the weak indirect recombination in bulk materials. Translational symmetry in the growth direction is broken by the monolayer potential. Thus, optical transitions which are indirect in **k** space in the bulk material acquire some admixture of direct character.[21] For the InAs monolayer the PL intensity in the indirect case well above the crossover pressure (lines M_1 and M_2) is still about two orders of magnitude lower compared to the intensity maximum of the direct emission band M (see Fig. 4). A similar ratio is obtained for bulk GaAs under pressure,[10] indicating that the perturbing effect of a monolayer on the PL oscillator strength is not significant in the indirect case.

We have also measured the temperature dependence of the PL spectra under various constant pressures. The most striking behavior is displayed by the lines M_1 and M_2 in the vicinity of the band crossover. Figure 5 shows several PL spectra measured at 4.5 GPa and for different temperatures ranging from 5 to 70 K. Surprisingly, the lowest-energy peak M_2 is quenched at about 25 K. This temperature dependence, which is consistent with a very small thermal activation energy of 2–3 meV, speaks against line M_2 being of extrinsic origin. For comparison, the luminescence intensity from carbon acceptor states in GaAs vanishes only above 100 K. We also find that the direct transition of the InAs monolayer (line M) picks up intensity with increasing temperature. In the spectra of Fig. 5 it is clearly observed at temperatures around 70 K.

The experimental results show that the M_1 and M_2 transitions are related to electronic states originating from InAs monolayers. A convincing assignment of these transitions is, however, not straightforward, because in contrast to the direct case the X-like conduction-band energy levels undergo a strain-induced splitting. For an unambiguous interpretation of the optical transitions observed in the indirect case, it is therefore necessary to calculate quantitatively the contributions of both confinement and strain on the energy levels for the InAs monolayer in GaAs as discussed in the next section.

C. Tight-binding calculations

In previous reports[4,8] the identification of the transitions seen in PL and PL excitation spectra was made on the basis of calculations within the envelope-function model. Its applicability to the case of a monolayer is conceptually questionable. Furthermore, it yields the electronic levels only in the vicinity of the Γ point, whereas in the present case the most relevant results are obtained in the pressure range above the Γ-X crossover. We have therefore calculated the electronic structure of the monolayer system using the tight-binding (TB) approach. The TB method provides a real space picture of the electronic interactions which give rise to the band structure. Moreover, using only a few interaction parameters it is possible to obtain the energy bands and the electronic wave functions in the entire Brillouin zone.

Here we have employed an empirical TB method with an sp^3s^* orbital basis[11] including first-nearest-neighbor interactions, which is known to yield sufficiently accurate conduction bands as compared to more sophisticated *ab initio* band-structure calculations.[22] In InAs the spin-orbit splitting energy Δ_0 of the valence band is comparable to the direct band at Γ (0.418 eV at 10 K). Thus, spin-orbit interactions are important and have to be incorporated into the calculations by including relativistic terms in the Hamiltonian and by an appropriate choice of interaction parameters.[23] The effect of hydrostatic pres-

sure is introduced in the theory by accounting for the dependence of the interaction parameters on interatomic distance. The scaling rule of the nondiagonal parameters is of the form

$$V_{ll'}(d) = V_{ll'}(d_0)\left(\frac{d}{d_0}\right)^{n_{ll'}}, \quad (2)$$

where d_0 (d) is the unstrained (strained) interatomic distance and the exponent $n_{ll'}$ depends on the pair of orbitals (l,l') involved.[24] We have adjusted the different exponents separately for bulk InAs and GaAs to reproduce the experimental pressure dependence of the direct gap at the Γ point and both indirect Γ-X and Γ-L gaps.[7]

The (001) growth direction of our particular structures is hereafter denoted as z. Under isomorphic growth conditions the InAs monolayer is subjected to a strong biaxial compression in the xy plane due to its large lattice mismatch to GaAs. Associated with the distortion is a shift of the energy levels according to the hydrostatic component of the biaxial strain and an additional energy splitting of the p orbitals which is proportional to the built-in strain and the shear deformation potential.[25] Because of the large p character, the valence band at Γ and the X and L points of the conduction band are the ones being primarily affected by the anisotropic strain. This results in an additional splitting between light- and heavy-hole bands. In order to get the correct X_{xy}-X_z splitting of the conduction band it is essential to modify the strain dependence of the s^*-p interaction, as was pointed out by Muñoz and Armelles[26]. Following the scheme of Ref. 26, we find that the X_{xy} states in InAs are pushed down by shear strain below the energy of the X minimum of GaAs whereas the InAs X_z zone-edge states shift upwards by about 0.5 eV.

In calculating the band structure we have constructed a TB supercell consisting of one strained layer of InAs and 28 layers of bulklike GaAs. For the matching of the energy bands of both materials we have used a valence-band offset of 0.25 eV as determined recently for the *strained* InAs/GaAs interface from the core-level shifts in x-ray photoemission experiments.[3] This value, which corresponds to the energy difference between the *centroids* of the valence-band maxima in the two materials, was added to the diagonal elements of the TB Hamiltonian for InAs. The resulting band alignment is of type I. In this way we have calculated the electronic subbands of the monolayer structure along the Γ-X and Γ-L directions and obtained both the energies and wave functions at certain $\mathbf{k}$ points of interest in the Brillouin zone. A more detailed account of the results obtained from the theoretical work will be given elsewhere.[27] For our present purpose it is sufficient to discuss the two limiting cases, namely, the level ordering of Γ states in the direct gap situation and in particular that of the X-like conduction-band states for the indirect gap situation.

Figure 6 is a schematic representation of the results obtained for the direct case corresponding to pressures below 4.2 GPa. The Γ point band edges along z are indicated for the conduction and valence bands. Thick horizontal lines represent the energy levels calculated for ambient pressure. We also show for each level the squared

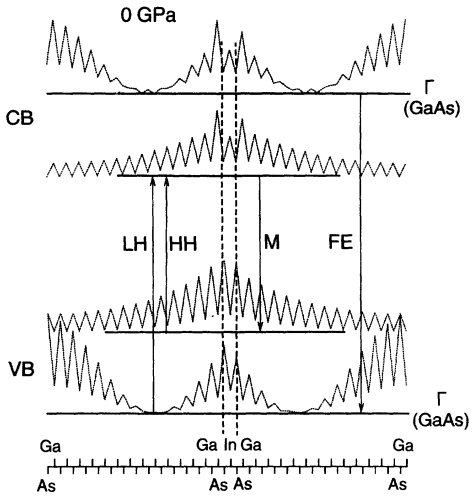

FIG. 6. Sketch of the Γ point conduction- and valence-band profile of the monolayer structure. The energy levels and the square moduli of the wave functions represent the results of tight-binding calculations (see text) for ambient pressure. Arrows indicate the assignment of observed optical transitions.

modulus of the corresponding wave function. These results clearly indicate the formation of electron and hole states bound to the InAs monolayer. For the lowest state of the conduction band the wave function is mainly localized at the monolayer, though it penetrates up to ten monolayers into the GaAs. The first excited state, on the contrary, is very much extended in GaAs with the same probability of finding an electron close to or far from the InAs layer. It is important to note that a spectral decomposition of the wave function into the orbital basis states shows that both conduction-band states have mainly s character. In the valence band there are two nearly degenerate levels with wave functions concentrated at the monolayer (for clarity we show in Fig. 6 only one wave function). These states originate from the heavy-hole bands whereas the light holes are completely delocalized.

The vertical arrows in Fig. 6 indicate the optical transitions observed in PL and absorption. The assignment is fully supported by the TB calculation. The calculated energy differences between InAs-like and GaAs-like states are compared to the measured ones in Table II. We find a good quantitative agreement between theory and experiment if one takes into account the difference in monolayer width and the fact that excitonic effects have been neglected in the calculations.

Except for the change in band gap energy, the situation represented in Fig. 6 holds in the pressure range up to the Γ-X crossover. The similarity of the linear pressure coefficients of the M and FE emission lines is explained by the small localization energies of electrons and holes and the large penetration of the wave functions in the GaAs barrier.

Results of the TB calculations for the indirect case at 8

GPa are displayed in Fig. 7. The relative energy position of the levels remains unaltered in the pressure range from 4 to 8 GPa. The most pronounced changes relative to the direct gap case (Fig. 6) occur in the conduction band. As discussed above, the built-in strain splits the X valleys of InAs and the X_{xy} levels fall below the energy of the X minimum in GaAs. The lowest conduction-band state, which has evolved from the X_{xy} levels of the InAs, is strongly localized at the monolayer. The next higher energy level, which belongs to the bulk GaAs, appears at only 8 meV higher energy (see Table II).

Based on these calculations it is now possible to interpret the PL spectra measured for the indirect case. We attribute the high-energy peak M_1 in the PL spectra to the type-II transition (see Fig. 7) from the X valley of GaAs to the heavy hole in the InAs layer. Its intensity exhibits a coupling behavior with peak M. Its energy separation from the indirect transition at X in GaAs is very close to the heavy-hole localization energy in the InAs monolayer.

The lower-energy peak M_2 is assigned to the type-I transition from the X_{xy} conduction valleys to heavy-hole states in InAs. The calculations predict, however, a smaller energy difference between the two indirect transitions than observed. The larger experimental energy difference of 28 meV between M_1 and M_2, which is nearly the same in both samples, can be understood if one takes into account that only phonon-assisted transitions are allowed for indirect recombination processes from X_{xy} to heavy-hole states. This is because the monolayer potential cannot couple in-plane X_{xy} states with that at Γ. The opposite is true for the type-II transition M_1, which corresponds to the zero-phonon line activated by interface scattering at the monolayer. The LO-phonon energy at the X zone edge of InAs is 25 meV.[7] We can therefore infer from the measured energies of the PL peaks that the X_{xy} level of the InAs layer is just 2–3 meV below the X minimum of GaAs, in good agreement with the theoretical results.

The small energy difference also explains the temperature dependence of the M_2 intensity. With increasing temperature electrons localized at the monolayer are thermally activated over the small barrier before they recombine. This gives rise to the quenching of the intensity of the M_2 emission line at relatively low temperatures. At somewhat higher temperatures the probability increases for carriers to occupy states at the higher-lying Γ minimum, and therefore direct emission picks up intensity.

Finally, we point out that, in contrast to the indirect case, below the crossover pressure we do not observe any emission band corresponding to type-II transitions between Γ states of GaAs and the InAs heavy holes which are *direct in k space*. This speaks for very short capture times into the InAs monolayer for Γ electrons in GaAs as compared to electrons in the X valleys. Conduction band electrons at Γ which are close to the InAs monolayer and which have an appreciable overlap with the heavy-hole wave function are captured before they recombine radiatively. The capture process appears to be far less effective for electrons in the X minima of GaAs. This is partly due to the large difference in electron effective masses of the Γ and X minima. Possibly the fact that the lowest conduction-band state which is strongly localized at the InAs monolayer (see Fig. 7) originates from strain-split X_{xy} levels also plays a role, since the capture of electrons from X_{xy} states of GaAs requires an additional scattering process.

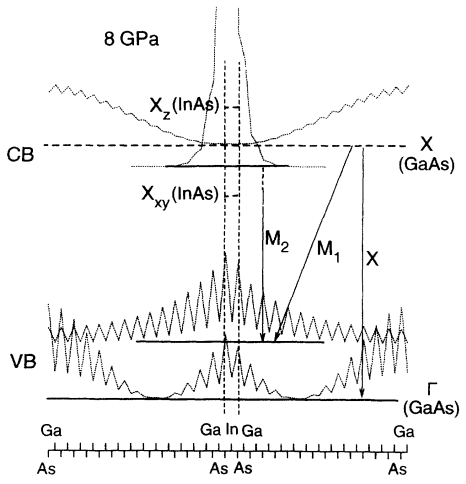

FIG. 7. X point conduction-band and Γ point valence-band profile above the band crossover pressure. The X-conduction-band minima of InAs split by the built-in biaxial strain are indicated schematically by dashed lines. The energy levels and the square moduli of the wave functions represent the results of tight-binding calculations (see text). Arrows indicate the assignment of the observed optical transitions.

IV. SUMMARY

We have investigated the dependence on pressure up to 8 GPa of the low-temperature photoluminescence emitted from monolayers of InAs embedded in bulk GaAs. At 4.2 GPa the structure undergoes a transition from a direct to an indirect band material with corresponding changes in optical emission spectra. For all pressures in the range of the present experiments the dominant features in the PL spectra are related to the InAs monolayer. Effectively, it acts as a sink for electrons and holes which then recombine radiatively, producing PL lines which are redshifted with respect to the fundamental band gap of GaAs. Of particular interest is the observation of *two* PL lines in the indirect gap case. The lowest energy emission corresponds to the type-I phonon-assisted transition between conduction-band X states and heavy holes at the InAs layer. The intensity of this line is quenched at 25 K due to thermally activated carrier delocalization. The second PL line is related to zero-phonon type-II transitions from the X-conduction-band minimum of GaAs to heavy-hole states in InAs. There is no counterpart to the type-II transition in the direct gap situation.

The interpretation of the experimental observations is supported by the results of tight-binding band-structure calculations. In this way we have obtained experimental evidence for the strong influence of InAs monolayers on the optical and electronic properties of bulk GaAs for the direct as well as the indirect gap case.

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