

Ultrahigh vacuum scanning tunneling microscopy/magnetic force microscopy study of ultrathin iron films grown on polycrystalline nickel oxide films

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The thickness dependence of the topographic and magnetic structure of ultrathin Fe films grown on polycrystalline NiO films under ultrahigh vacuum (UHV) conditions was studied to investigate the growth mechanism of the ferromagnetic film and the corresponding magnetic interaction with the antiferromagnetic substrate. Externally prepared NiO films of 60 nm thickness were cleaned by heating in UHV. Ultrathin layers of Fe in the range of 1–27 nm were deposited on top of the NiO film and were analyzed at specific coverages. Iron grows as a polycrystalline film with the grains increasing in size with the thickness. The contours of the underlying NiO crystallites were evident at low coverages but gradually disappeared as the Fe grains coalesced at thicker coverages. Magnetic force microscopy images of the 1 nm thick film show randomly oriented magnetic grains with an average domain size of 30 nm. With an increase in film thickness the size of the domains grows to about 200 nm at 15 nm of iron. At a film thickness of 19 nm cross-tie domain walls become visible, indicating the crossover of some parts of the film from random magnetic grains into continuous domains with in-plane magnetization. A further increase in the film thickness leads to larger in-plane domains, while there are some areas with localized grains on the surface. © 2002 American Institute of Physics. [DOI: 10.1063/1.1447216]

I. INTRODUCTION

Since early suggestions of using ferromagnetic/antiferromagnetic (FM/AFM) bilayer films for device applications,¹ the study of exchange biased ferromagnetic films on nickel oxide is still a topic of considerable interest from both scientific and technological viewpoints.² Considering the great variability in the preparation of NiO films, i.e., crystalline/polycrystalline, small/large grain size etc., it is perhaps not surprising that the mechanism of exchange biasing in this system is still not fully understood.³ In the case of polycrystalline films, progress has been achieved in determining the dependence of the exchange field and the coercivity on the preparation of NiO.^{4,5} The effects of texturing and thickness,⁶ Fe doping⁷ as well as the influence of patterning on both single crystal and polycrystalline NiO have been reported in the literature. However, very little attention has been devoted to understanding the relationship between the microscopic surface topography and the resulting magnetic domain structure. Understanding this correlation could facilitate the verification of the various models of exchange bias, as proposed in the literature.^{8,9} In this work, we present our results on the correlation of the topographic

and magnetic structure of *as grown* ultrathin iron films on a polycrystalline NiO substrate under ultrahigh vacuum (UHV) conditions.

II. EXPERIMENTAL PROCEDURES

The experiment was performed in a commercial UHV chamber equipped with an Auger electron spectroscopy (AES) unit, two FOCUS electron beam evaporators, an argon ion gun as well as a combined atomic force, magnetic force and scanning tunneling microscope. The base pressure inside the UHV chamber was better than 10^{-10} Torr. The nickel oxide substrates were prepared *ex situ* by rf sputtering 60 nm of NiO in the absence of oxygen gas onto silicon wafers with a native oxide layer. The base pressure and Ar pressure were 2×10^{-6} Torr and 2 mTorr, respectively. The NiO films were flushed with isopropanol and subsequently dried with nitrogen gas before being introduced into the UHV system, where further processing steps and the measurements were performed. For additional *in situ* cleaning, the substrates were heated to a temperature of 500 °C and the substrate was examined by AES. The process was repeated until the AES data showed only the signature of nickel and oxygen and was devoid of carbon contamination. Thin iron films were deposited by electron beam evaporation without an external field. The Fe deposition rate was established independently and maintained at 1 Å/min by monitoring the Fe flux. The film thickness, determined from the deposition time, was in-

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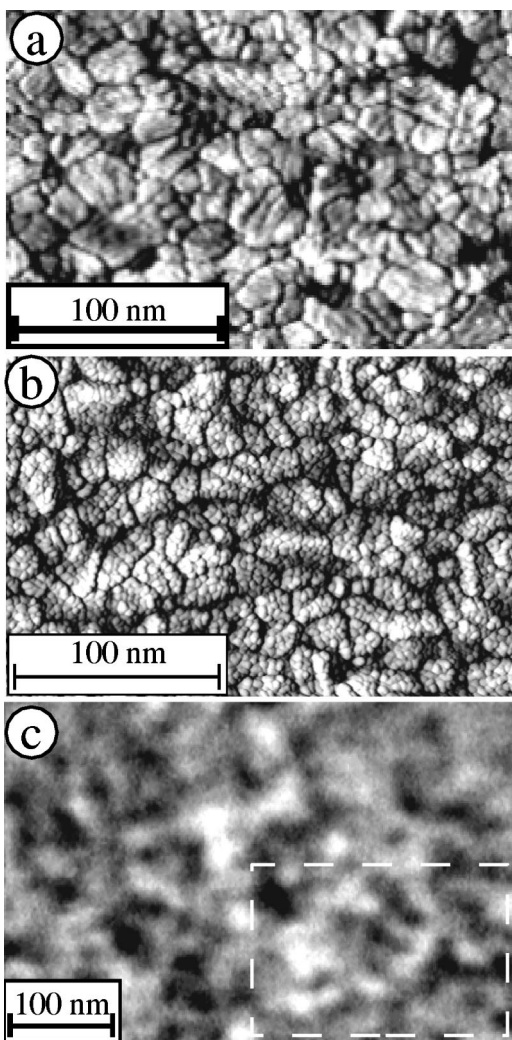


FIG. 1. STM images of pure NiO. (a) ($U_{\text{bias}}=8.5$ V, $I_T=80$ pA) and of a 1 nm thick Fe film on the same NiO substrate. (b) ($U_{\text{bias}}=-1$ V, $I_T=600$ pA). (c) MFM data of the 1 nm thick Fe film. The square in (c) marks the respective sizes of images (a) and (b).

creased in steps of 1–4 nm to obtain films in a thickness range of 1–27 nm. Between deposition steps the films were characterized by scanning tunneling microscopy (STM), atomic force microscopy (AFM) as well as magnetic force microscopy (MFM). The chemical composition of the iron films was also monitored by AES and showed no contamination over the period of measurement. The STM measurements were performed using etched tungsten wires as tips. Prior to use, the tips were cleaned within the UHV system using a home built electron beam heater. The tips for the MFM measurements were prepared *in situ* as well. Regular silicon tips were cleaned by argon ion bombardment for 60 min using a energy of 600 eV for the argon ions or by heating to 150 °C for 60 min. Afterwards, the tips were coated with a thin iron film 5–10 nm thick. The tips were tested on a 200 nm thick permalloy film. The magnetization is oriented perpendicular to the film surface and it shows a distinct serpentine domain pattern with 300 nm wide domains.¹⁰

III. RESULTS

Figure 1 shows STM and MFM scans of the bare NiO

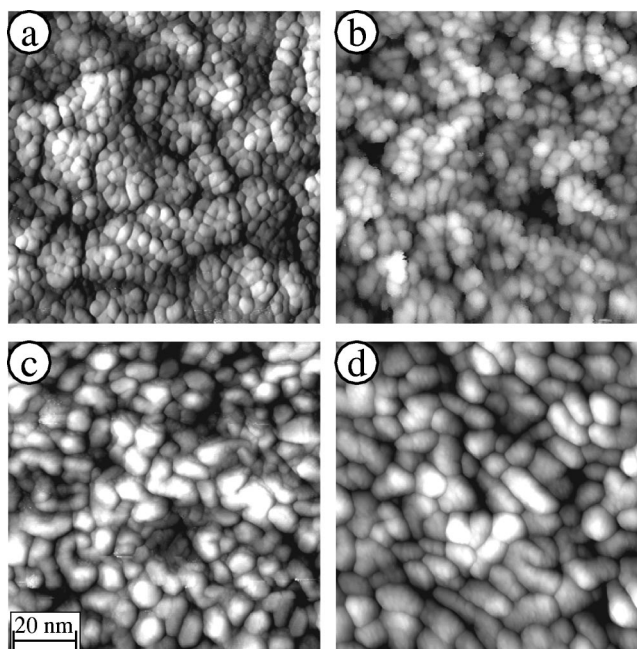


FIG. 2. (a)–(d) STM images of 1, 2, 19 and 27 nm thick Fe films on NiO. Each image is 100×100 nm².

substrate and of a 1 nm thick Fe overlayer, which exhibits the initial stage of film growth. The STM image of the bare NiO substrate [Fig. 1(a)] shows mostly flat crystallites with diameters in the range of 10–30 nm. This is consistent with the crystallites observed by other authors using TEM on similarly prepared oxides, which indicated columnar growth mode.⁴ The surface has an root mean square (rms) roughness of $\langle r_{\text{rms}} \rangle = 0.69$ nm. Figure 1(b) shows the same substrate after the deposition of 1 nm of iron. The iron film forms particles with diameters in a range of 2–4 nm that accumulate on top of the NiO crystallites so that the grain structure of the NiO substrate is still evident. Despite the low coverage of Fe, magnetic features can be detected by MFM, which is shown in Fig. 1(c). While it is hard to discern the individual magnetic grains or the orientation of the magnetization, we find that the size of the structures (i.e., the size of the bright and dark patches) is in the range of 15–45 nm, which is comparable to the size of one or a few of the NiO crystallites. In this determination we assume that some grains could have out-of-plane components.

The development of the morphology of the Fe overlayer with an increase in coverage is shown in Fig. 2. The size of the iron particles increases monotonically with the film thickness up to a diameter of 20 nm. Presumably, Fe particles coalesce as more iron is deposited, which eventually fills the void between the underlying NiO structure. While at a film thickness of 2 nm [Fig. 2(b)] the NiO structure can still be seen, the arrangement of the iron grains seems to be unrelated to the NiO crystallites at higher thickness of 19 nm [Fig. 2(c)] and 27 nm [Fig. 2(d)]. The rms roughness values in Figs. 2(a)–2(d) are $\langle r_{\text{rms}} \rangle = 0.73, 0.75, 0.75$ and 0.87 nm.

The evolution of the magnetic features with an increase in Fe film thickness from 1 to 27 nm is shown in Fig. 3. At the lowest coverage (1 nm) the magnetic structure appears to

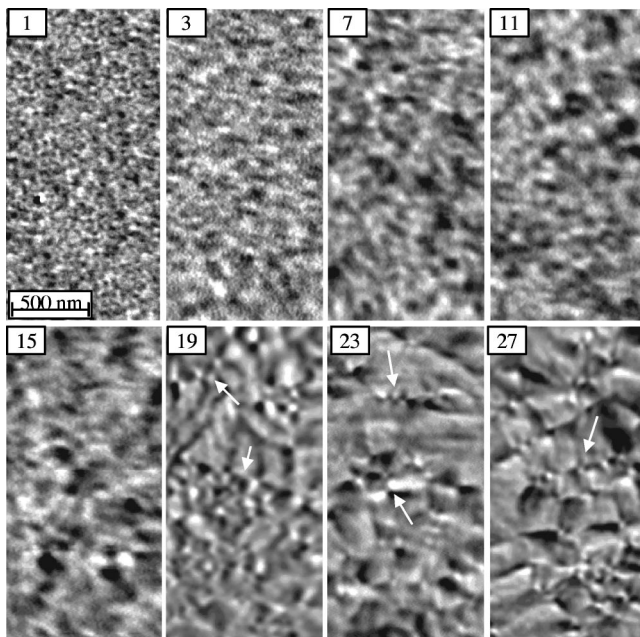


FIG. 3. MFM images of 1–27 nm thick iron films on NiO (each $1 \times 2 \mu\text{m}^2$). The numbers in the upper left corner denote the respective film thicknesses. The arrows in the last three images point to some of the cross-tie domain walls.

consist of small patches of randomly oriented ferromagnetic grains. The characteristic size is on the order of 30 nm. With an increase in coverage, the patches increase in size whereas the density reduces. At a film thickness of 15 nm, the average size of the magnetic features reaches 200 nm. At 19 nm, a dramatic reorientation of the magnetization is observed. Now, evidence of in-plane magnetization is revealed by the presence of cross-tie domain walls. However, there are still areas on the surface that exhibit random patches. This suggests the coexistence of in-plane and out-of-plane magnetization at 19 nm. With a further increase in the thickness of the Fe film the in-plane domains grow in size and we observe a large number of cross-tie domain walls.

IV. DISCUSSION

In the polycrystalline NiO substrate, the magnetization of the individual NiO crystallites depends on the respective crystallographic orientation which, in turn, appears to be random [cf. Fig. 1(a)]. The MFM image of the 1 nm Fe film shows randomly oriented regions whose size is of the order of that of the NiO crystallites. Furthermore, the STM image of the 1 nm thick Fe film [Fig. 1(b)] suggests reduced coupling between iron grains that belong to different NiO crystallites. In other words, Fe–Fe grain interaction is suppressed

by the trenches between the NiO grains. These observations suggest that the magnetization of the 1 nm thick Fe film is governed by the exchange coupling to the respective NiO crystallite. The MFM image thus reflects the magnetic structure of antiferromagnetic NiO.

With an increase in film thickness the trenches of the NiO surface become bridged by the Fe added, so the coupling between previously isolated Fe grains on the NiO crystallites increases. At this intermediate phase (3–15 nm of Fe) the domain structure is still dominated by the magnetic structure of the NiO film. However, neighboring segments with similar magnetic orientation form bigger domains. At a film thickness of 19 nm the magnetic stray field and the coupling within the Fe film become dominant, which forces the overall direction of the magnetization to be within the film. The magnetic structure becomes similar to the structure of a non-exchange biased film. A further increase in the film thickness leads to growing magnetic domains, while in some regions a complex domain pattern remains, probably still supported by the domain structure of the NiO substrate.

V. CONCLUSION

We have observed topographic and magnetic changes of ultrathin Fe films with an increase in thickness in the range of 1–27 nm. The magnetic structure starts out as an arrangement of randomly oriented domains of on average, 30 nm, which mainly reflect the magnetization of the NiO substrate. With growing film thickness the interaction between the iron crystallites increases which leads, up to a film thickness of 15 nm, to larger domains with average size of 200 nm. At 19 nm the magnetization changes to an in-plane domain structure as the magnetic stray field and the Fe–Fe interaction become dominant over NiO–Fe exchange coupling. These results, when taken with complementary measurements of the exchange bias field, could help elucidate the mechanism of exchange anisotropy.

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