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High-pressure study of optical transitions in strained $In_{0.2}Ga_{0.8}As/GaAs$ multiple quantum wells

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We have measured low-temperature photoluminescence (PL) and absorption spectra of $In_{0.2}Ga_{0.8}As/GaAs$ multiple quantum wells (MQW's) under hydrostatic pressures up to 8 GPa. In PL, only a single peak is observed below 4.9 GPa, corresponding to the $n=1$ heavy-hole (HH) exciton in the $In_xGa_{1-x}As$ wells. Above 4.9 GPa, new PL lines related to X-like conduction band states appear. They are assigned to the type-II transition from the X_Z states in GaAs to the HH subband of the $In_xGa_{1-x}As$ wells and to the zero-phonon line and LO-phonon replica of the type-I transition involving the X_{XY} valleys of the wells. In addition to absorption peaks corresponding to direct exciton transitions in the wells, a new strong absorption feature is apparent in spectra for pressures between 4.5 and 5.5 GPa. This absorption is attributed to the *pseudodirect* transition between the HH subband and the X_Z state of the wells. This gives clear evidence for an enhanced strength of indirect optical transitions due to the breakdown of translational invariance in MQW structures. From experimental level splittings we determine the valence band offset and the shear deformation potential for X states in the $In_{0.2}Ga_{0.8}As$ layer. [S0163-1829(96)07443-7]

I. INTRODUCTION

The $In_xGa_{1-x}As/GaAs$ multiple quantum well (MQW) system has been extensively studied in recent years mainly due to its technological importance in heterostructure laser design.[1–5] In this system the $In_xGa_{1-x}As$ layers are strained provided the layer thickness does not exceed the composition-dependent critical value for lattice relaxation.[6] The electron states are modulated both by the strain and by the periodic potential along the growth direction. Various optical methods have been used to investigate the band structure and intersubband transitions in strained $In_xGa_{1-x}As/GaAs$ MQW's at ambient pressure[7–13] as well as at high hydrostatic pressure.[14–20] Most of the high pressure work is concerned with the direct transitions between subbands at the Γ point. Under sufficiently high pressure a reordering of different conduction band minima is induced in $In_xGa_{1-x}As/GaAs$ MQW's, known as the Γ-X crossover. X-related indirect emission bands have been reported,[15,18] but the assignment is still an open question.

Recent high-pressure photoluminescence (PL) studies of InAs monolayers embedded in GaAs have revealed spectral features, which have led to a consistent picture for the energy levels in such structures.[21] Inspired by these results we propose for the $In_{0.2}Ga_{0.8}As/GaAs$ MQW structure at pressures below and above the Γ-X crossover the band alignments schematically shown in Fig. 1.[22] The arrows indicate optical transitions, and their assignment will be discussed in detail below. The X valleys in the highly strained $In_xGa_{1-x}As$ layer are split into twofold degenerate X_Z and fourfold degenerate X_{XY} states.[23] The splitting of the X valleys is determined by the corresponding shear deformation potential. For biaxial compression the X_Z states are higher in energy. In the MQW's the X_Z states are folded back into the Γ point due to the lack of translational invariance along the growth axis. One would therefore expect some oscillator-strength enhancement for the pseudodirect optical transitions (X_3) in the wells. Up to now, however, no clear evidence for such an enhancement has been found in QW structures of indirect-band-gap materials.

In this work we report the pressure dependence of low temperature PL and transmission spectra of a $In_{0.2}Ga_{0.8}As/GaAs$ MQW system up to 8 GPa. The large band gap of the GaAs substrate compared to the energies of direct optical transitions between well states allows us to measure the absorption of the wells without the need to remove the substrate. The sharp exciton peaks observed in absorption broaden significantly above the pressure-induced Γ-X crossover. This effect is attributed to a reduction of the

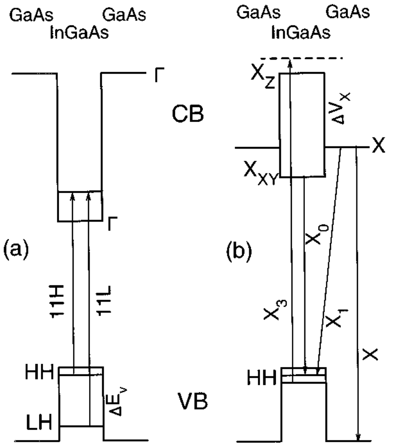

FIG. 1. Sketch of the band edge profiles of the $In_xGa_{1-x}As/GaAs$ MQW structure (a) below and (b) above the Γ-X crossover pressure. Arrows indicate observed optical transitions.

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exciton lifetime caused by Γ-X intervalley scattering processes. Above the Γ-X crossover new features are observed in PL spectra. These are assigned to the type-II transition from the X states in the GaAs barrier to the heavy-hole (HH) states in the $In_xGa_{1-x}As$ well and to type-I transitions between X_{XY} states and the HH states in the wells [compare Fig. 1(b)]. A striking result of this work is the observation of a pronounced absorption feature which is assigned to the pseudodirect transition X_3 from HH to X_Z states of the well. We propose an explanation for the large oscillator strength, which is based on the formation of a resonant state related to the X_Z level of the well. From the differences in X-related transition energies we determine the valence band offset ΔE_v for the strained $In_xGa_{1-x}As$/GaAs interface and the shear deformation potential for X states in the $In_xGa_{1-x}As$ layers under biaxial compression.

II. EXPERIMENTAL DETAILS

The $In_xGa_{1-x}As$/GaAs MQW sample was grown by molecular-beam epitaxy at substrate temperatures in the range of 520–580 °C. The substrate was Cr-doped semi-insulating GaAs (001). The layer sequence consists of an undoped GaAs buffer layer (0.7 μm thickness), 15 periods of undoped $In_{0.2}Ga_{0.8}As$ (8 nm) and GaAs (15 nm) layers, and an undoped GaAs cap layer. The indium concentration and the well widths were verified by double crystal x-ray diffraction and liquid-helium-temperature PL measurements.

For pressure experiments the sample was mechanically thinned to a total thickness of about 20 μm, and then cut into pieces of about 100×100 μm^2 in size. High pressure PL and optical transmission measurements were performed at $T=10$ K using a diamond-anvil cell in combination with a helium-flow cryostat. Condensed helium was used as the pressure-transmitting medium. Pressure was always changed at 300 K in order to ensure the best possible hydrostatic conditions. Pressure was measured using the ruby luminescence method[24] with temperature correction of the pressure calibration according to Ref. 25.

The PL spectra were excited by the 488 nm line of an Ar^+ ion laser at a power density of about 10 W/cm^2. For transmission measurements white light from a tungsten lamp was focused onto the sample forming a spot of about 35 μm in diameter. Both the PL and the transmitted light were analyzed by a 0.6 m single-grating spectrometer equipped with a GaAs photomultiplier operating in fast photon-counting mode. The transmission spectra were normalized in order to account for the spectral dependencies of all optical components.

III. RESULTS AND DISCUSSION

A. Optical transition energies and intensities

Figure 2 shows representative PL and transmission spectra of the $In_xGa_{1-x}As$/GaAs MQW sample measured at 10 K and at different pressures. At zero pressure three absorption bands are observed at energies below the direct band gap of GaAs. These features correspond to direct transitions 11H, 11L, and 22H involving the first and second electron and hole subbands. The emission line observed in PL spectra at zero pressure corresponds to the 11H transition, its Stokes shift is about 2 meV. With increasing pressure all the absorption and PL features shift to higher energy.

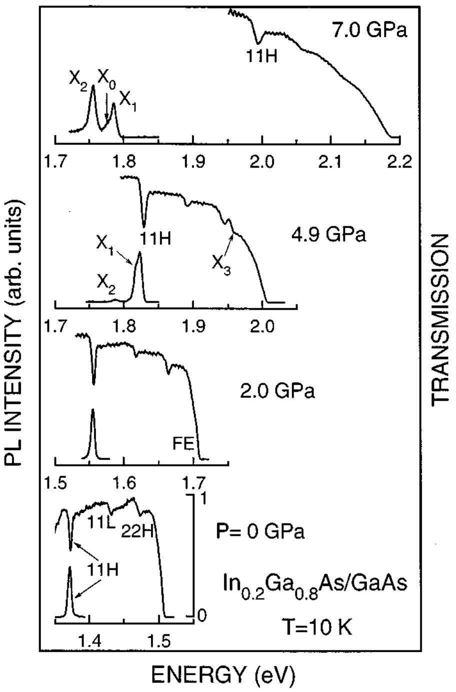

FIG. 2. Summary of photoluminescence and transmission spectra of the $In_xGa_{1-x}As$/GaAs MQW sample at 10 K and different pressures. The PL spectra have been normalized according to the respective strongest peak.

Near 4.9 GPa the intensity of the 11H PL peak decreases reversibly by about one order of magnitude within a pressure interval of less than 0.3 GPa. This behavior indicates the crossing of Γ- and X-related conduction band states. Two new emission lines (X_1 and X_2) appear above the crossover. These shift to lower energies with increasing pressure at a rate characteristic of Γ-X indirect gaps in III-V semiconductors. Near the conduction band crossover the absorption bands start to broaden significantly.

The pressure dependence of the main absorption and PL peak energies and of PL peak intensities is summarized in Figs. 3 and 4, respectively. For comparison, related energies for bulk GaAs are also shown in Fig. 3. These are the direct free exciton energy (FE) obtained from absorption edge measurements and the X-related exciton.[21] The solid lines in Fig. 3 represent the results of least-squares fits to the experimental data using first or second order polynomials. Fitted parameters are listed in Table I.

Figure 5(a) shows higher resolution PL spectra for pressures above 4.9 GPa. In addition to the two main peaks X_1 and X_2 there is a weak feature at the low-energy side of the X_1 peak. Deconvolution into two components was performed by least-squares fitting using Gaussian line profiles, as illustrated by the dashed lines in Fig. 5(a). The corresponding peak energies are plotted in Fig. 5(b) as a function of pressure. Within experimental uncertainty all three X-related PL peaks have the same pressure coefficient. The energy difference between the X_1 and X_0 transitions is about 8(4) meV. Figure 5(c) shows the ratios of peak intensities $I(X_1)/I(X_2)$ and $I(X_0)/I(X_2)$ as a function of pressure.

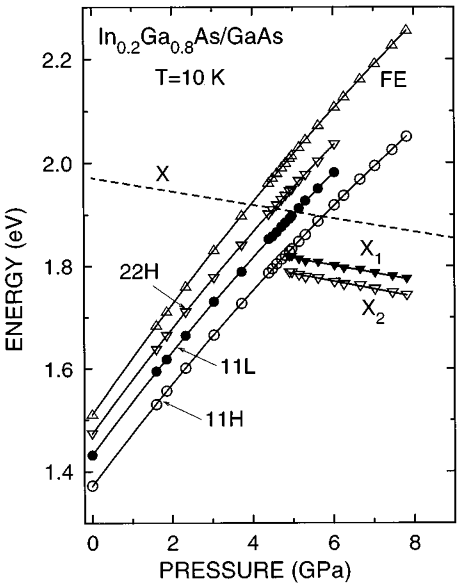

FIG. 3. Energies of PL and absorption peaks of the $In_xGa_{1-x}As/GaAs$ MQW sample as a function of pressure. The solid lines correspond to the results of least-squares fits. The dashed line represents the variation with pressure of the Γ-X indirect gap in bulk GaAs.

TABLE I. Coefficients describing the pressure dependence of PL and absorption peaks of the $In_xGa_{1-x}As/GaAs$ MQW sample obtained from least-squares fits of a quadratic expression $E(P)=E(0)+a_1\times P+a_2\times P^2$ to the experimental data.

Peak	$E(0)$ (eV)	a_1 (meV/GPa)	a_2 (meV/GPa^2)
11H	1.372(2)	104(2)	−2.1(5)
11L	1.432(2)	105(2)	−2.3(5)
22H	1.473(2)	107(2)	−2.3(5)
FE	1.510(2)	112(2)	−2.1(5)
11H(PL)	1.370(2)	103(2)	−2.1(5)
X_1	1.891(4)	−15(1)	
X_0	1.883(4)	−15(3)	
X_2	1.860(4)	−15(1)	
X_3	2.034(8)	−16(3)	
X	1.971(2)[a]	−13(1)[a]	

[a] Reference 21.

From 5 GPa to 8 GPa, the $I(X_1)/I(X_2)$ ratio decreases by almost two orders of magnitude, whereas $I(X_0)/I(X_2)$ is nearly independent of the pressure, except near the band crossover point.

B. Assignment of X-related PL transitions

An assignment of the X-related optical transitions can be made by taking into account the PL intensity variations under pressure. For $P\approx P_c$, the crossover pressure, the X_1 emission appears as strong as the 11H peak (see Fig. 4), whereas the X_2 emission is weaker by more than one order of magnitude. Above P_c the X_1 intensity decreases rapidly within about 0.3 GPa, whereas X_2 decreases only very little in intensity. Above 5.3 GPa both the X_1 and X_2 peaks have nearly the same intensities (on the logarithmic scale of Fig. 4) and the same decreasing tendency with further increase of pressure. The continuous intensity transfer from line 11H to line X_1 in the pressure range of the crossover is an indication of mixing effects between conduction band states induced by the potential step at the QW interface. This implies that the X_1 emission involves X_Z states. As schematically shown in Fig. 1(b), the X_Z levels of the $In_xGa_{1-x}As$ layer are pushed up in energy due to biaxial compression.[21] On the other hand, the GaAs layers act as *wells* for electrons in X_Z states. The peak X_1 is therefore attributed to the type-II transition from X_Z states in GaAs to the HH states in the $In_xGa_{1-x}As$ layers.

There is little intensity transfer between 11H and X_2 transitions. Furthermore, the X_{XY} valleys in the $In_xGa_{1-x}As$ well are located below the X states in GaAs. Thus the X_2

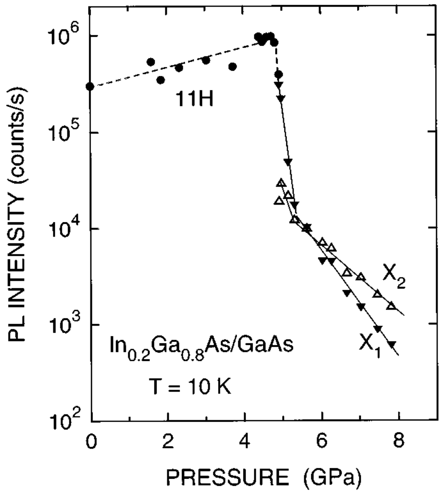

FIG. 4. Pressure dependence of the PL peak intensities of the 11H, X_1, and X_2 features.

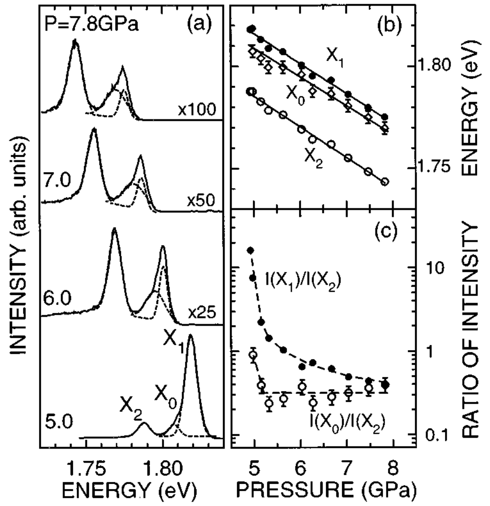

FIG. 5. (a) Photoluminescence spectra of the $In_xGa_{1-x}As/GaAs$ MQW sample at different pressures above 4.9 GPa. Dashed curves represent fitted Gaussian line profiles. (b) Pressure dependence of the PL peak energies. (c) PL peak intensities of transitions X_0 and X_1 relative to that of the X_2 transition. Dashed lines are a guide to eye.

peak corresponds to a type-I transition involving X_{XY} and HH states of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer. The fact that the ratio $I(X_0)/I(X_2)$ is nearly independent of pressure suggests that peaks X_0 and X_2 belong to the same type of transition. Furthermore, the energy difference between X_0 and X_2 emissions amounts to 23(4) meV, which is close to the energy of LO phonons at the X point of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ alloys.[26] Thus, we attribute the X_0 and X_2 bands to the zero-phonon line and the LO phonon replica of the type-I transition within the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layers, respectively. The appearance of the zero-phonon line is mainly attributed to compositional disorder in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ wells.

The above assignment is consistent with the temperature dependence of PL spectra above the Γ-X crossover. The two main features remain observable up to 80 K. However, the relative intensity of the X_2 emission decreases continuously with increasing temperature. This indicates a relatively small thermal activation energy for electrons in the X_{XY} valleys. In fact, within the proposed level scheme the X_{XY} states in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer are only 8 meV below the X minimum in the GaAs barrier.

C. Valence band offset

The valence band offset ΔE_v can be obtained from the energy difference of the peaks X_1 and X (X-related exciton in bulk GaAs) as indicated in Fig. 1(b). If we neglect the small confinement energy of the first HH state and the difference between the exciton binding energies in the MQW and bulk GaAs, ΔE_v can be determined directly from the data in Table I, being 80(4) meV. For a similar MQW structure it has been suggested[10,20] that the light holes are confined in the GaAs layer rather than in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ due to the large strain effects. In that case, ΔE_v should agree with the energy difference of the 11H and 11L absorption peaks [60(4) meV]. This 20 meV discrepancy in the ΔE_v values is larger than the magnitude of the terms neglected in our calculation. The interpretation in Refs. 10 and 20 is based only on the agreement between the measured peak energies and the results of a $\mathbf{k}\cdot\mathbf{p}$ calculation. In contrast, our assignment of X-related peaks results from the analysis of both the measured energies and the temperature dependence of intensities. We conclude that the 11L transition is still of type-I in the MQW structure investigated here.

D. Direct exciton absorption

Figure 6 shows experimental absorption spectra for different pressures in the energy range below the absorption edge of GaAs. The optical density D_O is given by $D_O = \log_{10}[I_0(\omega)/I(\omega)]$, where $I(\omega)$ and $I_0(\omega)$ are the transmitted and the reference intensity, respectively, at energy $\hbar\omega$.

The formation of two-dimensional (2D) excitons in semiconductor QW structures[27,28] is evidenced by the sharp peak-like structures which dominate the absorption near the band gaps. According to Shinada and Sugano,[29] for each direct allowed excitonic transition between hole and electron subbands the absorption coefficient can be expressed as a sum of two contributions: excitonic discrete lines and the exciton continuum. In MQW structures of ternary alloys, compositional disorder and well width fluctuations are responsible for an inhomogeneous broadening of the exciton absorption lines.[30] An expression for the excitonic absorption line shape in QW's including a Gaussian line broadening has been given elsewhere.[27]

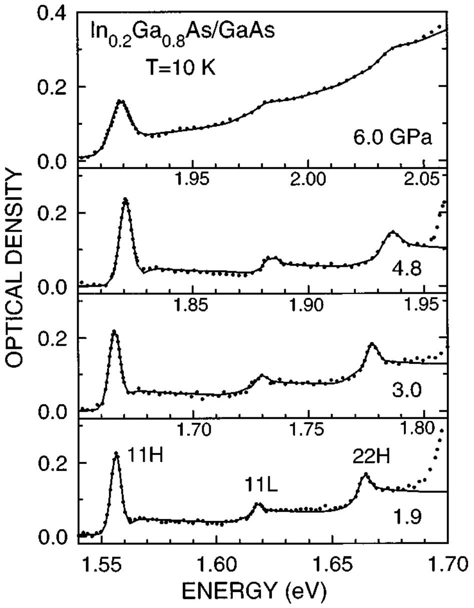

FIG. 6. Exciton absorption spectra of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ MQW sample for different pressures. The solid lines represent the results of line shape fits to the experimental data (see text for details).

The solid lines in Fig. 6 represent results of least-squares fits using the line profile function of Ref. 27. For each excitonic transition the adjustable parameters were the exciton ground state energy E_0, the exciton binding energy E_b, the linewidth Γ_1, and a normalization factor C_0. In fitting the absorption spectra above the Γ-X crossover, the indirect absorption tail of the GaAs substrate was taken into account by introducing a parabolic background term.[31] The results of the fits are more accurate for the 11H and 22H excitons because the peaks are better defined in the spectra. We discuss in the following only the 11H and 22H excitons.

The pressure dependencies of the half-widths (Γ_1) of the 11H and 22H exciton peaks are shown in Figs. 7(a) and (b), respectively. The inset shows the binding energy E_b of the 11H exciton as a function of pressure. The solid lines in Fig. 7 are a guide to the eye. For pressures below 4.9 (4.0) GPa the width of the 11H (22H) exciton peak exhibits only a weak dependence on pressure. Above 4.9 (4.0) GPa the 11H (22H) exciton peak broadens significantly. From Fig. 3 we can see that the lines corresponding to the energies of 11H (22H) and X_1 transitions cross at 4.9 (4.0) GPa. In other words, each time that an electron subband of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ wells crosses the X valley in the GaAs layers the corresponding exciton line starts to broaden. We therefore attribute the broadening of the exciton lines to Γ-X intervalley scattering of the carriers after the Γ-X crossover.

A pressure-induced exciton line broadening has been observed in bulk GaAs (Ref. 32) and Ge,[33] where it is well accounted for by phonon-assisted intervalley scattering theory.[34] The intervalley scattering involves, via electron-

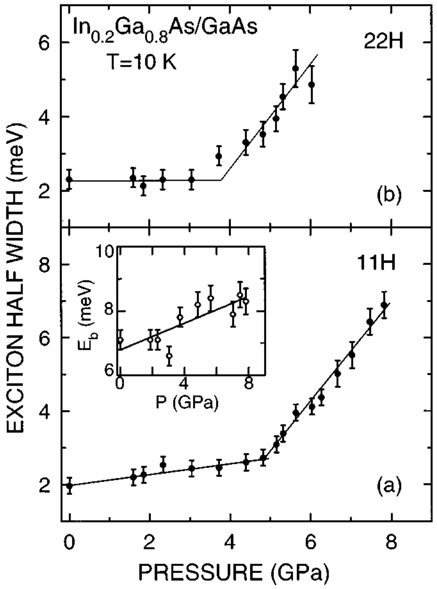

FIG. 7. Pressure dependence of the half-widths of (a) 11*H* and (b) 22*H* exciton lines. The solid lines are a guide to the eye. The inset shows the binding energy of the 11*H* exciton as a function of pressure.

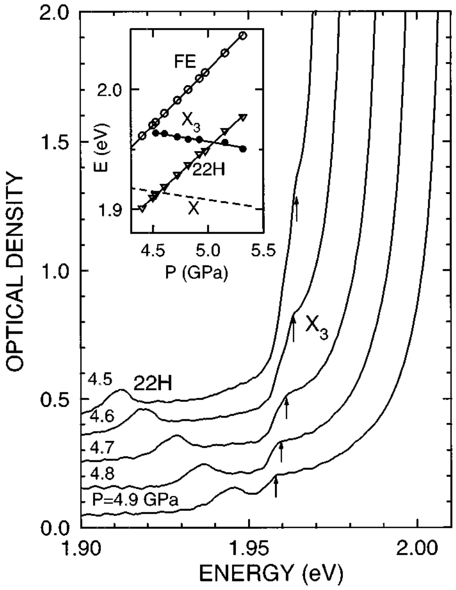

FIG. 8. Absorption spectra of the $In_xGa_{1-x}As/GaAs$ MQW sample for pressures above 4.5 GPa. Spectra have been shifted vertically for clarity. Arrows indicate the energy of X_3 absorption band. The inset shows the peak energy of transitions X_3, 22*H*, and FE as a function of pressure.

phonon interaction, the emission or absorption of proper zone-edge phonons allowed by symmetry. The scattering probability increases with the pressure-dependent Γ-X energy separation.[32,33] Using the phonon deformation potentials obtained for bulk GaAs,[32] we find a good agreement between calculated and measured linewidth increase of the 11*H* (22*H*) transitions for pressures lower than 6.5 GPa (5.0 GPa). Nevertheless, the linewidth increases further beyond these pressures. This is an indication that an additional scattering channel becomes active. As another possible mechanism for intervalley scattering in MQW structures, we may consider elastic scattering of carriers induced by interface roughness.

E. Pseudodirect absorption related to X_Z well states

Above 4.5 GPa a new structure labeled X_3 appears in transmission at energies higher than the PL peak X of GaAs. Figure 8 shows several absorption spectra measured at pressures, where the X_3 feature indicated by arrows is clearly observed below the steep absorption edge of GaAs. We note that the X_3 feature is about as intense as the 22*H* direct transition. The energy of the X_3 transition is taken from the zeros of the second derivative of the optical density with respect to photon energy. The inset shows the pressure dependence of the X_3 peak energy together with the 22*H* and FE energies. The dashed line in Fig. 8 represents the pressure dependence of the X exciton in bulk GaAs. The redshift of the X_3 transition energy with increasing pressure is characteristic of a Γ-X indirect transition but its intensity is comparable to that of the observed direct absorption peaks. Therefore, we attribute the X_3 band to the pseudodirect transition from a HH subband to the strain-split X_Z valley in the $In_xGa_{1-x}As$ layer, as indicated in Fig. 1(b).

The breakdown of translational invariance due the potential steps at the heterointerfaces folds back the X_Z states of the well to the Γ point inducing a mixing between conduction band states. As a consequence of the mixing, the X_3 absorption acquires in part the character of a direct allowed transition. However, given an $In_xGa_{1-x}As$ layer width of 8 nm and the typically small mixing potential of ≈ 1 meV, it would be difficult to explain the large oscillator strength of the X_3-related absorption simply by Γ-X mixing effects.

We propose a different explanation, which takes into account that the X_Z levels in the strained $In_xGa_{1-x}As$ layers are higher in energy compared to the X valley in GaAs [see Fig. 1(b)]. The $In_xGa_{1-x}As$ constitutes a potential *barrier* for electrons in the X valleys of GaAs having wave vectors $\vec{k} \| z$. For such a quantum mechanical system there exist states with energies larger than the barrier height ΔV_X, but with wave functions being fairly concentrated within the barrier. These states correspond to the *resonances* in the transmission coefficient of the barrier.[35] The first resonance occurs for a wave vector $k = \pi/W$, where W is the width of the $In_xGa_{1-x}As$ layers. The matrix element for direct optical transitions between heavy holes and the resonance state has the form[35]

$$M \propto \int_{\text{crystal}} \chi_e^*(z) \;\; \hat{e} \cdot \vec{r} \;\; \chi_h(z) dz \cdot \int_{\text{unit cell}} u_c^*(\vec{r}) u_v(\vec{r}) d\vec{r}, \quad (1)$$

where $\hat{e}$ is the polarization vector of the light, u_c, u_v are p-type Bloch functions of conduction and valence band states, and χ_e, χ_h are the electron and hole envelope functions of the second heavy-hole subband and the resonance state, respectively. Since the integral over the Bloch functions yields almost unity, the optical matrix element M is essentially given by the magnitude of the dipole moment taken between the envelopes. This matrix element has large values of the order of W, the width of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ quantum well.

F. Shear deformation potential for X states

From the energies of the X_3 absorption and X_0 emission bands we obtain a splitting of 150(10) meV between the X_Z and X_{XY} valleys in the strained $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer. The energy separation of the split X valleys can be expressed as[23,36]

$$X_Z - X_{XY} = -\Xi_u(1+2C_{12}/C_{11})\,\varepsilon_{xx}, \tag{2}$$

where Ξ_u is the shear deformation potential for X valleys, C_{11} and C_{12} are the elastic constants of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$, and ε_{xx} is the built-in strain of the wells. For the alloy C_{11} and C_{12} values are estimated by linear interpolation of the GaAs and InAs data.[26] The strain ε_{xx} is given by the lattice mismatch between $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ and GaAs. In this way we obtain a value of $\Xi_u = 5.5(4)$ eV for the shear deformation potential of X conduction band minima in $\mathrm{In}_{0.2}\mathrm{Ga}_{0.8}\mathrm{As}$. This result is in very good agreement with an interpolation of theoretical values of Ξ_u for GaAs (6.0 eV) and InAs (3.7 eV) calculated within a tight-binding approach.[36] A somewhat larger theoretical value of 7.8 eV is reported in Ref. 37.

IV. SUMMARY

We have investigated the effect of pressure up to 8 GPa on the low-temperature luminescence and excitonic absorption of an $\mathrm{In}_{0.2}\mathrm{Ga}_{0.8}\mathrm{As}$/GaAs MQW structure. At low pressures three absorption bands have been observed below the band gap of GaAs. These correspond to direct exciton transitions between quantum well subbands. When the confined electron states in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ wells become, with increasing pressure, subsequently degenerate in energy with the X conduction band edge, the exciton absorption peaks exhibit a pronounced broadening which is attributed to Γ-X intervalley scattering. In the pressure range of 4.5 to 5.5 GPa a new absorption feature is observed at energies higher than the indirect Γ-X gap of bulk GaAs but with the same negative pressure coefficient. We assign this feature to the *pseudodirect* transition from the second HH subband at Γ to a resonant state related to the X_Z levels in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer. In this way, we have obtained evidence for an enhanced strength of indirect optical transitions induced by the potential steps at heterointerfaces in a MQW structure. The dominant peak in the PL spectra for pressures $P < 4.9$ GPa corresponds to $11H$ transition. Above 4.9 GPa three X-related emission bands appear. Their assignment in terms of spatially direct and indirect $X \rightarrow \Gamma$ recombination is presented and the valence band offset is determined. From combined PL and absorption results we determine the energy splitting between X_Z and X_{XY} states in the strained $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layers and obtain an experimental value for the corresponding shear deformation potential.

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