Preparation and structural properties of thin films and multilayers of the Heusler compounds \( \text{Cu}_2\text{MnAl}, \text{Co}_2\text{MnSn}, \text{Co}_2\text{MnSi} \) and \( \text{Co}_2\text{MnGe} \)

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Abstract

We report on the preparation of thin films and multilayers of the intermetallic Heusler compound \( \text{Cu}_2\text{MnAl}, \text{Co}_2\text{MnSn}, \text{Co}_2\text{MnSi} \) and \( \text{Co}_2\text{MnGe} \) by rf-sputtering on MgO and \( \text{Al}_2\text{O}_3 \) substrates. \( \text{Cu}_2\text{MnAl} \) can be grown epitaxially with (100)-orientation on MgO (100) and in (110)-orientation on \( \text{Al}_2\text{O}_3 \) a-plane. The Co based Heusler alloys need metallic seedlayers to induce high quality textured growth. We also have prepared multilayers with smooth interfaces by combining the Heusler compounds with Au and V. An analysis of the ferromagnetic saturation magnetization of the films indicates that the \( \text{Cu}_2\text{MnAl} \)-compound tends to grow in the disordered \( B_2 \)-type structure whereas the Co-based Heusler alloy thin films grow in the ordered \( L2_1 \) structure. All multilayers with thin layers of the Heusler compounds exhibit a definitely reduced ferromagnetic magnetization indicating substantial disorder and intermixing at the interfaces.

Key words: Magnetic properties and measurements; Multilayers;

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1 Introduction

The new, rapidly evolving field of magnetoelectronics [1] started an upsurge of interest in ferromagnetic metals with full spin polarization at the Fermi level. In principal these so called half metallic ferromagnets are ideal for applications in tunnelling magnetoresistance (TMR)[2] or giant magnetoresistance (GMR) [3] elements and as electrodes for spin polarized current injection into semiconductors.

Half metallic ferromagnetic alloys are scarce, since usually the s- and p-type valence electrons contribute both spin directions at the Fermi level. From electronic energy band structure calculations on known several ferromagnetic oxides like CrO$_2$ [4] and La$_{1-x}$Sr$_x$MnO$_3$ [1]. Until now there are only a few intermetallic compounds known to have this unique property, all belonging to the Heusler group with the general formula $A_2BX$ (A=Cu, Co, Ni, B=Mn, Fe..., X=Al, Ge, Si)[5]. The basic ordered Heusler structure is a cubic lattice (space group Fm$ar{3}$m) with four interpenetrating fcc sublattices occupied by A, B or X- atoms respectively. There are several structural variants of the Heusler unit cell with different degrees of site disorder of the atoms on the A, B and X-positions. Among them the B2 structure with a random occupancy of the B and X-position and the completely disordered bcc structure with a random occupancy on the A, B and X-positions [5].

The ferromagnetic half metals known from theoretical electron energy band structure calculations are the compounds PtMnSb and NiMnSb [6] (so called half Heusler compounds since one A-sublattice is empty) and the compounds Co$_2$MnSi and Co$_2$MnGe [7]. Co$_2$MnSn and Co$_2$MnSb in a strict sense do not belong to this group, since they possess only about 90 % of spin polarization at the Fermi level, but they can be made half metallic ferromagnets by alloying [8].

In recent years the properties of thin films of the half Heusler compounds PtMnSb and NiMnSb have been studied intensely by several groups in order to elucidate their potential in the field of magnetoelectronics [9]. These compounds have also been tested already in TMR- and GMR-thin film devices [10], however with only moderate success until now. The main difficulty one encountered when preparing thin films of the half Heusler compounds is a high degree of site disorder. This leads, on the one hand, to strong electron scattering and a low electron mean free path which has a negative influence of the amplitude of the GMR [11]. On the other hand, it is expected that the site disorder destroys the full spin polarization at the Fermi level, which theoretically has been predicted only when assuming perfectly ordered $A_2BX$ structure with pure $L2_1$ type of site symmetry [12].

The half metallic ferromagnets from the Heusler group Co$_2$MnGe, Co$_2$MnSi,
and $Co_2MnSn_{1-x}Sb_x$ found much less attention in the experimental literature until now. Recently two groups published first investigations of GMR elements using $Co_2MnSi$ [13] and $Co_2MnGe$ [14]. Similar to the results on the spin valves based on the half Heusler compounds the amplitude of the GMR was found to be very low, the reason for this was not clear. We have presented our first experimental results on thin films of the Co-based Heusler compounds in [15]. In this paper we present in the first part a detailed study of the preparation and structural properties of thin films of the Heusler phases $Co_2MnSi$, $Co_2MnGe$, $Co_2MnSn$ and $Cu_2MnAl$. The latter Heusler phase is a ferromagnet but not half metallic [16]. We use it as a reference compound and as a seed layer for improving the growth of the Co-based Heusler alloys.

In the second part of the present paper we report on multilayers prepared by combining thin layers of two different Heusler compounds and Heusler compounds with non magnetic metals. Multilayers with Heusler compounds have rarely been studied in the literature until now, we only know of publications on [PtMnSb/NiMnSb] multilayers [17]. Our original intention to study Heusler-based multilayers was to search for an antiferromagnetic interlayer exchange coupling (IEC). Quantum interference models for the IEC suggest that it should exist in virtually any multilayer system combining ferromagnetic and non ferromagnetic metallic layers [18]. Experimentally, however, a prerequisite for the observation of the IEC is a high quality of the layered structure with flat interfaces. Our results to this end were negative until now. We could not find clear evidence for an antiferromagnetic IEC in the multilayer systems we report on in the next section. Instead, we use the multilayers here mainly as a tool for gaining insight into the magnetic properties of the Heusler alloys films in the limit of a very small thickness.

2 Preparation and Experimental

Our thin films and multilayers were deposited by rf-sputtering using pure Ar at a pressure of $5 \cdot 10^{-3}$ mbar as sputter gas. The base pressure of the sputtering system was $5 \cdot 10^{-8}$ mbar, the sputtering rate was 0.04 nm/s for the Heusler compounds, 0.06 nm/s for Au and 0.03 nm/s for V. For the growth of pure Heusler alloy thin films the temperature of the substrates was 470$^\circ$C, the multilayers were grown at a temperature of 300$^\circ$C. A systematic change of the process parameters showed that these values gave the best structural results.

Heusler alloy targets with 10 cm diameter have been made from single phase, stoichiometric ingots prepared by high frequency melting of the components in high purity graphite crucibles. The thin films of the present study were grown
on a-plane sapphire substrates or MgO (100)-substrates which were carefully
cleaned and ion beam etched prior to deposition.

During the sputter deposition process of the multilayers the substrates were
moved automatically between the two targets of the dual source discharge.

After finishing 30 periods of the multilayers we deposited a 2 nm thick Au-cap
layer at room temperature for protection against oxidation. We usually pre-
pared series of 10 multilayers simultaneously within the same run with either
the thickness of the Heusler compound or the thickness of the other metal
varied. The thickness covered typically a range between 0.6 nm and 3 nm
for each component. For preparing the constant layer thickness the substrate
holder was rotated in the symmetric position above the target, for the prepa-
ration of the variable thickness we made use of the natural gradient of the
deposition rate when the substrate holder is in an off-centric position.

The stoichiometric composition of our thin films was controlled using quan-
titative electron microprobe analysis. For Co$_2$MnSi there is a small Si-deficiency
(23 at.% Si instead of 25 at.%), probably caused by selective resputtering of
Si, for Cu$_2$MnAl we find some excess of Cu (52 at.% instead 50 at.%). For
the other two Heusler phases of the present study the thin films preserve the
stoichiometric composition of the targets to within the precision of the micro-
probe analysis of about 0.5 at.%. The structural characterization of all samples was carried out by a thin film
3-circle x-ray spectrometer using Cu $- K_{\alpha}$-radiation. The x-ray study com-
combined small angle reflectivity, $\Theta - 2\Theta$ Bragg scans and rocking scans with
the scattering vector out of the film plane. For selected samples Bragg scans
and rocking scans at glancing incidence with the scattering vector in the film
plane were also taken. The determination of the saturation magnetization was
performed by a commercial SQUID magnetometer (Quantum Design MPMS
system) at a temperature of 5 K and at a field of 0.4 T, which is far above the
coercive force for all samples under study here.

3 Results and discussion

3.1 Cu$_2$MnAl thin films

We first discuss the properties of thin films of the Cu$_2$MnAl Heusler phase.
In the bulk the Fm3m phase of this compound is not stable below 923 K
but decomposes into the phases $\beta - Mn$, $\gamma - Cu_9Al_4$ and Cu$_3$AlMn$_2$ [19].
Interestingly we found, however, that single phase thin films can be prepared
by sputtering on MgO and sapphire a-plane at 470° C and are metallurgically
stable even when annealed for a long time at this temperature. Thus the sput-
tering process and the epitaxial strain seems to establish stability conditions
definitely different from the bulk. **Fig.1** shows an x-ray Bragg scan over the whole angular range of a $\text{Cu}_2\text{MnAl}$ film with a thickness of 100 nm grown on MgO (100). One observes only the Heusler (200) and (400)-peak indicating perfect epitaxial (100)-growth. The out-of-plane rocking width of the (200) Bragg peak was determined to be 0.16°. In the inset of Fig.1 we present the in-plane rocking scan of the Heusler (200) reflection exhibiting 4 peaks at a distance of 90°, as expected for a single crystalline layer. The [010] direction of the Heusler film is rotated by an angle of 45° from the in-plane MgO-[010]-direction. An example of the growth of the $\text{Cu}_2\text{MnAl}$-phase on $\text{Al}_2\text{O}_3$ a-plane is shown in **Fig.2a**. One observes a perfect out-of-plane (220)-texture of the Heusler phase with an out-of-plane rocking width of 0.8° for the (220)-Bragg-peak. An in-plane rocking scan of the (220)-Bragg peak, however, reveals an in-plane polycrystalline structure.

The $\text{Cu}_2\text{MnAl}$ thin films prepared on MgO and on sapphire a-plane at 470° C possess a flat morphology. As an example we show low angle x-ray reflectivity spectra of the $\text{Cu}_2\text{MnAl}$-film in **Fig.2b**. One observes well defined thickness oscillations up to scattering angles of $2\Theta \approx 5°$. From a simulation of the reflectivity spectrum using the Parratt formalism [20] we derive a total thickness of 108 nm and estimate a roughness parameter of 0.6 nm. Atomic force microscopic images of the surfaces also show a very flat surface morphology with a roughness of about 0.7 nm (rms).

An important characterization of the metallurgical state of $\text{Cu}_2\text{MnAl}$ films is the degree of order between the sites A, B and X of the Heusler unit cell. In polycrystalline, bulk material the relative intensity of the superstructure Bragg reflection (111) is conveniently used to determine the order parameter $S$ for the site order between the B-(Mn) and X-(Al) positions, $S=1$ defining perfect order (L2_1-structure) and $S=0$ defining complete site disorder (B2-structure)[21]. Unfortunately for our epitaxial (100) or (110)-films the (111)-Bragg peak is not accessible by our triple axis x-ray spectrometer. Qualitatively the degree of site disorder can be deduced from the value ferromagnetic saturation magnetization [22]. $\text{Cu}_2\text{MnAl}$ single crystals with perfect site order $S \approx 1$ have a saturation magnetization $M_s = 98 \text{ emu/g}$ corresponding to a magnetic moment of about 4.2 $\mu_B/\text{Mn-atom}$, B2-type disorder leads to a decrease of $M_s$, since Mn-spins on the X-position do not couple ferromagnetically to the Mn-spins on the B-position. $\text{Cu}_2\text{MnAl}$ in the completely disordered B2-state exhibits spin glass order with a very low value of the magnetization [23]. For the single crystalline $\text{Cu}_2\text{MnAl}$ film on MgO we get $M_s = 40 \text{ emu/g}$ pointing towards a substantial degree of site disorder. For the film prepared on a-plane $\text{Al}_2\text{O}_3$ we get $M_s = 62 \text{ emu/g}$, this value comes closer to the bulk value for $M_s$.

The structural parameters and the saturation magnetization for the $\text{Cu}_2\text{MnAl}$ phase are summarized in **Table 1**. Note that the reduction of the moment correlates with a definite decrease of the lattice parameter.
3.2 Co$_2$MnSi, Co$_2$MnGe and Co$_2$MnSn thin films

The Co$_2$MnSi, Co$_2$MnGe and Co$_2$MnSn halfmetallic Heusler thin films grown directly on MgO or Al$_2$O$_3$ are polycrystalline and have a bad structural quality and a low value for the saturation magnetization. Only when using suitable metallic seed layers with a typical thickness of about 2 nm we could achieve textured growth and good structural quality. For the Co$_2$MnSn phase we found that the optimum seed layer for the growth on sapphire a-plane is Au with a lattice parameter mismatch of about 1%. V and Cu$_2$MnAl seed layers can also be used. In Fig. 3 we show an out-of-plane Bragg-scan of a Co$_2$MnSn film grown on an Au seed layer. One observes only the (220)- and the (440)-Heusler-Bragg-peak, evidencing pure (110)-texture. The rocking width of the (220) Heusler Bragg-peak is about 3° i.e. it is definitely larger than obtained for the Cu$_2$MnAl layers (see Table 1). The Co$_2$MnSi phase with similar structural quality can be grown on V and Cr seedlayers, Cr giving a slightly better growth quality since it has a lattice mismatch of 0.8% only. For the Co$_2$MnGe-phase V, Au and Cr-seed layers give comparably good structural quality. A summary of these results is given in Table 1. For all three Co-based Heusler compounds the thin films have a very flat surface morphology. As one representative example we show a small angle x-ray reflectivity scan of the Co$_2$MnGe film grown on a V-seedlayer in Fig. 4. One observes well defined finite thickness oscillations from the total layer superimposed by an oscillation from the V-seedlayer. From a fit using the Parrat formalism we estimate a roughness of about 0.5 nm for the interfaces. We also have tested systematically the growth of the Co-based Heusler phases on MgO (100) substrates. The films grown on the bare MgO-surface are polycrystalline. When using metallic seedlayers one can induce reasonable quality out-of-plane (100)-textured growth, however with a definitely larger mosaicity than for the growth on sapphire a-plane, as evidenced by the increased rocking width of the Bragg peaks (see Table 1). Contrary to the case of the Cu$_2$MnAl phase we could not achieve epitaxial growth for the Co-based Heusler alloys, the structure in-plane is always polycrystalline with a broad distribution of the (220) or (200) Bragg peak intensity for an in-plane rocking scan.

An important criterion for the magnetic quality of the thin films is the value of the ferromagnetic saturation magnetization $M_s$ which we have included in Table 1. The values for $M_s$ for the Co-based Heusler alloy thin films are close to the bulk values and for the Co$_2$MnSn and the Co$_2$MnGe-phase nearly coincide with them. This indicates the absence of sizable B2-type of site disorder, consistent with the fact that for these phases the ordered $L2_1$-type phase is very stable [24]. Only for the Co$_2$MnSi-thin film we observe a definitely smaller value of the magnetization in the film than in the bulk, which we would attribute to the deviation from the ideal stoichiometry for this film.

The standard growth temperature we apply for the growth of the films in
Table 1 was 470 °C. In thin film heterostructures the maximum temperature which is allowed for avoiding strong interdiffusion of the components at the interfaces is often definitely lower. Thus it is essential to know the change of site disorder and the sample quality when applying lower substrate temperatures. We prepared series of films of the Heusler alloys at lower substrate temperatures down to T=100 °C. We found that the structural quality, as judged from the Bragg reflection intensity, is only slightly worse when preparing at 300°C, at still lower preparation temperatures, however, there is a definite deterioration of the crystal quality. Simultaneously the ferromagnetic saturation magnetization is strongly reduced for the Cu₂MnAl phase (Fig.5) and moderately for the Co₂MnSn phase and the Co₂MnGe phase. This indicates an increasing degree of site disorder when lowering the preparation temperature.

3.3 Multilayers with Heusler alloys

In this section we want to elucidate the possibility to grow multilayers based on the Cu₂MnAl, Co₂MnSn and the Co₂MnGe compounds. In order to avoid excessive interdiffusion at the interfaces the substrate temperature during the preparation of the multilayers had to be limited to 300 °C, although at this temperature the ferromagnetic saturation magnetization of the Heusler compounds is already definitely reduced (see Fig.5). Actually at 300 °C multilayers with high structural quality of all these phases can be grown on sapphire a-plane by combining them with fcc Au. Fig.6a shows a small angle x-ray reflectivity scan of a [Cu₂MnAl(3nm)/Au(3nm)]₃₀ multilayer with a nominal thickness, as calculated from the sputtering rate, of 3 nm for Au and Cu₂MnAl combined of 30 periods. Above the critical angle for total reflection Θc the multilayer structure gives rise to superlattice reflections superimposed on the Fresnel-reflectivity. We observe superlattice reflections up to 4th order, revealing a good interface quality and low fluctuations of the layer thickness. From the reflectivity peak of order l at the angle Θl one can calculate the superlattice periodicity Λ by using the relation [25]

\[ \Lambda = l \cdot \frac{\lambda}{2(\sqrt{\Theta_l^2 - \Theta_c^2})} \]

(1)

From a fit we get Λ = 5.7 nm in good agreement with the nominal thickness. From simulations of the reflectivity curves using the Parratt formalism [20] we derive an interface roughness of about 0.6 nm. The out-of-plane Bragg scan (Fig.6b) close to the (220)/(111) fundamental Bragg reflection reveals that the multilayer possesses a pure (110) out-of-plane texture for Cu₂MnAl, and (111) texture for the Au-layers. Besides the fundamental Bragg peak from the average lattice, the multilayer exhibits a rich satellite structure caused by
the chemical modulation. Satellites up to the order $l=+3$ and $l=-4$ can be resolved, proving coherently grown superstructures in the growth direction. The position of the satellite peaks give the superstucture periodicity from the separation $\Delta(2\Theta)$ of the satellites of order $l$ from the fundamental Bragg peak [25]:

$$\Lambda = \frac{\lambda}{2 \cdot l \Delta(\Theta) \cdot \cos(\Theta)}$$

From this relation we get a superlattice period of 5.8 nm, in good agreement with the value derived from the small angle x-ray reflectivity. From the width of the satellite peaks at half maximum (FWHM) $\Delta(2\Theta)$ we can derive the out-of-plane coherence length of the superstructure $D_{coh}$ using the Scherrer equation

$$D_{coh} = \frac{\lambda}{\Delta(2\Theta) \cdot \cos(\Theta)}$$

We estimate $D_{coh} = 60$ nm i.e. comprising about 10 superlattice periods. The fundamental Bragg peak in Fig.6b is positioned at $2\Theta = 40.5^\circ$ i.e. at the middle position between the Au (111)-Bragg peak at $2\Theta = 38.5^\circ$ and the $Cu_2MnAl$ (220) peak at $2\Theta = 42.5^\circ$, as expected for a coherently strained superlattice. Multilayers of similar hight quality can also be grown combining the Heusler compounds $Co_2MnGe$ and $Co_2MnSn$ with Au.

In Table 2 we summarize the important parameters characterizing the different multilayers with the Heusler compounds we have grown successfully until now. As revealed by in-plane rocking scans all samples exhibit a broad distribution of Bragg peaks in-plane and thus in are polycrystalline multilayers rather than superlattices.

Multilayers combining the Co-based Heusler alloys with V-interlayers can also be grown. They possess sharp interfaces, however the out-of-plane crystalline order is definitely worse than that we have obtained for the multilayers with Au (see Table 2). We also have grown multilayers combining two different Heusler phases. In Fig.7a we present the small angle reflectivity scan and the large angle Bragg scan of the $[Cu_2MnAl(3nm)/Co_2MnGe(3nm)]_{30}$ multilayer as an example. In the reflectivity one finds sharp superstructure peaks up to the 4th order indicating a good quality of the layered structure with sharp interfaces. From a fit of the reflectivity curve we determined a superlattice periodicity $\Lambda = 6.3$ nm. The Bragg scan close to the (220)-peak exhibits one fundamental superlattice reflection at $2\Theta = 43.2^\circ$ and two weak satellite peaks giving a superlattice periodicity of 6.4 nm. From the FWHM of the satellite peaks we estimate an out-of-plane structural coherence length $D_{coh}$ of about 20 nm thus the superstructure in the growth direction is coherent over about 3 periods.
Coming to the magnetic characterization of the multilayers, we have mea-
sured the ferromagnetic saturation magnetization at 5 K for all multilayers
and summarized the results in Table 2. As discussed above, deviations of
\( M_s \) from the ideal bulk value can be taken as an indication of site disor-
der of the Heusler alloys. By comparison with the bulk value of the mag-
netization (see Table 1) one finds that most of the \( M_s \) values of the multi-
layers are definitely below the bulk \( M_{so} \). This partly can be attributed to
the lower preparation temperature of the multilayers (see Fig.5). For the
\([Cu_2MnAl(3nm)/Au(3nm)]_{30}\) multilayer the magnetization is only about 12% of
the bulk value, consistent with a strongly disordered B2-type of structure
and spin glass magnetic order. The \([Cu_2MnAl(3nm)/Co_2MnGe(3nm)]_{30}\) and the
\([Cu_2MnAl(3nm)/Co_2MnSn(3nm)]_{30}\) multilayers in Table 2 have a relative mag-
netization value \( M_s/M_0 > 1 \), where one should note that \( M_0 \) refers to the
saturation magnetization of the Co-Heusler alloy alone. This clearly shows
that the \( Cu_2MnAl \)-layers in the multilayers posses a substantial ferromagnetic
magnetization definitely larger than that observed for the single \( Cu_2MnAl \)
thin film prepared at the same temperature. The reduced values of the saturation
magnetization for Co-based Heusler multilayers in combinatin with V and Au
in Table 2 suggests an intermixing at the interfaces or an increased degree of
site disorder.

More detailed insight into the metallurgical state and magnetism at the in-
terfaces can be gained by varying the thickness of the Heusler layers in the
multilayers. **Fig.8** shows how the magnetic saturation magnetization in the
multilayers changes when decreasing the thickness of the Heusler layers while
keeping the thickness of the non magnetic layers constant at 3 nm. The fer-
romagnetic saturation magnetization breaks down for a thickness of typically
1.5 nm in all systems. This result suggests that at the interfaces of the mul-
tilayers there exists an intermediate layer of about 0.7 nm thickness which is
metallurgically strongly disordered and not ferromagnetic. We have recently
shown in a separate investigation [26] that the interfaces in \([Co_2MnGe/Au]\)
multilayers develop spin glass order at low temperatures leading to ferromag-
netic hysteresis loops with an unidirectional exchange anisotropy (so called
exchange bias effect). This result gives clear evidence for the existence of non
ferromagnetic interfaces. Quantitatively the decrease of the saturation mag-
netization depicted in Fig.8 depends on the combination of both metals, the
multilayer \([Co_2MnSn/V]\) developing the highest magnetization values in the
thickness range above 2 nm. In comparison the multilayer \([Co_2MnGe/Au]\) has
a rather low value of the saturation magnetization in this thickness range.
4 Summary and conclusions

In summary, we have shown that the Heusler phase $Cu_2MnAl$ can be grown with high structural quality directly on MgO (100) and sapphire a-plane. For the half metallic Co-based Heusler compounds $Co_2MnSi$, $Co_2MnGe$ and $Co_2MnSn$ it is possible to grow thin films with flat surfaces, pure out-of-plane (110) texture and the desired ordered $L2_1$ structure by using metallic seedlayers. Principally this makes these compounds possible candidates for applications in spin transport devices.

A crucial step in this direction is the test of the Co-based Heusler compounds in the limit of very thin films and in combination with other metallic layers. We have shown that for several combinations of the Heusler compounds and nonmagnetic metals high quality, coherent multilayers can be grown down to a thickness range of 1 nm for the Heusler phase. The magnetic measurements however reveal that, depending of the specific combination of materials and the thickness of the Heusler alloy layers, the saturation magnetization is strongly lowered compared to the bulk value. Eventually, for a thickness below typically about 1.5 nm, the Heusler layers are no longer ferromagnetic. This result indicates that typically several monolayers of the Heusler compounds at the interfaces are not ferromagnetic, probably caused by alloying and (or) strong site disorder. This is not unexpected, since an alloying at the interfaces can hardly be avoided in real thin film systems and the chemical conditions for the phase formations of a ternary compound at the interfaces are complex and virtually unknown.

We finally come to the question concerning the potential of the Co-based Heusler alloys in the field of magnetoelectronics in the light of the results presented here. The main problem will be to preserve the full spin polarization predicted for the perfectly ordered Heusler structure in very thin layers of real devices. We have shown that for the preparation temperatures allowed in thin film heterostructures the formation of site disorder cannot be completely avoided in the Co-based Heusler alloys. Site disorder in the interior of the Heusler film is a critical factor, since it must be expected that the full spin polarization is lost in disordered Heusler alloys [7]. The question, to what extend some site disorder is tolerable, i.e. leaves at least a high value for the spin polarization, cannot be answered quantitatively at the moment, since corresponding band structure calculations have not been published yet. The existence of non ferromagnetic interfaces, which seem to be present in all combinations of the Co-based Heusler alloys and other metals which we have studied until now, causes a second problem, which might be even more detrimental for the performance of spin transport devices. Necessarily the spin polarization at the Fermi level will completely vanish in a non ferromagnetic
interlayer. Since in GMR with the current in the plane (cip-geometry) the spin
dependent electron scattering at the interfaces is dominating [3], non ferro-
magnetic interfaces are expected to reduce the GMR-value strongly. This is in
accord with our first results of magnetoresistance measurements for spin valve
systems using the Co-based Heusler alloys, which reveal very small values for
the GMR effect [27]. Possibly one can overcome this problem by using the
GMR geometry with the current perpendicular to the plane (cpp-geometry).
In this geometry one can use much larger thicknesses of the ferromagnetic
layers and the spin asymmetry of the electron scattering in the interior of
the ferromagnetic layers gives an important contribution to the GMR [28].
However, concerning technical applications the cpp-geometry seems not very
useful. The alternative choice would be to search for other material combina-
tions or preparation methods with less interdiffusion at the interfaces.

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5 Figure Captions

Fig. 1
Out-of-plane x-ray Bragg-scan of a $Cu_2MnAl$ film on MgO(100). The inset shows the in-plane rocking scan of the Heusler (200) peak.

Fig. 2
(a) Out-of-plane Bragg-scan of a $Cu_2MnAl$ film grown directly on $Al_2O_3$ and (b) low angle x-ray reflectivity spectrum of the same film.

Fig. 3
Out-of-plane Bragg-scan of a $Co_2MnSn$ film grown on an Au seed layer.

Fig. 4
Small angle x-ray reflectivity scan or a $Co_2MnGe$ film on $Al_2O_3$ with a V-seedlayer

Fig. 5
Saturation magnetization of $Co_2MnGe$, $Co_2MnSn$ and $Cu_2MnAl$ versus the substrate temperature during preparation.

Fig. 6
(a) Small angle x-ray reflectivity scan of a multilayer $[Cu_2MnAl(3nm)/Au(3nm)]_{30}$ with a nominal thickness of 3 nm for Au and $Cu_2MnAl$ and (b) out-of-plane Bragg scan of the same multilayer. The numbers in the figure denote the order of the superlattice reflections and the order of the satellites
Fig. 7
(a) Small angle reflectivity scan of a \([Cu_2MnAl_{3nm}/Co_2MnGe_{3nm}]_{30}\) multilayer and (b) large angle Bragg-scan for the same sample.

Fig. 8
Ferromagnetic saturation magnetization of multilayers measured at 5 K as a function of the thickness of the Co-Heusler layers. The thickness of the other layer is kept constant at 3 nm.
### Table 1
Structural parameters and saturation magnetization measured at 5 K for the Heusler films grown on Al$_2$O$_3$ a-plane or MgO (100)

| Phase                  | Prep. temp. (°C) | Substr./seed layer | Texture | Rocking width (220)-peak (°) | Lattice parameter (nm) | Magnetization (emu/g) |
|------------------------|------------------|--------------------|---------|-------------------------------|------------------------|-----------------------|
|                        |                  |                    |         |                               | bulk       | film      | bulk   | film   |
| Cu$_2$MnAl             | 470              | Al$_2$O$_3$ a-plane| (110)   | 0.8                           | 0.5962      | 0.5907    | 98     | 62     |
| Cu$_2$MnAl             | 470              | MgO (100)          | (100)   | 0.16                          | 0.5922      | 0.5972    | 40     |        |
| Co$_2$MnSi             | 470              | Al$_2$O$_3$ a-plane/Cr| (110) | 4                             | 0.5654      | 0.5688    | 138    | 98     |
| Co$_2$MnSi             | 470              | MgO (100)/Cr       | (100)   | 10                            | 0.5670      | 0.5760    | 100    |        |
| Co$_2$MnGe             | 470              | Al$_2$O$_3$ a-plane/V | (110) | 3                             | 0.5743      | 0.5766    | 111    | 103    |
| Co$_2$MnGe             | 470              | MgO (100)/V        | (100)   | 5                             | 0.5803      | 0.5830    | 107    |        |
| Co$_2$MnSn             | 470              | Al$_2$O$_3$ a-plane/Au | (110) | 3                             | 0.6000      | 0.6003    | 91     | 87     |
| Co$_2$MnSn             | 470              | MgO (100)/Au       | (100)   | 6                             | 0.6011      | 0.6011    | 80     |        |

### Table 2
Structural parameters of the Heusler multilayers grown on Al$_2$O$_3$ a-plane and the relative saturation magnetization measured at 5 K. $M_0$ denotes the saturation magnetization of the bulk Co-Heusler compounds (see Table 1)

| Multilayer | Texture | Period length (nm) | Coherence length (nm) | Lattice-parameter (nm) out-of-plane | $M_s/M_0$ |
|------------|---------|--------------------|-----------------------|------------------------------------|-----------|
| [Cu$_2$MnAl(3nm)$_{30}$/Au(3nm)]$_{30}$ | (110)/(111) | 5.7 | 60 | 0.610 / 0.400 | 0.12 |
| [Cu$_2$MnAl(3nm)$_{30}$/Co$_2$MnSn(3nm)]$_{30}$ | (110) | 6.0 | 20 | 0.598 / 0.598 | 1.45 |
| [Cu$_2$MnAl(3nm)$_{30}$/Co$_2$MnGe(3nm)]$_{30}$ | (110) | 6.4 | 18 | 0.588 / 0.588 | 1.09 |
| [Co$_2$MnSn(3nm)$_{30}$/Au(3nm)]$_{30}$ | (110)/(111) | 5.9 | 50 | 0.615 / 0.400 | 0.68 |
| [Co$_2$MnSn(3nm)$_{30}$/V(3nm)]$_{30}$ | (110) | 6.3 | 30 | 0.596 / 0.299 | 0.71 |
| [Co$_2$MnGe(3nm)$_{30}$/V(3nm)]$_{30}$ | (110) | 6.2 | 35 | 0.584 / 0.292 | 0.70 |
| [Co$_2$MnGe(3nm)$_{30}$/Au(3nm)]$_{30}$ | (110)/(111) | 5.9 | 70 | 0.620 / 0.409 | 0.47 |
Fig. 1.
Fig. 2.
Fig. 3.

Fig. 4.
Fig. 5.
Fig. 6.
Fig. 7.
Fig. 8.