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Anisotropic magnetic properties of obliquely deposited Ni films

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Magnetic properties of evaporated nickel films, deposited onto 75 μm thick 300 H Kapton substrates by evaporation at oblique off-normal angles of incidence, α were investigated by SQUID magnetometry. We found that, in the film plane, the direction of easy magnetization lay perpendicular to the incidence plane for films deposited at $\alpha < 50^\circ$. At large αs, the easy axis changed to the direction parallel to the incidence plane. The anisotropy, coercivity and squareness of the hysteresis loops increased with an increase in α. The results may be qualitatively understood from the presence of an inclined columnar structure with shape anisotropy governing the demagnetization of the magnetic fields.

1. Introduction

Magnetic properties of thin films have been studied for many years. An important factor driving the interest in thin film magnetism is the capability to deliberately modify the structure of a material in order to influence its magnetic behaviour. The properties of a thin film differ from those in the bulk if the surface spins constitute a significant fraction of the total number of spins in the material, which is the case for thin films. Perhaps the most striking difference is the enhancement of the magnetic moments at the surface as compared to the bulk. The enhancement can be as large as 40%, as has been verified theoretically [1, 2] and experimentally [3] for the 3d elements Fe, Co and Ni. The exchange energies are also enhanced at the surface [2].

It is well known that obliquely deposited films exhibit a structure comprising columns inclined from the film normal towards the direction of the deposition flux and align along a direction perpendicular to the incidence plane. Clearly such films may have magnetic properties that can be qualitatively as well as quantitatively different from those of the bulk. The perpendicular alignment has been termed a bundle of columnar grains [4] or a row of columns [5]. The structural features may be understood, at least generally, by considering a self-shadowing mechanism [6] initially proposed for explaining crystalline alignment in early stages of film growth. Fig. 1 gives a schematic representation of films with inclined columnar features.

The columnar grains in obliquely deposited magnetic films induce a uniaxial magnetic anisotropy through the anisotropy of the demagnetizing field, due to their shape anisotropies, and the texture also induces a magnetic anisotropy through magnetocrystalline anisotropy [7, 8]. The anisotropy of the remanence has been reported to be equivalent to the anisotropy field obtained by a torque measurement [9]. Below we report on the magnetic anisotropy of obliquely evaporated Ni films.

2. Experimental details

Films for this study were prepared by evaporating pure nickel wire (99.9% purity) on to 75 μm thick 300 H Kapton substrates at a pressure less than 10^{-6} mbar using a conventional oil diffusion pump system. The Kapton substrates were cleaned ultrasonically and then dried in methyl alcohol before being loaded into the chamber. Tape was used to mask the ends of the substrate, thereby making sharp steps to enable accurate measurement of the film thickness by use of a surface profilometer. The substrates were attached to a substrate holder with a rotatilt arrangement was placed 15 cm directly above the evaporation source. The substrate holder could be turned so that the deposition angle α between the direction of the incident evaporated flux and the substrate normal could be set to any value between 0 and 90°.

Nickel wire was placed in a helical tungsten filament and was resistively evaporated by resistive heating of the filament. The deposition rate at normal incidence lay between 0.8 and 1.2 nms^{-1} and was monitored *in situ* by a quartz crystal microbalance. Film thickness was determined *ex situ* by a Tencor Alphastep 200 surface profilometer. The samples had a film thickness d between 30 and 250 nm. The profilometer is a stylus instrument with a capability of sensing vertical displacements over a surface to an accuracy of 5 nm provided a good step is made between the film and the substrate. We prepared samples with deposition angles in the $0 \leq \alpha \leq 85^\circ$ range. The samples were cut to size 0.5×0.5 cm for SQUID characterization.

Magnetization measurements were performed using a Superconducting Quantum Interference Device

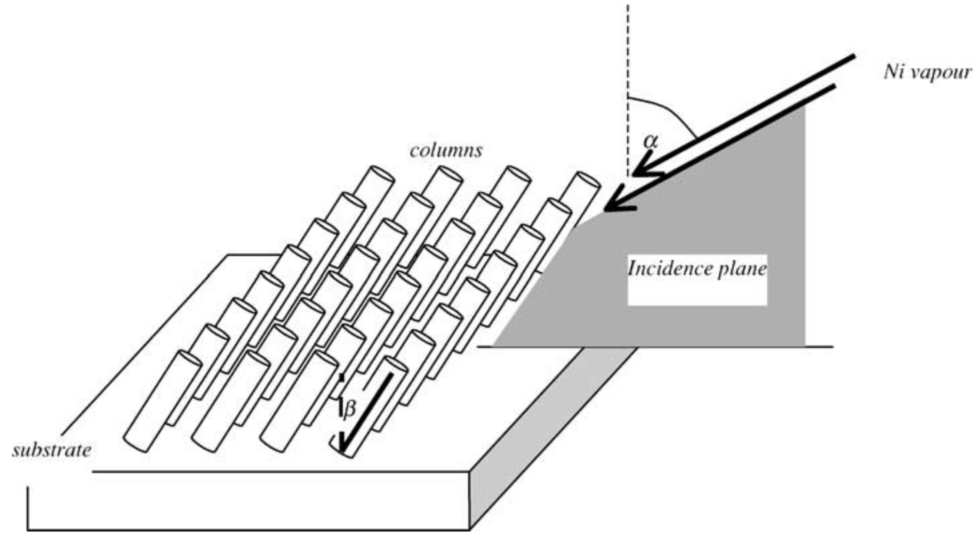


Figure 1 Definition of angles of deposition α and of inclination β of the columns, respectively.

(SQUID) magnetometer equipped with a 5.5 Tesla superconducting magnet operating in persistent current mode. A variable-temperature inset was used for measurements in the $35 < \tau < 300$ K range of the field dependence of the in-plane magnetic moment, denoted *m* (H), for Ni films made at different αs. Data were recorded by first saturating the magnetic moment of the film through a large negative field and then determining *m* (H) until saturation took place in the opposite field direction.

Microstructural investigations were performed using a high resolution Leo 1550 scanning electron microscope. The sample was fractured in the deposition plane and oriented such that the edge under investigation was directly opposite to the detector. Images of the cross-section of the films were then obtained.

3. Results and discussion

A cross-sectional micrograph of a fracture in the deposition plane showed well-defined microstructures with columns which are uniform in width from the substrate upwards. The column boundaries are separated by voids. A general resemblance to the schematic picture in Fig. 1 is obvious. Fig. 2 shows a typical cross-sectional morphology of a 250-nm-thick film evaporated onto glass at $\alpha = 85^\circ$. The films comprise four layers and were obtained through four consecutive evaporations. This method was used in order to obtain a sufficiently thick film and for studying the role of the conditions for the microstructural evolution. The columnar inclination of the first layer on the substrate is larger than for subsequent layers, which clearly illustrates the effect of the substrate. The alignment

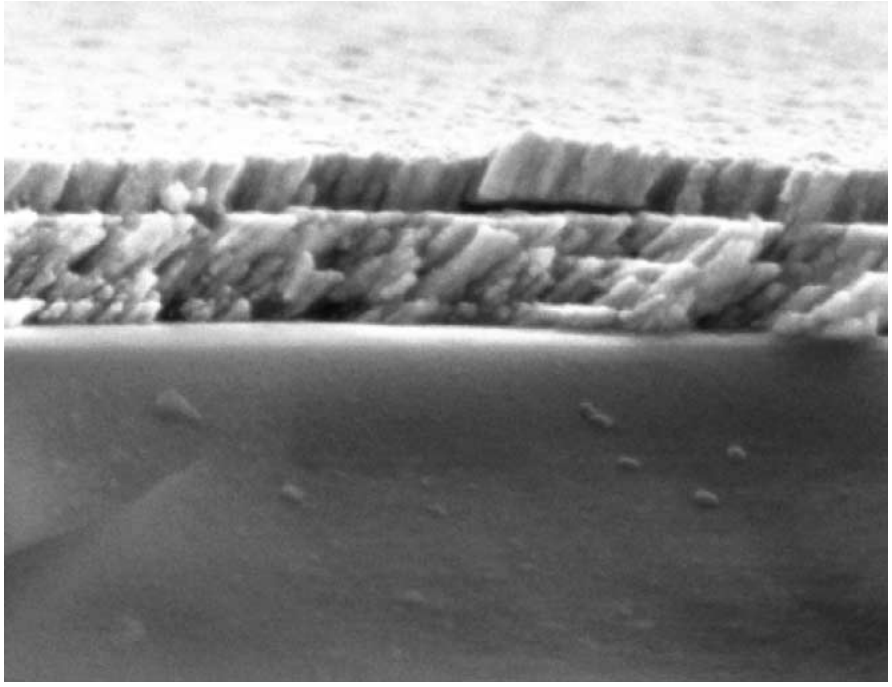
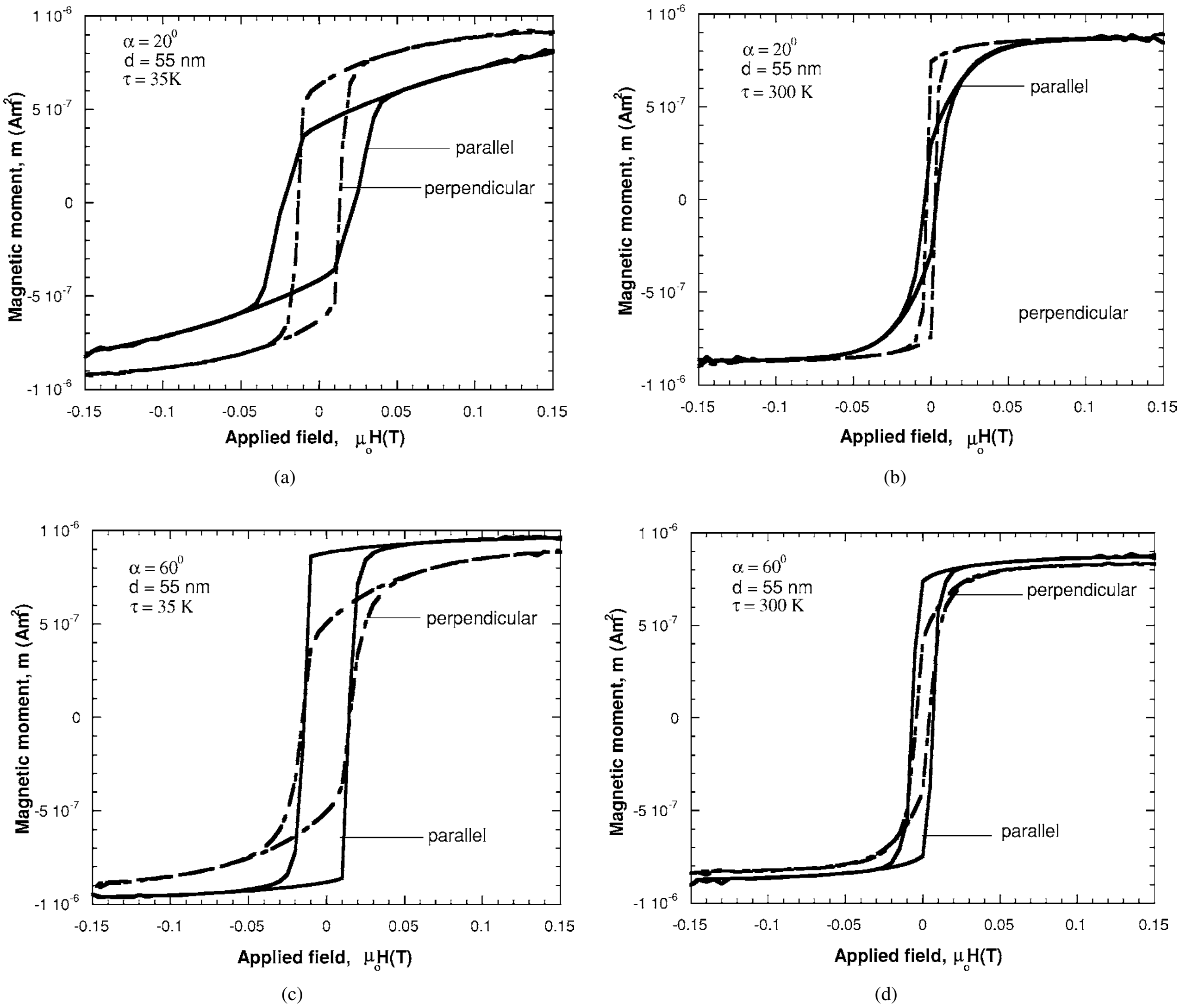


Figure 2 Scanning electron micrograph of a 250-nm-thick four layers of Ni film deposited on glass at 85° to the substrate normal and fractured in the deposition plane.



(a) (b) (c) (d)

Figure 3 Hysteresis loops of Ni films 55 nm thick evaporated at α being 20° and 60° on Kapton substrate and recorded for a field applied along the direction parallel and perpendicular to the incidence plane. Measurements were made at temperatures τ as indicated in the diagrams.

direction of the columns was nearly normal to the substrate for films deposited at $\alpha < 50°$, whereas an unambiguous inclination of the columns in the direction towards the evaporated flux could be seen above this magnitude of α.

Fig. 3a–d illustrate m (H) for for 55-nm-thick Ni films evaporated onto Kapton substrates at α being 20 and 60°. The data were recorded at τ equal to 35 and 300 K using the field applied both in the direction parallel and perpendicular to the incidence plane of deposition.

An enhanced anisotropy was found in the incidence plane. This effect can be explained by the formation of long chains of crystallites perpendicular to the direction of the incident vapour beam [6, 10]. The occurrence of the oriented crystallite structure is explained in terms of a self-shadowing mechanism [5]. Although the direction of the crystallites is known, the relation to the change in anisotropy is not immediately clear. Recent work on Cu evaporated on tilted Cu (001) substrates showed that steering may have an important influence on the morphology of growing films [11]. Steering originates from long range attractive forces between incoming and surface atoms, and leads to preferential arrival of atoms on top of islands, thereby forming anisotropic island shapes.

From the loops, the remanent magnetization measured parallel to the incidence plane $M_{r\|}$ is not zero and in addition the remanent magnetization measured perpendicular to the incidence plane $M_{r\perp}$ is smaller than the saturation magnetization M_s indicating the deviation of magnetization from the easy direction in the film plane. This fact suggests a stripe domain structure [7] which originates from the shape anisotropy of columnar grains formed.

In the analysis of the magnetic anisotropy, we have considered that the quantity $(M_{r\perp} - M_{r\|})/M_s$ is a measure of the magnetic anisotropy since the ratio M_r/M_s expresses the capacity to possess the magnetization in the direction of the magnetic field [7]. We have found the anisotropy constant to be positive and negative for films deposited at angles smaller and larger than 50° respectively. Positive or negative values of the in-plane anisotropy field correspond to the case of an in-plane easy axis of magnetization, perpendicular or parallel to the incidence plane of incoming atoms respectively.

The fact that the anisotropy is negative for films deposited at large angles indicates a geometric alignment of crystallites parallel to the incidence plane. The alignment induces the uniaxial anisotropy with the easy axis parallel to the incidence plane through the anisotropy of the demagnetizing field. The positive contribution of the columnar axis (c-axis) alignment is reduced by the negative contribution of the geometric alignment (shape).

4. Conclusion

We have presented a set of data on magnetic properties obliquely deposited Ni films. The results can be qualitatively understood from the presence of an inclined columnar microstructure with shape anisotropy of the inherent structural units affecting the demagnetization of the magnetic fields, thereby invoking field-dependent magnetization. In the film plane, the direction of easy magnetization lies perpendicular to the incidence plane at low incidence angles. The easy axis at high incidence angles changes to the direction parallel to the incidence plane. The magnetic anisotropy of obliquely deposited films is explained by considering the microstructure. At low incidence angles the column appears like a wall elongated in the direction perpendicular to the incidence plane. In this case the direction of the easy magnetization lies perpendicular to the incidence plane. However, at high incidence angles the column becomes needle-like, and the anisotropy changes direction parallel to the incidence plane. Our results highlight the extreme sensitivity of the magnetic behaviour of obliquely deposited films to the microstructural morphology.

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