

Helical Spin Arrangement in Chromium Metal*

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The lowest energy spin arrangements in a body centered cubic crystal with first, second, and third nearest neighbor exchange interactions are given. The results may explain the experimental neutron diffraction pattern for chromium single crystals as observed by Corliss, Hastings, and Weiss. The susceptibility of the helical array is calculated.

IT has been suggested by Yoshimori¹ and Villain² that helical spin arrangements may occur in magnetic crystals. Such an arrangement has been found in manganese dioxide by Erickson.³ We discuss here the spin arrangement observed in chromium single crystals by Corliss, Hastings, and Weiss.⁴ They interpret the neutron diffraction patterns obtained in terms of an antiphase domain structure. The existence of a helical array in Cr was suggested by Kaplan.⁵ The present note treats the stability of all possible lowest energy spin arrangements following the method used by Villain for Bravais lattices. In addition we suggest the use of susceptibility measurements to complete the information on the exchange integrals involved in the theory.

The origin of the helix is most easily understood in terms of a one dimensional model with antiferromagnetic interactions between second neighbors. If there is no other coupling, the spins split into two independent antiferromagnetic lattices. We now add a small coupling (of arbitrary sign) J_1 between first neighbors, and consider a small helical distortion of both sublattices, with 2θ as the angle between second

neighbor spins: the J_1 coupling decreases the energy to first order in θ , while the antiferromagnetic coupling increases the energy only to second order, so that the helix is indeed more stable (Fig. 1).

If we use a Heisenberg model for chromium, at least three different exchange integrals J_1, J_2, J_3 (connecting 1st, 2nd, and 3rd neighbors as shown in Fig. 2) are required to obtain a helix. The exchange energy is

$$\mathcal{H} = \sum_{i>j} 2J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

To discuss the stability of all possible spin arrangements we follow the method of Luttinger and Tisza⁶ as improved by Villain² for Bravais lattices. We define

$$g(\mathbf{k}) = \sum_j J_{ij} e^{i\mathbf{k} \cdot \mathbf{R}_{ij}}. \quad (2)$$

The stable configurations are:

$$S_i^\alpha = S \cos \mathbf{k}_c \cdot \mathbf{R}_i, \quad S_i^\beta = S \sin \mathbf{k}_c \cdot \mathbf{R}_i, \quad S_i^\gamma = 0, \quad (3)$$

where \mathbf{k}_c is such that $g(\mathbf{k}_c)$ is the absolute maximum of $g(\mathbf{k})$, and α, β, γ are three axes mutually orthogonal, but otherwise arbitrary. If \mathbf{k}_c is a general vector of the zone, (3) represents a helical arrangement. In our case, there are three possible helical arrangements: I $[k_x, (2\pi/a) - k_x, (2\pi/a) - k_x]$, II $(k_x, 0, 0)$, and III $[k_x, (2\pi/a) - k_x, 0]$ plus all similar ones obtained by symmetry. The stability domains are shown in Fig. 3, where we further assume $J_1 < 0$, which will turn out to be the correct sign for chromium. Neutron diffraction lines occur when the scattering vector $\mathbf{\kappa}$ is equal to $\pm \mathbf{k}_c + 2\pi\tau$ (where $2\pi\tau$ is a reciprocal lattice vector). For a helical arrangement the lines cannot in general be indexed in any multiple of the unit cell. The experimental results for chromium show that (II) is the actual

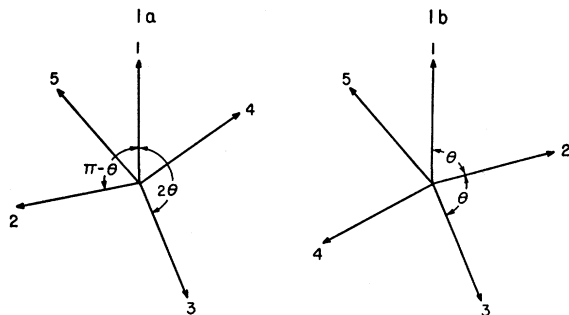


FIG. 1. (a) Proper helix viewed along axis with antiferromagnetic second neighbor interaction and small antiferromagnetic nearest neighbor interaction. The numbers refer to successive spins along the screw axis. (b) Proper helix viewed along axis with antiferromagnetic second neighbor interaction and small ferromagnetic nearest neighbor interaction.

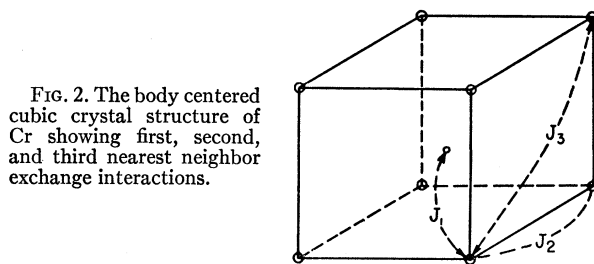


FIG. 2. The body centered cubic crystal structure of Cr showing first, second, and third nearest neighbor exchange interactions.

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¹ A. Yoshimori, J. Phys. Soc. (Japan) **14**, 807 (1959).

² J. Villain, J. Phys. Chem. Solids (to be published).

³ R. A. Erickson (to be published).

⁴ L. Corliss, J. Hastings, and R. J. Weiss, Phys. Rev. Letters **3**, 211 (1959).

⁵ T. A. Kaplan, Phys. Rev. **116**, 888 (1959).

⁶ J. M. Luttinger and L. Tisza, Phys. Rev. **70**, 954 (1946).

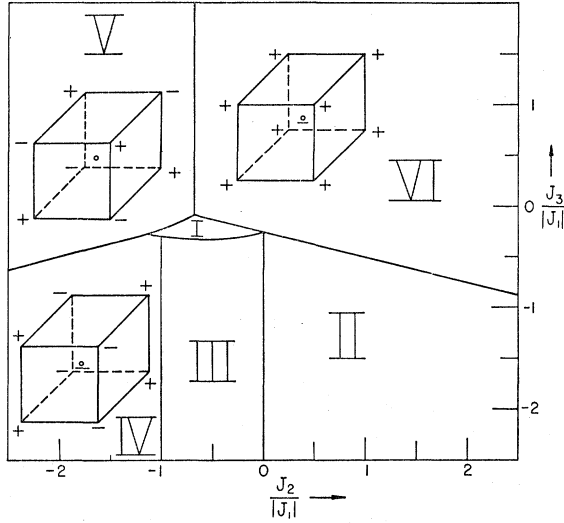


FIG. 3. The regions of stability of the various spin structures in the $J_2/|J_1|, J_3/|J_1|$ plane for antiferromagnetic coupling between nearest neighbors ($J_1 < 0$).

arrangement. The spins are parallel in each given (100) plane. (To ensure that they are, we need $J_2 > 0$.) The orientation in successive planes differs. The over-all coupling between next nearest planes is antiferromagnetic, as can be seen from Fig. 3 ($J_2 + 4J_3 < 0$). This could have been foreseen from our introductory argument. The length of k_c is given by

$$\cos(k_c a/2) = -J_1/(J_2 + 4J_3). \quad (4)$$

If we assume for instance $k_c a = 2\pi - \pi/14$ (which is close to the experimental value) we get $\cos(k_c a/2) = -0.97$. As regards orientation of the spin plane ($\alpha\beta$) with respect to the crystal axes, the intensity ratios observed by Corliss et al.⁴ are consistent with the following picture: there are three types of magnetic domains $(k_c, 0, 0)$, $(0, k_c, 0)$, and $(0, 0, k_c)$. In each domain k_c is parallel to γ ; this is a "proper" helix in Yoshimori's notation.¹ A consideration of the dipolar energies⁷ shows

⁷ M. H. Cohen and F. Keffer, Phys. Rev. **99**, 1128 (1955).

that proper helices are indeed favored from that standpoint. Other magnetocrystalline energies are probably more important, however, as emphasized by Kaplan.⁵ Any discussion of this effect, however, suffers from the lack of certainty in its sign.

Now, susceptibility measurements may possibly complete our information on the J 's. At $T=0$ the susceptibility tensor is (per atom):

$$\chi_{\alpha\alpha} = \chi_{\beta\beta} = \frac{g^2 \mu_B^2}{2[2J(k) - J(0) - J(2k)]}, \quad (5)$$

$$\chi_{\gamma\gamma} = \frac{g^2 \mu_B^2}{2[J(k) - J(0)]} \geq \chi_{\alpha\alpha}. \quad (6)$$

By applying a high field, parallel to one cubic axis, one could in principle increase the percentage of domains with the spin plane perpendicular to the field. The value of the Néel temperature is probably not reliable information toward determining the J 's since this model is limited to low temperatures. For iron⁸ J calculated from specific heat data and T_c is 0.011 ev; while J calculated by spin wave theory is 0.018 ev.

The following problems are still to be solved: (1) why are the magnetic properties of chromium so sensitive to impurity content or preparation techniques; (2) does the pitch change with temperature (with the above Heisenberg model, and within molecular field approximation, it does not); (3) what is the appropriate band picture for a helical spin distortion? This last problem might be worked out by an extension of the Herring technique for spin waves in metals.^{9,10}

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⁸ J. A. Hofmann, A. Paskin, K. J. Tauer, and R. J. Weiss, J. Phys. Chem. Solids **1**, 45 (1956).

⁹ C. Herring, Phys. Rev. **85**, 1003 (1952); **87**, 60 (1952).

¹⁰ P. -G. de Gennes, Compt. rend. **22**, 1730 (1956).