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Exchange bias on epitaxial Ni films due to ultrathin NiO layer

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Abstract. Exchange anisotropy refers to the effect that an antiferromagnetic (AF) layer grown in contact with a ferromagnetic (FM) layer has on the magnetic response of the FM layer. The most notable changes in the FM hysteresis loop due to the surface exchange coupling are a coercivity enhanced over the value typically observed in films grown on a nonmagnetic substrate, and a shift in the hysteresis loop of the ferromagnet away from the zero field axis. A typical observation is that the thickness of the antiferromagnet needs to exceed a critical value before exchange bias is observed. Here we report on the exchange bias properties observed in an epitaxial Ni/NiO system where a thin NiO layer forms spontaneously and is observed after annealing epitaxial Ni films MBE grown on MgO substrates.

PACS. 75.30Et Exchange and superexchange interactions – 75.30Gw Magnetic anisotropy – 75.70Cn Interfacial magnetic properties

1 Introduction

Metal-oxide [1,2] interfaces are of growing interest both from a fundamental and industrial point of view. The Ni/MgO interface is a model system for metal/oxide interfaces studies. All previous experimental work [1–12] agrees that the growth of Ni on MgO(001) at room temperature (RT) results in polycrystalline films. From their results, we readily conclude that the differences in the film morphology and the epitaxial orientation relationships are strongly dependent on different processing parameters such as the substrate temperatures, growth rate, deposition method, etc. Cube on cube (CC) epitaxy combined with a dislocation network, was reported for sputtered films prepared at 580 K and studied by high resolution transmission electron microscopy (HRTEM) [4]. Pure CC epitaxy followed by Ni(110) growth was reported for molecular beam epitaxy (MBE) films prepared between 700 and 900 K as well as at RT [13,14]. Interdiffusion and Ni (111) growth were observed at high temperature and high pressure [5]. A detailed study of the local electronic structure by a combination of experiment and theory has not yet been performed for the Ni/MgO interface. Therefore, it is possible that there is strong hybridization between Ni and O is present at the Ni/MgO interfaces and even NiO formation. We have applied several different techniques to study Ni films grown on (001) MgO substrates using MBE. As we will show below, our findings indicate that an ordered NiO layer forms at the interface between the Ni film and the MgO substrate which induces exchange bias on the adjacent Ni film.

2 Experimental

Ni films were grown epitaxially on MgO substrates in two different systems: an MBE VG 80 M system with a background pressure $<5\times10^{-11}$ torr with in situ RHEED and STM capability and also in a custom-made system adapted for in situ TEM studies. Ni was evaporated from a 99.999% pure source. MgO single crystals, were heat-treated in UHV at 800 °C for 1 hr prior the growth. It was found that the epitaxial growth was strongly dependent on temperature. Thus, prior to the growth the substrate temperature was lowered to the experimentally found appropriate deposition temperature [T = 150 °C for (001) Ni films] [14]. Heat transfer was by direct radiation between heater and MgO substrate. RHEED patterns were recorded continuously during deposition and during subsequent annealing of the films. In order to smoothen the films they were in situ annealed in UHV at 573 K (∼1/3 of the Ni melting temperature) for several hours. In situ RHEED and scanning tunneling microscopy (STM) confirmed surface smoothening. STM imaging also indicated turbulent step-flow features on the film surface, indicating growth defects that originated at the interface [15].

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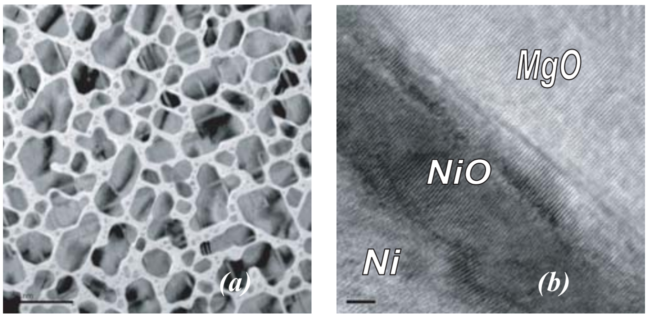

Fig. 1. (a) Plan view, in situ TEM during the early stages of Ni growth on (001) MgO. We observed islands with substantial faceting and no wetting layer yet. Bottom left scale corresponds to 50 nm (b) High resolution cross-sectional TEM image of (001)Ni film MBE grown on MgO and in situ annealed showing a rough interfacial layer between Ni and the MgO substrate. Bottom left scale corresponds to 2 nm.

3 Results

During the early stages of growth, RHEED showed that the MgO pattern faded and double-spots appeared and also faded rapidly. Only after a nominal growth of approximately 5 nm the crystalline Ni pattern emerged. In order to further characterize these early stages of growth additional in situ TEM studies were carried out in a separate MBE deposition system. It was found that the growth is characterized by islands that facet and don't form a full wetting layer until a nominal growth of 5 nm consistent with the RHEED observations and characteristic of highly strained growth (Fig. 1a). We postulate that an ultra-thin NiO interfacial layer is formed to relieve the strain due to mismatch between Ni and MgO. We also postulate that misfit dislocations piled up near the Ni/NiO interface significantly reducing the stress due to lattice mismatch between Ni and NiO [16]. In situ annealed thicker films (30 nm) were further characterized using cross-sectional TEM indicating that a rough interfacial layer (~7–8 nm thick) with crystalline order and lattice constant very close to that of NiO is formed between Ni and the MgO substrate. We also observe a region with high defect density in the adjacent Ni layer [Fig. 1b]. We used longitudinal magneto-optical Kerr effect (MOKE) to study the anisotropy in the magnetization reversal on the samples prior and after in situ annealing. Figure 2 shows the experimental azimuthal dependence of the coercivity in (001) Ni films as-grown (a) and in situ annealed at 300 °C (b). We notice that after annealing significant changes in the magnetic anisotropy are observed, namely the appearance of an additional uniaxial anisotropy superimposed to the expected 4-fold magnetocrystalline anisotropy [17]. We also performed polarized neutron reflectometry (PNR) studies on the films. Figure 3 shows experimental PNR data along with fitting models. The fitting parameters in the models assumed the presence of an interfacial NiO layer of approximately 8 nm thickness, thus consistent with the TEM image.

In order to establish if there was exchange bias in this system we obtained magnetic hysteresis loops (MHL) using longitudinal Kerr effect (MOKE), after field cooling below the blocking temperature for NiO (250 °C) along the [110] direction in the sample. Figure 4 shows the MHL, which in addition to exhibit enhanced coercivity along the film easy axis it also exhibits an exchange field of 16 Oe.

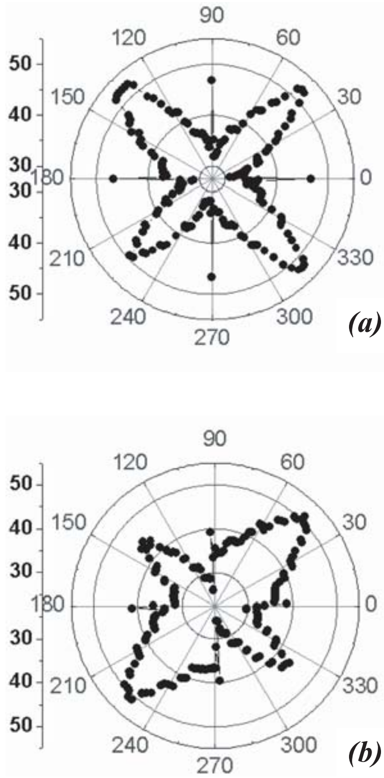

Fig. 2. Azimuthal dependence of the coercivity for (a) as grown (001) Ni film; (b) in situ annealed film. The films are 30 nm thick. The vertical axis corresponds to the coercive field (Oe).

4 Discussion

Both MgO substrate and Ni are materials which are face centered cubic, but have large lattice parameter mismatch (16.4%). We observe that although epitaxial films are obtained for this system despite this large mismatch, the epitaxy cannot be simply explained in terms of strained growth. Moreover recent theoretical investigations predict that Ni should strongly interact with MgO(001) [18,19] with large adhesion energy for Ni clusters (0.62 eV/atom) and strong bonding for an isolated Ni atom (1.24 eV). Thus, we believe that the formation of a NiO interfacial layer is favorable. The lattice constants of MgO and NiO are 4.213 Å, 4.177 Å respectively which has only 0.9% difference, but the constant of Ni is 3.52 Å with 16.4% of lattice parameter mismatch compared to MgO. We postulate that after this very thin intermediate layer is formed the crystal field of the (001) MgO substrate favors the subsequent cube on cube epitaxial Ni growth observed, but with high defect density.

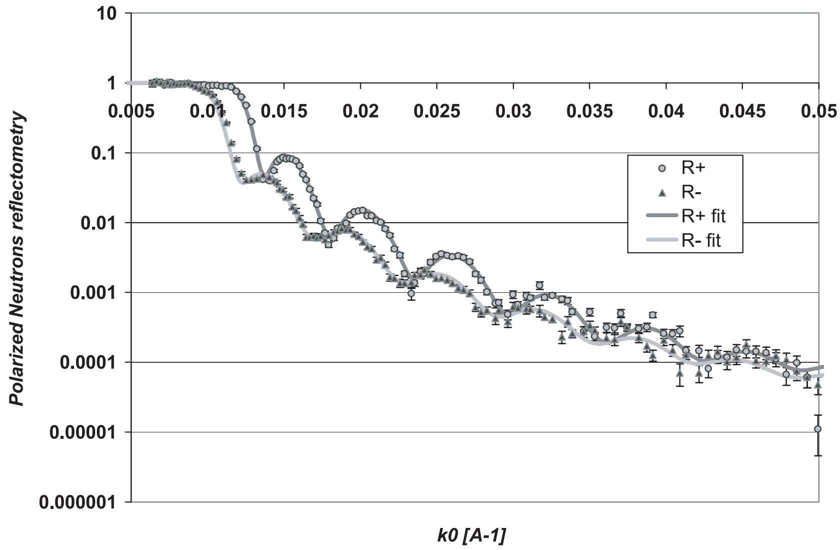

Fig. 3. Polarized neutrons reflectometry (PNR) measured at +/− 1 kOe and 295 K along with fits to the data. The vertical axis corresponds to the normalized intensities. The fits indicate the presence of a rough interfacial layer with scattering length density very close to that of NiO.

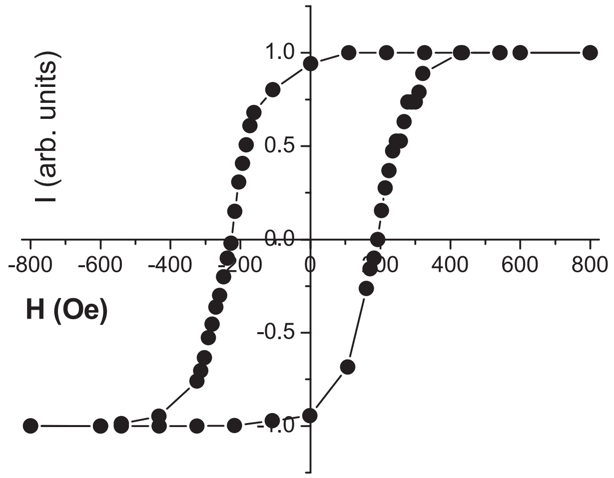

Fig. 4. Magnetic hysteresis loop (MHL) measured with longitudinal MOKE for an annealed and field cooled Ni/NiO/MgO sample along the easy axis. We note that the coercivity is enhanced and the exchange field is 16 Oe.

As mentioned above in situ annealing at 300 °C induces uniaxial magnetic anisotropy in the [011] direction [Fig. 1b]. We note here that a strong uniaxial magnetic anisotropy accompanying a lattice distortion along the [111] direction has been reported in NiO after annealing above its blocking temperature (250 °C) and posterior cooling down to RT [20]. Stress-induced AFM ordering may have occurred so that only one axis was picked in the sample thus breaking the symmetry and the distortion then is along one particular [011] projection of the [111] direction on the (001) plane [16]. Thus, our findings suggest the presence of a few ordered NiO layers at the Ni/MgO interface that grew *fcc* stabilized by the epitaxy on MgO. This NiO interfacial region becomes thicker (approximately 6–8 nm) and tetragonal distorted after further annealing at 300 °C, thus becoming apparent as an additional uniaxial anisotropy in the azimuthal MOKE plots. We speculate that the uncompensated moment of the distorted NiO layer lies parallel to the film surface and leads to a strong interfacial coupling between NiO and Ni inducing measurable exchange bias (16 Oe) on the Ni films as it is observed in Figure 4. Although previous reports indicate that a thickness of at least 10 nm is required for a NiO layer to induce observable exchange bias [21], we believe that the strain present in the self-assembled NiO layer in our samples may affect the interfacial coupling thus enhancing the exchange bias field.

5 Conclusions

From our experimental data we conclude that an ordered NiO interfacial layer is formed in epitaxial (001) Ni films MBE grown on MgO substrates. Our studies indicate a pinning field in (001) Ni films annealed above the blocking temperature for NiO and cooled down to room temperature in the presence of an applied field (10 kOe). PNR data also indicates the presence of this NiO interfacial layer. We postulate that a NiO layer is formed during growth and it becomes thicker during additional annealing at 300 °C. We also postulate that the annealing process is responsible for a lattice distortion in this interfacial layer. These distorted NiO layer may be responsible for the observed additional uniaxial anisotropy as well as the observed exchange bias. Additional PNR studies in (001) oriented samples are also currently in progress to further establish correlation between interfacial structural properties and the magnitude of the exchange bias pinning field.

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