Металлофиз. новейшие технол. / Metallofiz. Noveishie Tekhnol.
2015, т. 37, № 11, сс. 1477–1486
Оттиски доступны непосредственно от издателя
Фотокопирование разрешено только
в соответствии с лицензией

PACS numbers: 68.37.Ef, 75.50.Ss, 75.60.Lr, 75.70.Ak, 75.70.Rf, 75.80.+q, 85.70.Kh

Nanoscale Surface Deformation of the Granular $Co_{25}Ag_{75}$ Films

O. Yu. Gorobets[*,**], Yu. I. Yakymenko[**], A. F. Kravets[*],
O. M. Brukva[**], V. N. Zakharchenko[**], S. V. Mikhalko[*], I. A. Sizon[**]

[*]*Institute of Magnetism, N.A.S. of Ukraine and M.E.S. of Ukraine,*
36[b] Academician Vernadsky Blvd.,
03142 Kyiv, Ukraine
[**]*National Technical University of Ukraine 'KPI',*
37 Peremogy Ave.,
03056 Kyiv, Ukraine

Granular nanostructure $Co_{25}Ag_{75}$ films are analysed by means of the scanning tunnelling microscopy (STM) method. The phenomenon of giant film nanodeformation is detected with the 10% change of average height during the initial magnetization of the sample and with the 2% change after 8 days of relaxation during secondary magnetic field action. The changing of the shape of nanoscale granules and their displacement in the surface of film are observed during the process of nanodeformation under the action of magnetic field and in the aftereffect processes.

Key words: thin CoAg film, magnetostriction, magnetic aftereffects, magnetic deformation, nanodeformation.

Методою тунельної мікроскопії досліджено вплив магнетного поля на плівки ґранульованих наноструктур $Co_{25}Ag_{75}$. Було виявлено ефект гігантської нанодеформації з 10% зміною середньої висоти зразка впродовж первинного намагнетування і 2% зміною після 8 днів релаксації під час повторного намагнетування. Зафіксоване явище пов'язане із деформацією магнетних ґранул та їх зміщенням у немагнетній матриці під дією магнетного поля.

Ключові слова: тонка CoAg плівка, магнетострикція, магнетик після

Correspondence author: Igor Anatolievitch Sizon
E-mail: igor-sizon@mail.ru

O. Yu. Gorobets, Yu. I. Yakymenko, A. F. Kravets, O. M. Brukva, V. N. Zakharchenko, S. V. Mikhalko, and I. A. Sizon,
Nanoscale Surface Deformation of the Granular $Co_{25}Ag_{75}$ Films, *Metallofiz. Noveishie Tekhnol.*, **37**, No. 11: 1477–1486 (2015).

впливу, магнетна деформація, нанодеформація.

Методом туннельной микроскопии исследовано влияние магнитного поля на плёнки гранулированных наноструктур $Co_{25}Ag_{75}$. Был обнаружен эффект гигантской нанодеформации с 10% изменением средней высоты поверхности образца в течение первичного намагничивания и 2% изменением после 8 дней релаксации во время повторного намагничивания. Зафиксированное явление связано с деформацией магнитных гранул и их смещением в немагнитной матрице под действием магнитного поля.

Ключевые слова: тонкая CoAg плёнка, магнитострикция, магнетик после воздействия, магнитная деформация, нанодеформация.

(Received September 2, 2015)

1. INTRODUCTION

The intensively research of magnetic granular films (GF) which contain nanoscale magnetic granules, distributed in metal nonmagnetic structure began after discovery giant magnetic resistance effect in these granules [1]. Grained Co_xAg_{1-x} alloys with the highest magnetoresistance at room temperature (up to 25% in a magnetic field of 10 kOe) and giant magnetostriction among other granular systems occupy important place among GF. Probably, these effects are connected with special structure features of these materials, which have a limited solubility. The granular structure containing the Co granules, which are distributed in nonmagnetic structure of Ag, is formed during the process of simultaneous condensation and vice versa depending on the concentration. The granules of Co are well separated from each other below the topological percolation threshold (the volume part of Co in the Co–Ag GF is less than 35%). If the concentration is inverse, the granular structure is inverse too. The properties of these films are close to solid ferromagnetic films [5].

Therefore, the behaviour of granular Co–Ag films under the action of magnetic field (MF) is very interesting [6]. For this purpose, the granular films $Co_{30}Ag_{70}$ and $Co_{70}Ag_{30}$ were observed at the room temperature by means of the scanning tunnelling microscope (STM) REMT 100 with the source of static magnetic field, which has been placed into the zone of tunnel junction, parallel to the surface of the film [6]. Samples were obtained by evaporation of Co and Ag from two independent electron-beam sources. The thickness of the film is 200 nm. The technology features of production and research of the structure of the films are described in the paper [6]. In paper [6], the presence of giant magnetoresistance in such system, the hysteresis, the magnetic aftereffect effects and irreversible changes in the structural characteristics of the films surface of $Co_{70}Ag_{30}$ or $Co_{30}Ag_{70}$, under the influence of a magnetic field were confirmed. However, the quantitative characteristics of the deformations of the Co–Ag film surface were not investigated in the paper [6].

2. EXPERIMENT AND RESULTS

This work is devoted to research of these problems. In this paper, the influence of constant MF, applied in the plane of the $Co_{25}Ag_{75}$ film, on the state of surface was investigated by using the tunnelling microscopy methods. Similarly to the paper [6], the set of MF pulses (duration of the individual pulse is 80 s) was applied to the sample. The general algorithm of magnetic field application is presented in Fig. 1; the set of MF pulses is divided into two series of pulses (see Figs 2 and 3). The break (duration of 80 s) was made for the first series of pulses after applying each MF pulse before each STM scanning for investigation magnetic aftereffect effect (Fig. 1).

The sample was exposed to the action of the second series of pulses after the application of the first series of pulses (3 growing MF pulses). The scanning of the sample was carried out both directly during the second series of MF pulses and since different duration of relaxation after every MF pulse. Some scanning instability of surface elements and quality of scans were observed during magnetic field action.

The experiment for study action of MF on the surface structure of GF was conducted after 1 week of sample relaxation. The region of the

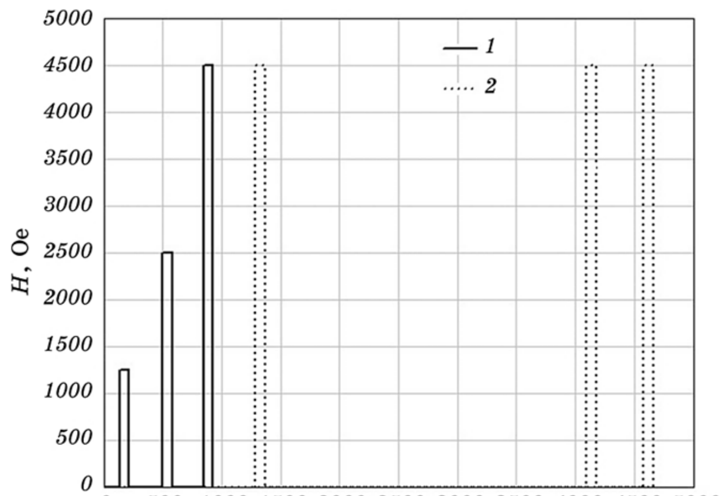

Fig. 1. The general algorithm of application of the magnetic field in the research: *1*—the first series of MF pulses, *2*—second series of MF pulses.

sample surface in this experiment was different from the surface region in the previous experiment. The drift of the scanning area was one of the major problems of wide and correct use of statistical software packages for the comparative analysis the STM scans. Therefore, special attention to the analysis of STM scans was paid to the formation of the comparable regions of the sample surface from the scanning data. As a result, the comparable areas were obtained from different scans with the possible shift in the horizontal plane from 1 to 3 pixels. Some differences in these comparable regions were observed.

These differences appear as the results of MF action or other random external factors, which could affect the quality of scans. Every scan was passed through median filter 3×3 pixels in Nova (Image Analysis 2.1.2) program for elimination of tiny occasional artefacts. The statistical analysis was performed several times for every scan for calculation of the mean value and standard deviation. In all cases, the value of random standard deviation of the film surface, connected to horizontal shift from 3 pixels to 1 pixel, was lower than value of scanning hardware error. The last was taken as the integrated methodological error

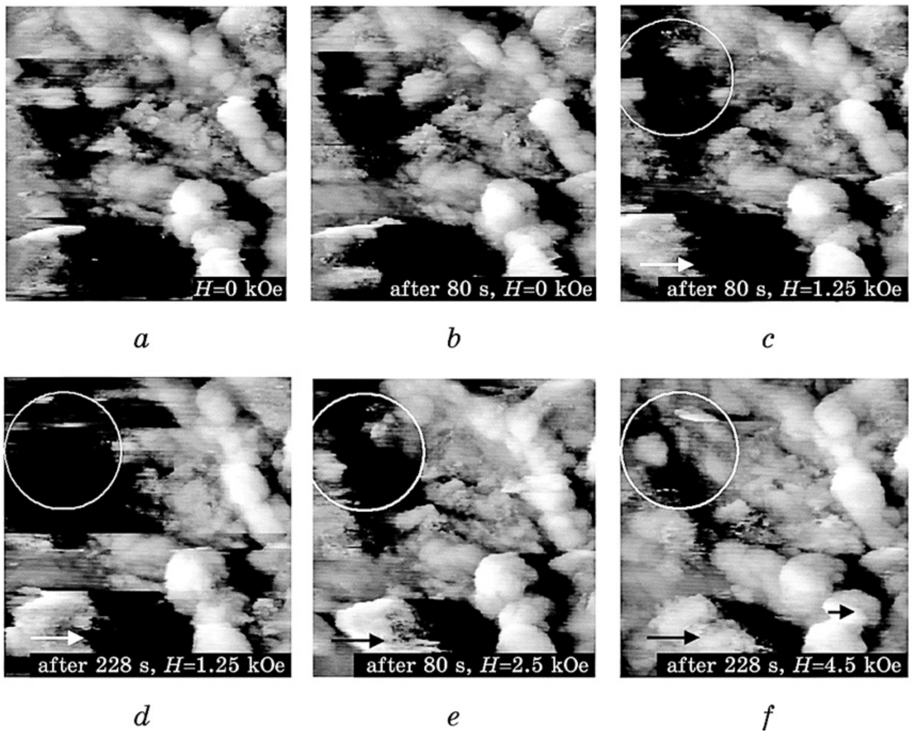

Fig. 2. The STM images of the $Co_{25}Ag_{75}$ film surface after MF action in the first research. The pictures have the same size ($x = 217$ nm, $y = 250$ nm).

for the subsequent calculation of total error (including random). The hardware methodological errors for each of the parameters was obtained by statistical comparison of the first and the second scans of the sample, these scans we made one by one with a 80 s pause before MF action (Fig. 2, *a*, *b* for the first investigation).

Visual scans comparison before the MF action indicates that the surface of film has been stable over time shape because Fig. 2, *a* and *b* are almost the same. Basic statistics average parameters of scans are very close to each other even with some horizontal shift of the identical surface regions within a few pixels. Figures 2, *b*–*d* show scans of film surfaces obtained after the action of MF pulse (duration 80 s) and after the pause (duration 80 s).

It is noticeable from visual comparison of scans after and before MF action that the surface morphology changes as a result of MF action.

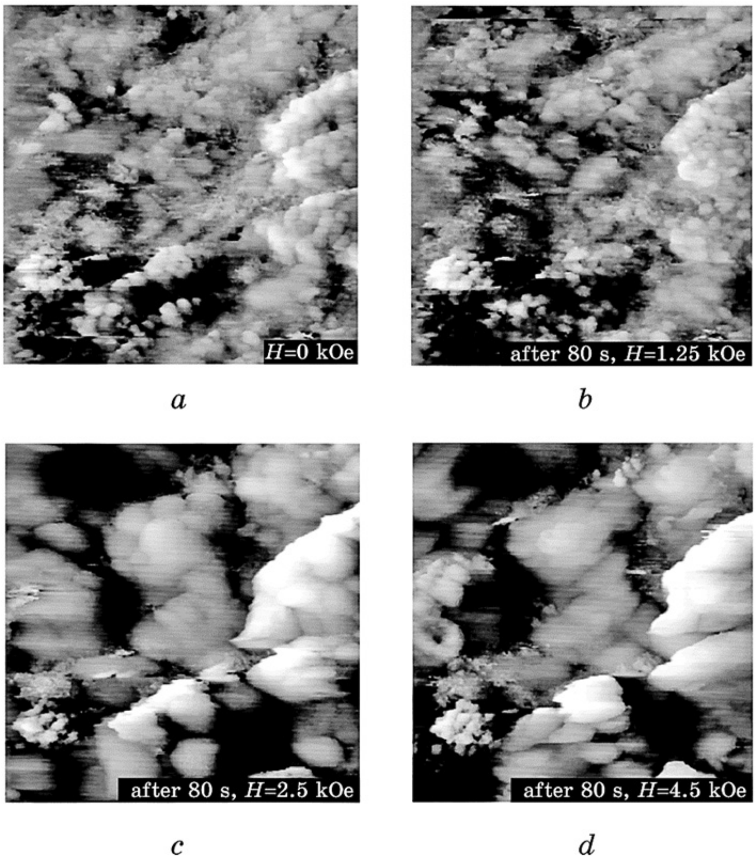

Fig. 3. The STM images of $Co_{25}Ag_{75}$ film surface after MF action in the second research. The pictures have the same size ($x = 262$ nm, $y = 286$ nm).

The morphology of films surface continues to change over time during the period at least several times larger than the MF pulse as one can see from the comparison of Fig. 2, *b* and *d*. For the quantitative analysis of the effect, the function $h(x, y, t)$ is usually used which shows height of film surface in the point with coordinates (x, y) and at the t time. The function h_c is the average h at the area of research. The temporal dependence of h_c is represented in Fig. 4. The value $\Delta h_c(x, y, \Delta t)$ is used to characterize the medium nanoscale deformation of the film surface

$$\Delta h_c(x,y,t,\Delta t) = h_c(x,y,t+\Delta t) - h_c(x,y,t), \quad (1)$$

which has physical meaning, namely, the changing of the point height

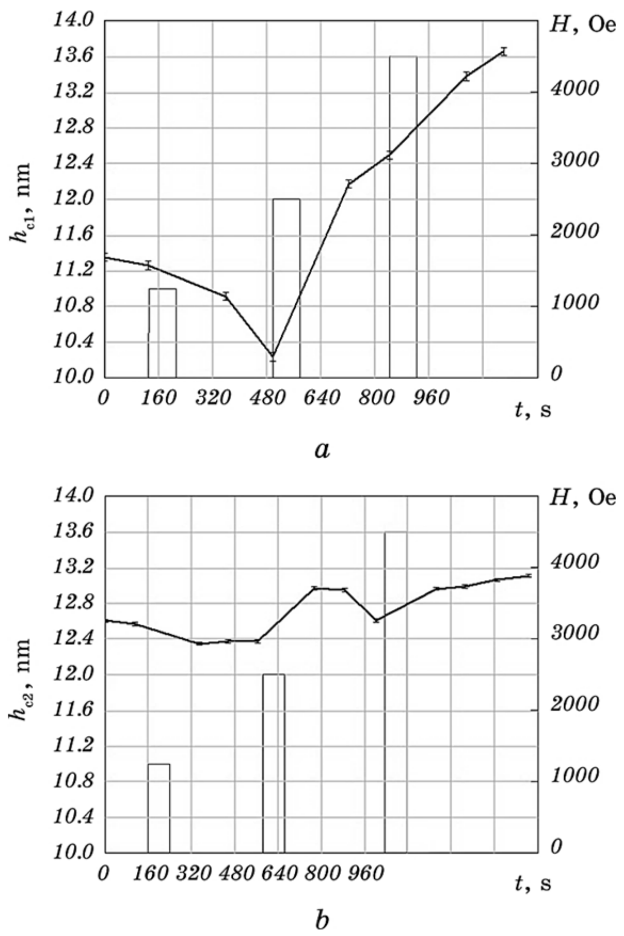

Fig. 4. Dependence of the average surface height h_c (bold line) on MF strength: the initial experiment (*a*), the second experiment (after 8 days) (*b*).

at the different times. And the rate of change of this difference is as follows:

$$V_n(t) = \Delta h_c(x, y, t, \Delta t)/\Delta t. \quad (2)$$

The temporal dependence of $V_n(t)$ is represented in Fig. 5.

The conventional variation coefficient is used for the quantitative description of the changes of film surface statistical parameters. The variation coefficient of average height is about 10% in the first experiment and it is about 2% in the second one.

The Figures 2 and 3 show that some nanogranules and their groups on the film surface are deformed and shifted in different directions.

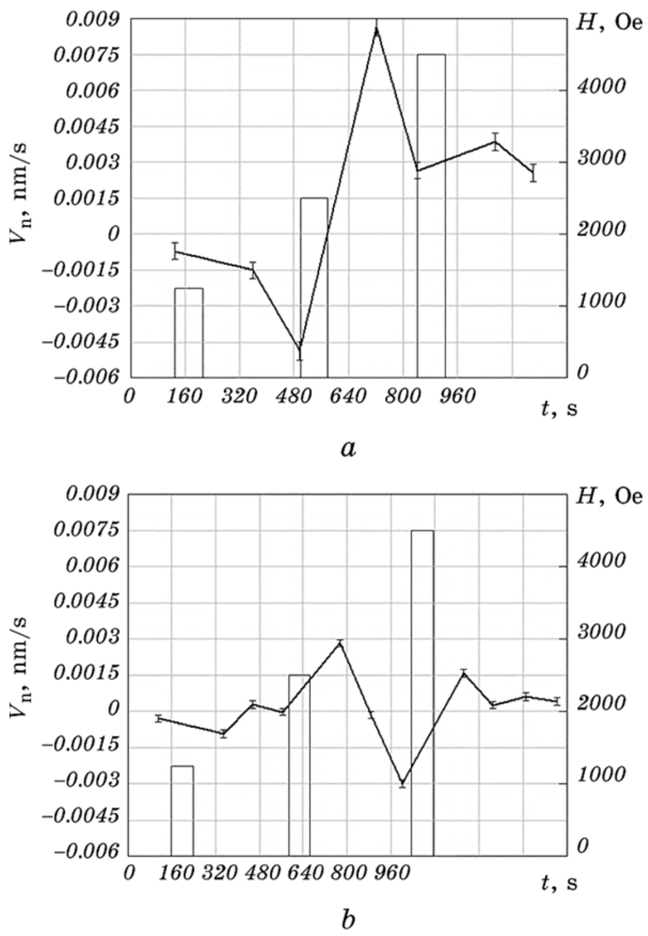

Fig. 5. Dependence of the rate of change average height V_n (bold line) on MF strength: initial experiment (*a*), second experiment (after 8 days) (*b*).

These deformed granules are several orders of magnitude bigger than the typical size of atoms on which the tunnel current action spreads. However, the influence of MF on tunnel current gives several orders of magnitude less contribution to the deformation than the typical magnitude of this deformation. Thus the observed effect can be attributed to irreversible changes in the characteristics of morphology of the $Co_{25}Ag_{75}$ film under MF action and magnetic relaxation processes, rather than to the MF influence on the tunnel current.

At the same time, the various types of GF deformation or individual nanogranules deformation can refer to the effect of giant magnetostriction, and/or be the result of Ag deformation due to the magnetostriction of neighbouring Co granules. At the same time shift of nanogranules, appearance and disappearance of new surface structures probably occur due to structural changes under the influence of the high-gradient MF. This MF is the stray magnetic fields of the neighbouring magnetized cobalt grains and its aftereffects.

The maximum changes of h_c and $V_n(t)$ values are obtained after application of the MF pulse as one can see from the comparison of average surface deformation (Fig. 4) and of its rate (Fig. 5). The maximum magnitude $V_n(t)$ in both experiments was obtained after MF action with strength 2.5 kOe. Further field pulses, even with greater strength have lesser effect. The rate of surface nanodeformation after MF action decreases after 225 s. At that time, the changes in the surface film structure continue to occur due to the magnetic aftereffect and possibly due to restructuring process (see. Fig. 2, *c, d* and Figs. 4, 5). The h_c seems to decrease due to the restructuring processes according to the graph. They increase or decrease the tendency of changes caused by magnetic aftereffect.

The similar multidirectional nature of the MF influence on deformation of the surface is observed both in the first and in the second research. In both cases, after the first MF pulse the average height of the surface of the film decreases, and after the second and third pulses the height usually increases. The absolute values of these changes are also different. The changes upward are about several times greater than downward in both cases.

As a result, after the 8-day period of relaxation, the new stable surface height h_c is about average value between the initial state before the first MF action and maximum value obtained during the initial investigation process. In addition, the GF surface structure has the new form (Fig. 3, *a*) which also indicates the presence of structural changes in the film surface as a result of MF action and its aftereffect. It is worth noting the considerable stability of the new surface structure, which can fundamentally change only after MF action with strength greater than 2.5 kOe.

The multidirectional deformation of individual elements and regions are also observed. The amplitude of the average h_c changes was about 3 nm during the initial magnetization depending on MF in the range from 0 to 4.5 kOe. It corresponds to the elongation of the sample in the normal direction to the GF surface (the film thickness is 200 nm) at level 15000 ppm (1.5%), which is close to giant magnetostriction effect in modern materials based on Tb and Dy, and in shape-memory materials based on martensitic transformation [7–9]. The specified amplitude decreased by 5 times after 8-day relaxation but remained one order of magnitude greater than the known value of cobalt giant magnetostriction.

As noted, all deformations of the sample several times greater at first MF application than the deformations of the sample which was exposed to MF before and where some magnetic aftereffect and structural changes occurred.

It is possible that after certain number of magnetization cycles, especially in strong fields, the resulting structural changes are much closer to the known values of cobalt magnetostriction.

Graphs show that the level of resulting errors do not affect on the reliability of the dependencies. They do not affect the apparent correlation of moments when the MF acts on the sample with the moments of time when deformation and deformation rate significantly increase or reach maxima (see Fig. 4, *a* and *b*).

3. CONCLUSIONS

It is the first time when the significant effect of MF on average height h_c of $Co_{25}Ag_{75}$ film surface have been observed during the initial magnetization. After 8 days of relaxation the result of the second MF action is several times less than initial effect, however it does not disappear. The absolute amplitude of the average height changes h_c in MF strength range from 0 to 4.5 kOe during the initial magnetization is close to the value of giant magnetostriction in modern materials based on Tb and Dy, and in shape-memory materials based on martensitic transformation [7–9]. The magnetic aftereffects are observed. In particular, the partial relaxation of average surface deformation in the direction of the initial value is observed. But in general, the deformation is irreversible at the 8-day time period.

The maximum rate of changes was detected after second MF pulse ($H = 2.5$ kOe) mainly in the direction of increasing the average surface height h_c with field increasing. The abrupt deformation of film surface (the shape changing of nanogranules, the granules shift in the plane of MF application) occurs after second MF pulse.

The effect of giant nanoscale deformation of $Co_{25}Ag_{75}$ film surface under MF action is similar to giant magnetostriction effect. It is probably the complex result of different types of film surface deformation under high-gradient MF action on the magnetic granules in the sample and their structure and due to magnetic aftereffects.

REFERENCES

1. A. E. Berkowitz, J. R. Mitchell, M. J. Carey, A. P. Young, S. Zang, F. E. Spada, F. T. Parker, A. Hutten, and G. Tomas, *Phys. Rev. Lett.*, **68**: 3745 (1992).
2. S. Honda, N. Nawate, M. Tanaka, and T. Okada, *J. Appl. Phys.*, **82**: 764 (1997).
3. K. P. Belov, *Soros Educational Journal*, **3**: 112 (1998) (in Russian).
4. N. P. Lyakishev, *Diagrammy Sostoyaniya Dvoynykh Metallicheskikh Sistem [State Diagrams of Binary Metal Systems]* (Moscow: Mashinostroenie: 1996), vol. 1–2, p. 992 (in Russian).
5. Yu. G. Pogorelov, G. N. Kakazei, J. B. Sousa, A. F. Kravets, N. A. Lesnik, M. M. Pereira de Azevedo, M. Malinowska, and P. Panissod, *Phys. Rev.*, **60**: 12200 (1999).
6. Yu. I. Gorobets, A. F. Kravets, V. N. Zakharchenko, and O. M. Brukva, *Research Bulletin of NTUU 'KPI'*, **6**: 115 (2001) (in Ukrainian).
7. L. JingHua, J. ChengBao, and X. HuiBin, *Sci. China*, **55**: 1319 (2012).
8. O. Söderberg, A. Sozinov, and V. K. Lindroos, *Enc. Mat.: Sci. Techn.*, **5**: 67 (2004).
9. A. Behera and S. C. Mishra, *Int. J. Adv. Appl. Sci.*, **2**: 205 (2013).