MBE growth of ferromagnetic single crystal Heusler alloys on (001)Ga$_{1-x}$In$_x$As

J.W. Dong$^a$, J. Lu$^a$, J.Q. Xie$^a$, L.C. Chen$^a$, R.D. James$^b$, S. McKernan$^c$, C.J. Palmstrøm$^a$, *

$^a$Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455, USA
$^b$Department of Aerospace Engineering and Mechanics, University of Minnesota, Minneapolis, MN 55455, USA
$^c$Center for Interfacial Engineering, University of Minnesota, Minneapolis, MN 55455, USA

Abstract

Ferromagnetic Ni$_2$MnGa and Ni$_2$MnGe have been grown on GaAs(001) and Ni$_2$MnIn on InAs(001) by molecular beam epitaxy. In situ reflection high energy electron diffraction, ex situ X-ray diffraction and transmission electron microscopy selected area electron diffraction indicate the growth of pseudomorphic single crystal (001) Ni$_2$MnGa on (001) GaAs. Superconducting quantum interference device magnetometry measurements show the films to be ferromagnetic with in-plane magnetization and Curie temperatures of $\sim$340, $\sim$320, and $\sim$290 K for Ni$_2$MnGa, Ni$_2$MnGe and Ni$_2$MnIn, respectively.

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1. Introduction

The recent ability of making ferromagnetic semiconductors [1] and the observation that the spin coherence length can be as large as 100 $\mu$m in a semiconductor [2] have stimulated the integration of magnetic materials and semiconductors to make novel devices that utilize the carrier spin as well as its charge [3]. A key element to develop “spintronic” devices is the ability to electrically inject spin polarized carriers into an otherwise unpolarized semiconductor. Recently, two groups have reported the spin polarized injection of electrons from a magnetic semiconductor at low temperature [4,5]. There has been only limited success injecting carriers with large spin polarization into a semiconductor from a ferromagnetic metal contact [6,7]. This is believed to result from poor interface properties, both electrical and magnetic, and from the lack of sufficient spin polarization of the ferromagnet at the Fermi level [8]. For diffusive transport, recent theories suggest that close to 100% spin polarization
at the Fermi level (half-metallic ferromagnetism) will be needed for significant spin injection [9]. Although a number of ferromagnetic compounds have been predicted theoretically to be half-metallic, experimentally 100% spin polarization at the Fermi level has not been observed [10]. However, contacts utilizing ballistic transport are more likely to inject high spin polarization [11]. For spin-polarized ballistic transport, matching of the band structure of the ferromagnetic metal and the semiconductor is very important [12,13]. Kilian and Victora [13] predicted that the band structure alignment between the Heusler alloy Ni$_2$MnIn and InAs would enhance the injection of the minority spins. Their calculations show that the minority spins are situated at the $\Gamma$ point in Ni$_2$MnIn and the majority spins are far away from the $\Gamma$ point. This is what makes the transmission coefficient much larger for the minority than the majority spins, suggesting that Heusler alloys may be a good choice as a ferromagnetic contact for spin injection. Recently the epitaxial growth of Ni$_2$MnGa [14–16] and Co$_2$MnGe [17] on GaAs(001) have been reported.

Fig. 1 shows a plot of the lattice parameter versus transition element for a number of Heusler alloys with the $L2_1$ crystal structure. The mismatch for Ni$_2$MnGa, Ni$_2$MnGe and Co$_2$MnGe to GaAs are 3%, 0.83% and 1.5%, respectively. Ni$_2$MnIn is closely lattice matched to InAs (mismatch $\sim$0.16%). In this paper, we report on the amazing compliancy of Ni$_2$MnGa grown on GaAs(001), the molecular beam epitaxial (MBE) growth of Ni$_2$MnGe on GaAs(001) and Ni$_2$MnIn on InAs(001) and their magnetic properties.

2. Experimental

In order to minimize Mn contamination of the compound semiconductor growth chamber, the GaAs and InAs buffer layers were grown in a separate growth chamber from the Heusler alloy growth. GaAs layers were grown on GaAs(001) substrates at 580°C in a modified VG V80H MBE system. The samples were arsenic capped by allowing the sample to cool overnight facing the liquid nitrogen cryopanel with a chamber pressure $\leqslant 5 \times 10^{11}$ mbar. The arsenic capping was performed with a sample temperature $<-10^\circ$C using an As$_4$-flux. In the case of buffer layers for Ni$_2$MnGa growth an additional six monolayer thick Sc$_{0.3}$Er$_{0.7}$As was grown at 350°C prior to the arsenic capping. The InAs buffer layers were grown at 460°C on InAs(001) substrates and arsenic capped in a similar manner to the GaAs buffer layers.

After arsenic capping, the samples were removed from the MBE system and immediately remounted on Mo sample holders for a Riber 1000 MBE system and loaded into the load lock. After inserting the arsenic capped sample into the Riber 1000 growth chamber, the arsenic cap was removed by heating to 300°C for 10 min. Reflection high energy electron diffraction was used to ensure complete removal of the As-cap prior to Heusler alloy growth. The growth procedures used varied for the different Heusler alloys. For Ni$_2$MnGa growth on the six monolayer thick Sc$_{0.3}$Er$_{0.7}$As interlayer, the sample was cooled from 300°C used for the arsenic cap removal to 200°C. Alternate layer epitaxy was used to grow five sequential monolayers of Ni and Mn+Ga to yield a Ni/Mn-Ga/Ni/Mn-Ga/Ni monolayer growth sequence at 250°C. Following this growth sequence, the sample was heated to 300°C prior to the subsequent growth of Ni$_2$MnGa by codeposition to a total thickness of 900 Å. The Ni$_2$MnGa growth rate was $\sim$0.09 µm/h. This growth procedure is similar to that described in reference [14]. The same growth procedures were used to grow Ni$_2$MnGe directly on GaAs without a Sc$_{0.3}$Er$_{0.7}$As interlayer. For Ni$_2$MnIn direct codeposition on the arsenic decapped InAs(001) was done at 80°C. After the growth, the
sample was annealed at 150°C for 10 min. For all samples, a thin ∼20 Å thick Al capping layer was deposited at room temperature prior to removal from the vacuum system.

3. Results and discussion

The X-ray diffraction pattern from the Ni$_2$MnGa (900 Å)/Sc$_{0.3}$Er$_{0.7}$As/GaAs(0 0 1) heterostructure shows sharp (0 0 2) and (0 0 4) diffraction peaks for both Ni$_2$MnGa and GaAs with the intensity of the (0 0 4) peak being greater than the (0 0 2), indicating a high quality single crystal epitaxial film. From these data, the out-of-plane lattice parameter was determined to be 6.18 Å, which is similar to that found for thinner (300 Å) pseudomorphic films [14]. The pseudomorphic growth is attributed to a new stress-induced Martensitic phase [14]. Rutherford backscattering channeling minimum yield was 6.5%, which is also indicative of an epitaxial single crystal film.

A high resolution transmission electron microscopy (TEM) micrograph of the sample is shown in Fig. 2. The abrupt six monolayer thick Sc$_{0.3}$Er$_{0.7}$As layer is clearly visible. Vertical lattice fringes can also be seen in the Ni$_2$MnGa film. In addition, there is some contrast from layers in {112} type orientation. Although, detailed analysis has not been made, this contrast is likely to come from stress or composition variations in the film. The inset in the figure shows a convergent beam electron diffraction pattern obtained from both the GaAs and the Ni$_2$MnGa film. The discs in the diffraction pattern are clearly split in the vertical direction (see upper discs), while no splitting is observed in the horizontal direction. These data are consistent with the Ni$_2$MnGa being tetragonally distorted and pseudomorphic on the GaAs substrate. This is a surprising result and corresponds to a thickness ≥ 50 times larger than the expected critical thickness for a system with 3% mismatch. These results are supported by first principle calculations of the mechanical properties of Ni$_2$MnGa, which show that Ni$_2$MnGa has a very wide energy minimum as a function of c/a ratio [18].

Fig. 3 shows reflection high energy electron diffraction (RHEED) patterns from the Ni$_2$MnGe and Ni$_2$MnIn surfaces after growth. The $2 \times 2$ reconstruction obtained from the Ni$_2$MnGe with the electron beam incident along the (1 1 0) and (0 1 0) directions of the GaAs suggest a (2×2) surface reconstruction, which is the same as that observed for Co$_2$MnGe [17]. X-ray diffraction data from Ni$_2$MnGe films show an out of plane lattice parameter of 5.896 Å, which is slightly larger than the bulk value ($a_0 = 5.70$ Å), indicating possible pseudomorphic growth. The RHEED pattern with the electron beam along (1 1 0) obtained from
the Ni$_2$MnIn surface (Fig. 3b) indicates that the surface unit cell is half that expected for the $L_2_1$ Mn–In terminated surface with a weak $3 \times$ reconstruction. The patterns would be consistent with a Ni terminated surface, a disordered Mn–In terminated surface, or a different crystal structure.

The out of plane lattice parameter determined from the diffraction scan (Fig. 4) assuming the cubic $L_2_1$ structure is 6.15 Å, which, as in the case of Ni$_2$MnGe, is slightly larger than the bulk value (6.068 Å). However, for the $L_2_1$ structure, the X-ray diffraction intensity for the (0 0 4) peak should be higher than the (0 0 2), which is not the case. This suggests that the Ni$_2$MnIn is growing in a different crystal structure. One possibility would be a disordered $L_2_1$ structure, which would result in a CsCl structure. In this case the (0 0 2) and (0 0 4) peaks should be relabeled as (0 0 1) and (0 0 2), respectively. The lattice parameter of NiIn is 3.06 Å, which is close to half the Ni$_2$MnIn lattice parameter determined from Fig. 4. However, in the case of NiGa growth on GaAs(0 0 1), the hexagonal phase Ni$_2$Ga$_3$ can be grown with a pseudo cubic surface structure [19]. Hence, an alternative explanation would be the growth of an (1 0 1 1) oriented hexagonal phase with the Ni$_2$In$_3$ structure and the Ni$_2$MnIn peaks would be identified (1 0 1 1) and (2 0 2 2) for the ones labeled (0 0 2) and (0 0 4) in Fig. 4, respectively. The expected planar spacing is 3.09 Å for Ni$_2$In$_3$, which is close to that observed. Further studies are required to clarify the Ni$_2$MnIn crystal structure, composition and possible pseudomorphic growth on InAs.

An in-plane magnetic hysteresis loop obtained from Ni$_2$MnIn is shown in Fig. 5a. Similar loops were obtained from Ni$_2$MnGa and Ni$_2$MnGe. No strong in-plane anisotropy was found for the Ni$_2$MnGa films. Fig. 5b shows the relative magnetization versus temperature obtained using a superconducting quantum interference device magnetometer. From these data, the Curie temperatures, $\sim$290, $\sim$320, and $\sim$340 K, and low temperature saturation magnetization, $\sim$200, $\sim$400, and $\sim$450 emu/cm$^3$, were determined for Ni$_2$MnIn, Ni$_2$MnGe and Ni$_2$MnGa, respectively.

4. Conclusions

Heteroepitaxial ferromagnetic Ni$_2$MnGa/GaAs (0 0 1), Ni$_2$MnGe/GaAs (0 0 1), and Ni$_2$MnIn/InAs (0 0 1) structures have been grown by molecular beam
epitaxy. The films show in-plane magnetization with Curie temperatures close to room temperature.

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