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Structural and magnetic properties of strongly carbon doped Fe–Co thin films

G. Giannopoulos [a,*], L. Reichel [b,c], A. Markou [d], W. Wallisch [e], M. Stöger-Pollach [f], I. Panagiotopoulos [d], V. Psycharis [a], S. Fähler [b], J. Fidler [e], D. Niarchos [a]

[a] INN, NCSR Demokritos, Athens 15310, Greece
[b] IFW Dresden, PO Box 270116, 01171 Dresden, Germany
[c] TU Dresden, Institute for Materials Science, 01062 Dresden, Germany
[d] Department of Materials Science and Engineering, University of Ioannina, Ioannina 45110, Greece
[e] Vienna University of Technology, Institute Solid State Physics, Vienna 1040, Austria
[f] Vienna University of Technology, University Service Center for Transmission Electron Microscopy, 1040 Vienna, Austria

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ABSTRACT

In the framework of the ongoing research for novel rare earth free permanent magnet materials, the alloy Fe–Co–C has attracted interest from theorists, since carbon could induce a magneto-crystalline anisotropy. In this work structural and magnetic properties of strongly doped magnetron sputtered thin films were investigated. Au–Cu buffers on MgO (100) substrates were used in order to promote epitaxial FeCo with 001 orientation. By adding carbon as a third element a tetragonal distortion was observed, according to structural measurements. An anisotropic behavior was induced in the magnetic properties of the system, where the magneto-crystalline anisotropy constant K_u value was estimated in the order of 0.8×10^6 J/m^{3}f or 3 nm thick Fe–Co(C) magnetic layer.

1. Introduction

Fe–Co is proposed as a possible candidate alloy for permanent magnet applications due to its high intrinsic magnetic moment. Apart from high magnetic moment, high magneto-crystalline anisotropy is required for permanent magnets. In this work the effect of carbon doping on the stabilization of metastable tetragonal phases, with strained unit cells and high magneto-crystalline anisotropyin sputtered Fe–Co thin films is studied. According to theoretical calculations FeCo alloy can support large uniaxial magnetic anisotropy energy (MAE), saturation magnetization M_s, while chemical disorder affects MAE significantly [1–3]. Tetragonal distortion can be induced via coherent growth of Fe–Co layers on different substrate or buffer materials. This was shown in various experimental studies. However, only ultrathin Fe–Co layers with a maximum thickness of 15 monolayers exhibit a perpendicular magnetic easy axis [4–6]. This strong magneto-crystalline anisotropy (MCA) was attributed to the tetragonal strain, expressed by the $(c/a)_{bcc}$ ratio of lattice distortion in the range between 1.1 and 1.25 according to Burkert et al. [1]. The reason why a strong MCA was not observed in thicker films is the lattice relaxation, which leads to a reduction of c/a to its equilibrium value of 1, as demonstrated with in situ measurements [7]. From these experimental studies, the chosen buffer layer was always considered being the key parameter to distort the Fe–Co lattice. For this purpose, a variable composition Au_xCu_{1-x} alloy library has been prepared based on combinatorial sputtering methods, in order to achieve Taylor made lattice parameter values [8]. Several single crystal substrates have also been employed to create an appropriate lattice mismatch [9–11]. Adding a third element, e.g. carbon, was recently proposed to stabilize the strain in Fe–Co and thus to hamper a complete lattice relaxation [12]. Remarkable magneto-crystalline anisotropy values were predicted for ternary Fe–Co–C system, according to these DFT calculations [9]. Carbon atoms occupying the interstitial positions in the FeCo lattice cause a tetragonal distortion. These suggestions from DFT theory were already confirmed for Fe–Co with low additions of C of approximately 2 at% by Reichel et al. [7]. In their $(Fe_{0.4}Co_{0.6})_{0.98}C_{0.02}$ films, a strain of $c/a = 1.03$ was observed which led to K_u of 0.44 MJ/m³. In this study, we introduce Fe–Co films with higher additions of C and demonstrate the consequences of the lattice strain, towards achieving a high tetragonal distortion of the unit cell.

* Corresponding author.
E-mail address: g.giannopoulos@inn.demokritos.gr (G. Giannopoulos).

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2. Experimental details

An ATC 2200-V high vacuum magnetron sputtering system supplied from AJA Inc. was used to prepare the samples, with a base pressure of 4×10^{-9} Torr. The depositions were performed on single crystalline MgO (100) substrates. The layer structure consists of a 3 nm Cr seed layer, a 30 nm Au–Cu buffer and the Fe–Co (C) magnetic layer. Two different stoichiometry buffers were deposited at 300 °C, $Au_{30}Cu_{70}$ and $L1_0$ ordered $Au_{50}Cu_{50}$ with cubic and tetragonal symmetry respectively. Fe–Co–C films of different thickness and carbon percentage were grown on these buffers at 300 °C. Additionally, $Fe_{45}Co_{55}$ composition was studied, which was verified by EDX measurements for the thicker films. The thickness was varied between 1 nm and 50 nm and carbon percentage from 0% to 20%. Carbon percentage was confirmed by Rutherford Backscattering Spectroscopy. X-ray diffraction (XRD) in Bragg–Brentano geometry was conducted in a Siemens D500 diffractometer. Additional pole figure measurements to study the lattice symmetry of the films, were carried out on an X'pert four circle goniometer, also using Cu-Ka radiation. The magnetic properties of the films were examined with a MPMS SQUID magnetometer at room temperature. Transmission electron microscopy (TEM) investigations were performed on a Titan3 80-300 microscope, equipped with C_5 corrector and Schottky field emission electron source and Quanta 200 3D DualBeam. The surface topology mapping was also examined using an Asylum Research Cypher Atomic Force Microscope (AFM). Two sputtered 3 nm thin Fe–Co films, doped with a different carbon concentrations (0 and 20 at%, respectively) were studied in order to analyze their magnetic behavior by means of energy loss magnetic chiral dichroism (EMCD) in order to analyze their magnetic behavior. The specimens were prepared in cross section for TEM investigations by focused ion beam (FIB) lift-out specimen preparation technique (Quanta 200 3D DualBeam FIB). Two FIB protection layers consisting of Cr and Pt–C were added prior to the milling process. The TEM lamella thickness was in the range of 100–180 nm. The final thinning was done by a Technoorg-Linda GentleMill to achieve a sample thickness smaller than 30 nm. For the EMCD experiments a FEI TECNAI F20 equipped with a Gatan GIF Tridiem energy filter was used. The magnetic fingerprints of the samples were analyzed by detecting magnetically induced chiral electronic transitions by employing the EMCD technique [13]. This way the magnetic moments of the Fe atoms can be resolved with a spatial resolution of better than 2 nm [14].

3. Results and discussion

Based on previous work on Au_xCu_{1-x} combinatorial sputtering depositions [8,15], the Fe–Co magnetic layer was deposited on different crystallographic buffer layers, $Au_{50}Cu_{50}$ with $a=0.396$ nm and $Au_{30}Cu_{70}$ with $a=0.375$ nm. The Au–Cu layers grow epitaxially on MgO (100), as indicated by the diffraction patterns (Fig. 1) where only high intensity (001), (002), (003), (004) reflections are visible. It also allows an epitaxial growth of Fe–Co [7]. The epitaxial relation for the whole layer setup, including Fe–Co, was confirmed by texture measurements. Fe–Co (001)[110]//Au–Cu(001)[100]//MgO(001)[100].

Variable stoichiometry Fe_xCo_{1-x} alloy shows interesting deviation in terms of structural and magnetic properties on different buffers [16,17] or when a third element is added as a dopant (W) [18]. $Fe_{45}Co_{55}$ composition (in the most promising compositional range according to theoretical predictions [12]).

The films were studied both structurally and magnetically on both types of Au–Cu buffer layers mentioned above. In the case of $Fe_{45}Co_{55}$ composition, epitaxial growth on both Au–Cu buffers

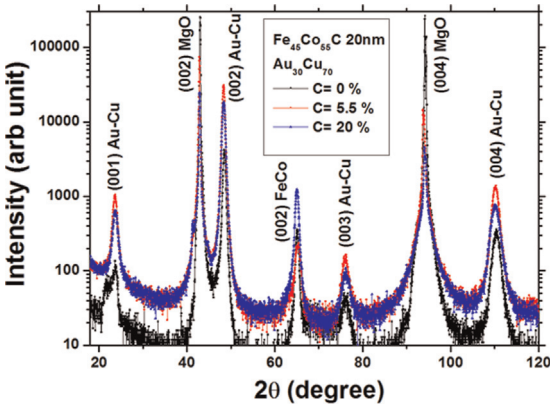

Fig. 1. X-ray diffraction (XRD) patterns of $Fe_{45}Co_{55}$–C thin films with variable C content and of pure FeCo magnetic layers, showing (00ℓ) reflections. Buffer layer was $Au_{30}Cu_{70}$.

with the c-axis along the film normal was observed as in XRD diffraction patterns, very strong intensity (00ℓ) peaks appear. Additionally the effect of Fe–Co magnetic layer thickness and carbon percentage on strain with the increase of the critical thickness for pseudomorphic growth were studied. From XRD a shift of the Fe–Co(002) reflection was observed, as it is shown in Fig. 2, with respect to the theoretically expected value at 65.31° (pdf 049-1567). In Fig. 2 representative XRD patterns of FeCo (C) films of 20 nm thickness are presented where the nominal carbon percentage in the alloy varied between 0% and 20%. The average calculated strain due to the lattice mismatch with the buffer layer and carbon addition is in the order of ~1%. For this film thickness, as it is shown in Fig. 2, no significant peak shift is observed in dependence on C content, when compared to binary $Fe_{45}Co_{55}$ on $Au_{30}Cu_{70}$ buffer.

This must be attributed to a tetragonal strain of the Fe–Co unit cell. XRD pole figure measurements confirmed such a tetragonal strain of the lattice of 1.03 in 10 nm thick $Fe_{45}Co_{55}$ films with 20% C. This value is already a strong indication that only a low amount of the added carbon contributes in straining the Fe–Co lattice. PLD prepared films with 2 at% C [7] showed higher shifts of the 002 reflection than our magnetron sputtered films. PLD is known as a further from equilibrium conditions method, due to the higher energies of the deposited species. Thus, for our sputtered films, it may be concluded that a large amount of the additional C does not enter in the Fe–Co phase lattice. The (nominally) 10 nm thick $Fe_{45}Co_{55}$ with 20% C film exhibited a c/a ratio of 1.03. From measurements of films with different thicknesses a relaxation of strain with increasing film thickness was observed as shown in Fig. 3. It is

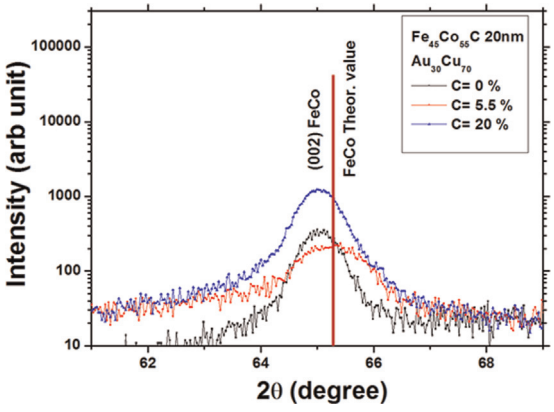

Fig. 2. X-rays diffraction showing the peak shift as a function of carbon percentage in 20 nm thick $Fe_{45}Co_{55}$ films.