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Growth and microwave properties of layered ferrogarnet structures

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The ferrogarnet structures consisting from one- to three- layers of monocrystalline yttrium-iron garnet $Y_3Fe_5O_{12}$ (YIG) films, two- layered YIG - La,Ga:YIG and two- layered $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$-YIG structures were grown by liquid-phase epitaxy (LPE) method on gadolinium - gallium garnet $Gd_3Ga_5O_{12}$ (GGG) substrates of (111) orientation. The obtained layered ferrogarnet structures were studied by the methods of ferromagnetic resonance (FMR) and magnetostatic wave (MSW) interference. The two- and three- layered YIG structures have a wide FMR line width (ΔH). For the three- layered YIG structures with the total thickness of 68-102 μm ΔH = 5.7- 11.5 Oe. The line width ΔH = 0.34 – 1.22 Oe correspond to the two- layered $(Y,Sm,La)_3(Fe,Ga)_5O_{12}$ – YIG structures with thicknesses from 3 to 65 μm. Individual layers in all structures are characterized by similar or different saturation magnetizations ($4\pi M_s$).The frequency MSW separation in the YIG - La,Ga:YIG layered structure was observed. It was shown that the propagation losses of MSW in one- and two- layered structures increase with decreasing wavelength of MSW and transition to a two- layered structure.

Key words: layered ferrogarnet structures, ferromagnetic resonance, magnetostatic wave.

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Introduction

The problem of miniaturizing ultra-high-frequency (UHF) electronic devices cannot be achieved without qualitatively new materials, such as monocrystalline ferrogarnet films and layered structures. The employment of the ferrogarnet films for replacing of bulk monocrystalline materials allows the use of planar technology for making UHF- components. The operating principle of such components is based on the propagation of magnetostatic waves (MSW) in the ferrite material [1,2,3]. The propagation of the MSW characteristics depend on the degree of the internal magnetic field homogeneity in the crystal. The value of losses is directly related to the quality of ferrogarnet films, which is determined by the ferromagnetic resonance (FMR) line width (ΔH), homogeneity of chemical composition, the thickness, and the magnetic parameters over the film area.

The multilayer ferrogarnet structures are attracting attention due to the wide possibilities manipulating of the magnetostatic and spin waves characteristics [4,5,6]. Therefore, obtaining such structures and studying their physical properties is an actual task for microwave electronics. A layered ferrogarnet structure can be considered a structure which have layers width similar or different chemical composition and, consequently, magnetic parameters.

To determine the influence of layer magnetic properties in ferrogarnet structures on the propagation of MSW, we have grown structures consisting of one- to three- layers of yttrium-iron garnet $Y_3Fe_5O_{12}$ (YIG) films, two-layered YIG-La,Ga:YIG structures and two-layered $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$-YIG structures.

The one-layer monocrystalline YIG film, in particular, can be considered as having a layered structure, which is related to the heterogeneity of the film chemical composition across the thickness [7], which caused by the peculiarity of the technology of their growth. In fact, a ferrogarnet film has of three layers.

This is the transition layer on the boundary between the film and the substrate (FS) and the surface layer (SL) on the film-air boundary and the inner middle layer, which is the most homogeneous in chemical composition. The FS layer is enriched of Gd^{3+} and Ga^{3+} ions diffusing from the substrate during the growth of the ferrogarnet film. The surface layer is enriched of Pb^{2+} and Pb^{4+} ions entering from the $PbO\text{-}B_2O_3$ solvent [8]. Impurity ions modify the chemical composition of PP and PS layers, and hence the saturation magnetization ($4\pi M_s$) and the line width ΔH of these layers.

The Gd^{3+} and Pb^{2+} ions, which have large ionic radii, replace the Y^{3+} ions in the dodecahedral sublattice (c), and with the Pb^{4+} and Pt^{4+} ions enter into the octahedral sublattice (a), increasing the film saturation magnetization. The appearance of Pt^{4+} ions is due to their penetration into the film from the platinum crucible. The Ga^{3+} ions, having a tendency to tetrahedral positions (d), replace Fe^{3+} ions from them, leading to a decrease saturation magnetization of the film.

The spin magnetic moment of the Gd^{3+} ions, entering the (c) sublattice, has the opposite direction to the total moment of the a-d- sublattice. Therefore, the exchange interaction between the c- and a-d- sublattices decrease the magnetization of the film.

Therefore, we can conclude that Gd^{3+} ions have a complex effect on the $4\pi M_s$ ferrogarnet film parameter. The penetration of the Gd^{3+} into the YIG film increases the ΔH line width. As shown in [9], the ΔH line width of gadolinium ferrogarnet depends on the saturation magnetization and increases strongly near the compensation point. The same is observed in the $Y_{3-x}Gd_xFe_5O_{12}$ ferrite films [10]. The Pb^{2+} and Pb^{4+} ions in the ferrogarnet film make a no significant contribution to the increase of the ΔH line width. At the same time, their presence in the crystal lattice cause the appearance of Fe^{4+} and Fe^{2+} ions [11,12], respectively. The electronic exchange between Fe^{4+} and Fe^{2+} ions increases the value of ΔH [13].

It can be expected that as the film thickness decreases, the ΔH parameter will be increasingly affected by the thickness of the transition layers. In addition, the reduction in film thickness limits the use of microwave signals.

The obtaining of quality thick ferrogarnet films is related to the difficulties caused by the appearance of mechanical stresses in the film structures that reduce their magnetic properties. To solve this problem, we used of stepwise growth of film structures. According to the method described in [14], all layered structures were investigated by the method of interference of magnetostatic waves.

I. Experimental methods and results discussion

Ferrogarnet structures were grown by liquid-phase epitaxy (LPE) on monocrystalline substrates of gadolinium-gallium garnet $Gd_3Ga_5O_{12}$ (GGG) of orientation (111) from a supersaturated melt-solution (MS) of ferrite charge using the $PbO\text{-}B_2O_3$ solvent. The substrates were cut from a single crystal of GGG (GGG lattice parameter $a_s = 12{,}3821$ Å), grown by the Chohralsky method, with the following mechanical and chemical treatment to 14th purity class. The thickness of the substrates was 0,5 mm with a defect density of no more than 1 cm^{-2} per substrate. The growth of ferrogarnet structures has been carried out on an automated system. The temperature in the furnace zones was controlled to an accuracy of ± 0.1 K.

The two- and tree- layered YIG, $(Y,Sm,La)_3(Fe,Ga)_5O_{12}$ - YIG and YIG - La,Ga:YIG layered structures were grown in three and two steps, respectively.

The platinum crucibles were used to melt of the charge and grow layered structures. The saturation temperature (T_s) of the MS was determined as the temperature at which the film growth on the substrate began. After the homogenization process at T = 1420 K for 16 hours, the melt -solution was cooled to the growth temperature (T_g), which was lower than the saturation temperature by $\Delta T = T_s - T_g$ (degree of supercooling of the melt-solution).

The horizontally fixed substrate of GGG, together with the special mixer for MS, was in a one-way rotational motion with a frequency of approximately 120 rpm during the process of ferrogarnet layer growth and layered structure formation. The mixer design involves capturing the MS fresh portion and directing it to the substrate, thus forming a flat diffusion layer near the surface of the grown film. As a result, ferrogarnet layers and, consequently, multilayer structures had the highest in-plane homogeneity of such important parameters as ferrite layer thickness and ΔH line width [15].

The thickness h of the ferrogarnet layers was measured by the interference method for $h \leq 10$ μm, and the thickness of layers when $h > 10$ μm was measured using an optical microscope. The FMR line widths ΔH of ferrogarnet films and layered structures was determined by the nondestructive method in the frequency range of 14 - 15 GHz [16]. Their composition was determined using the Camebax electronic microanalyzer.

The melt-solution for the growth of layered YIG-structures had the following composition (in mol %): PbO – 86.46; B_2O_3 – 5.54; Fe_2O_3 – 7.37; Y_2O_3 – 0.63. We were growing these structures in several stages. At each stage, the degree of yttrium substitution by lead was different. As a result, we obtained layered YIG structures with different lattice parameters in each layer. The saturation magnetization YIG layers were changed by altering the growth conditions.

It is known that the greater the degree of supercooling of the MS, the greater the growth rate (f_g,) of the ferogarnet film. With an increase in the growth rate, the degree of lead ions entering the film increases, which changes the composition and, accordingly, the saturation magnetization of the ferrite film.

The growing regimes of the three- layer YIG structures and their associated parameters are shown in Table 1.

Table 1 shows that for all three samples, the growth rate of the YIG layer increases with an increase in the degree of MS supercooling (ΔT). Table 1 also shows the lattice parameters (a_f) of the individual layers and their thicknesses (h), the total thickness and line width ΔH of the three-layer YIG structure. As can be seen, the ΔH parameter of a three-layer YIG structure increases with an increase in its total thickness.

Table 1.

The growing regimes of the three- layered YIG structures and their associated parameter

№ sample	The growing regimes			The lattice GGG parameter a_s, Å	The layer lattice parameter a_f, Å	The thickness of the each layer, µm	Total thickness of the 3-layer structure, µm	ΔH, Oe
	T_g, K	ΔT, K	f_g, µm/min					
1	1193	10	0.53	12.3821	12.3767	10.0	68.0	5.7
	1188	15	0.82		12.3798	51.0		
	1173	25	1.30		12.3820	7.0		
2	1188	15	0.85		12.3768	23.0	83.0	7.3
	1193	10	0.54		12.3800	8.0		
	1173	25	1.34		12.3791	52.0		
3	1173	25	1.31		12.3769	59.0	102.0	11.5
	1188	15	0.83		12.3810	36.0		
	1193	10	0.53		12.3823	7.0		

The FMR spectra of a three-layered and one- layered YIG structure with the thickness of 68 µm and 10 µm are shown in Fig.1a and 1b, respectively.

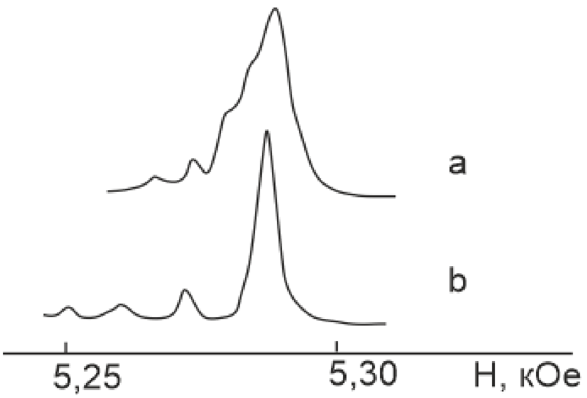

Fig. 1. The FMR spectra for a three- layered YIG structure with a thickness of 68 µm (a) and a one- layered YIG structure with a thickness of 10 µm (b).

As can be noted, the value of the ΔH parameter of three- layered YIG structures negatively affected by the heterogeneity of the chemical composition in the transition layers. In addition to the layered YIG structures, the two- layered ferrogarnet structures of other compositions were grown.

The ferrogarnet layers of the composition $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ were grown on the GGG substrates. These ferrogarnet monocrystalline layers have cylindrical magnetic domains (CMD). Since the crystal lattice parameter of this garnet ($a_{f(CMD)}$ = 12.373 Å) is close to the parameter of the YIG ($a_{f(YIG)}$ = 12.376 Å). This allowed to grow YIG layers with a thickness of $h \leq 65$ µm on $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ layers. In this way, we obtained two- layered $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$-YIG structures.

Tab. 2 shows the crystal lattice parameters, thickness and the FMR line width ΔH of YIG in $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$-YIG structure. As we can see, that the lattice parameters of these layers are similar in value (Tabl. 2), which reduces the mechanical stresses in both layers. Therefore, it is possible to grow rather thick YIG layers up to 65 µm with relatively small ΔH = 0.34-1.22 Oe.

According to the data in Table 2, it can be concluded that the $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ layer does not significantly influence on the ΔH of YIG layer.

For the obtained YIG- La,Ga:YIG layered structures, the thickness of the YIG layer was h = 10 µm, and La,Ga:YIG – h = 7 µm. The La,Ga:YIG films with a saturation magnetization of 4πMs = 1400 Gs were also grown directly on GGG substrates [17,18,19].

Fig 2a, 2b, and 2c show typical interference images of MSW for the layered YIG structures, one- layered La,Ga:YIG structure, and two-layered YIG-La,Ga:YIG structure, respectively

Since the saturation magnetization of the YIG ($4\pi M_s$= 1750 Gs) is much higher than the saturation magnetization of $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ ferrogarnet ($4\pi M_s$ = 200 Gs), their resonance frequencies are differ greatly in value.

For the $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ -YIG structures, the significant attenuation of the MSW is due to the presence of the boundary layer between the $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ and YIG layers, containing a significant quantity of the Sm^{3+} rare earth ions with a large orbital moment.

In Fig. 2c we observe the frequency separation of the interference images of MSW for the YIG-La,Ga:YIG structure. This can be explained by the different magnetizations of the YIG and La,Ga:YIG layers that form it. Obviously, the presence of a transition layer between layers with different magnetizations leads to distortions of the interference images MSW in the YIG-La,Ga:YIG structure (Fig. 2c) compared to those shown in Fig. 2a and 2b for individual layers.

Table 3 shows the calculated values of the MSW propagation losses for individual layers and layered structures, which were obtained from the values of the maximums and minimums of the interference image.

As can be seen from Table 3, the propagation losses of the MSW in one- and two- layered structures also increase with decreasing MSW wavelength and transition to a two- layered structure.

Table 2.

The crystal lattice parameters, thickness and the ΔH line width of the YIG layer in {Y,Sm,Lu}$_3$(Fe,Ga)$_5$O$_{12}$-YIG structure

№ sample	$a_{f(\mathrm{CMD})}$, Å	$a_{f(\mathrm{YIG})}$, Å	The thickness of the YIG layers (h), μm	ΔH, Oe
1	12.3774	12.3768	3.0	0.34
2	12.3774	12.3768	5.0	0.41
3	12.3772	12.3765	15.0	0.65
4	12.3771	12.3770	37.0	0.80
5	12.3771	12.3770	65.0	1.22

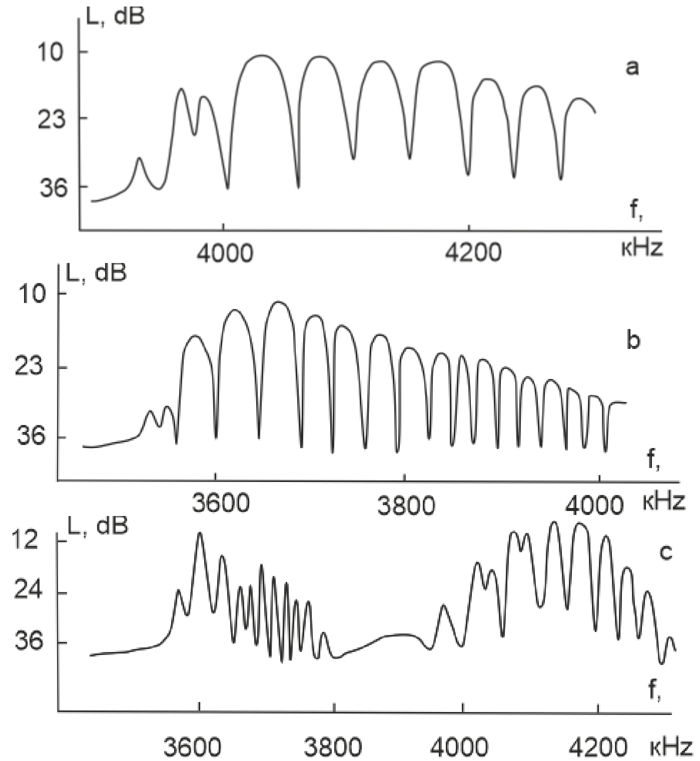

Fig.2. Typical interference images of MSW for the YIG structures (a), one- layered La,Ga:YIG structure (b) and two- layered YIG-La,Ga:YIG structure (c).

Table 3.

The MSW propagation losses in the one- and two- layered YIG structures, La,Ga:YIG layer and in two- layered YIG-La,Ga:YIG structures

The wave number k, см$^{-1}$	The propagation losses α, mm^{-1}			
	The one- YIG layer	The two- layered YIG structure	The one- La,Ga:YIG layer	The two- layered YIG- La,Ga:YIG structure
30	0.10	0.21	0.09	0.17
90	0.11	0.22	0.11	0.19
200	0.22	0.27	0.10	0.21

Conclusions

The method of epitaxial growth of layered structures from ferrogarnet monocrystalline layers with similar or different chemical compositions has been realized. The method of layer-by-layer growth makes it possible to produce ferrogarnet YIG structures with total thicknesses of 68 – 102 μm with the FMR line width ΔH = 5.7 – 11.5 Oe.

The composition of YIG layers can be changed due to the introduction of impurity ions Gd^{3+}, Ga^{3+}, Pb^{2+}, Pb^{4+}, Pt^{4+} by changing the technological growth conditions. The entry of impurity ions into the YIG layer increases with an increase in the degree of supercooling of the melt-solution.

In the two- layered {Y,Sm,Lu}$_3$(Fe,Ga)$_5$O$_{12}$-YIG structure no observed significant influence of {Y,Sm,Lu}$_3$(Fe,Ga)$_5$O$_{12}$ layer on the ΔH parameter of YIG layers due to a difference in the saturation magnetizations of ferrogarnet layers. The ΔH = 0.34 – 1.22 Oe correspond to the structures with thicknesses from 3 to 65 μm.

The frequency separation of the MSW in the two- layered YIG - La,Ga:YIG structure is established.

The MSW propagation losses increase from one- to two - layered YIG structure and from La,Ga:YIG to YIG - La,Ga:YIG structure with the decrease of MSW wavelength.

The use of two- layered ferro-garnet structures of YIG - La,Ga:YIG in MSW- devices provides an opportunity to expand the frequency range of their operation in comparison with single- layered ferrogarnet structures.

The obtained thick ferrogarnet structures of YIG are considered as three-layer structures that have a smaller range of operating frequencies compared to the two-layered YIG - La,Ga:YIG structure, but can be used for processing microwave signals of high power.

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Вирощування і мікрохвильові властивості шаруватих ферогранатових структур

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Методом рідкофазної епітаксії на підкладках з гадоліній-галієвого гранату $Gd_3Ga_5O_{12}$ орієнтації (111) вирощені ферогранатові структури, які мають від одного до трьох монокристалічних шарів з плівок залізо-ітрієвого гранату $Y_3Fe_5O_{12}$ (ЗІГ), двошарові структури ЗІГ- La,Ga:ЗІГ і $\{Y,Sm,Lu\}_3(Fe,Ga)_5O_{12}$ – ЗІГ. Одержані структури досліджені методом феромагнітного резонансу (ФМР) і інтерференції магнітостатичних хвиль (МСХ). Дво- і тришарові структури ЗІГ характеризуються розширеною лінією ФМР (ΔH). Для структур, що складаються з трьох плівок ЗІГ загальною товщиною 68 -102 мкм ширина лінії ФМР ΔH = 5.7 – 11.5 Е. Для структури $(Y,Sm,La)_3(Fe,Ga)_5O_{12}$ - ЗІГ товщиною від 3 до 65 мкм ΔH = 0.34 – 1.22 Е. Окремі шари в феритових структурах характеризуються різними за величиною намагніченостями насичення ($4\pi M_s$). Установлено частотне розмежування інтерференційних картин для МСХ в двошаровій структурі ЗІГ - La,Ga:ЗІГ. Втрати при поширенні МСХ зростають при переході від одно- до двошарових структур і при зменшенні довжини хвилі МСХ.

Ключові слова: шаруваті феромагнітні структури, феромагнітний резонанс, магнітостатичні хвилі.