

Magnetic Microstructure Aluminum-substituted Barium Hexaferrite for Microwave Devices mm-Wavelength Range

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The magnetic structure and cationic distribution was investigated in ferrite with a hexagonal structure $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{12}$ ($x \leq 2$). It is found that when $x = 2$ Al^{3+} ions are localized mainly in the positions of 12k (*a*-sublattice) and 4f₁ (*c*-sublattice). This contributes to change of number magnetically active connections and therefore the magnetic structure of ferrite and magnetic parameters. It was concluded about the possibility of using the results obtained with the choice of optimal composition of substrates for millimeter-wave devices.

Keywords: Hexaferrite, Magnetic microstructure, Cationic distribution, Microwave, Crystal structure.

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1. INTRODUCTION

Currently the problem of nano architectonic comes to the forefront at obtaining new crystalline materials for microwave magnetoelectronic. They include the production of thin layers of magnetic materials functioning in weak magnetic fields, or lack thereof. Such layers can be obtained by ordering of nanoscale structural elements as a result of coordinated interaction between the ferromagnetic nanostructures and solid surface (substrate), has a strong internal field of the magnetic anisotropy. Compliance with this requirement is only possible if a certain composition, structure and physical properties of the surface of the magnetic substrate.

A very promising material for such substrates are ferrites with a hexagonal structure [1]. They have a valuable combination of magnetic and dielectric properties capable of providing the required level of parameters required for the creation of microwave devices [2]. First of all, for their peculiar presence of high values of the magnetic anisotropy, adjustable level of saturation magnetization and dielectric loss. The crystal structure executes the important role in the formation of these properties. The crystal structure includes hexagonal layers, in which a portion of the oxygen ions is substituted with ions Ba^{2+} , Sr^{2+} , Ca^{2+} , etc. [3]. These layers provide a preferential orientation of the atoms of iron ions, and iron ions located in five-fold oxygen coordination, build a strong internal magnetic anisotropy field [4].

However, investigations of the hexagonal ferrites with different types of characterizing the cations (Ba, Sr, Ca) indicate the weak influence of cations on the energy of the magnetic anisotropy and the magnetization of the hexagonal ferrites. A more significant effect on these parameters have diamagnetic atoms replacing Fe ions and change the number and strength of the exchange coupling between the five existing magnetic sublattices. In addition, they are able to change the local distortions of the crystal lattice and consequently, reduce internal stresses and affect the magnetic structure of matter.

These ions are In^{3+} , Sc^{3+} , Al^{3+} and others [1, 5].

The paper studied the effect of Al^{3+} ions on the magnetic structure and magnetic properties of hexagonal ferrite $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}$.

The chemical composition of ferrites was set based on the presence of magnetic anomalies in the concentration dependences of the magnetic parameters and data of Mossbauer studies $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{12}$ [3, 5].

2. EXPERIMENTAL PART

The objects for the study were anisotropic and isotropic polycrystalline ferrites $\text{BaFe}_{12}\text{O}_{19}$ and $\text{BaFe}_{10}\text{Al}_2\text{O}_{19}$. The ferrites were manufactured by the known ceramic technology and by X-ray analysis, was not contain external phases. The magnetic parameters: specific saturation magnetization σ_s , specific residual magnetization σ_r , coercive force H_c , Curie temperature, anisotropy field H_a measured by standard techniques on a vibrating magnetometer VSM250 in the field of 20 kOe. Mössbauer investigations were conducted on the spectrometer Ms 1104 Em with automatic processing the spectra of the program Univem Ms in the range 87-300 K.

3. RESULTS AND DISCUSSION

Fig. 1 shows as an example Mössbauer spectra of isotropic and anisotropic ferrites samples $\text{BaFe}_{12}\text{O}_{19}$ and $\text{BaFe}_{10}\text{Al}_2\text{O}_{19}$.

From Fig. 1a and b shows that the spectra of samples $\text{BaFe}_{12}\text{O}_{19}$ represent a superposition of five sextets corresponding sublattices *a* (12k), *b* (2a), *c* (4f₁), *d* (4f₂), *e* (2b). Population sublattices correspond to their theoretical values for the hexagonal structure of M. The parameters of Mössbauer spectra of iron ions localized in these sublattices coincide with the data obtained in [6].

In contrast to the unsubstituted hexaferrites $\text{BaFe}_{12}\text{O}_{19}$ Mössbauer spectra of samples of substituted ferrites $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{12}$ represent a superposition of 7 sextets, corresponding sublattices *a*₁, *a*₂, *b*, *c*₁, *c*₂, *d*

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and e . The emergence of non-equivalent nodes $12k$ и $4f_1$ associated with the peculiarities of the localization Al^{3+} . According to the [3] it is assumed that the parameters of the substitution of Fe^{3+} ions Al^{3+} ions ($x < 5$) Al^{3+} ions localizes in positions $12k$ (a -sublattice), $2a$ (b -sublattice) or $4f_1$ (c -sublattice). It can be assumed that the Al^{3+} ions are localized mainly in the positions $12k$ and $4f_1$ comparing area sextets (Table. 1).

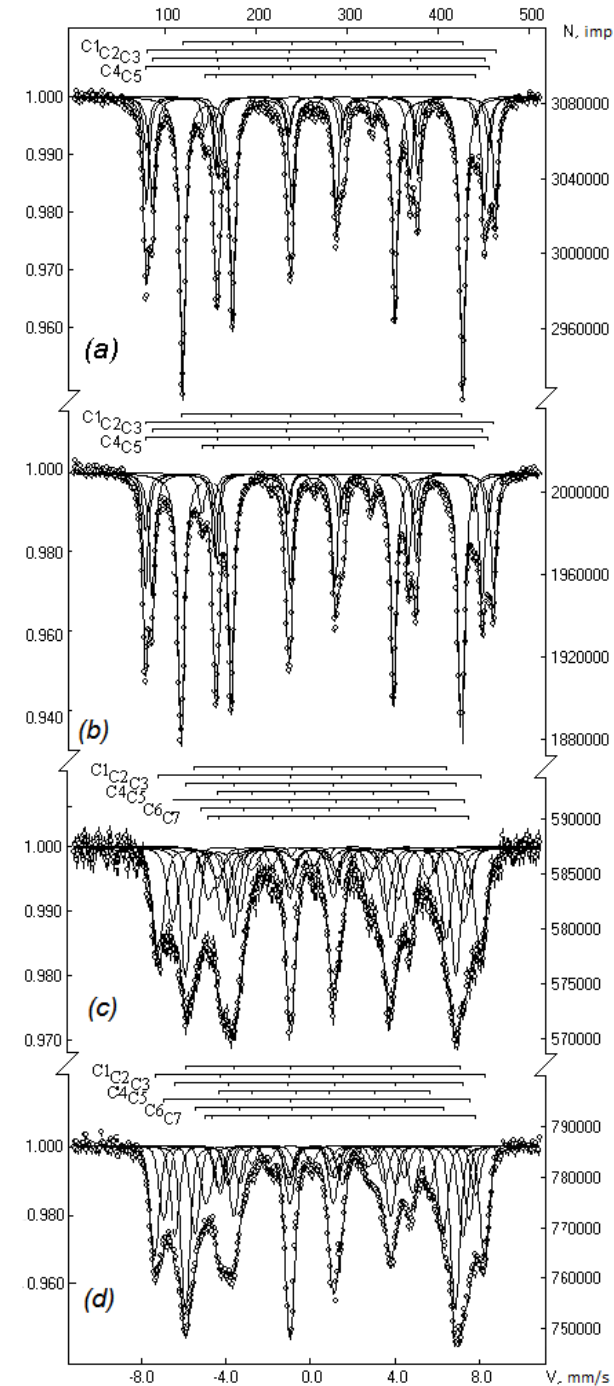


Fig. 1 – Mössbauer spectra of Ba-ferrites: a – isotropic $BaFe_{12}O_{19}$; b – anisotropic $BaFe_{12}O_{19}$; c – isotropic $BaFe_{10}Al_2O_{19}$; d – anisotropic $BaFe_{10}Al_2O_{19}$

This assumption is confirmed by the decrease in the local magnetic fields (H_L) to sublattices d ($4f_2$) and c ($2B_1$). The reduction values H_L in these sublattices indi-

cates a weakening of the main exchange relations, due to the substitution of Fe^{3+} ions ions Al^{3+} . From an examination of circuit of main exchange interactions follows that the decrease H_{eff} for sublattices d and e relatively unsubstituted ferrite $BaFe_{12}O_{19}$ associated with preferential localization of Al^{3+} ions in $12k$ and $4f_1$ -positions. Indeed, the sublattice c has the maximum number of (9) relations with a -under the grate. In turn, sublattice a has exchange connection with c -sublattice and 2 connection with d -sublattice.

Loss of magnetically bonds Fe_c-O-Fe_a and Fe_a-O-Fe_d leads to a decrease local magnetic fields at the nucleus Fe^{57} , the appearance of non-equivalent nodes $12k$ and $4f_1$ and as a consequence to a reduction sublattice magnetization and other parameters of the ferrite (Table 2).

From the Table 2 it is seen that the weakening of the exchange coupling leads to a decreases the Curie temperature of $177\text{ }^\circ\text{C}$, the specific saturation magnetization of $43\text{ Am}^2/\text{kg}$ compared with $BaFe_{12}O_{19}$ ferrite. However, based on the cationic distribution calculated reduction of the specific saturation magnetization of ferrite is substantially lower than observed experimentally. This indicates the presence of non-collinear magnetic structure in the ferrite $BaFe_{10}Al_2O_{19}$. Based on the ratio of integral intensities of the components of the Mössbauer spectra of anisotropic samples, it was found that the angle of deviation of the magnetic moments of the sublattices of ferrite is 44.5 degrees. The deflection angle of the magnetic moments retains its value, and the temperature is lowered to 87 K . Non-collinear magnetic structure is accompanied by a change in the shape of the magnetic hysteresis loop. As an example, Fig. 2 shows the magnetic hysteresis loop ferrite $BaFe_{12}O_{19}$ and $BaFe_{10}Al_2O_{19}$.

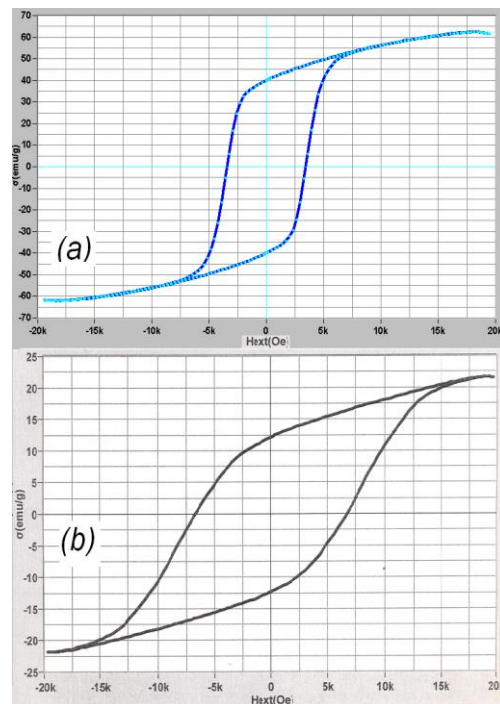


Fig. 2 – Hysteresis loops anisotropic hexagonal ferrites: a – $BaFe_{12}O_{19}$; b – $BaFe_{10}Al_2O_{19}$

Table 1 – Mössbauer parameters of the study of isotropic and anisotropic hexagonal ferrites BaFe₁₂O₁₉ and BaFe₁₀Al₂O₁₉

Ferrite	Sublattice, component spectrum	The isomer shift δ , mm/sec	Quadrupole splitting Δ , mm/sec	Magnetic field at the nucleus Fe ⁵⁷ H, kOe	Areas component S, %
Isotropic BaFe ₁₂ O ₁₉ powder	<i>a</i> -C1(Fe ³⁺)VI	0,35	0,42	414	49,9
	<i>b</i> -C4(Fe ³⁺)VI	0,33	0,00	507	8,8
	<i>c</i> -C3(Fe ³⁺)IV	0,26	0,22	491	18,6
	<i>d</i> -C2(Fe ³⁺)VI	0,39	0,20	516	17,6
	<i>e</i> -C5(Fe ³⁺)V	0,29	2,21	400	5,1
Anisotropic BaFe ₁₂ O ₁₉ powder	<i>a</i> -C1(Fe ³⁺)VI	0,35	0,41	414	47,7
	<i>b</i> -C4(Fe ³⁺)VI	0,32	0,02	506	7,8
	<i>c</i> -C3(Fe ³⁺)IV	0,26	0,21	488	19,4
	<i>d</i> -C2(Fe ³⁺)VI	0,39	0,18	514	19,4
	<i>e</i> -C5(Fe ³⁺)V	0,28	2,17	403	5,7
Isotropic BaFe ₁₀ Al ₂ O ₁₉ powder	<i>a</i> ₁ -C1(Fe ³⁺)VI	0,31	0,38	370	18,0
	<i>d</i> -C2(Fe ³⁺)VI	0,37	0,13	474	22,9
	<i>c</i> ₁ -C3(Fe ³⁺)IV	0,31	0,43	397	23,2
	<i>b</i> -C4(Fe ³⁺)VI	0,37	0,52	311	8,2
	<i>a</i> ₂ -C5(Fe ³⁺)IV	0,28	0,24	428	16,6
	<i>c</i> ₂ -C6(Fe ³⁺)IV	0,22	0,37	345	2,6
	<i>e</i> -C7(Fe ³⁺)V	0,29	2,1	383	8,4
Anisotropic BaFe ₁₀ Al ₂ O ₁₉ foil	<i>a</i> ₁ -C1(Fe ³⁺)VI	0,29	0,39	396	25,8
	<i>d</i> -C2(Fe ³⁺)VI	0,37	0,13	485	17,4
	<i>c</i> ₁ -C3(Fe ³⁺)IV	0,25	0,32	423	14,8
	<i>b</i> -C4(Fe ³⁺)VI	0,37	0,52	312	6,8
	<i>a</i> ₂ -C5(Fe ³⁺)IV	0,27	0,02	449	12,3
	<i>c</i> ₂ -C6(Fe ³⁺)IV	0,26	0,32	366	13,8
	<i>e</i> -C7(Fe ³⁺)V	0,29	2,21	400	8,9

Table 2 – Magnetic properties of anisotropic hexagonal ferrites

Ferrites	The specific saturation magnetization σ_s , Am ² /kg	Specific residual magnetization σ_r , Am ² /kg	Coercive force H_c , kA/m	The Curie temperature, K
BaFe ₁₂ O ₁₉	64,61	40,13	273	720
BaFe ₁₀ Al ₂ O ₁₉	21,68	12,13	532,3	543

The area of the hysteresis loop in the ferrite BaFe₁₂O₁₉ reduced by 8 times, and M_r / M_s 1.5 times (from $-0,641$ to $0,560$). Considering that the shape of the hysteresis loop depends on how the occur reversal processes, can be assumed that the substitution of Fe³⁺ ions Al³⁺ ions changing mechanical stresses or internal parameters of the spin-lattice relaxation. This is indicated by the changes the isomer shift and quadrupole splitting of the Mössbauer spectra for the sublattices *a* and *c*, which showed that the electronic structure transforms of Fe³⁺ ions as a result of changes in their local environment (Table 1). Indeed each cation located in the octahedral position 12k surrounded by six nearest neighbors (cations) from the trigonal axis of symmetry. If Al³⁺ ion occupies the tetrahedral position, the presence of the energy of the spin-lattice interaction is necessary to minimize its orderly arrangement of ions Al³⁺. In this case, Fe³⁺ ions are displaced position, which reinforce the relationship between the spin Fe³⁺ ions and the corresponding axis of the crystal field. It is the reason additional local the crystal lattice deformations and increase the anisotropy field in the ferrite BaFe₁₀Al₂O₁₂ experimentally observed. Thus, the joint

variation of local characteristics of the hexagonal ferrite with the *M* structure resulting from substitution of Fe³⁺ ions from the ion radius close Al³⁺ ions allows obtaining a material with combination of properties necessary for device millimeter waveband.

4. CONCLUSION

The investigations of hexagonal ferrites BaFe_{12-x}Al_xO₁₂ with isomorphic substitution of iron aluminum, allowed to establish a preferential distribution of ions Al³⁺ on the border of spinel and hexagonal blocks structure. Such localization of diamagnetic Al³⁺ contributed to a decrease in magnetization of hexagonal barium ferrite, a change in its magnetic structure and magnetic anisotropy field.

The results can be used in the selection of optimum compositions hexagonal ferrites substrate millimeter wave devices.

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