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Giant magnetoresistance (GMR) and ferromagnetic properties of DC and pulse electrodeposited Cu–Co alloys

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ABSTRACT

Cu–Co ferromagnetic alloys occurring as granular films and exhibiting giant magnetoresistance (GMR) property have been synthesized using both DC and pulse electrodeposition techniques. The growth process of these electrodeposits comprising multiple granules of disparate morphology, magnetic features exhibits critical dependence on electrodeposition conditions. Using ferromagnetic resonance and magnetic hysteresis data, we have attempted a correlation between evolution of these electrodeposits and their ferromagnetic properties with special emphasis on GMR property.

1. Introduction

Fascinating success of Fe–Cr superlattice magnetic multilayers exhibiting giant magneto-resistance (GMR) properties and finding applications as high density recording media, magnetic sensors and reading head etc., is the driving force for the sustained interest in exploring newer materials, their magnetic characteristics [1–4]. Application of GMR systems in memory, sensor devices has stimulated more interest for augmenting scope for newer materials, properties and hence newer avenues for applications. These types of magnetic multilayer structures are normally prepared through electrodeposition methods. This conventional electrochemical route — as a strategic technique has gained much practical significance in preparing such magnetic systems in terms of ease of preparation, cost, and control on synthesis parameters [5–7]. Motivated by the success of this technique, in this investigation we discuss the results concerning the material, magnetic characteristics of Co–Cu films prepared using both DC and pulse electro deposition methods.

2. Experimental aspects

Cu–Co alloys were electrodeposited on indium tin oxide coated glass substrate with resistivity in the range of 8 to 12 $\Omega\,cm^{-2}$ using the electrolyte composition comprising 0.04 M $CuSO_4$, 0.3 M $CoSO_4$ and 0.05 M tri-sodium citrate as chelating agent at pH 4. The deposition was carried out in a three-electrode cell system under a galvanostatic control using Parstat 2273 model electrochemical analyzer. Co–Cu films deposited at different conditions served as the working electrode (cathode) while a platinum sheet was used as the counter-electrode (anode). Throughout the electrodeposition process a saturated calomel electrode (SCE) was used as the reference electrode. In this investigation, depositions of alloys were carried out using both direct current (DC) and pulse current (square wave from frequency range 10–100 Hz with 10% duty cycle) techniques with the average current density of 10 $mA\,cm^{-2}$ (for a duration of 100 s in all cases). The samples are labeled as given in Fig. 1. The chemical purity and integrity of these electrodeposits were ascertained in terms of powder XRD pattern consistent with the standard file (Fig. 1) corresponding to the chemical composition $Co_{0.48}Cu_{0.52}$ (ICDD#50-1452) and their least squares refined crystallographic cell parameters (Table 1). Corresponding XRD line-width and intensity data were used to estimate the crystallite size (using Scherrer formula) and relative percentage of crystallinity.

The magnetic hysteresis curves were recorded using a vibrating sample magnetometer (DMS-1600) at room temperature with applied magnetic field in the range of +20 to −20 kOe. Ferromagnetic resonance (FMR) measurements were carried out using a Bruker X-band (9.83 GHz) electron paramagnetic resonance spectrometer. FMR spectra were recorded with the plane of electrodeposited films both parallel and perpendicular direction wrt the magnetic field axis of the electromagnet of the EPR system.

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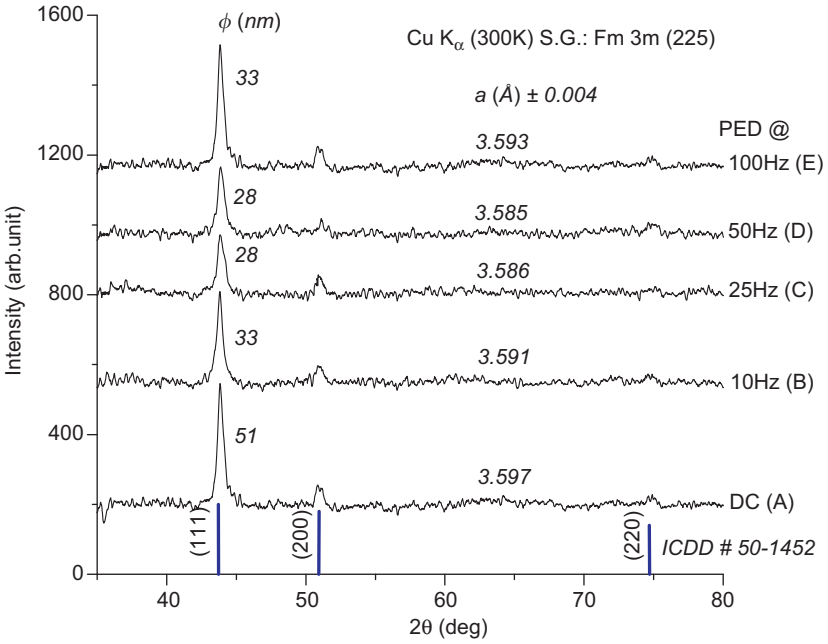

Fig. 1. XRD pattern of Cu–Co electrodeposits obtained under different conditions as labeled and in comparison with ICDD standard for $Cu_{0.52}Co_{0.48}$ composition. Crystallite size Φ in nm and fitted crystallographic cell parameter a in Å values are given under respective column heads.

Table 1
Materials and magnetic characteristics of Cu–Co electrodeposits.

Materials properties				Magnetic Properties				
Sample label: conditions; composition	Refined crystallographic cell parameter (a) $\delta = \pm 0.004$ Å	Crystallite size (Φ) nm	Relative % of crystallinity	VSM		FMR		MR=ΔR/$R_0 \times 100$%
				Coercivity (H_c) Oe	Remanence magnetization M_r X 10^{-3}	$K_{eff\|} \times 10^5$	$H_{an\|}$ (KOe)	
A: DC; $Cu_{0.94}Co_{0.06}$ Pulsed @	3.5974	50.5	94.3	60.4	3.85	6.75	1.65	1.77
B: 10 Hz; $Cu_{0.91}Co_{0.09}$	3.591	32.5	77.8	42.2	3.09	6.51	1.62	0.84
C: 25 Hz; $Cu_{0.90}Co_{0.10}$	3.5839	22.7	55.4	45.1	5.19	(i)6.68 (ii)1.48	(i)1.64 (ii)0.76	1.1
D: 50 Hz; $Cu_{0.88}Co_{0.12}$	3.5852	22.7	51.4	46.7	2.88	4.4	1.34	2.7
E: 100 Hz; $Cu_{0.80}Co_{0.20}$	3.5926	32.5	100	56.7	3.85	2.01	0.89	1.1

The magnetoresistance (MR) measurements were carried out using a Keithley Nanovoltmeter (6220) and power source (2182A) in four-probe method. During the MR measurements, direction of the magnetic field (0–6.3 kOe) was perpendicular to the applied current direction (transverse MR) which can be conveniently measured in current in plane configuration, commonly referred as CIP mode. That is in this CIP mode the current flow direction was parallel to the plane of the electrodeposited film(s). The percentage variation of MR values were calculated using the relation.

$$MR\ \% = \frac{R_H - R_0}{R_0} \times 100, \quad (1)$$

with R_H being the resistance with field and R_0 being the resistance without field of the given film. Furthermore, the growth of these electrodeposits and their morphologies were followed-up with the help of scanning electron microscope images (Hitachi S3000H), its EDAX facility for compositional analysis and also atomic force microscope (Agilent technologies 5500).

3. Results and discussion

3.1. Material properties of Cu–Co electrodeposits

The chemical integrity of the electrodeposits obtained using both DC and pulse deposited films yield nearly identical X-ray powder diffraction patterns consistent with the ICDD#50-1452 standard pattern corresponding to $Cu_{0.52}Co_{0.48}$ system (Fig. 1). Also the crystallographic unit cell parameter values of the different samples obtained using a standard least squares refinement procedure show good agreement with the standard values (Table 1) thereby confirming the chemical similarity and phase singularity of the electrodeposits being investigated. It is pertinent to note that in the $Cu_{1-x}Co_x$ alloy system, a wide variation (x=0.06 to 0.20) in the relative composition is possible and these samples are indexable under one kind of XRD pattern [8]. However, the crystallite size (Φ) determining the growth rate and relative percentage of crystallinity (%C) show profound dependence on the preparation conditions of these electrodeposits. The crystallite size is the highest for the film