Transport properties of cubic zero-moment ferromagnetic Mn_2Ru_xGa thin films

Naganivetha Thiyagarajah,[1] Yong-Chang Lau,[1] Karsten Rode,[1] Davide Betto,[1] Kiril Borisov,[1] M. Venkatesan,[1] J. M. D. Coey,[1] and Plamen Stamenov[1]
CRANN, AMBER and School of Physics, Trinity College Dublin, Dublin 2, Ireland

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The spin-dependent transport properties of cubic Mn_2Ru_xGa thin-films are studied as a function of the the Ru concentration, x and the substrate induced strain. We find that at Ru concentration $x \approx 0.7$, which shows practically zero magnetization, the spontaneous Hall effect at room temperature reverses sign and the spontaneous Hall angle is maximized. In addition, a small tetragonal distortion, $c/a \sim 2\%$, allows us to tune the compensation of the two Mn sub-lattices to a preferred temperature at, above or below room temperature. Having two handles on the zero moment half magnetic properties of Mn_2Ru_xGa opens up the possibilities for using this new class of material in various spintronic devices. We also present the initial work on magnetoresistive devices based on pseudo-spin-valves with Mn_2Ru_xGa electrodes.

I. INTRODUCTION

Cubic ferromagnetic Heusler compounds are a family of magnetic materials that often exhibit higher spin polarization at the Fermi level than binary ferromagnetic $3d$ alloys[1]. Some of the materials are half-metals with a gap in the spin-polarized density of states for one spin band which should make them ideal candidates for spin-valves or MTJs[2–5]. Since the prediction by van Leuken and de Groot in 1995, of a half-metallic material with two inequivalent magnetic sub-lattices whose moments cancel out[6], researchers have worked on fabricating such a material. While electronic structure calculations predicted several such compounds[7–9], fabrication of such materials had failed[8,10]. In 2014, Kurt *et. al.* reported the growth of thin films of Mn_2Ru_xGa, which was identified as a zero-moment ferrimagnet with high spin polarization and showed evidence of half-metallicity[11].

Here we report on the temperature, composition and thickness dependent transport properties of Mn_2Ru_xGa (MRG), which are at or near compensation point ($0.6 < x < 1.1$). Addition of Ru to the cubic Mn2Ga structure provides both states (12) and electrons (8). Based on the on the empirical Slater-Pauling rules, should result in perfect compensation for $Mn_2Ru_0 \cdot 5\,Ga$. However the addition of Ru is likely to change both the shape and position of the Mn bands leading to a more complex behaviour of the magnetic and spin-dependent transport properties. In addition the the tetragonal distortion (c/a) can also affect the band structure, hence we also look at strain as a possible control parameter in engineering the Mn_2Ru_xGa fully compensated half metallic system.

II. EXPERIMENTAL TECHNIQUES

MRG films of thickness 4 nm to 70 nm were grown on MgO (001) substrates by dc-magnetron sputtering at 250 °C substrate temperature and base pressure 2×10^{-8} Torr in a Shamrock deposition system. The films were co-sputtered from a Mn2Ga target and Ru target, and the Ru composition was controlled by keeping the Mn_2Ga sputtering power fixed while varying that of Ru. The MRG films were capped with a ~ 2 nm Al_2O_3 layer to prevent oxidation. The crystal structure and lattice parameters were determined by $2\theta - \theta$ and reciprocal space map (RSM) scans using a BRUKER D8 diffractometer with a $Cu - K\alpha$ source. In order to determine the Ru concentration x, we deposited four samples with varying Mn_2Ga target power along with a Ru film. The density and thickness of the samples were then measured using x-ray reflectivity. Based on the measured density and lattice parameters of these 5 control samples, we establish a relation between the x-ray density and the Ru concentration x against which all the samples are calibrated. Magnetization measurements were made using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. The transport measurements were conducted on unpatterned MRG films in a physical properties measurement system (PPMS) for temperatures from 10 K to 400 K. The maximum applied magnetic fields, $\mu_0 H$, for the two systems were 5 T and 14 T respectively. A summary of sample properties is provided in Table I. We also incorporated the MRG as the hard layer into a pseudo-spin-valve with the structure, MgO/ $Mn_2Ru_xGa(15)/Cu(2.8)/[Co(0.2)/Pd(0.6)]_6/Ta(3\,nm)$ in order to investigate the spin dependent transport. The MRG layer was grown at 250 °C, then cooled down to room temperature, and was subsequently transferred to a different deposition chamber for the Cu/[Co/Pd] multilayer deposition. Atomic force microscopy measurements of the MRG film showed a roughness of ~ 0.2 nm, free of pinholes.

TABLE I. Summary of sample properties. The temperature at which full compensation occurs, T_{comp} was defined by the temperature where $\partial\rho_{xy}/\partial T$ reaches its maximum.

Ru x	t nm	$c/a-1$ %	M_s μ_B	T_{comp} K
0.62	70	2.07	−0.09	100–200
0.69	70	1.76	0.03	200–300
0.73	70	1.83	0.07	300–360
0.77	70	1.92	0.09	> 360
1.09	70	1.82	0.07	> 360
1.12	70	1.84	-	387
1.01	34	1.92	-	335
0.98	16	2.24	-	280
1.09	8	2.90	-	214
1.07	4	3.60	-	< 10

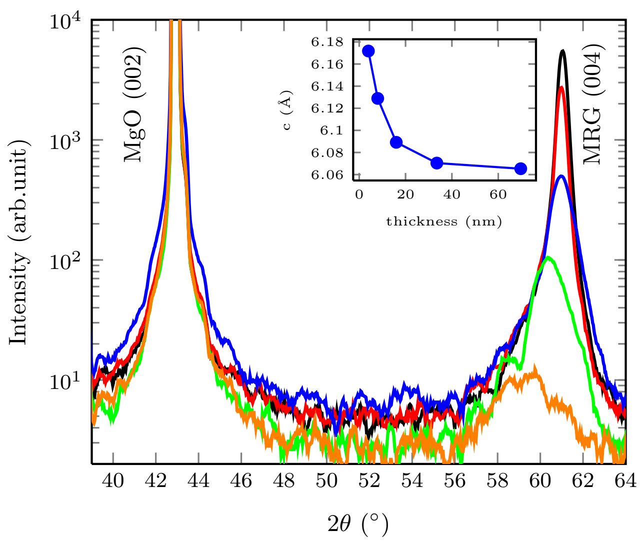

FIG. 1. XRD of thin films of Mn_2Ru_xGa of thickness from 70 nm to 4 nm grown on MgO substrates. Inset shows the dependence of the out-of-plane lattice parameter (c) on the thickness of the film, indicating that the substrate induced strain is increasingly relaxed as the thickness increases.

III. RESULTS AND DISCUSSION

The crystal structure of the cubic MRG films with different thickness and compositions were probed using $2\theta-\theta$ x-ray diffraction (XRD) as shown in Fig. 1. The out-of-plane lattice parameter, c, is between 0.598 nm and 0.618 nm, depending on the Ru concentration and film thickness (insert of Fig. 1). The in-plane lattice parameter, a, determined from reciprocal space maps was found to be 0.596 nm for all samples, which is precisely matched to that of the MgO substrate ($\sqrt{2}a_0\,(\mathrm{MgO}) = 0.5956\,\mathrm{nm}$). This confirms the cubic nature of the MRG films with a slight tetragonal out-of-plane distortion ($c/a-1$ between 1.8% and 3.6%).

Fig. 2 shows the magnetization measurement at 300 K of a typical MRG film of 70 nm near compensation of the magnetic sub-lattices. Clear out-of-plane anisotropy with a large coercivity of 1.2 T is evident. A small soft in-plane component is also clearly visible. As the Ru concentration is reduced from $x = 1.09$, the magnetization reduces, until it falls practically to zero ($12\,\mathrm{kA\,m^{-1}}$ or $0.07\,\mu_\mathrm{B}\,\mathrm{f.u.}^{-1}$) at $x = 0.68$ as shown in Fig. 4(a). We can attribute this to the almost perfect compensation of the two Mn sub-lattices at room temperature. On further reduction of Ru the magnetization again increases. We denote this as a negative magnetization, coincident with the reversal in sign in the room temperature spontaneous Hall effect (SHE) measurements as shown in Fig. 3(a). From the SHE measurements with varying Ru content, we extracted the coercivity, $\mu_0 H_c$, and spontaneous Hall angle (SHA) (defined as ρ_H/ρ) (Fig. 4(b)). As the magnetization approaches zero the coercivity clearly diverges (the sample closest to compensation at room temperature could not be saturated at an applied field of 5 T). The recorded spontaneous Hall angles for samples near compensation ($\sim 5\%$) are about a magnitude larger than those reported for other 3d ferromagnets at room temperature (0.2 to 0.3%)[12] and comparable to SHA recorded for amorphous rare earth transition metal alloys[13]. A high spontaneous Hall angle is indicative of much lower carrier concentrations and a high spin polarization.

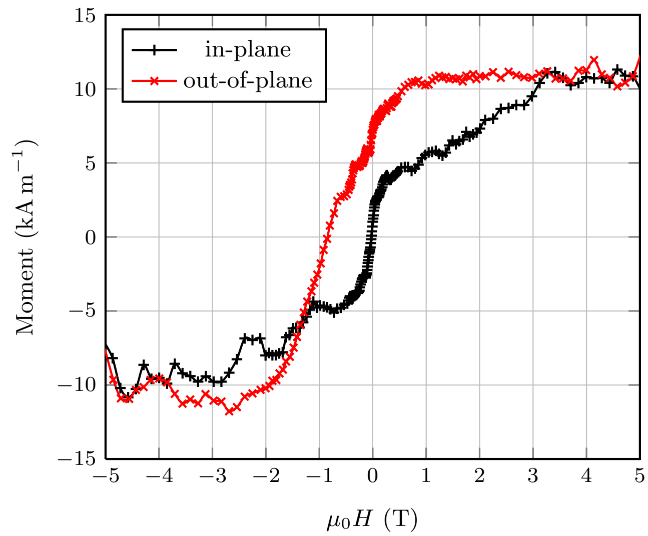

FIG. 2. In-plane and out-of-plane magnetization loops of Mn_2Ru_xGa sample of thickness 70 nm, measured in a SQUID magnetometer at 300 K.

As shown in Fig. 1, the MRG films are increasingly strained as the thickness of the film is reduced. It has been predicted that the magnetization may depend strongly on the lattice distortion since this would have an effect on the interaction between neighbouring atoms. We prepared Mn_2Ru_xGa samples of different thickness from 70 nm down to 4 nm and measured their SHE response at different temperatures from 400 K to 4 K in the PPMS. Fig. 3(b) shows a typical SHE response over the temperature range for the sample of 34 nm thickness. It can be seen that the coercivity diverges to ¿9 T at 350 K and the sign of the SHE loop reverses at 300 K. This indicates that the compensation temperature lies between

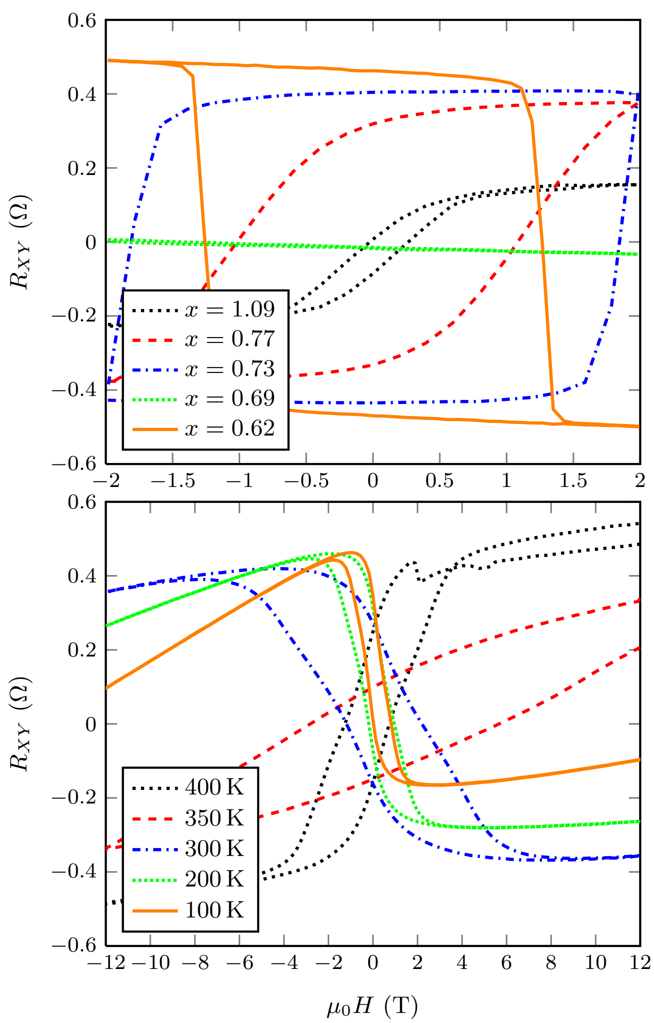

FIG. 3. SHE loops measured of Mn_2Ru_xGa for (a) various Ru compositions ($0.6 < x < 1.1$) and (b) temperatures between 10 K to 400 K, which illustrates the change of sign of the spontaneous hall coefficient between $x = 0.62$ and $x = 0.73$ and 300 K to 350 K respectively.

300 K to 350 K. By plotting the derivative of the Hall resistance w.r.t temperature, $\delta R_{XY}/\delta T$, as shown in Fig. 5(a), it can be seen that this compensation temperature shifts to lower temperatures as the thickness of the MRG is reduced. It is worth noting that the compensation temperature varies with both the Ru content and strain. Since the compensation is achieved by the cancelling out of the moment of the two inequivalent Mn sub-lattices, this shift in compensation temperature may be due to the slightly different temperature dependence of the two sub-lattices. As with samples with different Ru content, the extracted coercivity and SHA show maximum values near the compensation temperature for each thickness as shown in Fig. 5(b) and (c) respectively.

Finally we measured the magnetoresistance (MR) properties of the MRG/Cu/[Co/Pd] samples at different temperatures from 2 K to 300 K. The MR was measured

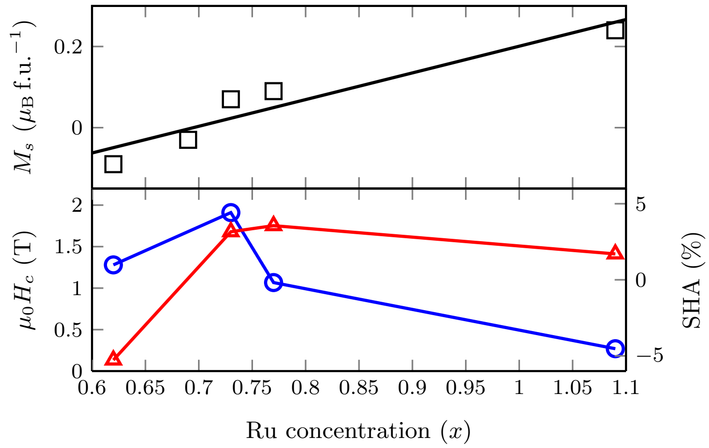

FIG. 4. (a) Extracted magnetization at 300 K (in μ_B f.u.$^{-1}$), for samples of thickness 70 nm with different Ru composition ($0.6 < x < 1.1$). The change of sign of the magnetization was established by SHE sign reversal at compensation. (b) Coercive field and spontaneous Hall angle as a function of Ru composition, extracted from SHE measurements carried out at 300 K, for the same MRG samples as in (a).

on unpatterned films in the current-in-plane configuration. A MR effect was cleared observed at 2 K, and persists even at room temperature as shown in Fig. 6. The observed MR is however quite low (~ 0.15) even at 4 K which may be due to two effects: Firstly considering the transfer between separate deposition chambers for the MRG and Cu/[Co/Pd] layers, some interfacial contamination or oxidation of the Mn can be expected. Secondly, based on the results shown for the thickness dependence of the MRG films, as discussed above, we find that the films are increasingly strained as the thickness of the film is reduced. This causes a variation in the spin-dependent transport properties and compensation of the two magnetic sub lattices, compared to the thicker films. Furthermore we assume that magnetic domains are present in the MRG film as in antiferromagnets; GMR is lost relatively quickly due to domain structuring and imperfect rotation of the magnetisation in the two electrodes, as evidenced by dispersed switching field range as shown in the electronic transport (Fig. 3, and 6).

IV. CONCLUSION

We have shown above that the spin-dependent transport properties of Mn_2Ru_xGa are tuneable with both the Ru concentration x and strain. Recent *ab inito* calculations[14] while providing some insight into the electronic structure, does not give convincing arguments explaining the variation of the transport properties both with varying Ru concentration x and strain. Above we have shown that for a Ru concentration $x \approx 0.7$, which shows practically zero magnetization, the sign of the spontaneous Hall effect is reversed, indicating the rever-

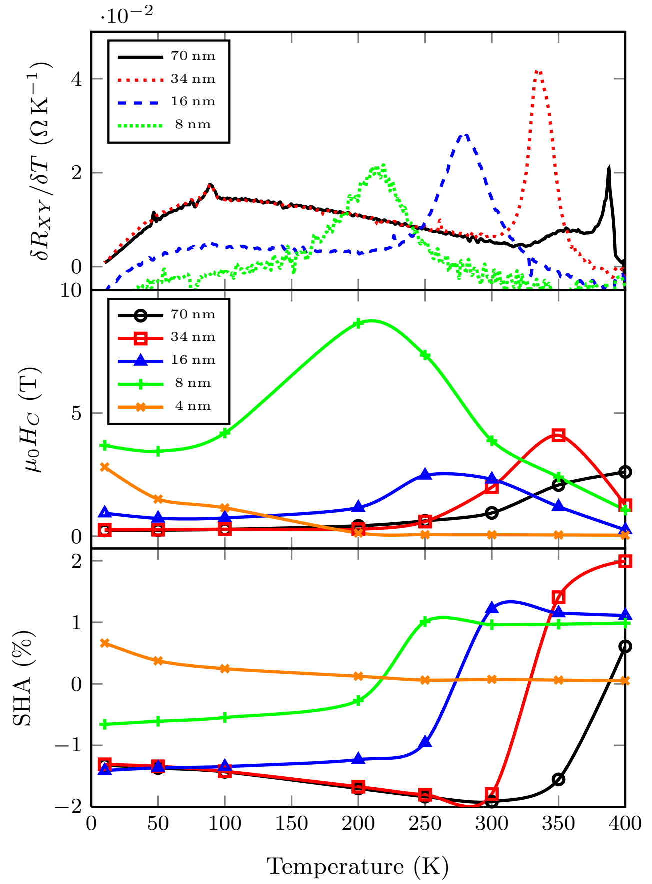

FIG. 5. (a) Variation of compensation temperature with the thickness of MRG film of same Ru concentration, given by the derivative of the resistance w.r.t temperature. The compensation temperature shifts to lower temperatures with decreasing thickness. (b) Extracted coercive field and (c) spontaneous Hall angle as a function of temperature for samples with the same Ru concentration ($x \sim 1.0$) and various thickness from 70 nm to 4 nm.

sal of the majority spin channel. Concurrently the the spontaneous Hall angle is maximised which would imply a reduction in the carrier concentration and high spin polarisation that point towards a half metallic state. We also show that by varying the tetragonal distortion at a particular Ru composition, we can tune the compensation of the two Mn sub lattices to be at a relavant temperature regime at above or below room temperature. The initial demonstration of magnetoresistance in pseudo-spin-valves with an MRG electrode indicates that while we are able to observe a MR effect, further understanding of the magnetic domain and micromagnetic structures are necessary for improving device performance.

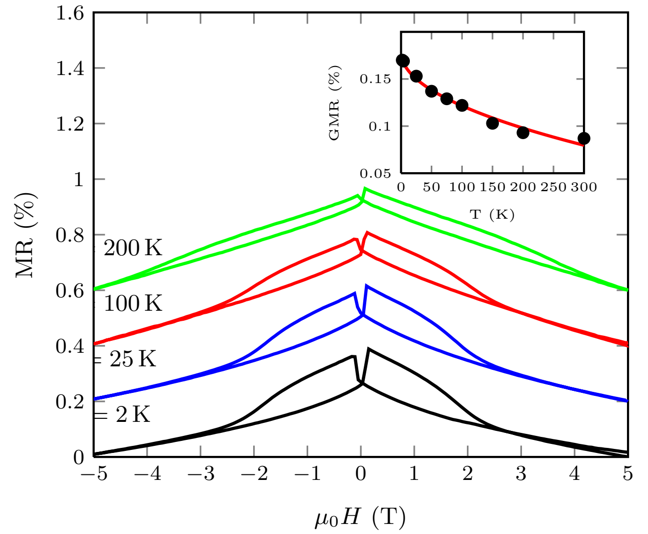

FIG. 6. MR of a pseudo spin valve $Mn_2Ru_xGa(15)/Cu(2.8)/[Co(0.2)/Pd(0.6)]_6/Ta(3\,nm)$ measured at various temperatures. The curves have been offset vertically for clarity. The inset shows the temperature variation of the GMR contribution with a fit to $T^{0.5}$ dependence.

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