

A POLARIZATION STUDY OF ORDERED GaInP₂

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ABSTRACT

Optical polarization in ordered GaInP₂ has been studied by low-temperature photoluminescence. By comparing our experimental results to the theoretical predictions, we show that to obtain quantitative information about ordering from the polarization of near-band-gap transitions, the effects of lattice mismatch, substrate misorientation, and excitonic transitions should be taken into account.

INTRODUCTION

Spontaneous ordering in III-V semiconductors, GaInP₂ for instance, brings about changes in their band structures from that of the corresponding disordered alloys [1], and causes changes in the polarization of their optical spectra [2]. Optical anisotropy caused by Cu-Pt ordering has been observed in various spectroscopic studies: photoluminescence (PL) and PL excitation [3], piezomodulated reflectance [4], reflectance difference [5], electroreflectance [6] and photocurrent [7]. Qualitatively, these studies have all shown the change in crystal symmetry as a result of ordering. Quantitatively, the dependence of polarization on the degree of ordering (η) has been used to obtain the parameter η [2] or to explain the dependence of reflectance difference on the parameter η [5]. Theoretically, the η -dependence of the optical polarization has been studied based on a model of band-to-band transitions at $k = 0$ and the strain effect due to the lattice mismatch between the epilayer and the substrate has been considered as well [2]. In this work we would like to show that there are two other aspects that can affect the quantitative analysis of the polarization measurements significantly but have not been considered in previous studies. They are substrate misorientation, which exists in most of the ordered samples, and excitonic transitions, which are the fundamental processes governing optical transitions. We also discuss the strain effect in real samples used in this work.

EXPERIMENT

The ordered Ga_xIn_{1-x}P₂ samples investigated in this work were grown by low- or high-pressure MOVPE on GaAs substrates with various misorientation angles (from 0° to 9°) towards a [111]B direction. The disordered samples were grown by low-pressure MOVPE on GaAs substrates 6° misoriented towards [111]A. The details about sample growth are described elsewhere [8]. The lattice constant of the GaInP₂ epilayer is nearly matched to that of GaAs at room temperature, but with a larger deviation at liquid helium temperature because of the difference in their thermal expansion coefficients. The lattice-matched composition is $x = 0.515$ and 0.520 at room temperature and liquid helium temperature, respectively. The actual compositions for the samples used in this study are in the range of $0.50 < x < 0.52$, corresponding to a range of biaxial strain $0 < |\epsilon_{\text{biaxial}}| < 0.15\%$.

PL was excited by an argon ion laser (488 nm line) and measured at 10 K. Assuming an untilted substrate, the polarization of the emission normal to the sample surface was measured parallel to $[\bar{1}10]$ and $[110]$ crystalline directions. The polarization analyzer is composed of a linear polarizer and a half wavelength retarder.

RESULTS

In this work, the ordering direction is assumed to be along the $[111]$ crystalline direction. The polarization, R , is defined as the intensity ratio of the PL peak polarized parallel to the assumed $[\bar{1}10]$ and $[110]$ directions on the sample surface. These directions deviate from the true crystalline $[\bar{1}10]$ and $[110]$ directions if the sample is grown on a misoriented substrate.

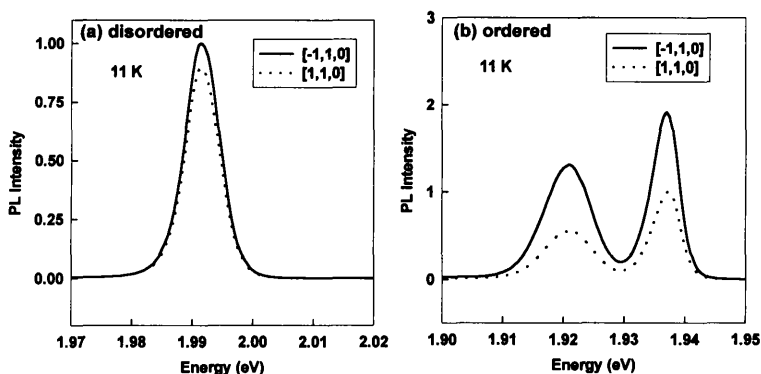


Fig.1. Photoluminescence spectra of GaInP₂ in two polarizations.

Fig.1a shows the PL spectra of a disordered sample for two polarizations. The weak polarization anisotropy is a result of residual ordering. In Fig.1b, the spectra for an ordered sample reveal a stronger polarization anisotropy for the two PL bands. The narrow band, seen only in the samples grown on substrates 6° misoriented towards $[111]_B$ under suitable conditions [8], is from intrinsic exciton recombination; the origin of the broad band, referred to as the “moving peak”, is uncertain, although it is suspected to involve spatially indirect free particle recombination [9]. We will refer to the former as the high energy peak and the latter as the low energy peak. This polarization measurement indicates that the hole involved in the recombination associated with the moving peak originates in an ordered region and, thus, carries the signature of the valence band symmetry for Cu-Pt ordering. Fig.2 is a summary of the polarization measurements for a series of samples with different degrees of ordering (all grown on 6° - $[111]_B$ misoriented substrates), and two disordered samples. On average, the polarization ratios are about 1.9 for the excitonic peak and 2.3 for the moving peak in the ordered samples.

The effect of substrate misorientation is investigated for five groups of samples. Samples in each group were grown at the same conditions but on substrates with different misorientation angles. The experimental results for the moving peak are shown in Fig.3, where the misorientation angle dependence predicted by theory is shown for comparison. Indeed, the trend

for samples in each group agrees with the theory, although there is a scatter in the values from one group to the other.

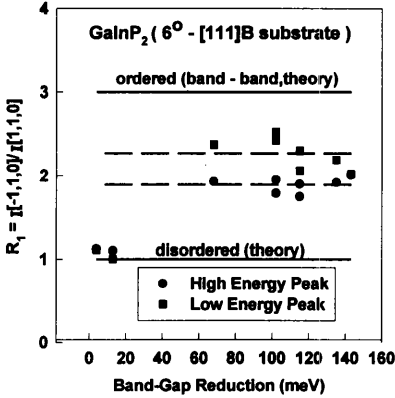


Fig.2. Polarization as a function of band-gap reduction.

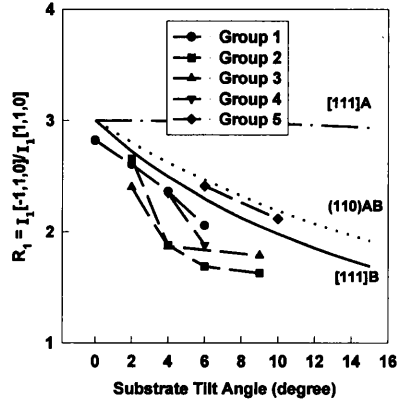


Fig.3. Polarization as a function of substrate misorientation angle.

DISCUSSIONS

In Cu-Pt ordered III-V semiconductor alloys, the top most valence band state is a heavy-hole-like (HH) state, and the polarization ratio R_1 , which is equal to 3 for band-to-band transitions from the band edge states, is independent of the degree of ordering [2]. This property can be exploited to examine other possible mechanisms that may cause the discrepancy between experimental results and the theoretical predictions. On the other hand, the polarization ratios for the light-hole-like (LH) band, R_2 , and the spin-orbit split-off (SO) band, R_3 , strongly depend on the degree of ordering. In principle, such a dependence can be used to measure the degree of ordering. Nevertheless, to do so, various corrections are required as will be discussed in the following text.

(1) The strain effect due to lattice mismatch: This effect has been studied in Ref.[2]. The sensitivity to strain depends on the states and polarization orientations as well as the composition range. For instance, the polarization R_1 for the HH state has a very weak strain dependence in the composition range $0.50 < x < 0.52$ ($|\delta R_1/R_1| < 2\%$); however, R_2 for the LH state has a more significant strain effect ($|\delta R_2/R_2|$ can be as large as 30% in the same x -range). The HH state would show a stronger strain effect if the polarization ratio between the [001] and [110] directions, for instance, was measured. Fig.4 shows the x -dependence of the polarization ratio for the HH and LH states. In general, for the samples grown with the intention of being lattice-matched to the GaAs substrate at room temperature or the growth temperature, small deviations in composition will not cause significant effects on the HH state polarization, but will cause more significant effects on that of the LH and SO states.

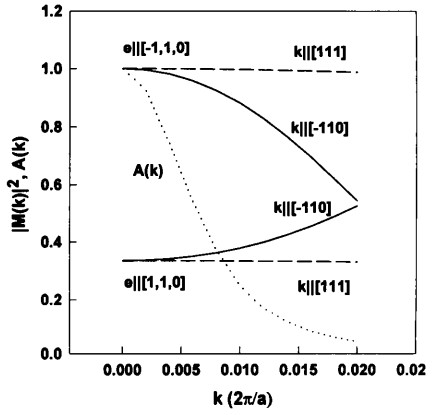
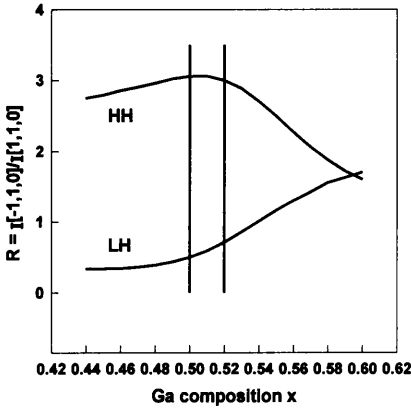


Fig.4. Polarization as a function of composition for the HH and LH states.

Fig.5. Matrix element $M_{cv}(\mathbf{k})$ and Fourier transform $A(\mathbf{k})$ as a function of \mathbf{k} .

(2) The effect of substrate misorientation: When the substrate is tilted toward one of the three directions, $[111]_A$, $[111]_B$ and $(110)_{AB}$ (a direction between the other two), by an angle $\delta\theta$, the polarization of the HH state is given as follows for the three tilted directions, respectively:

$$R_{1[111]A} = 3\left[1 - \frac{1}{3}\sin^2(\delta\theta)\right] \quad (1)$$

for the $[111]_A$ misorientation;

$$R_{1[111]B} = \frac{3}{1 + \sqrt{2}\sin(2\delta\theta) + \sin^2(\delta\theta)} \quad (2)$$

for the $[111]_B$ misorientation;

$$R_{1(110)AB} = \frac{2 + \cos(\delta\theta) - \sin(\delta\theta) + \frac{1}{2}\sin(2\delta\theta)}{2 - \cos(\delta\theta) + \sin(\delta\theta) + \frac{1}{2}\sin(2\delta\theta)} \quad (3)$$

for the $(110)_{AB}$ misorientation.

The $\delta\theta$ -dependence for the three misorientations is shown in Fig.3. For a typical value of $\delta\theta = 6^\circ$, $R_{1[111]A} = 2.995$, $R_{1[111]B} = 2.30$ and $R_{1(110)AB} = 2.47$. Note that the ratio 2.30 for the $[111]_B$ misorientation is exactly the same as that of the moving peak shown in Fig.2, which is supportive of its assignment as a band-to-band transition made in Ref. [9]. The effect of the tilted substrate is relatively weak for the LH state.

(3) Excitonic effect: The optical transition matrix element for an excitonic state at $k_{ex} = 0$ (k_{ex} is the wave vector of the excitonic state) is given as [10]:

$$|M_{ex}(k_{ex} = 0)|^2 = \left| \sum_{\mathbf{k}} A(\mathbf{k}) M_{cv}(\mathbf{k}) \right|^2 \tag{4}$$

where $M_{cv}(\mathbf{k}) = \langle \varphi_c(\mathbf{k}) | \mathbf{e} \cdot \mathbf{p} | \varphi_v(\mathbf{k}) \rangle$ is the matrix element for the direct band-to-band transition, and $A(\mathbf{k}) = \sum_{\mathbf{x}} \exp(-i\mathbf{k} \cdot \mathbf{x}) F_1(\mathbf{x})$ is the Fourier transform of the ground state exciton wavefunction $F_1(\mathbf{x})$. $A(\mathbf{k})$ is usually localized at $k = 0$, which is the justification for the effective mass approximation. If we assume that $M_{cv}(\mathbf{k})$ is a smooth function of \mathbf{k} , $|M_{ex}(k_{ex} = 0)|^2 \approx |M_{cv}(0)|^2 |F_1(0)|^2$, that is, the polarization of an excitonic transition is the same as that of the band-to-band transition at $k = 0$. However, in the Cu-Pt ordered alloys, the matrix element $M_{cv}(\mathbf{k})$ shows rather strong \mathbf{k} -dependence within the \mathbf{k} -range where $A(\mathbf{k})$ is significant. $M_{cv}(\mathbf{k})$ has been calculated by an eight-band $\mathbf{k} \cdot \mathbf{p}$ model [11] and $A(\mathbf{k})$ obtained from the exciton wavefunction $F_1(\mathbf{x}) = (a^2 b \pi)^{-1/2} \exp(-\sqrt{(x^2 + y^2)/a^2 + z^2/b^2})$ [12]. Fig.5 shows the \mathbf{k} -dependence of $M_{cv}(\mathbf{k})$ and $A(\mathbf{k})$ in two typical \mathbf{k} directions for the HH band. $M_{cv}(\mathbf{k})$ has a strong \mathbf{k} -dependence for \mathbf{k} in the plane perpendicular to the ordering direction, but a very weak dependence for \mathbf{k} parallel to the ordering direction. With the \mathbf{k} -dependence of $M_{cv}(\mathbf{k})$ and $A(\mathbf{k})$ taken into account, the polarization ratio for the excitonic transition becomes 2.8 instead of 3.0. Furthermore, for the case of a 6° [111]B misorientated substrate, the polarization ratio becomes 2.1, in good agreement with the experimental result shown in Fig.2. For the excitonic state derived from the LH state, the corresponding matrix element $M_{cv}(\mathbf{k})$ also has a strong \mathbf{k} -dependence, thus the excitonic effect is expected to be important as well.

CONCLUSIONS

To obtain quantitative information about ordering from optical polarization measurements, the following aspects should be considered: (1) strain caused by lattice mismatch, (2) substrate misorientation, (3) excitonic effect.

In particular for the polarization ratios between the $[\bar{1}10]$ and $[110]$ directions, the strain effect is weak for the HH state but relatively strong for the LH state; the effect of the substrate misorientation is strong for the HH state but weak for the LH state; the excitonic effect is significant for both HH and LH states.

ACKNOWLEDGMENTS

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