Effect of Magnetic Field on the Morphology and Structural Characteristics of Cobalt-based Thin Film Systems as Sensitive Sensor Elements

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(Received 25 May 2023; revised manuscript received 21 August 2023; published online 30 August 2023)

This work presents the results of a study of the influence of an external magnetic field on the surface morphology of three different thin-film magnetic structures were presented: a single-layer Co film, a granular film alloy based on Co and Ag, and a trilayer Co/Gd/Co film. Atomic force microscopy investigations at room temperature revealed a significant influence of the magnetic field on the surface morphology of the films and their structural characteristics (arithmetic mean Ra, root mean square Rq, structural entropy S). After the initial application of the magnetic field (at H = 0.01 T), a decrease in the surface roughness (Rq, Ra) was observed for all systems, which is associated with the destruction of system defects in the film samples and the transition of the structure to a new, more stable state. The most substantial reduction in roughness, with Rq decreasing by 19% and Ra by 16%, was observed in the granulated magnetic films based on Co and Ag. Further increases in the magnetic field practically had no effect on the structural characteristics, which remained constant even after relaxation over 18 hours. In the case of the single-layer Co films, with a subsequent increase in the magnetic field strength to 0.05 T, the values of structural characteristics (Rq, Ra, S) monotonously increased. Further intensification of the magnetic field to 0.1 T almost did not affect Rq, Ra, S. For the trilayer Co/Gd/Co film system, further increasing the magnetic field intensity to 0.1 T led to an increase in the roughness parameters Rq and Ra due to additional stresses in the upper Co layer. This effect was a result of the lower coercive force in the bottom Co layer compared to the upper Co layer. As a result, the lower Co layer magnetized at lower magnetic fields compared to the upper layer, which further increased the roughness of the film's surface.

Keywords: Sensor, Giant magnetoresistance, Granular thin films, Spin-dependent scattering, Electron beam evaporation; Co-evaporated technique, Magnetic aftereffect phenomenon.

DOI: 10.21272/jnep.15(4).04034

PACS numbers: 75.47.De, 75.70.Cn

1. INTRODUCTION

Magnetic film materials, exhibiting unique physical effects, are interesting for both the perspective of fundamental research and for practical applications. In nanosystems based on magnetic materials, it is possible to realize effects that are not achievable in homogeneous bulk materials. Magnetic film systems may be utilized in applications in spintronics (energy-independent magnetic memory elements and sensors), automotive engineering (rotation speed sensors and angle/position detectors), and biomedicine (biosensors, bi-molecular detection systems) [1].

The structures characterized by giant magnetoresistance (GMR) and tunnel magnetoresistance (TMR), perpendicular magnetic anisotropy [2, 3], and unique magneto-optical effects have gained the most popularity in the field of spintronics. The GMR effect is realized in multilayer films, multilayers, and granulated magnetic systems based on magnetic materials (e.g., Co, Fe, Ni, etc.) and non-magnetic materials (e.g., Ag, Cu, Au, etc.). The GMR effect is a result of spin-dependent scattering of electrons in conductivity at the interfaces of the magnetic and non-magnetic layers or magnetic granules and non-magnetic matrix.

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The development of functional elements based on magnetic film systems is accompanied by a number of unresolved issues caused by the lack of definitive information regarding diffusion processes [4], phase formation, and structural transformations under the influence of various physical factors, leading to changes in the physical properties of the entire system. One of these factors is the influence of a magnetic field on thin film samples. For instance, passing a spin current through thin magnetic films can result in a high-gradient magnetic field due to the small dimensions of these elements [5]. It was found that the magnetic field can affect the surface morphology and impact the structural characteristics of the film [6, 7]. The effects of magnetic field influence and magnetic after-effects, which significantly modify the structure and properties of nanomaterials, should be taken into account for designing nano-electronic devices operating under the influence of magnetic fields. Even minor changes in the structure or surface morphology of the films can lead to device failure [8]. Understanding such effects will ensure the stability of spintronic devices.

The effects of magnetic field influence on the physical properties and structure of bulk materials and thin
film systems was analyzed in works [6, 9, 10]. For instance, the influence of constant and variable magnetic fields on the yield strength, ultimate strength, stress relaxation, and other mechanical characteristics in bulk magnetic and non-magnetic materials was studied in work [9]. Special attention was given to investigating the positive and negative magnetoplastic effects under mechanical loading, depending on the intensity of the magnetic field. The influence of the magnetic field on the morphology and structural characteristics of magnetic thin film systems with a thickness of \( d \leq 50 \) nm was studied in the works [6, 11]. However, there are still several questions that require resolution.

This study is focused to experimental investigations of the influence of a magnetic field on the surface morphology and structural characteristics of a single-layer Co film, granulated thin film samples based on Co and Ag alloys, as well as a three-layer film Co/Gd/Co.

2. EXPERIMENTAL DETAILS

The film systems that were studied in this work were obtained by the electron beam evaporation in vacuum at a pressure of residual gases \( P = 10^{-4} \) Pa. The polished sheets of glass-ceramic (sitol) were used as substrates. The thickness of the film samples during the deposition was monitored by the quartz resonator method.

The deposition of films using the electron-beam evaporation method in a vacuum offers several significant advantages compared to other methods [8]. Firstly, the formation of nanostructures occurs in high vacuum, resulting in films with minimal oxide and nitride phases. Secondly, the thickness of material deposited by electron beam sputtering process can be easily controlled by quartz resonator method. Thirdly, adjusting the deposition parameters make possible to obtain the desired structure, ranging from amorphous to crystalline. The electron-beam evaporation method allows the production of metallic (magnetic and non-magnetic), semiconductor, and dielectric thin film materials.

Film alloys samples based on Co and Ag in the composition range (20 \( \leq c_{Co} \leq 80 \), where \( c \) is the concentration of \( c_{Co} \) in at.%) were obtained by co-evaporation from two independent evaporators [11]. The metal deposition rate was 0.2 nm/s for Co and 0.4 nm/s for Ag. The composition of the films was calculated by the method described in [11].

Three-layer Co/Gd/Co/S films (where \( S \) is the substrate) were obtained by layer-by-layer condensation on a substrate heated to a temperature \( T_0 = 460 \) K. The metal deposition rate was 0.2 nm/s for Co and 0.1 nm/s for Gd. The deposition of metals on a heated substrate allowed to obtain more structurally perfect samples with fewer defects.

The study of magnetic field influence on the change in the surface morphology of the films was carried out by atomic force microscopy on a Solver Pro device using a two-pass scanning technique. NSG01/Co cantilevers were used in the research.

The AFM method was used to scan a selected area of the film surface without an applied magnetic field and in a magnetic field from 0.01 T to \( H_{max} \leq 0.1 \) T. The magnetic field was directed in the film plane. In some cases, the processes of sample surface relaxation were studied after turning off the magnetic field and holding the sample without a field for a certain time. An analysis of the obtained images of the film surface morphology suggests that, due to the presence of a vibration-protective platform in the design of the Solver Pro microscope, it was possible to avoid displacement of the studied area during the research. In addition, to ensure scanning of the same section of the film, we snapped to control points on the surface of the sample.

The Solver Pro device was controlled and the results of the surface morphology and structural characteristics of the surface were processed using the Nova program. The study investigated the influence of a magnetic field on changes in the morphology and structural characteristics, such the height of the highest peak \( h_{max} \), the depth of the deepest cavity \( h_{min} \), the arithmetic mean \( R_q \) and the root mean square \( R_a \) surface roughness and structural entropy \( S \) of the films.

The arithmetic mean \( R_q \) of the surface is the average of the modulus of the measured deviations of the height of the points on the sample surface from its mean value. The roughness measurement characterizes changes in the height of the irregularities on the sample surface. The surface characteristic is calculated by the ratio:

\[
R_q = \frac{1}{N} \sum_{j=1}^{N} |r_j|
\]

The roughness height was calculated by the ratio:

\[
R_q = \sqrt{\frac{1}{N} \sum_{j=1}^{N} r_j^2}
\]

where \( N \) is the number of points on a AFM profile of the sample surface image; \( r_j = z_j - \bar{z} \) the values of the deviations of the \( j \)-th point height on this profile from its mean value.

The structural entropy \( S \) characterizes chaotic distribution of heights. The structural entropy \( S \) is derived from the Shannon ratio. For the case of the pixel intensity distribution \( I_i \), \( i = 1, \ldots, N \) is defined as:

\[
S = -\sum_{i=1}^{N} I_i \log I_i
\]

The crystal structure and phase state were investigated by TEM-125K transmission electron microscope. Samples for TEM were obtained by deposition of thin films on TEM grids coated with carbon film.

3. RESULTS AND DISCUSSION

The paper presents the results of research on the effects of magnetic field exposure on the morphology and structural characteristics of single-layer Co films, granulated thin film samples based on Co and Ag alloys, and three-layer Co/Gd/Co films. From the perspective of forming many physical properties of the films, their structural and phase states are crucial. Therefore, before studying the influence of magnetic fields on the morphology and structural characteristics of the films, their structure and phase composition had been analyzed.
3.1 Structural-phase States Investigations

The phase composition of single layer cobalt films is described in sufficient detail in ref. [12]. Cobalt films are chemically inactive, so oxidation processes are unlikely. Unannealed Co films have nanocrystalline structure and consists of two phases: hcp-Co + fcc-Co. The lattice parameter fcc-Co is $a = 0.355$ nm, and for hcp-Co it is $a = 0.251$ nm, $c = 0.410$ nm [13]. The presence of reflections from the crystallographic planes (111) and (200) fcc-Co is explained by stacking faults in hcp-Co.

The structural-phase state of granular film alloys based on Ag and Co has also been studied in detail. For example, in ref. [6, 11], the structural-phase state of film alloys with a thickness of $20 \leq d \leq 85$ nm was described in detail in a wide range of component concentrations $15 \leq c_{Co} \leq 80$ at. %. According to the phase diagram, the system based on Co and Ag [14] is two-component, since Co and Ag do not dissolve in each other. In a film system based on Co and Ag, a granular structure is formed, and the components have a clear separation limit.

It has been established that the structural-phase state of samples of film alloys based on Co and Ag depend on the concentration of the system components [11]. Thus, at low concentrations of Co in the Co-Ag alloy, the structural-phase state of the samples corresponded to hcp-Co granules with a size of $L = 3-22$ nm and were located in the fcc-Ag matrix. Granules of size $L = 2-5$ nm can combine into agglomerates of irregular elongated shape due to coalescence. Fig. 1 shows a TEM image of the nanostucture and diffraction pattern for a sample of a film alloy based on Co and Ag with a concentration of $c_{Co} = 39$ at. % in unannealed state. The sample thickness was $d = 35$ nm. The interpretation of the electron diffraction pattern showed that no solid solutions were formed in the film alloy samples. Since the film samples were unannealed, they are in a nonequilibrium state characterized by an increased content of various structural defects.

An increase in the concentration of Co in film alloys leads to the growth of magnetic granules to a size of $L = 5-30$ nm and their association into clusters. The gaps between the ferromagnetic clusters are filled by fcc-Ag. A further increase in the Co concentration (approximately to $c_{Co} = 80$ at. %) leads to the coalescence of clusters and the formation of a ferromagnetic film, the phase composition of which corresponds to hcp-Co.

The study of the structural-phase state of unannealed Co/Gd/Co/S films [13] showed lines of hcp-Co, fcc-Co and halo originating from quasi-amorphous Gd at a thickness of the Gd interlayer $d < 10$ nm. When the thickness of the Gd layer increases ($d > 10$ nm), the phase composition of the three-layer films becomes corresponding to hcp-Co + fcc-Co + hcp-Gd.

3.2 Influence of the Magnetic Field on the Structural Characteristics of Films

The effects of a magnetic field on the film surface morphology were studied by atomic force microscopy. The films were exposed to a magnetic field applied in the plane of the film at room temperature. The magnetic field was successively increased from 0 to 0.1 T.

The results of changing the surface morphology of a single-layer cobalt film 35 nm thick under the action of a magnetic field are shown in Fig. 2. As it can be seen from Fig. 2, the structure of the film surface is locally inhomogeneous. Such inhomogeneities can be associated with the method of obtaining film samples. Under the action of a magnetic field, inhomogeneities are displaced along the film surface (Fig. 2). The analysis of the obtained AFM images of the film surface allows us to state that there was no displacement of the film area under study at the moment the magnetic field was switched on and off. A series of studies showed that the structural characteristics of the surface of the cobalt film were extremely sensitive to the first impact of a magnetic field (Fig. 3). The results of the action of a magnetic field on the structural characteristics of the surface of an unannealed thin cobalt film 35 nm thick are shown in Fig. 3. After the first application of a magnetic field with a strength of $H = 0.01$ T, the arithmetic mean film surface roughness $R_{a}$ decreased by 6 %, and the root mean square roughness $R_{q}$ decreased by 8 %. This may be due to the decay of defects in hcp-Co. At the same time, the value of the structural entropy $S$ and the height of the highest peak $h_{\text{max}}$ remained almost unchanged. With an increase in the magnetic field $H$ to 0.05 T, the values of the structural characteristics $R_{a}$, $R_{q}$, $S$ monotonically increased, and at a field strength of $0.05 \leq H \leq 0.1$ T, they remained almost unchanged.

AFM studies of the surface morphology of samples of Co-Ag film alloys showed that the surface of the samples is locally inhomogeneous at the nanoscale. The surface structure of the samples was formed by Co granules and agglomerates based on them. It was the granule agglomerates that form the roughness of the granular surface of the film.

For a sample of granular film alloy Co$_{39}$Ag$_{61}$ upon the initial application of a magnetic field $H = 0.01$ T, a decrease in roughness $R_{a}$ by 19 % and $R_{q}$ by 16 % was observed (Fig. 4). The structural entropy of the system decreased by 4.5 %, and the height of the highest peak decreased fourfold. These changes remained nearly unchanged with gradual increments in the magnetic field intensity, step-by-step up to $H_{\text{max}} = 0.1$ T. Additionally, during the relaxation process (18 hours after field deactivation), the structural characteristics of the sample remained unchanged. Such an effect of the magnetic field on the surface structure of the granulated magnetic thin film system can be explained as follows.

In the initial unannealed state, the sample of the granular film alloy was characterized by an increased content of various structural defects. Even the first influence of a magnetic field with a strength of $H = 0.01$ T...
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Fig. 2 — 2D and 3D images of the surface morphology of Co thin film (d = 35 nm) before the effect of a magnetic field (H = 0 T (a, a’), under the action of a magnetic field H = 0.01 T (b, b’), H = 0.03 T (c, c’), H = 0.06 T (d, d’), H = 0.1 T (e, e’)).

Fig. 3 — Dependence of roughness values Ra and Rq of Co thin film (d = 35 nm) on the magnitude of the applied magnetic field.

Fig. 4 — Dependence of roughness values Rq and Rq of a thin film alloy Co39Ag61 (d = 35 nm) on the magnitude of the applied magnetic field.

Fig. 5 — 2D and 3D images of the surface morphology of thin film alloy Co30Ag70 (d = 85 nm) before the effect of a magnetic field (H = 0 T (a, a’), under the action of a magnetic field H = 0.01 T (b, b’), H = 0.03 T (c, c’), H = 0.06 T (d, d’), H = 0.1 T (e, e’)).

The study of the structural state of samples of granular film alloys showed that the shape of the agglomerates of Co granules is extracted, and in some cases even characterized by pointed edges (Fig. 1). When a magnetic field was applied to the tips of these agglomerates, magnetic fluxes are localized, i.e., high-gradient fields are concentrated. Thus, magnetic fluxes at the tips of Co granule agglomerates could increase by an order of magnitude. In this case, gradient amplifications of exchange local magnetic fields appear on structural inhomogeneities. These gradient fields, focused at sharp granules, interact more effectively with an applied external magnetic field than in the case of homogeneous structures. At the nanolevel, this affects the change in the nanostructure when an external magnetic field is applied. Fig. 5 showed the dynamics of changes in the surface morphology of an unannealed sample of a granular Co30Ag70 film alloy 85 nm thick before and after the application of a magnetic field of 0.01 T, 0.03 T, 0.06 T, 0.1 T. It is evident from the figure, the surface morphology of the sample changes with the variation in the intensity of the magnetic field.
In three-layer Co/Gd/Co/S films, the roughness is generally determined by the surface roughness of the upper Co layer. The study of the influence of the magnetic field on the structural characteristics of such films is of interest, since it was established [8] that the electrical conductivity of multilayer films depends on their magnetic domain structure of the surface roughness.

![Figure 6](image)

**Fig. 6** – Dependence of roughness values $R_a$ (a) and $R_q$ (b) of three-layer film Co(5)/Gd(5)/Co(25)/S on the magnitude of the applied magnetic field. In brackets, the film thickness is indicated in nm.

![Figure 7](image)

**Fig. 7** – The influence of the applied magnetic field on the change in the magnetic domain structure of the surface Co(5)/Gd(5)/Co(25)/S film at 0 T (a), 0.01 T (b), 0.015 T (c), 0.02 T (d), 0.06 T (e), 0.1 T (f).

As it can be seen from Fig. 6, during the initial application of the external magnetic field ($H = 0.01$ T), the roughness parameters $R_a$ and $R_q$ decrease. This behavior is analogous to what occurred in the single-layer Co film and in the samples of granulated Co-Ag alloys and may be attributed to the breakdown of the defect system in the film sample.

An increase in the intensity of the magnetic field caused an increase in the roughness parameters $R_a$ and $R_q$. The reason for the increase in roughness under the influence of a magnetic field may be the occurrence of additional stresses in the upper Co layer of the three-layer Co(5)/Gd(5)/Co(25)/S film.

The emergence of such stresses can be caused by a change in the upper Co layer of the magnetic domain structure, as shown in Fig. 7.

The upper Co layer had higher coercivity compared to the lower Co layer. This is confirmed by the results of work [15], which investigated the magnetic properties of single-layer Co films of different thicknesses. Then, the upper Co layer had a different direction of the easy magnetization axis compared to the lower magnetic layer. Under the action of the magnetic field, the domain structure of the lower layer changes, and, as a result, the surface roughness of the three-layer film increases.

4. CONCLUSIONS

The influence of a magnetic field on the surface morphology and structural characteristics (arithmetic mean $R_a$, root mean square $R_q$, structural entropy $S$) of three types of magnetic systems was experimentally investigated: single-layer Co film, granulated alloy based on Co and Ag, and three-layer Co/Gd/Co film. It was established that the initial application of an external magnetic field had the most significant influence on the change of structural parameters in the samples, specifically reducing the roughness parameters $R_a$ and $R_q$. This behavior of structural parameters is attributed to the breakdown of the defect system in the film samples under the influence of the magnetic field, leading the system to a new, more stable state. The largest influence of magnetic fields on the surface morphology and structural parameters of the film was observed in the samples granulated Co-Ag film alloys. For instance, during the first application of an external magnetic field in the granulated Co-Ag films, a reduction in roughness parameters $R_a$ by 19%, $R_q$ by 16%, structural entropy $S$ by 4.5%, and the height of the highest peak by 4 times were observed. In the case of homogeneous magnetic film structures, the effect was less pronounced. For the single-layer Co film, a decrease in roughness parameters $R_a$ by 6%, $R_q$ by 8%, and structural entropy $S$ by 1.3% was observed. A further increase of the magnetic field practically did not affect the structural characteristics of the Co-Ag film system, which remained constant even after relaxation for 18 hours. However, the surface morphology of the sample changed with the variation of the magnetic field intensity. In the single-layer Co films, with the application of a magnetic field of $0.01 < H < 0.05$ T, the values of structural characteristics $R_a$, $R_q$, and $S$ increased monotonically. A further increase of the magnetic field strength to 0.1 T had almost no effect on the values of the structural parameters. For the three-layer Co/Gd/Co film system, the application of an external magnetic field with intensities of $0.01 < H < 0.1$ T led to an
increase in surface roughness parameters \( R_s \) and \( R_d \) in the sample. This could be caused by the occurrence of additional stresses in the upper Co layer of the three-layer film due to the difference in the coercive force of the magnetic layers.

ACKNOWLEDGMENT

This work was funded by Project No. 9918 Magnetism in Ukraine Initiative. I. Shpetnyy acknowledge the EU NextGenerationEU financial support through the Recovery and Resilience Plan for Slovakia under the project No. 09I03-03-V01-00044. S. Vorobiov acknowledges the financial support provided by the Slovak Research and Development Agency under the contract APVV-20-0324. I. Shpetnyy and Yu. Shkurdoda would like to thank the Wolfgang Pauli Institute and personally Professor Norbert Mauser for their partial financial support under the WPI Thematic Program “Mathematics-Magnetism-Materials” (2021/2022).

REFERENCES


Вплив магнітного поля на морфологію та структурні характеристики тонкоклівкових систем на основі кобальту як чутливих елементів сенсорів

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У роботі представлені результати досліджень впливу зовнішнього магнітного поля на морфологію та структурні характеристики тонкоклівкових систем на основі кобальту як чутливих елементів сенсорів.

Матеріалом для продемонстрації альтернативного шляху використання магнітного поля на морфологію тонкоклівкових систем є магніт різних типів, в основному на основі Co та Ag та тришаровий шар Co/Gd/Co. Дослідження методом атомно-силової мікроскопії при кімнатній температурі показали суттєвий вплив магнітного поля на морфологію тонкоклівкових систем. При перших включений магнітного поля (при \( H = 0.01 \) Т) для всіх систем спостерігалось зменшення шириноси поверхні \( R_s \), що зумовлено розкладом системи дефектів у півкових зразках та перехідом структури зовнішнього шару. Вартість зменшення шириноси поверхні \( R_s \), що зумовлено розкладом системи дефектів у півкових зразках та перехідом структури зовнішнього шару, коливається від 19 до 16% зростається у грундувань магнітних півкових шарах на основі Co та Ag. Подальше збільшення магнітного поля практично не впливало на структурні характеристики, які зазналися стабілізації після релаксації протягом 18 годин. У свою чергу, в одноклівкових шарах Co, при подальшому збільшенні магнітного поля \( H \) до 0.05 Т значення структурних характеристик \( R_s \) монотонно збільшувалося. Подальше збільшення напруженості магнітного поля до 0.1 T майже не впливало на \( R_s \). У тришаровій шаруватій системі Co/Gd/Co подальше збільшення напруженості магнітного поля до 0.1 T спричинило зростання параметрів щорогових \( R_s \) та \( R_d \) в рахунок виникнення додаткових напруг за верхнім шаром Co. Такий ефект виникає на рахунок зменшення змінної кінетичної сприйнятності нижнього шару Co, порівняно з верхнім шаром Co. В результаті зазначені шари перетворюються в менших магнітних полях порівняно з верхнім шаром, що додатково підвищує щороговість поверхні півкових зразках.

Ключові слова: Сенсор, Гігантський магнітоімір, Гранульовані тонкі шари, Спін-залежне розсіювання, Електронно-променева випаровування, Одночасне осадження, Магнітна післядія.