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The growth and electronic properties of α-Sn thin films grown on InSb(100) and $(\bar{1}\bar{1}\bar{1})$ substrates by molecular beam epitaxy (MBE)

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The growth and electronic properties of α-Sn films grown on InSb(100) and $(\bar{1}\bar{1}\bar{1})$ are investigated by RHEED, Shubnikov–De Haas (SDH) and magneto-optical studies. Two new structures, p(2×2) and c(4×4) apart from the commonly observed two-domain (2×1), are observed on the (100) surfaces when the overlayer thicknesses approach 500 Å and 1000 Å respectively. The α-Sn/InSb$(\bar{1}\bar{1}\bar{1})$ surface exhibits a (3×3) reconstruction up to an overlayer thickness of 1000 Å and a (3×1) is developed on further growth. It is found that not only the surface structures of the α-Sn film vary with substrate orientations and the thicknesses of the overlayer, the charge concentration of the two dimensional electron gas (2DEG) at the interface also exhibits similar behaviour. The properties of the 2DEG are investigated by Shubnikov–De Haas (SDH) experiments and possible mechanisms responsible for the occurrence of the 2DEG are discussed. Magneto-optical data indicate the presence of carriers due to the α-Sn layer with m^* ranging from $0.028m_0$ to $0.034m_0$.

1. Introduction

The observation of a two-dimensional electron gas (2DEG) at the interface of a lattice-matched nonpolar–polar system, namely the α-Sn/InSb heterostructure [1], has already been reported. Results from samples with various film thicknesses seem to suggest that the electronic properties, in particular the 2DEG, may be related to the surface structures of the substrate and the overlayer. In this paper we will present a brief report of the growth and electronic properties of α-Sn films grown on InSb(100) and $(\bar{1}\bar{1}\bar{1})$ surfaces as studied by reflection high energy electron diffraction (RHEED) [2], Shubnikov–De Haas (SDH) and magneto-optical measurements. The properties of the 2DEG will then be discussed in the light of the various experimental results. A detailed analysis of the SDH and magneto-optical data for the α-Sn films will be reported elsewhere [3].

The growth of α-Sn and α-$Sn_{1-x}Ge_x$ thin films has recently been the subject of considerable interest following the pioneering work of Farrow et al. [4]. Several groups have independently reported the growth of high quality n-type α-Sn thin films on CdTe [5–9] and InSb [10] substrates. The confinement of carriers in the thin film (quantum size effect) has recently been reported [8], but apparently a 2DEG was not observed.

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2. Experimental

The samples investigated were 0.02–0.35 μm thick α-Sn overlayers grown on InSb(100) and $(\bar{1}\bar{1}\bar{1})$ surfaces (Cd doped to 10^{14} cm^{-3} for the p-type and Te doped to 10^{14} cm^{-3} for the n-type: both supplied by MCP, UK), using a three-chamber MBE machine. Details of the growth and sample preparation have been reported in earlier papers [1,2] and will not be repeated here.

3. Results and discussion

3.1. Surface structures of α-Sn(100) and $(\bar{1}\bar{1}\bar{1})$ films

Surface structures of the α-Sn films grown on InSb substrates have been studied by RHEED using a multi-azimuthal approach. Diffraction

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streaks in several Laue zones of the RHEED patterns obtained from three different azimuths are examined as a function of film thicknesses. This "multi-azimuthal, multi-Laue zone" diffraction analysis is essential for the study of complex surface structures in which several surface domains coexist at the same time. For instance, half order streaks along ⟨110⟩ azimuths together with an integral streak along [100] can both be found in the zeroth Laue zone of two-domain (2 × 1) as well as two-domain c(4 × 2) structures. The difference between these two structures can only be distinguished from the diffraction streaks in higher order Laue zones [2].

The growth of α-Sn on InSb(100)-(4 × 2) starts with the quenching of the substrate superstructure for the first 20 Å, leaving a bulk structure together with strong background in the RHEED patterns. Using a growth rate of about 0.5 μm/h, the surface structures of the α-Sn films undergo a thickness dependent change: two-domain (2 × 1), p(2 × 2) and c(4 × 4) structures are observed at film thicknesses of 20, 500 and 1000 Å respectively. When the film thickness exceeds 1000 Å, the surface structure becomes very complicated: 1D disorder accompanied by atomic steps is observed. The presence of these structures on the α-Sn(100) surface has been compared with that of Si(100), and a dimer model [2] has been proposed to account for the occurrence of these structures on the α-Sn(100) surface.

The growth of α-Sn/InSb($\bar{1}\bar{1}\bar{1}$)-(3 × 3) is simpler than that of the (100) surface: a (1 × 1) bulk structure is observed for the first 20 Å of growth and then a (3 × 3) structure takes over until the thickness exceeds 1000 Å. Further growth reduces the intensity of the 1/3 order streak along [$1\bar{1}2$] and a (3 × 1) structure begins to develop as the film thickness reaches about 1200 Å. The observation of the (3 × 3) structure agrees with that re-

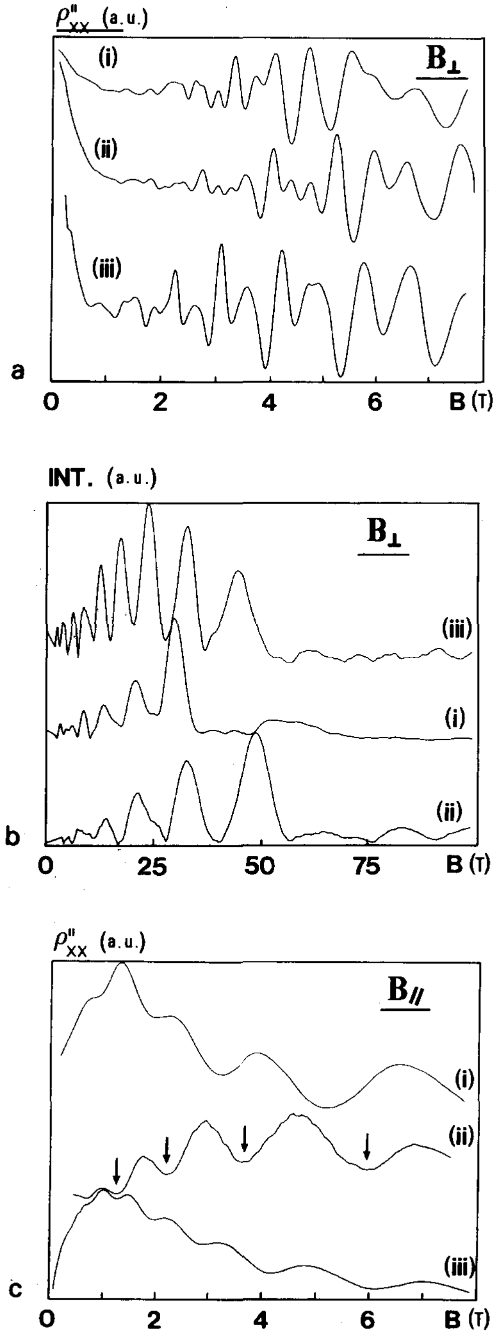

Fig. 1. (a) Experimental recordings of the second derivative of the magnetoresistance of an α-Sn/InSb structure at 4.2 K with the magnetic field applied perpendicular to the plane of the sample for the (i) 200 Å and (ii) 3000 Å thick film grown on InSb(100), and (iii) 1200 Å thick film grown on InSb($\bar{1}\bar{1}\bar{1}$) surfaces. (b) The corresponding FFT power spectrum for these three samples and (c) diamagnetic resonances due to depopulation of carrier into lower subbands when magnetic field is applied parallel to the plane of the sample. Depopulation fields of individual subbands are indicated by the arrows.

ported by Kasukabe et al. [11] but differs from the (1 × 1) structure observed by Hochst et al. [12].

Note that the surface structures of the α-Sn films depend on the overlayer thickness and substrate orientations. It is found that the 2DEG exhibits similar behaviour: the carrier concentration of the 2DEG seems to be affected by the orientation as well as the thickness of the sample (see section 3.2 below).

3.2. Shubnikov–De Haas results

Hall measurements indicated that the first few structures grown in the reactor were p-type but that later samples were n-type even for films only 200 Å thick. The presence of a high-density 2DEG is evident from the complicated magnetoresistance structures as depicted in fig. 1a. Note the remarkable difference between the peak structures observed from the thick (3000 Å) and the thin (200 Å) films. Due to the complicated oscillatory structures in the magnetoresistance, fast Fourier transform (FFT) of the magnetoresistance data is required to determine the $(1/B)$ periodicity and to identify individual subbands. The FFT power spectrum shown in fig. 1b indicates distinctly the existence of as many as nine subbands in the α-Sn/InSb($\bar{1}\bar{1}\bar{1}$) sample. When the magnetic field is applied parallel to the sample surface, the bottom of the electric and magnetic hybrid subbands will be shifted upwards through the Fermi level. As a result, charge carriers will be redistributed to the lower subbands. The reduction in inter-subband scattering gives rise to "diamagnetic resonances". Fig. 1c shows parallel field measurements which consistently demonstrate the presence of a large number of subbands in the thin film.

The results of "tilting" experiments in which the magnetic field is rotated to a variable angle θ with respect to the normal of the growth plane can be very informative. For a true 2DEG the fundamental fields of the Shubnikov–De Haas series should increase as $1/\cos\theta$, i.e. the separation of the Landau levels should decrease with increasing θ. On the other hand, spin-splitting should be independent of θ for isotropic conduction bands with the result that it should become progressively more significant as θ increases. Rotation measure-

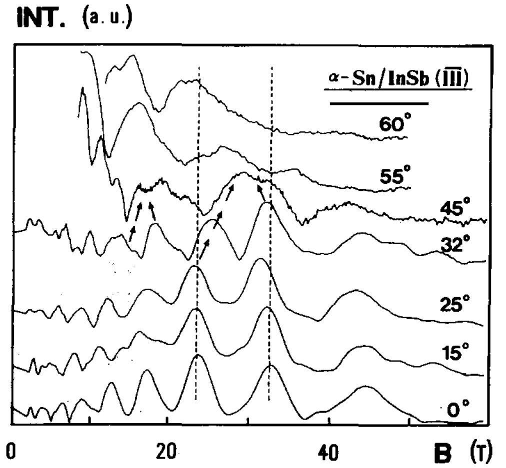

Fig. 2. The FFT power spectra for the α-Sn/InSb($\bar{1}\bar{1}\bar{1}$) sample with the magnetic field applied at angle θ to the normal of the surface plane. Note that the fundamental field for the subbands has been scaled to the $B\cos\theta$ term, i.e. for a perfect 2DEG the peaks should follow the dashed line. At about $\theta = 45°$ some peaks appear to merge together: this could be attributed to the change of Landau level spacing relative to that of the spin-splitting as θ increases.

ments were performed with α-Sn/InSb(100) and ($\bar{1}\bar{1}\bar{1}$) samples and the results for the ($\bar{1}\bar{1}\bar{1}$) surface are shown in fig. 2. With this sample, spin-splitting apparently became dominant at an angle of approximately 45°. Up to 32°, the Fourier peaks from the first two subbands showed little influence of spin whereas the spin-splitting terms for subbands with $i > 3$ became much weaker on rotation. Note that almost all of the subbands follows the $1/\cos\theta$ rule. On the other hand, experiments taken with small intervals in θ with spike-doped InSb grown at low temperature (240°C) showed some deviation from the $1/\cos\theta$ relationship at small angles [3]. While diffusion of Si dopant in these sample is expected to be low [13], it may still be significant on the scale of the cyclotron radius. Moreover, Fourier analysis data on δ-doped InSb samples indicated that the spin-splitting terms did not become dominant until $\theta > 50°$. The significant differences between the α-Sn structures and the δ-InSb layers suggest that the 2DEG in the former case involves the α-Sn layer itself and that little intermixing of the Sn and InSb has occurred away from the interface, i.e. an abrupt heterojunction is formed.

Substrates subjected to the cleaning procedure without subsequent epitaxial growth do not exhibit any SDH oscillations, demonstrating that the 2DEG is not created by substrate cleaning artefacts. The 2DEG charge density seems to be affected by the thickness of the α-Sn film in a complex manner: for film thickness below 3000 Å it increases with overlayer thickness but it shows an opposite trend when the overlayer thickness exceeds 3000 Å. Similarly, on reducing the thickness of a 3000 Å α-Sn film by etching in HCl for 2 min, the total charge density falls by about 10%. One possibility is that the shape of the potential well may change because the dielectric constant of the α-Sn films varies with film thickness [14]. Dielectric effects can also alter the potential of the well via the image force. We detect no SDH structures from films grown on n-type InSb substrates even using field modulation techniques. However, the effect may have been masked by the high conductance of the n-InSb substrate.

One important question remaining to be answered is the origin of the 2DEG. Quantum size effects cannot be fully responsible for the 2DEG: for a square well, the difference of the consecutive subband energies as well as their occupancy difference should increase with decreasing film thickness. Fig. 1b indicated that the reverse trend is true for α-Sn/InSb films and that the ratio of the subband occupancies E_n/E_{n+1} are relatively constant (~ 1.5) for both thin and thick films [3].

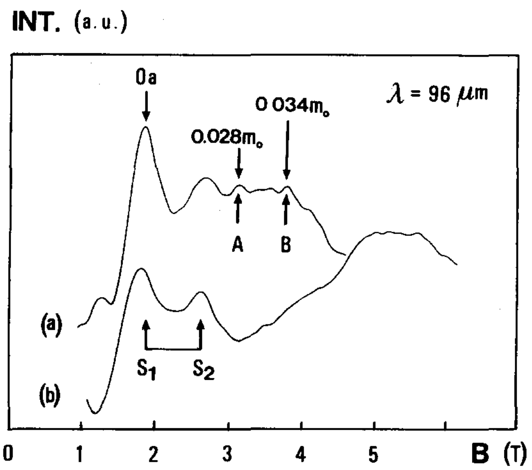

Fig. 3. The magneto-optical spectrum for (a) 3000 Å and (b) 200 Å thick α-Sn films grown on p-InSb(100) surface. Laser wavelength = 96 μm. The two peaks S1 and S2 are due to the bound hole (s–p) transitions of the p-InSb while the structures A and B are the cyclotron resonances of carriers in the bulk α-Sn film.

Hall measurements indicated that the apparent bulk carrier concentration of α-Sn films grown on low doped p-InSb(100) is of the order of 10^{17} cm^{-3}. At 4.3 K the Fermi levels for the α-Sn and p-InSb are about 20 and 1 meV above the valence bands of the two materials, respectively. Using the valence band offset of 400 ± 75 meV determined by core-level photoemission studies [15], the vacuum Fermi level of α-Sn would be about 420 ± 75 meV above that of InSb. In view of the narrow bandgap and high carrier concentration of the α-Sn layer, most of the band bending will occur in the wider bandgap InSb (250 meV) and the space charge region could extend several thousand ångströms into the bulk. However, a large concentration of interface states, which should vary with surface orientation, will alter the interfacial potential substantially.

3.3. Magneto-optical results

In order to understand the nature of the 2DEG as well as the properties of the carriers present in the bulk of the α-Sn films, high resolution magneto-optical experiments have been performed in the Faraday configuration. Fig. 3 shows the photoconductivity response from 3000 and 200 Å thick overlayers at a laser wavelength of 96 μm. The two structures labelled S_1 and S_2 are the bound hole transitions (s–p) due to the Cd dopant in the InSb substrate [3]. A broad hump at about 5 T can be observed in the thinner film which quenches when the film thickness increases. Similar behaviour is observed in the 118 μm wavelength spectrum. This structure is tentatively attributed to the 2DEG. On the other hand, two sharp peaks labelled A and B, which are also found in the 118 μm wavelength spectrum are identified as cyclotron structures having effective masses of $0.028m_0$ and $0.034m_0$, respectively. Note that these structures are almost unobservable in the thin sample, indicating that they are due to the α-Sn overlayer. An effective mass of $0.033m_0$ with broad resonance structure has also been observed in α-Sn/CdTe samples [16].

4. Conclusion

Surfaces of α-Sn(100) films exhibit two-domain (2×1), $p(2 \times 2)$ and $c(4 \times 4)$ structures depending on the film thickness while $(\bar{1}\bar{1}\bar{1})$ films show (3×3) and (3×1) reconstructions.

The properties of the 2DEG at the interface of α-Sn/InSb have been investigated by Shubnikov–De Haas measurements and data from various thicknesses of α-Sn overlayers grown on InSb(100) and $(\bar{1}\bar{1}\bar{1})$ indicate that cross-doping may not be the only factor responsible for the occurrence of the 2DEG. Although the α-Sn overlayers do affect the properties of the 2DEG, it is believed that interface states due either to the formation of a dipole layer when a nonpolar material is grown on a polar substrate [17,18], or to the localized states induced by the epilayer [19], may be important factors responsible for the occurrence of the 2DEG. In order to reveal the interfacial energy band diagram of this nonpolar/polar system, further information concerning the electronic and structural properties of α-Sn layers in the interfacial region is needed.

Due to the overlapping of transition energies of bound holes and cyclotron resonance structures, magneto-optical data could not unambiguously indicate the presence of carriers due to the InSb substrate. On the other hand, carriers due to α-Sn films with effective masses ranging from $0.028m_0$ to $0.034m_0$ are observed from thick film samples.

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