TEMPERATURE DEPENDENCIES OF MAGNETIZATION AND HYSTERESIS LOOPS OF COMPOSITE FILMS ($CoFeB+SiO_2$) WITH DIFFERENT COMPOSITIONS AND STRUCTURE

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Abstract. Temperature dependences of magnetization of composite films ($CoFeB+SiO_2$) with different concentrations of metal alloy *x*=45--85 at. % with different micro- and nanostructure at temperatures of 2--400 K in magnetic fields of 5 mT and 5 T were obtained. Hysteresis loops of magnetization of films with different x in magnetic fields up to 5 T at temperatures of 2, 100, 300, and 400 K were obtained. It was shown that films with granular structure are characterized by a wider hysteresis loop and have a stronger dependence of magnetization on temperature compared to films with granular-percolation structure and films with a structure in the form of a metal matrix with dielectric inclusions.

Keywords: *composite metal-dielectric films, magnetic structure, magnetization, temperature dependences, hysteresis loops*

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INTRODUCTION

The study of magnetic properties of composite materials represents one of the key areas in materials science and in modern physics especially in the context of the development of new technologies and devices [1-3]. Composite films consisting of combinations of magnetic and dielectric non-magnetic components have unique properties that can be used in various applications such as, magnetoelectronics, magnetic storage media and sensor technologies [4, 5]. Composite films: magnetic metal-dielectric have attracted much attention of researchers due to their unique ability to fine-tune magnetic properties and high sensitivity to external electromagnetic fields [6]. One of the interesting classes of composite materials are films containing CoFeB metal alloy and SiO_2dielectric material , in which there is a different combination of magnetic and non-magnetic phases [7]. Previous studies have shown that the magnetic properties of such composite films strongly depend on their structure, which can vary from granular to magnetic band structures and solid metal matrices with dielectric inclusions depending on the concentration of metal alloy x and the thickness of the films [8]. The purpose of this work is to establish the relationship of magnetic properties and their dependences on temperature and magnetic field for composite films ($CoFeB+SiO_2$) with their composition and micro- and nanostructure. For this purpose, experimental studies of temperature dependences of magnetization in the temperature range of 2-400 K at magnetic fields of 10 mT and 5 Tesla for composite films with different micro- and nanostructures were carried out in this work. Magnetization hysteresis loops were also obtained for these films at different temperatures of 2, 100, 300, and 400 K at changing magnetic fields from 0 to 5 Tesla.

FILM SPRAYING PARAMETERS, FORMULATIONS AND THICKNESSES

The object of study in this paper is composite films, which are amorphous metallic and dielectric micro- and nano-regions [9]. These films were synthesized at Voronezh State Technical University (VSTU) using advanced ion beam bombardment technologies [10]. The films were synthesized on a modernized vacuum sputtering post UVN-2M, specially adapted for ion-beam bombardment of different targets [11, 12]. As targets were used plates from metallic alloy $Co_{0.52}Fe_{(0.45)}\,B_{0.2}$and dielectric material SiO_2. The sputtering of films was carried out in the atmosphere of inert gas argon with pressure of 0.02 Pa, which provided stability of sputtering and minimized undesirable chemical reactions during sputtering of films. The substrate for the sputtered films was a polymer sheet made of polyethylene terephthalate of A4 format with dimensions 297×210 mm² and thickness 30 microns, which provided the necessary flexibility and temperature stability of the composite material. Before sputtering, the polymer substrate underwent an ionic cleaning process, which significantly improved the adhesion of the atomized target material. During the sputtering of the films, the hood with a fixed sheet of lavsan substrate was rotated at a rate of 1 revolution in 5 minutes, the time of the sputtering process was 120 minutes.

To analyze the chemical composition of composite films, the method of energy dispersive microanalysis (EDX) was applied using the AZTEC X-ACT attachment, manufactured by Oxford Instruments, integrated into the TESCAN MIRA3 SEM system. During the analysis, the obtained spectra were used to quantify the composition of the films, especially to determine the concentration of the metal alloy elements Co, Fe, B. To measure the thickness of the films, an image of the end of each sample was obtained, which allowed visualizing the boundary between the film and the substrate. A backscattered electron detector was used, which provided high-contrast images that were particularly sensitive to differences in the atomic number of the materials. The contrast between the film and substrate allowed the interface to be clearly defined, which enabled accurate measurement of film thickness. The results of the thickness and composition measurements of the films were systematized and presented in Table 1. The following notations are used in the table: d - film thickness, x - CoFeB metal alloy concentration expressed in atomic percent. The samples are ordered in the table, according to the sequence of sputtering on the polymer sheet, which allows to trace the changes in the characteristics of the films depending on time and conditions during their sputtering. The geometry and dimensions of SiO_2plates were selected such that almost all films, except for two films No. 1, 2 with x= 45 and 46 at. % were obtained with a post-percolation structure [12].

EXPERIMENTAL METHODOLOGY AND TECHNIQUE

Measurements of the magnetic moment of the films in the temperature range of 2-400 K and magnetic fields of 0-5 Tesla were carried out at the Center for Diagnostics of Functional Materials for Medicine, Pharmacology, and Nanoelectronics of the Science Park of St. Petersburg State University (SPbSU). Measurements of magnetization were carried out using MPMS 3 SQUID complex for automated measurements, including Superconducting Quantum Interference Device (SQUID) and thermostat with temperature control in the range of 1.9-400 K. Magneto-field dependences of the films magnetization at different temperatures 2, 100, 300, 400 K were measured in automatic mode when changing the magnitude and direction of the magnetic field. During all measurements, the magnetic field was directed parallel to the film surface. Such orientation of the magnetic field provided minimal influence of the demagnetizing field of the composite film and allowed to determine the magnetic moment of the film more accurately. After the measurements, the obtained values of the magnetic moment were converted into the magnetization of the sample, from which the temperature and magnetic-field dependences of magnetization (hysteresis loops) were plotted for each film with different composition and magnetic structure.

COMPOSITE FILM STRUCTURE

An atomic force microscope (AFM) INTEGRA PRIMA (NT-MDT, Russia) equipped with an MFM10 cantilever was used to obtain images of film surface topography and magnetic phase contrast (MPC). A silicon probe coated with a thin layer of CoCr magnetic alloy, 30 nm thick, was mounted on the MFM10 cantilever. The magnetic layer gives the probe additional sensitivity to magnetic fields on the sample surface. The radius of the probe tip was 40 nm, which provided a spatial resolution of magnetic structures of about 20 nm. During the measurements, the frequency of the external force acting on the cantilever with the probe was tuned close to the natural frequency of the cantilever in the frequency range from 70 to 90 kHz. The choice of frequency is important to optimize the sensitivity of measurements and minimize the influence of unwanted vibrations and noise. When scanning the film surface, the magnetic probe interacts with surface irregularities and with local magnetic fields that determine the phase of the cantilever vibrations at fixed time instants. The phase changes, or phase difference Δφ relative to some region of the film, were the data recorded during the AFM measurement. The recorded phase changes Δφ were converted into visual images that displayed the topography of the film and the distribution of magnetic homogeneous regions on the surface of the composite films. Next, the image of the surface topography of the films was subtracted from the total phase change image. After that, magneto-phase contrast (MPC) images were obtained (Fig. 1), in which regions with different sign Δφ have different directions of the local magnetic field of the scanned surface area. Information on the average sizes of granules and homogeneous regions can be obtained from the MFC image, since the magnetic probe does not only respond to changes in the magnetic fields above the film surface [7]. Estimation of the average granule sizes from the MFC image of film #3 in Fig. 1 gives values of 20-50 nm. Information on the size, shape and orientation of magnetic regions can also be obtained from the MFC image [7]. Analysis of the MFC images of composite films with different concentrations of metal alloy x showed significant differences in the magnetic structure (Fig. 1). As can be seen from Fig. 1, films #3 ($x =$ 51 at. %) and #5 ($x =$ 54 at. %) possess a granular-percolation structure. In the MFC image of fragment 3, a weak magnetic field is registered in the whole scanning window of the 10× 10 μm film^{2}with percolation regions of small sizes. This structure indicates the onset of percolation processes during film sputtering, when an accumulation of metal particles (granules) begin to form large regions capable of generating significant homogeneous magnetic fields over the film surface [7]. At a higher concentration of $x =$ 65 at. %, for films with a granular-percolation structure, magnetic bands with a length of more than 4 μm with weakly different magnetization in neighboring bands are observed (fragment 6). With increasing *x* up to 69 at. % the neighboring bands have strong difference in the generated magnetic field (fragment 7). Further increase of *x* leads to a significant broadening of these magnetic bands (fragment 8). For films with the highest concentration of metal alloy *x*= 84 at. %, the film images with a scanning window of 20× 20 μm^2show a homogeneously magnetized region of the film, along which run parallel bands that can be created by regions of other magnetization (fragment 10). This indicates that at high concentration of *x*, the magnetic field generated over the film surface becomes more homogeneous in large film microareas and corresponds to a metallic matrix with dielectric inclusions. Analysis of AFM images of MFC films obtained by AFM using a cantilever with a magnetic probe [3, 6] showed that the films have different micro- and nanostructures, including the magnetic structure (Fig. 1). Films with low concentration of metal alloy *x*= 45, 46 at. % have a granular structure (this structure is labeled G in Table 1). Films with concentrations of *x*= 51-65 at. % have a disordered granular-percolation structure (this structure is labeled GP1). Films with high concentration *x*= 67-77 at. % have a granular-percolation structure in the form of a banded zigzag magnetic structure (this structure is labeled GP2 in the table). At the highest concentration *x*> 85 at. %, the structure of the films is a matrix with dielectric inclusions (this structure is labeled MDI in the table).

RESULTS AND DISCUSSION

Fig. 2 shows the temperature dependences of the saturation magnetization $M(T)$ for composite films (CoFeB+SiO_2) with nine different concentrations of metal alloy *x* from the range 45 - 85 at. %, which are in magnetic fields of 5 mT and 5 Tesla. As can be seen from Fig. 2, a monotonic decrease in magnetization with increasing temperature from 20 to 400 K is observed for all films in 5 mT and 5 Tesla magnetic fields. For films #1, 2 with granular structure at small magnetic fields of 10 mTl, a more rapid decrease in magnetization with increasing temperature is observed than for the other films with granular-percolation structure. At high magnetic fields of 5 Tesla, a weak dependence of magnetization on temperature is observed for all films. As expected, the highest magnetization value in the whole temperature range is observed for film No. 10 with the highest concentration of metal alloy x= 85 at. %, which is characterized by a structure in the form of a metal matrix with dielectric inclusions. The small value of magnetization in large magnetic fields (Fig. *2b*) is characteristic of film No. 1 with the lowest concentration of x, which is characterized by a granular structure. Low-temperature magnetization maxima at temperatures of 10, 13, and 5 K are observed for films #1, 2, and 3, which have a granular structure, respectively (Fig. *2b*). The positions of these low-temperature maxima $M(T)$ are determined by the equality of the coercive field and the external magnetic field of 5 mTl. The different positions of the low-temperature maxima for films #1, 2 with granular structure are given by different values of the coercive field for these films, which also strongly depends on the temperature (Fig. 3).

Fig. 3 presents hysteresis loops of film magnetization from magnetic field up to 40 mTl for composite films (CoFeB+ SiO_2) with seven concentrations of metal alloy x at temperatures: 2 K (Fig. *3a*) and 300 K (Fig. *3b*). In the experiments, hysteresis loops for all films were obtained in fields up to 5 Tesla. For films No. 7, 8 and 10 with large

x= of 69, 79 and 85 at. %, having magneto-band structure and structure in the form of matrix with dielectric inclusions, the hysteresis loop is very narrow and changes weakly with temperature increase from 2 to 300 K. It should be noted that for films #1, 2 having granular structure, the hysteresis loops are very wide and show a significant effect of film temperature on them.

Fig. 4 shows the concentration dependences of saturation magnetization (a) and coercivity (b), equal to the width of the hysteresis loop, for composite films ($CoFeB+SiO_2$) with different structures at temperatures of 2, 100, 300, and 400 K. The structure of the films for different x concentration is summarized in Table 1. The saturation magnetization of films with granular and granular-percolation structure increases with increasing x concentration, while for films with metallic matrix structure, the magnetization changes weakly with concentration. The greatest growth of magnetization as a function of x is observed for films with granular-percolation structure, for which the volume of percolation or extended metallic regions increases. The behavior of the coercivity of the films at 2 K has a decreasing character for all concentrations x. The behavior of the coercivity $H_{(c)}$ $_{of}$the films at other temperatures of 100, 300 and 400 K has a different character and strongly depends on temperature. For films with concentrations x=46-51 at. %, including transition from granular structure to granular-percolation structure, a minimum of coercivity $H_c$is observed. For films with x= 55-75 at. %. with granular-percolation structure, an increase and further saturation of coercivity $H_c$with increasing concentration x at $T=400$ K is observed. For films with x=80-85 at. %. with metallic matrix structure, a slight decrease in the coercivity $H_c$with increasing x concentration is observed in the studies at all temperatures $T=2$, 100, 300 and 400 K.

CONCLUSION

The structure and magnetic properties of composite films ($CoFeB+SiO_2$) with different concentrations of metal alloy x= 45-85 at. % have been studied. The structure of composite films with increasing x concentration changes from granular structure with x = 45, 46 at. % to granular-percolation structure with x = 0.51-0.79, and with further increase of x from 0.8 the structure transforms into a metallic matrix with dielectric inclusions. Analysis of the behavior of the coercive force of the investigated composite films showed that its value is largely set by type of the composite films structure, which is determined by the concentration of metal alloy x and the thickness of the films. For films with concentrations x=45-60 at. %, including the transition from granular structure to granular-percolation structure, the coercive force $H_c$depends nonlinearly on the concentration of x, first decreasing, then increasing, and then entering saturation at high temperatures of 100, 300, and 400 K. The analysis of temperature dependences of magnetization and coercivity shows that the structure of the films, given by the metal alloy content of the films x, has a significant influence on the temperature behavior of the magnetic properties of composite films ($CoFeB+SiO_2$) . The greatest sensitivity to static magnetic fields is demonstrated by composite films with a magnetically banded structure with concentrations of x = 65-79 at. %, which emphasizes the prospect of their use in highly sensitive magnetic devices. The magnetization of films with a metal matrix structure depends weakly on temperature, which makes materials based on such films especially valuable for use in magnetic devices where high temperature stability is required.

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FIGURE CAPTIONS

Fig. 1. Magneto-phase contrast images of composite films (CoFeB+ SiO_2). The numbers of the figure fragments coincide with the numbers of the films given in Table 1. To the right of the images is a vertical color scale referenced to the phase difference values in milligrads.

Fig. 2. Temperature dependences of magnetization of composite films (CoFeB+ SiO_2) in magnetic fields of 5 mTl (*a*) and 5 Tl (*b*). The numbers of the curves coincide with the numbers of the films given in Table 1.

Fig. 3. Magnetization hysteresis loops of composite films (CoFeB+ SiO_2) at temperatures: 2 (*a*) and 300 K (*b*). The numbers of the curves coincide with the numbers of the films given in Table 1.

Fig. 4. Concentration dependences of magnetization (*a*) and coercivity $H_{(c)}$ *(b)*$_{of}$composite films (CoFeB+ SiO_2) at measurement temperatures *T*=*2*, 100, 300 and 400 K. The measurement temperatures of the corresponding parameters are given next to the curves.

Table 1. Compositions and thicknesses of composite films ($CoFeB+SiO_2$)

d, nm	*x*, at. %	B	Co	Fe	O	Si	Structure	Film number, №
556	46	4	22	20	38	16	G	1
684	45	4	22	19	39	16	G	2
741	51	4	25	22	33	16	GP-1	3
725	53	5	26	22	32	15	GP-1	4
761	54	4	26	24	31	15	GP-1	5
858	65	5	32	28	23	12	GP-2	6
864	69	5	34	30	20	11	GP-2	7
810	79	8	37	33	14	7	GP-2	8
759	83	10	39	34	11	6	MDI	9
680	85	10	40	35	9	6	MDI	10

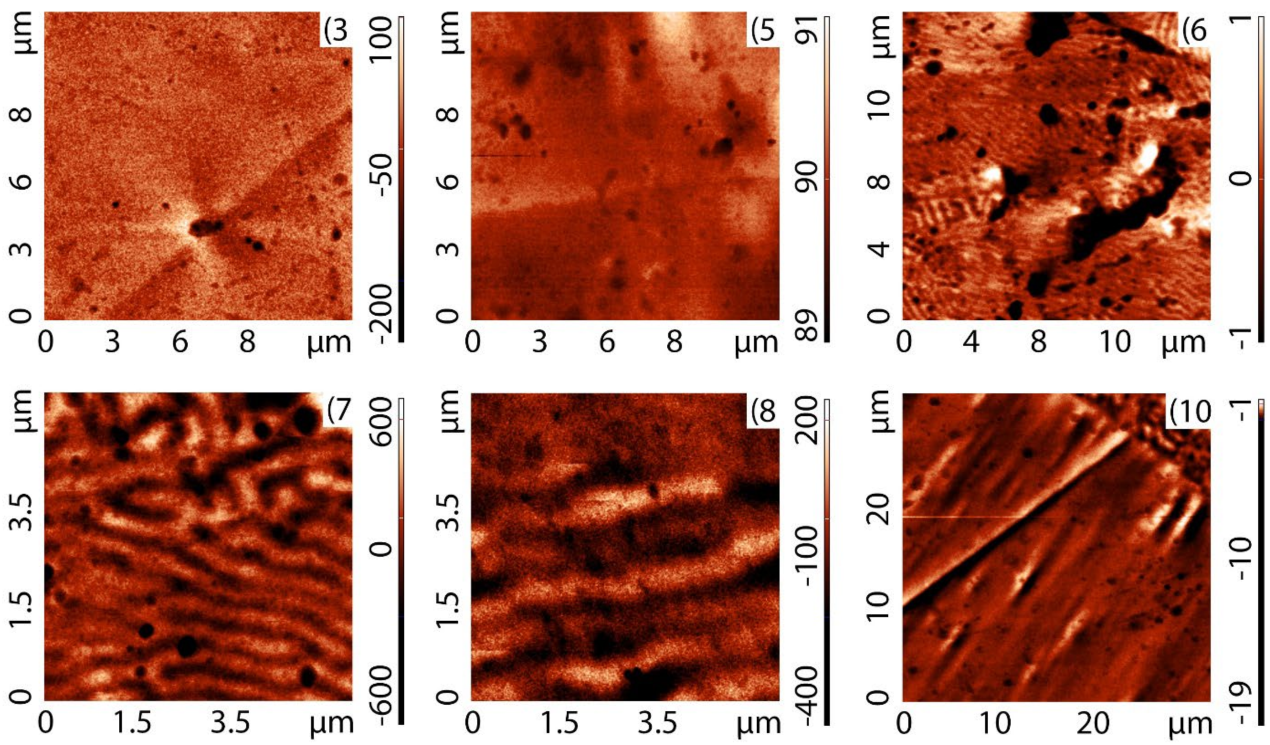
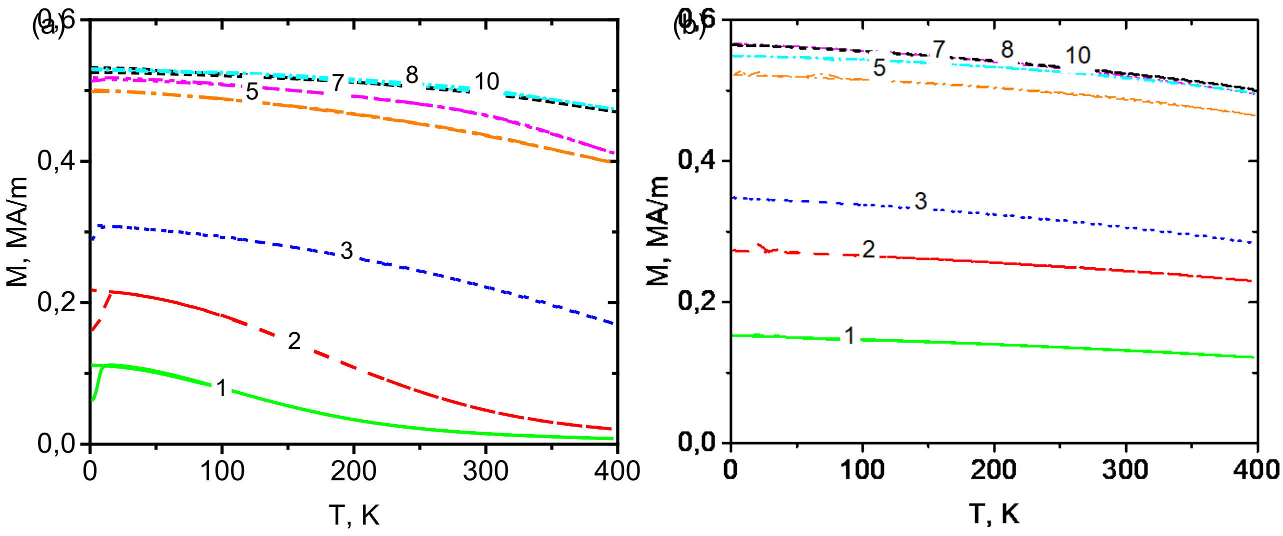
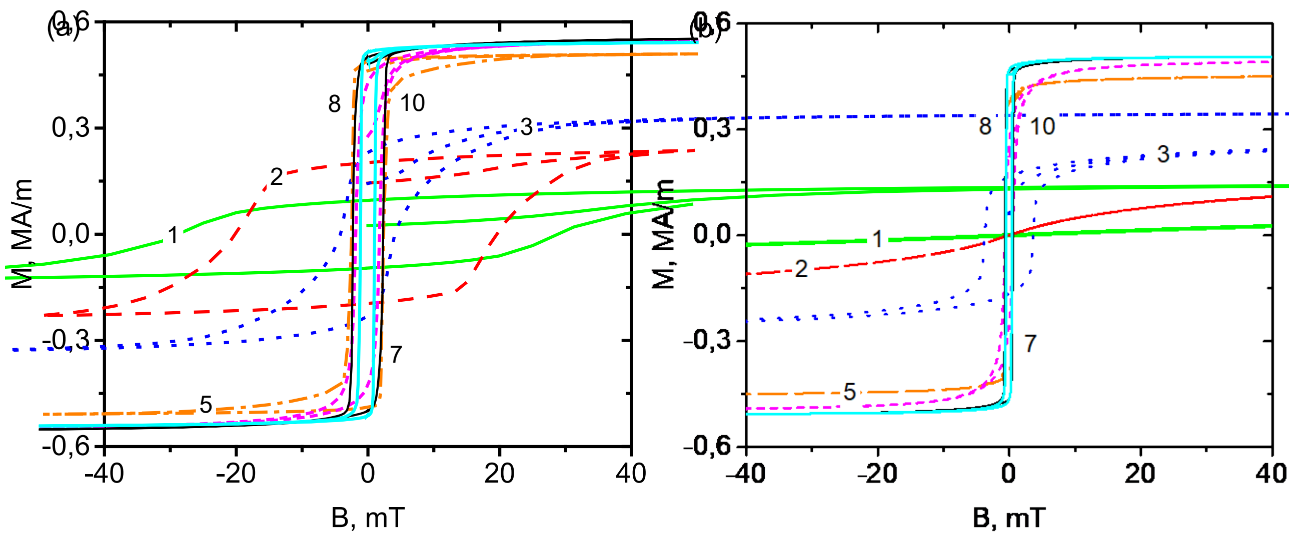


Fig. 1.



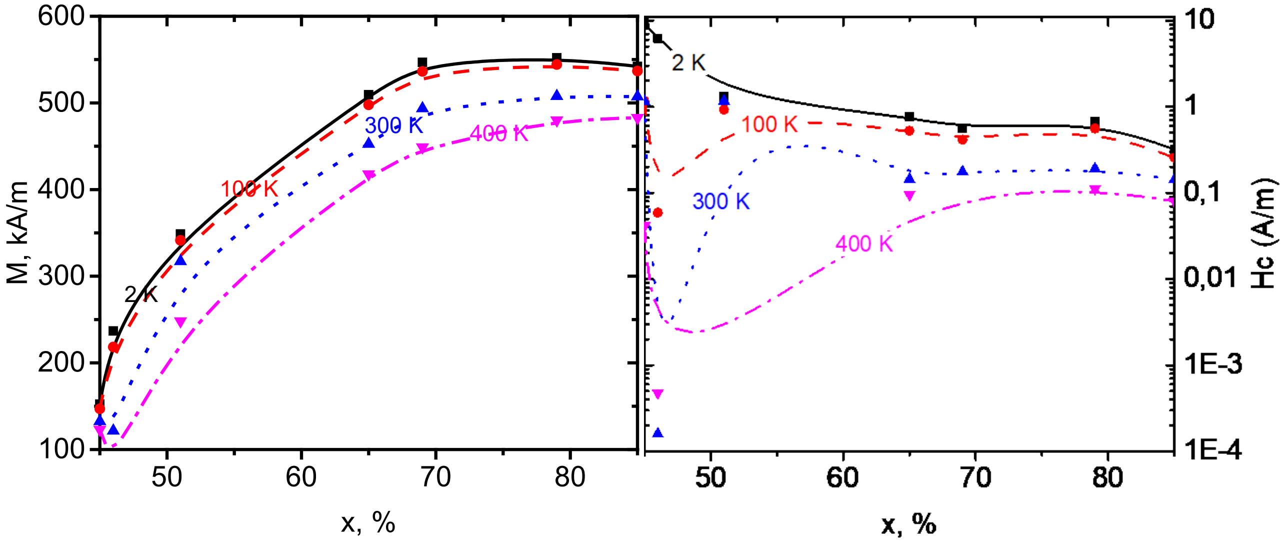
(a)
0,6
0,4
0,2
0,0
M, MA/m
0
100
200
300
400
T, K
1
2
3
5
7
8
10
(b)
0,6
0,4
0,2
0,0
M, MA/m
0
100
200
300
400
T, K
1
2
3
5
7
8
10

Fig. 2.



(a)
0,6
0,3
0,0
-0,3
-0,6
M, MA/m
-40
-20
0
20
40
B, mT
1
2
3
5
7
8
10
(b)
0,6
0,3
0,0
-0,3
-0,6
M, MA/m
-40
-20
0
20
40
B, mT
1
2
3
5
7
8
10

Fig. 3.



600
500
400
300
200
100
M, kA/m
50
60
70
80
x, %
2 K
100 K
300 K
400 K
10
1
0,1
0,01
1E-3
1E-4
Hc (A/m)
x, %

Fig. 4.