

Composition modulation and inhomogeneous strain field in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InP}$ strained layers

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Optical-absorption measurements have been carried out on tensile and compressive $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InP}$ strained layers. It is shown that the energetic dispersion of the heavy-hole relative to the light-hole subband $\sigma_{\text{HH}}/\sigma_{\text{LH}}$ is the key to knowing the origin of the microscopic inhomogeneities. So, $\sigma_{\text{HH}}/\sigma_{\text{LH}} < 1$ indicates the existence of composition inhomogeneities whereas $\sigma_{\text{HH}}/\sigma_{\text{LH}} = 2.8$ reveals an inhomogeneous strain field.

INTRODUCTION

III-V ternary semiconductors have been extensively studied as they are excellent materials for optoelectronics and high electron mobility devices. Their band-gap energy and electron effective mass can be tailored by choosing the molar fraction of the binary constituents. Usually epitaxial layers were lattice matched. That is, they were grown on a substrate with the same lattice parameter. In the past decade the possibility of growing strained layers has introduced another degree of freedom. The lattice mismatch produces a strain that, in turn, modifies the band structure.

High-quality homogeneous $\text{In}_x\text{Ga}_{1-x}\text{As}$ matched layers on InP are now grown to the point that only local statistical fluctuations of composition remain.¹ However, in strained layers, inhomogeneities do appear which are clearly observed as a contrast modulation in TEM (transmission electron microscopy).² Its microscopic origin is usually related^{2,3} to a composition modulation. Chemical microanalysis by x-ray³ together with theoretical calculations⁴ support this assertion. Our optical-absorption (OA) measurements were done with the aim of quantifying those composition variations. Surprisingly, the analysis of the OA spectra reveals another possible origin: the existence of an inhomogeneous strain field.

This paper is devoted to showing that near-band-gap optical-absorption spectra can clearly distinguish between both alternatives.

THEORY

A. Optical-absorption spectral shape

Following the paper of Elliot,⁵ the absorption coefficient near the band-gap edge is greatly enhanced by the excitonic effects. Considering nondegenerate parabolic bands, its spectral shape is given by

$$\alpha(h\nu) = A \frac{2\pi\sqrt{\epsilon_x}}{h\nu} \sum_{n=1}^{\infty} \frac{2\epsilon_x}{n^3} \delta \left[h\nu - E_g + \frac{\epsilon_x}{n^2} \right] + \frac{H(h\nu - E_g)}{1 - \exp(-2\pi\sqrt{\epsilon_x}/h\nu - E_g)}, \quad (1)$$

where ϵ_x is the exciton binding energy and $H(x)$ is the Heaviside function. The first term accounts for the transition into the discrete bound states whereas the second

one is related to the continuum. The band-gap energy E_g depends on the molar composition. For $\text{In}_x\text{Ga}_{1-x}\text{As}$ unstrained layers, we have at 4 K (Ref. 6)

$$E_g(x) = 1.519 - 1.608x + 0.50x^2 \text{ eV}. \quad (2)$$

The strain raises the degeneracy at the top of the valence band. The heavy-hole (HH) and light-hole (LH) valence-band energies relative to the conduction band change with the strain ϵ according to

$$\Delta E_{\text{LH}} = 3.9\epsilon, \quad (3)$$

$$\Delta E_{\text{HH}} = 10.8\epsilon \text{ (eV)},$$

where $\epsilon > 0$ ($\epsilon < 0$) accounts for compressive (tensile) samples. These formulas have been calculated with the elastic stiffness coefficients given in Refs. 7 and 8 for $x = 0.5$.

When the top of the valence band is no more degenerated the exciton has a bound state associated with every subband. For both bands the exciton binding energy takes the same value.⁹ So the absorption spectra would be described with four free parameters: the relative band amplitude $A_{\text{HH}}/A_{\text{LH}}$, the band-gap energy E_g , the band-gap splitting energy $\Delta E \equiv E_{g\text{LH}} - E_{g\text{HH}}$, and the exciton binding energy ϵ_x .

B. Band-gap dispersion energy

To account for the microscopic inhomogeneities we have to convolute the spectrum with a Gaussian distribution characterized by the standard deviation σ_{HH} and σ_{LH} . In fact, this is the feature that will allow us to distinguish which is the microscopic origin of the energy dispersion.

Suppose that the layer has a homogeneous composition but due to an unknown origin the strain changes from point to point. Following formulas (3) this strain field will give rise to an energy dispersion of

$$\begin{aligned} \sigma_{\text{HH}} &= 10.8\sigma_\epsilon, \\ \sigma_{\text{LH}} &= 3.9\sigma_\epsilon \text{ (eV)}. \end{aligned} \quad (4)$$

In this case, $\sigma_{\text{HH}}/\sigma_{\text{LH}} \approx 2.8$.

Now consider the case where the composition changes from point to point. Of course there will be a strain associated with the composition that can be calculated in the virtual-crystal approximation

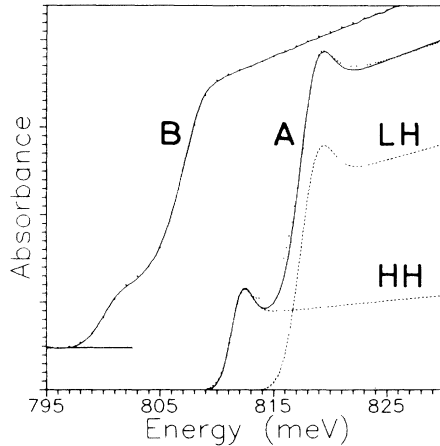


FIG. 1. Optical-absorption spectra measured at 10 K on *A* and *B* tensile samples (points). The calculated spectra are plotted altogether (solid line). Broken lines correspond to the HH and LH contributions.

$$\epsilon = \eta \frac{xa_{\text{InAs}} + (1-x)a_{\text{GaAs}}}{a_{\text{InP}}}, \quad (5)$$

where a_{AB} are the lattice constants and η accounts for the possibility of partial strain relaxation. By combining Eqs. (2), (3), and (5) the energy dispersion related to a composition modulation can be calculated:

$$\begin{aligned} \sigma_{\text{LH}} &= (1.608 - x - 0.26\eta)\sigma_x, \\ \sigma_{\text{HH}} &= (1.608 - x - 0.73\eta)\sigma_x. \end{aligned} \quad (6)$$

These formulas are valid near $x = 0.5$ (say $0.4 < x < 0.6$) and state that $\sigma_{\text{HH}}/\sigma_{\text{LH}}$ is always less than unity unless the layer is completely relaxed ($\eta = 0$).

In summary, the relative energy dispersion can be used to deduce which is the origin of microscopic inhomogeneities: $\sigma_{\text{HH}}/\sigma_{\text{LH}} \approx 2.8$ for a strain field and $\sigma_{\text{HH}}/\sigma_{\text{LH}} < 1$ for a modulation composition.

EXPERIMENT

The four samples analyzed in this work are $\text{In}_x\text{Ga}_{1-x}\text{As}$ epitaxial layers grown on InP. The *B*, *C*, and *D* epilayers were grown by MBE (molecular-beam epitaxy) and the *A* sample by VPE (vapor-phase epitaxy). Table I summarizes layer characteristics. The width has been determined through the period of the interference oscillations seen in the absorbance spectrum at energies below the band gap. The strain value comes from the band-gap splitting energy ΔE quoted in Table II.

The optical-absorption measurements have been carried out with a Fourier transform IR spectrometer. Sam-

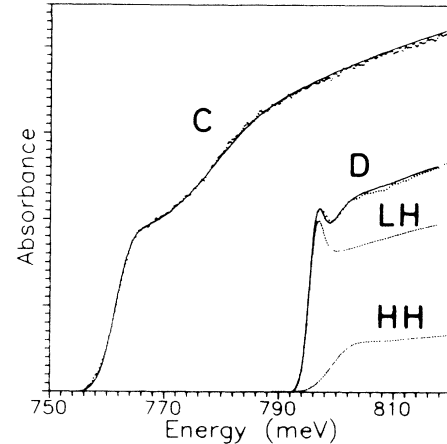


FIG. 2. Optical-absorption spectra on compressive *C* and *D* samples.

ples were illuminated with a tungsten lamp after a CaF_2 beamsplitter and the transmitted light was detected with a cooled InSb detector. A continuous flow helium cryostat allowed the samples to be held at 10 K.

RESULTS AND DISCUSSION

All the spectra plotted in Figs. 1 and 2 show a splitting in the absorption band edge that reveals the valence-band splitting due to an internal strain. Band-structure calculations¹⁰ give an optical matrix element between conduction and valence bands more intense in the case of LH than HH subbands. So in samples *A* and *B* (Fig. 1) the HH band lies higher in energy and determines the band-gap energy, indicating that the strain is tensile. On the other hand, samples *C* and *D* are compressive layers.

In order to obtain accurate values of band parameters all spectra have been fitted following the method described in Sec. I. In all cases the calculated spectrum fits reasonably well the measured one. The fitting parameters are summarized in Table II. Relative band intensities $A_{\text{HH}}/A_{\text{LH}}$ lie close to 0.35 in agreement with previous published results.¹¹ To complete the general description of spectra, a collateral feature must be noted with minor discussion. Surprisingly the exciton binding energy changes from sample to sample, the extreme values being 0.9 and 1.4 meV. These values contrast with the 2.0 meV predicted by the hydrogenic model and experimentally found by OA and photoluminescence by Goetz *et al.*¹² for $x \approx 0.5$. However, it seems rather usual to fit the OA spectra with a lower binding energy [in $\text{In}_x\text{Ga}_{1-x}\text{As}$ (Ref. 11), in GaAs (Ref. 13)]. This can be explained by the complex structure of the valence band not taken into account in formula (1). Nevertheless the origin of the

TABLE I. Sample characteristics. The strain value has been deduced from the band-gap splitting.

Sample	Growth	Substrate	Layer width	Strain	Type
<i>A</i>	VPE	Si doped	2.9	-8.8×10^{-4}	tensile
<i>B</i>	MBE	Fe doped	1.6	-9.3×10^{-4}	tensile
<i>C</i>	MBE	Fe doped	0.5	6.2×10^{-3}	compr.
<i>D</i>	MBE	Fe doped	2.0	6.3×10^{-4}	compr.

TABLE II. Fitting parameters. E_g is the band-gap energy, ΔE is the band-gap splitting, A_i is the amplitude of the light-hole and heavy-hole subbands, σ_i their corresponding energy dispersion, and ϵ_x the exciton binding energy.

Sample	E_g (meV)	ΔE (meV)	$A_{\text{HH}}/A_{\text{LH}}$	σ_{LH} (meV)	$\sigma_{\text{HH}}/\sigma_{\text{LH}}$	ϵ_x (meV)
<i>A</i>	807.2	-6.3	0.35	0.9	0.67	1.2
<i>B</i>	802.0	-6.6	0.37	1.7	0.93	0.9
<i>C</i>	733.9	44.1	0.40	2.3	2.5	0.9
<i>D</i>	797.7	4.5	0.35	1.2	2.4	1.4

variability from sample to sample remains obscure. In any case this problem does not affect the analysis that follows.

Let us now discuss the central point of our work: the energy dispersion of the band-edge absorption. When layers are perfectly homogeneous, it remains a dispersion in the exciton energy due to statistical fluctuations of molar composition into the volume of the ground exciton state.¹ This gives the limiting value of energy dispersion. Following Ref. 1 the lower limit on an $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer with $x \approx 0.5$ would be 0.66 meV for a completely relaxed sample. For an ideal strained layer ($\eta > 0$) this value would be even lower [formula (6)]. So our samples have spatial inhomogeneities other than the statistical fluctuations.

In samples *C* and *D* the energy dispersion of HH is greater than in the LH band. In fact, its relative value is close to the expected $\sigma_{\text{HH}}/\sigma_{\text{LH}} = 2.8$ for the case of inhomogeneous strain field. So we can conclude unambiguously that this is the dominant microscopic origin of the energy dispersion in these compressive samples. We can calculate the corresponding strain dispersion σ_ϵ with formula (4) (Table III). The Gaussian strain distribution is plotted in Fig. 3.

In samples *A* and *B* $\sigma_{\text{HH}} < \sigma_{\text{LH}}$. This feature is coherent with the interpretation that, in this case, the energy dispersion comes from a composition modulation. So use of formula (6) with $x = 0.5$ allows us to determine the dispersion in the indium molar composition σ_x from σ_{LH} (Table III). Moreover, one can deduce as well the

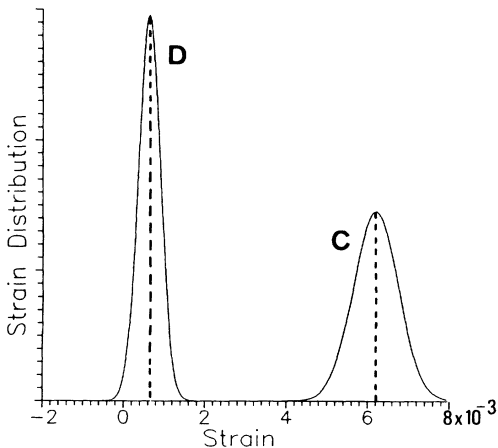


FIG. 3. The Gaussian strain distributions coherent with the energy dispersion present in compressive samples.

fraction of remaining strain η in the layers. In fact, the later calculation is subjected to great errors. A slight contribution of an inhomogeneous strain field would approach $\sigma_{\text{HH}}/\sigma_{\text{LH}}$ to unity. So the η value deduced by this procedure should be taken as a lower limit of the remaining strain.

The shape of the band-edge absorption is very sensitive to the energy dispersion. In particular, the exciton peak disappears when σ becomes large enough, as can be clearly seen by comparing the HH and LH subbands of sample *D* (Fig. 2). The error associated with $\sigma_{\text{HH}}/\sigma_{\text{LH}}$ is lower than 5%. So our conclusions are not weakened by the small discrepancies between the experimental and calculated spectra seen in samples *A* and *D*.

The presence of a composition modulation has been justified theoretically even when the growth conditions are spatially homogeneous.⁴ An inhomogeneous strain field in a layer with a homogeneous composition seems more difficult to justify. The easier possibility would be the following one. If the layer has a certain degree of strain relaxation with the appearance of dislocations, some domains in the layer could be more or less relaxed. However, this possibility has to be ruled out in sample *D* because no dislocations network has been seen in TEM observations. So the inhomogeneous strain field has to be associated with the internal structure of the layer.

The samples reported in this paper have been chosen from a number of samples where similar behavior was observed. The striking fact is that modulation composition seems to prevail always in tensile layers whereas in compressive ones an inhomogeneous strain field is always present. This behavior is perhaps quite general and should be tested with many other samples.

SUMMARY

The analysis of low-temperature optical-absorption spectra of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InP}$ strained layers can be explained by taking into account the existence of microscopic inhomogeneities other than statistical fluctuations.

TABLE III. Values of composition (σ_x) and strain (σ_ϵ) dispersion. η accounts for the remaining strain.

Sample	σ_x	η	σ_ϵ
<i>A</i>	0.9×10^{-3}	0.65	
<i>B</i>	1.6×10^{-3}	0.16	
<i>C</i>			5.5×10^{-4}
<i>D</i>			2.7×10^{-4}

It has been demonstrated that a composition modulation gives an energy dispersion smaller for the HH absorption band than for the LH ($\sigma_{HH} < \sigma_{LH}$). The contrary ($\sigma_{HH} = 2.8\sigma_{LH}$) holds when, the composition being homogeneous, the strain changes from point to point. This strain field has been found even in unrelaxed samples, so it is not related to a kind of inhomogeneous relaxation. Both possibilities, strain field and composition modulation, have been found in compressive and tensile samples, respectively. It has been suggested that this distinct be-

havior could be quite general.

To conclude, it should be emphasized that optical absorption is a very appropriate tool for these kind of problems and can be a very good complement to TEM observations. The conclusions given above could not have been reached by photoluminescence measurements. Thermalization prevents one from seeing the valence-band splitting at low temperature¹⁴ and the half-width of the free-exciton peak depends on the excitation conditions.¹⁵

¹E. F. Schubert, E. O. Gobel, Y. Horikoshi, K. Ploog, and H. J. Queisser, *Phys. Rev. B* **30**, 813 (1984).

²F. Peiro, A. Cornet, J. R. Morante, S. Clark, and R. H. Williams, *Appl. Phys. Lett.* **59**, 1957 (1991).

³P. Henoc, A. Izrael, M. Quillec, and H. Laundis, *Appl. Phys. Lett.* **40**, 963 (1982).

⁴F. Glas, *J. Appl. Phys.* **62**, 3201 (1987).

⁵J. Elliot, *Phys. Rev.* **108**, 1383 (1957).

⁶D. K. Gaskill, N. Bottka, L. Aina, and M. Mattingly, *Appl. Phys. Lett.* **56**, 1269 (1990).

⁷S. Adachi, *J. Appl. Phys.* **58**, R1 (1985).

⁸S. Adachi, *J. Appl. Phys.* **53**, 8775 (1982).

⁹B. Sermage and G. Fishman, *Phys. Rev. B* **30**, 813 (1984).

¹⁰T. C. Chong and C. G. Fonstad, *J. Quantum Electron.* **25**, 171 (1989).

¹¹E. H. Reihlen, D. Birkedal, T. Y. Wang, and G. B. Stringfellow, *J. Appl. Phys.* **68**, 1750 (1990).

¹²K. H. Goetz, D. Bimberg, H. Jurgensen, J. Selders, A. V. Solomonov, G. F. Glinskii, and M. Razeghi, *J. Appl. Phys.* **54**, 4543 (1983).

¹³M. D. Sturge, *Phys. Rev.* **127**, 768 (1962).

¹⁴I. C. Bassignana, C. J. Miner, and N. Puetz, *J. Appl. Phys.* **65**, 4299 (1989).

¹⁵E. F. Schubert and W. T. Tsang, *Phys. Rev. B* **34**, 2991 (1986).