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Spin polarization of single-crystalline Co_2MnSi films grown by PLD on GaAs(0 0 1)

W.H. Wang[a], M. Przybylski[a,b,*], W. Kuch[a], L.I. Chelaru[a], J. Wang[a], Y.F. Lu[a], J. Barthel[a], J. Kirschner[a]

[a] *Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany*
[b] *Solid State Physics Department, Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, Mickiewicza 30, 30-059 Krakow, Poland*

Abstract

Single-crystalline Co_2MnSi Heusler alloy films have been grown on GaAs(0 0 1) substrates by pulsed laser deposition. The best crystallographic quality has been achieved after deposition at 450 K. Spin-resolved photoemission measurements at BESSY reveal spin-resolved density of states that are in qualitative agreement with recent band structure calculations. The spin polarization of photoelectrons close to the Fermi level is found to be at most 12% at room temperature, in contrast to the predicted half-metallic behavior. We suggest that this discrepancy may be attributed to a non-magnetic surface region and/or partial chemical disorder in the Co_2MnSi lattice.

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Keywords: Half-metallic ferromagnets; Spin polarization; Heusler alloys

It was successfully proven recently that a magnetic-semiconductor-based system is feasible as the spin-electronic analog of the electro-optic light modulator, however only under conditions of low temperatures and high magnetic fields [1]. From the pragmatic requirement of functionality with small magnetic fields and at environmental temperatures, the concept of spin injection from a metallic ferromagnet into a semiconductor is still very attractive. Due to a recent work that imposed severe restrictions on the functionality of integrated ferromagnetic metal-semiconductor devices, this concept needs substantial modifications like an application of the materials showing 100% spin polarization of the injecting electrons [2]. Half-metallic ferromagnets have been proposed as ideal candidates for spin injection devices because

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[*] Corresponding author. Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany. Tel.: +49 345 5582969, fax: +49 345 5511223.

E-mail address: mprzybyl@mpi-halle.de (M. Przybylski).

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they have been predicted to exhibit 100% spin polarization. Notable among the half-metallic candidates are a number of half-Heusler alloys (NiMnSb, PtMnSb) and full-Heusler alloys (Co_2MnSi, Co_2MnGe, Co_2MnSn, Fe_2MnSi) [3,4]. Especially, the full-Heusler alloy Co_2MnSi has attracted strong interest due to a large minority spin band gap of ~0.4 eV at the Fermi level [5] and the highest Curie temperature (985 K) amongst the known half- and full-Heusler alloys [6]. In bulk form, Co_2MnSi has been stabilized in the cubic $L2_1$ structure with a lattice parameter of 5.67 Å, which implies a lattice mismatch to GaAs(0 0 1) of only 0.3%.

In this paper, we report on the magnetic characterization of single-crystalline Co_2MnSi thin films deposited by pulsed laser deposition (PLD) on GaAs(0 0 1) substrates. The best crystallographic quality of the films has been achieved after deposition at 450 K [7]. The films exhibit in-plane uniaxial magnetic anisotropy with the easy axis of magnetization along the [1 -1 0] direction superimposed on the four-fold anisotropy preferring magnetization along the four⟨1 1 0⟩ in-plane directions. Details concerning magnetization reversal for varying film thickness and at different temperatures will be published elsewhere [7].

Spin-resolved photoemission measurements were performed at BESSY in Berlin of the single-crystalline Co_2MnSi films of the thicknesses: 17, 23, 45 and 85 Å, which were grown at our Institute in Halle. After deposition, the samples were immediately transferred under UHV conditions (a pressure of about 4×10^{-8} Pa was maintained in the transfer chamber) to BESSY in Berlin. The spin polarization was probed at room temperature about 8 hours after sample preparation. Prior to the photoemission measurements the films were magnetized in an external magnetic field applied along the in-plane easy [1 -1 0] direction. The measurements were performed in remanence, the presence of which was regularly checked between and after scans of data acquisition using longitudinal magneto-optical Kerr effect (MOKE). Spin-resolved photoemission spectra were taken at the UE56/2 beam line. The spectra were taken for 45° incidence of the incoming radiation in the plane defined by the magnetization and surface normal, and normal emission of the outgoing photoelectrons.

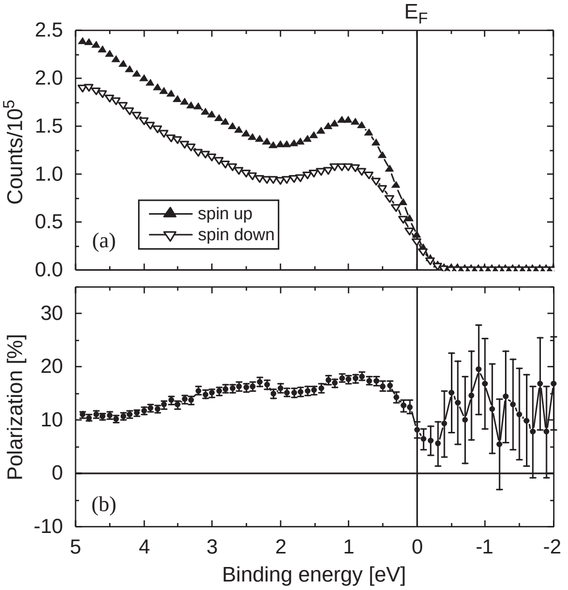

Fig. 1. (a) Spin-resolved photoemission spectra of 23 Å Co_2MnSi/GaAs (0 0 1) for 70 eV photon energy, taken for 45° incidence of the incoming radiation in the plane defined by the magnetization and surface normal, and normal emission of the outgoing photoelectrons; filled (open) triangles denote majority and minority spin spectra. (b) Spin polarization, defined as difference between majority and minority photoemission intensity normalized to the total intensity, of the spectrum of a.

A representative example of spin-resolved photoemission spectra of a 23 Å Co_2MnSi film on GaAs(0 0 1) is presented in Fig. 1a for photon energy of 70 eV. Spin-resolved photoemission measurements reveal spin-resolved density of states that are in qualitative agreement with recent band structure calculations (cf. Fig. 5 of Ref. [8]). As in these calculations, a peak around 1 eV binding energy is seen, accompanied by a gentle signal increase towards higher binding energies. In the whole probed binding energy range a prevailing majority-spin polarization is found. The spin polarization, defined as the difference between the partial intensity spectra for majority and minority spin normalized to the total intensity, is shown in Fig. 1b. The structure of the spectrum and its interpretation with respect to the electronic bands will be discussed elsewhere [9]. In this contribution, we concentrate on the spin polarization at the Fermi level, which is found to vary between 8.1% and 10.6% depending on the film thickness. The lowest spin-polarization is detected for the thinnest film (17 Å), which could be attributed to a reduced Curie temperature of the film of this thickness. An extrapolation to zero temperature was performed in order to exclude the magnetic size effect, which contributes to the size dependence of the spin polarization at a finite temperature. To follow this idea, a number of Co_2MnSi films of varying thickness, prepared exclusively for this purpose, were measured by MOKE at different temperatures [7]. The external magnetic field was always applied along the [1 -1 0] direction which was found to be the easy-axis of magnetization independently of the film thickness. The loops were of regular, rectangular shape, and saturated at fields of the order of tens of militesta. The Kerr rotation in remanence was considered as 100% of the saturation values. The results are plotted in Fig. 2. It is clearly seen that the magnetization depends most strongly on temperature for the thinnest films in accordance with the commonly accepted fact that, at finite temperatures, the film magnetization decreases with decreasing film thickness. However, the character and rate of this decrease are very controversial, both in experimental and theoretical approaches. There is no general theoretical formula to describe the temperature dependence of magnetization in ultrathin films. Nevertheless, for all our Co_2MnSi films, the temperature dependence of the Kerr rotation in saturation could be described, within reasonable limits and at low temperatures ($T < 0.6T_c$), by the Bloch formula: $M(T) = M(0)(1 - bT^{3/2})$, where $M(0)$ and b depend on the film thickness. In view of the limited accuracy of the measurements, possible deviations from this formula could not be evaluated. We intentionally fitted all our data to the same formula in order to obtain the phenomenological parameter b for each of our samples [10].

The average spin-wave parameter b, taken from the above quantitative description of the thermal reduction of the average magnetization, is used to describe the dependence of average magnetization on thickness at a finite temperature. The dependence of b on the reciprocal of the Co_2MnSi film thickness d, showing some systematic, is presented in Fig. 3 for all the samples for which the magnetization vs. temperature was followed. Linear b vs. $1/d$ dependence is a direct consequence of the increasing surface/interface contribution to the magnetization with decreasing film

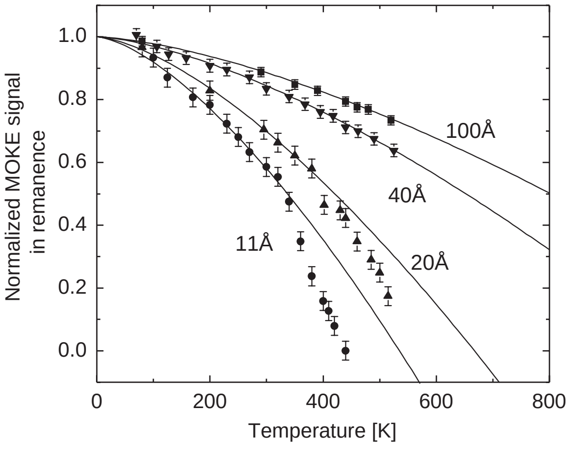

Fig. 2. Normalized values of Kerr signal in remanence vs. temperature describing the average film magnetization for Co_2MnSi films of various thicknesses. The results of fit to the $M(T) = M(0)(1 - bT^{3/2})$ formula, at $T < 0.6T_c$, are included (solid lines).

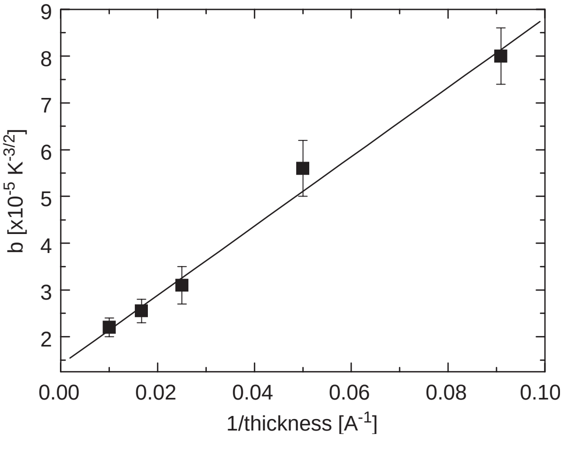

Fig. 3. Average spin-wave parameter b as a function of the reciprocal of the thickness d of the Co_2MnSi films grown on GaAs(0 0 1). A linear fit (solid line) is performed to the experimental values (dots).

thickness [10]. Based on the relation plotted in Fig. 3, the *b* parameter can be estimated for any thickness of the Co_2MnSi film.

Consequently, based on the temperature dependence of *M*, calculated using the Bloch formula and the experimentally determined thickness dependence of *b*, the values of the spin polarization can be extrapolated to 0 K. The procedure was previously successfully applied in the case of magnetic moments measured by X-ray magnetic circular dichroism (XMCD), and reported elsewhere [7,9]. The procedure applied for the spin-polarization close to the Fermi level, which was measured at RT only, leads to an almost thickness-independent value at 0 K. This value equals to $12.5 \pm 0.4\%$, in contrast to the predicted half-metallic behavior. In order to explain why the spin polarization we measured for our Co_2MnSi films is so low one has to recall all the technological and experimental limitations. We suggest that this discrepancy may be attributed to a non-magnetic surface region and/or partial chemical disorder in the Co_2MnSi lattice. A non-magnetic metallic phase would add an equal strength to the majority- and minority-spin components, effectively decreasing the polarization within the spin gap. It is also suspected that surface effects like a surface segregation and local atomic disorder are the causes of diminished spin polarization. This is due to the gap between the majority and minority spin states that can be closed by the introduction of disorder into the crystal lattice [11]. Even in the case of a single crystal of Co_2MnSi grown by the Czochralski method, an Andreev-reflection method indicated only a value of 56% of the spin polarization at 4.2 K [12]. One has also to keep in mind that due to the finite detection angle of the photoelectron spectrometer, the presented photoemission data do not represent a k-space sampling of the complete Brillouine zone.

In conclusion, we have epitaxially grown single-crystalline Co_2MnSi films on GaAs(0 0 1). At maximum spin polarization of only 12% at the Fermi energy was obtained from spin- resolved photoemission experiment after extrapolation of the RT data to 0 K. The discrepancy between 100% spin-polarization expected for the half-metallic Heusler alloy, and what was measured for Co_2MnSi films, we interpret as due to a non-magnetic surface region and/or partial chemical disorder in the Co_2MnSi lattice. This suggests that with more elaborate surface preparation, and/or in situ Co_2MnSi film growth, it would be possible to generate much higher spin polarization.

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