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

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Highly spin polarized Heusler alloys, NiMnSb and Co$_2$MnSi, 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 Ni$_{1.15}$Mn$_{0.85}$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 of theories, based on the Berry (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, which can be extracted from anomalous Hall measurements in well characterized systems.

The half Heusler alloy NiMnSb is ferromagnetic with a Curie temperature ($T_C$) of 728 K, and band structure calculations predict that it is half-metallic with the spin-polarized carriers holes derived from the Sb 6$s^2$ band. Further calculations have shown the transport spin polarization ($P_s$) of NiMnSb to be highly sensitive to atomic disorder and surface effects. 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 ($\rho_{xx}$) and Hall ($\rho_{xy}$) electrical resistivities of Ni$_{1.15}$Mn$_{0.85}$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 Ni$_{1.15}$Mn$_{0.85}$Sb, with thicknesses of 5, 45, 80, 110 and 400 nm, were grown on Si(100) by pulsed laser deposition at 200 °C from a stoichiometric target. All films were shown by energy dispersive x-ray analysis to be stoichiometric, formulated Ni$_{1+y}$Mn$_{1-y}$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 Å ± 0.02 Å, compared to 5.9320 Å ± 0.0028 Å 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
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.\textsuperscript{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 report\textsuperscript{12} 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 T $\equiv \mu_0 H \equiv 0$ T, because a slight curvature was observed in $\rho_{xy}$ 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 $\rho_{xx}$ is also shown in Fig. 2, for comparison. Note an increasingly strong low temperature upturn in $\rho_{xx}$ is observed with decreasing thickness.

In the stoichiometric bulk material $R_O$ remains positive at all temperatures below $T_C$.\textsuperscript{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.

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 calculations\textsuperscript{6} 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 $\mu_{e,h}B^2<1$), in this limit $R_O$ is given by Eq. (1), where $n$ and $p$ are the electron and hole carrier concentrations and $\mu_e$ and $\mu_h$ are the respective mobilities. In the high field limit ($\mu_{e,h}B^2>1$) 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 $\rho_{xy}$ at intermediate fields with an eventual change of sign.

$$R_O = \frac{p-n}{e(p+n)z}$$

$$z = \frac{\mu_e}{\mu_h}$$

Hence, the low-field Hall mobility is not necessarily representative of the majority transport carriers. For example,\textsuperscript{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\textsuperscript{6,15} but the refined parameters are strongly correlated and unique fit could not be obtained. This is consistent with the observation\textsuperscript{15} 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 $R_O$ 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 MR\textsuperscript{17} 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 $R_O$ with increasing even temperature, even in the thickest film, shows that at low temperature $p > n z^2$ and at high temperature $p < n z^2$. $z$ 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\textsubscript{2},\textsuperscript{15} $z$ is close to unity. The band structure of stoichiometric NiMnSb contains both holes and electrons,\textsuperscript{6} with holes dominating the Hall resistivity\textsuperscript{13,14} although the thermopower\textsuperscript{14} 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 Ni\textsubscript{1.15}Mn\textsubscript{0.85}Sb films will result in a large number of atomic site defects, which are predicted\textsuperscript{7} 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,\textsuperscript{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 ascribed\textsuperscript{19} to a scattering anisotropy, although there can also be an intrinsic\textsuperscript{20} (scattering independent) term, which is discussed in the following. In the scattering model, it was proposed\textsuperscript{13} that $R_S$ was derived from contributions from side-jump scattering and skew scattering, and that these terms were proportional to $\rho_{xx}^2$ and $\rho_{xx}$, respectively. In bulk NiMnSb, this model accounts for the experimental data at high temperatures, but there is a discontinuity in $R_S/\rho_{xx}$ at around 100 K.\textsuperscript{13,14} The inset to Fig. 3(b) shows a typical $R_S/\rho_{xx}$ vs $\rho_{xx}$ plot, from the 45 nm film $\rho_{xx}$ which is non-monotonic. At temperatures above and below the resistivity upturn two different straight lines are obtained. The $a$ and $b$ coefficients obtained from the fits to $R_S/\rho_{xx} = a + b \rho_{xx}$ for all films. Because the magnetization of the 5 nm 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) $R_S/\rho_{xx}$ vs $\rho_{xx}$ 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.
phase induced AHE have, thus far, been reported, one is related to spin chirality in magnetically frustrated systems and the other is associated with thermally induced topological defects that show an exponential temperature dependence 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 non-stoichiometric Heusler thin films becomes increasingly spin dependent scattering. Unlike the silver chalcogenides, there appears to be no correlation between the large positive MR found in these films and the sign reversal in the ordinary Hall coefficient.

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 stiochiometric 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 films and the sign reversal in the ordinary Hall coefficient.

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