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**Characterization of InMnSb epitaxial films for spintronics**

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**Abstract.** Dilute (III, Mn)V ferromagnetic magnetic semiconductors have potential applications in spintronic devices as magnetic field sensors, spin transistors and reconfigurable logic devices. For such applications ferromagnetism at room temperature is a practical requirement. Previous work has shown that In$_{1-x}$Mn$_x$Sb films grown on GaAs (100) substrates by atmospheric pressure metalorganic vapour phase epitaxy were single phase and had high temperature ferromagnetism for Mn concentrations up to 2%. Here we present a study of InMnSb thin films at higher Mn concentration (10 at%) showing ferromagnetic properties at room temperature. Aberration corrected and analytical (scanning) transmission electron microscopy techniques were used to study the structure and elemental distribution of the In$_{1-x}$Mn$_x$Sb/GaAs system. The crystalline quality of the film and the presence of phase separation is evaluated in the context of a cluster-mediated ferromagnetism model.

1. Introduction

Dilute magnetic semiconductors have attracted a lot of attention in recent years for their properties that potentially enables spin electronic applications including, spin transistors, logic devices and magnetic field sensors [1]. In fact, the replacement of transition metals atoms within a semiconductive matrix results in adding magnetic properties to the electronic properties of the matrix. In particular, the substitution of Mn atoms to group III in a III-V semiconductor introduces at the same time free holes for conduction as well as local magnetic moments. The In$_{1-x}$Mn$_x$Sb alloys are of special interest due to their light holes, small energy gap, and considerably higher carrier mobility than other III-Mn-V ferromagnetic semiconductors. These properties make In$_{1-x}$Mn$_x$Sb alloys ideal candidates for infrared spin photonics [2]. A fundamental requirement for the practical realization of spintronic devices is the Curie temperature ($T_c$) of the magnetic semiconductor to be above room temperature. Recent theory predicts that (III-V)Mn semiconductors, where Mn forms a shallow level, can lead to high $T_c$ [3]. The natural candidate for a ferromagnetic alloy grown on GaAs would be the (Ga, Mn)As alloy. This system avoids the strain effects at the film/substrate interface, however its $T_c$ is below 180K [4]. (In, Mn)Sb alloys, instead, have been reported to have higher $T_c$ than (Ga, Mn)As. In particular single phase InMnSb films have been demonstrated to have high temperature ferromagnetism for Mn concentrations up to 2% [5]. The low solubility of Mn has been already shown in (Ga, Mn)As dilute semiconductors where at high Mn concentration several secondary phase precipitates are formed [6]. Similarly, the (In, Mn)Sb system is expected to phase separate at high Mn concentrations. While in
general secondary phase formation is detrimental to the functional properties of spintronic devices, for the case of InMnSb some of the secondary phases, such as MnSb and Mn$_2$Sb, are still ferromagnetic with $T_c$ well above room temperature. In such semiconductors, following the quasi-chemical model for cluster-mediated ferromagnetism described in [3], it is predicted that the overall $T_c$ of the alloy should tend towards the highest $T_c$ of its constituent phases. In this work we study the crystalline structure of highly doped (10 at% of Mn) InMnSb films grown on GaAs and showing a $T_c$ above 500K [7].

2. Experimental
InMnSb thin film were deposited on semi-insulating GaAs (100) substrates in an atmospheric pressure metalorganic vapour phase epitaxy (MOVPE) chamber modified as reported in [8]. Reactants were optimized to obtain a nominal 10at% Mn concentration at the growth temperature of 420°C. TEM cross-section samples were obtained by mechanical polishing and room temperature Ar$^+$ Ion milling using Gatan PIPS up to electron transparency. Microstructural and chemical analyses were undertaken using a conventional 200kV JEOL 2011 Transmission electron microscope and a CEOS double aberration corrected JEOL 2200FS (scanning) transmission electron microscope [9] operating at 200keV, equipped with an in-column Omega type electron energy filter and a Thermo energy dispersive X-Ray (EDX) Si(Li) detector.

3. Results and Discussion
The Bright Field (BF) TEM image in figure 1(a) shows an overview along the [110] zone axis of the InMnSb film. The film has a cubic structure, with the following epitaxial relationship with respect to the GaAs substrate: [001]$_{\text{film}}$ || [001]$_{\text{GaAs}}$ and [110]$_{\text{film}}$ || [110]$_{\text{GaAs}}$, as shown by the selected area diffraction (SAD) pattern in figure 1(b) taken from an area that includes both the film and the substrate.

![Figure 1](image_url)

*Figure 1. (a) Overview BF image taken along the [110] zone axis. Labels G and T indicates film grains and twin crystal sections. (b) SAED pattern from an area including both the film and the substrate. The white dashed box refers to figure 2(a).*

The film consists of micrometer size grains separated by twin crystal regions as indicated in figure 1(a). Each one of the grains has a slightly different tilt respect to the substrate. This results in a small angular shift of the Bragg spots in the diffraction pattern. Figure 2(a) clearly shows, close to each other, the InMnSb multiple Bragg spots that correspond to the same Bragg reflection. Figure 2(b) shows an intensity profile across the (00-4) spots of InMnSb along a direction perpendicular to the [001] direction of GaAs. Each one of these peaks, labeled A, B, C, corresponds to different grains with +1.2 (A), -0.5 (B), and 2.5 (C) degrees tilt respect to the substrate.
The high resolution transmission electron microscopy (HREM) image in figure 2(c) shows the edges of two grains labelled as grain 1 (G1) and grain 2 (G2). The grains are separated by a twin boundary region with two mirror planes 10 nm apart and are rotated by 3 degrees with respect to each other. The twin boundary structure is highlighted in the diffractogram inset in figure 3(d). The tilt of the grains, the twin crystal sections together with a high density of misfit dislocations at the film/substrate interface contribute to reduce the strain caused by the 14% lattice mismatch between the substrate and the film. As expected, due the high level of Mn doping, the InMnSb film was found to have undergone phase separation. In particular, two main types of precipitates were found, interfacial structures extending into the substrate and structures buried within the film. Typical secondary phases are shown in the BF field TEM image in figure 3(a). V-shaped structures at the interface are indicated by white arrows. In figure 3(b) is reported an energy-filtered TEM (EFTEM) map of Mn. The map was obtained using the three window method [10] applied to the Mn-M$_{2,3}$ electron energy loss image electron energy loss edge at 49eV. Two pre-edge images (at 35 eV and 43) and one post-edge image (at 59 eV) were acquired. The width of the omega filter energy slit was set to 8 eV. Each 512 by 512 pixel image was recorded with an acquisition time of 45s. The images were cross-correlated to correct for spatial drift of the sample during the acquisition. The background component was calculated from the pre-edge images and removed from the post-edge image using the EFTEM analysis routine of Gatan Digital Micrograph™. Both interfacial and buried secondary phases are Mn-rich. This result is confirmed by annular dark field (ADF) STEM imaging and EDX analyses as shown in figure 3(c). The atomic number (Z) contrast dependence of the intensity in ADF images highlights the presence of secondary phases. Given the difference in atomic number of the InMnSb alloy components (Z$_{\text{In}}$= 49, Z$_{\text{Mn}}$=25 and Z$_{\text{Sb}}$=51), Mn-rich areas appear darker (box1 in figure3(c)) than InSb, and Sb-rich areas (box3 in figure3(c)) brighter that the GaAs substrate (Z$_{\text{Ga}}$=31 and Z$_{\text{As}}$=33). EDX analyses of the film were quantified using the thin film approximation with calculated Cliff-Lorimer factors. Elemental concentrations were obtained from the intensity of respectively Mn-K, Ga-K, As-K, In-L, and Sb-L peaks. Quantified results are reported in the table of figure 3(c). Mn was mainly found to be contained in the secondary phases while the surrounding matrix consists mainly of InSb. Interfacial secondary phases extending into the substrate were found to contain both Mn and Sb indicating Ga substitution with Mn and Sb, and formation of non-stoichiometric MnAsSb.

Figure 2. (a) SAD pattern analysis from the area indicated in figure 1(b). (b) Intensity profile across the InMnSb (00-4) Bragg reflection. (c) HREM image of an area including the edges of two Grains G1 and G2 separated by a twin boundary section labelled T. (d) Diffractogram with highlighted the mirrored structure characteristic of the twin boundary.
Figure 3. (a) BF field TEM image showing secondary phases buried within the film and at the substrate interface (indicated by white arrows). (b) Mn EFTEM map showing secondary phase. (c) ADF STEM image showing the two types of secondary phases. Boxes refer to the EDX analyses reported in the table on the right.

4. Conclusions
Analytical electron microscopy techniques were applied to study the structure and the elemental distribution in \text{In}_{0.8}\text{Mn}_{0.2}\text{Sb}/\text{GaAs}. The lattice mismatch between the deposited film and the substrate was found to be compensated by organizing the film in micrometer size grains interconnected by thin twin boundaries and through misfit dislocations at the substrate interface. The high Mn concentration was found to lead to Mn-rich secondary phases buried within an InSb matrix and at the interface with the substrate. Here the secondary phases are formed by Ga substitution with Mn and Sb resulting in non-stoichiometric \text{MnAsSb}. These findings indicate that Mn-rich precipitates contribute to the high Curie temperature observed in the two-phase semiconductor films.

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References