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Cathodoluminescence of $Ga_{1-x}In_xAs_ySb_{1-y}$ epitaxial layers

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Quaternary $Ga_{1-x}In_xAs_ySb_{1-y}$ layers lattice matched to a GaSb substrate are of interest for applications in the infrared range. In this work, the luminescence of $Ga_{1-x}In_xAs_ySb_{1-y}$ layers grown on GaSb by metal organic vapor phase epitaxy (MOVPE) with different In and As contents, has been studied by cathodoluminescence (CL) in a scanning electron microscope (SEM). CL images show a cellular structure which indicates the presence of dislocations decorated by recombination centers. Band gap values of 0.735 eV and 0.717 eV were measured from the CL spectra of two samples with diferent In content. This result is analyzed in the frame of existing theoretical models relating band gap with In and As content, in quaternary lattice matched layers. The band gap values are in good agreement with results of atomistic pseudopotential calculations.

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1. Introduction

Gallium antimonide and its related ternary and quaternary compounds are of increasing interest for long wavelength lasers, infrared detectors or thermo-photo-voltaic applications. In particular, quaternary GaInAsSb layers lattice matched grown on a GaSb substrate, have band gap energies, depending on the In and As content, in the approximate range 0.3 eV – 0.8 eV which is interesting for optoelectronic applications. In and As concentrations in $Ga_{1-x}In_xAs_ySb_{1-y}$ layers do not have independent values for lattice matched conditions on a GaSb substrate but there is a relationship between the In fraction, x, and the As fraction, y. A Vegard-like approximation [1] leads to the relationship y = 0.91x, while other, non-linear, relationship has been obtained by using an atomistic approach [2]. In both cases, diferent equations relating the band gap, E_g, to the composition x are obtained. For a certain composition of the layer, the estimated values of the band gap energy, using the mentioned approaches can show marked differences. In a previous paper [2] cathode luminescence of InGaSb was studied by SEM regarding the nature and distribution of defects that influence the luminescence. In this work cathodoluminescence (CL) in the scanning electron microscope (SEM) has been used to get information on the band gap energy and the presence of defects in $Ga_{1-x}In_xAs_ySb_{1-y}$ epitaxial layers with different compositions. The experimental results are compared with the band gap values estimated from the theoretical treatments of refs [1] and [2]. Quantitative comparison is limited by the fact that the equations refer to lattice matched conditions while the experimental results can be influenced by a certain mismatch, defects or some inhomogeneity arising during growth.

2. Experimental

$Ga_{1-x}In_xAs_ySb_{1-y}$ layers with a thickness of 300 nm, were grown on commercial Te doped (100) GaSb substrates, with carrier concentration of about 10^{17} cm^{-3}, from Wafer Technology, with 2º off orientations towards (110) at 575 ºC in an industrial size MOVPE reactor. The reactor pressure was 100 mbar and the growth rate about 1.5 μm/h. Details on the growth procedure have been reported elsewhere [4]. The layers exhibit a mirror-like surface morphology. The In and As content were measured by Rutherford Backscattering (RBS) and particle induced X-ray analysis (PIXE) [5] and the data are listed in Table 1. In the case of sample 1, layers with 2º off orientation towards (111)A and (111)B, labelled 1A and 1B respectively, have also been grown. An undoped GaSb layer grown on GaSb substrate has also been used as reference sample. The CL measurements were performed in a Hitachi S-2500 or a Leica 440 SEM at a temperature of 90 K and with electron beam energy of 15 keV, by using a cooled ADC infrared germanium detector.

Table 1. Data of the investigated samples.

Sample	Substrate orientation	y (As)	x (In)
1	(100) 2deg (110)	0.09	0.11
2	(100) 2deg (110)	0.17	0.18

3. Results and discussion

Secondary electron images of the samples are featureless and are not shown here. CL images of all samples investigated have a similar appearance showing a

not well defined cellular structure, with dark cells and bright walls (Fig. 1). The CL intensity at the walls is not uniform and some areas of enhanced intensity are observed. The cellular CL contrast has been often reported for other semiconductors e.g. [6] and is explained by a higher dislocation density in the walls. The dislocations getter nonradiative centers, impurities and/or point defects, causing the formation of areas with enhanced CL intensity surrounding the dislocations. The CL images do not show misfit dislocations, which are observable in mismatched structures as a cross-hatch pattern of dark straight lines. The dislocation related cell structure observed in these layers, has not been found in CL investigations of GaSb wafers, undoped or doped with different elements [7-9], of bulk GaInSb [10] and of AlGaSb epitaxial layers [11]. Possibly the relatively high In content of the layers of this work, and the presence of As, favours the formation of the luminescent cell structure.

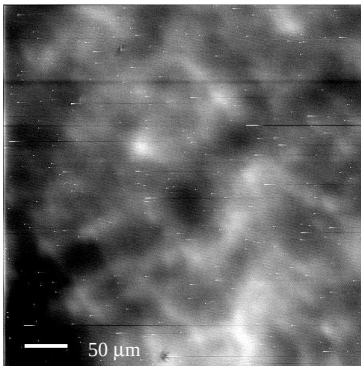

Fig. 1. Representative CL image from the quaternary layers showing the cellular-like structure.

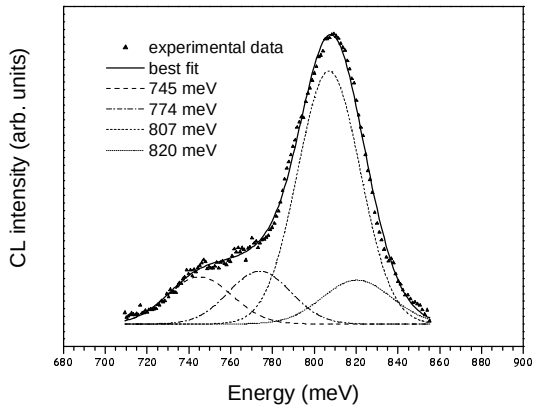

Fig. 2. CL spectrum of the reference GaSb layer.

Fig. 2 shows the CL spectrum of the reference GaSb layer. Gaussian deconvolution of the spectrum shows the presence of different well known emissions of GaSb. In addition to the dominant near band gap emission at about 807 meV and a weak above band gap band attributed to tail states, the native acceptor band (usually called band A) at about 770 meV and a band at 745 meV are also observed.

In the CL spectra of the quaternary layers the main band shifts to lower energies as expected in layers with In and As content. Fig. 3 shows the spectrum of sample 1. Gaussian deconvolution shows that a main band is peaked at 735 meV, which corresponds to the band gap emission of the alloy, and that weaker contributions, arising from the substrate, are present at higher energies. Samples 1, 1A and 1B, which have the same nominal In and As content, present dominant bands at 735 meV, 737 meV and 735 meV respectively, showing a correspondence of nominal composition with band gap energy measured by CL. In sample 2, with higher In content, the CL spectrum is shifted towards lower energies, and by deconvolution, a band at 717 meV is observed, which is attributed to the band gap of the quaternary layer.

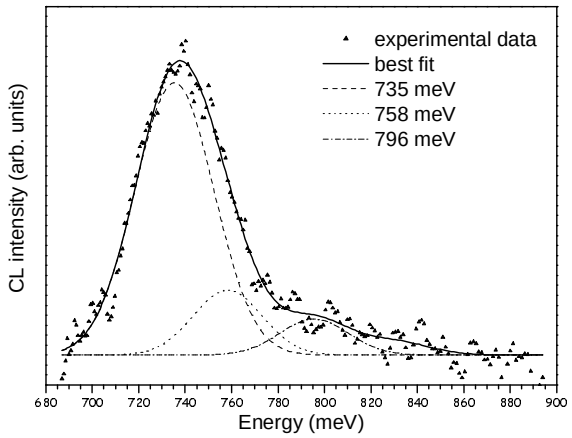

Fig. 3. CL spectrum of sample 1, which shows the main peak at 735 meV.

The relationship of In and As content (y = f(x)), with the energy band gap of GaInAsSb lattice matched to GaSb, has been investigated by several authors e.g. [3,12-14]. For lattice matched systems, the quaternary may be represented as a combination of two lattice-matched constituents, one of which must be a ternary while the other may be either a binary or a ternary. In our case, the system is $(GaSb)_{1-x}(InAs_{0.91}Sb_{0.09})_x$ and the results for the band gap energy lead to suggest different values of the bowing parameter, C. In the review of Vurgaftman *et al.* [1] the authors recommend a value of C=0.75 which is a composite obtained by averaging all the available results. In order to investigate the relation of the In and As content in our layers with the energy gap measured by CL we have used the dependence of E_g on x for $(GaSb)_{1-x}(InAs_{0.91}Sb_{0.09})_x$ system:

$$E_g\,(GaInAsSb) = (1-x)\,E_g(GaSb) + x\,E_g(InAs_{0.91}Sb_{0.09}) - x\,(1-x)\,C \quad (eV) \qquad (1)$$

with the energy band gaps at 90 K [1]. The x values obtained from eq. (1) listed in Table 2 differ by a factor of 2 or higher, from the x values measured by RBS/PIXE. Although no mismatch dislocations have been observed in the CL images, the possible deviation of the ideal ratio, y = 0.91 x, for lattice matching conditions, could certainly lead to errors in the determination of x by using the expression (1). However, the large deviation suggests that the equation (1) underestimates the x values.

Table 2. Experimental E_g and x and theoretical x values for the different samples.

Sample	Eg (CL) meV	x (RBS/PIXE)	x (eq.(1))	x (eq.(2))
1	735	0.11	0.05	0.11
1A	737	No data	0.055	0.108
1B	735	No data	0.05	0.11
2	717	0.18	0.07	0.14

For this reason the approach proposed by Magri et al. [3] has been considered. The atomistic elasticity via the Valence Force Field approach was used in [3] to obtain the function $y = 0.001 + 0.648x + 0.239x^2$, at which the quaternary alloy is lattice matched to GaSb. Atomistic pseudopotential calculations were used to study the evolution of the fundamental band gap as a function of the composition and the expression:

$$E_g = 0.804 - 0.5257x - 0.4322x^2 + 0.4175x^3 \quad \text{(eV)} \quad (2)$$

was obtained at 0 K. We have estimated the energy value at 90 K through the Varshni form by using a linear interpolation of the α and β parameters of GaSb and $InAs_{0.91}Sb_{0.09}$ [3]. The x values obtained from the above equation, by using the energy gap measured by CL, are listed in Table 2. Fig. 4 summarizes the results displayed in the Table 2. The lines correspond to the evolution of the bandgap with the In content according to eq. 1 (dashed line) and eq. 2 (solid line) at 90 K. The squares correspond to the value of the energy gap, 735 meV, in sample 1 and the circles to the the energy gap, 717 meV, in sample 2. In the x axis the In contents of both samples, measured by RBS, are marked for reference. It can be observed that the x values obtained from the dashed line (equation 1) are far from the experimental ones. On the contrary, the x value of sample 1, obtained from the solid line (equation 2) match exactly with the experimental value. This shows that the theoretical treatment of [3] appears suitable to describe the relationship between E_g, x and y in the samples studied in this work. In the case of sample 2, the agreement is clearly better when equation 2 is used. However, in this case the discrepancy is about 22 % which can be partly explained by the fact that sample 2 has a x/y ratio far from the condition given by the above y = f(x) function. In addition the theoretical treatment corresponds to perfectly random conditions and does not consider effects such as short range or defects.

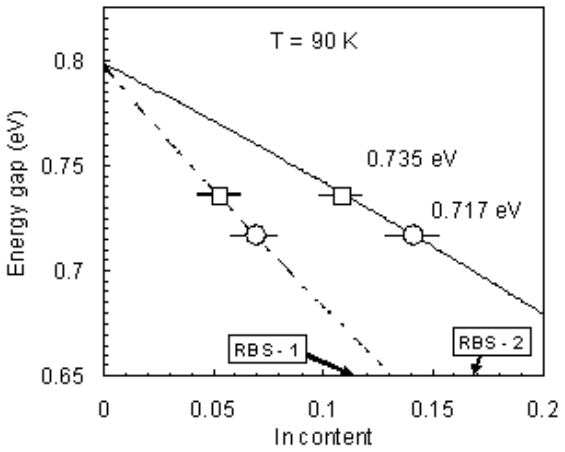

Fig. 4. Band gap energy of $Ga_{1-x}InAs_ySb_{1-y}$/GaSb as a function of the In content x extrapolated at 90 K. The dashed line is the quadratic function (equation 1) and the solid line corresponds to equation 2. The squares represent the energy gap measured by CL in sample 1 and the circles correspond to the energy gap of sample 2. The x values measured by RBS in both samples are marked, for comparison, in the In content axis.

4. Conclusions

$Ga_{1-x}InAs_ySb_{1-y}$ layers grown on GaSb by MOVPE with different x and y values, have been characterized by CL in the SEM. CL images show a cellular structure with bright cell walls related to gettering of defects by dislocations but no mismatch dislocations are observed. Band gap values of the layers, between 717 meV and 737 meV are obtained from the CL spectra. The results show good agreement with a theoretical relationship between E_g and In content in lattice matched layers.

Acknowledgements

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References

[1] I. Vurgaftman, J. R. Meyer, L. R. Ram-Moahn, J. Appl. Phys. **89**, 5815 (2001).
[2] M. F. Chioncel, C. Diaz-Guerra, J. Piqueras, N. Duhanian, T. Duffar, J. Optoelectron. Adv. Mater, **6**(1), 237 (2005).

[3] R. Magri, A. Zunger, H. Kroemer, J. Appl. Phys. **98**, 043701 (2005).
[4] A. Amarei, E. K. Polychroniadis, F. Dimroth, A. W. Bett J. Cryst. Growth **275**, el229 (2005).
[5] V. Corregidor, N. P. Barradas, E. Alves, N. Franco, L. C. Alves, P. C. Chaves, M. A. Reis, Ion beam analysis of GaInAsSb films grown by MOVPE on GaSb Nucl. Instrum. Methods Phys. Res. B (in press).
[6] B. Wakefield, P. A. Leigh, M. H. Lyons, C. R. Elliot Appl. Phys. Lett. **45**, 66 (1984).
[7] B. Méndez, P. S. Dutta, J. Piqueras, E. Diéguez, Appl. Phys. Lett. **67**, 2648 (1995).
[8] P. Hidalgo, B. Méndez, P. S. Dutta, J. Piqueras, E. Diéguez, Phys. Rev. B **57**, 6479 (1998).
[9] P. Hidalgo, B. Méndez, J. Piqueras, P. S. Dutta, E. Diéguez, Semicond. Sci. Technol. **14**, 901 (1999).
[10] M. F. Chioncel, C. Díaz-Guerra, J. Piqueras, J. Vincent, V. Bermúdez, E. Diéguez, J. Crys. Growth **268**, 52 (2004).
[11] J. Storgårds, B. Méndez, J. Piqueras, M. Chenot, F. Dimroth, A. W. Bett, J. Phys. Condens. Matter **16**, S251 (2004).
[12] F. Karouta, H. Mani, J. Bhan, F. J. Hua, A. Joullie, Rev. Phys. Appl. **22**, 1459 (1987).
[13] M. C. Wu, C. W. Chen, J. Appl. Phys. **71**, 6116 (1992).
[14] S. Iyer, S. Hegde, K. K. Bajaj, A. Abul-Fadl, W. Mitchel, J. Appl. Phys. **73**, 3958 (1993).

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