8.1. Introduction

Due to their potential applications in magnetic sensors and magnetic recording heads [163-166] soft magnetic nanostructures assumes significance from an applied point of view. Several strategies have been developed for the growth of nanostructured magnetic materials [167-171]. Nanolithography-based methods, solution-based approaches and template-based methods are some of them. Some of these methods, however, require high temperatures and special conditions while in other cases, they demand complex and tedious procedures. For instance, in template assisted growth of nanostructures, the selection of suitable catalysts and templates is not straightforward, and the removal of templates and the stabilization of unsupported nanostructures represent crucial issues that may compromise the structural and physical properties. The capability of obtaining ordered arrays of well-defined and periodic nanostructures in an accurate, fast, and inexpensive fashion would be of great interest not only from an applied perspective but also from a fundamental point of view.
Oblique angle vapour deposition offers advantages of fabrication of nanostructures over large areas, as required in many advanced technological and industrial applications [89,172-178]. Some other advantages of this technique are the non requirement of templates, relatively low temperatures and less harmful chemicals for the nanostructure fabrication [179].

Generally, the morphology of the nanostructures thus obtained is influenced by the substrate surface roughness and the growth conditions used for the film formation along with oblique angle, deposition rate, deposition time etc. The growth of nano structures will be the resultant of the competition between the smoothening due to adatom surface diffusion and roughening by self shadowing. For the synthesis of well defined nanostructures having appropriate separation and clear surface morphologies, an understanding on the interplay between the mechanisms involved in the growth process is essential. From an applied stand point, a detailed knowledge of the growth behaviour of the nanostructures on a solid surface will aid in synthesizing nanostructures with well defined roughness and geometry.

To date, ferromagnetic nano columns have been grown by vapour phase co-deposition and oblique angle vapour deposition [57-60]. Fe-Ni-Co nano columns were grown by the self organization of vapour phase co-deposited Fe-Ni-Co [58]. Nano columns with Co/Cu bilayers were obtained by two-source oblique angle vapour deposition [59]. The surface evolution of amorphous nano columns of Fe-Ni obtained by oblique angle vapour deposition on silicon substrate has been described in previous chapter and is published elsewhere[180]. It was found that the growth of nanostructures on a silicon substrate were more or less random and surface diffusion of adatoms led to the coarsening of the columns at higher deposition time.

There are many potentially attractive applications for these columnar films, if they can be prepared with the desired microstructure and inter column separation within the practical limits of time and expense. A critical issue concerning the
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The achievement of this goal is the control of nucleation. The nucleation events occur preferentially on defects and abnormalities on a substrate surface. While so much work has been devoted to understand the effect of deposition parameters on the morphology of elementary metal nano columns [120,158-160], there has been virtually no effort to understand the influence of substrate topography on the growth mechanisms and magnetic properties of nanostructures of amorphous alloys. A study relating the substrate surface roughness with column evolution and magnetic properties will be important not only from a fundamental perspective but also from an applied standpoint.

The main objective of the work presented in this chapter is to investigate the influence of substrate surface roughness on the morphology and the separation between the nanostructures in oblique angle vapour deposition. Fe-Ni based amorphous nano columnar structures were obtained on silicon and glass substrates having different initial surface features. Growth of columns on different substrates is studied using atomic force microscopy (AFM). Further, the evolution of magnetic properties with column growth is studied using atomic force microscopy and magnetic force microscopy (MFM) techniques. The combined use of AFM and MFM will aid in understanding the intricate relationship between the magnetic properties and the nanoscale sized surface features. The MFM measurements are supplemented with vibrating sample magnetometry (VSM) to correlate the average magnetic properties with microstructure.

8.2. Experiment

8.2.1 Preparation

Commercially available Metglas 2826 MB ribbon of composition Fe_{40}Ni_{38}Mo_{4}B_{18} was employed as a source material to deposit Fe-Ni thin films on silicon (coded as sample A) and glass substrates (coded as sample B). The substrates
were cleaned with acetone, ethanol and trichloroethylene and were immediately loaded into the vacuum chamber. The substrate was tilted in such a way that the angle between the surface normal to the substrate and the direction of incoming flux was at an oblique angle of 40°. No substrate rotation was provided. The films were deposited by thermal evaporation using a current of 25 A at a base pressure of 1x10⁻⁵ mbar onto substrates oriented at an oblique angle of 40° to the flux. The base pressure of ~ 1x10⁻⁵ mbar was achieved by a diffusion pump backed with a rotary pump. The source to substrate distance was 26 cm.

8.2.2 Characterization

The imaging of topography and magnetic domains was performed with a commercial AFM (Veeco Instrument, Multimode) operated in tapping plus lift mode. This ensures separation between the topographic and magnetic data. A commercial Si tip coated with a Co-Cr thin film (80 nm thick) that has magnetized vertically was used (Micro Masch NSC35/Co-Cr). The radius of curvature of the tip was less than 90 nm. The full tip cone angle was less than 30°. Images were collected at different tip to sample separation (lift height) ranging from 30-120 nm. Room temperature magnetization measurements were carried out using a vibrating sample magnetometer (DMS 1660 VSM) with an external field varying from −5 to +5 kOe.

8.3. Results and discussions

8.3.1 Evolution of morphology with substrate roughness

Characterization of the substrate surface was performed using Atomic force microscope. Figure 8.1 a & b shows 3-D AFM images of silicon and glass substrates. The best known parameter characterizing the morphology of a surface is its root mean square (rms) roughness. The rms roughness obtained from the AFM images for silicon and glass substrates were 0.77 nm and 3.38 nm respectively.
Figure 8.2 (a) and (b) shows the topographic images of films coated on silicon (coded as sample A) and glass substrates (coded as sample B) respectively. A clear difference in morphology of the films obtained on the two different substrates was observed. Lateral size of the columns on silicon substrate is small (~250 nm) as well as they are closely packed (average separation between the columns~240 nm). While well separated (average separation between the columns~570 nm) and larger columns were obtained on a glass substrate (lateral size around 450 nm). The measured rms roughness was ~3.16 nm and 8.64 nm for sample sets A and B respectively. In both cases the rms roughness were smaller than the total film thickness (~ 50 nm) which suggests that there are film deposits in between the columns.

The line scans shown in figure 8.3 illustrates the size of the nano columns on both silicon and glass substrates. Tip convolution effects result in an exaggerated column width and in actual case the width of the column could be much less [181].

Columnar growth is a result of atomic shadowing mechanisms that occur at the substrate surface [89]. During the initial stages of vapour deposition, adatoms condense on to the substrate and form individual separated islands or nuclei. When the substrate is tilted such that the incident vapour arrives at oblique angles, the
The topography of adatom nuclei results in geometrical shadowing over regions of the substrate, preventing the coalescence of nuclei into a continuous thin film layer. The nuclei capture the vapour flux that would have landed in the shadowed regions, resulting in the formation of columns.

**Fig. 8.2** a) 3-D AFM images for columns on (a) Si substrate (b) on glass substrate. MFM images for columns on (c) Si substrate (d) on glass substrate. Lift height in MFM scans is 60 nm.
Deposition on smooth substrates generally results in a pseudo-random arrangement of nucleation sites during the initial stages of film growth, producing a similar distribution of columns over the substrate surface [175]. On the other hand, if there is a small perturbation to a flat surface, the irregularities act as nucleation sites for the columnar structure [120]. The topographical variations define the shadow regions on the substrate during the initial stages of film growth so that adatom nucleation is forced to occur on the surface protrusions. The small perturbations on the flat surface increase with time because surface protrusions receive more flux than valleys. If the protrusions are high enough, their shadows extends to its neighbour and
suppresses the inter-seed film growth. This can be a reason for the decreased inter columnar competition in sample prepared on glass substrate, where the initial irregularities were 2-3 nm in height. In glass substrates the column evolution is defined by the topographic protrusions on its surface, while in silicon it is defined by the Fe-Ni clusters formed initially. In films deposited on glass substrates, the separation between the columns was defined by the position of the irregularities on the substrate surface. Random nucleation on a smooth silicon surface resulted in a randomly arranged nano columns.

8.3.2 Magnetic properties

8.3.2.1 Magnetic force microscopy studies

MFM with a cantilever vibrating normal to the sample is sensitive to the gradient of the tip-sample interaction force in the normal direction of vibration, that is, to \( \frac{\partial F_z}{\partial Z} \) [81,182]. The interaction force is \( F = q \nabla H \), where \( q \) is the tip moment and \( H \) is the field at the tip. When the tip is ideally hard (e.g. coated with Co-Cr) and of constant moment \( q_z \) directed normal to the sample surface, the MFM signal is proportional to \( q_z \frac{\partial^2 H_z}{\partial Z^2} \), that is, it is sensitive to the second derivative of the normal component of the sample field. The contrast in MFM image is thus proportional to the gradient of the magnetic force between the tip and the sample. Figure 8.2 (c) is an MFM image of sample A obtained at a lift height (tip–sample separation) of 60 nm. The contrast seen in the MFM images implies the presence of magnetic domains with out-of plane magnetic component. Sizes of the domains are larger than the width of individual columns which means that in this film there exists a magnetic interaction between the individual columns. In figure 8.2 (d) an MFM image of sample B indicates the existence of well separated circular domains. The microstructure of this
film is that of well separated larger columns and the MFM image from the corresponding scan area reveals that the out of plane magnetic component is only from individual columns. A one to one correspondence can be seen between the columns in the AFM image and circular domains in the MFM image of sample B (fig 8.2(b) & 8.2(d)). It is seen from the AFM images that the lateral size of columns for the two sample sets A and B are correspondingly 250 nm and 450 nm. While MFM images showed that the magnetic domain size are around 1.5 µm and 450 nm for the sample sets A and B respectively. This gives a clear indication that in sample A, magnetic correlation length is beyond the lateral size of individual columns. Thus in sample A the magnetic structure is not only determined by the individual columns but also by the magnetic interactions between them. The magnetic correlation length in sample B is within the column width itself which means that the columns in sample B are exchange isolated.

**Fig.8.4** MFM images obtained at a lift height of 100 nm under two different tip magnetization orientations
In order to ensure that the contrast in the MFM image of sample B is caused by the film magnetization and not due to topographical artefacts, the tip magnetization was reversed by 180° and a MFM scan was obtained from the same area. Because it was not possible to relocate the scanning probe exactly the same scan line after removal of the tip for remagnetisation, exact mirror symmetry between the traces could not be expected. Figure 8.4 (b) shows MFM image of sample B for a lift height of 100 nm obtained after reversing the tip magnetization. The phase shift is now positive and the contrast is now inverted from dark to bright [when compared with MFM image in figure 8.4 (a)] which is an indication to the fact that the contributions to the MFM images are a result of magnetic forces of the sample.

**Fig.8.5** a) Topography b) Phase image at lift height of 30 nm c) 60 nm and d) 100 nm for sample B.
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Figure 8.5 shows the topography and corresponding MFM images of samples B for a lift height of 30 nm, 60 nm and 100 nm. The plot of phase shift versus lift height (Figure 8.6) showed an exponential decay in agreement with our expectations. This decay in phase shift with lift height is due to the decay with distance of magnetic force from the sample.

**Figure 8.6** Scanning height dependence of phase shift in sample B

### 8.3.2.2 VSM studies

In order to gather more insight on the magnetic behaviour of the columns, room temperature magnetization measurements were performed on the two sample sets using a vibrating sample magnetometer. The measurements were carried out both in parallel (in-plane) and perpendicular fields (out-of-plane). Figure 8.7 and 8.8 shows the magnetization curves for sample sets A and B respectively. The saturation magnetization was found to be ~ 870 emu/cc in both cases. It is to be noted that a low field was only necessary to saturate the magnetization in the in-plane direction while a field as high as 5000 Oe could not saturate the material in the out of plane direction.
direction. AFM studies showed that the column height is smaller than the total film thickness implying that there is film deposits in-between the columns. Because of these deposits, the portion close to the substrate will become a continuous layer and the geometry of the whole system will be layer plus island type, in which islands are arranged on top of the layer. Due to the absence of significant magneto crystalline anisotropy, the magnetization direction will be largely influenced by the shape anisotropy. Within the layer, since the long axis is along the substrate plane, the magnetic direction will be in plane. On the other hand, in the islands, where the long axis is perpendicular to the substrate plane, the magnetic direction will be out of plane. The magnetization measurements using VSM will have contributions from the whole sample and one will only notice signatures of in plane magnetic direction because of the domination of the contributions emanating from the layer. However MFM being sensitive to the surface detects the out of plane component from the islands. The measurements show that the perpendicular magnetic component from the islands is small due to their small aspect ratio. The in-plane hysteresis loops show that the field necessary to saturate sample B (~2000 Oe) is double than that required for sample A (~1000 Oe). This can be correlated with the morphology of the columns prepared in two conditions.

In sample A the columns are packed close together (average inter-column distance around 240 nm) and MFM showed that there is a magnetic interaction between the columns. On the other hand, in sample B, the columns are well separated (average inter-column distance around 570 nm) and magnetic interaction between the columns is minimal. Since the columns of sample B are non-interacting, a larger magnetic field is required for achieving magnetic saturation. This field required for the sample A will be less due to the magnetic interaction existing in between the columns. Another feature to be noted from the in-plane hysteresis loop (Figures 8.7 & 8.8) is
that coercivity of sample B (~65 Oe) is larger than that of sample A (~40 Oe). This is due to the increased surface roughness of sample B.

![Hysteresis loop for nano columns on Si substrate](image1.png)

**Fig.8.7** Room temperature hysteresis loop for nano columns on Si substrate in parallel field. Inset shows the loop recorded in a perpendicular field.

![Hysteresis loop for nano columns on glass substrate](image2.png)

**Fig.8.8** Room temperature hysteresis loop for nano columns on glass substrate in parallel filed. Inset shows the loop recorded in a perpendicular field.
Small irregularities on the surface of a film inhibit the passage of a domain wall because the energy stored within a domain wall surrounding such a region is smaller than in an undisturbed domain wall and consequently the system energy must be increased to enable the domain wall motion. This is consistent with our previous observations in swift heavy ion irradiated Fe-Ni thin films [183].

**8.4 Conclusions**

Magnetic columnar thin films based on Fe-Ni were obtained by oblique angle deposition. Initial surface roughness of the substrate played a decisive role in the final morphology of the columnar structures. Thicker and taller columns were obtained on a glass substrate when compared to that on a smoother silicon substrate. Nucleation of Fe-Ni nano columns on a smooth silicon substrate were at random, while that on a rough glass substrate was defined by the irregularities on the substrate surface. The morphology of the resultant films determined their magnetic properties. Due to their small inter column separation, magnetic interaction was present for nano column arrays prepared on silicon substrates. On the other hand, well separated nano columns on glass substrate resulted in exchange isolated magnetic domains. These results also indicate that oblique angle deposition on a patterned substrate can result in well separated nano columns which can be promising for future high density recording applications.