

# Magnetic domain structures in Fe<sub>3</sub>O<sub>4</sub> thin films studied by magnetic force microscopy (MFM)

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**Abstract:** A magnetic long-range ordered domain structure was found for the first time on magnetite thin films prepared by molecular beam epitaxy (MBE) on MgO (001) substrates. Annealing was performed for 4 minutes in air. The domain structure deviates significantly from early observations [1-3]. The magnetic stripe-like structure indicates a weak perpendicular anisotropy in magnetite thin films. This perpendicular anisotropy is confirmed by magnetization measurements and supposed to arise from the interface strain between the thin film and the substrate. The analysis of magnetic domain structures shows that these results are in a good agreement with micromagnetic fundamentals.

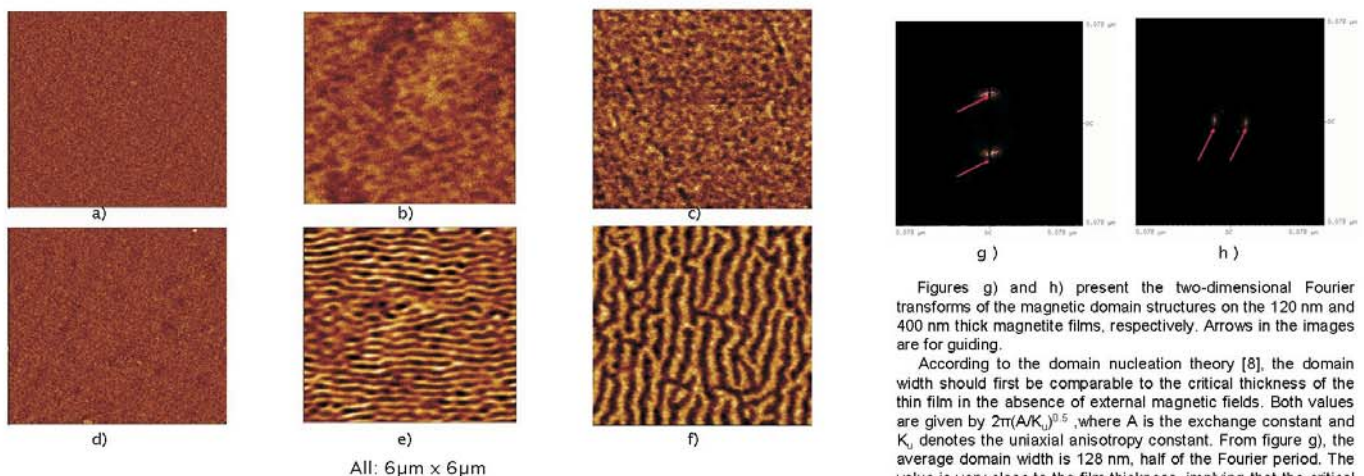
## 1 Introduction

The anomalous magnetic properties of magnetite thin films have attracted extensive interest for both technological and scientific reasons. Much emphasis has been put on single crystalline films grown on MgO (001) by MBE, sputter deposition and evaporation, due to the very similar spacing of the oxygen sub-lattice in MgO and Fe<sub>3</sub>O<sub>4</sub> (about 0.3 % mismatching). In all the magnetite thin films, similar magnetic properties, like very large saturation fields and out-of-plane magnetization in zero-field, were found independently of deposition methods. An anomalous anisotropy in magnetite thin films was detected by Mössbauer magnetospectroscopy [4] and ferromagnetic resonance [5]. Recently, Zhou et al. found that the annealing treatment could have significant effects on magnetization saturation and coercivity of magnetite films [6]. The influence of annealing on the magnetic properties of magnetite films can be detected by MFM. Annealing in air gives rise to a long-range ordered stripe-like domain structure, which is quite different from that of unannealed magnetite thin films [1-3].

In this study, we present MFM results for MBE-grown magnetite thin films with and without annealing treatment. The nucleation of magnetic stripe-like domains is been discussed based on the domain theory for magnetic thin films with a weak perpendicular anisotropy. The effects of film thickness and annealing treatment on the magnetic domain structure have been investigated.

## 2 Experiments and discussion

The magnetite films were epitaxially grown on MgO (001) substrates. The detailed sample preparation is described somewhere else [6]. Samples as-grown and annealed at 250 °C in air were measured by MFM. A cantilever with 30 nm CoCr coating was employed for measurements and the lift height in the MFM mode was 50 nm. MFM measurements were also carried out under external in-plane magnetic fields. Magnetization measurements were performed in both parallel and perpendicular direction to the film plane with a vibrating sample magnetometer.



Figures g) and h) present the two-dimensional Fourier transforms of the magnetic domain structures on the 120 nm and 400 nm thick magnetite films, respectively. Arrows in the images are for guiding.

According to the domain nucleation theory [8], the domain width should first be comparable to the critical thickness of the thin film in the absence of external magnetic fields. Both values are given by  $2\pi(A/K_u)^{0.5}$ , where  $A$  is the exchange constant and  $K_u$  denotes the uniaxial anisotropy constant. From figure g), the average domain width is 128 nm, half of the Fourier period. The value is very close to the film thickness, implying that the critical thickness is about 120 nm. Thus, 20 nm thick films are too thin to nucleate stripe-like domains.

When the films are thicker than the critical thickness, the domain width will be determined by:  $2(2D(A/K_u)^{0.5})^{0.5}$ , where  $D$  denotes the film thickness. From figure h), the domain width of the 400 nm thick film is 222 nm. The calculated value of  $A/K_u$  is a little larger for the 400 nm thick film than for the 120 nm. It might be due to a large anti-phase domain size in thick films [9], giving a larger exchange constant  $A$ .

Figures a), b) and c) are MFM images of as-grown magnetite thin films of 20 nm, 120 nm and 400 nm thickness, respectively. The corresponding MFM images of magnetite films annealed for 4 minutes are shown in Figures d), e) and f).

These images imply that the magnetic domain structures are significantly influenced by film thickness and annealing treatment. For the 20 nm thick films, there is no clear magnetic domain structure since the shape anisotropy is much larger than other anisotropies and it keeps the magnetization of films in-plane [7]. However, magnetic domain structures begin to be nucleated in the as-grown 120 nm and 400 nm thick magnetite films and the structures become long-range ordered stripe-like after a short annealing treatment. Such a stripe-like magnetic domain structure indicates a certain amount of out-of-plane magnetization and a weak perpendicular anisotropy in the thin film [8], which is in a good agreement with the magnetization canting phenomena found in Mössbauer magnetospectroscopy [4] and ferromagnetic resonance [5].

Film treatments (120 nm thickness)	Saturation field in plane (mT)	Saturation field Perpendicular (mT)	Calculated $K_{eff}$ ( $10^4$ J/m <sup>3</sup> )	Calculated angle from the film normal (°)
As-grown	220	506	4.5	9
4 min. annealing	627	637	15.6	20
180 min. annealing	648	687	17.2	25

In the table, the uniaxial anisotropy constants  $K_{eff}$  and the angles deviated from the film normal are calculated in a series of approximation equations [10]. The magnetocrystalline anisotropy constant  $K_1$  is of the order of  $10^4$  J/m<sup>3</sup> and can be neglected as compared to the shape anisotropy of  $1.4 \times 10^5$  J/m<sup>3</sup>. It shows evidently that magnetite films are relatively hard to saturate in the film plane after annealing. The  $K_{eff}$  is increased significantly by annealing and becomes larger than the shape anisotropy, which has been predicted by the strain model [7]. This is consistent with the finding that the annealing treatment could cause a larger strain by reducing the perpendicular lattice parameter [6]. Since the anisotropy is inclined to the film normal, a preferred perpendicular magnetization in annealed films is quite reasonable.

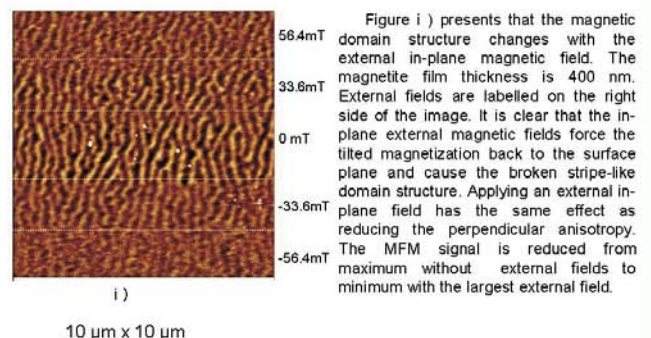


Figure i) presents that the magnetic domain structure changes with the external in-plane magnetic field. The magnetite film thickness is 400 nm. External fields are labelled on the right side of the image. It is clear that the in-plane external magnetic fields force the tilted magnetization back to the surface plane and cause the broken stripe-like domain structure. Applying an external in-plane field has the same effect as reducing the perpendicular anisotropy. The MFM signal is reduced from maximum without external fields to minimum with the largest external field.

## 3 Conclusion

A new magnetic domain structure has been found in annealed magnetite thin films. The stripe-like domain structure indicates a weak perpendicular anisotropy in thin films. This perpendicular anisotropy is confirmed by magnetization measurements. The source of it is supposed to be the interface strain caused by the lattice mismatch between Fe<sub>3</sub>O<sub>4</sub> and MgO single crystals. Annealing enhances domain nucleation. The domain widths are in a good agreement with the theoretical prediction.

## 4 Acknowledgement

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