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Magnetic microstructure of candidates for epitaxial dual Heusler magnetic tunnel junctions

A. Kaiser ^{a,}*, D. Banerjee ^a, A.D. Rata ^b, C. Wiemann ^a, S. Cramm ^a, C.M. Schneider ^a

a Forschungszentrum Jülich, Institut für Festkörperforschung IFF-9 and JARA-FIT, 52425 Jülich, Germany **b Leibniz-Institut für Festkörper- und Werkstoffforschung, Dresden, Germany**

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1. Introduction

Magnetic tunnel junctions (MTJ), i.e. two magnetic electrodes separated by a very thin tunneling barrier, are currently receiving great interest due to their technical importance in spintronics [\[1\].](#page--1-0) The applications range from hard disk read heads to memory cells in magnetic random access memories or magnetologic circuits. The tunneling magnetoresistance (TMR) effect in the diffusive limit can be quantitatively described by Jullière's model $[2]$ as

$$
TMR = \frac{R_{\uparrow\downarrow} - R_{\uparrow\uparrow}}{R_{\uparrow\uparrow}} = \frac{2P_1P_2}{1 - P_1P_2}
$$

with the spin-polarization values at the Fermi level of the two electrodes P_1 and P_2 and the resistances $R_{\uparrow\downarrow}$ and $R_{\uparrow\uparrow}$ of the antiparallel and the parallel magnetization configuration, respectively. In this simplified model half-metals with a spin polarization of 100% at the Fermi level are expected to show an infinitely high TMR effect. By the use of half-metallic $La_{2/3}Sr_{1/3}MnO₃$ electrodes a TMR effect of 1800% was already demonstrated at low temperatures [\[3\]](#page--1-0). At room temperature this effect vanishes due to the low Curie temperature of $La_{2/3}Sr_{1/3}MnO₃$. Another promising class of half-metallic materials with noticeably higher Curie temperatures is the group of full-Heusler compounds [\[4\].](#page--1-0) Several cobalt-based Heusler alloys with Curie temperatures well above

ABSTRACT

Heusler alloys are considered as interesting ferromagnetic electrode materials for magnetic tunnel junctions, because of their high spin polarization. We, therefore, investigated the micromagnetic properties in a prototypical thin film system comprising two different Heusler phases Co₂MnSi (CMS) and Co2FeSi (CFS) separated by a MgO barrier. The magnetic microstructure was investigated by X-ray photoemission electron microscopy (XPEEM). We find a strong influence of the Heusler phase formation process on the magnetic domain patterns. $SiO_2/V/CMS/MgO/CFS$ and $SiO_2/V/CFS/MgO/CMS$ trilayer structures exhibit a strikingly different magnetic behavior, which is due to pinhole coupling through the MgO barrier and a strong thickness dependence of the magnetic ordering in $Co₂MnSi$.

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room temperature have been predicted to show 100% spin polarization at the Fermi level [\[5\]](#page--1-0) and are therefore promising candidates for MTJ electrode materials. Dual-Heusler MTJs have already been realized with $Co₂MnSi$ electrodes and an AlO_x tunneling barrier reaching high TMR values [\[6\].](#page--1-0) By the use of epitaxially grown electrode-barrier combinations even higher values can be expected due to the reduction of defects and the onset of resonant tunneling mechanisms [\[7\].](#page--1-0) Magnesium oxide (MgO) provides compatible lattice constants to the above mentioned Heusler alloys allowing epitaxial growth of the MTJ structures [\[8\].](#page--1-0)

The full-Heusler compounds of the type $X₂YZ$ are among the materials predicted to exhibit half-metallic behavior with 100% spin polarization in the bulk [\[5\].](#page--1-0) $Co₂MnSi$ (CMS) and $Co₂FeSi$ (CFS) are two protagonists of this class, having similar lattice constants and providing structural compatibility to MgO. Both materials have very high Curie temperatures of 985 K (CMS, [\[9\]\)](#page--1-0) and 1100 K (CFS, [\[10\]](#page--1-0)), thereby fulfilling a prerequisite for their potential use in applications. The magnetic moment per formula unit is 5.07 μ_B (CMS) and $6 \mu_B$ (CFS), respectively. The experimental results described below show that both materials have clearly distinguishable coercive fields. This opens a pathway to combine CFS and CMS in one MTJ in the sense of a pseudo spin-valve structure, i.e., without using exchange bias materials for the reference magnetization direction.

Up to now many macroscopic studies of the magnetic, electronic and structural properties of Heusler-based MTJs have been carried out. From a fundamental point of view, but also with

^{*} Corresponding author.

E-mail address: [a.kaiser@fz-juelich.de \(A. Kaiser\).](mailto:a.kaiser@fz-juelich.de)

respect to technical aspects, the micromagnetic behavior of small elements cannot be neglected. The equilibrium magnetization of magnetic thin film elements is characterized by the minimum of the magnetic free energy, which is composed of several competing contributions, such as the exchange energy penalizing deviations from a homogeneous magnetization direction, the anisotropy energy describing the preferred orientation of the magnetization with respect to certain crystalline axes of the material, the Zeeman energy describing the interaction with the external field and the stray field energy connected with the demagnetizing field generated by the element itself [\[11\]](#page--1-0). The stray field H_d depends strongly on the dimensions and the shape of the magnetic element. Thus, by microstructuring the samples using lithographic techniques the influence of H_d on different element sizes can be studied and compared to the effect of the other competing energy terms—provided that these remain unchanged. In thin film layer stacks, an additional energy term comes in due to coupling mechanisms between the ferromagnetic layers, for instance, interlayer exchange coupling or Néel coupling. For the performance of MTJs, these coupling contributions may be very important and may have a considerable influence on the magnetic microstructure.

There is a variety of well-established techniques known for the investigation of the magnetic microstructure, such as magnetooptical Kerr microscopy, Lorentz microscopy, magnetic force microscopy, or scanning electron microscopy with spin-polarization analysis [\[12\].](#page--1-0) Only few of them, however, combine magnetic sensitivity with chemical selectivity. The chemical selectivity is an indispensible precondition for the separation of the magnetic contributions in a layered system. One of these techniques is photoemission electron microscopy (PEEM) in combination with polarized soft X-ray synchrotron radiation. It is a powerful tool to element-selectively investigate the local domain structure with both high spatial [\[13,14\]](#page--1-0) and temporal [\[15\]](#page--1-0) resolution, and has, therefore, been used as the main method in our studies.

2. Experimental details

Single films and trilayer structures have been prepared by magnetron sputtering onto silicon substrates with a 20 nm vanadium seed layer at room temperature, followed by an in-situ annealing step at 450 °C (single films) and 300 °C (trilayers with MgO), respectively. During the sputtering process a weak in-plane magnetic field of 7 mT was present in the deposition chamber. Later, the samples have not been exposed to any other magnetic field and have been studied in the as-grown state. The films have been capped with a thin vanadium layer to prevent oxidation of the layer stack. X-ray diffraction patterns showed the films to grow in a highly textured B2-phase. This is expected to result in a reduction of the spin-polarization and, therefore, a loss of the halfmetallicity as compared to the preferred $L2_1$ -phase [\[16\]](#page--1-0). The films have been subsequently microstructured by optical lithography and argon ion beam milling into squares with areas ranging from 2×2 to $100 \times 100 \mu m^2$. Prior to the microscopy experiments $M(H)$ -curves have been measured using vibrating sample magnetometry in order to determine the coercive fields and magnetic moments. By the use of synchrotron radiation the static magnetic microstructure of the samples was investigated element-selectively with an Elmitec PEEM III at the variable polarization undulator beamline UE56/1-SGM at BESSY-II (Berlin). For this purpose the photon energy was tuned to the corresponding L3-absorption edge of Mn, Fe, or Co and the XMCD asymmetry value $A = (I_{\sigma+} - I_{\sigma-})/(I_{\sigma+} + I_{\sigma-})$ was calculated for each pixel with $I_{\sigma\pm}$ being the intensity values for right and left circularly polarized light.

3. Results and discussion

3.1. Single films

Fig. 1 shows the $M(H)$ -curves of 20 nm thick single films of CMS and CFS measured with the magnetic field applied in the film plane. They exhibit clearly distinguishable coercive fields of 2.8 mT (CMS) and 6.5 mT (CFS) and magnetic moments per formula unit of 4.0 μ_B (CMS) and 5.2 μ_B (CFS). The reduction of the magnetic moments compared to the bulk values can be explained by atomic site disorder and interface effects. The hysteresis measurements have been carried out with the magnetic field applied along different in-plane angles in order to determine a possible global magnetic anisotropy in the samples. For both samples no sizable in-plane anisotropy could be found. This in-plane isotropy in the samples was also confirmed by FMR measurements [\[17\].](#page--1-0)

The magnetic structure of both materials [\(Fig. 2\)](#page--1-0) is strongly influenced by the polycrystalline nature of the films. In both films multidomain states are formed with the local magnetization direction laterally varying due to anisotropy fluctuations. However, a perpendicular magnetization direction is strongly suppressed by the demagnetizing field, which forces the average magnetization direction into the film plane. A direct comparison of the images in [Figs. 2\(](#page--1-0)a) and (b) reveals clear differences between CMS and CFS. Both films develop a magnetization ripple with the average magnetization vector pointing perpendicular to the domain walls [\[18\]](#page--1-0). The existence of magnetization ripple can be attributed to the polycrystalline nature of the films leading to a strong variation of the local magnetocrystalline anisotropy associated with each grain. The average wavelength of the periodic ripple structure can be correlated with the average crystallite size [\[19\].](#page--1-0) In our measurements the CMS film showed a more textured magnetization distribution with a preferred axis, giving rise to the ''feathery'' pattern. By contrast, the CFS films reveal a grain-type domain structure, which is possibly related to a smaller grain structure and higher disorder in the film. The average ripple wavelength in CMS is about $1.89 \mu m$ and significantly larger than the value for the respective CFS films $(0.68 \,\mu m)$.

In comparing with Fig. 1, it is also interesting to note that the shorter ripple wavelength in CFS is, in fact, related to a higher coercivity. This finding is consistent with the picture that the

Fig. 1. In-plane $M(H)$ -curves of 20 nm Co₂MnSi (CMS) and Co₂FeSi (CFS) thin films measured with vibrating sample magnetometry, showing the difference in saturation magnetization and coercive fields.

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