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Crystalline, magnetic and domain structure of epitaxial ferrite-garnet films (review)

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The article gives a literary review of the current state of research of ferrite-garnet films. The basic crystalline and magnetic characteristics of thin films of iron-yttrium garnet are considered. We analyzed the causes of the appearance of the labyrinth, stripe and cylindrical domain structure. It is shown that the domain structure of the yttrium iron garnet strongly depends on many parameters of the films, in particular on the thickness, structural perfection of the surface and the sample as a whole. The article reviews the most common methods of synthesis, ion implantation and post-growth treatment of ferrite-garnet films.

The study of dependencies between the conditions of obtaining, the chemical composition, the post-treatment conditions, the defective structure and the magnetic properties of ferrite-garnet films have great practical value for obtaining films with predetermined properties.

Key words: Domain structure, Yttrium-iron garnet, LaGa-substitution, magnetic properties, lattice parameter.

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Introduction

Magnetic garnets are widely used in devices of modern functional electronics. The scope of their use as an active medium traditionally includes devices on spin waves in the region of ultrahigh frequencies, elements of solid-state lasers, magneto-optical devices, magnetic recording devices, devices for weak magnetic fields [1, 2]. Great attention is given to epitaxial garnet films used in magnon spintronics [3] and magnon-optic hybrid systems [4]. The establishing of interconnections between crystal, magnetic and domain microstructure of magnetic garnet allows obtain materials with controlled physical properties [5].

I. The crystal structure

Ferrite-garnets are complex oxide compounds with the general formula $\{c_3\}[a_2](d_3)O_{12}$, space group $O_h^{10}\,(Ia3d)$. There are body-centered cubic structure formed by oxygen anions and three types of cavities– 24 dodecahedral, 16 octahedral and 24 tetrahedral (marked as {}, [] and (), respectively). Cavities are filled with cations which form dodecahedral (c), octahedral (a) and tetrahedral (d) sublattices. The first complex investigations of crystalline and magnetic properties of ferrite-garnet were carried out on the structure of yttrium iron garnet (YIG) $\{Y_3\}[Fe_2](Fe_3)O_{12}$ [6]. The stability of the garnet structure is a result of all cation position filling. At the same time the garnet unit cell has comparatively large volume so it's quite mobile. The presence of three types of cation positions provides great opportunities for isomorphic substitution and new materials formation with predicted structural and magnetic properties. Y^{3+} and Fe^{3+} ions can be replaced (partially or completely) by magnetic (Mn^{3+}, Cr^{3+}, Co^{2+}, Ni^{2+}) or non-magnetic (Al^{3+}, Ga^{3+}, Ge^{4+}, Si^{4+}, V^{3+}, Ca^{2+}, La^{2+}) ions. The garnet structure is characterized by free ordering of the coordination tetrahedral and this factor determines restrictions on the replacement cations sizes. The value of the ionic radius is a limiting factor of cations substitution which determines of its probability. The ion radii in the dodecahedral positions should be within 90-120 pm regardless of the electronic configuration. Spherical symmetry of the electron ground state of cations is important for tetra- and octa- positions filling.

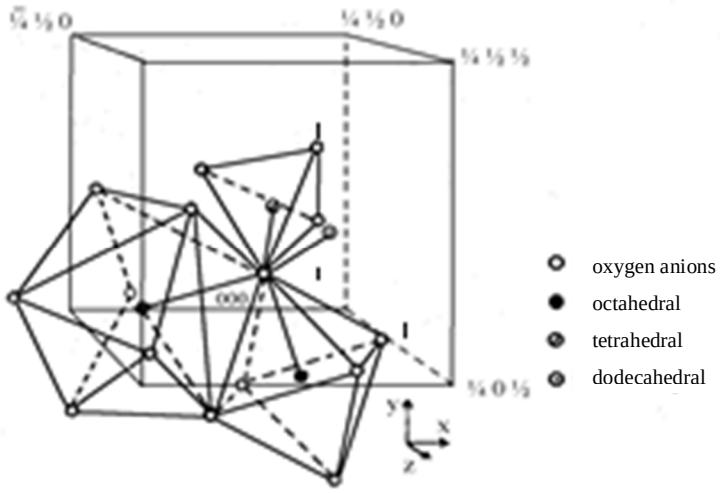

Fig. 1. YIG crystal lattice.

Lattice constant of multi-component garnets can be calculated using Vegard's law. The crystalline lattice of YIG single crystal is about a = 1.2374 nm and fluctuate within $\pm 2 \cdot 10^{-4}$ nm, depending on the growth method. [7].

II. Magnetic microstructure

Magnetic properties of oxides with a garnet structure can be explained by the Neel model antiferromagnetism [8]. For $\{Y_3\}[Fe_2](Fe_3)O_{12}$ ions in three different positions form three magnetic sublattice. The indirect exchange interaction between these sublattice are realized through oxygen anions. The resulting magnetization of ferrite-garnets films is a superposition of three sublattices magnetization were two iron sublattice are antiparallel. The value of the exchange interaction is determined by the bonding angle. The maximum interaction will be observed between the iron ions that in the octahedral and tetrahedral positions $Fe^{3+}(a) - O^{2-} - Fe^{3+}(d)$ where bonding angle is about 126.6°. For $Fe^{3+}(a) - O^{2-} - R(c)$ and $Fe^{3+}(d) - O^{2-} - R(c)$ positions the bonding angles are 122.2° and 120°, respectively . The exchange interaction between the same ions is small and is realized through two intermediate oxygen anions ($Fe^{3+}(d) - O^{2-} - O^{2-} - Fe(d)$) or additional non-magnetic cation ($Fe^{3+}(d) - O^{2-} - Ge^{4+} - O^{2-} - Fe(d)$). The magnitude and orientation of the resulting magnetic moment depends on the type of rare-earth ions in dodecahedral positions. The total magnetic moment of YIG at 0 K is $5\mu_b$ [9].

Rare-earth ions make a small contribution to exchange interaction between *a*- and *d*- sublattice therefore the Curie temperature for garnets doesn't depend of the type of rare earth ions and equal (565±20) K. [9]. The temperature of the magnetization disappearance for YIG is 559 K.

The temperature dependence of spontaneous magnetization is in good agreement with the Neel model. The *N*-type of $4\pi M_s$ temperature dependence with compensation point presence is typical for garnet compounds. The replacement of Fe^{3+} ions with nonmagnetic ions causes the saturation magnetization decreasing for *d* sublattice and increasing for *a* sublattice. The replacement of Fe^{3+} ions with Ge^{4+} is using for saturation magnetization reduction. The non-magnetic ions substitution into *a*- and *d*- sublattice violates inter-sublattice interaction and leads to Curie temperature decreasing.

III. Magnetic anisotropy

The orientation of magnetization vector is determined by uniaxial, cubic and rhombohedral anisotropy. There is also a growth-induced anisotropy as a result of two non-equivalent dodecahedral position selective filling [10]. Magnetostrictive anisotropy is a result of reverse magnetostrictive effect at the he presence of mechanical stresses [11]. The difference between film and substrate lattice constant is the typical reason of the stresses formation. The magneto-elastic anisotropy caused by ion implantation is used for growth anisotropy compensation and demagnetization energy reducing [12]. The maximal values of the uniaxial anisotropy do not exceed 10^4 J·m^{-3}. [13].

There are two types of magnetic anisotropy in epitaxial ferrite-garnets films with orientation (111) depending on the K_u uniaxial anisotropy and M_s saturation magnetization [14]. For $K_u/2\pi M_s^2 >> 1$ uniaxial anisotropy exceeds its own cubic anisotropy and the axis of easy magnetization is directed along the normal to the film (111) surface (films with cylindrical magnetic domains, $(YSmLuCa)_3(FeGeSi)_5O_{12}$). For $K_u/2\pi M_s^2 << 1$ the magnetization deviates to a small angle from the film plane due to cubic anisotropy ($Y_3Fe_5O_{12}$ and La - substituted YIG). For epitaxial ferrite-garnets films grown in (111) plane the easy axis of magnetization is perpendicular to the plane of the film but the angle between the magnetization vector and the normal to the film surface is about 75-85° [15].

IV. Domain structure of ferrite-garnet films

The elementary magnetic moments in the film are oriented along the axis of easy magnetization which leads to the demagnetization field appearance with the energy which describes as $W_m = M_0^2 d\, S_0$, where M_0 – magnetization of separate domain, d – domain thickness, S_0 – film area. The energy of domain walls is $W_d = s\, S_1 N$, $S_1 = Lh$; $N = L/d$ – number of domains. The total energy of the film is $W(d) = W_m + W_d$, or $W(d) = \frac{dM_0^2 V}{h_m} + \frac{sV}{d}$, where $V = L^2 h$. The minimum of full energy is observed when $d^2 = l_0 h$, where $l_0 = s/M_0^2$, so: $W = 2VM_0^2\sqrt{\frac{l_0}{h}}$.

There are three main types of the domain in YIG films - strip, labyrinth and cylindrical. For the experimental visual study of the domain structure the magneto-optical methods typically based on Kerr and Faraday's effects are used[16]. Strip and labyrinth domain structure will be formed if the thickness of a single crystal film with uniaxial anisotropy is less than l_0 ($h < l_0$) (Fig. 2).

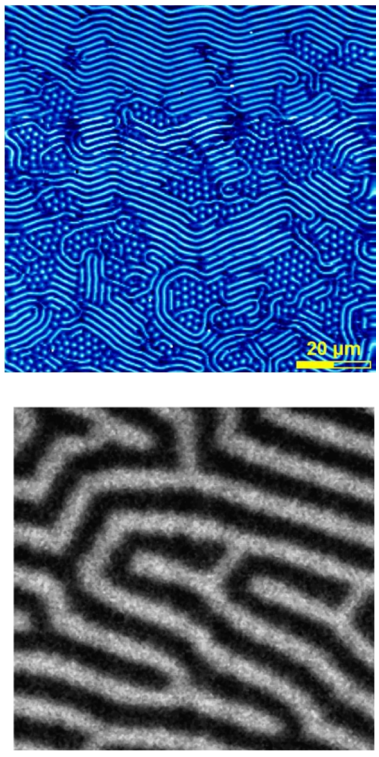

Fig. 2. Image of the labyrinth domain structure of the YIG films.

The main causes of the labyrinth structures formation – unfixed direction of the domain boundaries in the film plane, the bends of the of the film surface, accidental growth factors, effects associated with the lattice thermal motion. The labyrinth structure remains energetically beneficial in a small external magnetic field applied perpendicular to the film surface. If the equivalence of domain walls of different orientation is violated the structure of domains can be ordered and strip domain structure will form when the magnetization axis is deflected from the normal to the film surface, the rhombic anisotropy is presence or in the external magnetic field is applied (Fig. 3).

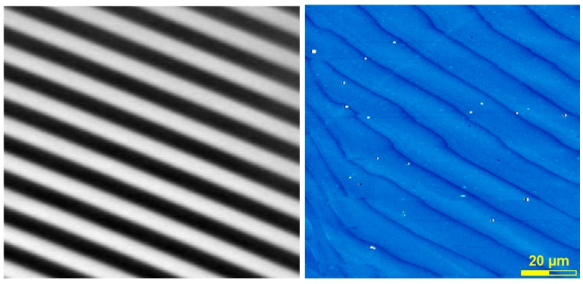

Fig. 3. Image of the strip domain structure of the YIG films.

The demagnetization field perpendicular to the film plane has a great influence factor on the strip domain structure formation.

Possibility of visual observation of the strip domain structure, can be explained by the inclination of the domain magnetization vectors to the epitaxial structure plane. The inclination of the magnetization vector may be related to the effect of the induced (growth) uniaxial anisotropy which is perpendicular to the plane of the film. The critical angle of inclination, which determines the possibility of visual monitoring of the strip domain structure, is close 19.5° [17].

For films of ZIG oriented in the plane by crystallographic indices (111) the appearance of normal components of magnetization vectors in the strip domain structure is due to the deviation of the axis of easy magnetization to the plane of the epitaxial structure (except for dislocations of a crystal lattice and induced anisotropy) [18].

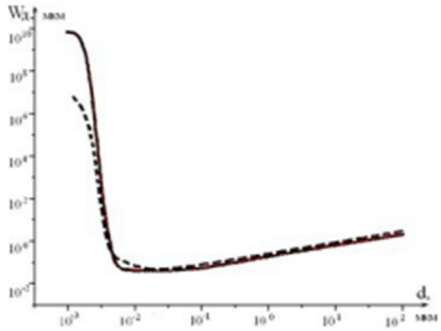

Fig. 4. Domain width W_d dependencies from the thickness of the epitaxial film: (simulation results) [18].

When applied a sufficiently large external magnetic field $\vec{B}$, number of domains with a magnetic moment $\vec{M}$ antiparallel to $\vec{B}$ decreases. Domains that are magnetized along the field $\vec{B}$, are trying to reduce the

energy of the sample and will increase in volume. Domains with the opposite direction of magnetization will be compressed in width and decrease in length until they turn into isolated cylinders. These domains are called Cylindrical Magnetic Domains (CMD) [7]. Thus, when $\vec{B}$ reaches a certain critical level, the domain splits into separate cylindrical domains, which, due to the magnetic dipole interaction, are evenly distributed over the film. A hexagonal lattice is also formed. The density of domains depends on the magnitude of the induction of the external magnetic field. The value of the magnetic field B_1, in which stable CMDs are formed, is called the field of elliptic instability of cylindrical magnetic domains.

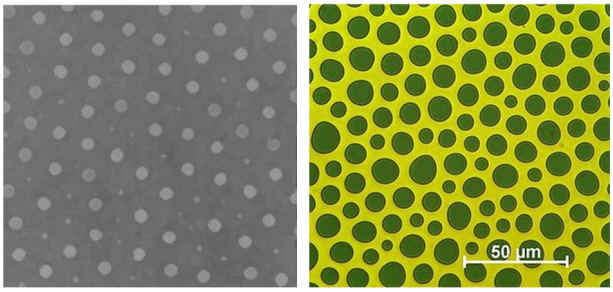

Fig. 5. The image of the cylindrical domain structure of the YIG.

In the interval of the fields $B_1 < B < B_2$, the energy of the CMD W lattice is lower than the energy of the labyrinth domain structure and the homogeneous state.
In other words, in this interval there are stable CMD (Fig. 5). When $B = B_2$, the energies of the CMD and the homogeneous state become equal, however, in the film there may be metastable CMD [19]. In this case, the dependence curve $W(d)$ has a local minimum at the value of the diameter of the CMD d_0. When value B increases, the value of d_0 decreases. After reaching the critical value d_0 (d_{cr}), the CMD collapses (magnetic field value $B = B_{kol}$). When $B = B_{kol}$, the CMD in the film is absent [20.]

The CMD can be used to create non-volatile magnetic memory elements where there are no moving parts. [11]. The problem with the practical application of CMD are the so-called "hard" domains [21]. A characteristic feature of these elements is the large number of vertical Bloch lines, which do not collapse at $B > B_{kol}$ [22]. To prevent the occurrence of "hard" CMD special technological measures are applied. These measures are aimed at creating a certain type of structure of the domain walls. Among them, ion implantation [23], growing of multilayered garnets structures, surface coating of ferrite garnets with a thin layer of soft magnetic material. Due to the bombardment of the film with high energy ions a locking magnetic layer is formed on its surface. Thickness of this layer is <1 μm. The magnetization of this layer due to the above mechanical stress is directed perpendicular to the magnetization of the CMD and is in the plane of the film. The effect of ion implantation on the dynamic characteristics of the CMD may be different. It depends on the composition of the ferrite-garnet film, and each specific case requires a separate study. A simple, but imperfect way of removing "hard" CMD is the annealing of ferrite-garnet films in an inert medium at 1100 °C.

V. Obtaining ferrite-garnet films

Qualitative characteristics of epitaxial ferrite-garnet films (which provide the possibility of their hunting as an active medium of spintronics), depends first of all on the magnetic properties (coercivity, magnetization saturation, magnetic permeability) of the surface layer. They depend on the crystalline microstructure of the films (homogeneity of the chemical and phase composition, defect, morphological structure). Therefore, the actual task is to obtain films with a predetermined crystalline (and, accordingly, magnetic) structure of the near-surface layer. Understanding the relationship between the structural and magnetic parameters of the films will be allowed to optimize methods of synthesis and further processing of epitaxial films iron yttrium garnet.

Today, the liquid-phase epitaxial method is technologically the most exhausted and widely used method of obtaining films of ferrite garnets. This method includes the following main stages: 1) moving the component of the solution to the edge of the diffusion layer adjacent to the front of crystallization; 2) independent diffusion of the component through the diffusion layer; 3) ordering the component of growth near the crystallization front; 4) desolvation of solvent ions and diffusion of the component; 5) diffusion of solvent ions through a diffusion layer in the melt solution.[11] The initial phase of the growth of the film is accompanied by the formation of a diffusion layer on the boundary of substrate - the solution-melt. In the main phase, the dependence of the thickness of the epitaxial film from the time of growth is linear [24]. The course of epitaxial growth from solution-melt is determined by the diffusion of garnet-forming components through the near-surface layer near the front crystallization. Another fact is the kinetics of the reactions of the inclusion of these components into the structure of the film at the front crystallization and the transfer of the component of the solution to the boundary of the diffusion layer. The process of growth is determined by such technological parameters as the degree of overcooling of the melt solution and the speed of rotation of the substrate [25]. The processing of films includes mechanical grinding; mechanical, chemical-mechanical and chemical polishing. In order for epitaxial growth to occur, it is necessary that the difference between the parameters of the film a_f and substrate a_s grating was minimal. When the difference between the film and substrate ($\Delta a = a_s - a_f$) lattice parameters is greater than 0.19 Å at 950°C, the epitaxial growth doesn't occur [24]. With smaller Δa in the epitaxial film, depending on its thickness, the microdefects are formed (different types depending on the sign Δa) [26]. Excessive stretching of the film ($a_f < a_s$) leads to the formation of cracks, excessive compression ($a_f > a_s$) leads to the formation of tubercles and it contributes to the destruction of the surface. It has been shown in [24] that for the growth of a defect-free

films, the allowance values of Δa at room temperature should be in the range from -0.012 ÷ -0.015 Å to +0.018 Å. The discrepancy between the parameters of the film and substrate lattice leads to the occurrence of dislocations of discrepancies and mechanical stresses that negatively affects to the characteristics of the ferrite-garnet film [27]. The choice of crystallographic orientation of the substrate surface is quite important. The substrate plane (111) is the most rough at the atomic level, therefore the rate of epitaxial growth is the greatest (compared with planes (110), (100) and (112)) [28, 29]. In general, the kinetics of epitaxial growth from a melt-solution is determined primarily by the diffusion of garnet-forming components through the near-surface layer near the front crystallization, the chemical reaction of joining particles from the melt solution to the film at the front crystallization, and the transfer of the component of the solution to the boundary of the diffusion layer [30, 31]. When using the method of liquid phase epitaxy, transitional surface layers are formed: film- substrate and film-air. The chemical composition of these layers differs from the main volume, which change the magnetic parameters of the film. The transition layers have a particularly significant effect on the parameters of submicron FGF.Methods of post-processing of ferrite-garnets

Ion implantation.

Application of ion irradiation to epitaxial films of YIG, allows predictably change the crystalline and, accordingly, magnetic structure of near-surface layers of FGF. This can cause magnetostrictive changes in the energy density of the magnetohydrogen anisotropy.

It also leads to a change in the effective magnetic thickness of the film, the change in the dispersion and the rate of propagation of the magnetostatic waves, and the change in the magnetization of saturation. In the manufacture of CMD materials, ion implantation is used to eliminate "hard" domains (by creating a parallel to the surface of the domain boundary) and the formation of ways to promote them [7].The mode of ion implantation is determined by such characteristics as type, dose and energy of implanted ions. Also important are the temperature of the target and the angle between the direction of flux of flowing ions and normal to the surface of the film [32].

Properties of the area affected by irradiation are determined by the nature of the distribution of radiation defects along the depth of the film. The broken layer can be conventionally divided into three sublayers: 1) the emission sublayer (thickness up to ≈ 1 nm) is formed when spraying particles from the surface of the film. In this sublayer ion implantation stimulates electronic emission, formation of oxygen vacancies, causes electroneutrality violation and a number of superficial effects (activation of chemical reactions, dissociation processes); 2) broken sublayer (thickness is determined by implantation conditions, usually <1 μm). It undergoes processes of primary and secondary radiation defect formation, the establishment of distribution profiles of rooted ions and displaced ion matrices, selective destruction of cation substrates, self-propagation by ion beam;3)excited sublayer (the thickness exceeds the ionic mileage). Properties of this sublayer are determined by processes occurring in the excited and, possibly, emission layers. Here there are fields of elastic deformation and processes of radiation-stimulated diffusion. The result of the ion implantation is a local increase in the interplane distance d in the direction normal to the surface of the film and the appearance of plane compression stresses: $s_s = \frac{E}{1-n}\frac{\Delta d}{d}$, where E – Young's module, ν – Poisson's ratio (for YIG $E=2\cdot10^5$ Mpa, ν=0,29 [7]). At doses of implantation (which can be neglected by the interaction of individual radiation defects and the formation of crystalline disordered regions along the ion track), we can assume that the stresses are proportional to the density of radiation defects. The compression stresses in the plane of the film cause a magnetostrictive change in the density of the energy of the magnetohydrogen anisotropy by the magnitude $\frac{3}{2}l_{111}s_s$, where λ_{111}- magnetostriction constant.

Annealing.

One of the ways for controlled change in the properties of an ion-implanted layer is annealing. This process is carried out to increase its thermal stability and correction of physical parameters (degree of defect, parameters of magnetic anisotropy). In the process of annealing (due to diffusion processes) there is a partial or complete elimination of radiation defects formed as a result of ion irradiation. Anneal is a multi-stage process, the conditions of which are determined by the implantation regimen and the parameters of the film. Annealing, depending on specific technological tasks, can take place both in the process of ion implantation (by varying the ion current density) [33], and after it - in air [34], in a vacuum or in a certain chemical atmosphere [35]. According to [36], the thermal annealing of point defects and their complexes is possible in three ways:

– by decomposing a complex defect on simple components with their subsequent annihilation or diffusion into the effluent;

– by moving the defect as a whole to the wastewater (surface, grain boundaries, dislocations, etc.).;

– by the interaction of mobile components (own atoms, impurity atoms, vacancies) with subsequent rearrangement and the formation of defects of a new type.

The process can be carried out both in the classical way - in the furnace, and with the help of local processing methods (laser radiation, electron beam, pulsed lamp).

Conclusions

Despite the extensive study of the properties of ferrite-garnet films, modern micro and nanoelectronics require new active media for magneto-optics and spintronics. And this needs the search for interconnections between the conditions of receipt, the chemical composition, the

conditions of post-processing and the heterogeneity of structural and magnetic properties in thickness.The current scientific task is to find the physical regularities of the formation of the crystalline, magnetic and domain structures of the FGF. These solutions will make it possible to develop methods for obtaining functional materials with the set of properties adapted to the specific technological narrow field.

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А.О. Коцюбинський

Взаємозв'язок кристалічної, магнітної та доменної структури епітаксійних ферит-ґранатових плівок (огляд)

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Здійснений літературний огляд сучасного стану досліджень ферит-ґранатових плівок. Розглянуті основні кристалічні та магнітні характеристики тонких плівок залізо-ітрієвого ґранату. Проаналізовано причини появи лабіринтної, полосової та циліндричної доменної структури. Показано, що доменна структура залізо-ітрієвого ґранату, сильно залежить від багатьох параметрів плівок, зокрема від товщини, структурної досконалості поверхні та зразка в цілому. Проведено оцінку найбільш поширених методів синтезу, іонної імплантації та пост-ростової обробки ферит-ґранатових плівок.

Дослідження залежностей між умовами отримання, хімічним складом, умовами постобробки, дефектною структурою та магнітними властивостях ферит-ґранатових плівок мають вагоме практичне значення для отримання плівок з наперед заданими властивостями.

Ключові слова: Доменна структура, магнітна мікроструктура, епітаксійна плівка, залізо-ітрієвий ґранат.