Long-range effective interactions and spin canting: A model for amorphous magnetism

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The onset of canting for a ferromagnet with one and two antiferromagnetic defect bonds has been studied analytically. For individual negative defect bonds which are unable to cause canting, there is a long-range effective dipolar-like interaction which determines whether or not the system will cant. An effective Hamiltonian is obtained which applies more generally. A canted state at low temperatures, but a collinear state at somewhat higher temperatures, can be expected on the basis of this effective Hamiltonian.

In this paper we consider the spin canting (or tipping) instability in classical two-component (XY) and three-component (Heisenberg) spin systems on square and simple cubic lattices when antiferromagnetic bonds J' are substituted for the host ferromagnetic nearest-neighbor exchange bonds J. These are the simplest possible ferromagnetic systems, and the simplest possible exchange defects that can be placed in such systems. We emphasize the case where individual defect bonds do *not* cause canting. Despite the short-range nature of the interactions, the system can become unstable due to effective long-range dipole-like interactions between spin distortions centered at the defect bonds and mediated by the ferromagnetic host. This is relevant to the degradation of magnetic order when ferromagnetic systems are subject to the alloying process, such as in amorphous magnets. The case where individual defects *do* cause distortions has been discussed by Villain,[1] who considered the "canted local state," following Nowik and Rosencwaig.[2,3]

Our considerations are analytical, but were stimulated by numerical simulations,[4,5] and we have performed additional simulations to verify certain aspects of the theory. It should be kept in mind that the theory has been developed for infinite systems, whereas the simulations have been performed for large periodic systems, which necessitates corrections due to neighboring cells.

For both XY and Heisenberg spin systems, the excess energy of a system with nearest-neighbor exchange constants J due to tippings of the spins $\delta\mathbf{S}_i$ from the aligned direction $\mathbf{S}_i^{(0)}=\hat{\mathbf{z}}$ is given by

$$\mathcal{H}_0=\tfrac{1}{2}J\sum_{\langle ij\rangle}(\delta\mathbf{S}_i-\delta\mathbf{S}_j)^2\ , \tag{1}$$

where $\langle ij\rangle$ denotes a sum over all nearest-neighbor pairs (no double counting). If a bond J' replaces the J bond connecting site a and site b, then the part of the perturbation quadratic in the spin tippings is given by

$$\delta\mathcal{H}=\tfrac{1}{2}(J'-J)(\delta\mathbf{S}_a-\delta\mathbf{S}_b)^2\ . \tag{2}$$

The sum of (1) and (2) gives a quadratic form whose eigenvalues must all be positive if the system is to be stable.

To study this stability, one searches for singularities of the Green's function for the operator K, defined by[5]

$$\mathcal{H}=\sum_{\langle i,j\rangle}K_{ij}\delta\mathbf{S}_i\cdot\delta\mathbf{S}_j\ . \tag{3}$$

With $G\equiv(\omega^2-K)^{-1}$, $G^0\equiv(\omega^2-K^0)^{-1}$, where K^0 is appropriate to $\mathcal{H}_0$, and $\delta K\equiv K-K^0$, we have the familiar equation

$$G=G^0+G^0\delta KG \tag{4}$$

or

$$[1-G^0\delta K]G=G_0\ . \tag{5}$$

A static instability occurs if, for $\omega=0$, G is singular but G^0 is not. In that case

$$\det[1-G^0\delta K]=0\ . \tag{6}$$

For the square and simple cubic lattices of side a_0, the eigenfunctions of K^0 are $\theta_i^{\mathbf{k}}=\langle i\mid\mathbf{k}\rangle=(1/\sqrt{N})e^{i\mathbf{k}\cdot\mathbf{r}_i}$, and the eigenvalues in $K_{ij}^0\theta_j^{\mathbf{k}}=\omega_k^2\theta_i^{\mathbf{k}}$ are given by

$$\omega_k^2=2J[\sin^2(k_xa_0/2)+\sin^2(k_ya_0/2)]$$

for the square lattice and

$$\omega_k^2=2J[\sin^2(k_xa_0/2)+\sin^2(k_ya_0/2)+\sin^2(k_xa_0/2)]$$

for the simple cubic lattice. In both cases,

$$(G^0)_{ij}=\frac{1}{N}\sum_k\frac{1}{\omega^2-\omega_k^2}e^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}\ , \tag{7}$$

where the sum $\sum_k$ is over the N points in each Brillouin zone.

For a single defect bond, (6) reduces to a 2×2 matrix; for two defect bonds of the same strength connected by a common site, a 3×3 matrix; and for two disconnected defects of the same strength bonds, a 4×4 matrix. In the latter two cases the determinants can be reduced to 2×2's.

With $\Delta\equiv(J'/J-1)$, the single defect bond problem leads to the condition $\Delta=-d$, where $d=2,3$ for the square and simple cubic lattices. Thus the critical value J_c' for stability with respect to a single negative defect bond is

$$J_c'/J = 1-d \ , \tag{8}$$

so that square lattices become unstable if $J'/J < -1$, and simple cubic lattices go unstable if $J'/J < -2$.[6] Calculations on a 30×30 ($d=2$) periodically repeated system yielded -0.9915 for this ratio, the lack of perfect agreement being due to both convergence and finite-size effects.

We now turn to the case where $J'<0$ but $J'/J > 1-d$, so that individual defect bonds do not cause the system to be unstable to canting. For the case of two negative defect bonds J' the instability condition is that

$$\left[1+\frac{\Delta}{d}\right]^2 = \Delta^2 I^2 \ , \tag{9}$$

with the effective interactions $I = J(2G^0_{ab} - G^0_{aa} - G^0_{ac})$ for the connected bonds $(\mathbf{a},\mathbf{b})$, $(\mathbf{b},\mathbf{c})$, and $I = J(G^0_{ad} + G^0_{bd} - G^0_{ac} - G^0_{bd})$ for the disconnected bonds $(\mathbf{a},\mathbf{b}),(\mathbf{c},\mathbf{d})$. (These are equivalent when $\mathbf{d}=\mathbf{b}$, since then $G^0_{bd} = G^0_{ad} = G^0_{ab}$ and $G^0_{bd} = G^0_{aa}$, due to the translational and fourfold rotational invariances of the unperturbed state.) Thus the critical value J_c' for stability with respect to two negative defect bonds is

$$J_c'/J = 1 - \frac{1}{d^{-1} + |I|} \ . \tag{10}$$

Explicitly, for $d=2$ and *connected* defect bonds: $I = \frac{1}{2} - (1/\pi)$ for opposite bonds (i.e., both along $\hat{\mathbf{x}}$ or both along $\hat{\mathbf{y}}$), and $I = (2/\pi) - \frac{1}{2}$ for adjacent bonds (i.e., one along $\hat{\mathbf{x}}$ and one along $\hat{\mathbf{y}}$). Thus

$$J_c'/J = 1 - \frac{\pi}{2} = 0.5708 \quad \text{(opposite bonds)} \ , \tag{11}$$

$$J_c'/J = -(\pi - 1)^{-1} = 0.4669 \quad \text{(adjacent bonds)} \ . \tag{12}$$

Calculations on a 30×30 periodically repeated system yielded -0.5683 and -0.4623 for these ratios. Thus, for $d=2$, defect pair induced canting will occur only for $-1 < J'/J < -0.4669$, where the upper limit holds only for adjacent bonds.

For $d=2$ and *disconnected* negative defect bonds (separated by $\mathbf{R}$), when the bond directions $\mathbf{b}-\mathbf{a}$ and $\mathbf{d}-\mathbf{c}$ are collinear (as in the case of opposite bonds), we employ

$$I_{xx} = \frac{1}{N}\sum_{\mathbf{k}} \frac{\sin^2 \frac{1}{2} k_x a_0 \cos \mathbf{k}\cdot\mathbf{R}}{\sin^2 \frac{1}{2} k_x a_0 + \sin^2 \frac{1}{2} k_y a_0} \ , \tag{13}$$

and when $\mathbf{b}-\mathbf{a}$ and $\mathbf{d}-\mathbf{c}$ are perpendicular (as in the case of adjacent bonds), we employ

$$I_{xy} = \frac{1}{N}\sum_{\mathbf{k}} \frac{\sin \frac{1}{2} k_x a_0 \sin k_y a_0 \cos \mathbf{k}\cdot\mathbf{R}}{\sin^2 \frac{1}{2} k_x a_0 + \sin^2 \frac{1}{2} k_y a_0} \ . \tag{14}$$

Asymptotically,

$$I_{xx} \to -\frac{1}{2\pi (R/a_0)^2} \cos 2\theta, \quad \text{as } R/a_0 \to \infty \ , \tag{15}$$

$$I_{xy} \to -\frac{1}{2\pi (R/a_0)^2} \sin 2\theta, \quad \text{as } R/a_0 \to \infty \ , \tag{16}$$

where $\mathbf{R} = (R\cos\theta, R\sin\theta)$, so that these look like two-dimensional dipole interactions. Indeed, they can be combined in the form

$$I \to -\frac{1}{2\pi (R/a_0)^2} (\hat{\boldsymbol{\mu}}_1\cdot\hat{\boldsymbol{\mu}}_2 - 2\hat{\boldsymbol{\mu}}_1\cdot\hat{\mathbf{R}}\hat{\boldsymbol{\mu}}_2\cdot\hat{\mathbf{R}}) \ ,$$

$$\text{as } R/a_0 \to \infty \ . \tag{17}$$

To treat the case $d=3$, for opposite and adjacent bonds one may employ the tables of Maradudin *et al.*[7] for the Green's function of a simple cubic lattice, which yields $|I_{\text{opp}}| = 0.2470$ and $|I_{\text{adj}}| = 0.2716$. Then (10) gives

$$J_c'/J = -0.7232 \quad \text{(opposite)} \ , \tag{18}$$

$$J_c'/J = -0.6531 \quad \text{(adjacent)} \ . \tag{19}$$

Thus, for $d=3$, defect-pair-induced canting will occur for $-2 < J'/J < -0.6531$, where the upper limit holds only for adjacent bonds.

For more distant spins, the $d=3$ analog of (17) is given by

$$I \to -\frac{1}{4\pi (R/a_0)^3} (\hat{\boldsymbol{\mu}}_1\cdot\hat{\boldsymbol{\mu}}_2 - 3\hat{\boldsymbol{\mu}}_1\cdot\hat{\mathbf{R}}\hat{\boldsymbol{\mu}}_2\cdot\hat{\mathbf{R}}) \ ,$$

$$\text{as } R/a_0 \to \infty \ . \tag{20}$$

Consider now the situation if the spins cant; in this case, one must determine the new, noncollinear, ground state. For a weakly unstable system, the spins will tip (or cant) by only a small amount before quartic terms in the transverse spin components stabilize the system. (The largest contribution will come from the vicinity of the spin defects, where the spins tend to counter align.) This can be modeled with an effective energy given by[4,5]

$$E = \tfrac{1}{2} a (M^2_{1\perp} + M^2_{2\perp}) + \tfrac{1}{4} b (M^4_{1\perp} + M^4_{2\perp}) + c\,\mathbf{M}_{1\perp}\cdot\mathbf{M}_{2\perp} \ , \tag{21}$$

where $|c/a| = |\Delta I/(1+\Delta/d)|$ in order to ensure that (21) go unstable for $M_{1\perp}, M_{2\perp} \neq 0$ when (9) is satisfied. In (21), $\mathbf{M}_{1\perp}$ and $\mathbf{M}_{2\perp}$ are *induced* source strengths, limited by the nonlinear term in b (which is very nearly proportional to $-J'$).[5] The asymptotic part of the spin distortion $\mathbf{S}_1(\mathbf{r})$ produced by the source $\mathbf{M}_{1\perp}$ is proportional to $\mathbf{M}_{1\perp}(\boldsymbol{\mu}_1\cdot\mathbf{r})/r^d$.

When canting occurs, the entire spin system is involved in dipole distortions centered about two bonds. The effective interaction between dipoles centered at the defect bonds is long ranged, despite the fact that the true interaction (between all spins, not just those associated with the defect bonds) is short ranged. Note that the spin "gauge" is decoupled from the spatial coordinates. In particular, the dipolar interactions of (17) and (20) [which enter c of (21)] involve vectors in real space, whereas the vectors $\mathbf{M}_{1\perp}, \mathbf{M}_{2\perp}$ of (21) are in spin space. Thus (21) is a specific realization of a system that is isotropic in spin space, with an interaction exchange constant c which has a dipolar form (in real space).

For the above problems, the energy scale near the threshold for canting is much lower than that for the or-

dering of the ferromagnetic background. Thus, when thermal effects are included, and one starts at very high temperatures, the ferromagnetic order will set in first. Only at a much lower temperature will there be an onset of spin canting, due to the interaction effects discussed above.[8] Thus, at elevated temperatures the system might appear to be a perfectly ordinary ferromagnet, with no hint of the more complex behavior to occur at much lower temperatures. Most likely, for a system containing a finite concentration of defects, the thermodynamic signature will be proportional to the defect concentration. We make no predictions as to the nature of this thermodynamic signature.

In the noncollinear phase there should be a collective mode of spin oscillation similar to the longitudinal mode of spin glasses;[9] however, this mode should not be tunable with an applied field, and thus will be difficult to observe by electron spin resonance. A macroscopic theory for this mode, in the presence of weak anisotropy, has already been developed.[10]

The generalization of (21) to a system with many identical, well-separated interacting defects is

$$E=\sum_i (\tfrac{1}{2}a\mathbf{M}_{i\perp}^2+\tfrac{1}{4}b\mathbf{M}_{i\perp}^4)+\tfrac{1}{2}\sum_{\langle i,j\rangle} c_{ij}\mathbf{M}_i\cdot\mathbf{M}_j\ , \qquad (22)$$

where c_{ij} is proportional to (17) or (20), as is appropriate, and the sum runs only over the defect sites. This neglects fluctuation energies associated with the ferromagnetic host, in favor of the exchange energies directly or indirectly due to the defect bonds (which cause a moment $\mathbf{M}_{1\perp}$ at site 1, mediated by the ferromagnetic host, to interact with a moment $\mathbf{M}_{\perp 2}$ at site 2). With more than two negative defect bonds present, it becomes possible for the system to go unstable with negative defect bonds that are more positive than given by the constraints following Eqs. (12) and (19), due to the collective interactions that can occur. Hence, a higher density of less negative defect bonds can cause canting to occur.

These results are clearly relevant to the problem of magnetic doping, which may introduce antiferromagnetic bonds. Perhaps of even more interest, they are relevant to the problem of magnetic order in magnetic alloys, where the nonferromagnetic dopant has a significant concentration. Moreover, the possibility of a canted state then leads to the case where the antiferromagnetic bonds have an even more extreme effect, and the system shows "reentrant" spin-glass behavior at low temperatures, or perhaps spin-glass behavior with no ferromagnetic-like regime at all.[8,11,12]

In closing, we note that the above considerations may have relevance to systems where J' is mediated by mobile impurities (electrons or holes). If the J' due to an individual impurity is insufficient to cause canting, one has the possibility of the impurities hopping about and, when they are close enough, for the effective interaction I [cf. (9)] to be strong enough to induce canting. This would lead to a dynamical magnetic structure. The case where J' alone can induce canting is perhaps relevant to the high-temperature superconductors,[13] where hopping may induce a dynamical J', to which the spin system must constantly adjust. In that case, the ratio of the rate of hopping to the rate of spin adjustment becomes a parameter that determines the physics of the problem: if the ratio is small, then the entities that hop are subject to effective interactions due to the adjustments of the spin system; if the ratio is large, the spin system is subject to rapid fluctuations in the exchange Hamiltonian, which could lead, among other things, to additional line broadening—or perhaps motional narrowing—in various types of spectra.

Note added in proof. After submitting our paper we received a preprint by J. Vannimenus, F. D. M. Haldane, C. Jayaprakash, and S. Kirkpatrick, *Ground State Morphology of Random Frustrated XY Systems,* also dealing with the onset of canting due to defect.

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[12]The expression "reentrant" is a misnomer, since no thermodynamic phase is reentered; rather, on lowering the temperature one passes from one phase with no spontaneous magnetization (paramagnet) to another phase with no spontaneous magnetization (spin glass), by way of a disordered ferromagnetic (perhaps more properly called ferrimagnetic) state, and canted disordered ferromagnetic (perhaps ferrimagnetic) state. A general reference on spin glasses is K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1987). It can be argued that the "transition" from the canted disordered ferromagnetic state is not a transition at all [see Ref. 7 and W. M. Saslow and G. N. Parker, Phys. Rev. Lett. **56**, 1074 (1986)]. Rather, the behavior in that case as the temperature is lowered is more akin to what happens in ^{3}He-A when a magnetic field is applied: for small fields one does not think of the system has having its symmetry changed.

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