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Structure and size effects in the magnetic anisotropy factors of Co-Ag nano granular films

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Abstract

Co-Ag granular films were prepared by electrodeposition method. The influence of the composition of thin films on their structure, granular size and magneic properties is presented. structural techniques (XRD and TEM) confirmed the heterogeneity of the deposits with a size distribution from superparamagnetic to ferromagnetic particles. Fcc-Ag and Fcc-Co were present in the coatings. The increase of the Co concentration in the film alloys leads to an increase in the size of magnetic granules and a simultaneous decrease of the distance between them. The in-plane easy magnetization axis is found for all samples. Using these results, we computed the first, second magnetic anisotropy factors. Values of the computed effective magnetic anisotropy factors higher than 0.53×10^6 erg/cm^3 have been found.

Keywords: Size effects, Magnetic properties, Nono Granular films, Perpendicular magnetic anisotropy

1. Introduction

Development and study of new magnetic films is one of the most important problems of modern nanotechnology and nanoelectronics. granular metallic solids have attracted much attention due to the discovery of giant magnetoresistance, enhanced coercivity (H_c), extraordinary Hall effect, giant magnetothermal properties, and other enhanced properties [1,2]. The magnetic granular films, in which magnetic (Co, Ni, Fe, Py) particles are embedded in the non-magnetic matrix (Ag, Cu, Au), display a giant magnetoresistance effect. Magnetic atoms located at the interfaces between the magnetic clusters and the non-magnetic matrix play an important role in magnetoresistance [1-4]. magnetic granular alloys have found a wide range of applications in nanoelectronics (e.g. non-volatile magnetic memory, sensors of magnitude the magnetic field and direction, magnetic resonance tomography) [5,6]. Among different magnetic granular films used for the development of effective magnetoresistive materials, granular Co-Ag thin films are characterized by high sensitivity and operating temperature reaching 200 ◦C[7]. Moreover, the percolation threshold concentration of the magnetic granules is larger for the Co-Ag system than for others, and thus a larger concentration of magnetic scattering centers would contribute to magnetoresistance [8,9]. Also, granular films based on Co and Ag exhibit high values of GMR and high sensitivity to the external magnetic field [10,11]. However, due to the complexity of granular Co-Ag microstructure, many of the physical properties of these films are yet to be clearly understood. The transport [12,13] and magnetic [1,8,14-16] properties of granular systems have been strongly related to the microstructure of the system, in particular, to the particle size, interparticle distance and the volume fraction of ferromagnetic element.

Most of the experimental work in these systems is focused on films having low ferromagnetic concentration, which usually consist of superparamagnetic particles, where wide variety of magnetic and transport behavior are observed as a result of dipolar interactions between the fine particles, since they are separated from each other by a large distance [17, 14]. Although there exist reports which present different characteristic features associated with the high ferromagnetic content, namely the presence of complex magnetotransport behavior, perpendicular magnetic anisotropy [1, 8,16,18, 19], and domain like structures larger than the particle size [14]. However, the nature of such effects is not yet fully understood. This is primarily due to the fact that a systematic study based on wide range of cobalt concentration and comprehensive investigation of such films using low temperature magnetotransport, dc-magnetization and magnetic domain imaging is lacking. Besides, there is still some controversy about how the magnetic and magnetoresistive properties depend on the structure and the phase of granular magnetic films [5].

As mentionea above, these nanogranular magnetic materials exhibits magnetic anisotropy that is, directional dependent magnetic behaviour. This anisotropy effect is very useful among the magneto-optic recording media. If

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the easy magnetization direction in the film is perpendicular to the plane, then it is called as perpendicular anisotropy. This perpendicular anisotropy is a potential candidate for many applications [20]. There is different possible mechanism which can account the perpendicular anisotropy effect like, shape anisotropy, magnetocrystalline anisotropy, the reduced symmetry of the structure at the interface and magnetostriction [20]. The overall anisotropy in the layers are influenced by the many factors [21, 22]: composition (degree of mixing of ferromagnetic and non-magnetic materials), temperature of measurement, film thickness and size of magnetic granules. Also, the fabrication method has a great influence on the magnetic properties. Customarily, electrochemical [9,22] and physical vapor deposition methods are in use (for example [5]). Electrodeposition has been revealed as an alternative to the physical deposition to grow high-quality materials because of its simplicity and lower cost [23].

In the ongoing miniaturization of magnetic data storage media, the superamaganetic limit, as well as interparticle interactions become critical and limiting issues. So, developing an elaborated understanding of the magnetic properties, especially the interparticle interactions are crucial to fully exploit such systems in the development of high performance magnetic materials and spin-dependent electronic devices [17]. Generally, the magnetization measurements on granular systems are used to get information about the interparticle interactions and the particle size .

Of relevant interest when using nanosized alloy is to optimize as to how to decrease the particle size of the material. However, the magnetic and magnetotransport properties of Co–Ag granular films strongly depend on the film thickness [1, 24-26]. Nevertheless, the size effects on magnetotransport of Co–Ag granular films have not widely studied. In this present work, attempts were made to deposit nanogranular Co-Ag with a range of component concentrations so as to study to study their magnetic anisotropy response to the magnetic field. Also, the influence of the magnetic granules size on magnetic properties of Co-Ag films with a range of component concentrations is revealed.

2. Experimental

2.1. Material

The electrodeposition of cobalt–silver coatings was performed from a freshly prepared 0.003 M $AgNO_3$, x M $CoCl_2$ (x=0.03, 0.09, 0.15), 3 M NaCl, 0.2 M H_3BO_3 solution, all chemicals being of analytical grade. The solution was freshly prepared with water first doubly distilled and then treated with a Millipore Milli Q system. Substrate is sputtered gold [Au(25 nm)/ Cr(5 nm)] on glass.

2.2. Preparation of Co-Ag films

In this work, electrodeposition of cobalt–silver films took place in a three-electrode cell. The sputtered gold substrates were used as cathode (working electrode). The anode (secondary electrode) was a parallelly placed platinum plat. Cobalt–silver films were coated under galvanostatic control at room temperature (25°C). The current density is 1 mA/cm^2. The pH value of the bath was kept constant at value of 2. Before and during electrodeposition process, the solution was de-aerated with argon. The film thickness for all experiments was chosen to be 200 nm.

2.3. Characterization

Film chemical compositions were determined by an energy dispersive X-ray spectroscopy (EDS). Crystal structure of Ag-Co thin films was studied by X-ray diffraction, using a Philips X'PERT-MRD diffractometer with Cu Kα radiation (λ=0.15418 A). Magnetic measurements were carried with a vibrating sample magnetometer (VSM) in the ±7000 Oe magnetic field range. The crystal structure of the samples was investigated via the transmission electron microscopy (TEM-125K). The images were studied by Anix Emica software.

3. Results and discussion

3.1. EDS spectra

EDS spectra was applied to investigate the compositions of the Co-Ag deposits. Figure 1, shows EDS spectra for Co-Ag films electrodeposited with a range of component concentrations. The results show that Co-Ag thin films electrodeposited from the solution composed of 0.003 M $AgNO_3$ and, 0.02 M, 0.09M and 0.15 M $CoCl_2$ coreespond to 19.27, 26.04 and 33.12 of Co (at.%) in alloy granolar films.

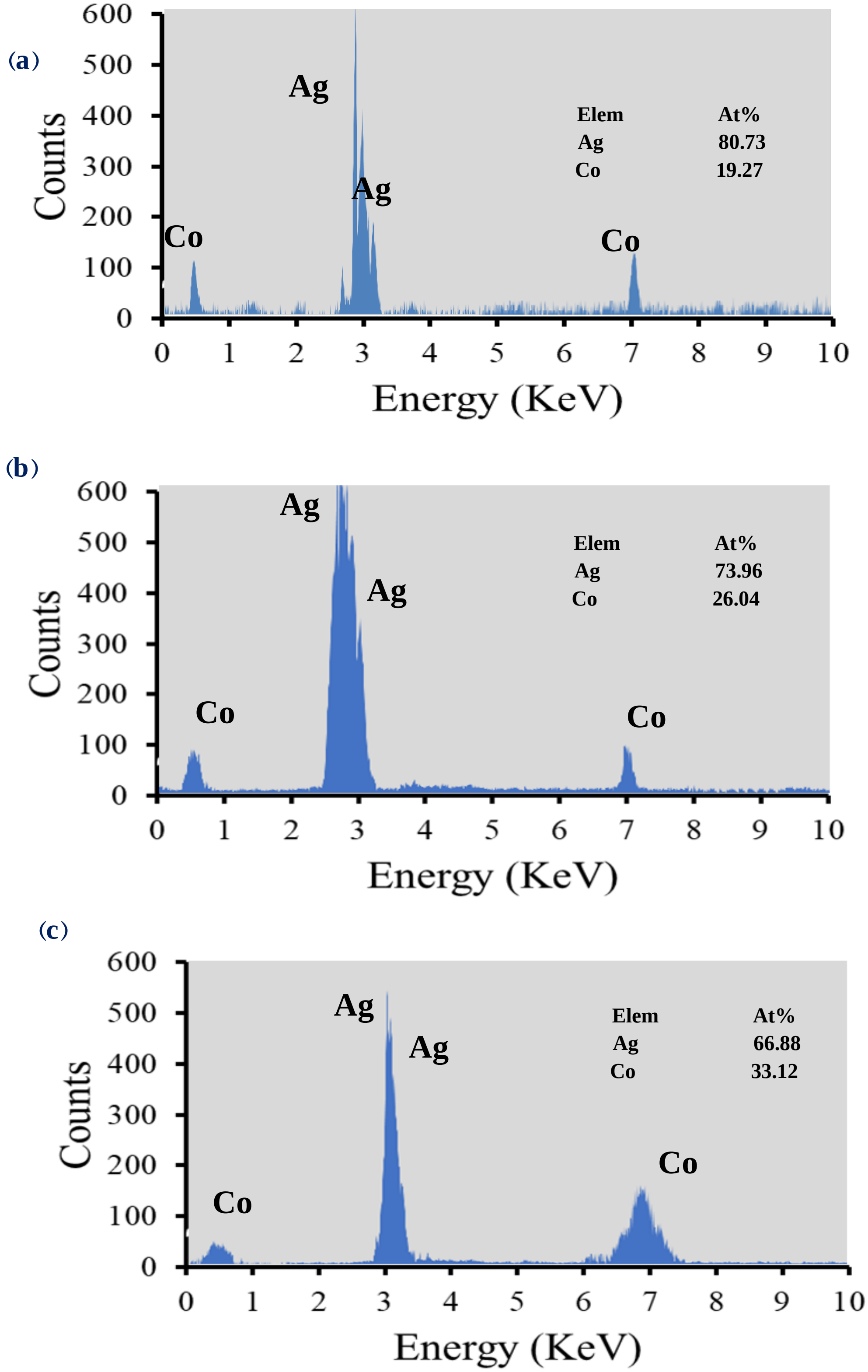

Fig. 1. EDS spectrum of Co-Ag thin films with different concentrations of Co obtained from the solution 0.003 M $AgNO_3$ and (a) 0.02 M, (b) 0.09M and (c) 0.15 M $CoCl_2$

3.2. XRD ananlysis

Fig. 2 shows the XRD diffractograms of Co-Ag film electrodeposited films with a range of component concentrations. These patterns exhibit diffraction peaks corresponding to the face-centered-cubic (fcc)-silver (JCPDS file No. 04–0783) and fcc-Co (JCPDS file No. 15–0806) structures. Same structure peaks for fcc-silver and fcc-Co have been observed by Zaman et al. for Co-Ag granular films electrodeposited at the temperature of 85 ºC [26-31]. However, no hcp-Co structure peak was observed as reported by Garcia-Torres et al., for Co-Ag granular films. [32]. The appearance of both Co and Ag peaks in XRD patterns shows that phase separation has occurred in the samples. This result is in consistent with that reported by Garcia-Torres et al. for Co-Ag films and is in contrast with that reported by Zaman et al. for Co-Ag films [26,27]. The existence of strong peak of Ag (111) shows that Ag served as the film matrix while Co particles are interspersed as small clusters through the film. As the XRD patterns show, increase in Co content in the samples causes Ag peaks broadening and Co peaks narrowing. Similar results have been observed by John et al. [18], Chandra et al. [32] and Garcia-Torres et al. [33] for Co-Ag films prepared by various deposition methods.

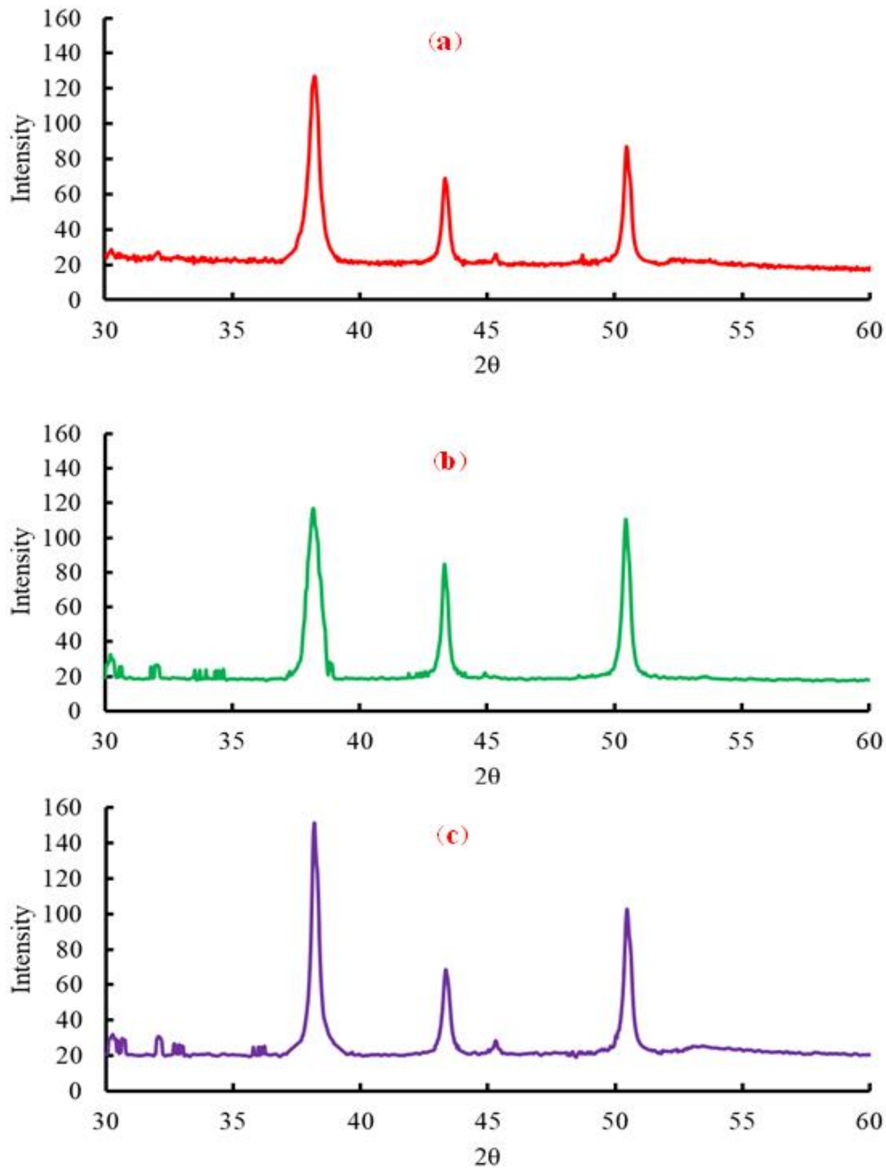

Fig. 2. X-ray diffraction result of Co-Ag thin films with different concentrations of Co (at.%): (a) 19.27, (b) 26.04 and (c) 33.12.

3.3. TEM study

The TEM studies and their analysis (by Anix Emica software) demonstrated that structural properties of produced films depend on their composition. Fig. 3 shows that Co granules are located in a matrix of Ag(Co). The size of the granules for samples with the concentration of 19.27 was L = 2–7 nm. Increasing the cobalt proportion in the alloy to 26.04 led to an increase in the size of the Co granules to L = 4–10 nm. For concentration of 33.12, the size of the Co granules increased to L = 5–23 nm. The increase of the Co concentration in the film alloys leads to an increase in the size of magnetic granules and a simultaneous decrease of the distance between them. Any further increase of the cobalt content in the film alloy leads to the formation of a continuous ferromagnetic film. According to Ref. [34, 35], larger granules show a single-domain ferromagnetic behavior whilst smaller granules are in the superparamagnetic state. Similar observations have been founded by Shpetnyi et al. [18] for Co-Ag films fabricated by the electron-beam co-evaporation and for Co-Cu films were obtained by the method of simultaneous evaporation [36,37] and by Stashkevich et al. for nanogranular (SiO_2)Co films and by de Moraes et al.for ZnSe–Co granularfilms [38].

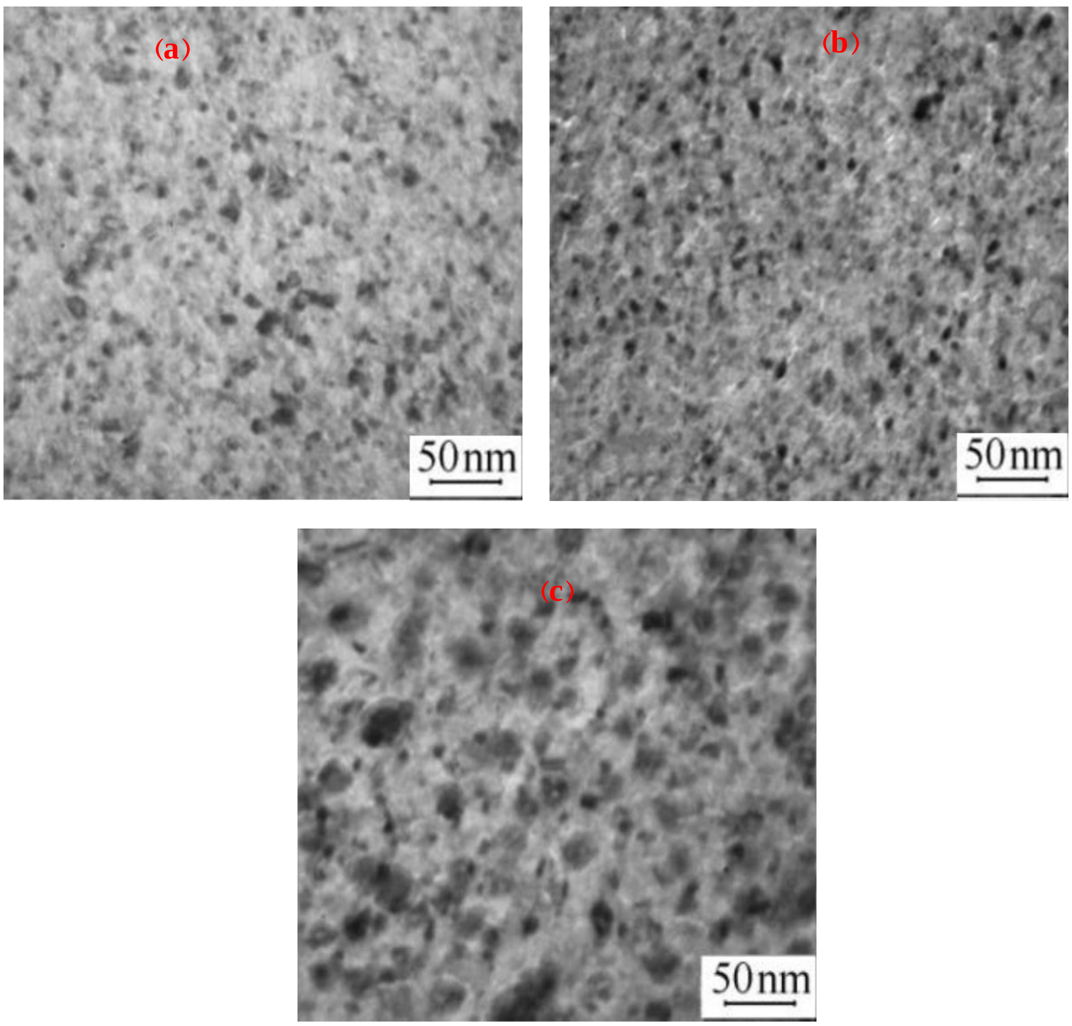

Fig. 3. TEM micrographs of a Co-Ag thin films with different concentrations of Co (at.%): (a) 19.27, (b) 26.04 and (c) 33.12.

3.4. VSM analysis

Fig. 4 demonstrate the magnetic hysteresis curves of granular films based on Co and Ag. The easy axis of the magnetization is assumed out of plane. As it can be seen from figure 4, the behavior of the magnetization curves depends on the composition of the film sample. As the concentration of cobalt in the alloy increases, the samples become magnetically anisotropic. This is confirmed by the different behavior of hysteresis loops for "in-plane" and "out of plane" geometries. The main reason for the appearance of anisotropy may be the magnetic interaction between the magnetic granules of cobalt. Another cause is surface anisotropy it could be suggested that the Co particles embedded in the Ag matrix are single-domain fine particles [39].

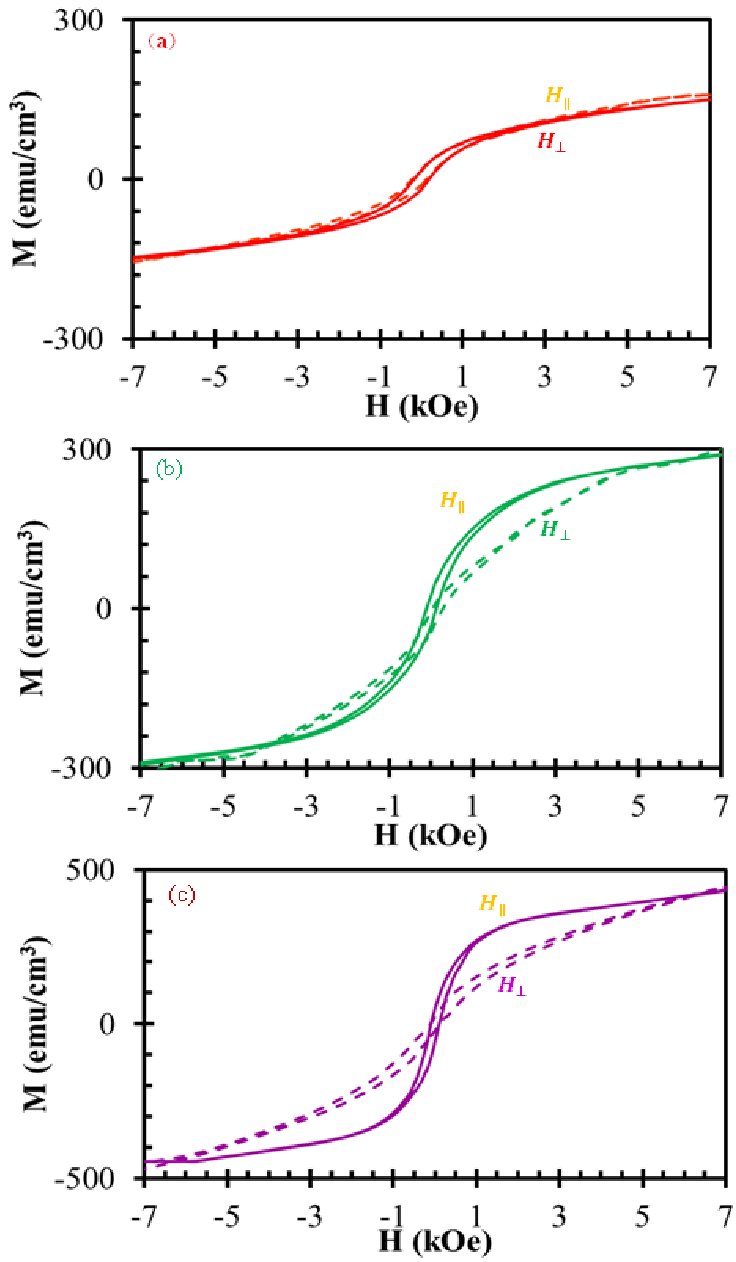

Fig. 4. Hysteresis loops of a Co-Ag thin films with different concentrations of Co (at.%): (a) 19.27, (b) 26.04 and (c) 33.12.

This perpendicular anisotropy has been observed in other granular films such as sputtered Ag-Co, Cu-Co, Co–Al–O and FeCoZr-CaF_2 [14, 18,19, 40-43]. In granulated materials the magnitude of the coercive force H_c depends on the size of magnetic granules [44, 45]. The enhancement of the coercivity in granular films with the concentration of cobalt is known to be due to an increase in size of the magnetic particles and an interaction of magnetic particles [40]. From the hysteresis loops and the volume of the samples we would be able to find out the magnetic anisotropy

field and therefore the anisotropy magnetic factors of the specimen. This is possible by accessing the saturation fields in both configurations (polar and planar) and using the saturation magnetization deduced from the magnetization curves and the volumes of the samples.
For a uniaxial anisotropy, the free magnetic energy of the sample can be written as [46]

$$E = K_0 + K_u \sin^2\theta + K_2 \sin^4\theta \quad (1)$$

Where θ is the angle between the magnetization vector M and the normal to the film plane in the spherical coordinate system, K_0 being the part of the magnetic energy independent of the orientation of the film. The magnetic anisotropy constant K_u may be written as:

$$K_u = K_1 - 2\pi(N_\perp - N_\parallel)M \quad (2)$$

$N_\perp$ and $N_\parallel$ being the effective demagnetizing factors, respectively perpendicular and parallel to the surface of the film.
For a thin film, the hard axis is parallel to [001], the easy axis being in the film plane, thus

$$N_\perp = 4\pi \quad \text{and} \quad N_\parallel = 0 \quad (3)$$

On the other hand, the shape energy may be written as

$$E_D = \frac{1}{2} M_s^2 (N_{xx} \sin^2\theta \cos^2\varphi + N_{yy} \sin^2\theta \sin^2\varphi + N_{zz} \cos^2\theta) \quad (4)$$

Besides, for a thin film with a uniaxial anisotropy, (1) may be written as

$$E = -\vec{M}.\vec{H} + 2\pi M_s^2 \cos^2\theta + K_1 \sin^2\theta + K_2 \sin^4\theta \quad (5)$$

Here, the first term arises from the Zeeman effect, the second term refers to the shape magnetic anisotropy (due to the demagnetizing field), and the third and fourth terms relay to the magneto crystalline anisotropy (due to the crystalline structure of the specimen).
The minimization of the energy ($\frac{\partial E}{\partial \theta} = 0, \frac{\partial^2 E}{\partial \theta^2} = \geq 0$) provides the saturation field H_{sat} and the anisotropy constants K_1 and K_2. When the applied field is perpendicular to the film plane $H_\parallel$[001] (Fig. 5), we get

$$H_{sat} = -2\frac{|K_{1eff}|}{M_s} \quad (6)$$

When the field is applied in the film plane ($H_\perp$ [001]) (Fig. 5), we get

$$H_{sat} = 2\frac{|K_{1eff}| + 2K_2}{M_s} \quad (7)$$

where $K_{1eff} = K_1 - 2\pi M_s^2$.

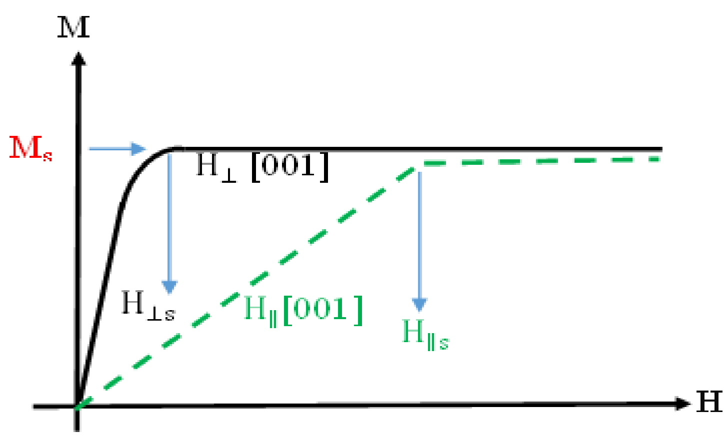

Fig. 5. Schematic M(H) curves displaying anisotropy field H_s, easy and hard axes.

Knowing H_{sat} from the hysteresis curves (as the figure 5) for the polar and the longitudinal configurations, and M_s values for each sample, the magnetic anisotropy constants are computed.
Fig. 6 display the evolution of K_{1eff} and K_2 versus cobalt content.We notice that K_{1eff} is always negative and K_2 is positive. The fact that K_{1eff} is negative infers that the easy axis lies in the plane of the film for all samples. Moreover, K_{1eff} values range from 0.53×10^6 to 8.5×10^6 erg/cm^3 when the cobalt content ranges from 19.27 to 33.12 at%. The deduced values of K, indicate that the perpendicular anisotropy is related to the volume fraction and hence to interparticle interactions. Its magnitude is minimum for samples with low values of Co concentration and becomes larger at larger Co concentration.
Similar K_{1eff} values have been reported by Chen et al. and Xiao et al. in their sputtered Co-Ag film [18, 19]. Kharmouche obtained K_{1eff} values of greater than 1.14×10^6 erg/cm^3 [47] for a Co-Cr films. In addition to being calculated with this method, magnetic anisotropy values are also calculated with Brillouin light scattering technique

[38]. The study based on the magnetization curves provides K_1 and K_2 constants; whereas the study based on the Brillouin light scattering technique provides the K_{ueff} ($K_{ueff} = \frac{1}{2} M_s H_a$, $H_{sat} = 2\frac{|K_{1eff}|+2K_2}{M_s}$), constant. The difference between these methods is due to the fact that the magnetization curves technique provides a global (average) value over the whole volume of the sample, whereas the Brillouin light scattering technique method is based on a local measurement, the area where the laser interacts with the matter.

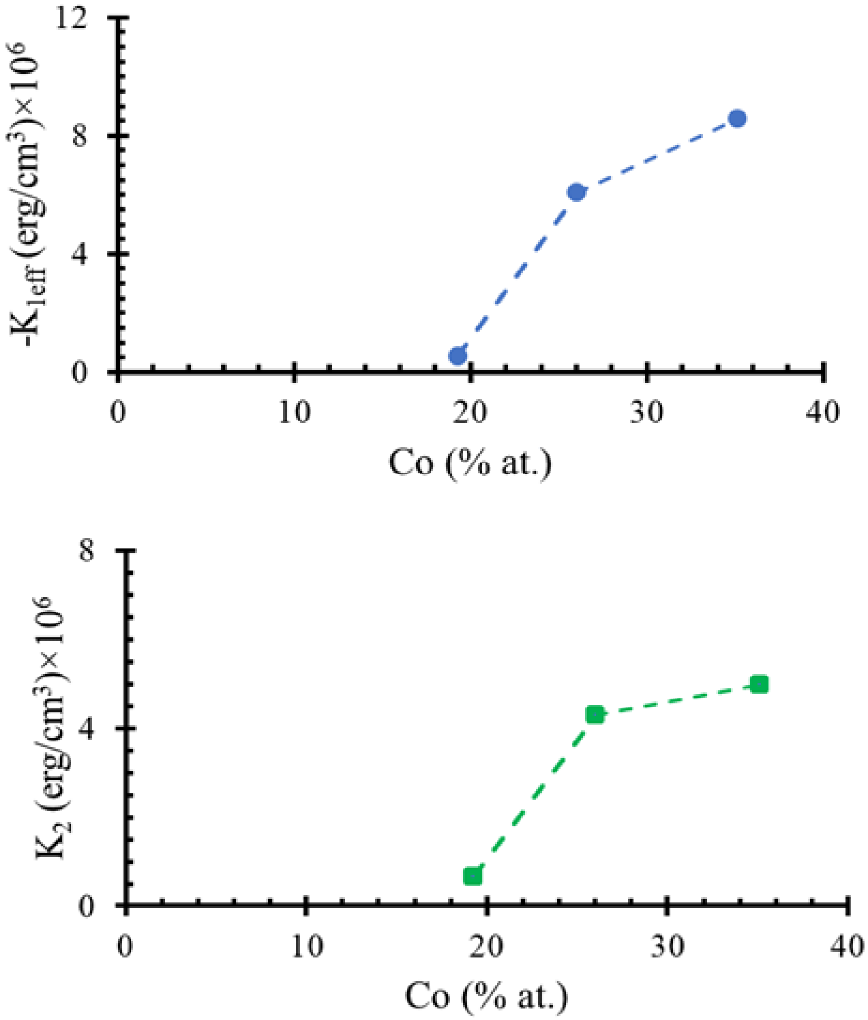

Fig. 6. Anisotropy constants K_{1eff} and K_2 versus cobalt content.

4. Conclusion

We have investigated the structural and magnetic properties of Co-Ag heterogeneous films prepared by electrodeposition. Fcc-Ag and and fcc-Co were present in the coatings. increase of the cobalt content in the films leads to broad of the cobalt peaks. With the increase of Co concentration indicates the increase in magnetic interactions with the increase in particle size. Magnetocrystalline anisotropy factors for coatings have been computed using experimental results performed by means of VSM. results show that the easy magnetization axis lies in the plane of the films for all samples. The study based on the magnetization curves provides anisotropy constants. K_{1eff} values range from 0.23×10^6 to 0.57×10^6 erg/cm^3 when the cobalt content ranges from 19.27 to 33.12 at%.

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Conflicts of Interest

The author declares no conflict of interest.

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