Quality Improvement by Annealing for Flash-evaporated Thin InSb Films

Md Abu Taher*, Yumiko Haga, Yoshio Nakamura and Osamu Nittono

Department of Metallurgy and Ceramics Science, Tokyo Institute of Technology, Tokyo 152-8552, Japan.

Thin InSb films were prepared at room temperature by a rapid vacuum evaporation (flash evaporation) method using indium antimonide compound as a source material. The characteristics of the films such as microstructure and electrical properties were investigated in terms of heat treatment procedure. The as-deposited InSb films were found to have polycrystalline structure, and their stoichiometry was much better at higher deposition rates. Their characteristics were strongly influenced by successive annealing. The evaporation of Sb from the film caused by annealing led to poor electrical properties of the films. In order to avoid the evaporation of Sb from the film, an attempt was made to cap an as-deposited film with SiO₂ layer before annealing. The microstructures as well as the galvanomagnetic properties were improved by introducing such capped-annealing. The highest Hall mobility of 2.1×10^4 cm²/Vs was obtained in an InSb film of 1.0 μm thick, when it was prepared with a deposition rate of 10 nm/s and followed by capped-annealing at 773 K.

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1. Introduction

Fabrication of electronic devices such as infrared devices, Hall effect devices, and magnetoresistance devices^{1,2)} often requires semiconductor films with high carrier mobility, small band gap, and high magnetic sensitivity. Among the compound semiconductors, InSb has the highest electron mobility and small band gap (0.17 eV at 300 K). Therefore, InSb films appear to be one of the most promising candidates for these applications.

InSb thin films have been prepared by various methods such as vapor-phase-transport reaction,³⁾ rf sputtering,⁴⁾ vacuum evaporation (including flash evaporation and three-temperature evaporation),^{5–12)} zone-crystallization (melt-regrowth of polycrystalline film),¹³⁾ molecular beam epitaxy (MBE),^{14–17)} and metalorganic chemical vapor deposition (MOCVD).^{18–20)} InSb films with carrier mobility comparable to that of bulk single crystals are usually obtained by epitaxial growth using MBE or MOCVD on single crystal substrates. The single crystal substrate and the MBE or MOCVD apparatus, however, cost a lot. Therefore, it is especially interesting to develop and study low cost procedures of producing high quality thin InSb films on low cost substrates.

Conventional vacuum evaporation techniques have difficulties in maintaining the stoichiometry of deposited InSb compounds.^{8,12,21)} This is because In and Sb have very different vapor pressures (at 800 K, 7.51×10^{-6} Pa for In and 1.066 Pa for Sb), and a loss of volatile Sb takes place during vacuum evaporation. However, such kinds of difficulties could be overcome by employing a rapid vacuum evaporation (or a flash evaporation).⁹⁾ In this method, when the source materials are flashly evaporated, the average vapor composition can be almost stoichiometric, resulting in maintaining the stoichiometry of deposited films. It is also reported that the films are produced with a good quality when deposited at high deposition rates.^{6,7,9)} Recently, Tomisu *et al.*¹²⁾ also reported that the value of the ratio Sb/(In + Sb) is 50% for the InSb

films prepared at the source temperature of 2030 K. This suggests that a high evaporation (or deposition) rate with a short deposition time can serve to reduce the deviation from stoichiometry.

In the present study, firstly we try to confirm the above speculation through a relationship between deposition rate and electrical property. Secondly, the characteristics of thus prepared films are investigated in terms of successive procedures including annealings without and with capping SiO₂ layer.

2. Experimental Procedure

Thin InSb films were prepared by evaporating high purity (99.999%) InSb compound in a high vacuum system less than 10^{-5} Pa. Pre-annealed Corning 7059 glass (size: 21 mm × 26 mm × 1.1 mm) was used as a substrate with a mask for electrical measurements (size: 3 mm × 19 mm with six terminals). The substrates were carefully cleaned before the deposition process: the glass substrates were first rinsed with water and dried, and then immersed into ethanol and rinsed ultrasonically for 600 s, and finally rinsed with acetone for 600 s and then dried. It is reported that higher substrate temperatures above 473 K lead to the deviation from stoichiometry of the InSb film.¹²⁾ In fact, we confirmed this in preliminary experiments. Thus, the substrate was not heated intentionally, but irradiated naturally from the crucible during deposition. The substrate temperature was estimated to be lower than 323 K from our previous experiments.²¹⁾ The present vacuum chamber has a heat-shielding wall having a small hole for evaporated vapor, which is set just above an alumina-coated tungsten basket (10 mm in diameter, 15 mm in depth). The distance between source material and substrate was about 10 cm. The source temperature and thus the deposition rate (0.5–20 nm/s) were adjusted by changing the electric current flow through the crucible and flow time. The source temperature was estimated to be higher than 2030 K by a pyrometer. In this study, the deposition rate was evaluated directly by a thickness monitor using a quartz crystal sensor

*Graduate Student, Tokyo Institute of Technology. Present address: Bangladesh University.

(CRTM-5000), set-up near the substrate. During deposition, the film thickness was always evaluated by a thickness monitor, and the films of 0.3 μm and 1.0 μm thick were prepared for measurements.

The deposited films were annealed at elevated temperatures between 623 and 773 K by using an image furnace (MILA-3000; ULVAC SHINKU-RIK0). Before annealing, the furnace was always pre-heated up to 873 K for cleaning the furnace, and the samples were annealed around 1×10^{-4} Pa.

The crystallinity was evaluated by X-ray diffraction analysis (XRD) using $\text{CuK}\alpha$ radiation for both as-deposited and annealed films. The surface morphology of the films was observed by a field emission scanning electron microscope (FE-SEM). The chemical composition of the films was examined by X-ray fluorescence (XRF) (RIX 2000, Rigaku) and electron probe microanalyzer (EPMA-8705, Shimadzu). A standard four-probe method was used to measure the electrical resistivity of the films. Van der Pauw Hall measurement was carried out for the films at room temperature in a magnetic field (H) of 1 T, and the electrical properties were evaluated together with galvanomagnetic properties including Hall mobility and magnetoresistance (MR), $\frac{\Delta R}{R_0} = \frac{R_H - R_0}{R_0}$, where R_0 and R_H are the resistances at a magnetic field of $H = 0$ and $H = 1$ T, respectively.

3. Experimental Results and Discussion

3.1 Optimization of deposition condition

The deposition rate is an important factor when we produce thin InSb films by vacuum evaporation techniques. In general, deposited InSb films tend to deviate from stoichiometry because the vapor pressures of Sb and In differ remarkably from each other. In this study we investigated microstructure as well as electrical property of as-deposited InSb films with respect to the deposition rate, and determined optimum deposition rates.

Figure 1 shows the dependence of Hall mobility (μ) and electrical resistivity (ρ) on the deposition rate (d), for the as-deposited films at room temperature. The Hall mobility increases and the resistivity decreases gradually with increasing deposition rate up to 10 nm/s. Over 10 nm/s, the Hall mobility decreases gradually, while the resistivity increases slightly with increasing deposition rate. These results are consistent with each other, because the two properties are inversely related to each other. The maximum Hall mobility of 400 cm^2/Vs was obtained for a film deposited at 10 nm/s. The resistivity also showed the lowest value at the corresponding rate. The results indicate that 10 nm/s is one of the most suitable deposition rates for the present rapid vacuum evaporation system. In this connection, the deposition rate, 10 nm/s, is much higher than the previously reported value (210 nm/min or 3.5 nm/s) at the crucible temperature of 2030 K.¹²⁾

In order to evaluate the crystallinity of the deposited films, several samples were examined by XRD. The results for three typical InSb samples deposited at 0.5, 5 and 10 nm/s are shown in Fig. 2. The film thickness was about 300 nm (or 0.3 μm). All the X-ray diffraction results prove that the films have polycrystalline structure. Figure 2(a) shows the result for a film deposited at 0.5 nm/s, which contains two

Fig. 1 Hall mobility and resistivity as a function of deposition rate for as-deposited films.

Fig. 2 X-ray diffraction results of InSb films prepared at various deposition rates: (a) 0.5 nm/s, (b) 5 nm/s and (c) 10 nm/s. All the films have the same thickness of 300 nm.

phases: InSb phase with peaks 111, 220, 311 and In phase with two minor peaks 101, 112. The InSb peaks become much stronger, but In peak decreases gradually with increasing deposition rate. In the film deposited at 5 nm/s, In 112 peak disappeared but a weak In 101 peak still remains. All the In peaks disappeared completely for the films deposited at 10 nm/s. Moreover, two additional peaks (331 and 422) for the InSb are clearly seen in this film, and the other InSb peaks appeared with appreciable intensities. The films deposited at higher deposition rates than 10 nm/s, although they are not reproduced here, showed a similar to but less sharp X-ray profile than that of the films deposited at 10 nm/s. The results also support that the film deposited at 10 nm/s is highly stoichiometric, resulting in high electron mobility as shown in Fig. 1.

The composition ratio of these as-deposited films was analyzed by XRF. The composition ratio of Sb is defined as $\text{Sb}/(\text{In} + \text{Sb})$, supposing that the total atomic percent of $(\text{In} + \text{Sb})$ is 100%. The value of the ratio $\text{Sb}/(\text{In} + \text{Sb})$ was $50 \pm 1\%$ when the film deposited at 10 nm/s. On the contrary, the ratio was found as small as 35% when the film deposited at 0.5 nm/s. For more confirmation, some samples were analyzed by EPMA. The composition ratio obtained by EPMA was almost the same ($\cong 50\%$) as that of XRF analysis. The above results indicate that the stoichiometric film

Fig. 3 Relationships between Hall mobility and annealing time for the films annealed at 623, 673, 743 and 753 K. Deposition rate: 10 nm/s.

can be obtained at 10 nm/s. Therefore, we determined that 10 nm/s is an optimum deposition rate in the present experiment. The reason why the nearly stoichiometric films were produced under these situations is not clear. It seems that a quasi-congruent evaporation took place under the present experimental conditions including room-temperature deposition and geometrical factors such as arrangements of crucible and substrate, source-substrate distance and a heat-shielding wall with a small hole (like a kind of quasi-Knudsen cell in MBE.) as well as the total deposition time of less than 30 s.

3.2 Improvement of film quality by annealing

3.2.1 Annealing without capping layer

The electrical and crystalline properties of the InSb films are strongly dependent on annealing temperature and time. In the present experiment, the annealing temperature varied from 623 to 773 K because the melting point of InSb is 798 K and thus solid-phase recrystallization can take place under these conditions.

Figure 3 shows the relationships between the annealing time and the Hall mobility for the films annealed at 623, 673, 743 and 753 K. The InSb films are not capped with SiO₂ layer before annealing. The Hall mobility increases after 300 s (5 min) annealing and then remains unchanged with annealing time for the films annealed at 623 K. However, in case of the films annealed at 673, 743 and 753 K the Hall mobility increases remarkably just after annealing and then decreases gradually with annealing time. This increase just after annealing may be due to structural relaxation caused by an initial annealing. A previous study¹²⁾ also showed a similar break point on the mobility vs annealing time curve for without-capping (hereafter referred to as uncapped) films prepared by using a rapid evaporation. We obtained the highest Hall mobility of 6.5×10^3 cm²/Vs for the film annealed at 743 K for 300 s. In this connection, this result is about six times higher than that of Tomisu *et al.*¹²⁾ It is also shown that the Hall mobility of the film annealed at 753 K is inferior to that of the film annealed at 743 K. This is mainly due to the vaporization of Sb from the film at higher temperatures, as will be confirmed below.

The film thickness (or average thickness) was also measured after annealing by a surface profiler (Dektak³ST). The change in film thickness is plotted as a function of annealing time for the films having initial thickness of 300 nm, as

Fig. 4 Thickness changes with annealing time for the films annealed at 743 and 753 K.

shown in Fig. 4. The film thicknesses were 255 and 240 nm after annealings at 743 and 753 K for 3.6×10^3 s (60 min), respectively. The XRF measurements also indicated that the composition ratios of Sb were 38 and 26% after annealings at 743 and 753 K for 3.6×10^3 s, respectively. Therefore, the decrease in film thickness is thought to be caused mainly by the vaporization of Sb from the film, though a small amount of In atoms is also ascribed to the decrease in film thickness.

Our previous study²¹⁾ showed a similar tendency that the Hall mobility decreased with increasing annealing temperature. The highest Hall mobility of 4.3×10^3 cm²/Vs was found for a film annealed at 733 K with the initial thickness of 300 nm. All the films of our previous study were prepared under the average deposition rate of 0.5 nm/s. Therefore, the results are explained by the same reason described above, and are consistent with the present results.

3.2.2 Annealing with capping SiO₂ layer

In this study the annealed uncapped films showed sixteen times larger Hall mobility than the as-deposited films. However, they showed lower values for longer annealing periods and at higher annealing temperatures. For further quality improvement of the films, a procedure needs to prevent Sb from self-evaporating during annealing. Therefore, an attempt was carried out to cap the InSb films with SiO₂ layer before annealing. For capping process, an InSb film was first deposited on a glass substrate, and then SiO₂ layer of about 250 nm thick was deposited on the InSb film in the same vacuum chamber. Thus prepared InSb films were then annealed in a vacuum. After annealing, the SiO₂ layer was removed by wet etching (HF: 20% solution), and gold contacts were then deposited on the terminals of such InSb films for electrical measurements. In preliminary experiments, this capped-annealing was found to be very effective for InSb films to be annealed up to 773 K without any change in Hall mobility, as will be shown later.

The effect of annealing on surface morphology of InSb film was also examined and the results are shown in Fig. 5, which shows SEM micrographs of the films annealed without and with capping SiO₂ layer. An uncapped film, when annealed at 723 K for 3.6×10^3 s, is composed of small grains resulting in less smooth surfaces, and a lot of voids are seen throughout the film (Fig. 5(a)). These voids are thought to have been formed during annealing process, probably ascribed mainly to

Fig. 5 SEM photographs showing surface morphology for the films without and with capping SiO₂ layer.

the evaporation of InSb, induced by initial vaporization of Sb atoms. The void size increases with increasing annealing temperature, as shown in Figs. 5(a) and (b), though the surface morphology appears to be suffered by the evaporation of InSb. The average size of the void is around 0.6 μm when the film was annealed at 753 K for 3.6×10^3 s. These voids may cause large electrical resistance or insulation for the films when annealed without capping SiO₂ layer. This can also explain why the Hall mobility decreases at longer annealing periods, as shown in Fig. 3. On the other hand, when annealed with capping SiO₂ layer, the films are composed of slightly rugged grains connecting with each other, as shown in Figs. 5(c) and (d). No voids are seen between adjacent grains in such capped films even when annealed at 723 K for 3.6×10^3 s. The XRF measurements also proved that the compositions of In and Sb in the capped films were nearly the same as those of as-deposited films. In this connection, neither appreciable decrease of the film thickness nor the ratio Sb/(In + Sb) was detected experimentally. The results indicate that the capping SiO₂ layer can prevent InSb from evaporating and hence the quality improvement of the films will be expected by a capped-annealing.

Figure 6 shows the relationships between the Hall mobility and the annealing time for such films annealed with and without capping SiO₂ layer. The Hall mobility of capped films increases instantly after short annealing time and then remains unchanged as the annealing time increases. This is quite different from the results of uncapped films. In the present study, the highest Hall mobility of 1.25×10^4 cm²/Vs was obtained for a capped film of 300 nm thick, when annealed at 773 K.

The carrier concentrations of the InSb films together with the resistivity, Hall coefficient and Hall mobility are listed in Table 1. In this evaluation, the majority of carrier is assumed to be electron.¹²⁾ As the annealing temperature increases, the carrier concentration decreases up to 743 K for uncapped films. Above 743 K, the carrier concentration slightly increases. On the other hand, the carrier concentration for capped films decreases gradually with increasing annealing temperature. The lower the carrier concentration, the higher the Hall coefficient and the Hall mobility. This trend, however, must be examined in more detail.

The magnetoresistance (MR) effect was also investigated at room temperature in a magnetic field of 1 T. It is well known that the magnetoresistance is associated with the specimen

Table 1 Electrical properties of InSb films with respect to annealing temperature.

Film type	Annealing temperature* K (°C)	Carrier concentration n (cm ⁻³)	Resistivity, ρ (Ω -cm)	Hall coefficient, R_H (cm ³ /C)	Hall mobility, μ (cm ² /Vs)
Uncapped film	623 K(350)	2.34×10^{17}	2.31×10^{-2}	26.71	1.15×10^3
	673 K(400)	1.61×10^{17}	9.51×10^{-3}	38.84	4.08×10^3
	723 K(450)	1.44×10^{17}	9.12×10^{-3}	43.12	4.76×10^3
	733 K(460)	1.28×10^{17}	8.65×10^{-3}	48.71	5.62×10^3
	743 K(470)	1.17×10^{17}	8.34×10^{-3}	53.33	6.39×10^3
	753 K(480)	1.41×10^{17}	1.10×10^{-2}	44.20	3.91×10^3
Capped film	623 K(350)	2.18×10^{17}	2.12×10^{-2}	28.54	1.34×10^3
	673 K(400)	1.56×10^{17}	9.45×10^{-3}	39.86	4.21×10^3
	723 K(450)	1.51×10^{17}	8.28×10^{-3}	41.36	4.99×10^3
	733 K(460)	1.25×10^{17}	7.36×10^{-3}	49.91	6.77×10^3
	743 K(470)	1.16×10^{17}	6.69×10^{-3}	53.54	7.99×10^3
	763 K(490)	1.09×10^{17}	5.75×10^{-3}	56.93	9.89×10^3
	773 K(500)	9.99×10^{16}	4.99×10^{-3}	62.56	1.25×10^4

*Annealing time was 15 min. Film thickness: 300 nm.

Fig. 6 Relationships between Hall mobility and annealing time annealed with and without capping SiO₂ layer. Deposition rate: 10 nm/s, film thickness: 300 nm.

factors such as thickness inhomogeneity, specimen shape and metallic inclusions.^{2, 22–24}) In the present study, all the samples have the same thickness of 300 nm and the same shape (the length between adjacent electrodes; $l = 4$ mm and the width; $w = 3$ mm) with the length to width ratio of $(l/w) = 1.3$. The highest magnetoresistance of $\Delta R/R_0 = 60\%$ and 75% was obtained for uncapped films annealed at 743 K and capped films annealed at 773 K, respectively. As shown on SEM micrographs reproduced in Fig. 5, the uncapped films possess considerably rough surfaces with a lot of voids throughout the film, which may result in the inhomogeneity of carrier concentrations when subjected to a magnetic field. On the other hand, the capped films show considerably continuous surfaces without any voids, which result in homogeneity of carrier concentration throughout the films. Thus, a lower value of magnetoresistance for uncapped film may be due to such inhomogeneity. A room temperature MR value of 155% has been reported by Oshita *et al.*²⁾ for InSb films prepared by a source-temperature-programmed evaporation method. Therefore, further quality improvement will be expected by successive optimum procedures including longer annealings.

A significant increase in diffraction intensity of XRD was also seen for the annealed capped films, although the results

Fig. 7 Dependence of the Hall mobility on film thickness, when annealed at 773 K for 60 min (3.6×10^3 s).

were not reproduced here. This indicates that the crystallinity of the film including grain sizes can be improved remarkably after annealing at elevated temperatures. In fact, individual constituent grains become slightly large with less rugged shapes and are connecting densely with adjacent grains, as shown in Fig. 5(d). Thus, the film quality improvement can be mainly reflected on such morphological improvements caused by annealing with capping SiO₂ layer.

The Hall mobility is also reported to depend on the film thickness.^{6, 12)} Therefore, in order to confirm this, we investigated the Hall mobility of the capped films as a function of film thickness. Figure 7 shows the variation of Hall mobility with film thickness for the films thinner than 1.0 μm thick. The Hall mobility increases with increasing film thickness, as theoretically expected.^{13, 25)} The highest Hall mobility of 2.1×10^4 cm²/Vs was achieved for InSb films with the thickness of 1.0 μm . In this connection, the single InSb crystal can show an electron mobility of 7.6×10^4 cm²/Vs.¹⁷⁾ This result suggests us that thicker films can show a higher Hall mobility. Thus, we also measured the Hall mobility for several thicker films than 1.0 μm . As a result, no linear increase in Hall mobility was observed for thicker films with a slightly large fluctuation of data, probably due to the lack of homogeneity in thicker films. It is reported, however, that for thicker films

than 1.0 μm the Hall mobility increases gradually with increasing film thickness.^{6,12,13,25} This point must be further examined in detail, because if the sample preparation conditions were guaranteed for the present films, a further high mobility will be expected by suitable procedures including annealing time and temperature.

4. Conclusions

InSb thin films were deposited on glass substrates by a rapid vacuum evaporation. The electrical and structural properties of the films were investigated in terms of heat treatment procedures. XRD, XRF and electrical measurements revealed that the film quality strongly depends on the deposition rate, and that the film deposited at 10 nm/s is highly stoichiometric. It was also found that the evaporation of Sb took place at higher annealing temperatures and for longer periods in a vacuum. Capped-annealing procedures can cause InSb films anneal up to 773 K in a vacuum. No further evaporation of Sb was observed for such capped films even for long annealing periods. The Hall mobility of $1.25 \times 10^4 \text{ cm}^2/\text{Vs}$ was obtained for a film annealed at 773 K with the thickness of 300 nm. It was also shown that, if the optimum preparation conditions are given, thicker InSb films can give a much higher mobility: in this study the highest electron mobility of $2.1 \times 10^4 \text{ cm}^2/\text{Vs}$ was successfully achieved for the films of 1.0 μm thick, when annealed at 773 K for $3.6 \times 10^3 \text{ s}$. Further detailed experiments are now progress for thicker InSb films.

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