Polycrystalline GaSb thin films grown by co-evaporation*

Qiao Zaixiang(乔在祥)†, Sun Yun(孙云), He Weiyu(何炜瑜), He Qing(何青),
and Li Changjian(李长健)

(Key Laboratory of Opto-Electronic Information Science and Technology of the Ministry of Education, Key Laboratory of Photo-Electronics Thin Film Devices and Techniques of Tianjin, Institute of Photoelectronic Thin Film Device and Technology, Nankai University, Tianjin 300071, China)

Abstract: We report optical and electrical properties of polycrystalline GaSb thin films which were successfully grown by co-evaporation on soda-lime glass substrates. The thin films have preferential orientation of the (111) direction. SEM results indicate that the average grain size of GaSb thin film is 500 nm with the substrate temperature of 560 °C. The average reflectance of GaSb thin film is about 30% and the absorption coefficient is of the order of 10^4 cm^{-1}. The optical bandgap of GaSb thin film is 0.726 eV. The hole concentration shows a clear increasing trend as the Ga-evaporation-temperature/ Sb-evaporation-temperature ($T_{\rm Ga}/T_{\rm Sb}$) ratio increases. When the Ga crucible temperature is 810 °C and the antinomy crucible temperature is 415 °C, the hole concentration of polycrystalline GaSb is 2×10^{17} cm^{-3} and the hole mobility is 130 cm^2/(V·s). These results suggest that polycrystalline GaSb thin film is a good candidate for the use as a cheap material in TPV cells.

Key words: substrate temperature; band gap; co-evaporation; polycrystalline GaSb films
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1. Introduction

Optical devices which utilize GaSb layers are important for a wide variety of optoelectronic applications, especially in thermophotovoltaic (TPV) systems[1–4]. The GaSb cell is recognized as key for the development of TPV because it responds out to longer wavelengths than the silicon solar cell thus providing higher power densities in combination with manmade heat sources. The bulk crystal growth was focused on GaSb by different techniques: vertical feeding method (VFM)[5, 6] to obtain polycrystalline ingots and Czochralski (CZ)[7–9] method to obtain monocrystalline ingots. The film growth is mainly focused on quaternary GaInAsSb on GaSb substrates by MOVPE[10], MBE[11, 12] or LPE[13–15] method.

However the important problem concerning the use of GaSb in the TPV technology is the high cost of these cells, which is related to the GaSb substrates itself, the monocrystalline epitaxial layers, and the processing on the device. One possibility of reducing the wafer cost is the use of polycrystalline thin film materials and cheap substrates for the reduction of the processing cost.

In this work, polycrystalline GaSb thin films grown on cheap soda-lime glass substrates by co-evaporation are studied. X-ray diffractometer (XRD) is used for structural characterization. Scanning electron microscopy is used for topological characterization. The optical and electronic analysis is performed with the spectrophotometer and Hall technique.

2. Experiment

Thin films of GaSb were deposited on thoroughly cleaned soda-lime glass substrates at different substrate temperatures ($T_{\rm s}$) using an evaporation system. The base pressure of the chamber was better than 1×10^{-4} Pa before deposition. In this system, vapors of the individual Ga(99.9999%) and Sb(99.9999%) elementary source materials were ejected through a nozzle (a perforation on the top lid) of the crucible. The evaporation rates of source material were controlled by setting crucible temperature. The substrate rotated at 30 rpm during deposition. The deposition chamber maintained at a pressure of about 4×10^{-4} Pa. The deposition rate was maintained at 20 nm/min.

Film thickness was measured using a surface profiler (AMBIOS XP-2). The surface morphologies were observed using field emission scanning electron microscope (FESEM, JSM6700F). The structure of the films was characterized by XRD (Philips PANalytical X'Pert, CuKλ, λ = 0.154056 nm). The optical reflectance and transmission spectra of films were recorded by the UV-VIS-IR spectrophotometer (VARIAN CARY5E). Mobility and carrier concentration were measured using a Hall automatic measuring system (ACCENT HL5550 LN2, USA).

3. Result and discussion

3.1. Structural property and surface morphology of polycrystalline GaSb thin films

In Fig. 1, the XRD patterns of GaSb films with different thicknesses deposited at 500 °C are presented. The XRD patterns are recorded in the range of 2θ = 15–85 ° using CuKα radiation. The XRD spectra revealed that the thin films are polycrystalline structure. The thin films are dominated by three principal orientations: (111), (220) and (311).

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† Corresponding author. Email: zxqiao@eyou.com

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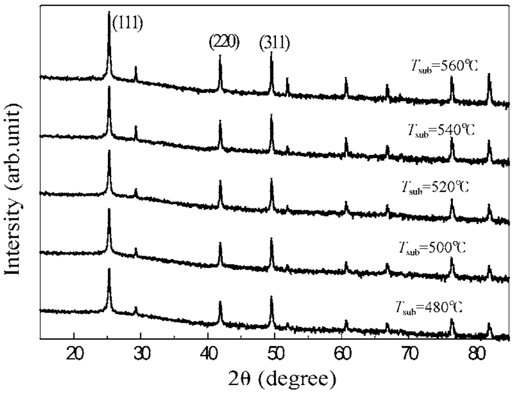

Fig. 1. XRD patterns for GaSb thin films of different substrate temperatures.

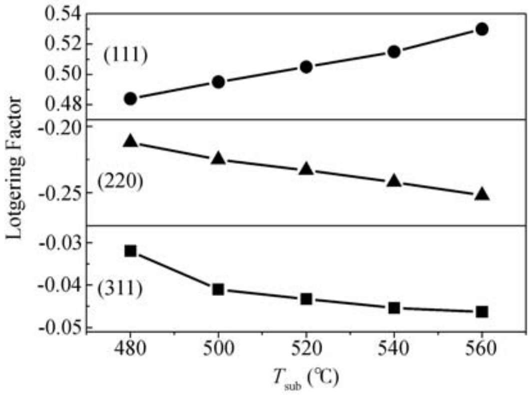

Fig. 2. Dependence of Lotgering factor of poly-GaSb samples on substrate temperature.

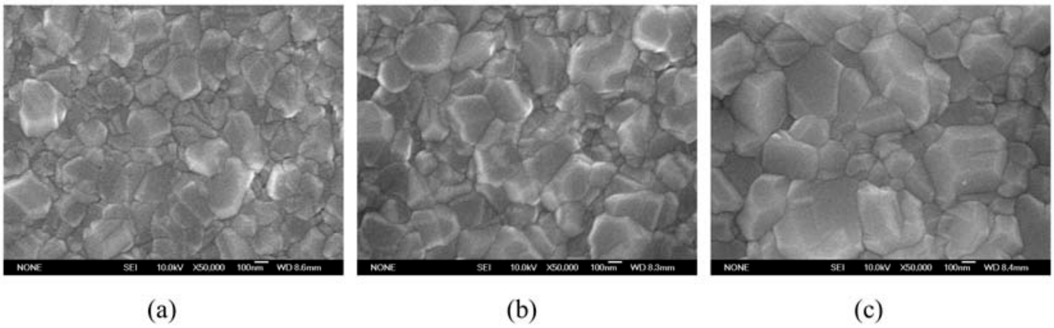

Fig. 3. SEM images of poly-GaSb samples grown at (a) 480 °C, (b) 520 °C, and (c) 560 °C.

Using the Lotgering method, the extent of orientation can be estimated by XRD intensity[16]. $\Sigma I(hkl)_{\rm ideal}$ is defined as the sum of XRD intensity of non-oriented plane, $I(hkl)_{\rm ideal}$ as the intensity of special plane, then P_0 can be derived by

$$P_0 = \frac{I(hkl)_{\rm ideal}}{\sum I(hkl)_{\rm ideal}}. \tag{1}$$

For oriented sample, define the intensity as $\Sigma I(hkl)_{\rm obsd}$ and $I(hkl)_{\rm obsd}$, and the P can be calculated by

$$P = \frac{I(hkl)_{\rm obsd}}{\sum I(hkl)_{\rm obsd}}. \tag{2}$$

Lotgering orientation factor is defined as

$$f = \frac{p - p_0}{1 - p_0}. \tag{3}$$

Figure 2 is the Lotgering orientation factor f, changed with heating temperature. It shows that, as the temperature increases from 480 to 560 °C, the Lotgering orientation factor of (111) increases linearly from 0.48 to 0.53, while the Lotgering orientation factor of (220) and (311) are negative and decreased. This result suggests that all the GaSb films have highly (111) preferential orientations.

Figure 3 shows the dependence of SEM maps at a magnification of 50 000 on substrate temperature when the thicknesses of thin films are about 1000 nm. The grains become densely packed as shown in the SEM surface micrograph. Grain size is generally inferred from the SEM. As expected grain size increases with temperature, the average grain size is 100, 250, and 500 nm at substrate temperature of 480, 520, and 560 °C. Dong[12] reported that the gain size of polycrystalline GaSb films grown by MBE at 520 °C was about 250 nm, so here the grain size of thin films grown by co-evaporation appears as large as the results grown by MBE.

3.2. Optical property of polycrystalline GaSb thin films

Figure 4 shows the transmission and reflection spectra of GaSb thin films at the substrate temperature of 560 °C. The average reflectance of GaSb thin film is about 30%, which demonstrates the index of GaSb refraction is about 3.8.

The absorption coefficient α was determined from transmission and reflectance spectra and the film thickness d, by[17]

$$\alpha = \frac{-1}{d} \ln \frac{\left[(1-R)^4 + 4T^2R^2\right]^{\frac{1}{2}} - (1-R)^2}{2TR^2}, \tag{4}$$

where T is the transmission and R is the reflectance for any intermediate photon energy. The GaSb film thickness d is 1040 nm.

Figure 5 shows the absorption coefficient of the GaSb film at the substrate temperature of 560 °C. The magnitude of absorption coefficient is high and is of the order of 10^4 cm^{-1}, which agrees with the bulk monocrystalline GaSb material data referred from Ref. [18]. The absorption coefficient increases sharply with photon energy beyond the fundamental absorption edge. The high absorption coefficient and tunable

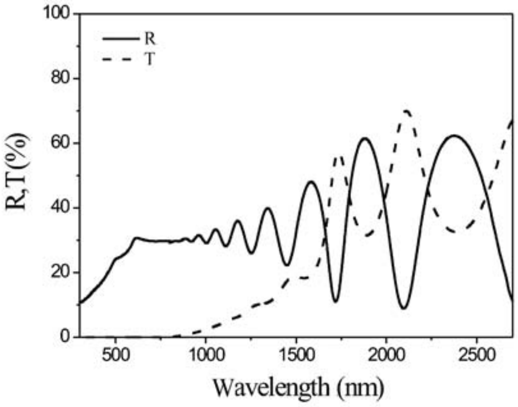

Fig. 4. Transmission and reflection spectra of GaSb thin film at the substrate temperature of 560 °C.

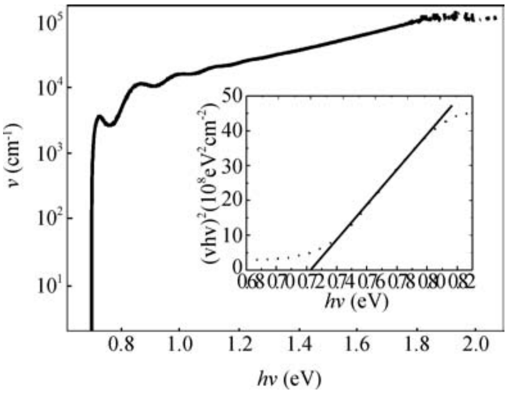

Fig. 5. Absorption coefficient of the GaSb film.

bandgap of GaSb films will be an added advantage in respect of their applications in photovoltaic devices.

As well known, GaSb is a direct gap semiconductor for which the dependence of the absorption coefficient on the radiation energy, $h\nu$, follows Tauc relation[19]:

$$\alpha h\nu = A\left(h\nu - E_{\mathrm{g}}\right)^{\frac{1}{2}}, \tag{5}$$

where h is the Planck constant, ν is the radiation frequency, d is the thickness of GaSb thin film, E_{g} is the band gap energy, and A is a constant that depends on the nature of the radiation. This relation indicates that the fundamental absorption edge of the GaSb is due to the allowed direct transitions among parabolic bands. The linear fit of $(\alpha h\nu)^2$ versus $h\nu$ allows us to obtain the value of A and E_{g}, as shown in Fig. 5. The E_{g} of polycrystalline GaSb thin film is 0.726 eV. This result suggests that GaSb film has an appropriate E_{g} for TPV application.

3.3. Electronic property of polycrystalline GaSb thin films

The polycrystalline GaSb samples were grown with varied Sb to Ga beam flux ratios at the same growth temperature of 560 °C. All the samples measured with Hall technique were p types. This behavior could be explained by Ga antisites ($\mathrm{Ga_{Sb}}$). Ga is a double acceptor since Ga has two less valence electrons than Sb[11]. Figure 6 shows the hole concentration and mobility of these samples as functions of the $T_{\mathrm{Ga}}/T_{\mathrm{Sb}}$ ratio. The hole concentration shows a clear increasing trend as the $T_{\mathrm{Ga}}/T_{\mathrm{Sb}}$ ratio increases, and the hole mobility shows decreasing trend as the $T_{\mathrm{Ga}}/T_{\mathrm{Sb}}$ ratio increases. When the Gallium crucible temperature is 810 °C and the antinomy crucible temperature is 415 °C, the hole concentration of polycrystalline GaSb is 2×10^{17} cm^{-3} and the hole mobility is 130 cm^2/(V·s). This sample is a good base layer material for the GaSb TPV cell structure.

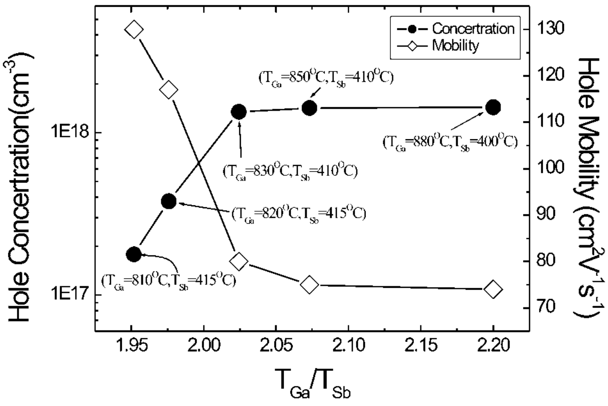

Fig. 6. Dependence of hole concentration and mobility of polycrystalline GaSb samples on $T_{\mathrm{Ga}}/T_{\mathrm{Sb}}$.

4. Conclusion

GaSb thin films have been successfully prepared by co-evaporation method on soda-lime glass substrate. X-ray diffraction studies indicated that the films are polycrystalline and have preferential orientation in the (111) direction. The grain size is increased with increasing thickness. SEM results indicate that the average grain size of GaSb thin film is 100, 250, and 500 nm at the substrate temperature of 480, 520, and 560 °C. The absorption coefficient of GaSb thin film is of the order of 10^4 cm^{-1}, and the optical bandgap of polycrystalline GaSb thin film is 0.726 eV. The hole concentration shows a clear increasing trend for increasing $T_{\mathrm{Ga}}/T_{\mathrm{Sb}}$ ratio. When the Ga crucible temperature is 810 °C and the Sb crucible temperature is 415 °C, the hole concentration of polycrystalline GaSb is 2×10^{17} cm^{-3} and the hole mobility is 130 cm^2/(V·s). These results suggest that polycrystalline GaSb thin film is a good candidate for use as a base layer material in TPV cells.

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