Optical Characteristics of Dilute Nitride of InSb Bulk Crystal

Manisha Joshi Deshpande[1], Dilip Maske[2], BrijMohan Arora[3], Dattatray Gadkari[4]

[1] Jai Hind College, Churchgate, Mumbai, Mithibai College, Vile Parle (W), Mumbai, India
[2] Ruparel College, Mahim, Mumbai, India, Mithibai College, Vile Parle (W), Mumbai, India
[3] I. I. T. B. Mumbai, India
[4] Mithbai College, Vile Parle (W), Mumbai, India

Abstract- Addition of highly electronegative Nitrogen to Indium Antimonide substantially reduces its band gap. Theoretically, it is expected that one percent of nitrogen should reduce the band gap by about 100meV. Thus by reduction in the bandgap, the resulting material will be suitable for 'far infrared' detection. Three bulk crystals of InSb:N were grown by vertical directional solidification technique. The grown ingots were found detached from the ampoule wall. The physical properties and microstructures are studied. The smooth appearance of the crystal, uniform diameter across the length and other studies confirm the excellent quality of the crystal. Wavelength dispersive spectroscopy (WDS) was carried out for compositional analysis. The analysis confirmed the successful incorporation of nitrogen in the compound. The graph of absorption vs. energy was plotted to study shift in energy band gap. The deviation in the shift to the smaller wavelength is attributed to high (~1018/cm3) carrier concentration. The corresponding Moss-Burstein shift in the energy gap is calculated. The calculated values of the energy band shift due to all effects are summed up and compared with the observed value.

Index Terms- InSb, bulk crystal, energy band gap, Moss-Burstein effect, dilute nitride

I. INTRODUCTION

The materials used for study of the 8-12 μm atmospheric transmission band are limited in number and they have their own limitations. The addition of arsenic to InSb leads to band gap reduction but large amount of arsenic is required to achieve the result. Large amount of arsenic can affect the quality of crystals. Mercury cadmium telluride is another option. However, HgCdTe has inherent problems of thermal stability and poor compositional uniformity over the large areas, and the material parameters change with time. [1] The addition of nitrogen drastically reduces the band gap of InSb. One atomic percent of nitrogen reduces the band gap of InSb by 100 meV. [2, 3] Energy band gap of Indium antimonide is 180 meV at 300K. (Cutoff wavelength 7μm at 300K and 5.5μm at 80K) With a very small addition of nitrogen, the band gap of InSb can be manipulated to suitable lower value which will make the material useful for 8-12 μm atmospheric range. Indium antimonide bulk crystals grown by Vertical Directional Solidification (VDS) technique are found to have good quality [4, 5]. Defect density of such crystals is low and crystals are found detached from the ampoule wall at the end of growth process. This method is suitable to grow dilute nitrides for such as in VDS technique continuous flow of nitrogen or presence of nitrogen plasma [3] is not required. Using this method three bulk crystals of dilute nitride Indium Antimonide are grown with nitrogen atomic percentage of 0.1% and 0.2% and 0.5% respectively.

II. MATERIALS AND METHODS

Three ingots of dilute nitride of Indium antimonide with percentage of nitrogen as 0.1%, 0.2% and 0.5% are grown by VDS technique. The VDS system consists of a vertical single zone furnace. The advantage of VDS-technique in addition to detachment of ingot is that the unidirectional growth process can control the crystal nucleation. The process also eliminates the need of seed inside the ampoule. The Indium, Antimony and Indium nitride powder were sealed in a quartz ampoule at pressure 10-3 bar. Before sealing, argon gas is flushed more than 10 times. Very small amount of nitrogen changes the band gap of InSb. For percentage above 1, the material will have negative band gap. Therefore the exact percentage of Indium nitride powder was added to Indium and Antimony. High quality (In, Sb) (6N) were used as the source materials.

Each growth was carried out as follow. The material was kept at temperature 8000C about 20 hours for synthesis and homogeneous mixing of melt composition. The phase diagram of ternary material InSbN is not available. The growth was started at 5000C. The ampoule was lowered at rate 3mm/hr with temperature gradient in the range of 18oC/cm to 20oC/cm. These values are optimized to ensure quality growth. [6] To ensure uniformity in the composition, ampoule was rotated with the speed 10 rpm. At 3500C, the downward movement of the crystal was halted. The crystal was kept at this temperature for 10 hours. The ingot was found detached from the quartz ampoule on growth. As a result of which it was easy to remove from the quartz ampoule. The process of removal of the ingot from the ampoule is shown in figures1. This detached growth in closed quartz ampoule is an indication of good quality crystal growth [7].

We got ingot of about 10 mm diameter which is slightly smaller than inner diameter of the quartz ampoule and 30mm length for each growth. The surface of the crystal was rough. The ingot had conical shape at one end. On the other side i.e. at the end point of growth the ingot had concave shape. The figure 1 shows the ingot with nitrogen percentage 0.2%.

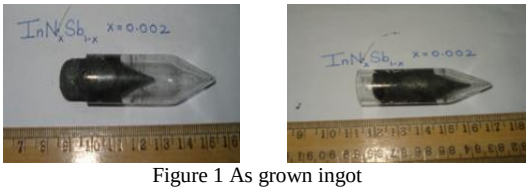

Figure 1 As grown ingot

III. RESULTS AND DISCUSSION

A. Compositional Analysis

Compositional analysis is done using WDS (Wavelength diffraction spectroscopy which is more accurate than electron energy dispersive X ray analysis), at IIT, SAIF. The instrument is Field Emission Gun-Scanning Electron Microscopes (FEG-SEM) with Model JSM-7600F with Resolution1.0 nm (for 15 kv). The incorporation of nitrogen is very uniform at weight percentage 0.25 %, decreasing only at end. But other ingots, where 0.1% and 0.5% of Indium nitride was added, showed non uniformity over the nitrogen content after growth.

B. Optical characterization

As by adopting the above practices all major constructs of a research paper can be written and together compiled to form a complete research ready for Peer review. Optical measurements are carried out on wafers from each growth and compared with undoped InSb wafer. The transmission is less than undoped InSb as each wafer is thick. The wafers are of thickness 0.5 mm.

The instrument used for the purpose of FTIR is PerkinElmer Spectrum-65. Absorbance versus energy graphs for each wafers are plotted to determine the energy band gap in each case. For sample with 0.1%, the band gap reduces to 0.168 eV and for the sample with percentage 0.5%, the band edge is at 0.180 eV. From figure2, the transmission of InSb is reduced to 30% where as transmission of InSbN samples show 50% drop in transmission percentage due to presence of defects.

The absorption coefficient is also defined as the ratio of the absorbed photon energy per unit volume and unit time to the energy- flux density.

$$T = I/I_0 = (1 - r) \quad \exp(-\alpha x) \qquad (1)$$

Where T is transmittance, r is the light reflection coefficient which is calculated from known value of refractive index (=4) of InSb. The transmittance v/s wavenumber graph plotted for different values of nitrogen content ranging from zero to 0.005. (Figure 2)

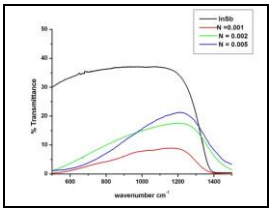

Figure 2 Graph of Transmittance v/s Wavenumber

In case of high defect density, similar type of band tails are observed, which is commonly described as Urbach tail. The direct way is to measure absorption coefficient below the energy gap. The exponential increase at the absorption edges is given by Urbach rule (equation 2).

$$\alpha(h\nu) = \alpha_0 \exp[(h\nu - E_0) / Eu] \qquad (2)$$

Where α_0 and E_0 are material parameters and E_u is the Urbach energy describing the width of the exponential absorption edge [8].

Shift in energy band gap towards higher energy is observed in all three samples as mentioned in the table1. This effect is observed due to high carrier concentration. The apparent shift towards lower wavelength is calculated as Moss-Burstein shift. The value of energy shift is given by the formula (eq. 3)

$$E_F = \frac{\eta^2 k_F^2}{2m_c} = \frac{\eta^2}{2m_c}\left(3\pi^2 n\right)^{2/3} \qquad (3)$$

Where n is carrier concentration and the m_c (value of effective mass of an electron) is calculated using following formula (equation 4)

$$m_c^2 = \frac{ne^3}{\varepsilon_0 C^3 n_\tau (2\pi)^2 \mu (\frac{\alpha}{\lambda^2})_{slope}} \qquad (4)$$

Where m_e is the effective mass of electron, C is speed of light, n_τ is refractive index of InSb, μ is mobility of electrons and α/λ^2 is found from slope of the graph (as shown in figure 3) of absorption versus wavelength square [9]. The carrier concentration and mobility values are obtained from Hall Effect and resistivity by Van der Pauw method.

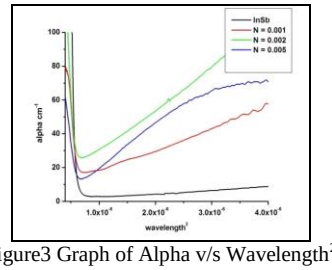

Figure3 Graph of Alpha v/s Wavelength^2

The result of calculations for energy band gap, Urbach tail and Moss-Burstein energy shift is tabulated in the table 1.

$InSb_{1-x}N_x$ x =	Carrier conc. $X10^{18}$ /cm^3	Mobility $cm^2/V.s$	Band gap meV	Urbach energy meV	Moss Burstein energy meV
0.001	0.5	3836	168.7	18.4	23
0.002	1	7800	172.5	16.9	46.7
0.005	0.5	3530	180	31	39.6

Table 1 Energy Band Gap Calculation

IV. CONCLUSION

Thus we can conclude that there was successful incorporation of nitrogen in the dilute nitride of Indium Antimonide grown by VDS technique. The energy band gap is calculated by plotting graph of absorption in cm-1versus energy in eV. From the graph Energy changes due to Urbach Tail and Moss-Burstein shift is also calculated and tabled. These effects are attributed to large carrier concentration. From table1energy band gap reduction can be seen for sample 2 with nitrogen content 0.002% whereas sample with nitrogen content does not show much reduction. This can be due to non uniform distribution of nitrogen in sample with nitrogen content as 0.5%.

ACKNOWLEDGMENT

I am thankful to all those who helped me in characterizing the samples. WDS has been done at TIFR and IITB, SAIF center.

REFERENCES

[1] M. Oszwaldowski, T. Berus, J. Szade, K. Jozwiak, I. Olejniczak, P.Konarski. "Structural properties of InSbBi and InSbAsBi thin films prepared by the flash-evaporation method", Crystal Res. Technology. 36 (2001) 8-10 1155-1171

[2] T. Ashley, T. M. Burke, G. J. Ryce, A. R. Adams, A. Andeev, B. N. Murdin, E. P. O'Reilly, C. R. Pidgeon. "InSb1-x Nx growth and devices" Solid State Electronics.47 (2003) 387-394

[3] T. D. Veal, I. Mehboob and C. F. McConville. "Negative Band gaps in Dilute InNxSb1-x Alloys." Physical Review Letters. Vol. 92, no. 13. (2004)

[4] D. B. Gadkari, B M Arora, Transaction of MRS- Japan 34(2) (2009) 571-574

[5] D. B. Gadkari, K. B. Lal, and B. M. Arora Indian Patent number: 139/BOM 1999 and the Gazette of India No. 8 Feb 21, 2004 patent number 192132,

[6] D. S. Maske, P. S. More, M. D. Deshpande, R. Choudhary and D. B. Gadkari, Archives of Physics Research, "Characterization of InSbBi Bulk Crystals Grown at Various Growth-rates by Vertical Directional Solidification (VDS) "2012, 3 (1):15-20

[7] D. B. Gadkari. J. Chem. Chem. Eng. 6 (2012) 250-258

[8] Physics and properties of narrow band gap semiconductors, Junhao Chu, Arden Sher pp 340,372. Springer publications

AUTHORS

First Author – Manisha Joshi Deshpande. M. Sc. (Physics) Jai Hind College, Churchgate, Mumbai, Mithibai College, Vile Parle (W), Mumbai, India, manishauj@gmail.com
Second Author – Dilip Maske M. Sc. (Physics) Ruparel College, Mahim, Mumbai, India, Mithibai College, Vile Parle (W), Mumbai, India, dsmaske@gmail.com
Third Author – Brij Mohan Arora Ph. D. I. I. T. B. Mumbai, India arora.brijmohan@gmail.com
Fourth Author – Dattatray Gadkari Ph. D. Mithbai college, Vile Parle (W), Mumbai, India dr_gadkari@yahoo.com

Correspondence Author – Manisha Joshi Deshpande. manishauj@gmail.com, contact number 9819210188.