Direct measurement of the magnetic anisotropy field in Mn–Ga and Mn–Co–Ga Heusler films

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Abstract
The static and dynamic magnetic properties of tetragonally distorted Mn–Ga based alloys were investigated. Static properties are determined in magnetic fields up to 6.5 T using SQUID magnetometry. For the pure Mn₁.₆Ga film, the saturation magnetisation is 0.36 MA m⁻¹ and the coercivity is 0.29 T. Partial substitution of Mn by Co results in Mn₂.₆Co₀.₃Ga₁.₁. The saturation magnetisation of those films drops to 0.2 MA m⁻¹ and the coercivity is increased to 1 T.

The time-resolved magneto-optical Kerr effect (TR-MOKE) is used to probe the high-frequency dynamics of Mn–Ga. The ferromagnetic resonance frequency extrapolated to zero-field is found to be 125 GHz with a Gilbert damping, α, of 0.019. The anisotropy field is determined from both SQUID and TR-MOKE to be 4.5 T, corresponding to an effective anisotropy density of 0.81 MJ m⁻³.

Given the large anisotropy field of the Mn₂.₆Co₀.₃Ga₁.₁ film, pulsed magnetic fields up to 60 T are used to determine the field strength required to saturate the film in the plane. For this, the extraordinary Hall effect was employed as a probe of the local magnetisation. By integrating the reconstructed in-plane magnetisation curve, the effective anisotropy energy density for Mn₂.₆Co₀.₃Ga₁.₁ is determined to be 1.23 MJ m⁻³.

Keywords: Heusler films, magnetic anisotropy, time-resolved magneto-optical Kerr effect, Hall effect, coercivity

(Some figures may appear in colour only in the online journal)

1. Introduction
There has been a recent resurgence in research on Heusler alloys, in particular Mn-based ferrimagnetic compounds, for both magnetic storage and spin-transfer-torque applications due to the ability to widely tune the magnetic properties with varying composition [1–6]. The addition of Co to Mn–Ga allows the subtle tuning of magnetic properties, such as uniaxial anisotropy and saturation magnetisation [4]. These films can possess very high uniaxial anisotropy [7, 8]. They are promising for future rare-earth free permanent magnets [9]; highly stable magnetic recording elements scalable...
down to 10 nm bit size [3]; spin-polarised electrodes for tunnel magnetoresistance devices [10, 11]; as well as active elements in next generation spin-transfer torque devices such as THz-band spin-transfer-oscillators, due to their high ferromagnetic resonance frequencies and low Gilbert damping [7].

In these films, anisotropy fields of several tens of Teslas have been reported [3, 4, 6]. Combined with their low magnetisation, typically below 0.5 MA m−1, traditional SQUID magnetometry and conventional ferromagnetic resonance (FMR) techniques are not so well suited for magnetic characterisation. In the majority of recent reports the anisotropy field is extrapolated from the intersection of the low field linear slope of the hard-axis magnetisation curve to the easy-axis saturation magnetisation. This intersection occurs at a field which is, in general, beyond the machine limit. For SQUID magnetometry this requires careful subtraction of the diamagnetic background signal [12]. It should also be pointed out that most studies to date were performed on thick samples, while more technologically relevant thinner films have shown reduced anisotropies [12]. Finally, measurement responses to standard SQUID- and FMR-based studies are also directly proportional to the total magnetic moment present. An alternative approach is to exploit electrical or optical detection techniques to characterise such materials. The direct measurement of the anisotropy field is required especially if there are additional, for example in-plane, anisotropy components to be considered in the material under investigation.

The time-resolved magneto-optical Kerr effect (TR-MOKE) has already been successfully used to characterise the high frequency dynamics of Mn–Ga thin films, showing precession frequencies between 200 GHz and 300 GHz [7]. The frequency of oscillation as a function of the applied field can be fit with the Kittel formula. Such measurements therefore yield not only the ferromagnetic resonance (FMR) frequency and the Gilbert damping parameter, α, but also the effective anisotropy field and energy density (μ0Hk and Keff, respectively). Provided the saturation magnetisation is known, the intrinsic uniaxial anisotropy Ku can be determined from

\[ K_{\text{eff}} = K_u - \frac{1}{2} \mu_0 M_S^2, \]

where \( \mu_0 M_S^2 \) represents the demagnetising energy of an extended thin-film.

The extraordinary Hall effect (EHE) is a very useful characterisation tool for these perpendicularly magnetised materials [13]. When a current is applied along a certain direction in a film, the transverse resistivity (ρxy) is directly proportional to the out-of-plane magnetisation component (Mz) via the extraordinary Hall coefficient, REHE [14]. For perpendicular anisotropy materials, if we apply an external field along the easy axis of the film and switch the magnetisation and therefore ρxy, we will obtain an electrical equivalent of the magnetic hysteresis loop. Similarly, when the field is applied along the hard axis, the saturation of the magnetisation can be seen as a gradual change and saturation of ρyx. From the hard axis data, the anisotropy field, \( \mu_0 H_K \), can be determined. EHE allows for characterisation of these high-anisotropy films where what is normally accessible from standard magnetometry for several reasons: firstly, it is a transport technique and not a magnetometry technique, which means it can be used to measure the magnetic response of volumes of material and/or patterned structures which would be otherwise undetectable; secondly, an inherent advantage is that ρyx exhibits an inverse thickness dependence meaning that the EHE signal is larger for thinner films; thirdly, REHE in ferromagnetic materials is large meaning that even though MS may be low, ρyx can be high. These advantages make EHE an ideal probe of the magnetisation at lower thickness and/or confined geometry.

In the search of materials with higher perpendicular anisotropy and lower saturation magnetisation for spin-transfer-torque applications, alternative characterisation techniques, such as those outlined above, will become increasingly more important. To exemplify this, we investigate \( L1_0 \) Mn1.6Ga (Mn–Ga) and Mn2.6Co0.3Ga1.1 (Mn–Co–Ga) using TR-MOKE and EHE in high magnetic fields. Both materials possess high uniaxial anisotropy. In particular, the chosen composition of Mn–Co–Ga has been shown to exhibit very low saturation magnetisation while retaining high anisotropy [4]. They therefore represent ideal samples for determining the usefulness of these techniques as their magnetic properties are favourable for applications while at the same time it may prove difficult to determine them with more traditional methods, especially at reduced thicknesses [13]. We show that, in particular, EHE at high magnetic fields is an ideal method for determining the anisotropy field of such Heusler systems.

2. Experimental details

Tetragonal Mn–Ga and Mn–Co–Ga thin films were grown by ultra-high-vacuum sputtering on single crystal MgO(001) substrates. Specific details on the sample fabrication and growth conditions can be found in [4]. The stacking structure of the multi-layered films are: MgO(100) substrate/Cr(40)/Mn1.6Ga(30)/Mg(0.4)/MgO(2.0)/AlOx(1.3) and MgO(100) substrate/Mn2.6Co0.3Ga1.1(30)/Mg(0.4)/MgO(2.0)/AlOx(1.3) as sketched in figures 1(a) and (b), respectively. The thickness, in nm, is appended in brackets after each material. The topmost three layers were added to simulate a tunnelling barrier and to protect the sample from oxidation.

Low-field magnetization data were obtained by magnetic field dependent magnetisation measurements at 300 K, up to an applied external field of 6.5 T using conventional SQUID magnetometry. TR-MOKE was used to characterise the high frequency dynamic properties of the Mn1.6Ga film. An 800 nm pump beam with a power of 31.2 mW at the sample, leads to ultra-fast demagnetisation of the sample. The laser pulses were 100 fs in length with a repetition rate of 5.2 MHz. A fixed-delay probe beam of 400 nm, with a power of 105 μW at the sample is then used to probe the excited dynamics via lock-in detection. The diameter of the spot sizes of the pump and probe beams were 17 μm and 5 μm, respectively. The pump and probe fluences, calculated from the incident power, beam diameter and repetition frequency are 1.32 mJ cm−2 and 0.05 mJ cm−2, respectively. Time resolution is obtained by a
variable delay line on the pump beam which allows for the measurement of changes in magnetisation both before and after the demagnetisation process.

EHE was measured initially using a DC current of 1 mA at room temperature in a magnetotransport set-up capable of applying fields up to 1.6 T. Subsequent EHE measurements were performed in a cryostat at 77 K using pulsed magnetic fields at the High Magnetic Field Laboratory located in the Helmholtz–Zentrum Dresden–Rossendorf. The pulsed magnet which was used had a rise time of 7 ms, a fall time of 24 ms and a maximum field of approximately 60 T. A sinusoidal AC current of 1 mA was applied to the sample with a frequency of 47.62 kHz. The transverse voltage was measured during the magnetic field pulse and the signal was locked-in post experiment via a digital lock-in program.

EHE scans along the easy and hard axes, in combination with the evaluated $M_S$ from SQUID, allows for the direct determination of the anisotropy field, $\mu_0 H_K$, as well as the effective anisotropy energy, $K_{\text{eff}}$, by integration of normalised magnetisation loops.

### 3. Magnetic properties

The initial magnetic characterisation of the thin films using SQUID is shown in figure 2. The pure Mn–Ga film—shown in figure 2(a)—exhibits sharp hysteric switching when the field is applied in the out-of-plane direction (closed red circles) with a coercive field, $\mu_0 H_c$, of 290 mT. The data clearly shows that the easy axis is along the film normal. A small in-plane component appears in the hard axis measurements consistent with an intrinsic canted moment on the $2b$ Mn sublattice [6]. The saturation magnetisation, $M_S$, is 0.36 MA m$^{-1}$. In-plane (open black squares) magnetisation measurements give an anisotropy field, $\mu_0 H_K$, of 4.5 T, which was also verified by vibrating sample magnetometry (VSM) up to 14 T (not shown here). These values correspond to an anisotropy energy density $K_{\text{eff}}$ of 0.81 MJ m$^{-3}$.

The addition of Co into Mn–Ga, shown in figure 2(b), leads to a reduction of the $M_S$ to 0.2 MA m$^{-1}$ and an accompanying increase of $\mu_0 H_c$ to 928 mT. The anisotropy field is also increased to a value beyond 5 T. For the in-plane curve, the
The same diamagnetic background as in the out-of-plane measurement was subtracted from the raw data, however as can be seen from the data, an accurate determination of the anisotropy field is not possible. VSM magnetometry up to 14 T was unable to confirm the anisotropy field due to a low magnetisation signal (not shown). We note that, as opposed to the Mn–Ga film, we have a soft magnetic component in both the in-plane and out-of-plane magnetisation curves indicating that we do not have the same canted moment as in the pure film. Rather, the data seems to indicate a segregated phase lacking any anisotropy axis. This could be initially attributed to Co clusters in the film or a lack of full epitaxial growth due to the lack of a seed layer.

The $M(H)$ hysteresis curves of both materials are nearly rectangular. The energy product is thus given by $E = B_r \times H_c$, where $B_r$ and $H_c$ are the remanence and the coercive field, respectively. The out-of-plane energy products are 104 kJ m$^{-3}$ for Mn$_{1.6}$Ga and 165 kJ m$^{-3}$ for Mn$_{2.6}$Co$_{0.3}$Ga$_{1.1}$. The addition of Co leads to a reduction of the maximum energy product, $BH_{\text{max}}$, from about 40 to 10 kJ m$^{-3}$. The values of the energy product and $BH_{\text{max}}$ for Mn$_{2.6}$Co$_{0.3}$Ga$_{1.1}$ are of the same order as bulk Mn$_3$Ga (see [15]).

Figure 3 shows the EHE curves obtained from 5 mm × 5 mm square films measured in the van der Pauw geometry. For both films, the coercivity is identical to that obtained from SQUID measurements. The magnitude of $\rho_{xy}$ for Mn–Ga is much lower than that obtained for the Mn–Co–Ga film. Although the underlying Cr layer is responsible for current shunting, it is expected in any case that pure Mn–Ga films show a lower Hall signal with improving crystal quality [13]. It can be seen that the EHE curve for the Co–Mn–Ga films does not trace out the additional change in magnetisation close to zero field as seen in figure 2(b). As has been calculated for Co-doped Mn–Ga films, Co substitutes Mn randomly at both the $2b$ and $4d$ positions, fills the minority band at $E_F$ and leads to the localisation of electrons in the minority band [16]. Assuming that the soft magnetic component seen in SQUID is related solely to Co substitution, it is likely that, due to the increased electron localisation, the contribution of Co to the magnetotransport is diminished, therefore the same soft phase is not reproduced in the EHE measurement.

4. Time-resolved MOKE

TR-MOKE was used to evaluate the effective anisotropy of the Mn–Ga film. A magnetic field of 0.65 to 1.3 T was applied at 60° to the film normal in order to cant the magnetisation away from the easy axis. This provides a projection of the precession along the film normal which is then measured in the polar MOKE geometry.

Figure 4(a) shows several TR-MOKE spectra at different applied magnetic fields as well as the fitted curves from which the frequency and damping are extracted. It can be seen in this figure that the magnetisation, after the initial demagnetisation pulse at $t = 0$, oscillates around the effective field direction leading to a characteristic oscillation of the optical signal. The ferromagnetic resonance frequency ($f_{\text{res}}$) and the Gilbert damping parameter ($\alpha$) are extracted by fitting these curves with the Kittel formula for the uniform mode. $f_{\text{res}}$ and $\alpha$ are plotted in figures 4(b) and (c), respectively. The precession frequency was found to vary between 136 GHz and 142 GHz in the aforementioned field range. The slope of the fitted line in figure 4(b) is 11.4 GHz T$^{-1}$. The Gilbert damping parameter...
The uniform precession mode is the measurement. This, combined with a single peak in the FFT of excellent agreement with the value obtained from the SQUID estimated. The extracted value of 4.5 T obtained from fitting is in field the effective anisotropy of the film can also be evaluated. As previously mentioned, the anisotropy field of the Mn–Co–Ga film is beyond the accessible range of both the SQUID and the VSM (maximum field of 6.5 T and 14 T, respectively). For the Mn–Co–Ga sample, it was also not possible to obtain any ferromagnetic resonance data from the TR-MOKE which is most likely due to the inability to obtain a large enough canting angle for polar MOKE detection. We therefore determine the anisotropy field using EHE as a detection method.

Figure 5(a) shows the EHE curves for both in-plane and out-of-plane applied fields up to 35 T. The EHE response with the field applied out-of-plane (open red circles) is, again, identical to that obtained from SQUID measurements with sharp switching of the magnetisation visible at approximately 1 T. When the magnetic field is applied in the plane of the sample (closed black triangles) the magnetisation gradually cants to the plane of the film. Since the EHE signal is only sensitive to the out-of-plane component of magnetisation, Mz, the in-plane component of the magnetisation, Mx, must be reconstructed. This is achieved using the transform, sin(cos⁻¹(Mz)) because the projection of the magnetisation along z is simply cos(φ), where φ is the angle of the magnetisation to the film normal (see figure 5(b) inset and figure 1(b)).

Figure 5(b) shows both the Mz component (open red circles), the reconstructed in-plane Mx component (open black squares) and the canting angle, φ (blue dashed line). Here, both Mz and Mx have been normalised to the maximum. It can be seen from the figure that the Mx component is almost totally saturated at 12 T and is completely saturated at 18 T. The anisotropy energy is obtained by evaluating the integral \[ \int_0^{M_s} \mu_0 H, dM_z \text{ between zero and saturation, i.e. the area enclosed by the reconstructed } M_z \text{ curve and the } y\text{-axis and multiplying this by } M_S. \] Beyond saturation the Mx component of magnetisation begins to increase. This is attributed to a small offset between the applied field direction and the film plane. Although the change is rather large in Mx, it corresponds to a misalignment of less than 5°.

From the integration of the area between the reconstructed in-plane and out-of-plane loops, a value of \( K_{\text{eff}} \) of 1.23 MJ m⁻³ is obtained. Although the TR-MOKE results were inconclusive with the Mn–Co–Ga sample, the given value of the anisotropy field and saturation magnetization yield precession frequencies of the order of 350 GHz.

6. Conclusion

We have investigated the static and dynamic magnetic properties of Mn1.6Ga and Mn2.6Co0.3Ga1.1 films. As expected, these materials possess a low saturation magnetisation and high uniaxial anisotropy, usually beyond the accessible field range available in typical SQUID magnetometers. Consequently, standard magnetometry was combined with both the time resolved magneto-optical Kerr effect and the extraordinary Hall effect in high magnetic fields to ascertain exact values for the magnetic anisotropy of both types of thin films.

For the pure Mn1.6Ga alloy, we find a magnetic anisotropy energy of 0.81 MJ m⁻³, with a saturation magnetisation of 0.36 MA m⁻¹. The partial substitution of Mn by Co increases the effective anisotropy energy to 1.23 MJ m⁻³ and decreases the saturation magnetisation to 0.2 MA m⁻¹.

The TR-MOKE and extraordinary Hall effect were utilised to directly probe the anisotropy fields of both materials. The measured values are 4.5 T and 18 T for Mn1.6Ga and Mn2.6Co0.3Ga1.1, respectively.

In summary, we have shown that both the time-resolved magneto-optical Kerr-effect and the extraordinary Hall effect in high magnetic fields are extremely useful techniques in determining the anisotropy energy for materials which cannot be saturated in standard magnetometers such as SQUID. Such indirect techniques can be readily applied to technologically relevant high anisotropy materials for spin-transfer-torque applications.
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