Hall effect in laser ablated Co$_2$(Mn,Fe)Si thin films

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Abstract
Pulsed laser deposition was employed to grow thin films of the Heusler compounds Co$_2$MnSi and Co$_2$FeSi. Epitaxial growth was realized both directly on MgO(1 0 0) and on a Cr or Fe buffer layer. Structural analysis by x-ray and electron diffraction shows for both materials the ordered L$_2^1$ structure. Bulk magnetization was determined with a SQUID magnetometer. The values agree with the Slater–Pauling rule for half-metallic Heusler compounds. On the films grown directly on the substrate measurements of the Hall effect have been performed. The normal Hall effect is nearly temperature independent and points towards a compensated Fermi surface. The anomalous contribution is found to be dominated by skew scattering. A remarkable sign change in both normal and anomalous Hall coefficients is observed on changing the valence electron count from 29 (Mn) to 30 (Fe).

1. Introduction
Due to their expected half-metallicity the use of Heusler alloys has been considered for a number of spintronic applications [1–3]. Especially the fabrication of magnetic tunnelling junctions with Heusler electrodes has advanced rapidly over the last few years. Recent experiments with Co$_2$MnSi electrodes and an amorphous AlO$_x$ barrier suggest a spin polarization of nearly 90% for the relevant layers at the interface between electrode and barrier [3]. However, this value is reached only at temperatures below 4 K. At room temperature these junctions show a transport spin polarization comparable to elements with conventional ferromagnetic electrodes, despite a Curie temperature of 985 K. It has been proposed that this decrease in spin polarization is due to the unfortunate position of the Fermi energy near the edge of the half-metallic gap [3, 4]. The calculations in [4] further predict that by replacing Mn with Fe the additional electrons will cause a shift of $E_F$ across the half-metallic band gap, while the size of the band gap as well as the general structure of the bands near $E_F$ will stay nearly unchanged. For Co$_2$FeSi the Fermi level is then expected to be located close to the bottom of the conduction band of the minority electrons.

The complexity of tunnelling junction experiments is likely to introduce additional, barrier dependent contributions to the tunnelling currents which will obfuscate intrinsic material properties [5, 6]. In contrast, the measurement of the Hall effect in ferromagnets is a well-established and comparatively simple procedure, which has already been applied to various Heusler alloys [7–9]. Although it is not possible to directly map the band structure or measure spin polarization, the electron doping in the Co$_2$(Mn,Fe)Si system is expected to cause changes in the normal as well as in the anomalous regime.

Since the Hall voltage scales with the inverse thickness of the measured sample, it is very advantageous to possess high quality thin films. In this paper we report pulsed laser deposition (PLD) of Co$_2$MnSi and Co$_2$FeSi on MgO(1 0 0) substrates. In contrast to the more widely used sputter deposition no buffer gas is needed and UHV conditions can be sustained during the whole preparation process. The ability of this technique to grow thin Heusler films has been demonstrated by Wang et al [10]. We will discuss the effect of Cr and Fe buffer layers on film growth and magnetic properties. It is also possible to achieve epitaxial growth directly on the insulating substrate. Using a standard photolithographic procedure the latter set of films was patterned with a Hall bar structure. The results from these experiments will be discussed in the second part of this paper.
2. Film growth

Film growth was realized by PLD from stoichiometric targets. As light source a KrF excimer laser ($\lambda = 248$ nm) with pulse energies ranging from 300 to 700 mJ was used. The choice affects the growth rate, which increases from 0.2 to 2 Å min$^{-1}$. Deposition was carried out in a UHV chamber ($p_{\text{base}} \lesssim 2 \times 10^{-10}$ mbar). A RHEED system allowed in situ surface analysis. Since no buffer gas was used in the growth process, the formation of melt droplets is observed (see figure 1). For high pulse energies the volume of these droplets is comparable to the film volume. This prohibits the determination of bulk magnetic properties in these films. The situation is drastically improved with lower laser energies. At 300 mJ only a small number of droplets are visible. They contribute to less than 5% of the sample volume. The films were deposited on MgO(1 0 0) substrates. In order to overcome the relatively large lattice mismatch of 5.6%, two strategies were employed. One set of samples was deposited directly on the substrate. In this case high substrate temperatures are required to grow the compounds epitaxially. Alternatively the Heusler alloys were deposited onto an intermediate Fe or Cr buffer layer at room temperature and subsequently annealed at 400–500 °C. The buffer layers were also grown by PLD in the same chamber. In order to protect the films from oxidation they were covered with an Al capping layer before transferring them out of the vacuum system.

3. Structural and magnetic properties

The crystal structure of the Heusler films was analysed by x-ray and electron diffraction. RHEED images as presented in figure 2 reveal a smooth and ordered surface. For the films grown on a Cr or Fe buffer $\omega$–$2\theta$ scans of the specular as well as $\phi$ scans of the off-specular reflections show epitaxial, single-phase growth already after deposition. However, the $\{111\}$ reflections cannot be detected, indicating growth not in the fully ordered L$_2_1$ but in the B2 structure. After annealing above 300 °C the superstructure reflections can be detected. For an annealing time of 60 min the maximum relative intensity is found between 400 and 500 °C. Above 500 °C the scattered intensity of all reflections starts to diminish. Comparison with reports for other Heusler films suggests that this decrease is caused by interdiffusion with the buffer layer [11, 12].

If the film is deposited directly onto the substrate at room temperature, no long-range crystallographic ordering is observed. For these films, a post-annealing procedure does not induce crystalline growth either. Instead substrate temperatures of 400 °C and above are required during
deposition. Like the films deposited on a metallic buffer layer, these samples exhibit \((100)\)-oriented and \(L_2\) ordered growth. This is in accordance with sputtered \(\text{Co}_2\text{FeSi}\) films we reported earlier [13, 14], but in contrast to reports from Inomata et al who found long-range crystallographic ordering already at room temperature [15].

Bulk magnetic properties were analysed with a SQUID magnetometer. As shown in figure 3, the film magnetization is correlated with their crystalline quality: for as-deposited films on a metallic buffer layer, the saturation magnetization of both compounds is reduced by about 10–15% compared with the films annealed at 400–500°C. For these temperatures the magnetization assumes values consistent with the Slater–Pauling rule for Heusler alloys (see figure 4) [16]. A further increase in the annealing temperature causes a rapid drop in the magnetization. For the films fabricated without a buffer layer the saturation magnetization increases by 25% from the disordered films to the Slater–Pauling values at substrate temperatures of 600°C and above.

4. Hall effect

The availability of epitaxial thin films grown directly on insulating substrates allowed the measurement of the Hall resistivity. For this purpose films were patterned into a Hall bar geometry using a standard photolithographic procedure. A dc current was applied and the measured voltage was antisymmetrized with respect to the external field in order to eliminate ohmic contributions to the signal. To rule out effects from both sample oxidation and a metallic Al cap samples with and without capping layer were compared. These results showed no significant difference.

Figure 5 shows Hall resistivities for \(\text{Co}_2\text{MnSi}\) and \(\text{Co}_2\text{FeSi}\) measured at various temperatures. Film thickness in both
cases was 50 nm. The graphs show the typical behaviour of ferromagnetic materials. Below saturation the signal is governed by the anomalous Hall effect and the increase in the sample magnetization. For higher fields the normal Hall effect caused by the Lorentz force from the external field determines the value of the resistivity. This behaviour is conventionally summed up in the formula $\rho_{xy}(B, M) = R_0 B + R_S \mu_0 M$ with the normal and anomalous Hall coefficients $R_0$ and $R_S$.

The value of the normal Hall coefficient assumes a different sign for the two materials. In order to interpret these values one has to take into account the multi-sheeted structure of the Fermi surface of the Heusler alloys. In this case $R_0$ depends on the electron- or hole-like nature as well as the effective mass tensor for each individual band [17]. But despite numerous theoretical and experimental investigations of Co$_2$MnSi and Co$_2$FeSi, these properties are not known so far. In the simple Fermi sphere model the relation $R_0 = 1/nq$ yields 5 holes/f.u. for Co$_2$MnSi and 25 electrons/f.u. for Co$_2$FeSi. Despite the limited use of this model two aspects are noteworthy. First, these apparently high charge carrier concentrations indeed point towards partially compensated electron-like and hole-like sheets of the Fermi surface. Second, it appears in this picture that the charge carriers on Co$_2$FeSi have a more electron-like character compared with Co$_2$MnSi. However, due to the additional d electron Co$_2$FeSi should be closer to a completely filled d band. Therefore the opposite behaviour of the normal Hall coefficient is expected. This disagreement highlights the need for a detailed knowledge of the Fermi surface.

The anomalous Hall effect in both materials also shows opposite signs—in the low temperature regime as well as in the temperature dependence. In order to understand the nature of the anomalous Hall effect the anomalous Hall coefficient $R_s$ is plotted as a function of the longitudinal resistivity in figure 6. For both materials a linear dependence was found. This implies that the temperature dependence of the anomalous Hall effect is governed by skew scattering. The opposite sign of the slopes is possibly connected to the apparent charge deduced from the normal Hall effect.

5. Summary

It has been shown that PLD is a suitable method for the deposition of high quality Heusler alloy thin films: epitaxial growth of Co$_2$MnSi and Co$_2$FeSi on MgO has been realized. The detection of the L2$_1$ crystal structure as well as the adherence of the Slater–Pauling rule for Heusler alloys are consistent with half-metallicity. Since it was possible to omit the use of a buffer layer, electronic transport measurements could be performed. The dependence of the normal Hall coefficient on the electron number in the compounds is counter-intuitive and points towards a complicated topology of the Fermi surfaces. The evolution of the anomalous Hall coefficient is consistent with skew scattering as the source of the magnetization dependent contribution.

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