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THERMOMAGNETIC ANALYSIS AND DOMAIN STRUCTURE IN THE PHASE TRANSITION REGION OF Ni-Mn-Ga AND Co-Ni-Ga SHAPE MEMORY ALLOYS

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Abstract. The recently developed ferromagnetic shape memory alloys (FSMA) exhibit large field induced strains thereby rendering new potentials for application in transducers, actuators and other novel devices. Magnetically controlled strain in FSMA is based on the reorientation of the twin structure of martensite under applied magnetic field. A detailed study of both martensitic and magnetic domain structure is presented for oriented single-crystalline bulk and texturized powdered samples embedded in polymer matrix, and polycrystalline bulk and rapidly quenched ribbon alloys. Optical microscopy including magneto-optical indicator film technique was employed alongside with X-ray and AFM/MFM studies for the characterization of the coexisting structural and magnetic domains. It is shown that only 180° magnetic domains exist in twin plates because martensite possesses uniaxial magnetic anisotropy having magnetization vector ***M*** oriented along easy *c*-axes at angles of ±45° with respect to the twin boundaries. Due to magnetostatic coupling the 180° magnetic domains of neighbouring twins cooperate with each other forming continuous macrodomains running through the whole crystallite or single crystal sample and changing the direction of ***M*** by ±90° in a zigzag fashion at each intersection of the twin boundary.

1. INTRODUCTION

Combination of ferromagnetism and structural phase transitions in Heusler alloys is perspective for the production of new devices based on the magnetic field control of the size and shape of the actuator active elements. The structural phase transitions in these elements proceed by the transformation of high-temperature austenite cubic phase into a tetragonal low-temperature martensite phase [1]. Extremely high (up to 10%) magnetically induced deformations were shown to exist in the family of $Ni_{2+x}Mn_{1-x}Ga$ alloys [2]. Also important, though studied in less detail, are some other groups of Heusler alloys, in particular, those on the basis of Co–Ni–Ga [3–5].

Detailed experimental studies of the regularities of formation and realignment of both martensitic and magnetic domain structure (DS) are necessary for modelling and simulation of magnetically induced phenomena in these alloys. These questions are studied intensively by a number of research groups [6-12]. However the available data are still fragmentary and need to be extended and generalized. In the present work we focus our attention on the study of martensite and magnetic DS of mono-, poly-, and nanocrystalline $Ni_{2+x}Mn_{1-x}Ga$ and $Co_{2+x}Ni_{1-x}Ga$ alloys.

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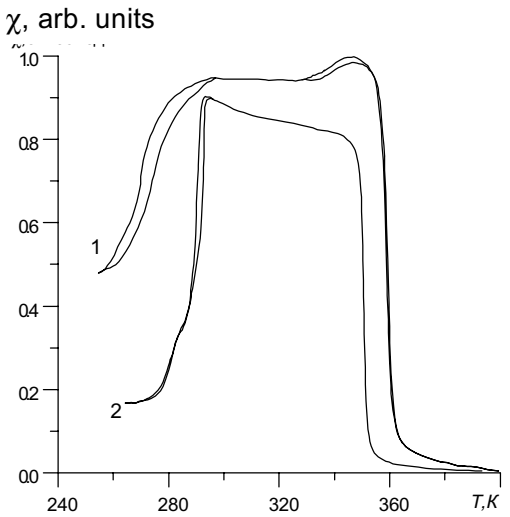

Fig. 1. Temperature dependence of the initial magnetic susceptibility χ for polycrystalline $Ni_{2.12}Mn_{0.88}Ga$ samples. Curve 1 – before, 2 – after homogenization at 800 °C for 100 hours.

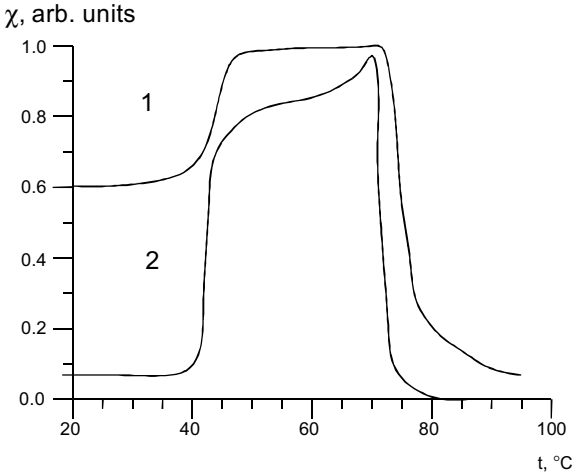

Fig. 2. Temperature dependence of the initial magnetic susceptibility χ for rapidly quenched $Ni_{53}Mn_{23.5}Ga_{23.5}$ ribbons annealed at 800°C for 5 hours. Curve 1 – field parallel, 2 – perpendicular to a ribbon length.

2. EXPERIMENTAL

Polycrystalline $Ni_{2+x}Mn_{1-x}Ga$ and $Co_{2+x}Ni_{1-x}Ga$ alloys with *x* from 0.12 to 0.19 were prepared by argon arc melting and homogenised by 100 hours of annealing at 800 °C followed by water quenching. Single crystalline samples were grown by the Bridgeman method in alumina crucibles from arc melted buttons. The initial charge was kept in vacuum of 10^{-4} mm Hg at 1200 °C for degassing. Next the chamber was filled with pure argon to minimize the evaporation of manganese and the temperature was raised up to 1350 °C. The crystals were grown at the rate of 20 mm per hour.

Nanocrystalline ribbons with a thickness of 30 – 40 μm were obtained by rapid quenching of the melt with a rate of the order of 10^5 K/sec onto a rotating copper wheel. Amorphous and nanocrystalline films with a thickness of 0.5 – 5 mm were prepared by magnetron sputtering.

The temperatures of martensite and magnetic transitions were obtained by the method of thermomagnetic analysis by the temperature dependence of the initial magnetic susceptibility. The martensite and magnetic DS were observed in polarized light with the aid of a digital differential optical microscope. Complementary methods of magnetic powder patterns, Kerr microscopy and magnetooptic indicator films [10,11] were used to reveal the magnetic domain structure.

3. RESULTS AND DISCUSSION

3.1. Thermomagnetic analysis (TMA)

In Fig. 1 the curves of initial magnetic susceptibility temperature dependence $\chi(T)$ for polycrystalline textured alloy $Ni_{2.12}Mn_{0.88}Ga$ are shown before and after homogenization at 800 °C. It is seen that the annealing results in some changes of the characteristic points of TMA curves, decrease of the hysteresis of these curves and sharpening of the magnetic order – disorder (nonhysteretic right parts of the TMA curves) and structural austenite–martensite transitions (left hysteretic parts).

The TMA curves presented in Fig. 1 are typical for single- and polycrystalline ferromagnetic Heusler alloys and play an important diagnostic role, because they give a clear indication of not only the existence of the structural phase transitions and its characteristic temperatures (start and finish points of the direct and reverse martensite transitions), but also provide information on the relative volume of the martensite phase and the material homogeneity.

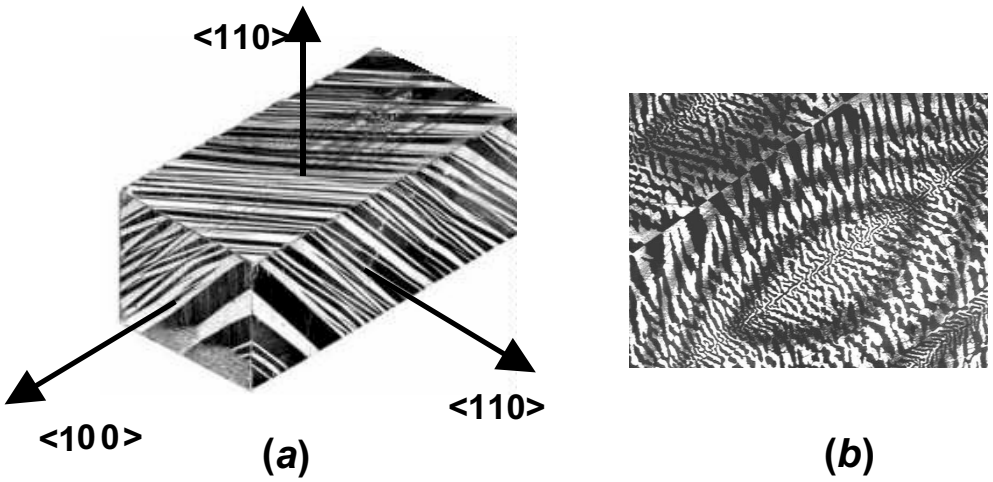

Fig. 3. Martensite relief of an oriented $Co_{48}Ni_{22}Ga_{30}$ single crystal with a size of 3×3×6 mm (a) and respective interference bands observed with the aid of plane parallel indicator film applied to the sample surface (b).

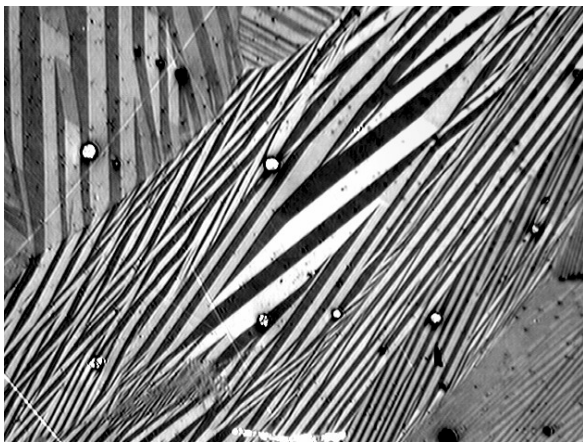

Fig. 4. Martensite structure of polycrystalline $Ni_{2.16}Mn_{0.84}Ga$ revealed by polarized light microscopy on the mechanically polished surface of the sample, ×120.

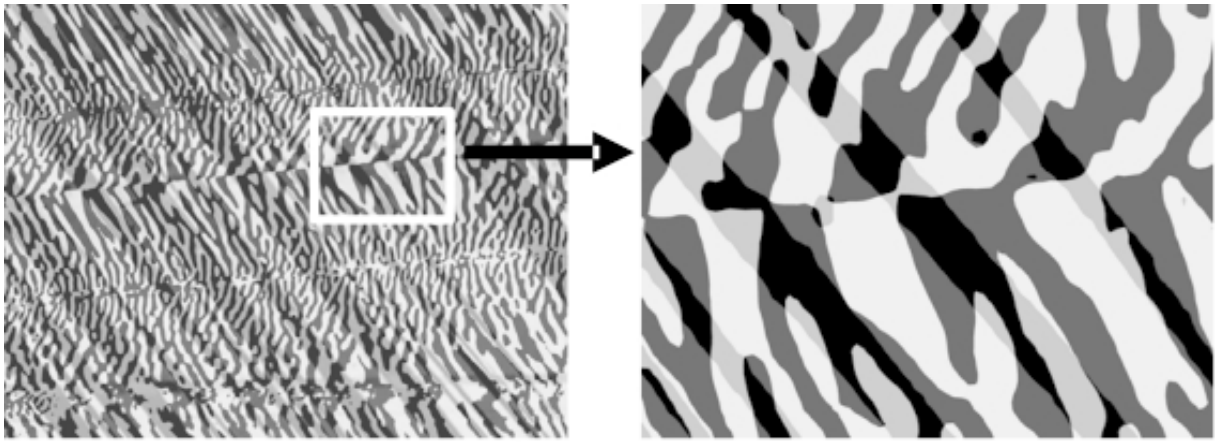

Fig. 5. Martensite relief on the (110) plane of $Co_{48}Ni_{22}Ga_{30}$ single crystal and its relationship with the magnetic DS.

The TMA curves of rapidly quenched ribbons are similar to those of bulk single- and polycrystals. The $\chi(T)$ curves vary with the change of the heat treatment regimes and the measurement conditions (orientation of the measuring field with respect to the sample long dimension, Fig. 2). The

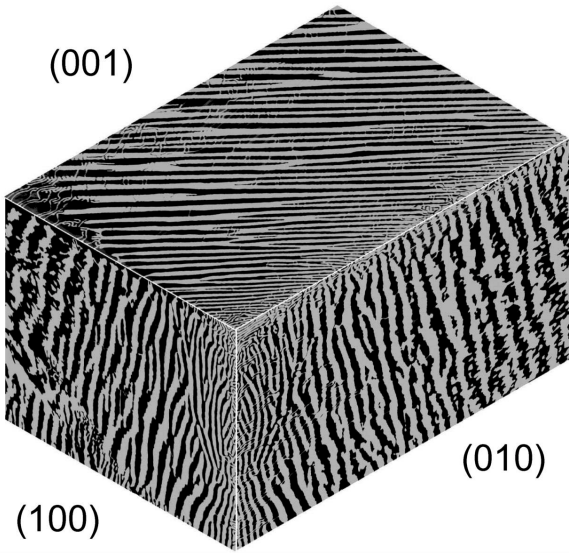

Fig. 6. 180-degree structure of magnetic macrodomains on the {100} planes of $Ni_{49}Mn_{29.7}Ga_{21.3}$ single crystal with linear dimensions of 1.2×1.5×2.5 mm.

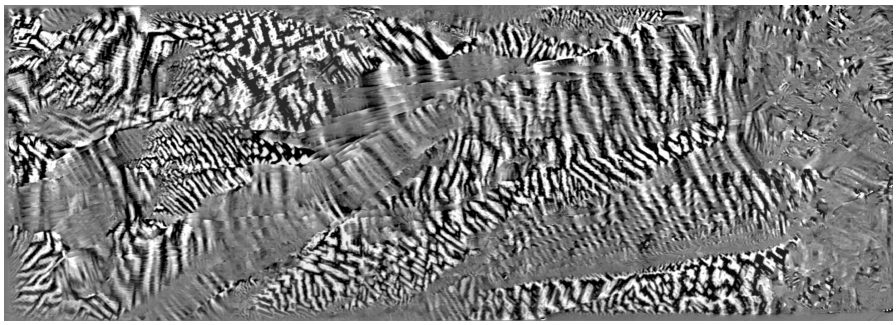

Fig. 7. Magnetic DS of textured $Co_{48}Ni_{22}Ga_{30}$ polycrystal revealed with the aid of ferrite-garnet indicator film. The texture axis is oriented parallel to the long side of the sample having dimensions of 3.3×3×9.4 mm.

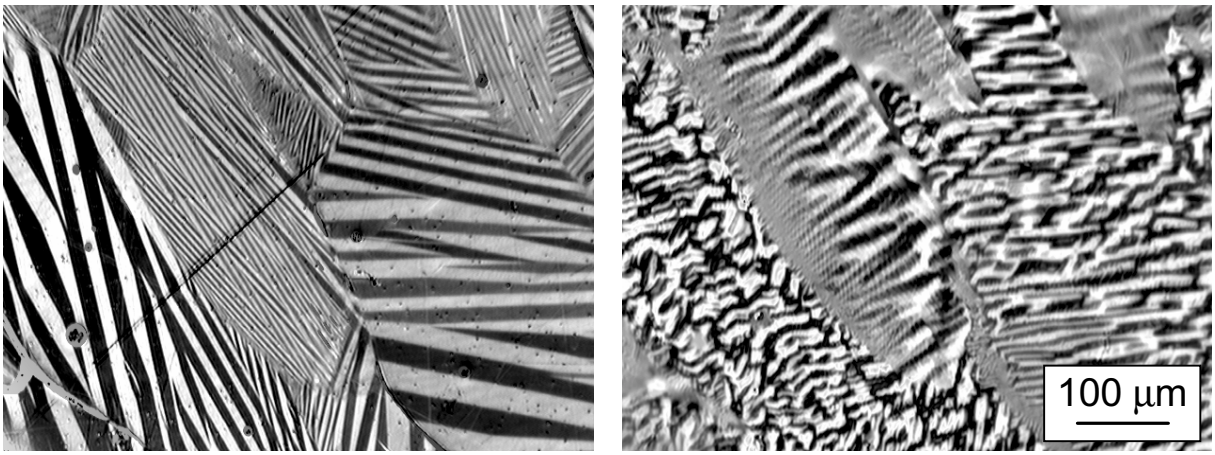

Fig. 8. Visualization of the martensite and magnetic DS of $Co_2Ni_{0.85}Ga$ polycrystal. Left – martensite structure observed in polarized light, right – magnetic DS of the same area of the sample revealed with the aid of magnetooptic indicator film.

latter effect is due to the shape anisotropy of the ribbon samples (different demagnetizing factors along the ribbon length and perpendicular to it). In spite of the difference in the absolute values of the measurement signal for these two cases the corresponding characteristic start and finish marten-

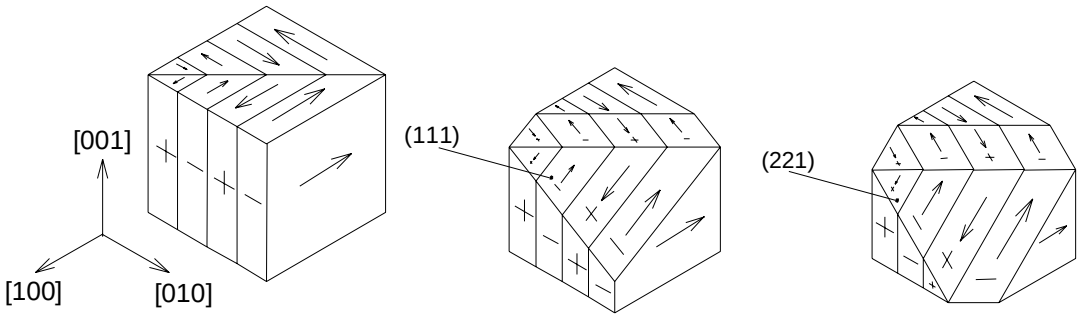

Fig. 9. Magnetic domain substructure forming by sectioning of the crystal with (hh1) planes. Cubic volume element (at the left) is divided into two parts by the martensite boundary lying in the (110) plane. (hh1) planes are obtained by the rotation of (001) plane with respect to the $[\bar{1}10]$ direction. Arrows and (+) and (-) signs characterize the magnetization directions of 180° magnetic domains.

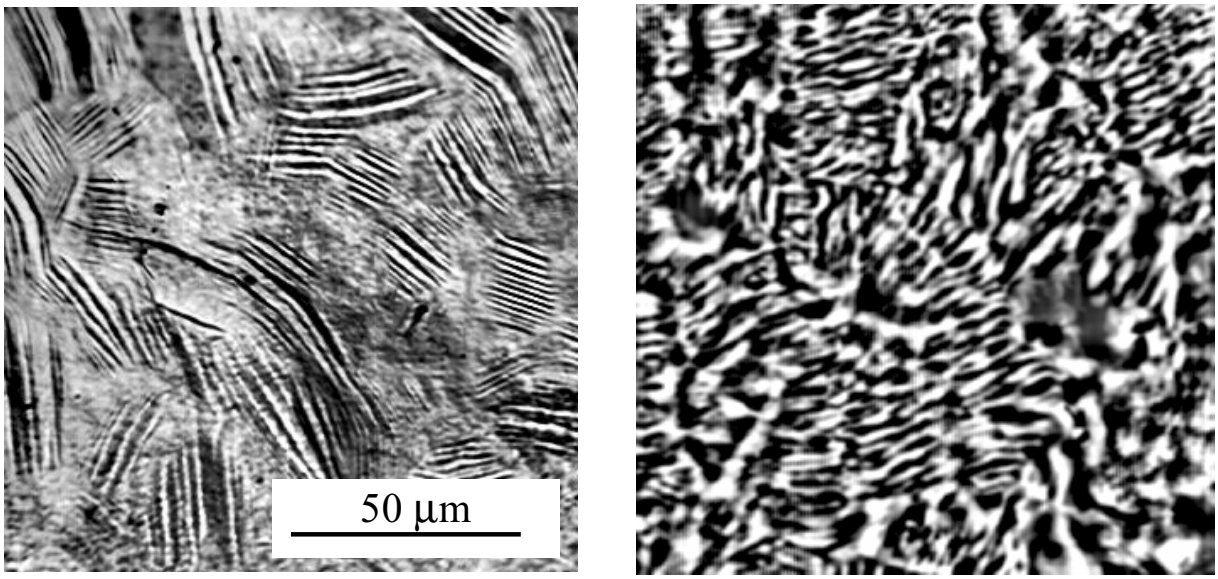

Fig. 10. Martensite and magnetic DS of the same area of rapidly quenched $Ni_{53}Mn_{23.5}Ga_{23.5}$ ribbon.

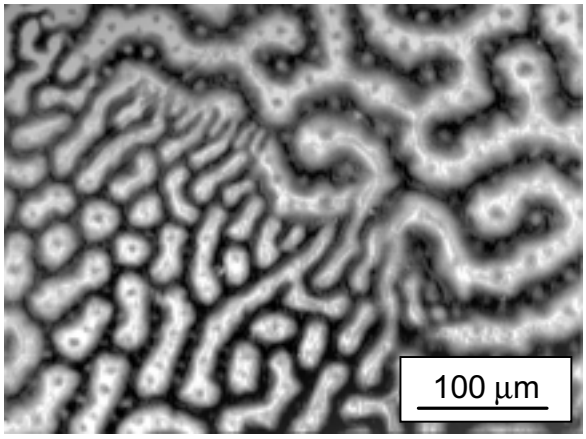

Fig. 11. 180-degree structure of magnetic domains of nanocrystalline $Ni_{52}Mn_{24}Ga_{24}$ film 5 μm thick prepared by magnetron sputtering.

site and austenite transition temperatures are close to each other.

3.2. Martensitic and magnetic domain structure

Figs. 3a and 3b show the surface relief of the martensite structure on different crystallographic planes of an oriented single crystal $Co_{48}Ni_{22}Ga_{30}$. The martensite plates pass continously through the whole crystal thus giving an evidence of the high perfection of the sample. By contrast, in polycrystalline samples more complex distribution of crystallite orientation and internal stresses results in an appearance of various intersections of different twin systems.

The martensite relief shown in Figs. 3a and 3b is formed after direct martensite transformation of the sample having a flat surface prepared by polishing at high temperature corresponding to the

austenite state. Otherwise, if the sample is flat polished in the martensite state, no relief is observed if the sample is not subjected to thermal cycling through the phase transition points. In this case the martensite structure is revealed with the aid of polarized microscopy making use of the optical anisotropy of the martensite phase (Fig. 4).

To reveal the magnetic DS and its relation to the martensite twins the method of ferrite-garnet magnetooptic films was used. Fig. 5 demonstrates the martensite relief and magnetic DS on the (110) plane of a $Co_{48}Ni_{22}Ga_{30}$ single crystal, while Fig. 6 shows the magnetic DS of the $Ni_{49}Mn_{29.7}Ga_{21.3}$ single crystal.

In polycrystalline samples with arbitrary crystallographic orientation of the crystallite surfaces the magnetic DS configuration becomes more complicated due to the effect of magnetostatic fields at the grain boundaries (Figs. 7 and 8)

4. CONCLUSIONS

The presented experimental results show that the main feature of the martensitic and magnetic DS of the ferromagnetic Heusler alloys under study is their interdependence. 180-degree magnetic domains are continous within the limits of the crystallites, which, in their turn, are divided into plane-parallel martensitic twin domains. The magnetization vector of magnetic domains becomes modulated by the martensite domains, because the *c*-axes of the latter, being the easy axes of magnetization, are oriented at the angles of 90° with respect to each other. As a result, the boundaries between martensite domains also play the role of 90-degree magnetic domain walls free of charges (of the Bloch type with div $M = 0$) (Fig. 9).

The configuration of the martensite and magnetic domain structures on different crystallographic planes depend on their orientation. The (001) plane is free of charges, and the magnetic domain walls are oriented symmetrically at 45° with respect to the martensite boundaries. In the (100) and (010) planes there are domains with magnetization vector alternating in the directions parallel and normal to the sample surface. For other arbitrary orientations the angular relations between the lines of domain wall intersections with the surface become more complicated.

The martensite structure of annealed rapidly quenched ribbons is qualitatively the same as for bulk samples (Fig. 10). The average magnetic domain width is considerably decreased to the values of the order of 1 mm. Observation of the magnetic DS of nanocrystalline films prepared by magnetron sputtering give an indication of uniaxial texture in a direction normal to the film plane (Fig. 11).

The general scheme of the coexisting martensite and magnetic DS of the investigated ferromagnetic Heusler alloys is well described by the model developed earlier for the case of polytwinned CoPt-type alloys [13].

REFERENCES

[1] A.N. Vasil'ev, V.D. Buchel'nikov, T. Takagi, V.V. Khovailo and E.I. Estrin // *Physics-Uspekhi* **46** (2003) 559.

[2] M.A. Marioni, R.C. O'Handley, S.M. Allen, S.R. Hall, D.I. Paul, M.L. Richard, J. Feuchtwanger, B.W. Peterson, J.M. Chambers and R.Techapiesancharoenkij // *J. Magn. Magn. Mater*. **290–291** (2005) 35.

[3] M. Wuttig, J. Li and C. Craciunescu // *Scr. Mater*. **44** (2001) 2393.

[4] C.M. Craciunescu and M. Wuttig // *J. Optoelectr. Adv. Mater.* **5** (2003) 139.

[5] C. Craciunescu, Y. Kishi, T.A. Lograsso and M. Wuttig // *Scr. Mater.* **47** (2002) 285.

[6] A.A. Likhachev and K. Ullakko // *Phys. Letters*. **A275** (2000) 142.

[7] Q. Pan and R.D. James // *J. Appl. Phys.* **87** (2000) 4702.

[8] Y. Murakami, D. Shindo and K. Oikawa // *Appl. Phys. Lett.* **82** (2003) 3695.

[9] Y. Kishi, C. Craciunescu, M. Sato, T. Okazaki, Y. Furuya and M. Wuttig // *J. Magn. Magn. Mater*. **262** (2003) L186.

[10] O.M. Korpusov, Yu.M. Smirnov, V.V. Koledov, A.B. Zaletov, S.A. Chigirinsky and D.S. Yulenkov // *Vestnik TvGU, ser. Fizika* **4** (2004) 81, In Russian.

[11] O.M. Korpusov, R.M. Grechishkin, V.V. Koledov, V.V. Khovailo, T. Takagi and V.G. Shavrov // *J. Magn. Magn. Mater.* **272-276** (2004) 2035.

[12] V.A. Chernenko, A.R. Lopez, M. Kohl, M. Ohtsuka, I. Orue and J.M. Barandiaran // *J. Phys.: Condens. Matter* **17** (2005) 5215.

[13] N.I. Vlasova, G.S. Kandaurova and N.N. Schegoleva // *J. Magn. Magn. Mater.* **222** (2000) 8474.