Spin ordering and magnetic crystallographic anisotropy in hexagonal ferrites of the Ni_2Sc_xW system

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New angular block magnetic structures in hexagonal ferrites of the $BaNi_2Sc_xFe_{16-x}O_{27}$ system were investigated by neutron-diffraction, ferromagnetic-resonance (FMR), and magnetic measurements. At Sc contents $x < 1.3$ a microheterogeneity in distribution of the Sc is observed and is responsible for the existence in the studied samples of regions having different magnetic-order periods. At $x \geq 1.3$, the $2d$ positions are completely filled with Sc^{3+} ions and the magnetic structure is canted and homogeneous over the sample. The behavior of such compositions in FMR can be described phenomenologically by a noncollinear block model with a quasicubic type of anisotropy within the blocks and with weakened exchange interaction between them.

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The determination of the relation between the magnetic crystallographic anisotropy, the spin ordering, and the cation distribution in such complicated ferrimagnets as hexagonal ferrites is an important physical task in the study of the nature of magnetic interactions. The possibility of synthesizing new compounds of the ferrite type are determined by the extent to which we know their physical properties with different variants of cation substitutions in the crystal lattice. Particular significance is assumed by the investigation of the magnetic characteristics of new types of ferrimagnets and the study of their magnetic structure.

Recent neutron-diffraction investigations of the magnetic structure of hexagonal ferrites of the type **M**, **Y**, and **Z** with different isomorphic substitutions have revealed a new type of magnetic-order formations, called magnetic block structures.[1] The present paper is devoted to a study of new angular magnetic block structures in hexagonal ferrites of the system $BaNi_2Sc_xFe_{16-x}O_{27}$ (Ni_2Sc_xW), which were previously known as collinear ferrimagnets, with the aid of neutron-diffraction, magnetostatic, and radio-frequency (based on ferromagnetic resonance) methods.

The investigated compounds crystallize into a space group $P6_3/mmc$ with the initial composition ($x = 0$) having unit-cell parameters $a = 3.88$ Å and $c = 32.85$ Å[2].

EXPERIMENTAL PART

1. Quasimonocrystalline (textured) ferrites in which the Sc^{3+} ion content per molecule, x, ranged from 0 to 4 (in steps of 0.1 in the interval $0 < x < 1$, steps of 0.2 at $1 < x < 1.6$, and steps of 1.0 at $2 < x < 4$) were synthesized by the topotactical-reaction method with texturizing pressing, in a magnetic field, of a mixture (alcohol suspension) of oxides prepared beforehand, including a strongly anisotropic component (the ferrite Ni_2W) that becomes oriented along the field. The pressed samples were subjected to a final annealing in formation temperature intervals that were optimal for each composition (in the range 1200–1350°C). The axis of the texture was oriented along the [001] direction. The samples were prepared in the form of prisms measuring $5 \times 5 \times 30$ mm and spheres of diameter 1 and 0.5 mm.

The high degree of the texture (0.8 and higher) and the sufficiently high density of the samples (more than 4 g/cm^3 or 0.75–0.8 of the x-ray value) at a high resistivity (10^6–10^8 Ω-cm) and relatively simple manufacturing technology have made it possible to carry out the entire set of investigations and to obtain for the magnetic structure and the anisotropy practically the same information is obtainable for single crystals. A phase analysis based on microscopically ground sections and with the URS-50IM x-ray diffractometer revealed practically no extraneous inclusions. The intensity of the reflections of the extraneous phases did not exceed 3% of the reflections of the main phase, which was the ferrite **W**.

2. Measurements of the magnetic crystallographic anisotropy were carried out by the FMR method in the frequency range 30–50 GHz in magnetizing fields up to 20 kOe. The sample was placed in a flow-through resonator on a rotating holder in such a way that the axis of the texture could make an arbitrary specified angle with the direction of the external field. The required sample temperature was ensured by blowing heated liquid-nitrogen vapor. The investigations have shown that replacement of the iron in ferrites of type **W** by diamagnetic Sc^{3+} ions, just as in type-**M** ferrites.[1] influences not only the magnitude but also the character of the magnetic anisotropy.

An increase of the Sc^{3+} ion concentration decreases monotonically the anisotropy field $H_A^{\parallel}$ and the first constant $K_1 = \frac{1}{2} H_A^{\parallel} M_S$ associated with it down to zero (at $x = 2.0$ and $T = 293°K$) (Fig. 1). The saturation magnetization M_S decreases also monotonically, but less rapidly, at 293°K. The temperature dependences of the anisotropy constants and of the magnetization are similar, and this is the cause of the weak temperature dependence of $H_A^{\parallel}$ (Fig. 1b).

Up to an Sc^{3+} ion concentration $x = 1.3$ the behavior of the ferrite in FMR is well described by two magnetic-anisotropy constants and the collinear ferrimagnetic model. The second constant is negative in this interval, and at low concentration it is much smaller in magnitude than the positive first constant, becoming comparable with the latter only near $x = 2.0$, where the positive contribution to the constant K_1 from the single-ion anisotropy of Fe^{3+} in the positions 2d decreases to the

value of the negative contribution of the magnetic dipole interaction.

The negative sign of the constant K_2 indicates that the magnetic structure can be either uniaxial, at $K_1 > |K_2|$ (with a local energy minimum observed in the interval $|2K_2| > K_1 > |K_2|$ in the direction of the basal plane (001), or else planar, at $K_1 < |K_2|$ (with a local minimum in the [001] direction in the interval of values $|K_2 > K_1 > 0$), but not "conical." This pertains, of course, only to the collinear model of the magnetic structure.

Table I yields values of the anisotropy parameters for ferrites of three compositions (the magnetizations and the anisotropy constants were recalculated for samples with x-ray density). The values of the fields and of the anisotropy constants were calculated from the resonance conditions for a uniaxial ferromagnet:

$$\nu/\gamma_{\parallel}=H_{res}^{\parallel}+H_A^{\parallel}, \quad (\nu/\gamma_{\perp})^2=H_{res}^{\perp}(H_{res}^{\perp}-H_A^{\perp}), \tag{1}$$

$$H_A^{\parallel}=2K_1/M_S, \quad H_A^{\perp}=(2K_1+4K_2)/M_S; \tag{2}$$

where ν and H_{res} are the FMR frequency and the resonant field, γ is the gyromagnetic ratio, M_S is the saturation magnetization, and the subscripts $\parallel$ and $\perp$ denote the orientation of the external field relative to the texture axis.

At concentrations $x > 1.3$, especially at low temperatures (see the anisotropy curves of the resonant fields on Fig. 2), the FMR data are difficult to interpret if one is to stay within the framework of the single-sublattice collinear model, for in this case the anisotropy constants depend on the field, which is a situation typical of canted structures. Therefore the values of the anisotropy constants in Table I for $x \geq 2$ are not given, or are included in parentheses as tentative ones, with an indication that they were calculated formally from relations (1) and (2) for a fixed frequency (42.6 GHz at T = 80 and 190°K, 34.5 GHz at T = 293°K), and γ is assumed to equal 2.8 MHz/Oe.

3. The real picture of the spin ordering was made more precise by comparing the data of the neutron-diffraction investigations and the magnetization measurements. The temperature dependences of the magnetization (Fig. 3a) exhibit maxima typical of ferrites that have a tendency to form canted structures. Indeed, at temperatures below the maxima one observes in all cases a structure different from the Gorter model, with a noncollinear ordering of the spin, as is evidenced by the anomalously large static susceptibility in sufficiently strong field (up to 20 kOe, oriented along the [001] axis), which could not be attributed only the imperfection of the ferrite texture (Fig. 3b).

4. The neutron-diffraction investigations were carried on textured samples of the system of hexagonal ferrites Ni_2Sc_xW with Sc^{3+} ion contents x =0, 0.4, 0.6, 0.8, 1.0, 1.4, 2.0, and 4.0. The neutron diffraction patterns were obtained under conditions of joint angular displacement of the sample and of the counter in a ratio $\theta:2\theta$ (reflections of the $00l$ series). The wavelength of the employed neutrons was 1.15 Å. In addition to the $00l$ reflections, we analyzed the magnetic contributions in the $h0l$ reflections at 80 and 293°K. The magnetic contributions were determined by heating the samples above the corresponding Curie points.

a) Ni_2W(x = 0). The neutron-diffraction patterns were obtained from textured samples and single crystals. The series of the $00l$ and $h0l$ reflections on the neutron diffraction patterns of the investigated compounds there were present only the structure reflections allowed by the space group $P6_3/mmc$. The $00l$ reflections are due

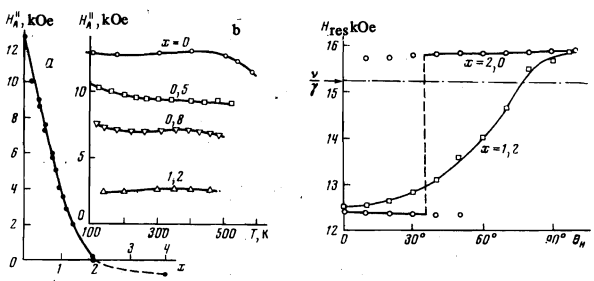

FIG. 1 FIG. 2

FIG. 1. The dependence of the anisotropy field of ferrites of the Ni_2Sc_xW system on the content of the Sc^{3+} ions (a) and on the temperature (b).

FIG. 2. The anisotropy of the resonant field for the ferrites Ni_2Sc_xW with Sc^{3+} ion contents x = 1.2 and x = 2.

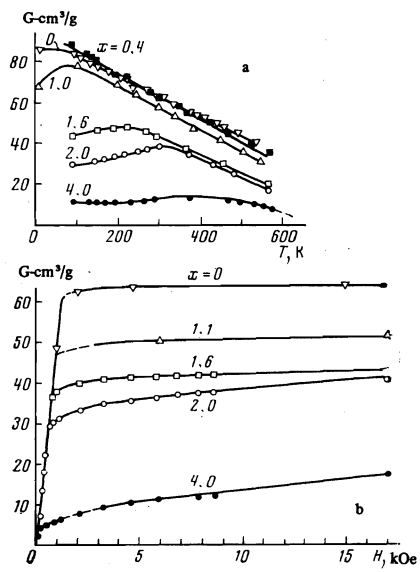

FIG. 3. Dependence of the magnetization on the temperature (a) and on the applied field (b).

TABLE I. Parameters of magnetic crystallographic anisotropy for the hexagonal ferrites Ni_2Sc_xW

Ferrite-composition parameter x	T=80 K					T=190 K					T=293 K				
	M	$H_A^{\parallel}$	$H_A^{\perp}$	K_1	K_2	M	$H_A^{\parallel}$	$H_A^{\perp}$	K_1	K_2	M	$H_A^{\parallel}$	$H_A^{\perp}$	K_1	K_2
	G	kOe		10^5 erg/cm³		G	kOe		10^5 erg/cm³		G	kOe		10^5 erg/cm³	
1.2						390	2.4	2.96	4.8	−0.4	265	2.7	1.4	3.4	−0.8
2.0	168	2.9	1.04	(2.5)	(−0.8)	—	—	—	—	—	218	<0.2	<0.2	<0.2	<0.2
4.0	51	2.9	0.9	—	—	53	1.0	1.0	—	—	63	0.7	1.4	(0.22)	(0.11)

exclusively to nuclear scattering of the neutrons. The reflections h0l contained appreciable magnetic contributions. Estimates of the magnetic contributions to the diffraction picture show that the magnetic moments of Ni_2W have a collinear ordering of the magnetic moments in accordance with the scheme shown in Fig. 4. This scheme of the orientation of the magnetic moments is confirmed by results of the magnetic measurements performed in the present study.

Starting from the values of the experimental nuclear contributions in the reflections 00l (textured samples), we investigated the possible variants of the arrangement of the Ni^{2+} ions in the lattice of the compound Ni_2W. We used in the calculation fifteen 00l reflections. The best agreement between the experimental and calculated values of the reflection intensity is obtained for the variant in which the Ni^{2+} ions are located in 4f positions (z = 0.073).

b) Ni_2Sc_xW (x = 0.4; 0.6; 0.8; 1.0). Replacement of the Fe^{3+} ions by Sc^{3+} ions in the lattices of the investigated ferrites noticeably alters the form of the neutron-diffraction patterns. Superstructure reflections of magnetic origin appear on the patterns, and the positions, shapes, and intensities of the reflections vary from composition to composition and with temperature. Figure 5 shows neutron diffraction patterns of the investigated samples at 80, 293, and 760°K. The form of the diffraction patterns from a number of samples of the same composition turned out to be the same. However, an analysis of the neutron-diffraction patterns gives grounds for assuming the presence of microheterogeneity in the distribution of the substituting ions, similar to that observed in the hexaferrite Al_xM.[3] Investigations with an x-ray microanalyzer revealed practically no inhomogeneity in the distribution of the scandium over the cross section of the sample.

According to[4], a canted block structure is realized in the ferrite Ni_2Sc_2W, with angular ordering of the spin axes of the blocks B. This is due to the breaking of the exchange bond in the 2d position in which the Sc^{3+} ions have been substituted. If the substitution in this position is incomplete, then regions with different orderings of the block spin axes are produced in the lattice, in accord with the microinhomogeneity of the distribution of the Sc^{3+} ions. Schemes of possible magnetic ordering in the regions are shown in Fig. 6. Depending on the distribution of the scandium ions over the 2d positions, the canted structure can be made up of the spin axes of "superblocks" that include two, three, or four "elementary" blocks B (see Fig. 6). There can also exist an ordering in which blocks B alternate along the z axis with blocks 2B, 2B alternate with 3B, etc. As seen from Fig. 6, the period of the magnetic order is in this case a multiple of the dimension of the block B, or is a multiple of half the unit chemical cell of the investigated ferrite.

The weight of the magnetic contributions in the reflections from the regions with the smaller periods increases with the content of the Sc^{3+} ions. The superstructure reflections of the higher periods (2c, 3c, 4c) are noted also on the neutron diffraction patterns of the composition with x = 1. This means that the Sc^{3+} ions are located in other positions of the lattice in parallel with the occupation of the positions 2d.

c) Ni_2Sc_xW (x = 1.4, 2.0, 4.0). The diffraction patterns of the 00l reflections of compositions with Sc^{3+} ion contents x = 1.4 and 2.0 per formula unit contain magnetic contributions that are distributed over the structure reflections 00l with even and odd l for the space group $P6_3/mmc$ and the period c of the unit cell of the ferrite W. In these samples, all the 2d positions are filled with Sc^{3+} ions.

The magnetic structure of these compounds corresponds to a noncollinear block ordering made up of the spin axes of the blocks B. It can be described with the aid of the polar angles θ_1 and θ_2 between the z axis of the cell and the spin of blocks B_1 and B_2 (see Fig. 4), and also the azimuthal angle φ between the projections of the spin axes of the blocks B_1 and B_2 on the basal

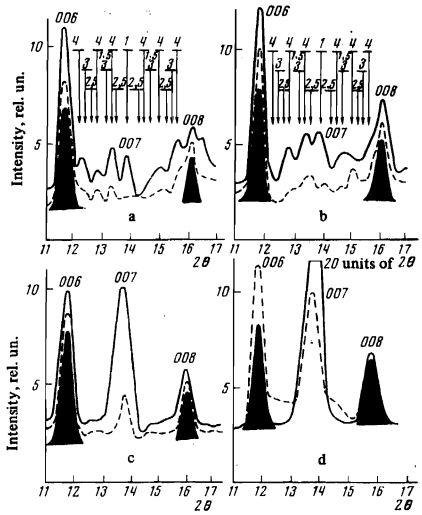

FIG. 5. Neutron diffraction patterns of the investigated compositions at 80, 293, and 760°K: a) $Ni_2Sc_{0,4}W$, b) $Ni_2Sc_{0,6}W$, c) $Ni_2Sc_{1,0}W$, d) $Ni_2Sc_{2,0}W$. Solid lines—diffraction patterns at 80°K, dashed—at 293°K; the nuclear compositions are blackened.

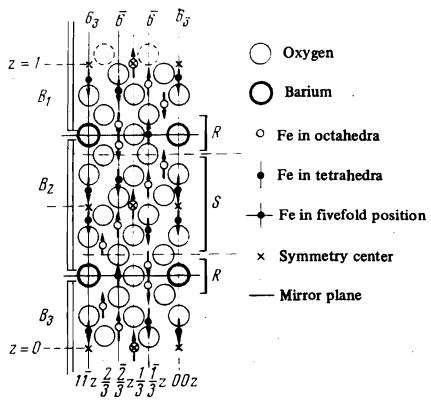

FIG. 4. Unit cell of hexagonal ferrite W. The arrows show the orientations of the magnetic moments of the ions.

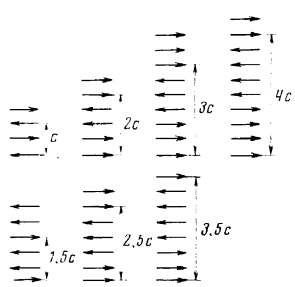

FIG. 6. Schemes of possible orderings of the spin axes of blocks B in the system of hexagonal ferrites Ni_2Sc_xW.

plane. Table II gives the values of the angles θ and φ for compositions with x = 1.4 and 2.0 at 80 and 293°K. It lists also the corresponding R factors from the reflections 00l.

Figure 7 shows the temperature dependence of the average value of the cosine of the angle θ between the z axis and the easy-magnetization axes for the investigated composition, as obtained from magnetostatic measurements. From a comparison of the data of Fig. 7 and Table II we can see a satisfactory agreement between the values of the angles θ obtained from magnetic and neutron-diffraction measurements.

An investigation of the composition with x = 4.0 has shown that a very complicated spin ordering takes place in this compound, and its interpretation calls for the use of single-crystal samples.

In calculations of the magnetic contributions to the diffraction patterns of compositions with x = 1.4 and 2.0, we used a cation-distribution variant in which the Sc^{3+} ions are placed in positions 2d (two ions per cell) and 4f (z = 0.208). The Ni^{2+} are placed in positions 4f (z = 0.073). This is the same cation-distribution variant that was used in the calculations of the diffraction-reflection intensities in[4], in which, however, a misprint occurred.

5. The assumed cation distribution was verified by the agreement between the data of the magnetic and neutron-diffraction investigations. Thus, for the variant assumed above for the placement of the Ni^{2+} ions, the calculated value of the specific magnetization σ is 85 G-cm^3/g, which coincides with the experimental value 84.7 G-cm^3/g obtained at 4.2°K. The complicated character of the disposition of the Sc^{3+} ions can be seen from the change of the magnetization at low temperatures with changing composition. It is seen from Fig. 3a that in the region of low substitution, the saturation magnetization at low temperatures is higher than that of the nonsubstituted ferrite Ni_2W. This means that the Sc^{3+} ion occupy partially the positions of Fe^{3+} ions with a spin directed against the magnetization vector (type A), namely, tetrahedral positions of the spinel (S) block 4f (z = 0.092) and 4e (z = 0.056) or octahedral positions $4f_R$ of the hexagonal barium block (R) (see Fig. 4). The tendency of the Sc^{3+} ions to occupy octahedral positions makes the second variant more probable.

Figure 8a shows plots of the specific magnetization against the concentration of the Sc^{3+} ions when Fe^{3+} is substituted only in positions of type A (curves 2) or type P (with spin along the magnetization vector, curve 3), and also the experimental values of σ extrapolated to magnetic fields that ensure establishment of a collinear Gorter structure (curve 1). These data were used to plot the relative number of Sc^{3+} ions in positions of type A against the total number of ions (Fig. 8b). We see that the number of substituted positions of this type does not exceed $x_1 = 0.32$ per formula unit of 16% of the total number of $4f_R$ positions, if the tetrahedral positions are not substituted. This maximum value is reached at x = 1. The decrease of the ion concentration in positions $4f_R$, calculated for x > 1, can be caused by the inaccuracy of the extrapolation of the magnetization.

TABLE II. Values of the angles θ and φ, and of the R factors for the compositions $Ni_2Sc_{1.4}W$ and Ni_2Sc_2W.

	$Ni_2Sc_{1.4}W$		Ni_2Sc_2W			$Ni_2Sc_{1.4}W$		Ni_2Sc_2W	
	T=80 K	293 K	80 K	293 K		T=80 K	293 K	80 K	293 K
θ_1, deg.	65	29	55	43	φ, deg.	120	60	180	90
θ_2, deg.	65	29	125	43	R, %	10.1	13.1	6.1	12.3

It follows from the obtained neutron-diffraction data that the remaining part of the Sc^{3+} ions at low concentration replaces predominantly Fe^{3+} in P-type 2d positions. It is seen from Fig. 8b that a concentration x > 1.3 the positions of type 2d are fully occupied in this case. The validity of this assumption is confirmed by the strong concentration dependence of the anisotropy field at x < 1.3 and by the decrease of its slope at x > 1.3 (Fig. 1a). This indicated a turning-off (possibly partial) of the strongest of the mechanisms that decrease the anisotropy field as a result of the decrease of the number of paramagnetic Fe^{3+} ions in the fivefold (2d) coordination that are responsible for the positive uniaxial anisotropy.[5]

When the concentration rises above x = 1.3, as shown by neutron diffraction, the Sc^{3+} ions become uniformly distributed in the block S, primarily over the central octahedral positions $4f_S$ (z = 0.208), and then 6g and 12k.

DISCUSSION OF RESULTS

The noncollinear model of the magnetic structure of the investigated compounds permits a more correct interpretation of the FMR data.

Indeed, as shown above, for crystals of the hexagonal space group $P6_3/mmc$ the negative constant K_2 does not admit of the existence, within the framework of the ferromagnetic model, of the observed deviations of the spin axes from the [001] directions. However, when the structure is broken up along the symmetry plane into autonomous sublattice blocks having a rhombohedral point group whose symmetry allows the presence of terms of the type $K_2' \sin^3\theta\cos\theta\cos 3\varphi$, in the

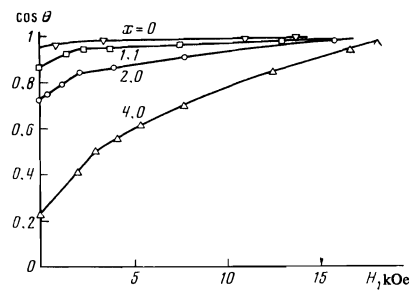

FIG. 7. Cosine of the angle between the block axes and the [001] axis as a function of the applied field.

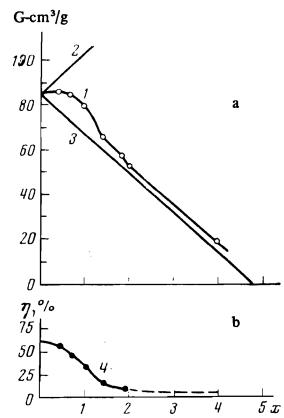

FIG. 8. Illustrating the cation distribution of the Sc^{3+} ions: 1—experimental concentration dependence of σ, 2—occupation of positions of type A with magnetic moment antiparallel to the magnetization, 3—occupation of positions of type P with magnetic moment paralllel to the magnetization, 4—relative number of ions in type-A position.

expression for the anisotropy energy, the magnetic moment can be directed along the generatrix of a cone with axis [001] if K_2' differs from zero (in analogy with the situation with the In-substituted ferrite of type M[6]). In the case of a collinear structure (i.e., for identical values of θ and φ) this term, which enters in the anisotropy energy with different signs from blocks connected by the symmetry plane, is cancelled out over the crystal.

In accordance with the obtained cation distribution, the cause of the separation of the magnetic structure into blocks is apparently the weakening of the indirect exchange interaction $4f_R-2d-4f_R$ when the Fe^{3+} ions in these positions are replaced by Sc^{3+} ions, and the fact that this interaction is weaker, at high Sc^{3+}-ion concentration, than the weak interactions of the $4f_R-4f_R$ antiferromagnetic exchange[7] and the magnetic dipole interaction. An important factor that stabilizes the structure in this case is the magnetic crystallographic anisotropy, including anisotropic terms of higher orders.

To analyze the competing interactions that are allowed by the crystal symmetry in a hexagonal type-W ferrite, we consider these interactions phenomenologically. The expression for the free interaction of the magnetic block structure can be written in the form

$$\begin{aligned} F=&M_s^2[(\lambda+a)\sin\theta_1\sin\theta_2\cos(\varphi_1-\varphi_2)+(\lambda-2a)\cos\theta_1\cos\theta_2] \\ &+d_\perp(\sin\theta_1\cos\theta_2+\cos\theta_1\sin\theta_2)+d_\parallel\sin\theta_1\sin\theta_2\sin(\varphi_1-\varphi_2) \\ &+K_1(\sin^2\theta_1+\sin^2\theta_2)+K_2(\sin^4\theta_1+\sin^4\theta_2) \\ &+K_2'(\sin^3\theta_1\cos\theta_1\cos3\varphi_1-\sin^3\theta_2\cos\theta_2\cos3\varphi) \\ &-HM_S\{\cos\theta_H(\cos\theta_1+\cos\theta_2)+\sin\theta_H[\sin\theta_1\cos(\varphi_H-\varphi_1) \\ &+\sin\theta_2\cos(\varphi_H-\varphi_2)]\}, \end{aligned} \tag{3}$$

where the subscripts of the polar and azimuthal angles θ and φ pertain to the magnetization vectors of the blocks B_1 and B_2 and to the external applied field, while M_S pertains to the saturation magnetization of the blocks at the given temperature.

The first energy term is responsible for the isotropic and anisotropic symmetrical (pseudodipole Van-Vleck) exchanges, and also to the magnetic dipole interaction between the blocks, the anisotropy of the dipole and pseudodipole interactions being given by the coefficient a. The second and third terms are the symmetry-allowed antisymmetrical exchange interactions with Dzyaloshinskiĭ vectors perpendicular to the direction of the [001] axis. The quantities λ, $d_\perp$, $d_\parallel$, and a depend on the degree of competition of the antiferromagnetic $4f_{R\uparrow}-4f_{R\uparrow}$ and ferromagnetic $4f_{R\uparrow}-2d\downarrow-4f_{R\uparrow}$ interactions of the blocks S.

The value of K_1 is determined primarily by the one-pion anisotropy of the Fe^{3+} ions in the position 2d of the trigonal symmetry (positive contribution) and by the magnetic dipole interaction (negative contribution), and also by the one-ion anisotropy of the cations that have a near-cubic environment, in octahedral (positive contribution) and tetrahedral (negative contribution) positions. The latter component is of the same order as K_2 and K_2' and can be readily separated together with them as a quasicubic term,[6] if it is borne in mind that the cubic-anisotropy constants K_1, K_2, and K_2' in a spherical coordinate system have a ratio $^2/_3:-^7/_2:\sqrt{2}/3$ (K_2 and K_1 have equal signs, while K_1 is positive for Fe^{3+} ions). The last term represents the Zeeman interaction of the magnetic moments of the blocks.

It is obvious that in the case of collinear magnetic order in accordance with the Gorter scheme ($\theta_1=\theta_2$; $\varphi_1=\varphi_2$), which obtains in the unsubstituted Ni_2W_2, the constants λ, a, $d_\perp$, $d_\parallel$, and K_2'' do not determine the statics and dynamics of the spin angular momenta.

As the ferrite Ni_2W is diluted with Sc^{3+} ions, conditions are achieved for a transition of the collinear ordering from the magnetically-uniaxial to planar, owing to the decrease of the algebraic sum of the positive contribution made to K_1 by the one-ion anisotropy of Fe^{3+} in the positions 2d and the negative contribution of the dipole interaction to the value $2|K_2|$, which determines the order of magnitude of the quasicubic contribution of the ions to the trigonally distorted polyhedra of the S block. The energy of the dipole interaction between the S blocks (from the sum of the dipole interactions) together with the energy of the exchange coupling realized via the ions in the $4f_R$ positions turns out to be smaller in the case of an antiparallel arrangement of the spin axes of the blocks. Therefore, simultaneously with the flopping of the angular momenta towards the basal plane, the directions of the spin axes are rotated. As shown above, the appearance of canting in the ordering of the magnetic moments of the blocks causes the azimuthally-dependent trigonal term of the quasicubic anisotropy of the S blocks with the constant K_2' different from zero. This energy component has minima when the orientation of the spin moments along three symmetrical positions of the generators of the cone with the [001] axis, having projections in the direction of the [110] axes of the crystal. For two neighboring S blocks, these positions are rotated through $\pi/3$, and this contributes to the formation of a helical block structure or any other structure that differs from a pure antiparallel one. Simultaneously with K_2', the invariants $d_\perp$ and $d_\parallel$ of the Dzyaloshinskiĭ-Moriya antisymmetrical exchange, which formally play jointly a role analogous to that of K_2', become different from zero. Subsequently, therefore, to determine the order of magnitude of the constants in first-order approximation, the terms with $d_\perp$ and $d_\parallel$ can be left from the anisotropy energy and included artificially in K_2'.

With account of the fact that the exchange and dipole interactions are proportional to the square of the magnetization M_S, and the one-ion contribution to the constant K_1 is linear in the magnetization (judging from the weak dependence of $H_A=2K_1/M_S$ on the temperature—Fig. 2), the angle between the spin axes of the blocks should increase in the absence of an external field with decreasing temperature and with increasing concentration of the Sc^{3+} ions. It is seen from (3) that at values of the anisotropy constant comparable with λM_S^2 and at a nonzero constant $d_\parallel$, the projections of the magnetic moments of the blocks should have the same sign ($\theta_1=\theta_2$) if $|\varphi_1-\varphi_2|\lesssim\pi/2$ and be of opposite sign ($\theta_1=\pi-\theta_2$) at $|\varphi_1-\varphi_2|>\pi/2$. This agrees well with the complicated evolution of the magnetic structure revealed with changing temperature by the neutron-diffraction data.

Let us consider the case of practical importance when the field is applied along the [001] direction. An external field of sufficiently large magnitude $H\gtrsim d_i/M_S$, K_2/M_S, K_2'/M_S stimulates the formation of a structure with $\theta_1=\theta_2$, causing in certain cases a first-order transition upon magnetization. Introducing the simplification $\varphi_1=\varphi_2=\varphi$, which does not change the qualitative picture of the magnetization of the ferrite, we can easily find the equilibrium conditions in this case:

$$\sin\theta=0,$$

$$H=2\cos\theta(B\cos 2\theta-A)-\sin\theta\cos 3\varphi B'(2\cos 2\theta+1), \quad (4)$$

where

$$A=(2a-\lambda)M_s+\frac{K_1+K_2}{M_s}, \quad B=\frac{K_2}{M_s}, \quad B'=\frac{K_2'}{M_s}.$$

The second relation gives a ferrite-magnetization equation of sixth degree:

$$H=2m[2B(2m^2-1)-A]+B'(1-m^2)^{1/2}(1-4m^2), \quad (5)$$

which agrees well with the curves of Fig. 7 if it is assumed that the constants are of the same order of magnitude and that $(2a-\lambda)M_S$ exceeds it for $x > 2$ ($m = M_Z/M_S$). The root of this equation gives the value of the residual magnetization or the initial cone apex angle (at $H = 0$).

The quantity $(A-B)M_S$ plays the role of the first constant in the simplified analysis of the behavior of the magnetization in the one-sublattice model. It is seen from (5) that it determines the saturation field as $m \to 1$:

$$H_K=-2A. \quad (6)$$

From the data of Fig. (7) and from (6) we can find the value of $M_S^2(2a-\lambda)$. For $x = 2$ and $x = 4$ it is equal to $\sim 6.7\times 10^5$ and $\sim 2.1\times 10^5$ erg/cm^3, respectively.

Assuming that the components of the anisotropy energy with the anisotropy constants K_2 and K_2' are mainly responsible for the anisotropy of the S blocks of one-ion nature, we can connect these constants by a relation that is valid for complexes of cubic anisotropy:

$$K_2=\tfrac{7}{12}K_0^c, \quad K_2'=\tfrac{2}{3}K_0^c;$$

here K_0^C is the cubic anisotropy constant in the traditional expansion in terms of the direction cosines in a Cartesian coordinate system. The value of K_0^C, which connects the constants K_i in the region of small substitutions of Sc in the S block, is of the same order of magnitude as the anisotropy constant of the spinel ferrite; we set it equal to one-half (per block sublattice) of the constant of nickel spinel, i.e., 0.45×10^5 erg/cm^3 at 80°K. Then the transition to the noncollinear structure will be characterized by the relation $K_1 < \tfrac{7}{6}K_0^C \approx 0.52\times 10^5$ erg/cm^3, and $K_2' = 0.21\times 10^5$ erg/cm^3.

It can be shown that the magnitude of the initial splitting in the FMR spectrum for Ni_2Sc_2W is equal in this case to $\gamma H_{c1} \approx [2(A-B)\cdot 4K_2'/M_S]^{1/2} \approx 3$ kOe. In the weak-field region, the FMR frequency is quadratically related to the field applied along the [001] axis.

In ferrite saturation fields, i.e., at $H > H_{c2}$, the dependence of the resonance frequency on the field is similar to the resonance conditions for a uniaxial ferromagnet with an effective anisotropy field $H_{c2} = 2K_1/M_S + 2aM_S$, which takes into account the gain in the energy of the dipole interaction of the blocks when the magnetic moments are arranged along the [001] axis and the local minimum of the energy of the one-ion anisotropy in this direction. Extrapolating the measured $\nu(H)$ dependence to fields $H_{c2} \approx 16$ kOe and using the relations for the anisotropy fields, we can separate the constants of the isotropic antiferromagnetic exchange interaction of the blocks and the dipole (or quasidipole) anisotropy. For the composition with $x = 2$ these constants turn out to be of the same order: $\lambda M_S^2 = 11\times 10^5$ erg/cm^3 and $aM_S = 4.2\times 10^5$ erg/cm^3; we note that they are somewhat smaller than the intrablock dipole interactions which enter with a negative sign in the constant K_1 (16×10^5 if no account is taken of the Sc substitutions).

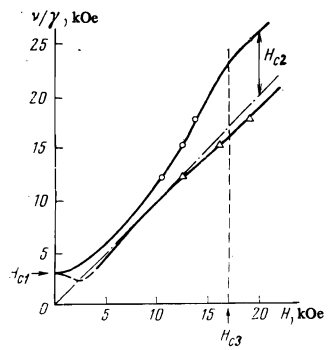

FIG. 9. Dependence of the FMR frequency on the external field.

Figure 9 shows the dependence of the FMR frequency on the external field, constructed from the obtained parameters for the ferrite Ni_2Sc_2W. When the field is oriented perpendicular to the [001] axis, at $H > H_{c1}$, the FMR frequency is practically proportional to the external field and depends quite weakly on the parameters of the investigated compositions with $x \geq 2$. The reason is that the crystal is practically isotropic for the magnetizations of each individual sublattice when the latter is oriented in the (001) plane of the crystal.

When the external-field vector is rotated near an axis lying in the basal plane, the orientation and magnitude of the resultant magnetization of the sample experience a discontinuity when the field passes through the direction of the spin axis of one of the blocks. This is clearly shown by the orientation dependence of the resonant field (Fig. 2). The angle $\theta_H = 45°$, at which the discontinuity of H_{res} is observed, correlates with the direction $\theta = 55°$ of the spin axes of the blocks, as obtained from neutron-diffraction data, if account is taken of the fact that investigations by the FMR method were carried out in relatively strong external fields.

Thus, the results of investigations by the FMR method do not contradict the model of the block magnetic structure as obtained from neutron-diffraction data, and constitute a switching of the magnetization of the blocks to a state with magnetic-moment projections of equal sign and magnitude when the field is applied along and across the [001] axis, and further increase of the field leads to a gradual decrease of the angle between the sublattices.

From the data on neutron diffraction, from measurements of the magnetization, and from investigations by the FMR method, we obtained the principal mechanisms of the anisotropic interactions and determined their constants. Thus, simultaneous use of the aggregate of the aforementioned methods has made it possible to determine almost completely the magnetic structure of the hexagonal ferrites of the system under consideration.

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235