Mat. Res. Soc. Symp. Proc. Vol. 696 © 2002 Materials Research Society

Surface reconstruction and induced uniaxial magnetic fields on Ni films

R. A. Lukaszew, B. McNaughton[1], V. Stoica[2] and R. Clarke[2]
Department of Physics and Astronomy, University of Toledo, Toledo, OH 43606

[1]NSF-REU Summer 2001, Physics and Astronomy, University of Toledo.
[2]Randall Laboratory of Physics, University of Michigan, Ann Arbor, MI 48109

ABSTRACT

Metal ceramic interfaces are important in applications as diverse as magnetic storage media to support catalysts. Understanding the correlation between the films structure and its properties is very important for applications. For example the lattice mismatch between substrate and epilayer during heteroepitaxial growth induces strain as well as defects on the structure of the films that in turn affect the magnetic properties.

We have previously shown that it is possible to obtain epitaxial (001) and (111) Ni films grown on MgO substrates. [1] In particular we observed that the crystalline quality of the films improved considerably after 10 nm of film growth and annealing of the films indicated significant reduction of the surface roughness. In addition, after annealing (001) Ni films we observed nano-patterning of the surface through self-assembly of periodic "stripes".

We will now present our analysis of the magnetic properties of these films, in particular the azimuthal dependence of the magnetization reversal, using MOKE, and we will attempt to correlate our findings with the surface characterization obtained with STM.

INTRODUCTION

The magnetic anisotropy, of epitaxial thin films is dominated by the crystallographic structure of the metal/substrate interface as well as the surface quality. For many practical applications (e.g. spin-dependent tunneling devices, [2]) the roughness at the surface must be very small in order to ensure the integrity of the subsequent epilayers. Thus, we have considered the growth of magnetic films on MgO substrates, which can be prepared with very smooth surfaces. [3]

Theoretical studies have indicated that for Ni films grown on MgO substrates, Ni is expected to strongly interact with MgO [4]. Various researchers have studied the orientation of Ni films on MgO substrates under various growth conditions [5], and some reports indicate that Ni may form an epitaxial relationship with Ni[001]//MgO[001] and Ni(010)//MgO(010) for films deposited using dc sputtering on MgO substrates held at 100^{o}C [6]. There are also reports on epitaxial growth of *fcc* metals on surfaces with hexagonal surface symmetry such as MgO (111) [7]. Sandström et al. [8] have shown that at growth temperatures between 300^{o}C and 400^{o}C it is possible to grow smooth <111> oriented single domain epitaxial films on MgO substrates, utilizing dc magnetron sputtering in an ultra high vacuum (UHV) chamber.

We have reported previously that we have been able to grow epitaixal and smooth Ni films on MgO using MBE. [1] Here we report on our preliminary studies on the magnetic switching behavior of these thin films, in particular in the (001) orientation.

EXPERIMENTAL DETAILS

The Ni films were grown in an MBE VG 80 M system with a background pressure <5 x 10^{-11} torr. Ni was evaporated from a 99.9999 % pure source. The deposition rate was 0.5Å/sec. The substrates used in the experiment were 0.5 mm thick, 1 x 1 cm^2 pre-polished MgO (001), which were heat-treated in UHV at 800°C for 1 hr. The combination of flat polished substrates and the UHV heating cycle to allow the surface layers to regain crystalline order has been proven to permit growth of single crystal metal films as well as exhibiting sharp reflection high-energy electron diffraction (RHEED) from the MgO surface.

Prior to initiating the growth, the substrate temperature was lowered to the appropriate deposition temperature for metal growth. Heat transfer was by direct radiation between the heater and MgO substrate. The RHEED patterns were recorded continuously during deposition and during subsequent annealing of the films. The surface morphology of the as-deposited and annealed films was determined *in-situ* with scanning tunneling microscopy [(STM) RHK model STM100]. Our films were typically 10-15 nm thick and the surface morphology of the "as deposited" films was mounded. [Fig. 1(a)] The structural characterization of the films is extensively discussed elsewhere. [1]

We have previously observed that when annealing epitaxial metallic thin films at a temperature 1/3 of the bulk melting point (K) mass transport mechanisms favor surface diffusion thus further enhancing the surface smoothness. [9] Our present studies have shown that in the case of Ni films the surface smoothness is indeed improved with annealing (from 0.5 nm rms to 0.2nm rms). In addition, in the case of (001) Ni, the surface also exhibits periodic "stripe" nano-patterning. [Fig. 1(b)] This periodic stripe nano-patterning is not present in (111) Ni films grown and annealed under the same conditions, [Fig. 1(c)] and therefore the possibility of surface contamination with residual gases to explain it in the (001) films, although not impossible, is unlikely. To understand the periodicity observed in the stripes, we have considered the possible effect of strain. The lattice misfit between MgO and Ni is 16%. However, it has been postulated [6] that an in-plane super-cell matching (commensuration) between the film and substrate with a_o(Ni) x 6 = 2.0446nm and a_o(MgO) x 5 = 2.1066 nm will reduce the misfit to ~0.8%. The critical thickness needed to relieve such a small strain may be quite large.

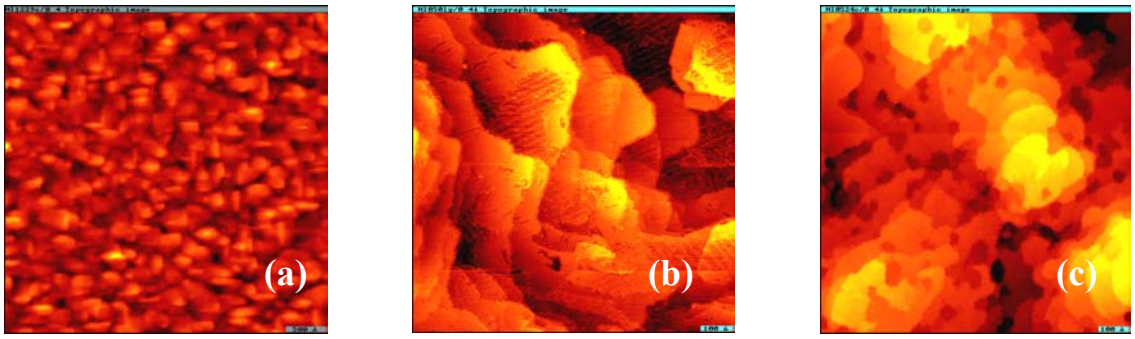

Figure 1. Epitaxial Ni films MBE grown on MgO. (a) (001) Ni film MBE grown on (001) MgO at 373K, not annealed; (b) the same Ni film annealed at 573K (note the "stripe" surface reconstruction); (c) epitaxial (111) Ni film MBE grown on (111) MgO and annealed at 573K, note the absence of surface reconstruction. Bottom scale: 10 nm. [1]

Still, some authors [10] claim that super-cell matching itself cannot give rise to the formation of single crystalline Ni layers, as it has been shown that in other cases interfacial periodic reconstructions can exist that allow for single crystal growth. Our observations seem to support this. Annealing the films may have relaxed the surface and evidenced a reconstruction with periodicity related to the size of the postulated super-cell (i.e. 2.1 nm). Further structural studies including cross sectional TEM are currently under way to confirm this mechanism.

The azimuthal dependence of the magnetization reversal was investigated *ex-situ* with MOKE. Our studies indicate that for epiaxial (001) Ni films that were not annealed the magnetization reversal follows the overall symmetry of the magnetocrystalline anisotropy as indicated by azimuthal plots of the coercive field (Figure 2), while for the annealed films, the azimuthal dependence of the reversal shows superimposed uniaxial anisotropy (Figure 3).

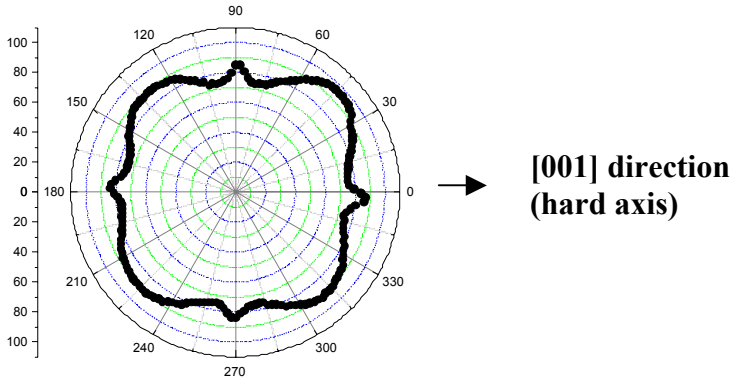

Figure 2. Azimuthal dependence of the coercive field for a 10nm (001) Ni film epitaxially grown on MgO and not annealed. The vertical axis is in Oe. Note the overall four-fold symmetry with "spikes" in coercivity along the hard axes directions.

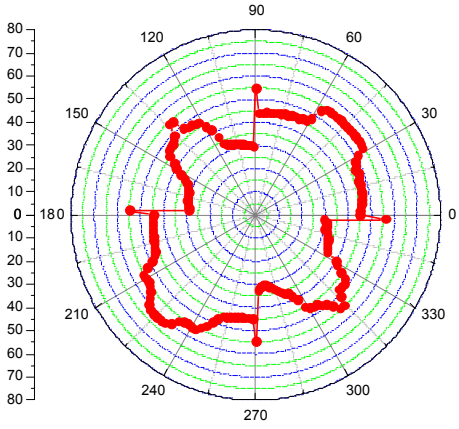

Figure 3. Azimuthal dependence of the coercive field for a 10nm (001) Ni film epitaxially grown on MgO and annealed at 573K. Note the superimposed uniaxial anisotropy.

DISCUSSION

In order to understand the observed results we applied a phenomenological model that we have previously developed [11] starting with the traditional coherent rotation approach. [12] We start by considering the free energy density. For example, for *fcc* structures in the (001) crystallographic orientation we may write:

$$E(\theta_M, H) = K_1 \cos^2(\theta_M) \sin^2(\theta_M) - H M_s \cos(\theta_M - \theta_H) \quad (1)$$

where K_1 represents the magnetocrystalline anisotropy [13] M_s the saturation magnetization, H the external applied field, and θ_M (θ_H) are the angles between the Ni [001] axis and the magnetization (external field), respectively. Stable orientations of the magnetization are found by minimizing $E(\theta_M,H)$. To accurately describe the free energy for each sample, the anisotropy constants were determined from separate experimental measurements using ferromagnetic resonance (FMR). [14]

For the field applied along the easy axis, the simple coherent rotation model predicts switching of the magnetization from <110> to <110)> at $H_c = \pm 2 K_1/ M_s$; however, the observed coercive field is much smaller. Thus we allow the system to form domains such that the moment reversal can occur at lower field by domain-wall nucleation. In this new scenario, we assume that the magnetization may "jump"[15] between two local free-energy minima when the gain in energy exceeds the cost of forming a domain wall:

$$|\Delta E| \geq E_w \quad (2)$$

E_w can be calculated straightforwardly using a standard form [16], which includes the usual exchange and anisotropy contributions. Using experimentally derived values for K_1/M_s (from FMR data), and assuming the bulk value for M_s, we were able to predict the coercive field and found it in close agreement with our experimental values.

To explain the singularities in coercive field observed for the external field applied along a hard axis direction we notice a unique feature for this situation, namely that what was initially a potential local minimum in energy density (for example for $\theta_M = 0^o$) at large H, transforms to a maximum as H is decreased, and two degenerate minima start to develop in close proximity and equidistant from each side of this new maximum. A similar situation occurs for $\theta_M = 180^o$. As mentioned before, this condition only appears for the applied field along the hard axis. Therefore, we see that as the external field is decreased from saturation, the system is no longer stable in the single-domain state, but a continuous transition to a multiple domain state occurs before the magnetization reversal is completed. The system spontaneously breaks up randomly in two types of symmetric domains, with the magnetization in each domain oriented along the direction of one of the two minima at each side of the hard axis direction, akin to a second-order phase transition. As the external field varies from H_{sat} to $-H_{sat}$, our model predicts that the magnetization in each domain type will at first coherently rotate. When the jumping criterion (2) is satisfied (i.e. when the energy gain exceeds the cost of forming a domain wall for each domain) the magnetization jumps to the opposite set of minima, through nucleation of domain-walls. Since twice as many walls must now be nucleated, the energy requirement is higher than it would have been without the symmetry breaking, hence the 'spikes' in the coercive field.

As mentioned above, STM studies on the annealed (001) Ni surface, indicated nano-patterning of the surface through self-assembly of "stripes". Such uniaxial "stripe" reconstructions generally must have two types of energetically degenerate domains, rotated by 90 degrees with respect to each other. Domains where the reconstruction points in 90 degrees different directions can be seen in the STM image [Fig. 1 (b)]. If the thin film sample were composed of a 50/50 mixture of stripe domains where the reconstruction is oriented in the two different symmetry directions, a macroscopic sample will show four-fold symmetric magnetic anisotropy. But if one "stripe" domain dominates (e.g. during annealing one domain grew at the expense of the other), then the net effect will be of a superimposed uniaxial magnetic anisotropy on the four-fold magnetocrystalline anisotropy as indeed we observe. We have used MOKE to study the magnetization reversal, which is a surface sensitive technique and therefore suggests the correlation between the observed surface nano-patterning and the magnetic properties. In addition, other authors have already discussed the importance of monolayer-scale surface features in determining the magnetization reversal. [17] Thus further studies to assert the validity of the proposed mechanism are currently in progress.

In order to simulate the experimental magnetic data, we have introduced a third term in equation (1) to take into consideration a uniaxial magnetic anisotropy. The mathematical form of this term is: $Ku \cos^2(\pi/4 - \theta_M)$ where Ku is the uniaxial anisotropy constant. In order to fit the experimental data, this constant was assumed in our simulations to be one order in magnitude lower than the experimentally determined magnetocrystalline anisotropy (from FMR data), and led to the azimuthal dependence of the coercivity shown in Figure 4.

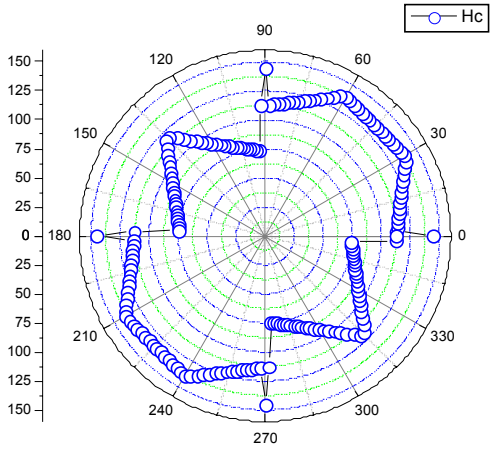

Figure 4. Simulated azimuthal dependence of the coercive field for an annealed (001) Ni film.

We note the remarkable similarity in the main features of the simulated plot with the experimental one validating the model used to describe the magnetization reversal. In addition we also see that these results suggest inverse scaling of the anisotropy constants noticing that the magnetocrystalline anisotropy is related to the crystal lattice constant and

the uniaxial anisotropy constant may be related to the period of the stripe surface reconstruction, which is one order larger than the latter (i.e. the interatomic distance). Further research to establish this correlation is currently on its way.

In conclusion, our simple magnetization reversal model describes the experimentally observed magnetic anisotropy but significant questions remain concerning the correlation between this uniaxial magnetic anisotropy and the observed surface morphology after annealing. Work to determine the underlying cause of the observed magnetic uniaxial anisotropy is currently in progress.

References and Notes

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