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The Magnetic Microstructure of YIG / GGG Films: Mossbauer Studies in the External Magnetic Fields

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The Mossbauer studies of yttrium iron garnet (YIG) films grown on the gadolinium gallium garnet (GGG) substrate was realized for different films thickness (2.85 and 5.42 μm) at the applied external magnetic field. It is shown that the main changes in the magnetic microstructure of YIG/GGG films are occurred at magnetic field value up to 2.20 kOe. The changes of the hyperfine magnetic fields on the iron nuclei have been investigated and its isotropic component was analyzed. It was determined that the domain structure changes (in particular, the domains width enlarging) under the applied magnetic field increase with the film thickness decreasing.

Keywords: Yttrium-Iron Garnet, Mössbauer spectroscopy, Magnetic field microscopy, Magnetic domain.

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Introduction

Epitaxial YIG/GGG films are widely used as material for magnetic spin waves generation in microwave techniques in the frequency range of 10^8-10^{11} Hz and for magnetical and optical applications as modulators, analyzers, correlators, deflectors [1]. The high performance of materials practical application involves the minimizing of electromagnetic waves extinction with high magnetic Q-factor. The solution of this problem involves the YIG magnetic properties understanding especially its domain structure transformation with external influences. The aim of this work is the investigation of epitaxial YIG/GGG films domain structure and its dependence on the external magnetic field and films thickness. Another task is to find the relationship between the magnetic microstructure parameters and the domain structure of films with different thicknesses using Mössbauer spectroscopy.

I. Materials and methods

YIG films was grown on gadolinium gallium garnet substrate (GGG) with (111) crystal orientation from melt solution of Y_2O_3 - Fe_2O_3–La_2O_3-Ga_2O_3 garnet forming oxides and PbO – B_2O_3 solvent. When growing films the substrate was horizontally placed in the melt solution with rotation at a frequency of 60 rpm. The growth temperature was in a range of 1236-1248 K. Growing procedure was performed on the "Karat" Scientific Production Enterprise (Lviv). The five-zone furnace for the liquid phase epitaxy (LPE) was used. The superficial layer (film-air interface) was etched with a mixture of nitric and acetic acids with the next flushing with deionized water. For increasing of the Mössbauer effect probability iron oxide Fe_2O_3 enriched with Fe^{57} isotope up to 10 at. % was used.

The investigation of magnetic and electrical superfine interactions in LaGa: YIG/GGG was obtained by the method of conversion electron Mössbauer spectroscopy (CEMS). The calibration of the spectra were realized relatively to α-Fe. The zero-level instability and registration error did not exceed 0.05 mm/s, the line width for the α-Fe was about 0.29-0.30 mm/s. ^{57}Co source in chromium matrix was used. Atomic force microscopy (AFM) was used for investigation of a film morphology. The magnetic domain structures were observed by the magnetic force microscopy (MFM) [2].

II. Results and discussion

The characteristics of ferrimagnetic ordering in YIG depends on the electrostatic fields on the iron nuclei formed both lattice ions and its own electron shells. The non-equivalent positions of iron in YIG lattice correspond to different values of angle θ between the direction of the effective magnetic field on the ^{57}Fe nucleus and the direction of the resulting electric field gradient (EFG) in which this nucleus is located. Since the

tetrahedral positions have symmetry axis of fourth order, and octahedric ones of the third order, then the garnet structure have 7 crystalline and magnetic non-equivalent sites for Fe^{3+} ions with 7 components of the Mössbauer spectrum correspond to these positions. For monocrystalline YIG films epitaxially grown on a (111)-oriented GGG substrate the number of non-equivalent positions of Fe^{3+} ions is reduced to three. The polar angle between the directions of the EFG on the nucleus and the direction of the gamma rays (axis [111]) is 54°44′ for all ^{57}Fe nuclei located in *d*-sublattice (tetrahedral site). At the same time, for 1/4 of the ^{57}Fe nuclei in *a*-sublattice (octahedral site) this angle is zero and for another 3/4 of ^{57}Fe nuclei in *a*-sublattice this angle is 70°32′. In this case, two types of interactions can be distinguished which have approximately equal intensity - electric quadrupole and magnetic dipole ones.

The method of conversion electron Mössbauer spectroscopy was used for the separation and identification of close non-equivalent crystallographic positions of resonance nuclei. The measurements were carried out in external magnetic fields of varying intensity. This made it possible to investigate the processes of forming the resulting magnetic moment vector and to study its behavior when changing the magnitude of the magnetic field [3]. The information about magnitude and direction of the Heff allow predicting the domain structure properties as well as its re-magnetization, angular spin configurations and relaxation processes. Applying of an external magnetic field parallel to (111) easy magnetization direction allowed to separate the anisotropic part of a hyperfine magnetic field on the nucleus when the magnetization vector orientation changes relatively to the axes of crystalline symmetry [4]. The direction of external magnetic field formed by a system of neodymium magnets was perpendicular to the sample surface. In this case the magnetic moments of individual iron ions are oriented along the direction of the external magnetic field. Four different values of applied field strength were 1.50, 2.20, 2.55 and 2.80 kOe, respectively.

CEMS spectra of the YIG/GGG samples with a thickness of 2.85 and 5.42 μm in external fields are shown in Fig. 1.

The presence of the second and fifth spectral lines in the absence of an external magnetic field indicates the close orthogonal nature of the mutual placement of the propagation direction of γ-quanta and the axis of light magnetization.

The analysis of Mosssbauer spectra lines intensity allows to calculate the angle β between the magnetic moment of the Fe^{57} nucleus and the perpendicular to the sample plane [5]: $I_{1,6} : I_{1,6} : I_{1,6} = 3 : \frac{4\sin^2 b}{1+\cos^2 b} : 1$. The applying of an external magnetic field up to 2.8 kOe causes the reorientation of film's resulting magnetization vector normally to sample plane. Experimental spectra in both cases were approximated by the superposition of four sextets and one doublet component that corresponds to the Fe^{3+} ions in the paramagnetic state. Each component was described such parameters: amplitude, isomer shift δ, magnetic field H_{eff}, axial component of EFG U_{zz}, polar β and azimuthal α angles of the H_{eff} orientation relative to second and fifth lines of EFG; polar θ and azimuthal φ angles of the γ-quantum beam orientation. The application of external magnetic field simplifies the finding of parameters set for each non-equivalent position. The magnetization vectors orientation of YIG samples magnetic sublattices were calculated for external magnetic field different intensities.

The relative content of iron ions in the paramagnetic state observed for both films is about 2.5-3.0%. The paramagnetic phase is a result of oxygen non-stoichiometry and the presence of impurity atom from the melt solution at the final stages of the epitaxy. The Pb^{2+} and Pb^{4+} occupy octahedral positions by displacing Fe^{3+} with a probability of 0.4 and 0.3 respectively [6]. Another uncontrolled admixture is the Pt^{4+} ions, which occupy only *a* -position. CEMS spectra is a result of emission of conversion electrons from the near-surface layer up to 100 nm.

The major changes in the magnetic microstructure occur prior to external field intensity of 2.20 kOe. It also follows from the analysis that this conclusion does not depend on the thickness of the film. It was determine that

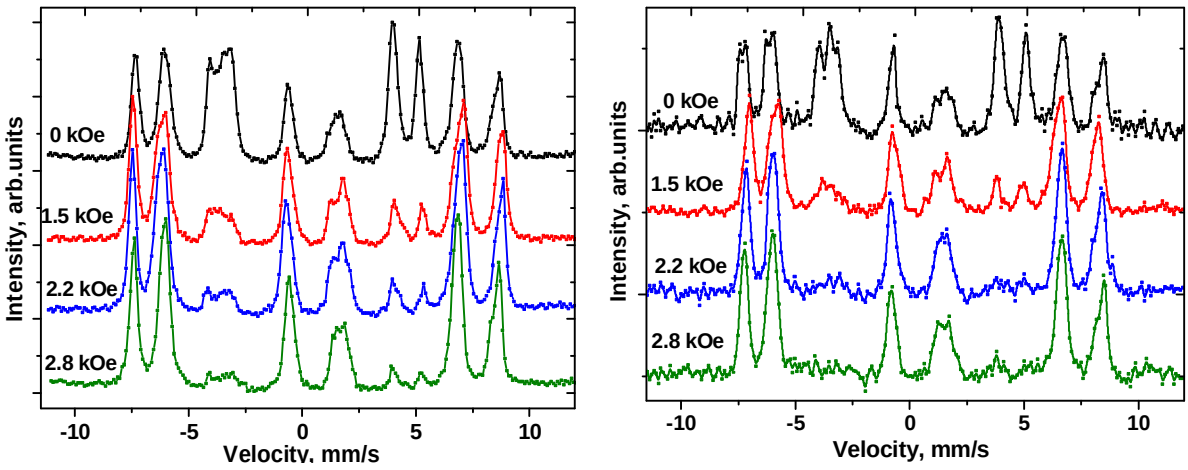

Fig. 1. Mössbauer spectra of the YIG/GGG films with a thickness of (a) 2.85 and (b) 5.42 μm in external magnetic field.

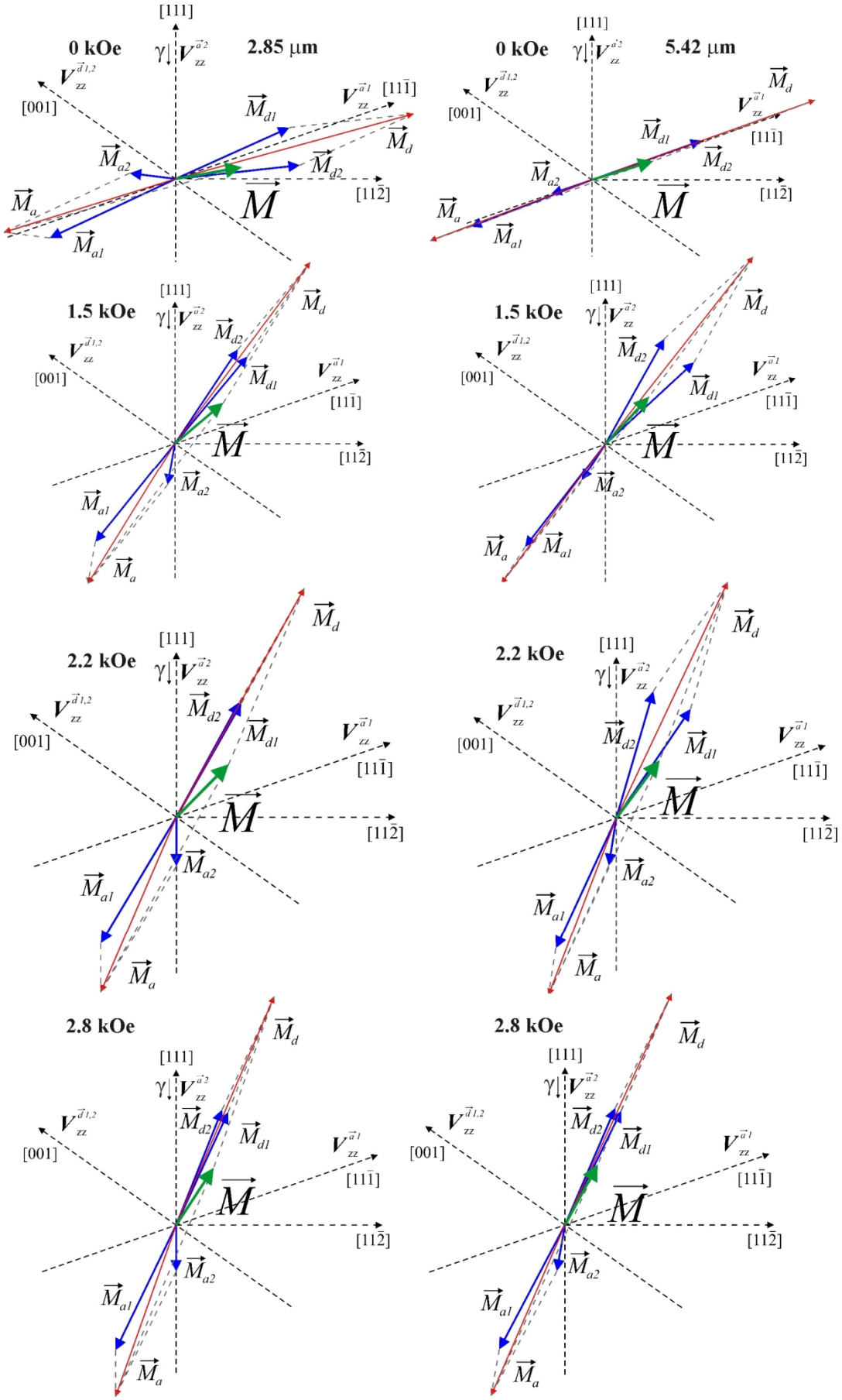

Fig. 2. The magnetization orientation for YIG epitaxial film of different thickness (2.85 and 5.42 μm) in external magnetic field.

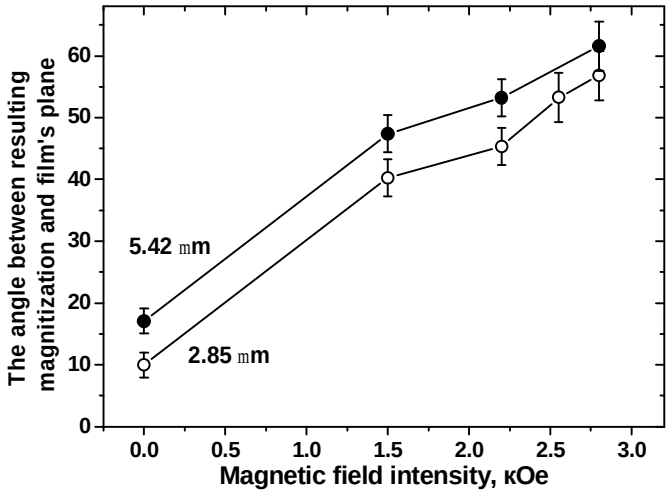

Fig. 3 The orientation of the resulting magnetization depending on the external magnetic field intensity for epitaxial films YIG/GGG with different thicknesses.

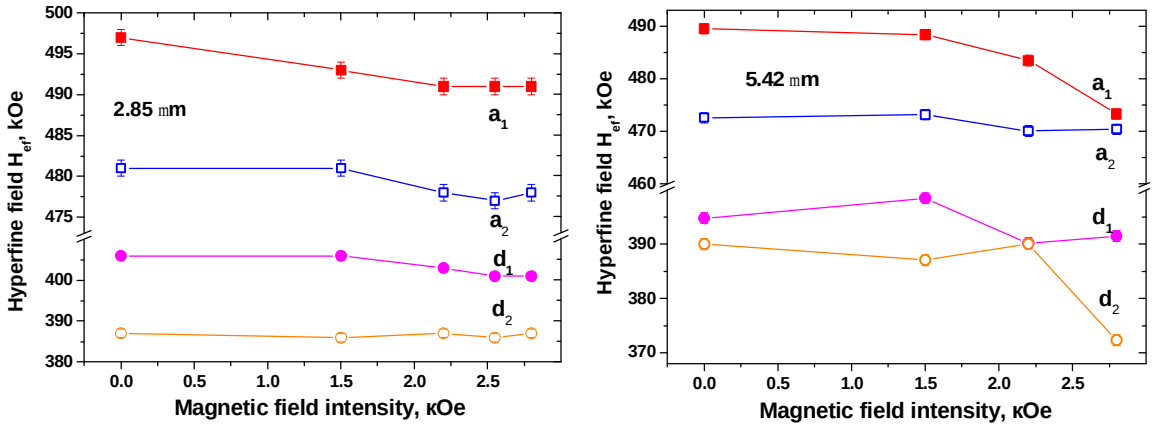

Fig. 4. The dependence of the hyperfine magnetic fields on the Fe^{57} nuclei in various magnetic non-equivalent positions of YIG/GGG on the external magnetic field intensity for film thickness of 2.85 (a) and 5.42 μm (b).

the resultant magnetization vector orientations in the absence of an external field are close to film plane (10^o and 16^o for 2.85 and 5.42 μm film thicknesses). The increasing of external magnetic field causes the non-linear increasing of angle between sample plane and resultant magnetization vector with saturation at about $70\text{-}75^o$ (fig.).

The effective magnetic fields on the Fe^{57} nuclei for two octahedral and two tetrahedral positions for both film thicknesses decrease with the external magnetic field increasing in the range of 0-2.8 kOe (Fig. 4). It is worth noting that for the case of a thicker sample, the absolute values of the effective magnetic fields are relatively lower, and the rate of their decrease with the increase in the external field is higher.

The hyperfine magnetic field on the resonant nucleus is formed from several components, which characterize the interaction between the magnetic moment of the nucleus nd the magnetic field of surrounding electrons [7]. The interaction of s-electrons with the nucleus makes the maximum contribution to the value of H_{eff}. The super-transfer field H_{ST} component is responsible for the interaction between magnetic ions separated by paramagnetic ligands. This field is determined by the number and geometry of the Fe-O-Fe chains. There are also fields formed by the orbital momentum and spin moment of the atomic electron *s*-shell (H_L and H_S, respectively). The field of dipole-dipole interaction of the Mosssbauer nucleus and nuclei of surrounding atoms (H_{SD}) also makes its contribution. The contribution of the Lorentzian and demagnetizing fields, for the films of ferrite garnets due to the small spin-orbital interaction, is negligible [8]. Among the listed terms, only the orbital HL and spin dipolar HSD components have anisotropic character in relation to the spatial orientation of the magnetic moment. The field H_L caused by the orbital motion of electrons, in our case is close to zero, since the iron ions in the garnets are in the orbital singlet state L = 0 [9]. The anisotropic component of the spin dipole field is the function of the angle β between the main components of the EFG tensor and the orientation of the magnetic moment on the nucleus: $B^{SD}(b) = B_A\left(3\cos^2 b - 1\right)$ [9].

The relationships between the hyperfine fields H_{ef} for the iron nuclei in the a_1-position and the corresponding angles β are shown in Fig. 5. For the case

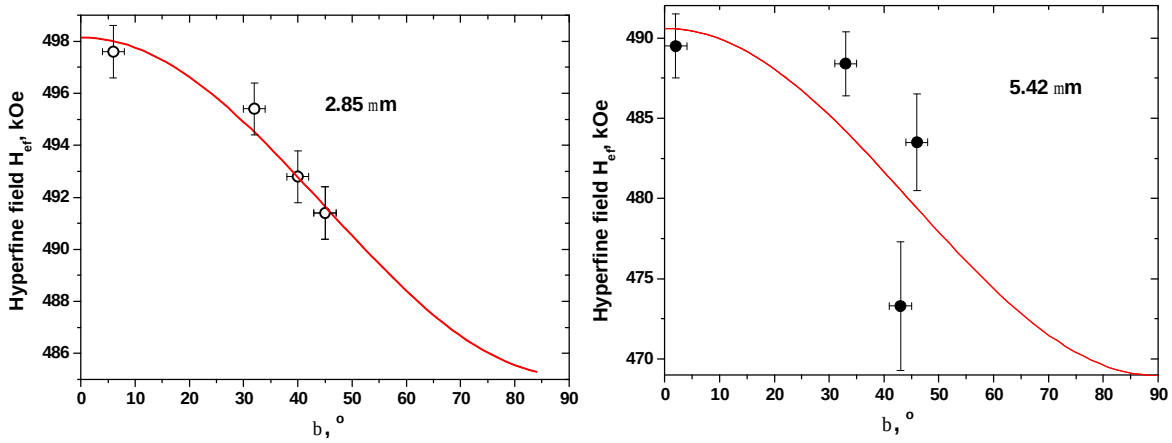

Fig.5. The hyperfine field on the Fe^{57} nucleus in the a_1-position as a function of the angle β between the main components of the EFG tensor and the orientation of the magnetic moment on the nucleus.

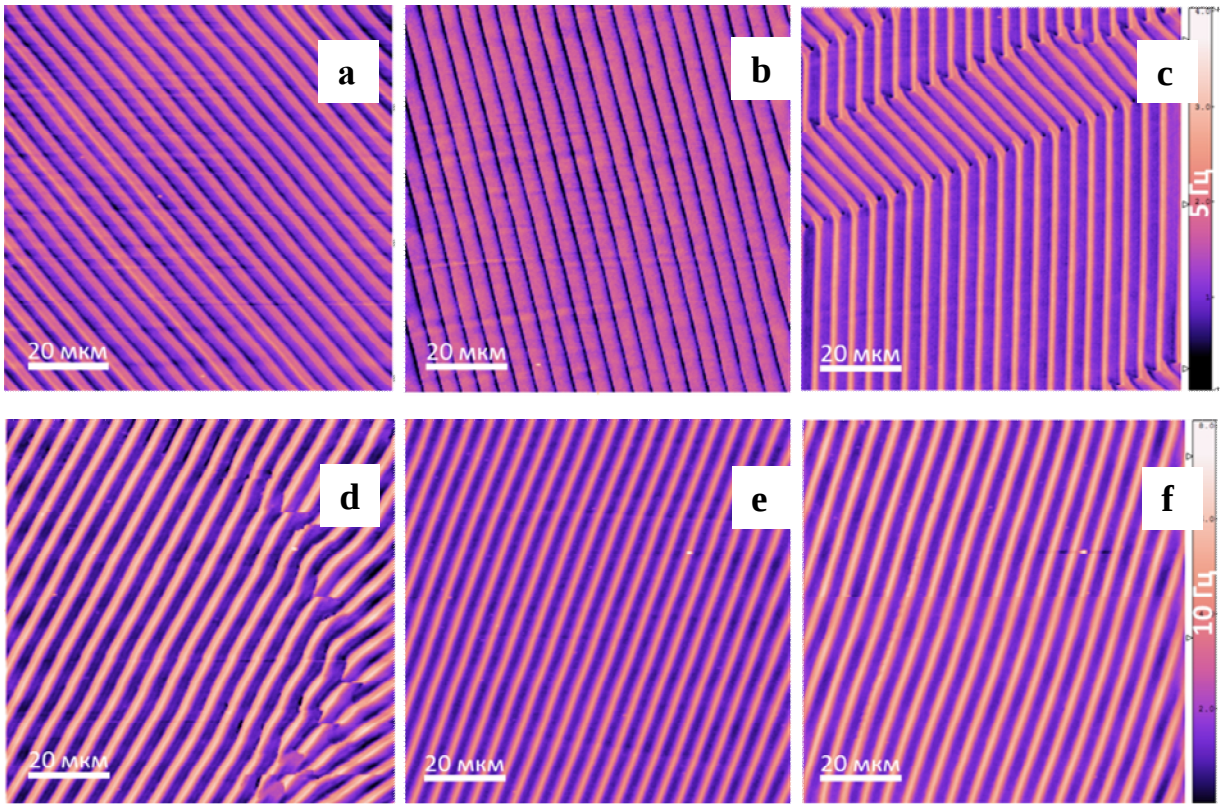

Fig. 6. Magnetic force microscopy maps of the field gradient distribution of the YIG/GGG epitaxial films with a thicknesses of 2.85 and 5.42 μm (before the external magnetic field is applied (a, d), in the field (b, e) and after the field action is terminated (c, f)).

of film thickness of 2.85 μm the accuracy of the fixation of the experimental spectrum made possible the fitting procedure. The calculated value of hyperfine field isotropic component is 489.5±0.5 kOe with the value of the parameter B_A =4.3 ± 0.5 kOe. For the case of film thickness of 2 5.42 μm the accuracy obtained results is lower and calculated values are 476.2±6.6 kOe for isotropic part of the hyperfine field and 7.2±5.5 kOe for B_A parameter.

The observed increasing of isomeric shifts values for iron at the increasing of external magnetic field intensity for both samples can be explained by changes in the electron density on the Fe^{57} nuclei. The isomeric shift depends on the difference in charge nucleus radii in the principal and excited states and electron densities on the resonant nucleus in the absorber and source. The increasing in isomeric shifts indicates a decrease of *Fe-O* chemical bond covalence and can be explained by the growth of the distance of the exchange interaction and the change in the surrounding of the iron atoms, the decrease in the overlapping of the Fe^{3+} and O^{2-} electron shells, and, consequently, the localization of the wave

function of *s*-electrons on the Fe^{57} nucleus.

The domain structure of the investigated samples is shown in Fig. 6. The observed domain walls (Bloch and Neel walls) correspond to gradual change in the direction of the magnetization vector between the two domains. The shift of the domain wall under the magnetic field is a result of the orientation change of local magnetic moments in the domains separated by this domain wall. The external magnetic field leads to rotation of the magnetization in the Bloch walls. The comparison of magnetic domains distribution on the surface of films with different thickness was performed before the external magnetic field is applied (a, d), during applying (b, e) and after the field application (c, f). Scanning in a magnetic field allows investigating the processes of samples` magnetization redistribution. The magnetic induction value was 4 mT. For film thickness of 2.85 μm the broadening of domains is observed at application of a magnetic field and after elimination there is a bend of the domain walls. For the epitaxial structure with thickness of 5.42 μm the influence of the external field is weaker, the domain broadening is not observed, however, the alignment of the domain structure takes place. The alignment or bending of the domain structure are caused by the presence of Bloch vertical lines in the domain walls, which, when moving domains, slow down the movement of those sections of the wall where they are located. There are also Bloch points, which are characterized by the fact that all possible orientations of magnetization are on the sphere of an infinitely small radius with a center at the Bloch point.

An example of the Bloch point is the magnetic configuration in the vertical Bloch line at the junction of two of its aeas having opposite directions of magnetization rotation around the z axis. At the Bloch point, the wall and the Bloch line "pull" to zero. Bloch lines and points determine the observed changes in the YIG domain structure under the action of a magnetic field [8].

Conclusions

1. Comparison of the Mössbauer studies results of the magnetic microstructure of YIG/GGG films of different thicknesses (2.85 and 5.42 μm) when applying an external magnetic field was done. It was established that the main changes in the magnetic microstructure are observed until the intensity of the external field is 2.20 kOe.
2. The changes of the hyperfine field on the iron nuclei have been observed. The isotropic component of hyperfine field values are 489.5 ± 0.5 and 476.2 ± 6.6 kOe for films YIG/GGG with thicknesses of 2.85 and 5.42 um, respectively.
3. It was established that the transformation of the band domain structure (in particular, the increase of the domain width) at the applied external magnetic field is more manifested for a film of a smaller thickness, which can be interpreted as a result of decreasing the influence of the block structure of the film when its thickness increase

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[1] S.B. Ubizskii, Journal of Magnetism and Magnetic Materials 195, 575 (1999).
[2] I.M. Fodchuk , I.I. Gutsuliak , R.A. Zaplitniy , S.V. Balovsyak , I.P. Yaremiy, O.Yu. Bonchyk, G.V. Savitskiy, I.M. Syvorotka, P. M. Lytvyn, Semiconductor Physics Quantum Electronics & Optoelectronics 16(3), 246 (2013).
[3] B.K. Ostafiichuk, V.M. Tkachuk, O.M. Tkachuk, V.D. Fedoriv, Metallofyzyka y noveishye tekhnolohy 22(11), 11 (2000).
[4] Y.P. Suzdalev, Dynamycheskye effekty v hamma– rezonansnoi spektroskopyy (Atomyzdat, M.,1979).
[5] J. Piekoszewski, L. Dąbrowski, J. Suwalski, S. M. Ągwa, physica status solidi (a) 39(2), 643 (1977).
[6] B.K. Ostafiychuk, V.A. Oleynik, V.M. Pylypiv, B.T. Semen, L.M. Smerklo, B.I. Yavorskiy, V.I. Kravets, I.V. Koval, AN USSR, Institut metallofiziki 1, 91 (1991).
[7] V.Y. Nykolaev, V.S. Rusakov, Messbauerovskye yssledovanyia ferrytov (Yzd-vo Mosk. un-ta M. 1985).
[8] V.A. Bokov, Fyzyka mahnetykov: Ucheb. posobye dlia vuzov (Nevskyi Dyalekt, SPb., 2002).
[9] M. Bonnet, A. Delapalme, H. Fuess, M. Thomas, Acta Crystallographica Section B 31(9), 2233 (1975).

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Мессбауерівське дослідження магнітної мікроструктури плівок ЗІГ/ҐҐ: при накладанні зовнішнього магнітного поля

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Проведено порівняння результатів месбауерівського дослідження плівок залізо-ітрієвого гранату (ЗІГ), вирощених на підкладці галій-гадолінієвого гранату (ҐҐ) різної товщини плівок (2,85 і 5,42 мкм) при накладанні зовнішнього магнітного поля. Показано, що основні зміни магнітної мікроструктури плівок ЗІГ/ҐҐ відбуваються при значенні зовнішнього магнітного поля 2,20 кЕ. Досліджено зміни надтонких магнітних полів на ядрах заліза та проаналізовано його ізотропне компоненту. Встановлено, що трансформація доменної структури (зокрема, збільшення ширини доменів) при накладанні зовнішнього магнітного поля збільшується зі зменшенням товщини плівки.

Ключові слова: залізо-ітрієвий гранат, месбауерівська спектроскопія, магнітно-силова мікроскопія, доменна структура.