THE ANOMALOUS INTENSITY DISTRIBUTION IN SATELLITES IN NEUTRON DIFFRACTION INVESTIGATIONS OF BLOCK HELICOIDAL STRUCTURES

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The intensity anomalies of a number of superstructure reflections observed in the neutron diffraction patterns of hexagonal ferrite single crystals of the $BaSc_xFe_{12-x}O_{19}$(M) system at 78 and 4.2°K are explained on the basis of a block magnetic helix model. It is shown that the anomalies are due to the superposition of ferro- and antiferromagnetic reflection satellites which occurs when the nuclear unit cell is divided into two magnetically identical blocks whose spin axes make a sufficiently large angle with one another.

NOTWITHSTANDING the satisfactory agreement between the calculated and experimental values of the intensities in studies of the system of scandium substituted type-M hexagonal ferrites,[1,2] the method of calculation could not explain a number of instances in which the experimental value of the intensity of the left satellite was lower than that of the right satellite. This contradicted the usual ideas about satellites in helix structures. Thus, for example, the observed intensity of the superstructure 003^+ reflection was higher than that of the 003^- for all compositions (Fig. 1), this anomalous redistribution of the intensity being more and more appreciable with decreasing scandium concentration.

Until the discovery in hexagonal ferrites of block helicoidal structures with large values of the angle of rotation of the spin axes from block to block, investigators were dealing with various types of helices which had one feature in common—the angle of rotation of the spins on going from one spin layer to the next was small. Depending on the nature of the positions of the spins (or their helical component in the case of the conical helix) with respect to one another in two neighboring layers one can distinguish two characteristic types of helices.

a) The positions of neighboring spins S are close to ferromagnetic (Fig. 2a). The repeat distance of the helix is given by the expression

$$\tau = \frac{2\pi}{\varphi}\Delta r, \qquad (1)$$

where φ is the angle of rotation of the spin on going from one spin layer to the next ($\varphi \sim 0$) and Δr is the distance between neighboring spin layers. In this case one should observe on the neutron pattern of a series of planes perpendicular to the axis of the helix splitting into satellites of the contributions of the fundamental nuclear-magnetic reflections.

b) The positions of the spins in two neighboring layers are close to antiferromagnetic ($\varphi \sim 180°$; Fig. 2). Cox et al.[3] considered this type of helix as a superposition of two helices of type a) shifted in phase. In this case splitting of superstructure ("antiferromagnetic") reflections should occur. The repeat distance

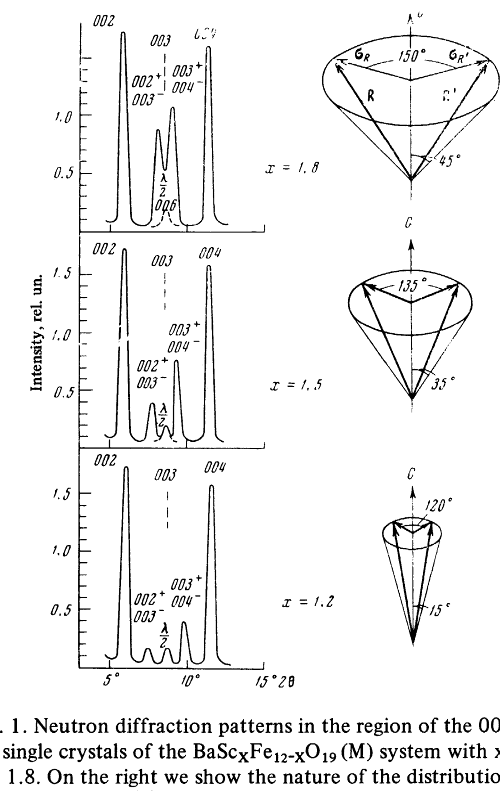

FIG. 1. Neutron diffraction patterns in the region of the 003 reflection for single crystals of the $BaSc_xFe_{12-x}O_{19}$ (M) system with x = 1.2, 1.5, and 1.8. On the right we show the nature of the distribution of the total moments (**R** and **R**′) of magnetically identical blocks of the unit cell and of their basal projections ($\sigma_{\mathbf{R}}$ and $\sigma_{\mathbf{R}'}$).

is given by the expression

$$\tau = \frac{2\pi}{\pi - \varphi}\Delta r. \qquad (2)$$

However, if the helix is not formed by small rotations of the spins of neighboring layers, but as a result of rotation by a considerable angle ($0 < \varphi < 180°$) of the spin axes of definite blocks of the unit cell, then one should observe splitting of the magnetic intensity of both ferro- and antiferromagnetic reflections. Precisely such a case is encountered in the investigation of the $BaSc_xFe_{12-x}O_{19}$ system.

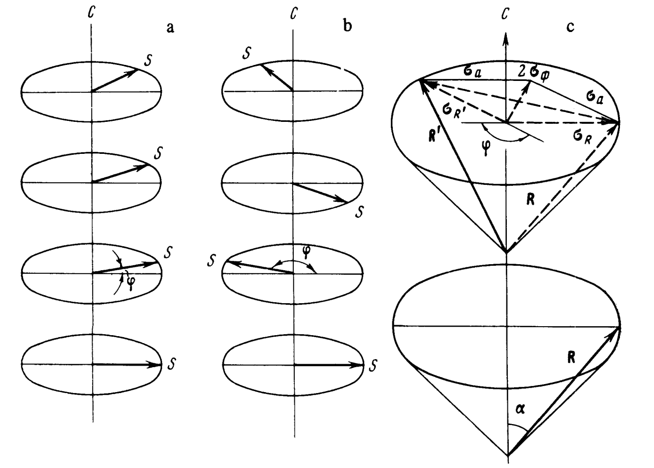

FIG. 2. Types of helices: a–planar with $\varphi \sim 0$, b–planar with $\varphi \sim 180°$, c–block conical with a large angle of rotation.

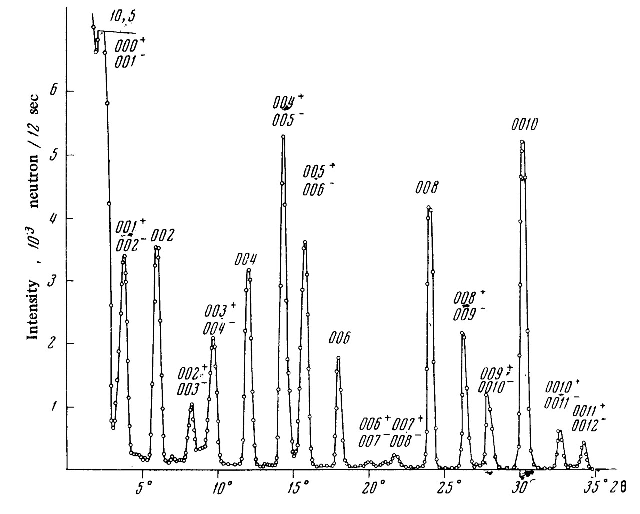

FIG. 3. Neutron diffraction pattern of a single crystal of $BaSc_{1.5}(M)$ at 4.2°K (λ = 1.22 Å).

Let us recall that in crystals of this system[1,2] the nuclear unit cell along the C axis is divided into two magnetically identical blocks (R and R′) whose total magnetic moments form a conical helix. Depending on the concentration of scandium ions the value of the angle of rotation from block to block of the basal component of the total magnetic moment is within the 120–150° range. The helix is formed by the addition along the C axis of unit cells with subsequent rotation of the spin axes of their blocks. In this case in the basal plane the total moments of the blocks **R** and **R′** have both appreciable ferro- (σ_f) as well as antiferromagnetic (σ_a) components (Fig. 2c) which must be taken into account in calculating the diffraction pattern.

Using expressions (1) and (2), we obtain for the ferro- and antiferromagnetic components of the helix two repeat distances:

$$\tau^{\Phi} = \tau_{002n} = \frac{2\pi}{\varphi}\frac{c}{N}, \tag{3}$$

$$\tau^{a} = \tau_{00(2n+1)} = \frac{2\pi}{\pi-\varphi}\frac{c}{N}, \tag{4}$$

where c is the lattice parameter of the unit cell along the hexagonal axis and N is the number of blocks (or rotations) per unit cell. Using this definition of the repeat distances, the Bragg equation and the relation for the interplanar distances for the satellites with the corresponding repeat distance, one readily obtains the expression

$$\sin\theta_{00(2n+1)^-} - \sin\theta_{002n^+} = \frac{\lambda}{4c}(2-N). \tag{5}$$

It follows hence that in the case in which the unit cell divides into two blocks (the right-hand side vanishes), the right-hand "ferromagnetic" satellite of the 002n reflection coincides with the left-hand satellite of the "antiferromagnetic" 00(2n + 1) reflection. Thus each superstructure reflection appearing when the unit cell divides into two magnetic blocks must be ascribed not one "ferromagnetic" set of indices, as in[1,2], but two corresponding sets as follows from Fig. 3.

In calculating the intensities of superstructure reflections of the basal series the value of the magnetic interaction vector | **q** | was determined from the expressions

$$q_{002n^{\pm}} = \frac{1}{2}\sin\alpha\cos\frac{\varphi}{2}, \quad q_{00(2n+1)^{\pm}} = \frac{1}{2}\sin\alpha\sin\frac{\varphi}{2}, \tag{6}$$

where the factor ½ was introduced to take into ac-

Table I

hkl	$\lvert F_{nuc}\rvert$, $\lvert qF_m\rvert$	$I_{theor}E_pE_s$	I_{exp}
000^+	1.56	27.23 } 62.41	65.03
001^-	3.39	35.18	
001^+	3.25	51.35 } 57.08	56.22
002^-	0.64	5.73	
002	2.75	39.66	52.65
002^+	0,63	2.69 } 28.90	19.34
003^-	2.21	26.21	
003^+	2.13	22.99 } 53.41	44.63
004^-	2,62	30.42	
004	3.93	48.44	52.05
004^+	2.51	21.63 } 111.72	116.69
005^-	7.47	90.04	
005^+	7.39	88.16 } 99.43	88.43
006^-	1.83	11.27	
006	3.87	38,06	35.63
006^+	1.73	8.31 } 12.42	8.24
007^-	1,20	4.11	
007^+	1.14	3.45 } 16,02	11.18
008^-	2.24	12.57	
008	8.54	90.36	98.03
008^+	2,05	8.96 } 65.21	61.50
009^-	5.90	56.25	
009^+	5,59	51.32 } 57,14	54.30
0010^-	1.68	5.82	
0010	10.25	100,66	103.03
0010^+	1.51	4.16 } 9.69	14.62
0011^-	1.76	5.53	
0011^+	1.70	5.01 } 5.04	12.99
0012^-	>0	0.03	

Table II

$BaSc_xFe_{12-x}O_{19}$	T, °K	φ, deg	α, deg	τ_{2n}, Å	τ_{2n+1}, Å	Agreement factor
$x = 0$	78	—	0	—	—	0,065
$x = 1.2$	78	120	15	36.4	69.9	0.094
$x = 1.5$	78	135	35	31.2	92.2	0.111
	4.2	135	60	31,2	92.2	0.106
$x = 1.8$	78	150	45	29.7	141.0	0.127

count the splitting of the magnetic intensity into two satellites and α is the half-apex angle of the cone of the helix. The calculation of the structure factors and the account of extinction were carried out by the usual methods. In Table I we present as an example the calculated and experimental data for the composition $BaSc_{1.5}(M)$ at 4.2°K.

An investigation of the composition $BaSc_{1.5}(M)$ at 4.2°K showed that the half-apex angle of the cone changes appreciably with changing temperature whereas the repeat distance of the helix remains unchanged. Table II presents the comparative characteristics of the conical helices for the investigated compositions and the corresponding values of the agreement factors.

It should be noted that the method under consideration explains the increase in the "anomalous" redistribution of the intensities in satellites with decreasing content of scandium ions. In fact, with decreasing replacement concentration the angle φ decreases leading to an increase of σ_f compared with σ_a (Fig. 1). Under these conditions the splitting of the comparatively large contributions to the 004 and 008 reflections and the combining of their intensities with those of the $00(2n+1)^{+}$ reflections leads with decreasing scandium concentration to a larger and larger "redistribution" of intensity into the series of "right-hand" satellites.

[1] O. P. Aleshko-Ozhevskiĭ, R. A. Sizov, V. P. Cheparin, and I. I. Yamzin, ZhETF Pis. Red. 7, 207 (1968) [JETP Lett. 7, 158 (1968)].

[2] O. P. Aleshko-Ozhevskiĭ, R. A. Sizov, I. I. Yamzin, and V. A. Lyubimtsev, Zh. Eksp. Teor. Fiz. 55, 820 (1968) [Sov. Phys.-JETP 28, (1969)].

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