Journal of Alloys and Compounds 369 (2004) 213–216

Journal of ALLOYS AND COMPOUNDS

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Surface morphology, structure and magnetic anisotropy in epitaxial Ni films

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Abstract

Understanding the correlation between film structure and its ferromagnetic properties is very important for applications. To this end, we have investigated epitaxial and smooth Ni films grown on MgO substrates using molecular beam epitaxy (MBE) and dc sputtering. To establish correlation between film morphology and structure with magnetic properties, we have used in situ and ex situ scanning tunneling microscopy (STM) images along with studies on the azimuthal dependence of the magnetization reversal utilizing longitudinal Kerr effect (MOKE). Our MOKE azimuthal studies on annealed MBE-grown (0 0 1) Ni films indicate additional uniaxial anisotropy possibly related to surface nano-patterning, superimposed to the expected four-fold symmetry due to magneto-crystalline anisotropy. This uniaxial anisotropy is absent in non-annealed MBE-grown (0 0 1) Ni films as well as sputtered annealed and non-annealed (0 0 1) Ni films. Conversely, neither annealed MBE-grown nor sputtered (1 1 1) oriented Ni films exhibit surface nano-patterning and their structural and magnetic properties exhibit identical azimuthal dependence.
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Keywords: Scanning tunneling microscopy; Molecular beam epitaxy; Anisotropy

1. Introduction

It is already well established that the magnetic properties of epitaxial thin films, particularly the anisotropy [1], are dominated by the crystallographic structure of the film, as well as the metal/substrate interface and the surface morphology [2]. We have previously reported that epitaxial single-crystal magnetic thin films may be used in spin-dependent tunneling applications [3]. For this kind of application, the interfacial roughness must be very small in order to ensure the integrity of the subsequent layers. Thus, one interesting possibility is the growth of magnetic films on MgO substrates, which can be prepared with very smooth surfaces [4]. We have already shown that the epitaxial growth of (0 0 1) Ni on (0 0 1) MgO using molecular beam epitaxy (MBE) is not trivial due to a large lattice mismatch of ∼17% between the metallic epilayer and the underlying substrate [5]. There are several possible mechanisms that may allow the observed epitaxy as well as the distinct surface nano-patterning present on annealed (0 0 1) Ni films, namely interfacial commensuration [6], a network of defects to relief strain, [7] or even stabilization of new faces that minimize the mismatch [8]. To our knowledge, no clear identification of the dominant mechanism responsible for the observed epitaxy in the (0 0 1) Ni/MgO system has yet been reported. We have also shown that (1 1 1) Ni films can be grown epitaxially on (1 1 1) MgO as well, and in this case we have identified a strain relaxation mechanism via defects and dislocations of the crystalline lattice during the early stages of growth. In an attempt to further understand the Ni–MgO epitaxial system, here we report on a comparative study between (0 0 1) and (1 1 1) films obtained using dc sputtering and similar films grown with MBE. We have extended our investigation to include the effect of in situ annealing on the structure and magnetic properties of the films. Our studies involve surface morphology depiction as well as structural characterization of single domain Ni films deposited on MgO substrates and the correlation with magnetic anisotropy during magnetization reversal.

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doi:10.1016/j.jallcom.2003.09.087

2. Experimental

MBE-grown Ni films were deposited in a VG 80 M system with a background pressure $<7\times10^{-9}$ Pa. Ni was evaporated from a 99.999% pure source. The deposition rate was 0.5 Å/s. The film's final thickness in all cases was in the range of 25–50 nm. The substrates used in the experiment were heat-treated in ultra high vacuum (UHV) at 800 °C for 1 h. In situ reflection high energy electron diffraction (RHEED) patterns were recorded continuously during deposition and during subsequent in situ annealing of evaporated films. A discussion of the MBE growth, surface morphology and structural characterization of these films can be found elsewhere [5]. Systematic studies on the optimal conditions for single crystalline growth, determined by monitoring the RHEED pattern in real-time, indicated that the optimal substrate temperature was in the range 100–200 °C for (0 0 1) oriented Ni films and ∼300 °C for (1 1 1) oriented ones.

Sputtered films were deposited in UHV chamber (background pressure $<7\times10^{-8}$ Pa) using dc magnetron sputtering under identical conditions for the growth rate and substrate temperatures as for the MBE growth. In situ annealing conditions were the same for evaporated and sputtered films in both crystallographic orientations. The single crystalline quality of the sputtered films was confirmed via ex situ high resolution X-ray diffraction (XRD) [9]. Ex situ scanning tunneling microscopy (STM) was used to compare the surface morphology of the MBE-grown and sputtered films.

3. Discussion

Various researchers have studied the orientation of Ni films sputtered on MgO substrates using different growth conditions [10,11] and some reports indicate that Ni forms an epitaxial relationship with Ni[0 0 1]//MgO[0 0 1] and Ni(0 1 0)//MgO(0 1 0) for films deposited on MgO substrates held at 100 °C [12]. In addition, Sandström et al. [13] have also shown that at growth temperatures between 300 and 400 °C it is possible to grow smooth ⟨1 1 1⟩ oriented single domain epitaxial films on (1 1 1) MgO substrates, utilizing dc magnetron sputtering in an ultra high vacuum chamber. Here, we present our studies comparing the structure and magnetic properties of epitaxial Ni films deposited under optimized growth conditions using MBE as well as dc sputtering on MgO substrates in (0 0 1) and (1 1 1) crystallographic orientations.

For MBE-grown (0 0 1) oriented films, we observe that the RHEED pattern of the MgO substrate fades immediately when the growth starts while a diffuse pattern corresponding to epitaxial Ni starts to develop. This pattern becomes sharp and well defined only after ∼10 nm of film growth. The epitaxial relationship is [0 0 1] Ni//[0 0 1] MgO, and (0 1 0) Ni//(0 1 0) MgO, indicating single domain crystallographic structure in all azimuthal orientations. In addition, ex situ high resolution XRD confirmed good epitaxial quality for these films with no indication of texture. Conversely, MBE-grown (1 1 1) Ni films, exhibited a streaky RHEED pattern indicative of good epitaxy after only a few monolayers of growth [5].

In situ annealing of the films at mild temperature (one-third of the melting point, which is known to be the optimal temperature to activate surface diffusion) yields significant reduction of the surface roughness, and in addition, appears to induce nano-patterning of the surface via self-assembly of periodic stripes in the (0 0 1) evaporated films (Fig. 1(a)). The rms surface roughness of the as-grown films was 0.5 nm while the rms surface roughness of the annealed films was 0.2 nm, including nano-patterning with periodicity of 2.1 nm. One possible explanation for the observed nano-patterning is in-plane super-cell matching (commensuration) at the interface between the film and

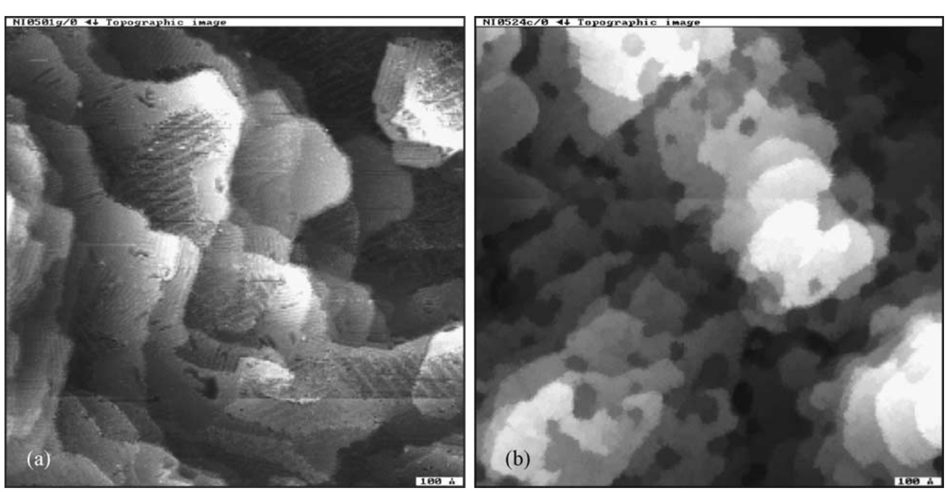

Fig. 1. (a) STM image of a 35 nm (0 0 1) Ni film after annealing (scale bar = 10 nm). We notice stripe surface nano-patterning with two distinct orientations at 90° with one another (scale bar = 10 nm). (b) STM image of the surface of a 30 nm (1 1 1) Ni film (scale bar = 10 nm). We notice vacancy islands in many terraces, associated with dislocations.

substrate with $a_0(\text{Ni}) \times 6 = 2.0446$ nm and $a_0(\text{MgO}) \times 5 = 2.1066$ nm to reduce the misfit to ∼0.8%. Annealing the films may have relaxed the surface evidencing a reconstruction with periodicity related to the size of the postulated super-cell (i.e. 2.1 nm). Conversely, ex situ STM indicated no surface reconstruction on annealed sputtered (0 0 1) Ni films. We attribute this to the fact that sputtered atoms/clusters are deposited with higher energy on the substrate and this may hinder the possibility of motion on the surface to reach lower energy sites, thus affecting the growth mode and posterior relaxation of the surface. Further structural characterization studies using cross-sectional transmission electron microscopy (TEM) are in progress to establish the actual character of the metal/ceramic interface in these films.

We have performed measurements of the magnetization reversal using longitudinal magneto-optic Kerr effect (MOKE). A typical square hysteresis loop along the easy axis for a 30 nm (0 0 1) Ni film is shown in Fig. 2(c). To investigate the in-plane magnetic anisotropy, we performed azimuthal measurements of the coercive field along various in-pane orientations of the applied field (Fig. 2(a) and (b)). As expected, the four-fold symmetry of the magnetic anisotropy in the (0 0 1) oriented samples is revealed in these plots. We have identified that the 'spikes' in coercivity observed along magnetic hard axes arise due to a second order type of transition in the reorientation of the magnetization under the application of an external field. A detailed description of the phenomenological model explaining this mechanism can be found elsewhere [14].

Both types of films exhibit typical four-fold symmetry expected for (0 0 1) oriented *fcc* epitaxial films, but we also notice the superposition of an additional strong uniaxial anisotropy in the case of annealed evaporated films that is absent in the annealed or non-annealed sputtered films. The orientation of this additional anisotropy is coincident with one of the directions in the stripe surface nano-patterning described above for annealed MBE-grown (0 0 1) Ni films. This finding suggests correlation between the uniaxial magnetic anisotropy and the surface nano-patterning, assuming

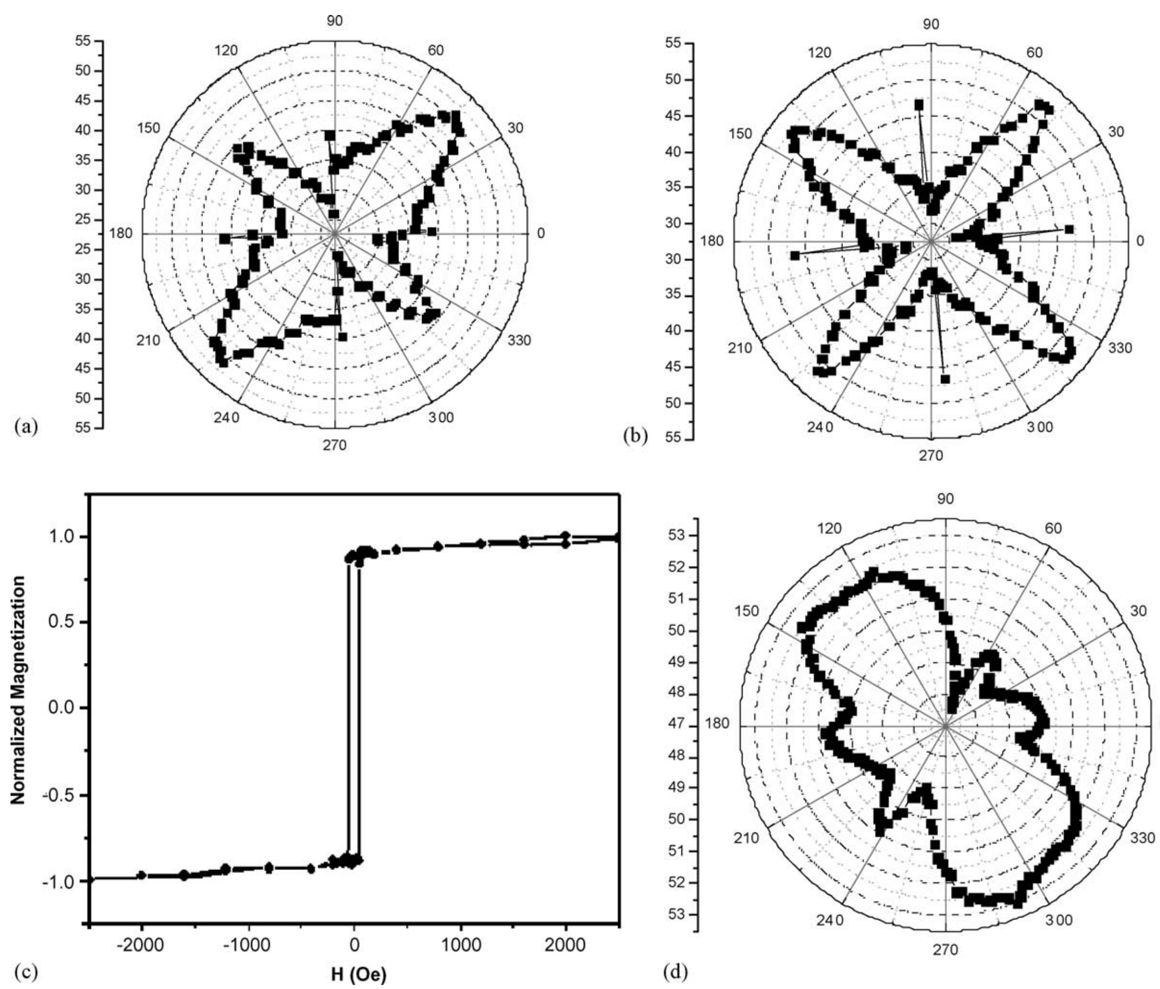

Fig. 2. Azimuthal dependence of the coercive field for a 30 nm (0 0 1) Ni film epitaxially grown on MgO and annealed at 573 K. The vertical axis is in Oe. We notice a significant uniaxial anisotropy superimposed to the four-fold anisotropy. The same graph for a 30 nm (0 0 1) Ni film sputtered on MgO and annealed at 573 K. We only notice four-fold symmetry. Typical hysteresis loop along the easy axis for a 30 nm (0 0 1) Ni film. Azimuthal dependence of the coercive field for a 30 nm (1 1 1) Ni film MBE grown on (1 1 1) MgO. We notice six-fold anisotropy and additional small uniaxial anisotropy due to strain.

that one of the stripe domains prevailed over the other during annealing as appears to be the case according to the STM images. Thus, our magnetic anisotropy studies suggest important differences in the interfacial structure and surface morphology between sputtered and MBE-grown (0 0 1) Ni films, which otherwise exhibit very similar crystallographic structure.

For the case of MBE-grown (1 1 1) Ni films the RHEED pattern indicates single domain epitaxial growth with only the first few monolayers strained with lattice parameter close to that of MgO. The growth proceeds relaxed and RHEED indicates that the subsequent layers exhibit bulk Ni lattice parameter. STM imaging of a 25 nm film (Fig. 1(b)) shows large islands with inner vacancies associated with dislocations. Thus, the rapid strain relaxation was achieved via formation of a large number of defects and dislocations in this case. Ni films were also sputtered under similar conditions on (1 1 1) MgO and their good epitaxial quality was confirmed with ex situ XRD. Our azimuthal MOKE studies indicate that the coercivity and the magnetization switching anisotropy are identical in sputtered and MBE-grown (1 1 1) films of the same thickness. The magnetic anisotropy exhibits six-fold symmetry due to the second order magneto-crystalline anisotropy (we note here that the first order magneto-crystalline anisotropy does not have a projection on the plane of the samples) and superimposed a small uniaxial anisotropy due to strain (Fig. 2(d)). These findings indicate that the surface morphology is less sensitive to the growth mode in the (1 1 1) oriented films.

4. Conclusions

We have been able to grow epitaxial single-domain smooth Ni films on MgO substrates in both (0 0 1) and (1 1 1) orientations using MBE and dc sputtering. Structural characterizations confirm the epitaxial orientation of the films with respect to the substrate previously reported by other authors for samples grown using sputtering techniques [6–10]. We have extended our studies to include the effect of in situ annealing on the structure, surface morphology and magnetic properties of the films. STM imaging of the (0 0 1) annealed Ni surface evidenced formation of a periodic stripe surface nano-patterning only on evaporated films, therefore related to the specific growth mode and structure of the films. The magnetic anisotropy of these films is also significantly affected by in situ annealing showing additional uniaxial anisotropy superimposed to the expected four-fold anisotropy for *fcc* epitaxial (0 0 1) oriented films, possibly related to the observed surface nano-patterning. Further structural and magnetic studies are in progress to further characterize this system.

No surface reconstruction was observed for (1 1 1) Ni films grown on MgO substrates and annealed under similar conditions. In addition, for the (1 1 1) case, strain relaxation occurred during the initial stages of growth via formation of lattice defects and dislocations. No significant difference was observed in the magnetic properties of MBE-grown and sputtered (1 1 1) Ni films.

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