Transport properties of cubic zero-moment ferromagnetic Mn_2Ru_xGa thin films

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We have studied the spin-dependent transport properties of cubic Mn_2Ru_xGa thin-films as 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.

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[?,?,?,?] . 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[?] , researchers have worked on fabricating such a material. While electronic structure calculations predicted several such compounds[2–4], fabrication of such materials had failed[3,5]. In 2014, Kurt *et. al.* reported the growth of thin films of Mn_2Ru_xGa (MRG), which was identified as a zero-moment ferrimagnet with high spin polarization and showed evidence of half-metallicity[6].

Here we report on the temperature, composition and thickness dependent transport properties of MRG, which are at or near compensation point ($0.6 < x < 1.1$). Addition of Ru to the cubic Mn_2Ga structure provides both states (12) and electrons (8). Based on the on the empirical Slater-Pauling rules, should result in perfect compensation for $Mn_2Ru_{0.5}Ga$. 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 tetragonal distortion (c/a) can also affect the band structure, hence we also look at strain as a possible control parameter in engineering the MRG 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. 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/ MRG(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.

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). The out-of-plane lattice parameter, c, is between 0.598 nm and 0.618 nm, depending on the Ru concentration and film thickness. We find

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	0.07	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

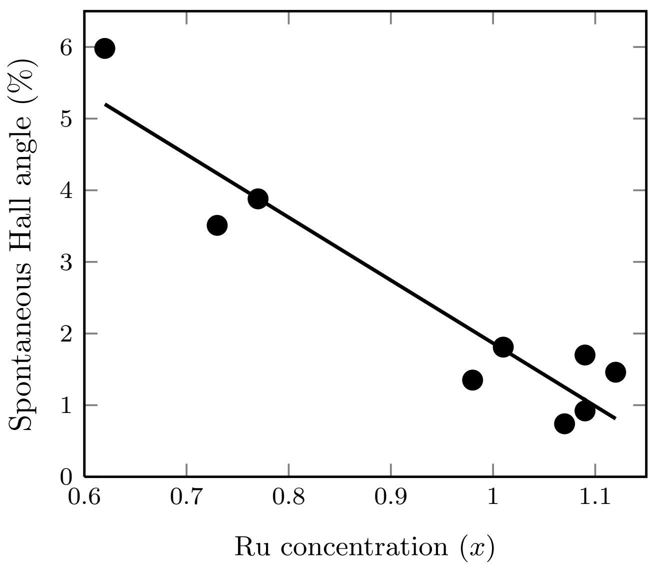

FIG. 1. Evolution of the spontaneous Hall angle (SHA) as a function of Ru composition, x, extracted from SHE measurements. The solid line is a linear fit of the data set.

that c increases exponentially with reducing film thickness. 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 ($\sqrt{2}a_0$ (MgO) = 0.5956 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%).

SQUID magnetometry shows clear out-of-plane anisotropy in all the samples we have studied. A small soft in-plane component is also present. As the Ru concentration is reduced from $x = 1.09$, the magnetization reduces, until it falls practically to zero (12 kA/m or 0.07 μ_B f.u.$^{-1}$) at $x = 0.68$. 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, coincident with the reversal in sign in the room temperature spontaneous Hall effect (SHE) measurements. 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. ??(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 SHA 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%)[7] and comparable to SHA recorded for amorphous rare earth transition metal alloys[8]. A high SHA is indicative of much lower carrier concentrations and a high spin polarization.

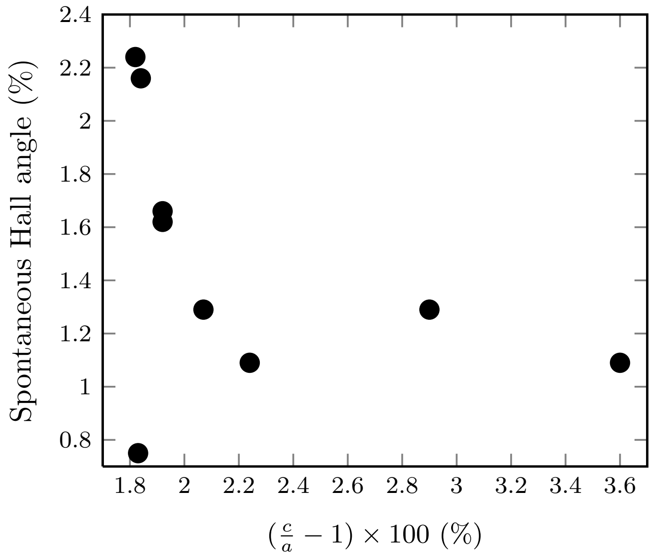

FIG. 2. Evolution of the spontaneous Hall angle (SHA) as a function of c/a ratio extracted from SHE measurements for samples with their SHA translated to a virtual $x = 1.0$ Ru concentration.

As shown in Fig. ??, 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 MRG 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. ??(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 $\sim$ 9 T at 350 K and the sign of the SHE loop reverses at 300 K. This indicates that the compensation temperature lies between 300 K and 350 K. By plotting the derivative of the Hall resistance w.r.t temperature, $\delta R_{XY}/\delta T$, as shown in Fig. 3(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

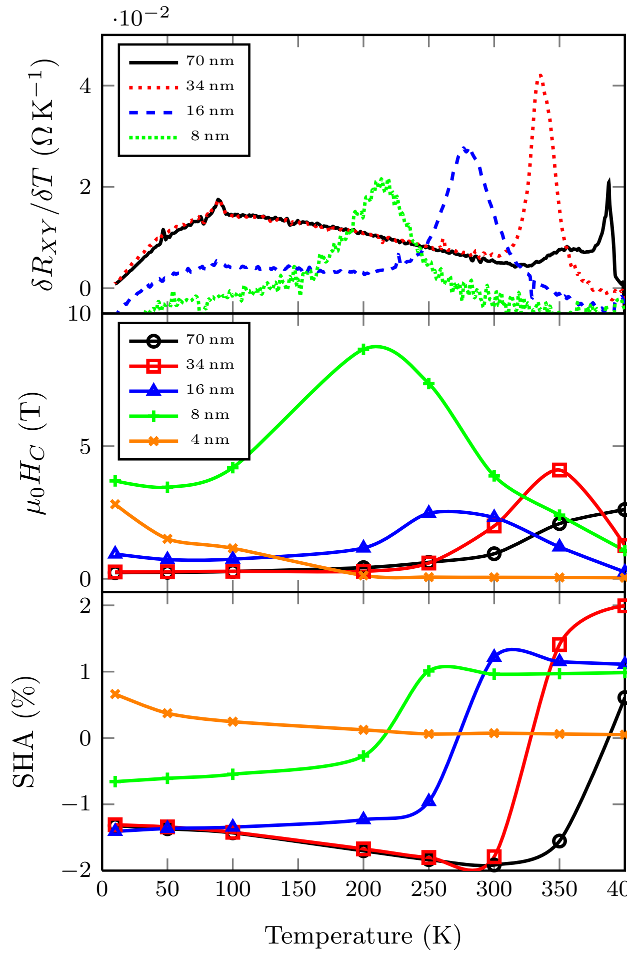

FIG. 3. (a) Variation of compensation temperature with the thickness of MRG film of same Ru concentration, given by the derivative of the resistance w.r.t temperature. The compensation temperature shifts to lower temperatures with decreasing thickness. (b) Extracted coercive field and (c) spontaneous 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.

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. 3(b) and (c) respectively.

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[9] 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 reversal of the majority spin channel. Concurrently 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.

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