Classical Spin-Configuration Stability in the Presence of Competing Exchange Forces*

T. A. KAPLAN
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts
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It is pointed out that Yafet-Kittel triangular arrangements are not stable in the cubic spinel. The stability criterion used is that the classical Heisenberg energy should not decrease for small, but otherwise arbitrary, spin-deviations from the configuration of interest. It is found that the Yafet-Kittel-Prince configuration can probably be stabilized by a sufficient tetragonal distortion of the pattern of B-B interactions. In addition, the classical ground state is found for the antiferromagnetic body-centered cubic lattice with first, second, and third neighbor antiferromagnetic interactions (with parameters J_1, $J_1\sigma_2$ and $J_1\sigma_3$): the spin $\mathbf{S}(\mathbf{R}_n)$ at lattice point $\mathbf{R}_n$ is independent of time, is always parallel to one plane, P, and the angle made by $\mathbf{S}(\mathbf{R}_n)$ with a fixed line in P is of the form $\mathbf{k}\cdot\mathbf{R}_n$ for $\mathbf{R}_n$ a cube corner, and of the form $\mathbf{k}\cdot\mathbf{R}_n+\pi$ for $\mathbf{R}_n$ a body-center position with the vector $\mathbf{k}$ determined by the σ_i. The neutron diffraction pattern for such a "spiral" configuration (with $\sigma_2\sim0.6$, $\sigma_3\sim0.1$, for example) bears a close relationship with the unusual pattern obtained by Corliss, Hastings, and Weiss with a single crystal of chromium.

YAFET and Kittel[1] showed that, for spinels with large enough antiferromagnetic A-A and/or B-B interactions compared to the A-B interaction, certain triangular spin arrangements would have a lower energy (as calculated from the molecular-field theory) than the conventional collinear configuration. As far as we are aware, there has been no discussion of the possibility that there might exist configurations of still lower energy. The main purpose of this note is to point out that the Yafet-Kittel (Y-K) configurations generally are not stable in the cubic spinel. The stability of other configurations is also discussed.

The stability criterion used is that the classical Heisenberg energy should not decrease for small, but otherwise arbitrary, spin deviations from the spin configuration of interest. This is a straightforward extension of the usual molecular-field theory of the ground state—the present approach reduces to the latter if we restrict ourselves to the very small class of spin deviations in which large numbers of spins are assumed to be rigidly parallel (the sublattice division). Furthermore, this criterion must be satisfied to have a stable, semiclassical spin-wave solution for the excited states.

We have considered the special case of the Y-K configurations suggested by Prince[2] for copper chromite (configuration C). The application of the stability criterion goes, in outline, as follows. The energy, E, consists of nearest-neighbor A-B interaction terms with exchange integral J_{AB}, and nearest-neighbor B-B interactions (the A-A interactions being neglected). To take into account a tetragonal distortion,[2] we assume $J_{BB}\neq J_{BB}'$, where J_{BB} and J_{BB}' are the B-B exchange integrals effective, respectively, within, and between, planes perpendicular to the c-axis ([001]). (All J's are are <0.) In C, there are three coplanar spin directions, z_1, z_2, z_3 (Fig. 1); the A-spins (magnitude S_A), are in z_1, one set of B-spins (B_2) is in z_2, the other (B_3) in z_3. [All B's in one (001) plane are parallel, the direction alternating $z_2-z_3\cdots$ from one (001) plane to the next. All B's have magnitude S_B.] If we write the spin-vectors for the A, B_2, and B_3 sites in terms of their $\mathcal{S}_1$, $\mathcal{S}_2$, $\mathcal{S}_3$ components, respectively, configuration C is realized when the respective x and y_i components are zero, the z_i components positive. The energy for small deviations from C is then obtained (as in standard spin wave calculations) by expanding E in terms of the x and y_i components, keeping up to quadratic terms: $E=E(\theta)$ +linear terms+quadratic terms, where $E(\theta)$ is the energy of C. In the quadratic part there are no cross $(x-y_i)$ terms. We choose θ to minimize $E(\theta)$; then the linear terms vanish and θ satisfies $\cos\theta=\xi$ when $\xi<1$, $\theta=0$ otherwise, where $\xi=3J_{AB}S_A/4J_{BB}'S_B$. (This checks with the results of Yafet and Kittel when $J_{BB}=J_{BB}'$.)

Again following spin-wave methods, we expand the x and y_i components in Fourier series, introducing $\mathbf{k}$ (rationalized, reduced reciprocal vectors). After a further transformation (utilizing the symmetry between the two crystallography different tetrahedral sites) we find $\Delta E=E-E(\theta)$ to be a sum of two sets, X and Y, of six-variable quadratic forms, arising from the x and y_i spin-components, respectively. (Six corresponds to the number of spins per primitive unit cell.) The

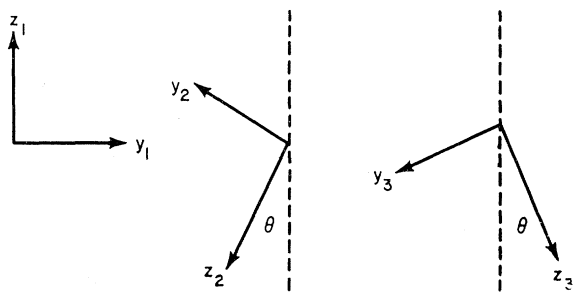

FIG. 1. The coordinate systems, $\mathcal{S}_i$; the x axes are out of the paper.

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[1] Y. Yafet and C. Kittel, Phys. Rev. **87**, 290 (1952).

[2] E. Prince, Acta Cryst. **10**, 554 (1957).

variables are real and the matrices, $M(\mathbf{k})$, of the forms are real and symmetric.

A necessary and sufficient condition for $\Delta E>0$ is that the eigenvalues of the $M(\mathbf{k})$ be positive. For $\mathbf{k}$ in [110], the forms can easily be factored into 1-, 2-, and 3- variable forms, the last of which, in X, shows instability for $\xi<1$ in the cubic case ($J_{BB}=J_{BB}'$): the determinant of its matrix is <0 for all $k\neq 0$. If (and only if) $J_{BB}'\geqslant 3J_{BB}$, the form is positive for all $k\neq 0$.

By considering other $\mathbf{k}$'s—all values of $\mathbf{k}$ along [001] and small values in arbitrary directions—we are led to suggest that $J_{BB}'\geqslant 3J_{BB}$ is probably sufficient for stability of the triangular configuration ($\xi<1$). The instability of the Y-K-Prince configuration of course implies that all the (degenerate) Yafet-Kittel triangular configurations arising in the absence of A-A interactions are unstable in the cubic case. Even for the collinear, Néel case ($\xi\geqslant 1$) in the cubic spinel, we find the configuration to be unstable when $\xi<\xi_0$, with $\xi_0\sim 1.1$.

From these results we conclude (1), that the temperature-region of validity of Anderson's[3] suggestion (as to the nonexistence of long-range-ordered triangular configurations on the octahedral sublattice) can be extended to absolute zero, and (2), a spin-wave calculation based on C as a ground state can be sensible only if there is lattice distortion.

The stability criterion has also been applied to the previously suggested[1] triangular spin configuration on a two-dimensional, triangular lattice. We found this stable.

The interesting question concerning the classical ground state when the "intuitive" guess is unstable, is, of course, a difficult one. We have considered this for a simpler system, namely an antiferromagnetic body-centered cubic lattice, the "basic" spin-configuration (C_0) having the spins at cube corners (A) "up", those at the cube centers (B) "down." First, second, and third neighbor antiferromagnetic interactions are assumed (with parameters $J_i>0$). Our stability criterion yields the condition $m(\mathbf{k})\geqslant 0$ where $m(\mathbf{k})$ is the lowest branch of the appropriate eigenvalue spectrum. For example, for small $\mathbf{k}$ this gives $J_1-J_2-4J_3\geqslant 0$ [somewhat stronger than the (intuitive) molecular-field criterion]. By considering the corresponding eigenvectors, (which show how the spin-system "wants to deviate" from C_0 when $m(\mathbf{k})<0$), we were led to consider the class, $C_\mathbf{k}$, of equilibrium configurations given by $\mathbf{S}_{n^\nu}=\pm(\xi\sin\theta_{n^\nu}+\zeta\cos\theta_{n^\nu})$, where $\nu=A$ or B, $+$ for $\nu=A$, $-$ for $\nu=B$, $\theta_{n^\nu}=\mathbf{k}\cdot\mathbf{R}_{n^\nu}$, $\mathbf{R}_{n^\nu}$ and $\mathbf{S}_{n^\nu}$ are, respectively, the position and spin-vector at site $n-\nu$, and ξ and ζ are orthonormal vectors. In other words, the angle between an A-spin and ζ increases uniformly as one moves along $\mathbf{k}$, going from 0 to 2π in a distance $\lambda=2\pi/k$; similarly for the B's. We find, surprisingly, that $E(\mathbf{k})-E(0)=bm(\mathbf{k})$, (exactly), where b is a positive number, and $E(\mathbf{k})$ is the energy of $C_\mathbf{k}$. Hence, if C_0 is unstable with respect to small deviations characterized by wave-vector $\mathbf{k}$, $C_\mathbf{k}$ will be of lower energy.[4]

The considerations of the previous paragraph may be of interest in connection with a striking neutron-diffraction pattern recently obtained with a single crystal of chromium,[5] where each magnetic peak expected for C_0 is split into a number of peaks, with angular separation of a few degrees. The diffraction pattern for $C_\mathbf{k}$ with $\lambda\cong 30$ lattice constants will show a similar splitting. Furthermore, for an appropriate choice of the J_i, e.g., $J_2/J_1=0.58$, $4J_3/J_1=0.424$, a minimum occurs in $E(\mathbf{k})$ at about this wavelength. However the reduction in energy (from that of C_0) is $\approx 10^{-4}E(0)$, so that smaller forces (e.g. dipole-dipole) must be considered.[6]

In connection with the spin waves, we mention a preliminary result which shows that triangular configurations may be expected to exhibit rather unusual spin-wave spectra. Namely, for configuration C with $\mathbf{k}$ in [001], we find two acoustic branches, one quadratic, the other linear in k for small k (when the net spin is nonzero).

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Note added in proof.—After submitting the present paper for publication, the author became aware of papers by Yoshimori [J. Phys. Soc. Japan **14**, 807 (1959)] and Villain (to appear in J. Phys. Chem. Solids) in which the concept of "spiral" (or "helical") spin-configurations is also suggested (in connection with different materials).

[3] P. W. Anderson, Phys. Rev. **102**, 1008 (1956).

[4] We have shown that $C_\mathbf{k}$ will include the lowest state, using the method of J. M. Luttinger and L. Tisza [Phys. Rev. **70**, 954 (1946)]. We are indebted to P. W. Anderson (private communication) for suggesting this approach.

[5] Corliss, Hastings, and Weiss, Phys. Rev. Letters **3**, 211 (1959).

[6] Preliminary calculations indicate that the pseudodipole energy with the same sign as the magnetic dipolar energy will prefer C_k over C_0 (for the J_i of our example), and furthermore will tend to put $\mathbf{k}$ in a cubic direction with $\mathbf{k}\cdot\xi=\mathbf{k}\cdot\zeta=0$ (in agreement with the diffraction results[5]). Other forces are being investigated.