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Analysis of Spin Polarization in Half-Metallic Heusler Alloys

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Abstract
Half-metals have recently gained great interest in the field of spintronics because their 100% spin polarization may make them an ideal current source for spintronic devices. This project examines four Heusler Alloys of the form $Co_2Fe_xMn_{1-x}Si$ that are expected to be half-metallic. Two magnetic properties of these alloys were examined, the Anisotropic Magnetoresistance (AMR) and the Anomalous Hall Effect (AHE). These properties have the potential to be used as simple and fast ways to identify materials as half-metallic or non-half-metallic. The results of these measurements were also used to examine the spin polarization and other properties of these materials.

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I. INTRODUCTION

Spintronics is an emerging area of study in the field of condensed matter physics that attempts to exploit electron spin and magnetic moment in electronic devices. Essentially, where traditional electronic devices use only charge degree of freedom, spintronic devices use spin properties in addition to charge. Spintronic devices have a wide variety of applications including memory, detection, and computing. Spintronic devices are driven using spin injection. Spin polarized electrons are first injected into a semiconductor through a ferromagnetic contact. This spin signal then travels through the semiconductor and can be detected at a second ferromagnetic contact. In the experiments described in this paper, specific properties of ferromagnets were examined. For this reason, current was expected to short straight through the ferromagnet rather than process through the semiconductor. Previous experiments using half-metallic ferromagnets have shown large spin signal. This means that they could potentially be used as ideal current sources in spintronic devices. More information about the properties of half-metals as well as easy methods for determining the half-metallicity of ferromagnets are desired as half-metals begin to be introduced into spintronic devices. In this work, we examined four Heusler alloys of the form $Co_2Fe_xMn_{1-x}Si$ where x had the values 0, 0.33, 0.66, and 1. As the amount of iron increases they are expected to change from half-metal to non-half metal, with a critical point at $x = 0.6 - 0.8$ [1]. We examined two magnetic properties of ferromagnetic materials, Anisotropic Magnetoresistance and Anomalous Hall Effect, which are expected to have a sign dependence on the half-metallicity of a material. We hope to examine these properties for potential use in determining whether a compound is half-metallic as well as examining spin polarization of half-metallic compounds.

A. Half Metals

Half metals are materials that are predicted to have 100% spin polarization at the Fermi level, meaning that, at the Fermi Level, the materials have either all spin-up or all spin-down electrons, illustrated in Fig. 1. This is unlike ordinary metals like iron that have a combination of both spin types and this property makes half metals ideal for achieving high efficiency in spin injection. It means that half-metals can achieve perfect or near-perfect spin-polarized current which may enhance spin-dependent properties [2]. In fact, many experiments have measured large values of magnetic effects such as tunneling magnetoresistance (TMR) and giant magnetoresistance (GMR) [3].

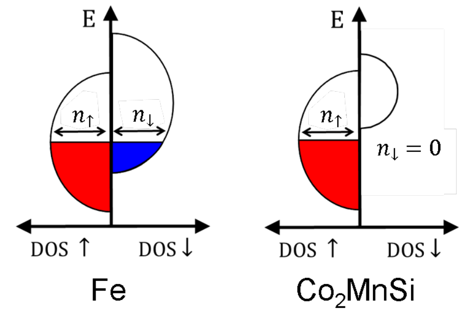

FIG. 1: Simplified density of states for ordinary ferromagnet and half-metallic ferromagnet. The location of the Fermi level is represented by the point where the images change from colored to uncolored. The amount of spin-up electrons at the Fermi level is proportional to the width of the red semicircle at this point. Similarly for spin-down and the blue semicircle. In iron, an ordinary metallic ferromagnet, both spin-up and spin-down electrons are present at the Fermi level. In Co_2MnSi, a half-metal, only spin-up electrons are present. $n_{\uparrow/\downarrow}$ is the number of spin-up/spin-down electrons. DOS stands for density of states.

B. Anisotropic Magnetoresistance

Magnetoresistance is the change in resistance of a material due to an applied magnetic field. The anisotropic magnetoresistance (AMR) is a contribution to the magnetoresistance that occurs in ferromagnets and is dependent on the orientation of the material. In ferromagnets, the resistivity of the material changes with respect to the angle between the magnetization of the material and an applied current. The AMR effect is caused by $s - d$ scattering, a result of the spin-orbit interaction. More specifically, when s state electrons scatter to the d state they become immobile, causing a resistance [2]. The sign of the AMR effect is dependent upon the nature of this scattering. When the dominant scattering is between $s_\uparrow$ and $d_\downarrow$ or $s_\downarrow$ and $d_\uparrow$ the sign will be opposite to that when the scattering is from $s_\uparrow$ to $d_\uparrow$ or $s_\downarrow$ to $d_\downarrow$. This is because $s_\uparrow$ to $d_\downarrow$ or $s_\downarrow$ to $d_\uparrow$ scattering has a different angle dependence that $s_\uparrow$ to $d_\uparrow$ or $s_\downarrow$ to $d_\downarrow$. As a result of this scattering effect, the sign of the AMR effect has been shown to switch between metallic and half-metallic ferromagnets because the high spin polarization of half metals determines that the scattering will always be between states of the same spin [1]. The AMR effect is quantified by the AMR ratio, defined as

$$\frac{\Delta\rho}{\rho} = \frac{\rho_{\parallel} - \rho_{\perp}}{\rho_{\perp}}$$

where $\rho_{\parallel}$ is the resistivity when current flows parallel to the magnetization and $\rho_{\perp}$ is the resistivity when current flows perpendicular to the magnetization [2]. Previous experiments done on Heusler alloys similar to those we examined showed the expected change in sign of the AMR ratio with half-metallicity. The experiments of Yang et al.on $Co_2Fe_xMn_{1-x}Si$ films showed a clear change in sign of the AMR ratio when x was increased from 0.6 to 0.8 [1]. This effect can be seen in the opposite curves in Fig. 2.

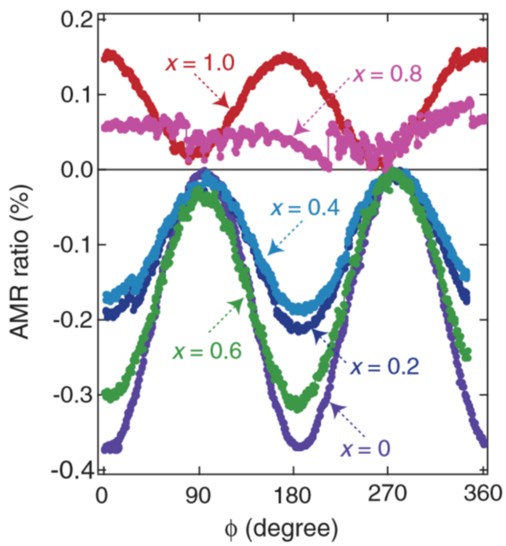

FIG. 2: Plot of AMR ratios for different Heusler compounds of the form $Co_2Fe_xMn_{1-x}Si$ for different values of x. ϕ is the relative angle between current and magnetization. Curves show opposite sign of AMR ratio for $x < 0.8$ [1].

C. Anomalous Hall Effect

The Hall effect is a phenomenon in electronics in which an electric field appears across a sample after application of a magnetic field perpendicular to the current through the sample. This corresponds to a voltage difference across the sample. The anomalous Hall effect (AHE), which appears in ferromagnets, is an additive term in the Hall resistivity that depends on the magnetization of the material. The total Hall resistivity is expressed as:

$$\rho_{\mathrm{H}} = \rho_{\mathrm{OHE}} + \rho_{\mathrm{AHE}} = R_{\mathrm{OHE}}B + \mu_0 R_{\mathrm{AHE}}M$$

where OHE and AHE refer to the ordinary Hall effect and anomalous Hall effect, respectively. ρ refers to resistivity, R refers to Hall coefficients, B is the magnetic induction, μ_0 is the vacuum permeability and M magnetization [4]. The mechanism for the anomalous Hall effect is still not fully understood and there are multiple possible theories for the origin of the effect. It is thought that the AHE may, like the AMR effect, be related to spin scattering, and would thus reveal information about Fermi level spin states. Previous work studying $Co_2Fe_xMn_{1-x}Si$ showed a change in sign of the AHE with increasing x [5].

II. METHODOLOGY

A. Device Fabrication

Each ferromagnetic film was grown on a gallium arsenide (GaAs) substrate with a small capping layer. The ferromagnets were Heusler alloys of the form $Co_2Fe_xMn_{1-x}Si$ where $x = 0, 0.33, 0.66, 1$. Devices were then fabricated using tools in the Minnesota Nanocenter Cleanroom. Three major steps were used in device creation. First, the pattern of the ferromagnet was defined using a combination of wet etching and ion milling that cut through parts of the capping layer. Next, silicon nitrite (SiN) was deposited to insulate the device, covering most of the surface apart from a few areas where contacts were placed. Finally, a layer of titanium and gold was deposited using e-beam evaporation to create large contacts for wire bonding. Our devices used a simple hall bar geometry with one large ferromagnetic arm and various gold contacts through which we could apply current and measure voltage. Completed devices were on the order of 600 microns square in size (Fig. 3).

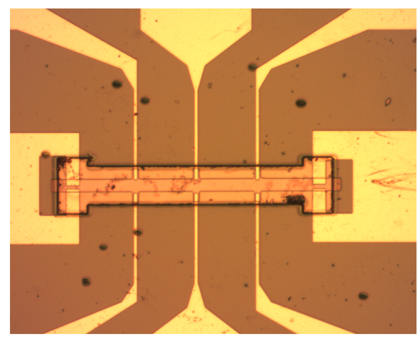

FIG. 3: Microscope image of completed devices showing ferromagnetic arm, gold contacts, and SiN layer. The length of the ferromagnetic arm is about 600 μm and the width is about 80 μm.

B. Testing

Devices were tested in a Quantum Design Physical Property Measurement System (PPMS) which permitted sensitive vacuum and temperature control as well as the ability to rotate both the sample and the magnetic field. To measure the AMR effect, we used a 4-terminal measurement geometry. The orientation of our measurements can be seen in Fig. 4. At high magnetic field, the magnetization of a ferromagnet will become aligned with the direction of the applied magnetic field. We applied a strong magnetic field to the sample and rotated it, thus rotating the magnetization of the sample. We then applied a current through the ferromagnetic arm and measured voltage in the same direction as the applied current. Finally, we measured the change in resistance due to the

AMR effect, which is dependent on the angle between applied current and magnetization. All AMR measurements were done at room temperature. To measure the AHE we applied a current through the ferromagnet and measured voltage perpendicular to the applied current. We applied a magnetic field perpendicular to the surface of the device, in the out-of-plane direction, sweeping from high negative field to high positive field, which gradually forced the magnetization to point in the direction of the field. We then used voltage measurements to examine the change in hall resistivity as a function of the applied magnetic field. We performed this test on each Heusler sample at three different temperatures, 60K, 100K, and 300K. We also gradually varied the temperature of a pure Fe sample between 30K and 300K and performed the test multiple times. We used these results to examine the temperature dependence of the AHE.

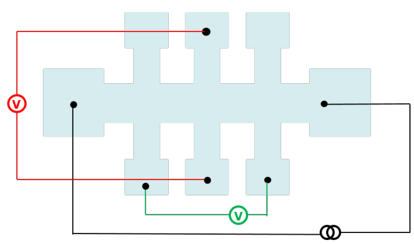

FIG. 4: Typical orientation for measurements. Current applied through the length of the ferromagnet in both measurements. Voltage measurement for AMR shown in green, AHE in red.

III. RESULTS AND DISCUSSION

A. Heusler AMR and AHE

We examined four Heusler samples as well as a pure iron sample in our measurements. In angle sweep measurements we found an AMR sign change between the Fe sample and the Heusler compounds, which is consistent with theoretical predictions for half-metallic Heuslers. However, we did not find a change in sign between Co_2FeSi and the rest of our samples (Figs. 5, 6, and 7). Co_2FeSi is not believed to be half-metallic and a sign change was expected based on previous experiments. We also measured much lower AMR ratios for the Heusler samples than for the Fe sample. These values are shown in Table 1.

We were also unable to observe the expected sign change in AHE of the Heusler samples (Figs. 8 and 9). Our temperature measurements did not show a significant temperature dependence on the magnitude of the AHE in Heusler samples. We did observe a strong temperature dependence in AHE measurements of the Fe sample (Fig. 10). AHE measurements were taken at different temperatures between about 20 K to over 300 K. At high temperatures (close to room temperature) the sign of the AHE was opposite that of the Heusler samples, but at low temperatures the sign switched. This temperature dependence is likely caused by magnetoresistive effects in the semiconductor, which have less of an effect on the resistance measurement in the ferromagnet at lower temperatures. This indicates that the true sign we measured was the same as the sign of the Heusler alloys. The temperature dependence also suggests the possibility that much of the applied current was flowing through the semiconductor, rather than shorting straight through the ferromagnet as we intended. This means that unintended effects from the semiconductor may have had an effect on our results. These results suggest that the use of a doped semiconductor may have had an effect on the measurments. Later experiments on semi-insulating GaAs examine whether the use of an undoped semiconductor improves the results. It is also a possibility that previous predictions of the density of states of some of the Heusler Alloys are incorrect. This would change the results that we expect from both of these tests.

	AMR Ratio
Fe	0.1%
Co_2FeSi	-0.004%
Co_2MnSi	-0.002%

TABLE 1: Measured AMR ratios for iron and two Heusler compounds. Co_2MnSi is expected to be half-metallic while Co_2FeSi is not.

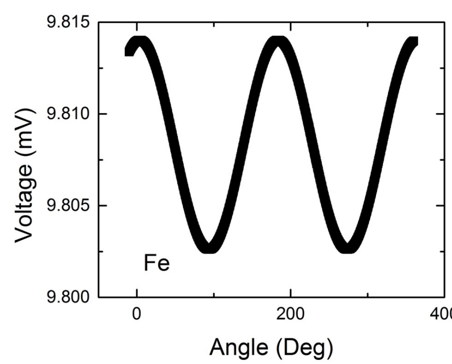

FIG. 5: Angle sweep measurements of AMR effect in Fe. x-axis shows the angle between the direction of the applied magnetic field (and hence the magnetization of the sample) and the applied current, in degrees. y-axis shows the voltage measurements, proportional to the resistivity of the sample, in millivolts. Plot shows the change in resistivity as a function of the described angle, showing the AMR effect in the sample.

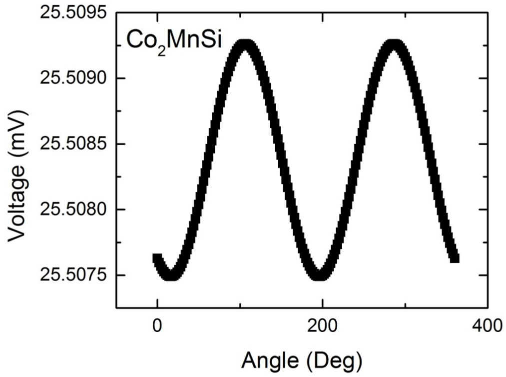

FIG. 6: Angle sweep measurements of AMR effect in Co_2MnSi. x-axis shows the angle between the direction of the applied magnetic field (and hence the magnetization of the sample) and the applied current, in degrees. y-axis shows the voltage measurements, proportional to the resistivity of the sample, in millivolts. Plot shows the change in resistivity as a function of the described angle, showing the AMR effect in the sample. The sign of the AMR effect is opposite that of Fe, shown by the fact that peaks in this plot correspond to troughs in Fig. 5.

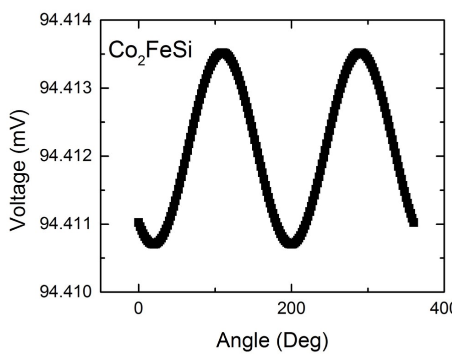

FIG. 7: Angle sweep measurements of AMR effect in Co_2FeSi. x-axis shows the angle between the direction of the applied magnetic field (and hence the magnetization of the sample) and the applied current, in degrees. y-axis shows the voltage measurements, proportional to the resistivity of the sample, in millivolts. Plot shows the change in resistivity as a function of the described angle, showing the AMR effect in the sample. The sign of the AMR effect is the same as that of Co_2MnSi, which is contrary to expectations.

B. Further Tests

Further tests were performed using new samples grown on undoped semiconductors in order to examine if an in-

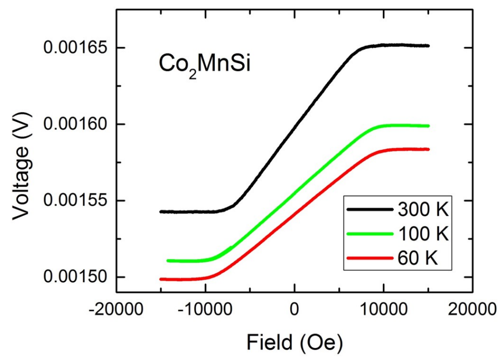

FIG. 8: Measurements of AHE in Co_2MnSi. x-axis shows the applied magnetic field in Oersted. y-axis shows the voltage measurements, proportional to the resistivity of the sample, in volts. Measurements were taken at three different temperatures, 300K 100K and 60K. The sign of the AHE effect can be seen in the sign of the slope of the plots.

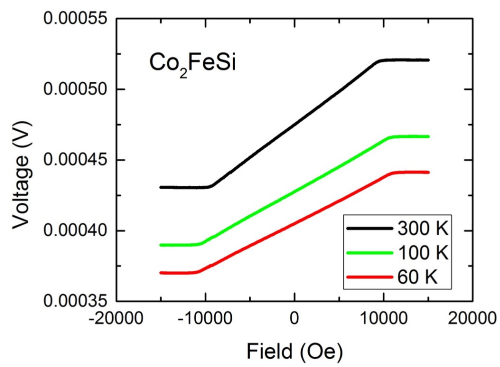

FIG. 9: Measurements of AHE in Co_2FeSi. x-axis shows the applied magnetic field in Oersted. y-axis shows the voltage measurements, proportional to the resistivity of the sample, in volts. Measurements were taken at three different temperatures, 300K 100K and 60K. The sign of the AHE effect can be seen in the sign of the slope of the plots, and when compared to Fig. 8 shows the same sign as that of Co_2MnSi. This is opposite of expectations.

sulating substrate will have an effect on these measurements. Recent measurements were taken by members of the Crowell group on Heusler samples grown on a semi-insulating GaAs substrate. As with our first measurements, Co_2MnSi and Co_2FeSi showed the same sign of the AMR ratio, however the values of the AMR ratio were much higher than in previous tests. The values are shown in Table 2.

Further examination indicates that, contrary to our previous expectation, both compounds may be

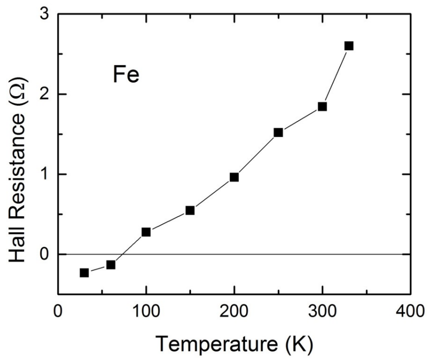

FIG. 10: Temperature dependence of AHE in Fe. x-axis shows temperature in K. y-axis shows Hall Resistance in ohms. A sign change of the Hall Resistance occurs at low temperature, indicating that the sign of the AHE effect in Fe is the same as in the Heusler alloys. This is contrary to expectations.

	AMR Ratio
Co_2FeSi	0.088%
Co_2MnSi	0.081%

TABLE 2: Later measurements of AMR ratios for Co_2MnSi and Co_2FeSi samples grown on insulating substrate.

half-metallic. The difference between them may be that Co_2MnSi is majority spin (spin up) dominated while Co_2FeSi is minority spin (spin down) dominated at the Fermi level (Fig. 11). This could explain why we did not observe a sign change, as both materials would have s-d scattering from $s_\uparrow$ to $d_\uparrow$ or $s_\downarrow$ to $d_\downarrow$.

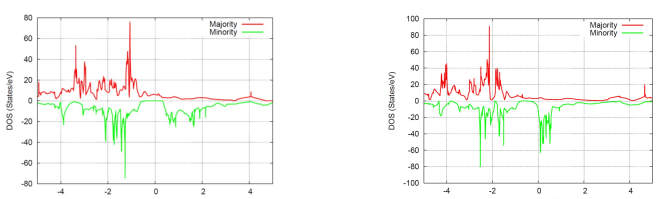

FIG. 11: Density of states for Co_2MnSi (left) and Co_2FeSi (right).

IV. SUMMARY AND CONCLUSIONS

We measured a change in sign of the AMR effect between iron and the heusler alloys, which agrees with our predictions based on the spin polarization of half metals. However, one sample, Co_2FeSi did not agree with previous experiments. The observation of a clear anomalous hall signal was promising, but the sign did not change as we predicted. Further tests performed by the Crowell group using new samples grown on undoped semiconductors showed the same signs of the AMR effect as measured previously, but with much higher AMR ratios for the Heusler alloys. This suggests that the semiconductor may have some effect on the measurements. It also indicates that our understanding of the spin polarization of the Heusler alloys may be incomplete, suggesting the possibility that Co_2MnSi and Co_2FeSi are both half-metallic. Further studies of these and other half-metals are necessary to provide conclusive results.

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