Mn$_2$As Films and MnGa/Mn$_2$As/MnGa Trilayers on GaAs(001)

W. Van Roy, H. Akinaga,* and S. Miyanishi**

Joint Research Center for Atom Technology–Angstrom Technology Partnership (JRCAT-ATP), 1-1-4 Higashi, Tsukuba, Ibaraki 305-8562

*Joint Research Center for Atom Technology–National Institute for Advanced Interdisciplinary Research (JRCAT-NAIR), 1-1-4 Higashi, Tsukuba, Ibaraki 305-8562

**Institute of Materials Science, University of Tsukuba, 1-1-1 Tennoudai, Tsukuba, Ibaraki 305-0006

We have succeeded in growing for the first time Mn$_2$As thin films on GaAs(001) using molecular beam epitaxy. Bulk Mn$_2$As is a layered antiferromagnetic material. Single-crystalline films with the $c$ axis perpendicular to the substrate were grown directly on the GaAs substrates, as well as on MnGa seed layers, where MnGa is a ferromagnet. We also grew MnGa/Mn$_2$As/MnGa trilayers, and we found interlayer coupling with oscillatory spacer layer thickness dependence. The coupling strengths at room temperature were up to $-0.63 \text{ mJ/m}^2$ (antiferromagnetic) and $+0.53 \text{ mJ/m}^2$ (ferromagnetic). The oscillation period was between 0.95 and 1.55 nm, which is close to the period of the antiferromagnetic order in Mn$_2$As (1.25 nm). The sign of the magnetoresistance was correlated with the sign of the interlayer coupling in the same way as was found earlier for MnGa/(Mn, Ga, As)/MnGa trilayers.

Key words: ferromagnetic/antiferromagnetic/ferromagnetic multilayer, oscillatory interlayer coupling, magnetoresistance, Mn$_2$As, MnGa$_{1-x}$Ga$_x$ (x = 54, 60), molecular beam epitaxy

1. Introduction

The integration of magnetic materials into semiconductor technology offers the possibility of using the electron spin as an additional degree of freedom, and many interesting device concepts have already been proposed. Multilayers of ferromagnetic metals and semiconductor spacer layers show interlayer coupling and giant magnetoresistance similar to systems with a metallic spacer layer. In addition, the coupling through a semiconducting spacer layer can be induced by external excitations such as heat, and possibly also light and the injection of an electric current. However, the growth of high quality ferromagnet/semiconductor multilayers remains a technological challenge. The Fe/Si system, which has been studied most, suffers from a high chemical reactivity even at room temperature, and intermixing could only be suppressed by depositing the films at 40 K and performing all measurements without heating up the samples to room temperature.

We have already shown the growth of several Mn-based ferro- and ferrimagnetic films on III-V substrates by molecular beam epitaxy (MBE). We have also reported MnGa/(Mn, Ga, As)/MnGa trilayers. Where two ferromagnetic MnGa films are separated by a nonmagnetic spacer layer. Although the spacer layer consisted nominally of the semiconductor GaAs, we observed a large diffusion of Mn into the spacer layer, and a mixture or intermediate phase between GaAs and the antiferromagnetic metal Mn$_2$As was formed. To gain a clear understanding of the magnetic behaviour, it is essential to grow well characterized single phase spacer layers. In this report we will show the epitaxial growth of single crystalline Mn$_2$As films and MnGa/Mn$_2$As/MnGa trilayers on GaAs(001), and discuss the interlayer coupling and the magnetoresistance of the trilayers.

δMnGa has a tetragonal crystal structure with a strong magnetocrystalline anisotropy forcing the magnetic moments along the $c$ axis (Fig. 1). Thin films grown epitaxially on GaAs(001) by MBE have their $c$ axis perpendicular to the surface. Bulk Mn$_2$As has the tetragonal Cu$_2$Sb crystal structure, which is a layered structure consisting of alternatingly one Mn plane and two (Mn + As) planes perpendicular to the $c$ axis (Fig. 1). When the unit cell is rotated through 45° around the $c$ axis, the atom stacking in the basal Mn plane is identical to the stacking in δMnGa, with a lattice mismatch of only $-1.3\%$. The lattice mismatch with GaAs ($a_0/\sqrt{2} = 0.400$ nm) is $-4.9\%$. Mn$_2$As has a layered antiferromagnetic structure where the orientation of the moments reverses three times within

![Fig. 1 Crystal structures of Mn$_2$As and MnGa.](image-url)
the unit cell. The magnetic unit cell is twice as high as the structural cell.

Since MnGa and Mn$_2$As are both magnetic, one may expect a direct exchange coupling between them, which may influence the coupling between two MnGa films separated by a Mn$_2$As spacer layer. However, the moments of MnGa (in the basal plane) and Mn$_2$As (parallel to the c-axis) are orthogonal to each other, and the precise interaction cannot be predicted easily.

2. Experimental Procedure

The samples were grown by molecular beam epitaxy (MBE) in a modified Riber 32P system equipped with standard effusion cells for Ga and Mn, and a valved cracker cell (EPI) for As which allows very fast transients in the As flux. The base pressure of the chamber was in the low 10$^{-11}$ Torr range, and remained less than 2×10$^{-10}$ Torr during the growth of MnGa. All Mn$_2$As films reported here have been grown with a beam of cracked As$_2$ molecules. During the growth of the MnGa films, the cracking zone was cooled down again to limit the background pressure. A linear shutter in front of the substrate was used to prepare wedge-shaped spacers. We used epi-ready GaAs(001) substrates on which a 100 nm GaAs buffer layer was deposited in a separate growth chamber. The structural and compositional properties have been determined by reflection high energy electron diffraction (RHEED), semi in-situ Auger electron spectroscopy (AES), and X-ray diffraction (XRD) θ-2θ scans. The magnetic properties have been evaluated by transport measurements (magnetoresistance and extraordinary Hall effect EHE) at room temperature (RT) in the van der Pauw configuration (CIP). The applied field was perpendicular to the surface.

3. Results and Discussion

3.1 Mn$_2$As single films

The growth of Mn$_2$As films has been performed directly on the GaAs(001) substrate, as well as on top of 10–20 nm thick MnGa films. Because of the good matching between the basal planes (Fig. 1), MnGa presents an excellent seed layer for Mn$_2$As, and films grown in this way had a very good quality. We used a beam equivalent pressure (BEP) ratio of $\phi$$_{\text{As}}$: $\phi$$_{\text{Mn}} = 3:1$, a growth rate of $\sim$60 nm/h, and substrate temperatures in the range $T_{\text{sub}} = 200$ to $300^\circ$C. At $T_{\text{sub}} = 200^\circ$C the nucleation of Mn$_2$As was polycrystalline, but in the range $T_{\text{sub}} = 250$–$300^\circ$C we obtained single crystalline films. At $T_{\text{sub}} = 250^\circ$C the crystal quality slowly degraded for film thickness larger than $\sim 10$ nm, as indicated by RHEED. At $T_{\text{sub}} = 300^\circ$C the RHEED pattern showed no sign of degradation and remained very steady up to at least 70 nm. XRD measurements confirm that films grown at 300$^\circ$C had the highest quality. The films had their c axis perpendicular to the substrate, and the lattice constant was c = 0.6242 ± 0.0005 nm for all samples, indicating that the films were relaxed. For all growth temperatures AES measurements showed roughly a 2:1 ratio between Mn and As, and there was no trace of Ga for thicknesses larger than the information depth of 1–2 nm.

We have also attempted to grow Mn$_2$As directly on the GaAs substrate. As-rich c (4×4) and Ga-rich (4×6) reconstructed surfaces were used, as well as crystalline MnGa templates of 1.5 and 2.5 bilayers thick. In all these cases we observed an interface between the nucleation of Mn$_2$As and hexagonal MnAs when the growth conditions were similar as above (BEP ratio $\phi$$_{\text{As}}$: $\phi$$_{\text{Mn}} = 3:1$, substrate temperature $T_{\text{sub}} = 250^\circ$C). Preliminary results indicated that by using arsenic-poor growth conditions ($\phi$$_{\text{As}}$: $\phi$$_{\text{Mn}} = 1.5:1$), it is possible to nucleate exclusively the Mn-rich Mn$_2$As phase on GaAs c (4×4).

3.2 MnGa/Mn$_2$As/MnGa trilayers

MnGa/Mn$_2$As/MnGa trilayers with wedge shaped spacer layers were grown using the growth conditions that were established for thick Mn$_2$As films on MnGa (BEP ratio $\phi$$_{\text{As}}$: $\phi$$_{\text{Mn}} = 3:1$, $T_{\text{sub}} = 250$–$280^\circ$C). The wedges had a step-like profile with step heights of 1/3 and 1/6 of a unit cell of Mn$_2$As (0.21 and 0.10 nm, resp.) and terrace widths of 4 nm.

For the growth of both MnGa films we used the

![Fig. 2 Magnetotransport properties at RT of a [10 nm Mn$_{0.6}$Ga$_{0.4}$/t nm Mn$_2$As/10 nm Mn$_{0.6}$Ga$_{0.4}$] trilayer with a wedge-shaped spacer. The applied magnetic field was perpendicular to the surface. Two spacer layer thicknesses are shown: t = 0.33 unit cells Mn$_2$As = 0.21 nm [(a), (c)] and 1.00 cell Mn$_2$As = 0.62 nm [(b), (d)]. The top panels show the EHE hysteresis loops, including minor loops where only the magnetization of the soft Mn$_2$Ga$_{0.4}$ film reverses. The shift of the minor loops indicates AF coupling for the 0.33-cell spacer (a) and F coupling for the 1.00-cell spacer (b). The bottom panels show the magnetoresistance, where for clarity only the major hysteresis loops are shown. In the sample with a 0.33-cell spacer and AF coupling, the resistance is smallest when the magnetizations are aligned AP (c). In the sample with a 1.00-cell spacer and F coupling, the resistance is smallest when the magnetizations are aligned P.](image-url)
existing technique involving the deposition of an amorphous template at low temperature \((\leq 20^\circ \text{C})\), solid phase crystallization, and growth of MnGa at \(T_{\text{sub}} = 230^\circ \text{C}\). Both MnGa films had slightly different composition and underwent a different annealing treatment (54 at\% Mn and 10 min at \(T_{\text{sub}} = 340^\circ \text{C}\) for the bottom MnGa layer, 60 at\% Mn and 5 min at \(T_{\text{sub}} = 310^\circ \text{C}\) for the top MnGa layer). This resulted in a different coercive field, and allowed the easy determination of both ferromagnetic (F) and antiferromagnetic (AF) coupling from the hysteresis loops. Finally the samples were covered by a 15 nm thick GaAs cap layer to prevent oxidation.

Figures 2(a), (b) show the EHE hysteresis loops of two parts of the same \(10 \text{ nm MnGaAs}/t \text{ nm MnAs}/10 \text{ nm MnGaAs}\) sample with a MnAs wedge grown at \(T_{\text{sub}} = 250^\circ \text{C}\), for \(t = 0.21 \text{ and } 0.62 \text{ nm (0.35 and 1.0 unit cells, resp.)}\). The minor loops are shifted over a field \(\mu_0 H_e = -0.139 \text{ and } 0.118 \text{ T}\), indicating strong AF and F coupling, resp. The coupling energies \(J = \mu_0 M_{\text{MnGa},1} t_{\text{MnGa},1} H_e = -0.63 \text{ and } +0.53 \text{ ml/m}^2\), where \(M_{\text{MnGa},1} \approx 450 \text{ kA/m and } t_{\text{MnGa},1} = 10 \text{ nm}\) are the magnetization and the thickness of the soft MnGa layer.

Figure 3(a) shows the spacer layer thickness dependence of the interlayer coupling for two wedges grown at \(T_{\text{sub}} = 250^\circ \text{C}\) (step heights 1/3 and 1/6 unit cell), and one wedge grown at 280°C (step height 1/3 unit cell). The two samples grown at 250°C show a very good reproducibility. The oscillation period is \(\sim 1.5 \text{ unit cells (0.95 nm) for the samples grown at } 250^\circ \text{C}\), and \(\sim 2.5 \text{ unit cells (1.55 nm) for the sample grown at } T_{\text{sub}} = 280^\circ \text{C}\). The origin for this difference is not clear at present. It might be due to a difference in crystal quality or stoichiometry of the spacer layer, or possibly also to different sticking coefficients, resulting in different effective layer thicknesses.

The shape of the thickness dependence is similar to the case of non-magnetic metallic spacer layers, and the oscillation period, especially for spacer layers grown at \(T_{\text{sub}} = 250^\circ \text{C (0.95 nm)}\), agrees very well with the periods of \(\sim 1.0 \text{ nm found for many transition metals}\). However, this does not allow us to conclude that the origin of the coupling is RKKY or quantum interference like. The oscillation period of 1.5–2.5 structural unit cells matches approximately with the magnetic unit cell, which is 2 times the structural unit cell. Therefore we cannot rule out the possibility that direct exchange coupling between the magnetic moments of MnGa and MnAs is responsible for the interlayer coupling, in spite of the fact that the easy magnetic axes are orthogonal. The oscillation shows only one component to the interlayer coupling. This is in contrast with our earlier results on (Mn, Ga, As) spacers where the thickness and temperature dependence of the coupling indicated the coexistence of two coupling mechanisms, presumably related with the presence of both GaAs and MnAs in the spacer layers.

Figures 2(c), (d) show the magnetoresistance (only the major hysteresis loop) for the same samples as shown in Figs. 2(a), (b). It is obvious that parallel (P) and anti-parallel (AP) alignment of both magnetizations result in a different resistance. Furthermore, the sign of the resistance difference \(\Delta R = (R_P - R_{AP})\) depends on the sign of the interlayer coupling. For F coupling we observe the smallest resistance for P alignment [Figs. 2(b), (d)], whereas for AF coupling the resistance is smallest for AP alignment [Figs. 2(a), (c)]. The results for the entire wedge are shown in Fig. 3(b), indicating that the magnetoresistance shows exactly the same sine-like dependence on the spacer layer thickness as the interlayer coupling. When the magnetoresistance is plotted versus the interlayer coupling, Fig. 4, the resulting curve is very close to a straight line through the origin. There appears to be a small phase shift between the coupling and the magnetoresistance, but this may an artifact of the measurement or analysis technique.

![Graph](image)

Fig. 3 (a) Dependence of the interlayer coupling strength \(J\) at room temperature on the spacer layer thickness for gallery \(\text{MnGaAs/MnAs/MnGaAs}\) trilayers. The MnAs spacer layers were grown at \(T_{\text{sub}} = 250^\circ \text{C (O, O) and 280°C (O, O)}\) in the form of a wedge. Positive values for \(J\) indicate ferromagnetic coupling. (b) Spacer layer thickness dependence of the magnetoresistance \((R_P - R_{AP})/R_{P,0}\) for one of the samples shown above (O).

![Graph](image)

Fig. 4 Correlation between the magnetoresistance \((R_P - R_{AP})/R_{P,0}\) and the interlayer coupling energy \(J\) for the sample shown in Fig. 3(b). The points nearly fall on a straight line through the origin.


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addition, a reference area with a 6 nm thick spacer layer at the end of the wedge had both interlayer coupling and magnetoresistance $\Delta R$ equal to zero, suggesting that the curve should indeed go through the origin of the plot.

This correlation is identical to what we observed for the (Mn, Ga, As) spacer layers. The physical origin of this correlation is not clear yet, but at least two possible explanations have been proposed. Although the initial formulations of the spin-valve model for giant magnetoresistance did not predict such a correlation, it was shown recently by Barnas and Bruynseraede that quantum size effects also create a component of the magnetoresistance that oscillates with the spacer layer thickness, and that the oscillations have the same period as the indirect exchange coupling. In the special case where the scattering is spin-independent, it was predicted that the magnetoresistance $\Delta R/R$ should change sign in the same way as our measurement results. However, their model makes the implicit assumption that the coupling is of quantum interference or RKKY origin, which is not at all clear in our samples. As we showed above, there is the possibility that the coupling though $\text{Mn}_2\text{As}$ is caused by direct exchange coupling between the magnetic moments of MnGa and MnAs. In the samples with a (Mn, Ga, As) spacer layer, the coexistence of two coupling mechanisms suggests that the coupling is not simply of quantum interference origin, but rather related to the magnetic contamination in the spacer layer. Therefore it is not clear that the model of Barnas and Bruynseraede is applicable to our samples.

Alternatively, we have proposed a frustration magnetoresistance mechanism which is independent of the nature of the interlayer coupling. It relates the frustration of the interlayer coupling to a disturbance of the magnetic moments near the spacer layer and an increase in resistance. That is, when the alignment of both magnetizations satisfies the interlayer coupling (e.g., a multilayer with F coupling is aligned P), all moments are at rest along the single easy axis of MnGa (the c axis), the system is highly ordered, and the resistance is low. When the alignment is opposite to the preferred alignment (e.g., a multilayer with P coupling is aligned AP), then the moments near the spacer layer are tilted slightly to preserve some degree of P alignment. This creates some disorder and increases the scattering of the conduction electrons, resulting in a higher resistance. This mechanism is independent of the specific origin of the interlayer coupling, and it can explain our results also if the oscillations of the interlayer coupling are caused by the internal antiferromagnetic ordering of $\text{Mn}_2\text{As}$.

4. Conclusions

We have succeeded in growing for the first time epitaxial Mn$_2$As thin films on GaAs(001) substrates by MBE. Bulk Mn$_2$As is a layered antiferromagnetic material. Single crystalline films with their c axis perpendicular to the surface can be grown directly on the GaAs substrate, or on a MgGa seed layer, with the highest quality for the latter case. We have used these new Mn$_2$As films to grow ferromagnetic/antiferromagnetic/ferromagnetic MnGa/Mn$_2$As/MnGa trilayers. These trilayers show strong and oscillatory interlayer coupling with strengths up to $-0.63 \text{mJ/m}^2$ (antiferromagnetic) and $+0.53 \text{mJ/m}^2$ (ferromagnetic). The oscillation period of 0.95 to 1.55 nm is close to the period of antiferromagnetic order in Mn$_2$As (1.25 nm). At present we cannot yet attribute the origin of the interlayer coupling to direct exchange coupling between MnGa and Mn$_2$As, or to RKKY type coupling (quantum interference). The sign of the magnetoresistance in the MnGa/Mn$_2$As/MnGa trilayers shows the same correlation with the sign of the interlayer coupling that was observed earlier for MnGa/(Mn, Ga, As)/MnGa trilayers.

Acknowledgment. This work, partly supported by the New Energy and Industrial Technology Development Organization (NEDO), was performed in JRCAT under the joint research agreement between NAIR and ATP.

References


Received Oct. 28, 1997; Accepted Feb. 2, 1998