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ORIGINAL PAPER

Magnetic Domain Studies of Cobalt Nanostructures

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Abstract The pulsed laser deposition technique associated with a low energy cluster beam is used to deposit cobalt thin films with a thickness 100–200 nm and cobalt dots of a diameter 100–200 nm on silicon substrates. The deposited thin films of Co are composed of clusters of a size 10–50 nm, with very few large grains as revealed by atomic force microscopy. The observations performed by magnetic force microscopy on as-grown thin films reveal randomly distributed out-of-plane magnetic domain structures. These magnetic domains are aligned linearly by applying an external magnetic field either perpendicular or parallel to the substrate during the deposition. In addition, the effect of film thickness and roughness on multidomains is reported. The increase of roughness resulted in the decrease of magnetic domain width from 200 to 100 nm. This decrease is accompanied by the appearance of instability in the stripe domain pattern. Well separated cobalt dots of diameter in the range of 100–200 nm are also deposited on silicon substrates, which show arc-like multidomains. The domains seem to be oriented along the long axis of the dots. The domain structure of Co nanodots is similar to that of Co thin films indicating strong magnetic coupling of clusters.

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1 Introduction

The study of magnetic domains and its relationship with the morphology of nanoparticles and thin films is of technological and scientific interest. Thin films and nanoparticles of cobalt synthesized by various methods have been found to be model systems for magnetic studies due to their domain structures and the versatility of their magnetic properties [1, 2]. Cobalt based compounds are also popular for magnetic recording media because of their higher recording density and higher coercivity [3]. The magnetization process of Co thin films, multilayer with other noble metals, submicron sized particles, and their patterned structures have been the subject of extensive investigations as they are both promising for industrial applications and important for understanding the fundamental aspects of surface magnetism [4–8].

The nanostructured ferromagnetic particle arrays are interesting systems for understanding the switching behavior due to high coercivity of the submicron size magnets with potential applications. An assembly of nanomagnets with large uniaxial anisotropy is required in various future magnetic devices such as ultra high density recording media and bias magnets in monolithic microwave integrated circuits. Magnetization of a single-domain particle can be in two discrete states, either up or down which can be switched using an external field. Therefore, each particle is able to code a single bit in a microstructured recording media [3, 9]. According to theory as well as experiments, ferromagnetic particles below a critical size possess single domain structure [6]. Therefore, the domain structure and coercivity depends

strongly on the size of the grains and the morphology of thin films.

In the literature, the studies on magnetic domains of Co are based on thin films deposited using conventional techniques like sputtering [3, 10–12], e-beam evaporation [5, 13], molecular beam epitaxy (MBE) [14] and nanoparticle patterned arrays fabricated using lithography. The structural and magnetic properties of thin films depend on specific method of preparation, deposition conditions, and substrate, etc. [15]. In this paper, we report the topographical and magnetic domain structures of cobalt nanodots of diameter 100–200 nm and thin films of thickness 100–200 nm prepared by pulsed laser deposition (PLD) using a low energy cluster beam. In this study, magnetic force microscopy (MFM) has been used to image the domain structure of nanodots because of its high resolution and sensitivity. These results are compared with the hysteresis curves traced using vibrating sample magnetometer (VSM).

Cobalt granular films of various thickness and roughness have been deposited. The evolution of magnetic domains with the thickness of films and the effect of roughness on the formation and stability of domains under the action of external magnetic field has been analyzed. This is important because the formation and stability of magnetic domains is a key issue for applications in magnetic recording technologies. The roughness and imperfections influence the nucleation and pinning of domain boundary walls and affect the magnetic order within the domains [12].

2 Experimental

2.1 Pulsed Laser Deposition System

A cluster generator is used to deposit Co thin films. The PLD system equipped with a cluster generator allows to deposit high density granular films of various grain sizes. It is also intended for the deposition of low flux or isolated nanoparticles. In the previous paper we have reported the details of technical data and some preliminary results on Co and Cu nanoparticles and films [16].

Cobalt target is irradiated by an excimer laser. The resulting plume is rapidly cooled down by a pulse of helium gas delivered by an electromagnetic valve. Triggers of the laser and the pulse valve are adjusted in order to synchronize the laser beam impact on the target with the helium gas puff. The upper helium pressure is 10 bars and the opening duration of the valve is 280 µs. Then the mixture of gas and aggregates is ejected through a nozzle into a high vacuum chamber with background pressure equal to 10^{-8} mbar. At the exit of the nozzle, clusters have a low kinetic energy leading to a low energy cluster beam deposition regime (LECBD) [17]. They are deposited on a Si (100) substrate located in front of the nozzle exit, perpendicular to the cluster beam. The films deposited over a large thickness range 50–200 nm consist of grains of different size 10–70 nm depending on the sample without island growth. We have also deposited Co dots of 100 to 200 nm in diameter to study their individual domain structures.

2.2 Magnetic Force Microscopy (MFM)

Picoscan atomic force microscope from Molecular Imaging is operated in lift mode with topography and magnetic images collected simultaneously. Commercial magnetic cantilevers from nanosensors with resonance frequency in the range 50 to 70 kHz are used to sense the stray magnetic field just above the sample surface. Cantilevers are vibrated at their resonance frequency and the oscillation phase shift at certain height above the sample surface during scanning constitutes magnetic image. The lift height varies from 50 to 120 nm above the surface of the sample and is optimized in order to reduce the influence of topography on the magnetic response. The bright and dark contrast in the MFM image corresponds to the repulsive or attractive forces which in turn represents domains with up or down out-of-plane magnetization, respectively.

2.3 Vibrating sample magnetometer (VSM)

The macroscopic magnetic response of the films was determined using a vibrating sample magnetometer VSM with 8 T maximum field, in the 15–300 K temperature range. The hysteresis loops were registered by applying the field both in-plane (IP) and perpendicular-plane (PP) to the surface of the film.

3 Results and Discussion

Figure 1a shows the topography of the Co film of thickness 100 nm deposited on a silicon substrate. The grain size varies from 40 to 70 nm with an average diameter of about 50 nm. Normally, each MFM image is accompanied by a topographic image since they are recorded simultaneously. However, it is observed that the larger grains are composed of many smaller ones. Earlier we reported the deposition of the grains of size below 10 nm [16]. The other details of the films/nanoparticles are listed in Table 1.

Figure 1b shows MFM image taken on the films consisting of smaller grains of 50 nm average diameter (Sample 1) which reveal the random stripes. These random domains are aligned on application of external magnetic field as shown in Fig. 1c. The randomly oriented domains have already been observed in the absence of external magnetic fields in cobalt thin films, various cobalt alloys, and magnets of rare

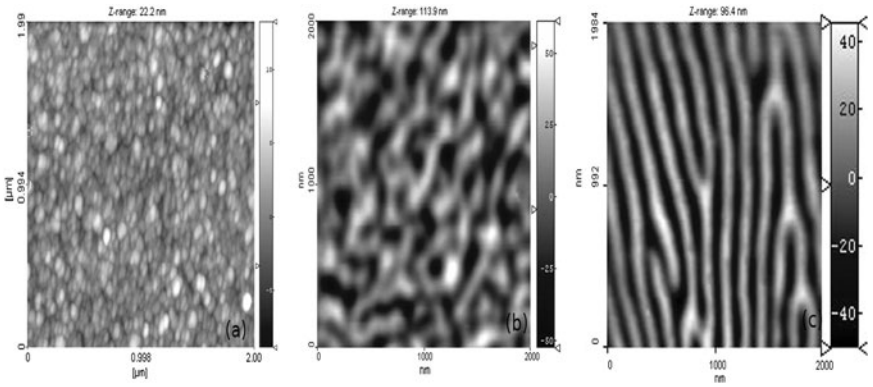

Fig. 1 AFM/MFM images (**a**) topographic and (**b**) Magnetic image of thin film of cobalt with average grain size of 50 nm. (**c**) Magnetic image of the film subjected to magnetic field shows alignment of domains

Table 1 Representing the important parameters; thickness, grain size and roughness of the films and magnetic domain width as measured by MFM. The values represent the range with minimum and maximum for a given film. The numbers in the brackets are the average values measured for about 10 frames of images on different locations of the films

	Sample 1 as deposited	Sample 2	Cobalt dots
Thickness (nm)	100	200	25–30 nm
Grain size (nm)	40–70 (50)	500–800(600)/smaller 50	140–200 nm
Roughness (nm)	15–18 (nm)	100–125(108)	90 nm
Domain pattern	Random/twisted	Strips with fluctuation near large grains	Arc like
Domain width	180–225 (200)	80–125 (100)	200–275 nm

earth compounds [15, 18]. As measured from MFM images, the width of domains is about 200 nm. It is to be noticed that the domain size is much larger than that of elementary grains. We could not obtain MFM signal for the film with a thickness of 50 nm. Such vertical multidomains exist only for films with thickness 100 nm and above. It is in good agreement with the prediction that the magnetization turns from in-plane to out-of-plane only above a certain critical thickness [19]. This kind of thickness dependence has been observed for a MBE grown Co film, for which the critical thickness was evaluated to be around 60 nm [14].

Figure 1c display highly aligned domains for sample 2. This is obtained by placing a magnet of 50 mT and size ($60 \times 20 \times 15$ mm^3) just behind the substrate during the deposition in the growth chamber. Such an alignment has also been achieved by subjecting the sample with random domains to a magnetic field before imaging with MFM. Periodically spaced parallel stripe domains are aligned along the direction of the applied field in order to minimize the Zeeman energy [20]. Using MFM, the average domain width is measured to be about 100 nm.

MFM records the magnetization component which is perpendicular to the film surface. Thus, the present results confirm the existence of perpendicular magnetization component of films. This is in agreement with the existence of stripe domains derived from in-plane magnetization measurements. The AFM topographic images of thinner films, below 50 nm, were found to be very similar to those of thicker Co films. However, in this case, no magnetic domain could be seen by MFM, implying that the magnetization is fully aligned in-plane (IP). At very small thicknesses of the films, the out-of-plane component of the magnetization disappears under the influence of shape anisotropy [12, 16].

The above results may be compared with the Hysteresis curves shown in Figs. 2a and 2b, registered using Vibrating sample magnetometer (VSM). When the magnetic was field applied perpendicular to the film plane (PP configuration), the magnetization variation is linear up to magnetic saturation which is reached at 0.6 T, a value which is almost temperature independent. The PP coercive field is approximately twice that of IP coercive field as shown in Fig. 2b. The IP hysteresis loop of a 100 nm Co cluster film was measured at 15 K after cooling the sample under field (FC) 2T and in zero field (ZFC). No loop shift was found on the FC cycles. The exchange bias effect is often found when Co particles are coated by antiferromagnetic CoO oxide [21]. In agreement with TEM observations [22], the lack of exchange-bias effects in the present systems suggests that the amount of CoO at the surface of the Co clusters is negligible.

As can be seen from the topographic image in Fig. 3a, sample 2 consists of few large isolated grains with an average diameter of 600 nm and a mean roughness of 108 nm. In comparison, to sample 1, this sample has 10 times larger roughness. Actually, the roughness has been increased by increasing the film thickness, which is about 200 nm, dou-

ble that of sample 1. From MFM observation as in Fig. 3b, we could see clear stripe domains. The measured width of parallel domains is about 100 nm which is half of the domain width for the sample 1. Generally, the domain width is of the order of the thickness of the film and should vary like square root of thickness as reported [15]. It has to be noticed that the larger grains in sample 2 are covered by a bed of smaller clusters with an average size of 50 nm. We observe that the stripe pattern originates effectively by the layer of 50 nm grains as in the case sample 1. However, the fluctuation in the domain pattern and the compression is due to the underlying larger grains and increased roughness of the sample. From the one to one correspondence of topographic and magnetic images (Figs. 2a and 2b), one can clearly notice the fluctuation near the larger grains. For higher roughness of the films, domains become more unstable and it has not been possible to image them under MFM. Generally, the instability in surface magnetism can be caused by several factors like morphology of the films, defects, external magnetic field etc. Such an experimental result is modeled and simulated by Joo-Voo-Kim et al. [23]. Also, fluctuation of stripe domains due to external magnetic field is studied by Demand et al. [24]. It is observed that the parallel stripe domains fluctuate into zigzag path, and finally break down to magnetic bubbles with the increasing strength of external magnetic field applied perpendicular to the stripe domains.

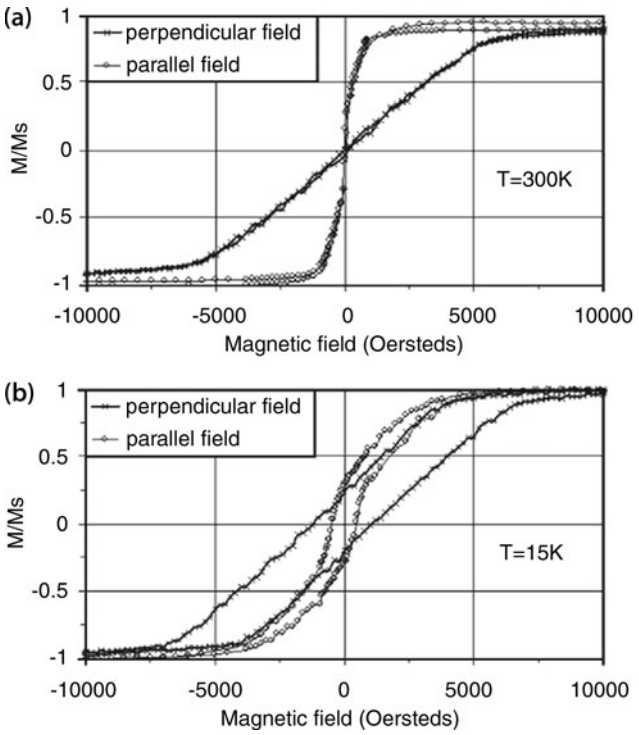

Fig. 2 Magnetization hysteresis curves registered on a cobalt film using VSM with both in-plane and perpendicular plane fields at temperature 300 K and 15 K

Figures 4a, 4b and 4c show the topographic, phase and MFM images of Co dots, respectively. From the topographic image, the diameter of large dots is scattered around 200 nm and that of smaller ones is scattered around 140 nm. The average height of the dots is evaluated to be 50 nm and 25 nm for the large and small dots, respectively. It is evident from the MFM image that each dot is characterized by a pair of arc-like domains of one orientation with a domain in the middle of opposite orientation of magnetization. From the higher magnification topographic images, it appears that these dots are supported on a thin layer of small cobalt clusters at the bottom. The total domain width measured at the center of each dot in the MFM image is about 270 nm and 200 nm for the large and small dots, respectively, which is much larger compared to the diameter of the dots. The dots were deposited in presence of an external magnetic field and, therefore, all the domains are oriented in the vertical direction. However, close observation reveals that in certain cases as marked on the images by arrows the orientation of domains depends on the morphology of the dots. For nonspherical dots, the domains seem to preferably

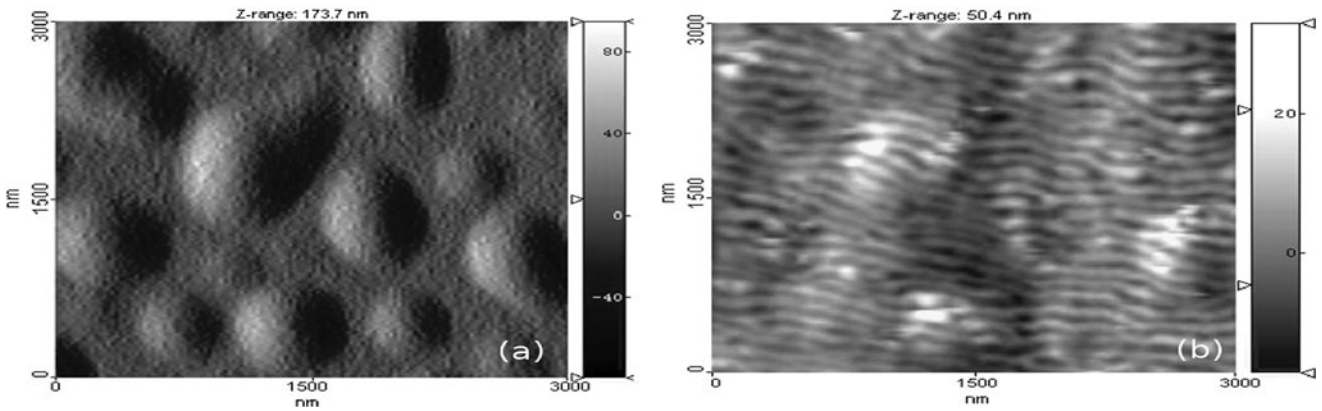

Fig. 3 AFM/MFM images (**a**) topographic and (**b**) magnetic image of thin film of cobalt consisting of larger grains with average grain size of 600 nm

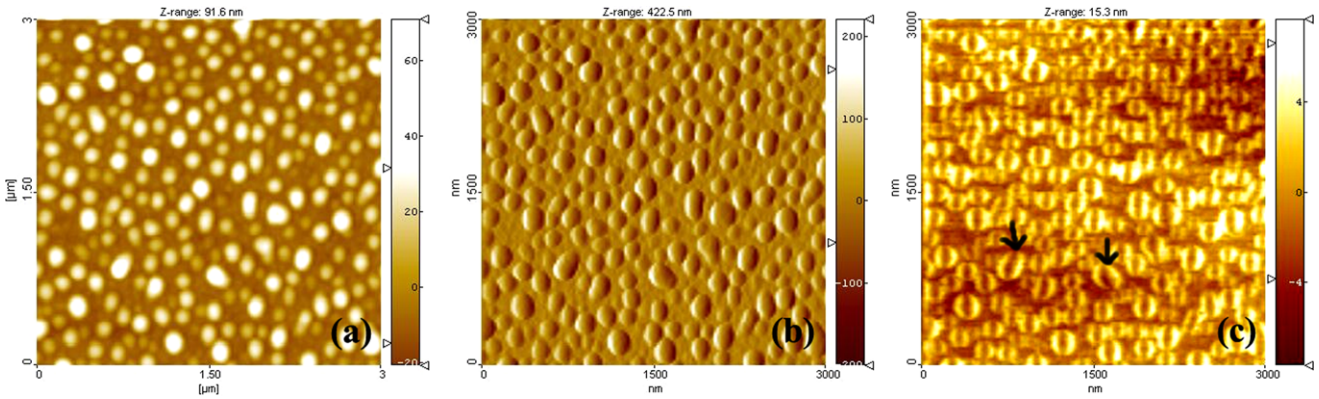

Fig. 4 (**a**) Topographic (**b**) phase and (**c**) MFM images of cobalt dots with average grain size of 200 nm

be oriented along the long axis of dots as marked by arrows on the images.

Our observations can be compared with some of the important reported results [3, 9]. Most of the reported single-domain studies were carried out on patterned arrays of Co dots prepared by lithography [9]. It is reported that single-domains were registered on 50 nm particles whereas, the isolated particles larger than 200 nm show multidomain structure. Also, in the case of elliptical dots of size $140 \times 250 \times 7$ nm, the magnetization direction is aligned parallel to the long axis [20, 25, 26].

From the figures, it can be observed that the multidomain structure of dots is similar to that of films consisting of high density small clusters of 10–50 nm in diameter. This indicates a strong magnetic coupling of small clusters in the case of films. Therefore, it has not been possible to observe a local magnetic structure of 10 nm clusters in the films, unless they are well separated.

4 Conclusions

Cobalt thin films and nanodots are deposited using low energy cluster beam obtained with pulsed laser ablation technique. The deposited films show random magnetic domain structure as imaged under magnetic force microscopy. The random domains were arranged forming straight domains using an external magnet. The MFM results agree well with the data obtained from hysteresis loop measurement using VSM. Cobalt dots of size about 100–200 nm show arc like magnetic domain structure. The domain structure of films as well as cobalt dots indicate exchange coupling of smaller cobalt clusters in the film.

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