

Research Article

Structural Characteristics and Magnetic Properties of Al₂O₃ Matrix-Based Co-Cermet Nanogranular Films

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Magnetic micro- and nanogranular materials prepared by different methods have been used widely in studies of magneto-optical response. However, among them there seems to be nothing about magnetic nanogranular thin films prepared by a *rf* cosputtering technique for both metals and insulators till now. This paper presented and discussed preparation, structural characteristics, and magnetic properties of alumina (Al₂O₃) matrix-based granular Co-cermet thin films deposited by means of the cosputtering technique for both Co and Al₂O₃. By varying the ferromagnetic (Co) atomic fraction, *x*, from 0.04 to 0.63, several dominant features of deposition for these thin films were shown. Structural characteristics by X-ray diffraction confirmed a cermet-type structure for these films. Furthermore, magnetic behaviours presented a transition from paramagnetic- to superparamagnetic- and then to ferromagnetic-like properties, indicating agglomeration and growth following Co components of Co clusters or nanoparticles. These results show a typical granular Co-cermet feature for the Co-Al₂O₃ thin films prepared, in which Co magnetic nanogranules are dispersed in a ceramic matrix. Such nanomaterials can be applied suitably for our investigations in future on the magneto-optical responses of spinplasmonics.

1. Introduction

Cermet, or ceramic metal, is a kind of composite material in which many component phases coexist. Normally, it includes metallic and ceramic phases that are separated and mixed. Al₂O₃ matrix-based granular Co-cermet can form by means of immersing segregated Co nanoparticles into an alumina matrix, Al₂O₃. In other words, this is a kind of nanocomposite consisting of metallic nanoparticles embedded in an insulating (or even metallic) matrix. Alumina-based ceramics (Al₂O₃) possess excellent physical and chemical properties, as well as good mechanical resistance and thermal stability. Nanogranular composite materials display a particularly rich variety of interesting physical properties, including conducting, optical, and magnetic [1].

The materials of this type are quite suitable for different aims of research. For example, formerly, Au tiny particles dispersed into a dielectric medium have been considered as optical media to study on plasmonics [2]. In the field of magnetic materials, the Co-cermet thin film is also called magnetic granular film (MGF) because of the ferromagnetic and nanogranular structural characteristics of Co. Such materials are estimated to open a new opportunity for developing novel soft magnetic materials [3]. In electronics and spintronics, Al₂O₃ matrix-based Co-cermet MGFs, such as Co-Al₂O₃ films, have been also known to have the spin-dependent magnetic tunneling effect [4, 5] and a very typical feature by the Coulomb blockade phenomenon for the intergranules magnetic tunnel [6]. Furthermore, the MGF material has been recently reported to still exhibit rectification [7]. In

addition, MGFs are predicted; they can also be appropriate for studying magnetic field-dependent super- k dielectric granular and spin-caloritronic materials.

Recently, interesting phenomena in nanomagnetic materials have been observed, such as spin-plasmonic propagation in a system including gold-plated ferromagnetic microparticles [8], excitation of surface plasmons on magnetoactive materials [9], or enhancement of surface plasmon of metal nanocomposite films [10]. However, among the materials structured from metallic nanoparticles dispersing in a dielectric matrix for studying on spinplasmonics or other nanomagneto-optical responses, those prepared using radio-frequency (rf , 13.56 MHz) cosputtering techniques for both ferromagnetic metals and dielectric isolators seem to be unreported as yet. For example, CoFe-Al₂O₃ granular films, which are similar to our Co-Al₂O₃ granular films, were coevaporated using e -beam evaporation for studies of IR reflectance and magnetorefractive effects [11]. Recently, some kinds of nanomaterials for the magnetic photonics or plasmonic resonance studies were fabricated using different techniques, such as sol-gel method for magnetic photonic crystals based on Fe₃O₄ nanoparticles [12], spray pyrolysis technique for Au nanoclusters embedded in titania (TiO₂) matrix [13], and thermal evaporation method and then annealed at high temperature for Au nanoparticles [14], and so forth. Based on those studies, we were motivated to prepare Al₂O₃ matrix-based Co-cermet MGF materials using rf cosputtering technique. These cermet films are believed to be able to excellently satisfy to study plasmonic resonance phenomena, and especially the spin-dependent optical response as an approach to the field of spinplasmonics.

In this paper, we report on the preparation and analysis of several typical characteristics of deposition and the structural and magnetic properties of Co-Al₂O₃ cermet films as a function of Co content.

2. Experimental Details

The samples of Co _{x} (Al₂O₃)_{1- x} cermet films were deposited onto 200 μ m thick lamellar glass substrates via rf cosputtering technique (Alcatel SCM 400) at ambient temperature. The background vacuum was about 10⁻⁹ bar and argon gas of 5N purity was used as the sputter gas kept constant at a pressure of $\sim 5 \times 10^{-6}$ bar during the sputtering time. A 4N-alumina disk of 7.5 cm diameter with 4N-Co pieces pasted on its surface was used as a target, and a sputtering power of 300 W was applied. The target-to-substrate distance is 7.5 cm, and the rf power density was evaluated to be about 7 W/cm². The Co and Al₂O₃ depositing rates were 0.5 and 0.2 $\text{\AA}/\text{s}$, respectively, and an average depositing rate for Co-Al₂O₃ was determined to be about 0.3 $\text{\AA}/\text{s}$. The film thickness was varied from ~ 90 to 100 nm and was measured using an Alpha-Step IQ by KLA Tencor. Co contents with $x = 0.04$ –0.63, which correspond with 4–63 Co at%, were determined via energy dispersive X-ray spectroscopy (EDS), and the morphology of the film surfaces was observed via scanning electron microscopy (SEM) by using JEOL JSM-541. In particular cases, the surface morphology of several selected samples was also observed

using an atomic force microscopy (AFM) model FlexAFM by Nanosurf. The crystalline structural characteristics of the Co-Al₂O₃ cermet films were investigated via X-ray diffraction (XRD) by using a Philips X'Pert PRO with Cu-K α radiation ($\lambda = 1.5406 \text{\AA}$) for two theta values from 10° to 80°. Measurements of the magnetic properties were carried out at room temperature by using a magnetometer MicroMag 2900/3900, essentially at magnetic fields parallel to the film plane and up to 14 kOe.

3. Results and Discussion

The Co-Al₂O₃ thin films were deposited via cosputtering from a combined target including different Co (A_{Co}) and Al₂O₃ ($A_{\text{Al}_2\text{O}_3}$) areas that have different sputtering rates, such as 190 and 40 $\text{\AA}/\text{s}$, respectively [15]. Other factors still affect the deposition processes of the thin films, such as reflection, resputtering, and desorption, and the sputtering yield of Co is higher than that of Al₂O₃, which corresponds to 1.4 versus 0.4 atoms/ion, respectively [15]. Therefore, the Co content in the thin films is not identical to that in the target. Figure 1 shows a relationship between the Co content x determined via EDS in the thin films (in at%) and Co : Al₂O₃ area ratio in target; $\sigma = A_{\text{Co}}/A_{\text{Al}_2\text{O}_3}$ (in area percent). The experimental data fitted very well to a fourth-order law, which is defined approximately by the equation $x \approx 5 \times 10^{-7} \sigma^4 - 7.5 \times 10^{-6} \sigma^3 + 2 \times 10^{-3} \sigma^2 + 0.40\sigma$. The result shows also that, for $x \leq 0.4$, the Co atom amount in the films versus Co area in the target can obey very well a first-order relation; that is, $x \approx 0.40\sigma$. This means the Co amount increased more proportionally with a trend of an uncompleted fourth-order polynomial function, or almost exponentially with a law of $x \sim \{\exp[(0.02\sigma + 0.5)] - 1\}$. The fourth-order relation between A_S/A_T areal ratio of substrate electrode (A_S) and target electrode (A_T) and the V_T/V_S potential ratio of the potential on the target (cathode) sheath (V_T) and substrate sheath (V_S), that is, $V_T/V_S = (A_S/A_T)^4$ [16], mainly governs this behavior of growth rate of Co atom amount in the film. The increase in Co-chip area stuck on the Al₂O₃ target, or the increase of the σ ratio, is also synonym with the strong enhancement of an effective potential fallen on cathode, that is, V_T sheath potential, in the sputtering process for Co because the sputtering yield of Co is very high compared to that of Al₂O₃. Thus, the ion bombardment of the substrate for resputtering and desorption of Co atoms, which had been absorbed on substrate, was minimized.

Figure 2 shows a typical XRD pattern selected for the sample with $x = 0.27$. The XRD pattern presents a superposition of the signal caused by the Co-Al₂O₃ thin film (blue full line), called diffractogram, and the contribution of the glass substrate (illustrated by the dotted line), or from other background spectra/signals. The figure shows that the Al₂O₃ matrix dominantly had a typical amorphous-like phase or a low crystalline structure with a large hump at around $2\theta \sim 25^\circ$ and a small one at $\sim 43^\circ$. Three strongest standard peaks for bulk α -Al₂O₃ crystal corresponding to (104), (113), and (116) Miller indexes are indicated by orange lines in Figure 2. These standard peaks pertain to the rhombohedral

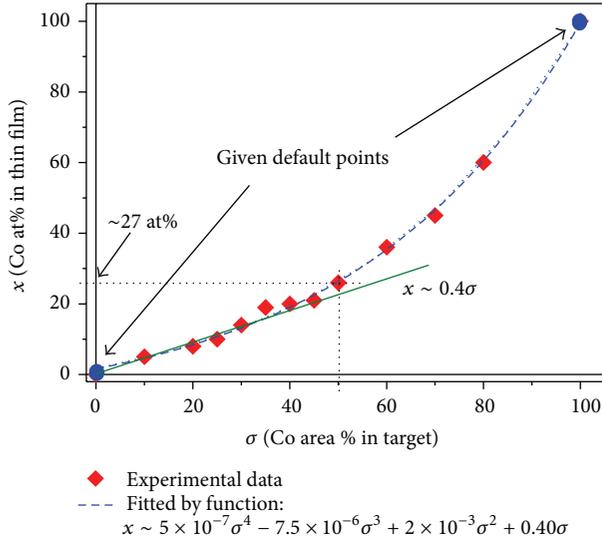


FIGURE 1: Co content of Co-Al₂O₃ thin films, x , as a function of Co : Al₂O₃ area ratio in the target, σ . Points in zero (origin) and 100% coordinates are as given default data.

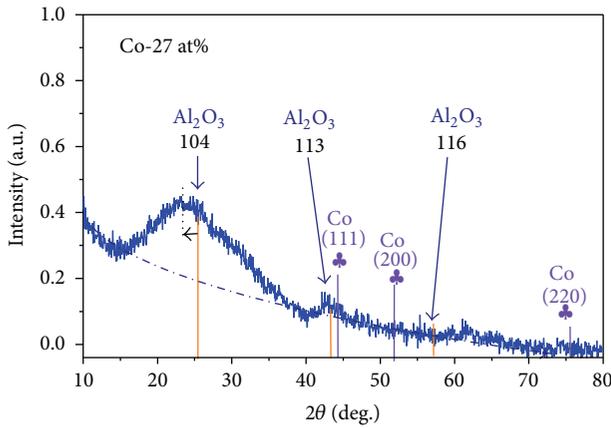


FIGURE 2: XRD pattern of Co _{x} (Al₂O₃)_{1- x} thin film typically selected with $x = 0.27$. Dotted line presents background signal from the glass substrate. Violet and orange lines show standard XRD lines for Co and Al₂O₃, respectively. The shift from the Al₂O₃ (104) line of the big hump is pointed out.

system of the α -Al₂O₃ for structure as indicated in the JCPDS-International Centre for Diffraction Data Card number 46-1212. However, the coincidence of these peaks with the amorphous-like humps of Al₂O₃ shows that the Al₂O₃ matrix of the samples varied from a rhombohedral crystal structure into a disordered variant. Alternatively, the large hump also presents a type of the so-called fine crystalline structure. When Scherrer's formula is used

$$D = \frac{0.94\lambda}{\beta \cos \theta}, \quad (1)$$

where D is the average crystallite size, λ is the X-ray wavelength of CuK α radiation, β is the full width of half maximum (FWHM) of the diffraction peaks, and θ is the diffraction

angle, the mean size of the Al₂O₃ fine crystallites was found to be about 1 nm or ~ 10 Å. Given below the ion radii of Al³⁺ and O²⁻ in an Al₂O₃ molecular, each fine crystallite includes only about 10 molecules. Hence, the Al₂O₃ matrix can be considered as a mixture of fine crystallite and amorphous phases.

On the other hand, a rather clear shift was observed for the Al₂O₃ humps from standard peak positions towards the left side, which corresponds to a lower 2θ angle. The results seem to indicate a tensile stress in the Al₂O₃ matrix. This lattice deformation or disorder in the rhombohedral structure may be due to Co atoms or clusters dispersing into the matrix because the ion radius of Co²⁺, ~ 0.075 Å, is larger than that of Al³⁺, ~ 0.054 Å, but smaller than that of O²⁻, ~ 0.140 Å (in terms of atomic radius, Co ~ 0.152 Å and Al ~ 0.118 Å and O ~ 0.048 Å) [17]. In other words, the shift of the Al₂O₃ humps is indicative of the existence of Co atoms in the rhombohedral lattice of Al₂O₃ or Co clusters in the Al₂O₃ matrix. The indication of the shift for matrix diffraction peaks has been used as an experimental evidence supporting, for example, a continuous solid solution of interstitial intermetallics [18], or an insertion of atoms of a certain metal (e.g., Co) into a matrix metal (e.g., Cu) by means of ion implantation [19]. A similar trend of the shift of the MgO Bragg diffraction peaks (i.e., matrix phase) compared to the peak positions in the pure MgO XRD patterns when adding another metals (e.g., Al, Cr, Ti, and Zr) to the MgO matrix phase has been also observed [20].

As seen in Figure 2, almost no clear signature of the Co peaks was observed, except for a very small hump at around $2\theta \sim 44^\circ$. The three strongest standard peaks of Co-fcc crystal corresponded to (111), (200), and (220) Miller indexes, as indicated by violet lines with rosette figures in Figure 2 (after JCPDS-ICDD Card number 01-089-7093), where the Co-fcc (111) line coincides with the small hump right beside the Al₂O₃ (113) hump. There are two main arguments for this absence of Co peaks. First, X-ray fluorescent radiation from Co was the one used to be excited in the sample by the Cu-K α radiation of the used X-ray source. The background of an XRD pattern is composed of many variant factors, in which there are fluorescent radiation and small particles [21]. Co is a ferromagnetic element that is known to cause the strongest X-ray fluorescent for Cu-K α radiations. Thus, the fluorescent effect may cause the weakest Co diffraction peaks so that the peaks become almost invisible in relation to the background in the XRD spectra [22]. Second, this reason can occur simultaneously with the first reason but can also be more dominant: the shift of the Al₂O₃ humps as mentioned above, the size of Co clusters is very small, and/or the Co atoms are broadly dispersed into the Al₂O₃ matrix. Such similar feature of no diffraction peaks in the XRD has been also observed for amorphous or nanocrystalline CoFe clusters in a SiO₂ silica matrix, with cluster sizes of about 1 nm–4 nm observed and estimated based on their TEM images [23]. The Co particle sizes of the Co-cermet films are hard to evaluate from the XRD data by using Scherrer's formula because of the very weak X-ray reflection of Co. However, a raw estimation can be carried out from the superparamagnetic (SPM) behavior of the Co-Al₂O₃ thin

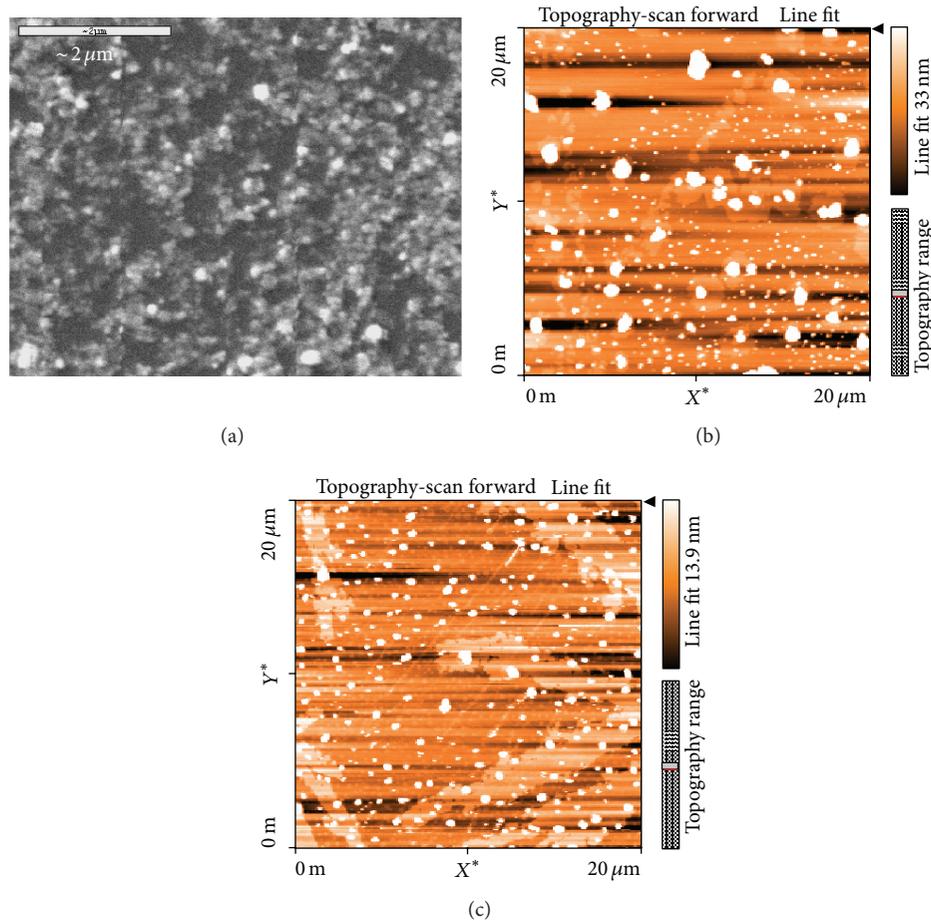


FIGURE 3: (a) Surface SEM image of as-deposited $\text{Co-Al}_2\text{O}_3$ thin film with $x = 0.27$. (b) AFM image $20 \mu\text{m} \times 20 \mu\text{m}$ for the surface of the as-deposited $\text{Co}_{0.27}\text{-Al}_2\text{O}_3$ film and (c) of the film after annealing at 250°C for 1 hour.

films, which we will discuss below. The result shows the same sizes. A “redundant” amount of about 5% volume of Co atoms that will not participate in the formation of Co particles/clusters is mentioned. These “redundant” Co atoms in the amorphous state created the shift in the XRD peaks. A similar shift behavior of the diffraction peaks caused by the doping of Co atoms into TiO_2 nanoparticles has been pointed out [24]. For metal-insulator systems, such as $\text{Co-Al}_2\text{O}_3$, separation and isolation of metallic clusters or particles in the oxide matrix can occur easily, because the large difference between the surface energies of Co and Al_2O_3 leads to an isotropic three-dimensional growth of Co [25, 26]. However, it is difficult to determine the size of Co particles by using Scherrer’s formula (1) in this situation due to no clear Co XRD peaks, as seen. But it can be estimated by means of magnetization behaviors in case of superparamagnetism, as seen hereinafter.

Figure 3(a) shows a rather rough surface topographic morphology of the as-deposited $\text{Co-Al}_2\text{O}_3$ thin film selected at $x = 0.27$ as an example. Figures 3(b) and 3(c) present the surface AFM images (size: $20 \mu\text{m} \times 20 \mu\text{m}$) captured in the amplitude type for the same sample, which correspond to the

as-deposited and after annealing at 250°C for 1 h, respectively. The rough surface is caused by the low atomic mobility due to deposition at room temperature. The as-deposited thin films seem to be improved after annealing. As seen from the line fit indicated in Figures 3(b) and 3(c), the height amplitude range was reduced from 33 nm to 14 nm.

Dominant features in the change in the magnetic phases from paramagnetic- to superparamagnetic- and ferromagnetic-like behaviors with increasing Co content, which correspond to $x < 0.12$, $0.12 \leq x < 0.37$, and $x \geq 0.37$, respectively, were observed for all $\text{Co}_x(\text{Al}_2\text{O}_3)_{1-x}$ granular thin films prepared. For systems with Co granules immersed in nonmagnetic matrixes, the ferromagnetic behavior begins to appear at room temperature with a concentration at ~ 35 Co at% [27]. Figure 4(a) presents only several typical normalized $M(H)$ magnetization curves selected for the $\text{Co}_x\text{Al}_2\text{O}_3$ thin films with $x = 0.06, 0.16, 0.27$, and 0.49 , which are representative of the three regions of magnetic phases. A paramagnetic-like behavior at room temperature with an unsaturated sign of up to about 1.4 T (Tesla) was observed, and no hysteresis was observed in the case of $x = 0.06$. This result can imply that the matrix of the film was sprinkled with

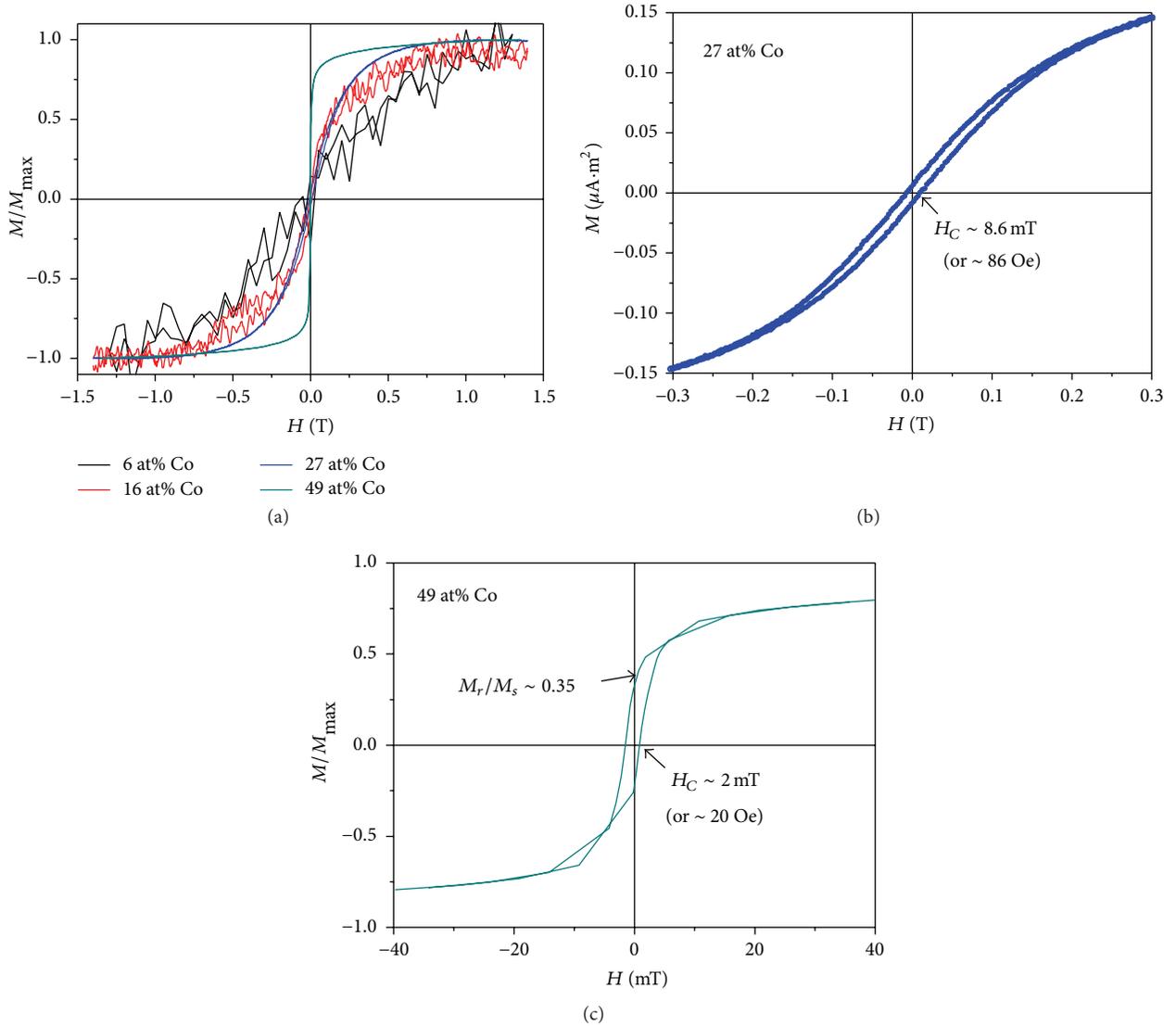


FIGURE 4: Room temperature magnetization curves $M(H)$ of $\text{Co}_x(\text{Al}_2\text{O}_3)_{1-x}$ thin films with (a) $x = 0.06, 0.16, 0.27$, and 0.49 . (b) $M(H)$ curve extracted partly in a range ± 0.3 T for the case of Co-27 at% thin film presents a narrow hysteresis loop with $H_C \sim 8.6$ mT and very small remanence. (c) A small hysteresis loop with $H_C \sim 2$ mT and ratio $(M_r/M_s) \sim 0.35$ for the case of Co-49 at% thin film.

Co atoms or with very small atomic agglomerates. In fact, because Co clusters formed only part of the Co material and the remainder is dispersed in non- or weak-magnetic states buried within the Al_2O_3 matrix, the true concentration of the magnetic clusters is somewhat lower than the one determined from EDS [28]. For Co- Al_2O_3 films, Co atoms are atomically dispersed in the Al_2O_3 matrix and, as mentioned above, can hold an amount up to 5 vol.% [29]. The $M(H)$ curve in this case can reflect a dominant feature of the spin glass state because Co atoms seem to behave individually. Moreover, a drastic competition between magnetostatic energy, thermal fluctuation, and RKKY-type interaction is reality. For $x = 0.16$ and 0.27 , the $M(H)$ curves expressed in Figure 4(a) show typical room temperature SPM behaviors. This type expresses a granular system of almost noninteractive single domain spherical ferromagnetic particles. For uniform particle size d ,

the magnetization curve $M(H)$ is described by the Langevin function:

$$M(H) = M_S L(\alpha) = M_S \left(\coth \alpha - \frac{1}{\alpha} \right), \quad (2)$$

with $\alpha = \frac{M_S V H}{k_B T}$,

where M_S is saturation magnetization of ferromagnetic particles with volume $V = (4\pi/3)(d/2)^3$, k_B is Boltzmann constant, and T is temperature. In fact, there are size distributions in SPM particles. Thus, the total magnetization is better described as a weighted sum of Langevin functions under the form of (2) [30]. However, a small hysteresis behavior of SPM status, with coercivity up to $H_C \sim 8.6$ mT, but with a very low remanence, $M_r < 5\%$, was observed for the case

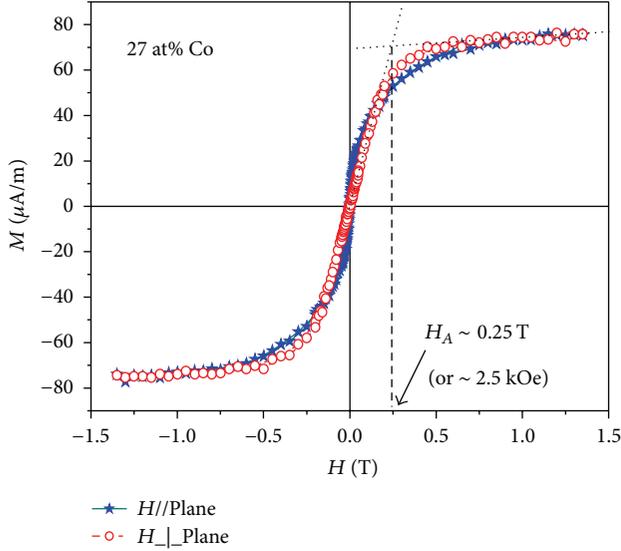


FIGURE 5: Magnetization curves $M(H)$ measured in both configurations with H parallel and perpendicular to the plane of $\text{Co}_{0.27}(\text{Al}_2\text{O}_3)_{0.73}$ thin film. An anisotropic field H_A of about 0.25 T (~ 2.5 kOe) for the perpendicular configuration can be determined.

of $x = 0.27$, as seen in Figure 4(b). This result proves that there can be a weak interaction between Co particles, such as a dipolar intergranular interaction that resulted in anisotropy in the system [31]. It can be also due to a slight shape and/or crystal anisotropy in these particles [32]. Figure 5 presents a typical investigation of hysteresis loops for the case of $x = 0.27$ with both configurations of H parallel and perpendicular to the plane of this sample. As seen, two magnetization curves are almost identical. This shows a rather isotropic behaviour for the Co particles. However, a linearity at low H region (< 0.25 T) of the magnetization curve measured in the perpendicular configuration presents a little in-plane anisotropy with the anisotropic field H_A of about 0.25 T (~ 2.5 kOe). In other words, the Co particles have an somewhat oblate spherical form.

With the above features, Co particle sizes can be assigned to the range of about 1 nm–5 nm in diameter [32]. Given formula $T_B = VK_{\text{Co}}/25k_B$ below, where the magnetization will be stable, for constant size spherical particles, the blocking temperatures T_B of the SPM Co-cermet systems can be roughly established. For $k_B = 1.381 \times 10^{-16}$ erg·K $^{-1}$ and $K_{\text{Co}} = 4.1 \times 10^6$ erg/cm 3 at room temperature for Co [33] for particles that are distributed from 1 nm–5 nm in particle size, T_B will be in a broad range from 0.6 K to 80 K. For instance, for $d = 3$ nm, $T_B \sim 17$ K. Such systems will correspond to the SPM relaxation time $\tau < 10^5$ s [32]. Using the ion radius as has been given for Co^{2+} , the number of Co atoms in each particle in this range of size can be estimated to be about 250×10^3 to 35×10^6 . Several studies on systems of Co-based granular thin films have also confirmed these sizes via transmission electron micrograph (TEM) [29, 34] or scanning tunneling microscopy (STM) [35, 36], and via calculations from the Langevin fitting of SPM M - H data [29, 37]. Therefore, in

this case, a state called the spin cluster state and/or a Co-rich particle state of Co atoms in the Al_2O_3 matrix can also show up. The oscillation-type RKKY interaction between Co clusters can be brought into play, similar to the type of granular metallic alloys, such as Fe-Cr alloys [38].

When the Co content is high as 49 at%, a ferromagnetic type in the $M(H)$ curve showed a narrow hysteresis loop with $H_C \sim 2$ mT, and the ratio of remanent and saturation magnetization (M_r/M_S) was estimated to be almost ~ 0.35 , as inserted in Figure 4(c). These behaviors of the weak coercivity and the remanence near $0.5 M_S$ prove that the Co particles can still be in ferromagnetic nature at room temperature with an essential single domain form and are randomly dispersed. Both coercivity and ratio (M_r/M_S) = 0.5 (theoretically) are signature/evidence for a random dispersion of noninteracting single domain particles but in weakly blocked state of particle magnetization [1, 39]. For granular metal systems, there exists a so-called percolation threshold (x_p) of the volume fraction at which the first continuous metal particle is formed [1, 40]. This threshold is around 50%–60% for the Co particles in an amorphous Al-O matrix [1, 34]. For higher Co contents with $x = 0.49$ and below the percolation threshold, the Co particles are inclined to agglomerate into larger Co-rich regions or masses, in which each composed particle is still almost separated from the others but is coupled (or blocked) magnetostatically together in a ferromagnetic-type state, which is similar to real ferromagnetic entities. To some extent, the ferromagnetic-type property with a weak coercivity of such assemblies of nanoparticles can therefore be considered as a so-called “superferromagnetic” (SFM) state, as called at first by Bostanjoglo and Roehkel [41]. This way of looking at the SFM state is similar to the case of calling the SPM-type state. Thus, the magnetization process for such systems should occur in two steps: (i) in low-field region, about several tens of Oersted, orientation of magnetization of the large ferromagnetic masses and (ii) in higher-field region, orientation along the applied field of the disorder magnetization of the clusters/particles that are dispersed in the matrix and/or in the ferromagnetic masses. These two steps will correspond with two magnetization components that can be separated for each ferromagnetic and paramagnetic contribution [27].

When the particle concentration is further increased, the magnetic interparticle interactions become nonnegligible and one may find a crossover from single-particle blocking to collective freezing. When the Co content is high enough over the percolation threshold, such as for $x = 0.63$, the Co clusters/particles are impinged on each other or the infinite cluster/particle occupies almost the whole volume of the sample [31, 33], where the magnetic properties are close to the ones of bulk ferromagnetic Co films.

4. Conclusion

We have fabricated the Co- Al_2O_3 films by *rf* cosputtering onto glass substrates with a rather high *rf* power density. We found that at such, the sputtering condition, the relationship between the Co contents in the Co- Al_2O_3 films, and the Co area ratio in the target obeyed a quaternary polynomial

function, and these films hold dominant features of a typical nanocomposite cermet. The Co-Al₂O₃ films, which can be called as Al₂O₃ matrix-based Co-cermet nanogranular films, have shown that the amorphous Al₂O₃ ceramic matrixes were sprinkled with Co magnetic atoms, clusters, or nanoparticles, depending on Co contents. The important manifestations of a cermet-type nanogranular structure are the shift of XRD peaks from standard peaks for pure matrix phase, for example, Al₂O₃, due to internal microstress caused by Co atoms and/or clusters or particles, and magnetic behaviours among the paramagnetism pass superparamagnetism to weak ferromagnetism for magnetic phase, for example, Co, due to spin glass or cluster glass states caused by ferromagnetic superfine particles. Such cermet-type nanogranular films are suitable to study on spinplasmonics.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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