Temperature dependence of photocurrent spectra for $AgInS_2$ epilayers grown by hot wall epitaxy

Seungnam Baek*, Kwangjoon Hong**

*Department of Physics, Chosun University ,** Department of material science & Engineering, Chosun University

Abstract A silver indium sulfide ($AgInS_2$) epilayer was grown by the hot wall epitaxy method, which has not been reported in the literature. The grown $AgInS_2$ epilayer has found to be a chalcopyrite structure and evaluated to be high quality crystal. From the photocurrent measurement in the temperature range from 30 K to 300 K, the two peaks of A and B were only observed, whereas the three peaks of A, B, and C were seen in the PC spectrum of 10 K. These peaks are ascribed to the band-to-band transition. The valence band splitting of $AgInS_2$ was investigated by means of the photocurrent measurement. The temperature dependence of the energy band gap of the $AgGaSe_2$ obtained from the photocurrent spectrum was well described by the Varshni's relation, $E_g(T) = E_g(0)$ eV-(7.78×10^{-4} eV/K)$T^2/(T + 116$ K K). Also, Eg(0) is the energy band gap at 0 K, which is estimated to be 2.036 eV at the valence band state A and 2.186 eV at the valence band state B.

Keywords: silver indium sulfide ($AgInS_2$) epilayer, photocurrent spectrum, energy band gap, hot wall epitaxy

1. Introduction

$AgInS_2$ has been known to be a ternary compound semiconductor, of which band gap is wide and belongs to the visible region of the spectrum. Ordinary, ternary chalcopyrite crystals are currently of technological interest since they show promise for application in the areas of visible and infrared light-emitting diodes, infrared detectors, optical parametric oscillators, upconverters, and far infrared generators. of $AgInS_2$, the splitting of the valence band was known to be dominated by the uniaxial lattice compression. Therefore, the uppermost valence bands of $AgInS_2$ are profoundly influenced by the proximity of the d levels of the valence band of the noble-metal [1-3]. Then, both p and d orbital of $AgInS_2$ are known to be partially lifted due to the degeneracy. These states participate in the photoconduction process, resulting in extending the photoresponse on the higher energy side. Such studies were carried out for a single crystal of the chalcopyrite $AgInS_2$ that was grown by chemical transport and for others grown by Bridgman method. However, optical properties and the epitaxial growth of $AgInS_2$ have not been well understood. Only several researchers ,have investigated the electrical properties and the polycrystalline $AgInS_2$ film growth. In this paper, we first tried to grow the $AgInS_2$ epilayers using hot wall epitaxy (HWE) which is designed specifically to grow epilayers under the condition of the close thermodynamic equilibrium. The electric and optical measurements of the grown $AgInS_2$ epilayer have been carried out at the temperature ranging from 10 to 300 K. Also, the valence band splitting and the temperature dependence of energy band gap have been determined with using the photocurrent (PC) spectra.

2. Experimental Procedure

The starting materials were used as the shot-type Ag, In, and S to grow the $AgInS_2$ epilayers and the purity of

these materials is 6N. After being weighed in stoichiometric proportions, those materials were sealed in a quartz tube whose lining was coated with carbon. The sealed ampoule was placed in a synthesis furnace and continuously rotated at a rate of 1 revolution per minute. In order to avoid the explosion of the ampoule due to the high sulfur vapor pressure, the temperature of the ampoule was gradually increased to 1050 ℃. This temperature was then maintained for 48 hours. After the growth of the polycrystalline $AgInS_2$ ingot, the $AgInS_2$ epilayers were grown on semi-insulating (100) GaAs by HWE method using the polycrystalline $AgInS_2$ ingot as source materials [12].

3. Results and Discussion

3.1 Temperature Dependence of the Photocurrent Energy Band Gap

Fig. 1 displays the variation of the energy band gap of $AgInS_2$ and the energy gap of the A and B peak obtained from the PC peaks as a function of temperature. As shown in Fig. 1, the energy gap variation of the A and B peaks corresponding to the split-levels of the $\Gamma_4(z)$ and $\Gamma_5(x)$ valence band shows a nonlinear relationship. Generally, the energy gap of these peaks varies proportionately to the square of the temperature when the measurement temperatures is much lower than the Debye temperature, whereas the energy gap varies linearly with the temperature when the measurement temperature is much higher than the Debye temperature. The energy band variation of these peaks as a function of temperature is well fitted numerically by the following formula:

$$E_g(T) = E_g(0) - \alpha T^2/(\beta + T), \quad \text{------------------------} \quad (1)$$

where α is a constant and β is approximately the Debye temperature. When α and β are taken to be 7.78×10^{-4} eV/K and 116 K, respectively, the curve plotted by eq. (1) closely fits the experimental values, as shown in Fig. 1. The Debye characteristic temperature $\theta_\perp$ [18] of $AgInS_2$ was found to be 117 K at 300 K and reasonable agreement with our result. Also, Eg(0) is the energy band gap at 0 K, which is estimated to be 2.036 eV at the valence band state A and 2.186 eV at the valence band state B. The energy band gap at 300 K fitted by eq. (1) was 1.868 eV. This band gap energy shows slightly smaller than the value obtained from the photoconductivity by Joshi et al., which is 1.91 eV at 300 K. But our observation is in good agreement with the value measured at 300 K by Shay et al., which was found from the electroreflectance measurement. Generally, the band gap energy of $AgInS_2$ at room temperature is known to be 1.87 eV.

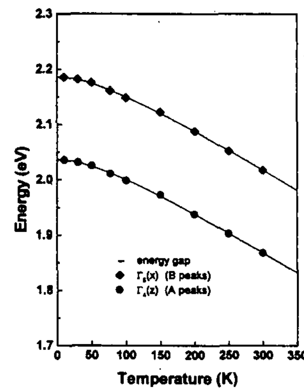

Fig. 1. Experimental values of the photocurrent peak energies and the energy band gap as a function of temperature.

4. Conclusions

The $AgInS_2$ epilayers on the GaAs substrate have been first grown with HWE method. The temperature dependence of the energy band gap for $AgInS_2$ was also first obtained from the PC measurement. The variation of the energy band gap with temperature was well described by the equation of $E_g(T) = Eg(0) - (7.78 \times 10^{-4})T^2/(116 + T)$. Also, Eg(0) is estimated to be 2.036 eV at the valence band state A and 2.186 eV at the valence band state B.

References

[1] C.M. Joseph, C.S. Menon, Semicond. Sci. Technol. 11 (1996) 1668.