Growth and Characterization of InSb Thin Films on GaAs (001) without Any Buffer Layers by MBE *

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We report the growth of InSb layers directly on GaAs (001) substrates without any buffer layers by molecular beam epitaxy (MBE). Influences of growth temperature and V/Ⅲ flux ratios on the crystal quality, the surface morphology and the electrical properties of InSb thin films are investigated. The InSb samples with room-temperature mobility of 44600 cm^2/Vs are grown under optimized growth conditions. The effect of defects in InSb epitaxial on the electrical properties is researched, and we infer that the formation of In vacancy (V_{In}) and Sb anti-site (Sb_{In}) defects is the main reason for concentrations changing with growth temperature and Sb_2/In flux ratios. The mobility of the InSb sample as a function of temperature ranging from 90 K to 360 K is demonstrated and the dislocation scattering mechanism and phonon scattering mechanism are discussed.

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Recently, InSb single-crystal thin films grown by molecular beam epitaxy (MBE) have received a great deal of attention due to its ultra-high electron mobility of ∼70000 cm^2/Vs, a low effective mass of $0.013m_e$, and a narrow band gap of 0.17 eV. With these characteristics, InSb becomes a unique Ⅲ–Ⅴ compound semiconductor suitable for several applications including infrared detectors,[1,2] high-speed devices[3–7] and magnetic sensors.[8–11] For practical applications, InSb films are necessary to be deposited on semi-insulating (SI) substrates. There has been no lattice matched SI substrates for InSb thin films, thus SI GaAs (001) substrates are an available choice due to their electrical isolation, low cost and mechanical strength. However, because of the lattice mismatch (14.9%) between GaAs substrates and InSb thin films, it is difficult to induce the dislocations in the InSb growing process.[12,13] Generally, buffer layers[14] are now in common use to solve this problem. Some groups took a two-step method consisting of a low-temperature InSb buffer layer[13,15,16] at the initial growing to filtering the dislocation. The InAlSb layer is also an alternative buffer[12,17,18] to reduce the misfit dislocation associated with lattice mismatch. The inserted buffer layer makes a high-quality InSb thin film despite the large lattice mismatch between InSb and the GaAs substrate. However, it is not cost efficient and makes the growing process complicated. Thus a few reports about using no buffer layer have also been published.[19,20] In this Letter, a series of InSb thin films are grown directly on GaAs (001) substrates to investigate the influence of growth temperature, V/Ⅲ flux ratios on the crystal quality, the surface morphology and electrical characteristics of InSb thin films.

All samples are grown on SI GaAs (001) substrates with an EPI GEN-Ⅱ solid-source MBE system. Arsenic and antimony are supplied by cracking cells to produce As_2 and Sb_2, respectively, and all fluxes are calibrated using beam-equivalent pressure (BEP) shown by a vacuum ion gauge at the substrate position. Quality and crystallinity of grown layers is monitored by reflection high-energy electron diffraction (RHEED) during the growth process. After desorbing surface oxide under As flux for 10 min at 600℃, which is measured by an infrared thermometer, the substrate continues to cool down to the InSb growth temperature T_g. Antimony shutter is opened and kept on for 3 min before the InSb thin films grows.[21] Finally, unintentionally doped 1 μm InSb layers are grown directly on the GaAs (001) substrate without a GaAs buffer layer with a growth rate 1 μm/hr. To optimize growth conditions, experiments are designed with different temperatures and BEP ratios of Sb/In. As listed in Table 1, samples A, B, C and D are grown at T_g of 400, 410, 420, 440℃, respectively, and a second series of samples (C1, C, C2, C3) are grown at the same temperature 420℃ with different Sb_2/In (BEP ratios) ranging from 8 to 15. The InSb layers of 1000 nm and 500 nm named as samples E and F, respectively, are grown with a GaAs buffer layer of 200 nm. It is noted that our InSb thin film is grown directly on the substrate without any InAlSb[22] or low temperature InSb[16] buffer layers.

Hall measurement using the standard van der

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Pauw configuration is carried out to characterize the electronic properties of InSb films with indium electrode for ohmic contact.[23] The surface morphology is evidenced and analyzed by atomic force microscopy (AFM). The InSb crystalline quality is evaluated by double-crystal x-ray diffraction (XRD), using a Cu Kα source and (004) reflection.

Table 1. InSb samples with their related room temperature (300 K) electron mobilities by Hall measurements with van der Pauw configuration.

Sample	T_g(℃)	Sb_2/In	Electron mobility 300 K (cm^2/Vs)
A	400	10	38960
B	410	10	39400
C	420	10	44600
D	440	10	25700
C1	420	8	42884
C2	420	12	38705
C3	420	15	35230

The ω–2θ scan of (004) double crystal XRD spectra for samples A, B, C and D is plotted in Fig. 1(a). From the angle difference between the diffraction peaks of the epitaxial film (001) InSb and the substrate (001) GaAs, the lattice constants of InSb along the [001] direction are calculated to be 6.466 Å, 6.467 Å, 6.467 Å and 6.466 Å of samples A, B, C and D, respectively. This indicates that the residual stress in the InSb film along the [001] direction is almost fully relaxed with the process of growth. Figure 2 shows the full width at half maximum (FWHM) of the XRD rocking curves of the InSb layers as a function of the growth temperature of InSb. The FWHM of the XRD rocking curves related to the crystal quality of the InSb thin films has a small increase with increasing the InSb growth temperature. However, sample C at a growth temperature of 420℃ has a sharp decrease with the lowest FWHM value of 581.5 arcsec, indicating the best crystal quality. Figure 3 shows the AFM images of the surfaces of samples A, B, C and D in an area of $1 \times 1\,\mu m^2$, respectively. Atomic step-and-terrace structure with similar results reported by Akira *et al.*[15] is clearly visible on the surfaces of samples A, B and C, indicating the 2D mode in the growth process of the InSb layer. However, the surface of sample D at a higher growth temperature presents granular stripes. It can be seen that the InSb thin films grown at lower temperature has a flat surface with lower rms roughness due to its microcrystal grain size. However, it has poor crystallinity for samples A and B probably because of insufficient ad-atom migration on the growing surface. On the other hand, sample D grown at a higher temperature has a rougher surface compared with other samples due to three-dimensional islands. The surface roughness is directly related to the interface roughness of the hetero-junction structure, resulting in the interface roughness scattering in the quantum well.[24] Thus the surface of InSb grown at a relative low growth temperature is suitable for hetero-structure devices. From the results of XRD and AFM, it can be assumed that sample C grown at 420℃ in this study has the best crystal quality and relatively low surface roughness, which is useful for InSb infrared detectors and quantum devices.

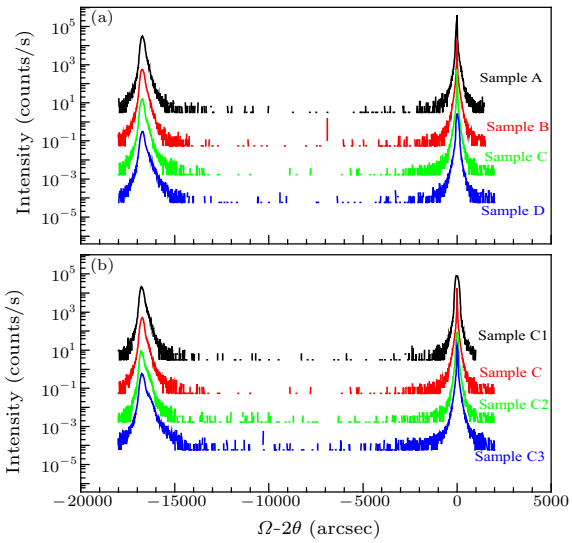

Fig. 1. Double-crystal x-ray diffraction scans of (a) samples A, B, C and D, (b) samples C1, C, C2 and C3.

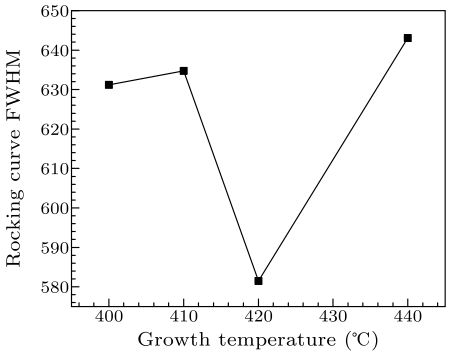

Fig. 2. The FWHM of the XRD rocking curves of the InSb thin film as a function of the growth temperature of InSb.

To compare the electrical property of samples at different temperatures, Hall measurement is carried out using the standard van der Pauw configuration. The room temperature (300 K) and 77 K electron mobility and concentrations of the InSb layers as a function of the growth temperature are shown in Fig. 4. When the growth temperature of InSb is 420℃, the InSb epitaxial layer of sample C has the highest room temperature and 77 K electron mobility (44, 600 cm^2/V·s and 946 cm^2/V·s). Meanwhile, when the growth temperature is higher or lower than 420℃, there is a significant decrease of both room temperature and 77 K electron mobility. Thus it is obvious that the InSb layer grown at 420℃ has the best crystal quality and electrical quality.

However, it can be seen in Figs. 4(a) and 4(b) that the electron concentrations of both 77 K and room temperature have an increasing trend as the InSb

growth temperature increases, unlike the results reported by Atsushi *et al.*[25] The un-doped InSb like an intrinsic semiconductor with approximately equal numbers of electrons and holes behaves like an n-type conduction, because the electrons are much lighter dominating the conductivity as the sign of the Hall constant. The native point defects in InSb are presumably resulted from the removal of isolated vacancies, which become supersaturated as the epitaxial layer is cooled. In the Sb-rich epitaxial layer, a probable mechanism is due to a formation of Schottky pairs and Sb anti-site (Sb_{In}) defects.[26,27] Thus in the Sb-rich InSb layers, it can be ascribed to the formation of more donors like VIn and SbIn defects as the InSb growth temperature increases higher. With increasing the growth temperature, it is deduced that the surface re-evaporation of In atoms increase and In lattice sites are available for Sb atoms, resulting in the formation of more In vacancy (V_{In}) and Sb anti-site (Sb_{In}) defects, which are both n-type native defects. Thus the electron concentration increases as the InSb growth temperature increases.

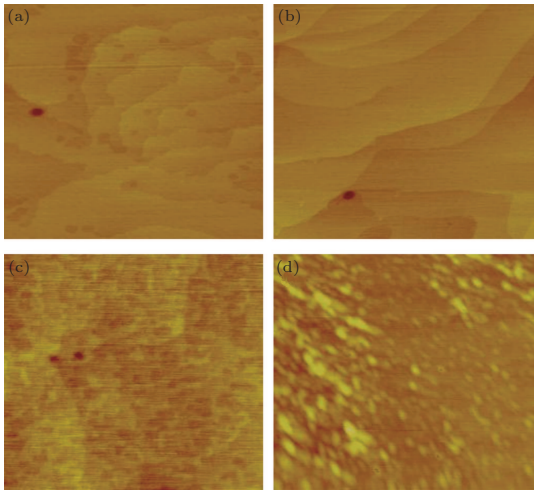

Fig. 3. AFM images of the surface of samples A (a), B (b), C (c) and D (d).

After optimization of the growth temperature, a set of samples (C1, C, C2 and C3) is grown to investigate the influence of V/Ⅲ flux ratios on InSb thin films. All the samples are prepared at the optimal growing temperature of 420℃. The FWHM of InSb layers with Sb2/In BEP ratio from 8 to 10 decreases from 803 arcsec to 533 arcsec. When the Sb2/In BEP ratio increases continuously from 10 to 15, the FWHM of the InSb layer becomes higher than 970 arcsec, which means the deterioration of InSb crystal quality. When the Sb/In flux ratio is 10, the InSb thin film has the lowest rms surface roughness. Although the surface morphology and crystal quality of the InSb epitaxial layer are influenced largely by the large lattice mismatch (14.6%) between the InSb thin film and the GaAs substrate, the surface roughness can be minimized by optimizing the growth temperatures and corresponding V/Ⅲ flux ratios. It is noted that the surface morphology and crystal quality of InSb thin film with no buffers is around a similar level with the InSb thin films using InAlSb buffer[28] or low temperature (LT) InSb buffer.[29] Based on the results and analysis of Zhang *et al.*,[30] during the initial stages of growing InSb thin film on GaAs substrates, misfit dislocations are formed from the InSb islands. Thus when the InAlSb or LT InSb buffer layers are grown on the GaAs substrate, misfit dislocations may also be formed at the island edges as the islands grow and coalesce. It is inferred that the interface between InAlSb or LT buffers and GaAs substrate acts as a threading dislocation nucleation source and introduces some additional threading dislocation.[29] It is a probable reason that the InSb layers with no buffers have no significant deterioration on surface morphology and crystal quality compared with InSb thin films with InAlSb or LT buffers.

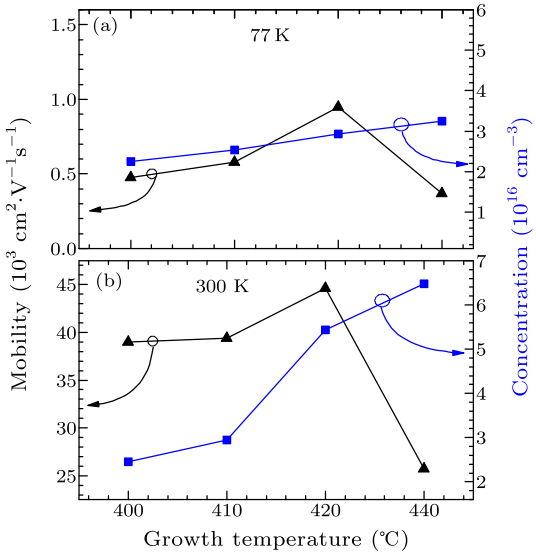

Fig. 4. (a) The 77 K and (b) room-temperature electron mobilities and concentrations of the InSb layers with increasing the growth temperature.

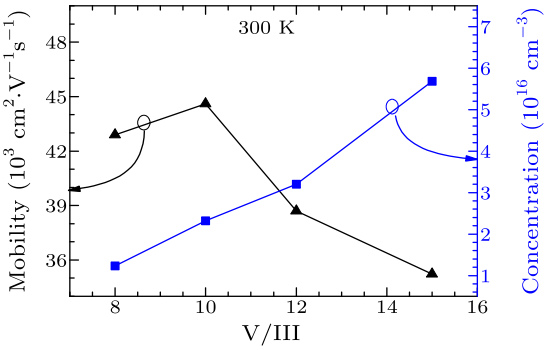

Fig. 5. Room-temperature (300 K) mobility and concentrations of the InSb layers versus the Sb_2/In BEP ratio.

The electrical mobility and concentration of the InSb layers as a function of the Sb_2/In BEP ratio at room temperature (300 K) are shown in Fig. 5. With increasing the Sb_2/In BEP ratio from 8 to 10, the InSb

room temperature electronic mobility increases from 42884 $cm^2/V{\cdot}s$ to the maximum value 44600 $cm^2/V{\cdot}s$. When the Sb_2/In BEP ratio increases higher than 10, the InSb electronic mobility presents the obvious downward trend. Simultaneously, the 77 K electronic mobility has the highest value with the Sb_2/In BEP ratio at 10, as shown in Fig. 6. The InSb electronic mobility dependence on Sb_2/In BEP ratio is consistent with the surface morphology and crystal quality results. As the Sb atom has a longer surface lifetime, excess Sb atoms are incorporated in the InSb films. On the other hand, the lack of Sb element can increase In droplets on the surface.[18] As a result, an appropriate V/Ⅲ flux ratio is very important for the surface morphology, crystal quality and electrical characteristics. In our samples, InSb epitaxial films present worse properties when the V/Ⅲ flux ratio is larger or smaller than the optimal value of 10. With the results discussed above, it is pointed out that the growing trend of 300 K electrical concentration with Sb_2/In BEP ratio increasing from 8 to 10 is attributed to the formation of more n-type donors like V_{In} and Sb_{In} defects. However, it is remarkable that the majority carrier of the InSb thin film with Sb_2/In BEP ratio of 8 changes from n-type to p-type conduction at 77 K. For unintentionally doped InSb thin films, the carrier is mainly derived from intrinsic excitation[31] and native defects. In the case of a low enough Sb_2/In BEP ratio, 8, in this study, the InSb layer behaves with n-type conduction at 300 K intrinsic excitation dominating the conductivity. When the temperature decreases, the intrinsic carriers freeze out[32] and the extrinsic properties of the InSb sample C1 begin to play a major role. It is deduced that sample C1 has a background of acceptors like V_{Sb} or InSb defects that start to dominate resulting in this InSb sample behaving like a p-type conduction. Compared with InSb thin films grown on LT InSb[15,16,29,31] or InAlSb[17,18,22] buffers, the room-temperature electrical mobility of our samples grown directly on GaAs substrates has a similar value. Figure 7 shows the mobility of sample C as a function of temperature obtained by Hall measurements. In Fig. 7, it is found that the electrical mobility of sample C takes a sharp decrease as the temperature decreases. It is attributed to the scattering effect associated with dislocations. According to the Pödör model[33] and the Dexter–Seitz model,[34] the electrical mobility limited by dislocation scattering scales with temperature. The inset of Fig. 7 shows the linear fit of electrical mobility versus temperature ranging from 90 K to 150 K. It is evident that the electrical mobility of sample C in the low temperature range is mainly limited by scattering associated with dislocations. As reported by Ayers[35] and Qadri and Dinan,[2] x-ray rocking curves can provide non-destructive measurements of dislocation densities. The threading dislocation density of sample C is $1.10\times10^9\ cm^{-2}$, a higher value than those results from other groups.[17] It is considered that dislocation is the reason for a relatively low mobility of sample C at low temperature, compared with those using buffer layers.[16] Additionally, the maximum value of electrical mobility around 300 K is determined by the competitive mechanism including phonon scattering and dislocation scattering.[36]

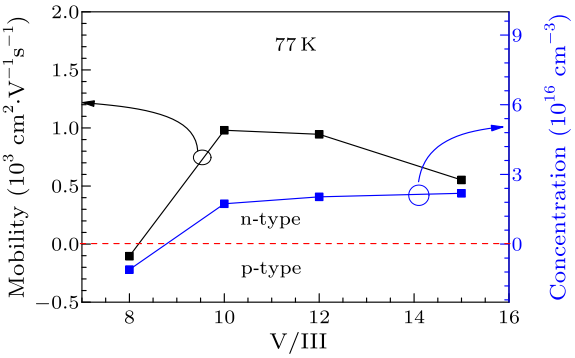

Fig. 6. The 77 K electron (hole) mobility and concentrations of the InSb layers versus the Sb_2/In BEP ratio.

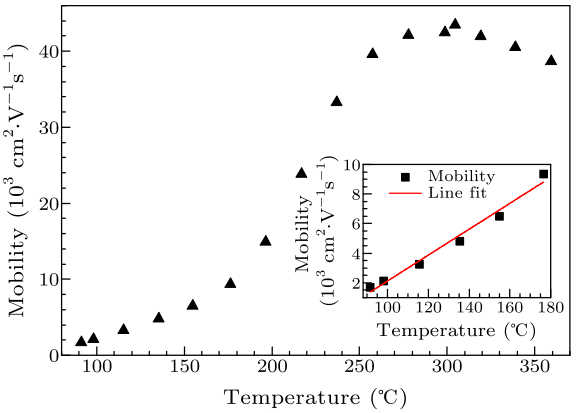

Fig. 7. Mobility of sample C as a function of temperature obtained by Hall measurements. The inset shows the linear fit of mobility versus temperature ranging from 90 K to 150 K.

However, it is found that the mobility of InSb thin films at 77 K is too much smaller than that at room temperature. The threading dislocation density of InSb samples according to the x-ray rocking curves shows a high value of $1.10\times10^9\ cm^{-2}$. It is assumed that the threading dislocation contains a part from the surface stages of the GaAs substrate after desorbing surface oxide rather than lattice mismatch between GaAs and InSb. Therefore, it is inserted with a 200 nm GaAs buffer layer before InSb growing. Samples E and F with InSb thickness of 1000 and 500 nm are grown on the GaAs buffer layers. Figure 8 shows the ω–2θ scan of (004) double crystal XRD spectra for samples E (black) and F (red). According to the FWHM of x-ray rocking curves[2,35] of samples E (330 arcsec) and F (420 arcsec), dislocation densities can be estimated $\sim5.36\times10^8\ cm^{-2}$ and $\sim7.85\times10^8\ cm^{-2}$, respectively. Then, a Hall measurement carried out at 77 K for samples E and F display the mobility as ∼2937 and ∼1240 cm^2/Vs, respectively. Thus the GaAs buffer is

commonly used to repair the surface on GaAs[37] substrate after desorbing surface oxide at 600℃. It can be speculated that the threading dislocations caused by the stages on the GaAs substrate surface are reduced with the GaAs buffer layer inserted. Thus the electron mobility at 77 K of InSb thin films is improved. Nevertheless, the lattice mismatch between GaAs and InSb still has an influence on electronic transfer performance under a very low temperature.

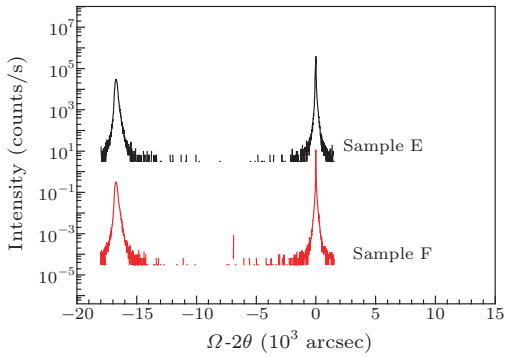

Fig. 8. Double-crystal x-ray diffraction scans of samples E (black) and F (red) with a 200 nm GaAs buffer layer.

In summary, the growth conditions of InSb layers directly on GaAs (001) substrates without any buffer layers by molecular beam epitaxy (MBE) are optimized including growth temperature and Ⅴ/Ⅲ flux ratios. We investigate the influence of growth conditions on the crystal quality, the surface morphology and the electrical properties of InSb thin film. The InSb samples with the mobility of 44600 cm^2/Vs are grown under optimized growth conditions. The effect of defects in InSb epitaxial on the electrical properties is researched, and we deduce that the formation of V_{In} and Sb_{In} defects is the main reason for the concentrations changing with growth temperature and Sb_2/In flux ratios. The mobility of the InSb sample as a function of temperature demonstrates the dislocation scattering mechanism in the low temperature ranging from 90 K to 150 K.

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