Thickness dependence of Hall transport in $\text{Ni}_{1.15}\text{Mn}_{0.85}\text{Sb}$ thin films on silicon

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Highly spin polarized Heusler alloys, NiMnSb and Co_2MnSi , attract a great deal of interest as potential spin injectors for spintronic applications. Spintronic devices require control of interfacial properties at the ferromagnet:semiconductor contact. To address this issue we report a systematic study of the ordinary and anomalous Hall effect, in $\text{Ni}_{1.15}\text{Mn}_{0.85}\text{Sb}$ films on silicon, as a function of film thickness. In contrast to the bulk stoichiometric material, the Hall carriers in these films become increasingly electron-like as the film thickness decreases, and as the temperature increases from 50 K toward room temperature. High field Hall measurements confirm that this is representative of the majority transport carriers. This suggests that current injected from a NiMnSb :semiconductor interface may not necessarily carry the bulk spin polarization. The films also show a low temperature upturn in the resistivity, which is linked to a discontinuity in the anomalous Hall coefficient. Overall these trends indicate that the application of Heusler alloys as spin injectors will require strictly controlled interfacial engineering, which is likely to be demanding in these ternary alloys.

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It has long been known that there is a component of the Hall resistivity of ferromagnets proportional to the magnetization, $\rho_{xy} = R_O B + R_S \mu_0 M$, where R_O and R_S are known, respectively, as the ordinary and anomalous Hall coefficients and $\mu_0 M$ is the magnetization. The recent development^{1,2} of theories, based on the Berry³ (or Pancharatnam) phase, that quantitatively describe the behavior of R_S in a number of different systems has resulted in a resurgence of interest in the anomalous Hall effect (AHE). The observation of a non-zero anomalous Hall velocity requires a finite spin polarization of the transport current and spin-orbit coupling. This holds out the intriguing possibility that the transport spin polarization can be extracted from anomalous Hall measurements in well characterized systems.

The half Heusler⁴ alloy NiMnSb is ferromagnetic with a Curie temperature (T_C) of 728 K,⁵ and band structure calculations predict that it is half-metallic⁶ with the spin-polarized carriers holes derived from the Sb $6s^2$ band. Further calculations have shown the transport spin polarization (P_t) of NiMnSb to be highly sensitive to atomic disorder⁷ and surface effects.^{8,9} Ideally spin injection for spintronic applications will require the carriers close to the injection interface to carry the bulk spin polarization.

A primary motivation for performing this study was to determine whether bulk-like transport could be achieved in thin films and hence evaluate NiMnSb as a potential spin injector. Here, we report a systematic study of the longitudinal (ρ_{xx}) and Hall (ρ_{xy}) electrical resistivities of $\text{Ni}_{1.15}\text{Mn}_{0.85}\text{Sb}$ films on silicon as a function of film thickness. We demonstrate that the film transport properties close to the interface vary quite drastically compared to bulk—like behavior and this has important implications for using this material in spintronic applications. Thin films of $\text{Ni}_{1.15}\text{Mn}_{0.85}\text{Sb}$, with thicknesses of 5, 45, 80, 110 and 400 nm, were grown on Si(100) by pulsed laser deposition¹⁰ at 200 °C from a stoichiometric target. All films were shown by energy dispersive x-ray analysis to be slightly off-stoichiometric, formulated $\text{Ni}_{1+x}\text{Mn}_{1-y}\text{Sb}$; $x = 0.15 \pm 0.05$, $y = 0.15 \pm 0.05$. The x-ray diffraction patterns were consistent with (220) oriented polycrystalline NiMnSb Heusler phase with lattice parameter $5.99 \text{ \AA} \pm 0.02 \text{ \AA}$, compared to $5.9320 \text{ \AA} \pm 0.0028 \text{ \AA}$ for the target. No second phase was observed. The rocking curve of the (220) reflection indicated that the out-of-plane alignment was imprecise, with a spread of orientations of 12 around [110].

Magnetotransport data were collected in a square geometry by the van der Pauw method. The geometry led to a

FIG. 1. Hall resistivity (open squares) vs field for 80 nm film at 50, 60, 70, 80, 90, 100, 110, 130, 150, 200, 250, and 290 K. Solid lines show fit to $\rho_{xy} = R_O B + R_S M$ at each temperature. Inset: Hall resistivity vs field for 5 nm film at selected temperatures.

strong mixing of the Hall and MR components, which were separated by their opposite symmetries with respect to inversion of the magnetic field. The temperature and field dependence of the magnetoresistance were reported previously.¹¹ The field dependence of the magnetization of the films was measured at the same temperatures and in the same geometry (field perpendicular to the film surface) as the Hall measurements, in an Oxford Instruments vibrating sample magnetometer. In this geometry the magnetic anisotropy of the films is dominated by the shape anisotropy. A reliable magnetization could not be obtained for the 5 nm film.

The Hall resistivity was measured for all the films at selected temperatures between 50 and 290 K, the data for the 80 nm film, which is typical of all the films, is shown in Fig. 1. An iterative procedure was used to fit the measured Hall resistivity to the expression for $\rho_{xy} = R_O B + R_S M$, using independently measured magnetization, which was measured at the same temperature, and in the same geometry (with field perpendicular to film surface). We previously used this method to report¹² the Hall transport of the thickest film. With this field orientation the demagnetization factor (N) is unity, hence the flux density, $B = \mu_0[H + 4\pi(1-N)M] = \mu_0 H$, where H is the applied magnetic field in A/m. The fitting procedure was limited to the range of data $1.5 \text{ T} \geq \mu_0 H \geq 0 \text{ T}$, because a slight curvature was observed in ρ_{xy} at larger fields, indicating that the low-field limit model (discussed in the following) becomes inappropriate above 1.5 T. The solid lines in Fig. 1 are fits to the data for the 80 nm film at a selection of temperatures. The temperature dependence of the low field limit ordinary Hall coefficient R_O obtained from this fitting procedure, for all the films, is shown in Fig. 2. The temperature dependence of ρ_{xx} is also shown in Fig. 2, for comparison. Note an increasingly strong low temperature upturn in ρ_{xx} is observed with decreasing thickness.

In the stoichiometric bulk material R_O remains positive at all temperatures below T_C .^{13,14} It is immediately apparent from Fig. 2(b) that the transport in all these films is different to that material, as R_O is increasingly negative as the temperature increases from 50 K, and as the thickness decreases.

FIG. 2. (a) Longitudinal resistivity vs temperature for the series of films. (b) Ordinary Hall coefficient R_O vs temperature, solid lines are a guide to the eye.

The crossover from positive to negative R_O corresponds to a crossover from hole dominated to electron dominated Hall transport, and hence that the Hall data must be considered within a two-carrier model. Band structure calculations⁶ predict that the spin polarized carriers are Sb holes, so the observation of electron dominated transport at room temperature in thin films suggests that NiMnSb may not be an efficient spin injector.

In a two-carrier system, R_O is only constant in the low field limit (when $\mu_{e,h}^2 B^2 \ll 1$), in this limit R_O is given by Eq. (1), where n and p are the electron and hole carrier concentrations and μ_e and μ_h are the respective mobilities. In the high field limit ($\mu_{e,h}^2 B^2 \gg 1$) the dependence on the mobility ratio z disappears and $R_O = 1/(p-n)e$; hence it becomes a direct measure of the majority carriers. Therefore, if the low-field Hall resistivity has been dominated by a high mobility minority carrier, then there must be a strong curvature of ρ_{xy} at intermediate fields with an eventual change of sign,

$$R_O = \frac{p - nz^2}{|e|(p + nz)^2}, \quad z = \frac{\mu_e}{\mu_h}. \quad (1)$$

Hence, the low-field Hall mobility is not necessarily representative of the majority transport carriers. For example,¹⁵ in CrO_2 there are a small number of high mobility holes and around 500 times more low mobility electrons, the low-field Hall is hole-like, and the high-field Hall is electron-like, in agreement with the thermopower. To investigate whether the low-field Hall is representative of the majority transport carriers, the high field Hall resistivity of the 5 nm sample was measured, this is plotted in the inset to Fig. 1. There is a slight curvature toward less negative slope with increasing field, but unlike CrO_2 (Ref. 15) neither a sign change nor the high field limit is reached by 8 T. This strongly suggests that the low-field Hall is representative of the majority transport carriers. The curvature can be fit to the two band model^{16,15} but the refined parameters are strongly correlated and unique fit could not be obtained. This is consistent with the observation¹⁵ that a reliable fit can only be obtained by relating the band parameters to the measured low-field limit,

high field limit and crossover point values. There is no feature in the temperature dependence of R_0 associated with the resistivity upturn; this suggests that the resistivity upturn is not due to a freezing out of carriers, but to a decrease in carrier mobility.

Detailed knowledge of the transport carriers as a function of thickness is important for understanding spin injection processes at ferromagnet:semiconductor interfaces. Four relations are required to determine the four band parameters, the Hall and the zero field resistivity provide two. Two-carrier transport analysis is routine in high mobility semiconductors, where the other two relations are obtained from the Shubnikov–de Haas oscillations and the MR. In these films that information is not accessible because the two-carrier MR is masked by the anomalously large positive MR¹⁷ and in metals Shubnikov–de Haas oscillations are only observed in extremely high fields. Therefore, only a qualitative analysis of the band parameters can be made. The sign reversal of the low field R_0 with increasing temperature, even in the thickest film, shows that at low temperature $p > nz^2$ and at high temperature $p < nz^2$. z is unlikely to change dramatically with temperature and n/p is almost certainly increasing with temperature. The small amount of curvature in the high-field Hall indicates that, unlike CrO_2 ,¹⁵ z is close to unity. The band structure of stoichiometric NiMnSb contains both holes and electrons,⁶ with holes dominating the Hall resistivity,^{13,14} although the thermopower¹⁴ indicates a crossover to electron-dominant transport. For the films studied here, a likely hypothesis is that the holes result from the bulk band structure and their concentration is only weakly temperature dependent, whereas the electron concentration seems to be derived partly from the band structure and partly from a thermally activated process, such as thermal excitation of donor states. The off-stoichiometry in these $\text{Ni}_{1.15}\text{Mn}_{0.85}\text{Sb}$ films will result in a large number of atomic site defects, which are predicted⁷ to affect the band structure, and the difference between the stoichiometric bulk and the 400-nm-thick film is likely to be a result of the stoichiometry. The increasingly electron dominated transport as a function of thickness is not attributed to off-stoichiometry in our films as this did not change systematically with thickness. The trend can only be explained by the increasing significance of electronic surface or interface states, arising from either the reduced symmetry at the interfaces or strain induced defects. Note that unlike the silver chalcogenides,¹⁸ there is no evidence of a crossover of majority carrier at the MR maximum (resistivity upturn).

Now let us turn to R_S . The anomalous Hall effect has historically been ascribed¹⁹ to a scattering anisotropy, although there can also be an intrinsic²⁰ (scattering independent) term, which is discussed in the following. In the scattering model, it was proposed¹³ that R_S was derived from contributions from side-jump scattering and skew scattering, and that these terms were proportional to ρ_{xx}^2 and ρ_{xx} , respectively. In bulk NiMnSb , this model accounts for the experimental data at high temperatures, but there is a discontinuity in R_S/ρ_{xx} vs ρ_{xx} at around 100 K.^{13,14} The inset to Fig. 3(b) shows a typical R_S/ρ_{xx} vs ρ_{xx} plot, from the 45 nm film ρ_{xx} which is non-monotonic. At temperatures above and be-

FIG. 3. (a) a and (b) b coefficients obtained from the fits to $R_S/\rho_{xx} = a + b\rho_{xx}$ for all films. Because the magnetization of the 5nm film could not be measured directly, the magnetization loop of the 45 nm film was scaled by volume to obtain R_S of the 5 nm film. Bulk values taken from Otto *et al.* (Ref. 13). Inset to (b) R_S/ρ_{xx} vs ρ_{xx} for the 45 nm film, dashed line is a guide to the eye. Solid lines show fits to $R_S/\rho_{xx} = a + b\rho_{xx}$ in regions above and below upturn.

low the resistivity upturn two different straight lines are obtained. The a and b coefficients obtained from linear fitting of R_S/ρ_{xx} vs ρ_{xx} above and below the upturn for all the films are shown in Figs. 3(a) and 3(b), respectively. Above the resistivity upturn, the coefficients are, within error, the same as the stoichiometric bulk values¹³ of $a = -6.5 \times 10^{-4} \text{T}^{-1}$ and $b = 21\,500 \text{T}^{-1} \Omega^{-1} \text{m}^{-1}$, which were previously interpreted as side-jumps dominating over skew scattering. However, below the resistivity upturn, the magnitudes of both the slope and the intercept increase dramatically as the thickness is increased from 5 to 110 nm, driven by the temperature dependence of ρ_{xx} . The implicit assumption in the traditional R_S/ρ_{xx} vs ρ_{xx} analysis¹³ is that R_S is only indirectly a function of temperature via its dependence on the resistivity (scattering). It appears that both in the stoichiometric bulk material, and these films, that assumption and the validity of that model breaks down around 100 K. No change was observed by room temperature in the sign of R_S in any of our films. In a number of simple ferromagnets (such as Fe, Co, Gd) the skew-scattering and side jump terms are of opposite sign, but it is not known if that is the case in our material.

It is important to note that the anomalous Hall effect arises from an asymmetric deflection of the carriers, resulting in an anomalous Hall conductivity, (σ^A); the anomalous Hall resistivity (ρ^A) is derived from the conductivity by $\sigma^A = \rho^A/(\rho_{xx}^2 + \rho_{yy}^2) \approx \rho^A/\rho_{xx}^2$ because $\rho_{xx} \gg \rho_{yy}$. Since a quadratic behavior of R_S in ρ_{xx} only requires a temperature independent σ^A , it is not clear that any inferences can be made about the scattering.

Recently, theories describing the anomalous Hall effect as an intrinsic Berry³ phase effect, have given a good quantitative agreement with experiment^{1,2} that was never achieved with the scattering model. Two types of Berry

phase induced AHE have, thus far, been reported, one is related to spin chirality in magnetically frustrated²¹ systems and the other is associated with thermally induced topological defects that show an exponential temperature dependence¹ around T_C . In our films there is no feature in the magnetization at the resistivity upturn temperature, and this temperature is far from the Curie temperature, so the anomalous behavior of R_S below the resistivity upturn is dissimilar to previously reported Berry systems. Although a Berry phase component cannot be ruled out, the change in σ^A at the resistivity upturn is probably due to a change in spin dependent scattering.

In summary, the room temperature electrical transport in non-stoichiometric Heusler thin films becomes increasingly electron dominated with decreasing thickness, in marked contrast to the spin-polarized holes predicted for the bulk

stoichiometric material. The thickness dependence is likely to be due to the increasing significance of interface or free surface electronic states, and indicates that controlled interfacial engineering will be required for the use of NiMnSb as a spin injector. The anomalous Hall conductivity cannot be interpreted within the traditional scattering model at low temperatures, because of an additional contribution that comes into play, which is attributed to a change in the spin-dependent scattering. Unlike the silver chalcogenides, there appears to be no correlation between the large positive MR found in these films¹⁷ and the sign reversal in the ordinary Hall coefficient.

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