JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. **5** No 4, 04016(4pp) (2013)

ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том **5** № 4, 04016(4cc) (2013)

The Study of Anisotropy and Domain Condition of Permalloy Thin Films

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(Received 01 October 2013; revised manuscript received 10 November 2013; published online 10 December 2013)

Hysteretic Loops, 1,5; 3,0; 4,5; 6 and 10 nm in thickness, obtained by magnetron sputtering of Ni81Fe19 alloy, were measured by means of vibration magnetometer. It has been detected that by increasing the film thickness from 3 to 10 nm coercive force (HC) increases as well. In the direction perpendicular to the axis of easy magnetization the loop form considerably differs from the right-angled one, which is caused by amplitude dispersion of anisotropy. The films, derived in scattered magnetic field of the Earth, are by magnetic parameter isotropic. The results of atomic force microscope investigation indicate to the granular structure of films and confirms the presence of non-magnetized areas among the examined films. The critical thickness at which permalloy films pass from multi-domain to single-domain state was 10 nm. Estimation of the critical thickness of the transition from single-domain state to superparamagnetic led to the values of 1.5-2 nm.

Keywords: Permalloy thin films, Magnetometer, Hysteretic loop, Coercive force, Single-domain particles, Superparamagnetic state.

PACS numbers: 75.70.Ak, 75.50.Bd

1. INTRODUCTION

Physical properties including magnetic properties of any material in the form of a film are substantially different from those in a massive state. The film sizes are limited to one direction, that is why, in the absence of induced anisotropy and in such a sample geometry, there becomes energetically favorable such a state in which moments of magnet, in the absence external magnetic field, lay in the film plane. This leads to such magnetic materials obtaining in-plane magnetic form anisotropy. In this case, all the directions in the film plane are equivalent. When applying a magnetic film in an external magnetic field there arises an easy magnetic axis in the film plane, due to the preferred magnetic moments alignment in the film plane along external magnetic field (longitudinal magnetic anisotropy).

Thus, the films may obtain some kind of anisotropy, which has considerable influence on the processes of remagnetization of thin film samples. Formation of all kinds of anisotropy depends on various factors: the film size and structure, crystal structure of the material, impurities and defects, elastic strains, external effects and etc.

In this respect it is of scientific interest to determine critical thickness, below which the film turns from a ferromagnetic state to superparamagnetic one (SP). Earlier in the process of Co films study it was shown that their magnetic order remains to a thickness of 0,7 nm [1]. Above all, there is a problem of high importance - the problem of determining the critical thickness at which the film turns from multidomain (MD) state to a single-domain state. (SD).

2. MATERIALS AND METHODS

The films 1,5; 3,0; 4,5; 6 and 10 nm were obtained with the help of ion-plasma sputtering of cathode from permalloy (19 % Ni and 81 % Fe) and condensation to a cold glass base in argon atmosphere at the pressure of residual gases equal to 1-10 Pascal. The film thickness was controlled by the time of desspositing with an accuracy of 0.3 nm by a known method [1, 2]. With this method of film condensation there occurs bulk island (granulated) structure [2].

The films disposition was carried out in the scattered magnetic field (without magnetic insulation of the Earth's field) and in the longitudinal magnetic field of 120 Oersted, which was created with the help of permanent magnet.

The hysteresis loops were measured with the help of high-sensitive vibration magnetometer [3], developed by V.I. Maksimochkin at the geophysics department of Moscow State university. Absolute Measurement Error equaled to not more than 5 microampere per meter at the minimal division value of this device being 2 5 microampere per meter. The measurements were carried out at the room temperature and external magnetic field intensity up to 600 Oersted repeatedly. The results in digital form were recorded into the computer memory, and then there was performed mathematical processing to determine parameters of the hysteresis loop.

3. THE STUDY'S RESULTS AND DISCUSSION

The film samples were subjected to magnetic measurements on a glass substrate in the form of squares 10 per 10 mm. When measuring magnetization and the hysteresis loops the film samples were placed in the measuring cell of the vibration magnetometer and then orientated relatively to the direction of the magnetizing field.

When magnetizing along the axis of easy magnetization the loop shape is nearly rectangular and saturation is achieved in magnetic fields with a strength of 30-50 A/m. The magnetization saturation in a

The article was reported at the International Conference «Physics and Technology of Nanomaterials and Structures», Kursk, 21-22 November, 2013

perpendicular direction is achieved for some of the films only at the strengths of 500-600 A/m. It was also found that the thinner the film is, the saturation state is achieved at lower intensities (Fig. 1).

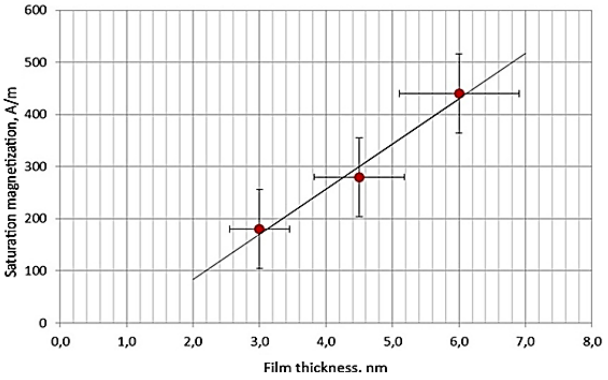

Fig. 1 – Dependence of saturation magnetization of the film thickness obtained by condensation in the magnetic field of 120 Oersted

The loops, measured parallel to the axis of easy magnetization, have a high coefficient of squareness $K_S = J_r/J_s \approx 1$, their coercive force (H_c) is 1,5 – 3 times less (depending on a film thickness) than that of the loops measured perpendicularly to the axis of easy magnetization. The coefficient of squareness, as well as the remnant magnetization of the latter is considerably lower $K_S = J_r/J_s = 0.3 \div 0.7$.

Experimental data analysis shows that the films obtained in a magnetic field of 120 Oersted, exhibit anisotropy of the parameters, which is conditioned by the formation of the easy magnetization axis at their condensing in a magnetic field of a permanent magnet. At the same time, the experimental data indicate the absence of parameters anisotropy in the films obtained in the scattered magnetic field of the Earth.

Attention should also be paid to a marked decrease in the saturation magnetization of the films with the films' decreasing thickness. This is likely due to the granularity of the films studied and due to the presence of non-magnetic areas inside the films and on their surface (voids spaces, surface irregularities, and nonmagnetic impurities, etc.). Another possible reason for decreasing the IS is associated with the approach of the film thickness to a certain critical value of the film transfer to a superparamagnetic state , the presence of which is confirmed by the absence of hysteresis loops for films with thickness of 1.5 nm. The transition state "ferromagnetic – paramagnetic" even at such small thicknesses should be observed at higher temperatures.

The coercive force decreases significantly with decreasing the permalloy film thickness from 195 A/m at a thickness of 10 nm to 15 A/m at a thickness of 3 nm (Fig 2). This result is relevant to the results of coercive force measurements in the research [4], in which it was found that with decreasing the film thickness of permalloy from 110 nm to 10 nm, the coercive force increased from 120 A/m to 197 A/m. The authors of the mentioned research did not study the films with less thickness. The analysis of our results and the results obtained in [4], suggests that the critical size for single-domain state of permalloy films is about 10 nm (Fig. 2).

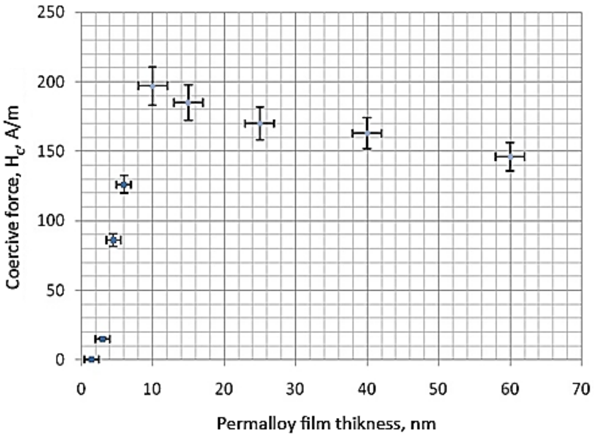

Fig. 2 – Dependence of the coercive force on the permalloy films thickness

As the Fig. 2 shows, the thickness at which the coercive force has maximum points is about 10 nm. With further decreasing of the film thickness there occurs approximation to superparamagnetic state, which, judging by the results of our measurements, occurs when the thickness of the studied films of permalloy is 1.5-2 nm (Fig. 1 and 2).

Many magnetic parameters of thin films are smaller than that of the materials in solid form. Deviations from magnetic parameters of bulk samples, in all cases, are the stronger - the smaller the film thickness is.

Decrease of the films magnetic moment was previously associated only with reduced thickness while maintaining the film magnetization. Our results suggest another reason for decrease in the films magnetization.

The results of atomic power microscopy showed [2, 5, 8] that a permalloy thin film consists of islets with an average size of from 2 to 6 nm, depending on the film thickness and deposition conditions.

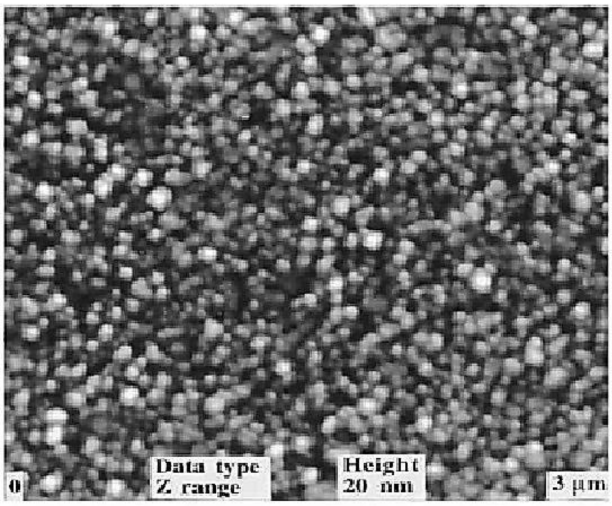

Fig. 3 – Atomic-power micrographics of the the permalloy films surface [8]

The grain sizes are proportional and close in values to the film thickness. Grains are an extremely small distances from each other (2.1 nm). The set of ferromagnetic particles of such small size may have superparamagnetic properties. The value KV (K is the constant of the magnetic anisotropy of the material, and V is the volume of the particle) of such particles may be similar or less than kT. That means that magnetisation vector, remaining equal on terrain clearance quantity $M_s \cdot V$, where M_s – the saturation magnetisation of the massive sample, which will be affected by the temperature fluctuations with characteristic time which is much smaller than the measurement time.

The magnetization of superparamagnetic particles has been studied in many papers for various nanoparticle systems in which the distance between the particles in all cases were large in comparison with the size of the particles, and therefore, the magnetic dipole-dipole interaction was not cosidered. In this conditions the dependence of such particles magnetization from the external field and temperature can be described by the Langevin function, which for small external magnetic field ($M_S VH \ll kT$) takes the form $M/M_s = M_S VH/3kT$, and for intense fields ($M_S VH \gg kT$) is expressed as $M/M_s = 1 - 3kT/M_S VH$, where M – magnetization of the system, and H – external magnetic field intensity. In [6], the dependence of saturation magnetization of antiferromagnetic nanoparticles is taken into account, which resulted in better agreement between theoretical calculations and experimental results. In this case, the dipole interaction between the particles is absent not only because of the great distances, but also because of the small magnetic moments of the separate antiferromagnetic nanoparticles.

At the analysis of effects of examination of the thin films consisting from magnetic nanoparticles, located in short distances (1-3 nanometers) it is necessary to consider interaction influence between moments of magnet of separate particles of a ferromagnetic. For each separate particle of a polycrystalline film such interaction can be viewed approximately as the interior magnetic field created by moments of magnet of neighbours. Such interior magnetic field will be peak when moments of magnet of all particles are oriented in one direction. If directions of the moments are proportioned in a random way it disappears because of spatial averaging. For some predominant orientations of the particle magnetic moments intensity of the internal field can be approximately determined [7] expression $H_i = H_D M/M_s$, where M / M_s – the relative magnetisation of system (film), a H_D – the field strength of the dipole interaction reached at orientation when the $M/M_s = 1$. Then intensity of an effective magnetic field in a film will be $H_r = H + H_D M/M_s$, where vectors H_r, H and H_D lie on the film plane and directed along H. In the first approximation, the relative magnetization of the system of nanoparticles can be expressed by the generalized Langevin function:

$$M/M_s = \mathrm{th}\alpha\,, \qquad (1)$$

Where $\alpha = M_S VH_r/kT = \{M_S V[H + (M/M_s)H_D]\}/kT$ – argument of Langevin function (the dimensionless parameter).

This expression can be written in a view:

$$\frac{M}{M_s} = \frac{T}{\theta}\alpha - \frac{H}{H_D}, \qquad (2)$$

where $\theta = M_S VH_D/k$ – blocking temperature.

Equations (1) and (2) expressing the relationship between M, T and H in a thin film consisting of islands of ferromagnetic material. The relationship between these values is most vividly represented in graphical form as the dependence of the ratio M/M_s on the parameter □ (Fig. 3). Equation (1) on this graph corresponds to the Langevin curve. For different values of the external field H and a fixed temperature equation (2) corresponds to a series of parallel lines with a slope $kT/M_S VH_D$.

Measurements of the film magnetization at specified temperature and external magnetic field intensity allow us to estimate a couple of parameters: the average grain volume, blocking temperature, the field strength of the dipole interaction. For example, the ratio of the external magnetic field to the dipole interaction field H/H_D determines the distance between points of intersection with the axis M/M_s and the origin. Out of the graph it is easy to define the H_D for a given value of H, if M_S is measured (achieved the state of the film magnetic saturation). From the slope of the line is possible to estimate the average volume of ferromagnetic particles in the thin film.

Magnetic state of the film (ferromagnetic or superparamagnetic) also can be determined by these graphs. At $H = 0$ line passes through the origin. According to the temperature this line may cross or not the Langevin curve. The slope of tangent to the hyperbolic tangent curve in the point corresponding to the origin, is equal to one therefore the intersection point will exist if $T < \theta$. If $T > \theta$ the spontaneous magnetization of the islands on the film will not exist in the absence of an external field (a superparamagnetic state). In this area, graphic dependences of the relative magnetization M / M_S of $H / (T - \theta)$ will be overlaid for all temperature values. However if $T < \theta$ then the film with island structure will experience the spontaneous magnetization (ferromagnetic state), if $H = 0$ it appears that $M/M_s > 0$. The field required for saturation has to be very large in the case where T is slightly smaller than θ.The film will magnetize to saturation even in low fields (like the large ferromagnetic body) only at very low temperatures.

The value of the characteristic temperature θ depends on the islets and the field intensity H_D. To roughly estimate the field dipolar interaction in theory, let us assume that it is comparable to a field in a spherical cavity inside a single domain [7] of the studied material. Since the film is not plane, but consists of separate islets, it is logical to introduce a "packing coefficient" that expresses an amendment to the density of the ferromagnetic material in the film and is equal to $k = 1 - \pi\Delta h/(6h)$, where Δh is an average size of the non-magnetic areas of the film, a h is an average film thickness. Then

$$H_D \approx k\frac{4}{3}\pi M_S. \tag{3}$$

According to our estimates, this efficient is equal to 2/3 for the films of thickness equal to 3.0 nm. Then, taking for calculation the value of M_S = 50 Oersted, measured by us for this permalloy sample, we get H_D ~ 140 Oersted. Calculations carried out on the experimental magnetization curves and graphics have led to similar values of the dipolar magnetic field strength.

If we take a blocking temperature equal to θ = 300 K, the permalloy film consisting of islets, the volume of which is less than $V = k\theta/M_S H_D = 8{,}0 \cdot 10^{-27}\, m^3$ will appear superparamagnetic and its behavior and the temperature range T > 300 K will be described by the Langevin function. The mentioned volume meets the linear size of the islet of about 2 nm. This calculation is consistent with the data of magnetic measurements on vibrating magnetometer, which showed no spontaneous magnetization and hysteresis loops for the film with an average thickness of 1.5 nm.

For the islets with a linear size of 3 nm, which are most often observed in the studied permalloy films with an average thickness of 3 nm, according to the calculations, the value of θ is already about 600 K.

4. CONCLUSION

1. The calculations show that the initial magnetic susceptibility is directly proportional to the square of the saturation magnetization and, ultimately, in accordance with the theory of ferromagnetism depends on the thickness of the permalloy film.

2. It is determined that with decreasing the film thickness squareness and residual magnetization of the loops decreases and the hysteresis disappears for the films with a thickness of ~ 1.5 nm. This means that the films rest ferromagnetic till a thickness of ~ 2.3 nm. With further decreasing of thickness the magnetic order disappears and there arises a superparamagnetic state of the film. That allows us to estimate the critical size of superparamagnetism 1.5-2 nm.

3. The coercive force decreases with decreasing the film thickness. Analysis of the obtained results and the research [9] show that the critical thickness dimension of the film at which the permalloy grain stay single-domain, is approximately 10 nm. At this thickness, the films coercivity maximum.

4. It has been found that the studied films have sufficiently high dispersion of anisotropy which increases while the thickness is decreasing. And the key role in anisotropy plays the amplitude dispersion.

5. It is shown that the main influence on the reduction of the saturation magnetization has an increase in the proportion of non-magnetic areas of the film with decreasing of its thickness. Basing on the results of measurements for permalloy films with different thicknesses there was evaluated a medium-size of non-magnetic areas. It is approximately $\Delta h \approx 2$ nm.

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