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B. Y. Boucher, R. Buhl, and M. Perrin

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Compounds and Oxides. II

G. P. RODRIGUE, *Chairman*

Three-Sublattice Ferrimagnetic Structure

B. Y. BOUCHER, R. BUHL, AND M. PERRIN
CEA, Saclay and C.N.R.S., Bellevue, France

We show that, in the hypothesis of molecular field theory, a "star" structure (one direction for each sublattice moment) can exist and be stable. Such a structure becomes collinear for some $T_1 < T_c$. From the variation of the magnetization of one sublattice vs temperature in the neighbourhood of $T = T_1$, we deduce the high-temperature structure. The neutron diffraction measurements relative to $(Mn_xNi_{1-x})(Ni_xMn_{2-x})O_4$ $(0.74 < x < 0.93)$ show the existence of a "star" structure which explains the spontaneous magnetization values.

LET us consider a metallic oxide consisting of three types of magnetic cations of average moments μ_1, μ_2, μ_3 in an ordered state for $T < T_c$, the resultant of which is defined by σ. Let α, δ, ζ, β, γ, ϵ be the interaction constants of interactions in the molecular field model corresponding, respectively, to the interactions 1-1, 2-2, 3-3, 1-2, 1-3, and 2-3.

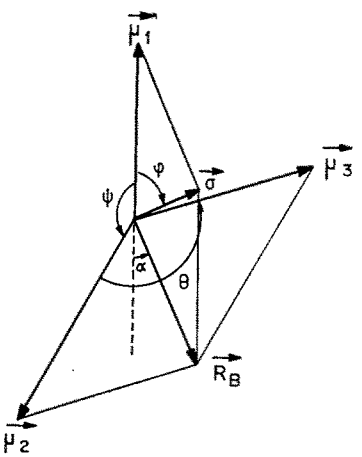

FIG. 1. Magnetic configuration of a three-sublattice structure.

EXISTENCE AND STABILITY OF STRUCTURES

The extremum of energy is obtained with the notations of Fig. 1 for $m_2 \sin\psi + m_3 \sin(\theta+\psi) = 0$ (1) and $m_1 \sin\psi - m_3 \sin\theta = 0$ (2) with $m_1 = \beta\gamma n_1\mu_1$, $m_2 = \epsilon\beta n_2\mu_2$, $m_3 = \epsilon\gamma n_3\mu_3$, and n_i representing the fraction of the surrounding ion i.

These equations have the following solutions (Fig. 1):

$$\sin\psi = \sin\theta = 0 \qquad (\theta, \psi = 0 \text{ or } \pi) \tag{1}$$

(solution for ferro- or ferrimagnetic collinear case)

$$\cos\psi = (-m_1^2 - m_2^2 + m_3^2)/2m_1m_2$$

$$\cos\theta = (m_1^2 - m_2^2 - m_3^2)/2m_2m_3$$

$$\cos(\theta+\psi) = (-m_1^2 + m_2^2 - m_3^2)/2m_1m_3. \tag{2}$$

(These solutions were foreseen by Niessen.[1]) [Case of a "star" structure, the Yafet-Kittel structure[2] being a particular case $(\beta = \gamma; |\mu_2| = |\mu_3|)$.]

The conditions, $\cos^2\theta < 1$ and $\cos^2\psi < 1$, can be written as $\{m_1^2 - (m_2+m_3)^2\}\{m_1^2 - (m_2-m_3)^2\} < 0$. The θ and ψ solutions (different from 0 or π) exist simultaneously; further, this equation shows that one can vary continuously from a "star" structure to a collinear one.

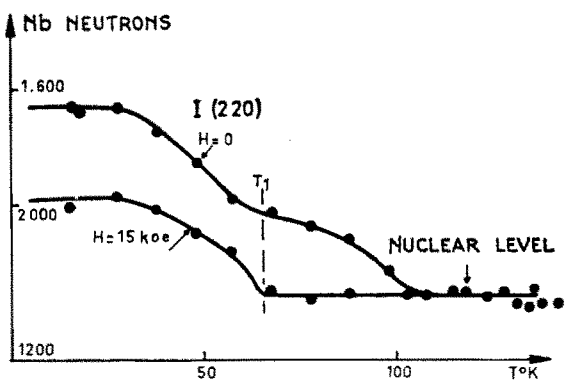

FIG. 2. Thermal variation of the (220) reflection.

Let us compare the energies of the different solutions

$$E_{\text{star}} - E_{\text{collinear}} = (\beta\gamma\epsilon)^{-1}(\sum_i U_i \mathbf{m}_i)^2 \quad \text{with} \quad U_i = \pm 1.$$

If $\beta\gamma\epsilon < 0$, the "star" structure is the more stable one.

THERMAL VARIATIONS

The temperature dependence of the magnetization for a sublattice can be written as

$$\mu_i = \mu_{i_0} B_{J_i}(\mu_{i_0} |\mathbf{h}_i| / kT).$$

In a "star" structure, the molecular field of a sublattice can be described as a function of its own magnetization. Then, this one varies independently of the other two until a certain temperature T_1 at which one of them will be sufficiently small so that the modulus of

[1] K. F. Niessen, Physica **19**, 1035 (1953).
[2] Y. Yafet and C. Kittel, Phys. Rev. **87**, 290 (1952).

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TABLE I. Shape of the magnetization thermal variation curve relative to one sublattice as a function of angles and field constants.

Values of ψ and θ for $T \geq T_1$	$\beta, \gamma > 0$ $\epsilon < 0$	$\gamma, \epsilon > 0$ $\beta < 0$	$\beta, \epsilon > 0$ $\gamma < 0$	$\beta, \gamma, \epsilon < 0$
$\psi=\pi \quad \theta=\pi$	case 2	case 1	impossible	case 2
$\psi=0 \quad \theta=\pi$	case 2	impossible	case 1	case 2
$\psi=0 \quad \theta=0F$	case 1	case 2	case 2	impossible
$\psi=\pi \quad \theta=0$	impossible	case 2	case 2	case 1

$\cos\theta$ and $\cos\psi$ would reach unity. At $T=T_1$, the collinear structure replaces the star structure, and, in this case, the molecular field is a function of the three sublattice magnetization. This gives rise to a singular point at $T=T_1$ on the curve depicting thermal variations of the magnetization of one sublattice. Two cases are possible: The slope to the right side of T_1 is smaller (case 1) or greater (case 2) than the slope on the left side. Table I summarizes the results as a function of the field constant signs and the disposition of moments at $T \geq T_1$.

APPLICATION TO THE CUBIC SPINEL $(Mn_xNi_{1-x})(Mn_{2-x}Ni_x)O_4$ $(0.74 < x < 0.93)$

The diffraction pattern indicates that the nickel and manganese atoms are distributed statistically in the B sites. If we suppose Mn_A–Mn_B interactions different from Mn_A–Ni_B interactions, we have a case where "star" structure is possible $(\beta \neq \gamma)$. But the disorder due to the statistical ionic distribution in B sites enables only the detection of an average moment R_B (in length and direction), sum of the moments of B-site ions.

The neutron diffraction patterns at 4.2°K demonstrate the presence of parallel moments in each of the A and B sites. The value of the B-site moments is clearly smaller than one could expect when the moments are really aligned. One is thus concerned with "average moments." When a magnetic field is applied along the diffusion vector, the magnetic intensities decrease or increase (according to the x values) without vanishing, despite the absence of anisotropy $(x > 0.80)$. This effect cannot be explained by a collinear model. The B-site moments are thus not aligned to those of A sites. The spontaneous magnetization determined by the neutron diffraction studies for the "star" model are in good agreement with the results obtained by direct measurements. The results are tabulated in Table II.

TABLE II. Observed values of μ_A, $R_B/2$, σ (Bohr's magnetons) and α as a function of inversion x.

x	μ_A	$R_B/2$	α	σ neutrons	σ measured
0.74	3.48	1.91	3°25′	0.40	0.35
0.80	3.75	2.0	8°10′	0.60	0.66
0.93	3.90	1.33	21°30′	1.73	1.75

For $T \simeq 90$°K, the patterns indicate a collinear structure; the magnetic intensities cancel out when the magnetic field is applied.

THERMAL VARIATION $(x = 0.93)$

With this model the (220) reflection depends only upon μ_A^2. The curve of thermal variation (Fig. 2) is of the type 2. Knowing the variation of spontaneous magnetization $\mu(T)$, the evolution of α and β angles are deduced as a function of temperature.

We have determined the resultant of the magnetic moments of the ions placed in the B sites at 4.2°K; if we make the hypothesis that the manganese ions are ferromagnetically coupled amongst themselves and that they carry at very low temperature a moment of $4\mu_B$ (the nickel ions being coupled in the same way with a moment of $2\mu_B$), the structure is then defined by $\psi_0 = 143°$ and $\theta_0 = 158°$.

The relations (1) and (2) give negative values to β, γ, ϵ. For $T \geq T_1$, Table I indicates that $\theta = \pi$, and from the variation of the angle ϕ, we deduce that the moments of manganese and nickel ions in the B sites are aligned respectively antiparallel and parallel to the moments of the A-site ions.

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