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Study of thermally evaporated thin permalloy films by the Fresnel mode of TEM and AFM

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ABSTRACT

Thin permalloy films 10 nm and 60 nm thick were investigated. They were thermally evaporated at an incidence angle of 0° in a vacuum of about 10^{-5} mbar. The magnetic structure of the films was observed with the Fresnel mode of transmission electron microscopy (TEM), while their morphological structure was revealed using atomic force microscopy (AFM). The magnetic structure consisted of domains typically 10–30 µm in size. The films were substantially magnetized in the plane of the film. The domain walls of Néel type as well as cross-tie walls occurred in the films 10 nm thick, while in the films 60 nm thick the presence of cross-tie walls was only observed. The presence of cross-tie walls in the films 10 nm thick is reported for the first time. The coexistence of Néel type and cross-tie walls in the films 10 nm thick means that their wall energies are comparable at this film thickness, and this statement is supported by the results of theoretical works. The morphological structure of the films was composed of nanocrystalline grains smaller than about 30 nm in size; the films 60 nm thick had grains somewhat larger in size than the films 10 nm thick. The random distribution of the magnetocrystalline anisotropy of the individual nanocrystalline grains is found to be practically averaged out by exchange interaction, which leads consequently to the strongly reduced effective magnetic anisotropy and the wide magnetic domains on a 10 µm scale.

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1. Introduction

Magnetic films are of large interest from both fundamental and technological points of view. On the fundamental side, they exhibit different magnetic properties, such as magnetic anisotropy, magnetic microstructure, coercivity and magnetoresistance, depending on their thickness, composition, crystalline structure and preparation conditions. From the technological point of view, magnetic films find a wide range of applications in the areas of magnetic and magneto-optic recording and magnetic sensors.

Permalloy is a soft magnetic material used in many applications. In particular, in this context, permalloy films attract large attention and are the subject of extensive studies in recent years. Permalloy films are commonly used in magnetoresistive sensors based on the intrinsic magnetoresistance of the ferromagnetic material (anisotropic magnetoresistance sensors) or on ferromagnetic/nonmagnetic heterostructures (giant magnetoresistance multilayers, spin valve and tunneling magnetoresistance devices). The applications of permalloy films include also magnetic random access memories, soft underlayers in perpendicular magnetic recording, and soft underlayer and interlayers with potential use in multilevel three-dimensional magnetic recording [1–3].

Permalloy films were widely investigated in the past. Nevertheless, studies which reported simultaneously the magnetic and morphological structures of these films were in fact rare. Moreover, in recent years permalloy films can be investigated in more detail, using more advanced and higher resolution techniques (e.g. Refs. [2–12]). For example, Gentils et al. [11] reported the presence of cross-tie walls in 17.5 nm thick permalloy films, while earlier studies showed that the mentioned walls occur for the film thickness in the range from 30–40 nm to 90–100 nm [13,14].

The present paper reports the results of a study of thin permalloy films 10 nm and 60 nm in thickness prepared by the process of thermal evaporation. The magnetic microstructure of the films was imaged using the Fresnel mode of transmission electron microscopy (TEM), and the morphological structure of the films was observed by atomic force microscopy (AFM). The main purpose of the paper has been to determine the types of domain walls in the investigated films. Because of the recent work of Gentils et al. [11], the film thicknesses were chosen just as

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Fig. 1. Images of Néel type walls (a) and cross-tie walls (b) in a permalloy film 10 nm thick, taken with the Fresnel mode of Lorentz microscopy.

10 nm and 60 nm, i.e. in the neighborhood of the film thickness 30-40 nm.

2. Experimental

The specimens under study were permalloy films 10 nm and 60 nm in thickness. They were thermally evaporated at an incidence angle of 0° (with respect to the surface normal) in a vacuum of approximately 10^{-5} mbar. A heater composed of two Al₂O₃ tubes threaded with tungsten wire was used. The films were deposited simultaneously on unheated glass substrates and NaCl crystals; the latter films were prepared for the purpose of studying by TEM. We chose glass as the substrate because glass, besides silicon, is one of the two most common substrates used for device applications [15], due to its non-magnetic nature, smoothness and uniformity.

The magnetic microstructure of the films was observed with the Fresnel (or defocus) mode of Lorentz microscopy [16,17] using a Tesla BS 540 transmission electron microscope. For this purpose, the NaCl substrates were dissolved away in water and then the films were caught on microscope copper grids. Our experiments [18] and those reported in the literature by other authors [19–21] show that the film structure is practically not affected by the specimen preparation method for TEM investigations. Experimental conditions concerning observation of the magnetic microstructure were optimized to improve the magnetic contrast in the images [16]. The morphological structure of the films was made visible by AFM using Omicron and NT-MDT instruments.

3. Results and discussion

TEM is a very powerful method for imaging the magnetic microstructure of thin films [16,17]. It offers high spatial resolution and high sensitivity to small variations in the magnetization, and consequently compares favorably with Bitter pattern technique [22,23]. Thanks to the mentioned features of TEM, the magnetization distribution and the character of domain walls present in the investigated films could be precisely determined.

The magnetic microstructure of the studied thin permalloy films was composed of domains. The typical sizes of the domains were in the range of $10-30\,\mu\text{m}$. The magnetization of the films was observed to lie essentially in the film plane; only in small regions within the cross-tie walls the magnetization was found to have an out-of-plane component.

In general, both Néel type and cross-tie walls occurred in the films 10 nm thick. This is evidenced in the images of Fig. 1, recorded by the Fresnel mode of TEM. The images in Fig. 1a and b show the presence of Néel type and cross-tie walls, respectively. The domain walls are visible as black and white lines. In case of the cross-tie walls, the cross ties serve to decrease the demagnetizing energy and are imaged as short bars perpendicular to the main wall (Fig. 1b). The cross-tie walls represent a transition between Néel and Bloch type walls.

It is well known that in thin magnetic films the type of domain walls is dependent on the film thickness [11,13,16]. In fact the energy (and consequently the character) of a domain wall in a thin film is most strongly influenced by the demagnetizing factor perpendicular to the plane of the film. Earlier studies of permalloy and cobalt films [13,14] showed that in films thinner than 30-40 nm the domain walls are of Néel type (in which the magnetization rotates in the plane of the film, and which is accompanied by free magnetic poles at the wall, inside the film), while in films thicker than 90–100 nm the domain walls are of Bloch type (in which the magnetization rotates in the plane of the domain wall, and which is accompanied by free magnetic poles at the intersection of the wall with the surface of the film). For the film thickness in the range from 30-40 nm to 90-100 nm cross-tie walls were found, the first interpretation and the name of which were given by the authors of Ref. [24].

The cross-tie wall has a lower energy than the simple 180° Néel wall because it consists mainly of energetically favorable 90° walls. Although the total 90° wall area is 3–4 times larger than the original 180° wall area, the total energy decreases because the specific



Fig. 2. Image of cross-tie walls in a permalloy film 60 nm in thickness, recorded by the Fresnel mode of TEM.



Fig. 3. AFM images of the morphological structure of 10 nm (a) and 60 nm (b) thick permalloy films.

energy of 90° walls is smaller than that of 180° walls by a factor that overcompensates the larger wall area of the cross-tie wall [13]. At first sight, the occurrence of cross-tie walls in 10 nm thick permalloy films, reported for the first time in the present paper, may be recognized as somewhat unexpected. In this context, however, it should be noted that recently cross-tie walls were also observed in 17.5 nm thick permalloy films in Ref. [11]. The reason for the coexistence of Néel type and cross-tie walls is that their wall energies are comparable at the mentioned film thicknesses. This statement is supported by the results of the theoretical works of Metlov [25,26] and Redjdal et al. [27].

According to the theoretical papers of Metlov [25,26], the film thickness corresponding to the transition between the Néel and cross-tie walls is given by the formula $L_{tr}^{(M)} \approx 2.3 \sqrt{A/(\mu_0 M_s^2)}$, where *A* is the exchange constant, *M*_s is the saturation magnetization and $\mu_0 = 4\pi \times 10^{-7}$ V s/(A m) is the vacuum permeability. Using standard material parameters for permalloy ($A = 8 \times 10^{-12}$ J/m, $M_s = 8.6 \times 10^5$ A/m [8]), one obtains $L_{tr}^{(M)} \approx 7$ nm. Whereas on the basis of the micromagnetic simulations performed in Ref. [27], Redjdal et al. found the film thickness for the Néel to cross-tie walls transition to be $L_{tr}^{(R)} \approx 14$ nm. As a consequence, these theoretical predictions are in very satisfactory agreement with our experimental observations of the coexistence of Néel type and cross-tie walls in the films 10 nm thick.

In the films 60 nm thick only cross-tie walls were present, in accordance with the earlier experimental results reported in the literature [13,24]. An example is presented in Fig. 2, which shows the image taken using the Fresnel mode of Lorentz microscopy. Note that the cross ties in the films 60 nm thick extended to considerably larger distances from the main wall and were less widely spaced than the cross ties in the films 10 nm thick (compare Fig. 2 and Fig. 1b). This is a consequence of the fact that the influence of free magnetic poles becomes larger as the film thickness increases.

Owing to the fact that the Fresnel mode of TEM is very sensitive to small variations in the magnetization, a ripple structure of the magnetization could be seen in all the images recorded (cf. Figs. 1 and 2). The ripple structure is caused by local variations of the magnetic anisotropy and reflects the irregular polycrystalline nature of the investigated films [11,28]. For magnetostatic reasons, the local magnetization direction is found to be stronger influenced by magnetic anisotropy variations perpendicular to the average magnetization of the film than parallel to it. In other words, the ripple contrast is predominantly perpendicular to the average magnetization direction and hence the magnetic easy axis [11,28]. Fig. 3a and b presents AFM images of the morphological structure of the permalloy films 10 nm and 60 nm in thickness, respectively. It is seen that the morphological structure is composed of grains, as expected. The sizes of the grains are at the nanometer range. In the films both 10 nm and 60 nm thick the grains are packed closely, i.e. the films are seen to be practically continuous, in agreement with the results obtained in Refs. [11,15]. The films 60 nm in thickness have grains somewhat larger in size than the films 10 nm thick. Note also that the root mean square (rms) values of the surface roughness were 0.2 nm and 0.5 nm for the films 10 nm and 60 nm thick, respectively.

Because the size of magnetic domains is much larger than the grain size of the films, thus each magnetic domain contains a lot of nanocrystalline grains. The exchange length is determined by $(A/K)^{1/2}$, where A is the exchange constant and K is the magnetic anisotropy constant (the magnetocrystalline anisotropy constant in the case of the studied films). For permalloy films the exchange length is found to be about 300 nm [13], i.e. it is considerably larger than the grain size of the studied films, the latter size being smaller than about 30 nm (cf. Fig. 3). This in turn proves that the random distribution of the magnetocrystalline anisotropy of the individual nanocrystalline grains is practically averaged out by exchange interaction. The grain size is so small that the magnetization cannot be oriented along the magnetic easy axis in each individual grain because the exchange energy and the magnetostatic energy at the grain boundaries would become too large [29]. As a consequence, the exchange interaction and the magnetostatic effect cause that the total effective magnetic anisotropy is strongly reduced in comparison with the magnetocrystalline anisotropy. And a decrease of the magnetic anisotropy results in increasing the width of magnetic domains, as demonstrated for example in Refs. [30-33]. In case of the studied permalloy films, the result of the strongly reduced magnetic anisotropy is the domain structure with wide domains on a $10\,\mu m$ scale, as can be seen in Figs. 1 and 2.

4. Conclusions

Using the Fresnel mode of Lorentz microscopy and AFM, a study was made of the magnetic and morphological structures of permalloy films 10 nm and 60 nm thick, deposited by thermal evaporation at an incidence angle of 0° in a vacuum of approximately 10^{-5} mbar. The morphological structure of the films was composed of nanocrystalline grains smaller than about 30 nm in size; the films were found to be practically continuous. In comparison with the

films 10 nm in thickness, the films 60 nm thick possessed grains somewhat larger in size.

The magnetic structure showed the presence of domains typically 10–30 μ m in size. The nature of domain walls was determined and a ripple structure of the magnetization within the domains was observed with high contrast and clarity. Both Néel type and crosstie walls were observed in the films 10 nm thick, and in the films 60 nm thick only cross-tie walls occurred. The presence of cross-tie walls in the films 10 nm in thickness was reported for the first time. The coexistence of Néel type and cross-tie walls in the films 10 nm thick means that their wall energies are comparable at this film thickness. This experimental finding confirms the predictions of the theoretical works of Metlov [25,26] and Redjdal et al. [27]. The films both 10 nm and 60 nm thick were magnetized in the plane of the film, except for small regions within the cross-tie walls in which the magnetization was found to be out-of-plane.

As the grain size of the studied films is significantly smaller than the exchange length of about 300 nm for permalloy, the random distribution of the magnetocrystalline anisotropy of the individual nanocrystalline grains is found to be practically averaged out by exchange interaction. As a consequence, the exchange interaction in connection with the magnetostatic effect leads to the strongly reduced total effective magnetic anisotropy in comparison with the magnetocrystalline anisotropy, and thereby to the magnetic domain structure with wide domains on a 10 µm scale.

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