Spin canting and reentrance

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(Received 13 June 1984)

A simple, non-mean-field explanation of spin canting and reentrance is presented and applied to spin-glasses. It is shown that the competition between ferromagnetic short-range order and a long-range interaction of the Ruderman-Kittel-Kasuya-Yosida (RKKY) type is responsible for the canting transition.

Ferromagnetism on a site-diluted lattice with ferromagnetic nearest-neighbor interactions is possible only if[1] the concentration c of the magnetic moments (spins) exceeds the percolation threshold c_p. If $c > c_p$, one finds a paramagnetic-to-ferromagnetic phase transition at a critical temperature T_c. Metallic spin-glasses, such as *Au*Fe, have not only a ferromagnetic short-range interaction[2] but also a long-range, Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction with randomly oscillating sign, and they exhibit[1,3] a second transition to a spin-glass state at a still lower temperature T_f. This transition is commonly called reentrance. Below T_f spin canting sets in, i.e., the spins rotate away from a certain axis determined by, say, the external field[4] and they acquire static transverse-moment components. Finally, at a still lower temperature T', torque experiments[5] reveal a strong irreversibility in the magnetization process, signalling the occurrence of many metastable states.

Up to a certain extent, these phenomena can be explained by means of a mean-field theory,[6] which predicts a series of transitions agreeing roughly with the ones indicated above. However, it is difficult to decide unequivocally which experimental feature is to be identified with a specific model transition. Moreover, one might wonder whether the transitions persist if the interactions are *not* infinite range. Within the context of equilibrium statistical mechanics[7] we present in this paper a simple physical argument that proves the possibility of both reentrance and spin canting for realistic interactions.

The relevant interactions in a metallic spin-glass consist of a fairly strong, ferromagnetic short-range part (whose influence increases with c) and an RKKY-type interaction which is weaker, but has a much longer range. The conduction electrons of the metallic host, which have a finite mean free path R, mediate the RKKY interaction. Though R greatly exceeds the average minimal distance between two spins, it places an upper bound on the range of the interaction.

To simplify the discussion we start by assuming that the spins are on a regular, hypercubic lattice in $d=3$ dimensions, and take the XY Hamiltonian

$$H=-J_0\sum_{i\neq j}\vec{S}(i)\cdot\vec{S}(j)+2\epsilon\sum_{\substack{i\neq j\\ |i-j|\leq R}}\frac{1}{|i-j|^{d+\sigma}}\vec{S}(i)\cdot\vec{S}(j)\ , \tag{1}$$

where the $\vec{S}(i)$ are classical two-component spins. The first sum in (1) is over nearest neighbors. The second, which contains *anti*ferromagnetic terms only, models the long-range part of the interaction and gives rise to frustration.[8] As yet there is no randomness. We can write $\vec{S}(i)\cdot\vec{S}(j)=\cos(\theta_i-\theta_j)$. By assumption, $0<\epsilon<<J_0$.

There is a stable ferromagnetic phase with a critical temperature T_c for $\epsilon=0$. It persists for ϵ small and T just below $T_c(\epsilon)$. We now show that the ferromagnet becomes unstable as we lower T. More precisely, for T low enough there is a "canted" state with a lower free energy.

According to the variational principle[9] an equilibrium state ρ *minimizes* the free energy $u(\rho)-Ts(\rho)$ where $u(\rho)$ and $s(\rho)$ are the energy and entropy per spin, respectively. If we show that there is another state $\tilde{\rho}$ with $s(\rho)=s(\tilde{\rho})$ but $u(\rho)>u(\tilde{\rho})$, then ρ cannot be an equilibrium state anymore. We write $\langle\mathcal{O}\rangle_\rho=\mathrm{Tr}(\rho\,\mathcal{O})$ for any observable $\mathcal{O}$; in the classical case the trace is replaced by an integral. Though our notation suggests a large but finite system, we implicitly assume that the infinite-volume limit has already been taken.[7]

Suppose it were possible to have a *ferromagnetic* equilibrium state ρ which was stable down to *low* T. Every equilibrium state may be decomposed uniquely into its ergodic components,[7] so we may assume that ρ itself is ergodic and, hence,

$$\lim_{|i-j|\to\infty}\langle\vec{S}(i)\cdot\vec{S}(j)\rangle_\rho=\lim_{|i-j|\to\infty}\langle\vec{S}(i)\rangle_\rho\cdot\langle\vec{S}(j)\rangle_\rho=m^2\ . \tag{2}$$

Since ρ is ferromagnetic, the spontaneous magnetization m is strictly greater than zero for $T<T_c$ and increases as T is lowered. Plainly,

$$\langle\vec{S}(i)\cdot\vec{S}(j)\rangle_\rho>\tfrac{1}{2}m^2,\ \text{ if } |i-j|>k(\epsilon)\ . \tag{3}$$

To simplify the ensuing formulae it is assumed that $k(\epsilon)=1$; this is not a serious restriction.

The angle θ_i characterizes the spin at i. Let $R_M^{\pm}\theta_i=\theta_i\pm i_1(\pi/M)$ for $i=(i_1,\ldots,i_d)$ and M a positive integer. The operation R_M rotates all the spins in a layer perpendicular to the 1 axis through π/M with respect to the previous layer. Whereas the state ρ itself is translationally invariant, the *canted* state $R_M\rho$ defined by $R_M\rho(\mathcal{O})=\langle R_M\mathcal{O}\rangle_\rho$ for any observable $\mathcal{O}$, is periodic with period $2M$ in the 1 direction and has no spontaneous magnetization. Accordingly,

$$\tilde{\rho}=\tfrac{1}{2}(R_M^{+}\rho+R_M^{-}\rho) \tag{4}$$

has no spontaneous magnetization either. It has a periodic long-range order of period $2M$. The quantity M, which is still at our disposal, may be thought of as a typical *cluster size.*

Since the entropy is affine,[10]

$$s(\tilde{\rho})=\tfrac{1}{2}s(R_M^+\rho)+\tfrac{1}{2}s(R_M^-\rho)=s(\rho) \ .$$

Here we also utilize the fact that R_M is induced by a unitary transformation, which leaves the entropy invariant. Turning to the energy, we use the formula

$$\tfrac{1}{2}[\cos(x+y)+\cos(x-y)]=\cos(x)\cos(y)$$

and find, with $\tilde{\rho}$ on the left and ρ on the right,

$$\langle \vec{S}(i)\cdot\vec{S}(j)\rangle_{\tilde{\rho}}=\langle \vec{S}(i)\cdot\vec{S}(j)\rangle_{\rho}\cos[(i_1-j_1)\pi/M] \ . \quad (5)$$

In passing we note that precisely the same formula holds in the quantum case. Taking advantage of (5) we now compare $u(\tilde{\rho})$ with $u(\rho)$ and show that $u(\tilde{\rho})<u(\rho)$ for suitable M and T low enough.

In the ferromagnetic state ρ the mean energy $u(\rho)$ is in first approximation given by

$$u(\rho)=-zJ_0U+\epsilon m^2\sum_{|j|\leq R}{}' |j|^{-(d+\sigma)} \ , \quad (6)$$

where $U=\langle \vec{S}(i)\cdot\vec{S}(j)\rangle_{\rho}$ is the interaction energy between nearest neighbors (there are z of them) and the primed sum indicates that $j=0$ has to be excluded. There is a $U_0>0$ such that $U\geq U_0$ for $T\leq 2T_c$ because ρ is ferromagnetic.[11] On the other hand, in the canted state we get

$$u(\tilde{\rho})=-zJ_0U\cos(\pi/M)+\epsilon m^2\sum_{|j|\leq R}{}' |j|^{-(d+\sigma)}\cos(\pi j_1/M) \quad (7)$$

so that

$$u(\rho)-u(\tilde{\rho})=-zJ_0U[1-\cos(\pi/M)]+\epsilon m^2\sum_{|j|\leq R}{}' |j|^{-(d+\sigma)}[1-\cos(\pi j_1/M)] \ . \quad (8)$$

The first term in the right-hand side of (8) is negative for all M. The second is positive. Their sum may have either sign; the sign depends on M and $m=m(T)$. We now exploit our freedom to vary T and choose M suitably.

There are two interesting cases, $0<\sigma<2$ and $\sigma\leq 0$, which we consider in turn. We first take $0<\sigma<2$ and suppose $M>>1$. Then the first term may be written $-(\tfrac{1}{2}zJ_0U\pi^2)M^{-2}\equiv -aJ_0UM^{-2}$. Since M is large, the summation over j in the second term may be replaced by a d-dimensional integral. Making the change of variables $\vec{x}\rightarrow M\vec{x}$ we then find the dependence upon M,

$$M^{-\sigma}\int_{|\vec{x}|\leq R/M} d\vec{x}\,|\vec{x}|^{-(d+\sigma)}[1-\cos(\pi x_1)] \equiv M^{-\sigma}\phi_\sigma(R/M) \ . \quad (9)$$

The function $\phi_\sigma(R/M)$ *in*creases from zero as M decreases from infinity, and approaches a finite value b for $M\lesssim R$, say. And this is precisely the range of M we are interested in. The point is that as $(R/M)\rightarrow 0$, the function $\phi_\sigma(R/M)$ decreases as $(R/M)^{2-\sigma}$, so that the last term in (8) behaves like $(\epsilon m^2R^{2-\sigma})M^{-2}$, i.e., like the first term —except for the sign. For large M we want the ferromagnetic phase to dominate and, therefore,

$$zJ_0U\gtrsim \epsilon m^2R^{2-\sigma} \ . \quad (10)$$

The antiferromagnetic interaction was required to be "weak" and we now have made this more precise.[12] Just below T_c the ferromagnetic phase dominates. Hence, $M\lesssim R$.

Collecting terms we obtain

$$u(\rho)-u(\tilde{\rho})\approx -aJ_0UM^{-2}+\epsilon m^2bM^{-\sigma} \ , \quad (11)$$

where a and b are geometrical constants independent of M. We maximize the right side of (11) with respect to M so as to find a positive maximum $(\sigma<2)$ provided

$$m^2=(2aJ_0U/\sigma b\epsilon)M^{\sigma-2} \Rightarrow M\sim (J_0U/\epsilon m^2)^{1/(2-\sigma)} \ . \quad (12)$$

Since $M\lesssim R$, condition (12) cannot be realized if $m(T)$ is too small, i.e., just below T_c. However, $m(T)$ increases as T decreases and at a certain $T=T_f<T_c$ condition (12) can be realized so that $u(\rho)-u(\tilde{\rho})\geq 0$. A canted state with period approximately R appears and we have reentrance. Surprisingly, the cluster size M *de*creases as the temperature is lowered. This argument might provide an explanation of the mechanism postulated by Malozemoff, Barnes, and Barbara[13] who assume that the cluster size diverges as T approaches T_f *from below.* If at low temperatures the period M becomes comparable with the distance between the spins and the assumption $M>>1$ breaks down, then we expect many equivalent, low-lying states separated by relatively small energy barriers and, thus, a crossover to irreversibility. There is no reason for yet another phase transition. One might argue that at low T also the classical approximation breaks down and one has to take quantum spins instead. However, the whole setup, in particular Eqs. (5)–(11), needs no modification for quantum spins.

Including a magnetic field along a certain direction in the xy plane does not change the argument either. The field stabilizes the ferromagnetic phase through a negative, constant term in the right-hand side of (11) but, as the temperature is lowered, the total energy difference still can become positive, i.e., canting is advantageous.

Reasoning much as before, one easily verifies that for $\sigma\leq 0$ the right side of (11) is maximized if $M\approx R$, whatever $m(T)$ and, thus, T. Depending on ϵ we may get a reentrance, but below T_f no T-dependent canting.

For Heisenberg spins in not too strong an external field one may use the above arguments down to T_f. Without anisotropy the system then will undergo a transition into a "spin-flop" state with the spins perpendicular to the external field, except for a small longitudinal component. Below T_f, the transverse components are frozen in a canted state whose period M is still determined by (12). Anisotropy[14] is expected to modify the picture, however. Indeed, this may be illustrated by the rich variety of phenomena which occur in heavy rare-earth metals.[15] Note, however, that in these metals the ferromagnetic phase is stable only at *low* temperatures and that the present theory deviates rather strongly from the traditional type of argument, whether spin-wave or mean-field, in that the short- and long-range interactions are treated on a different footing.[11]

In a spin-glass we have *site* disorder and, therefore, the gist of our argument still applies. To see this, replace $\vec{S}(i)$ in (1) by $\xi_i\vec{S}(i)$ where $\xi_i=1$ or 0 according to whether the site i is occupied or not. Arguing as before and applying the ergodic theorem[16] one finds that $u(\rho)-u(\tilde{\rho})$, a *spatial* average, is still given by the right-hand side of (8) provided one makes the substitutions $J_0\rightarrow cJ_0$ and $\epsilon\rightarrow c\epsilon$. The point is that we want to show that the ferromagnet is unstable at low temperatures. In fact, the precise form of the short-range interaction is immaterial. The only requirement is that it is mainly ferromagnetic and percolates through the lattice. This type of interaction certainly favors short-range ferromagnetic order[1,2] as it indeed occurs in spin-glasses. In our model, the long-range RKKY interaction $(\sigma \geqslant 0)$ breaks the long-range ferromagnetic order at $T_f < T_c$ through canting, but, owing to the disorder, the system now decomposes into clusters with different canting period. Again we must expect an equation of the form (11) to compare the energies of the ferromagnetic and the canted, spin-glass state. The physics behind this is a simple energy argument: *canting respects the short-range order,* which is distorted at low cost of energy, while energy is gained via a long-range, RKKY-type interaction. Accordingly, we expect spin waves, i.e., delocalized elementary excitations, to persist below T_f. Recent neutron scattering measurements[17] are consistent with this observation. If, at very low temperature, the canting period becomes comparable with the distance between the spins, our argument suggests the occurrence of many metastable states and, hence, a crossover to irreversibility. A more detailed explanation of the canting structures would critically depend on the nature of the anisotropy, which is not very well understood yet.

The relevance of the interplay between ferromagnetic short-range order, and long-range interactions has been confirmed by Cable, Werner, Felcher, and Wakabayashi[18] who showed the presence of a long-period spin modulation in *Cu*Mn, with concentration dependent period M. Fixing the temperature and increasing the concentration c, they found that M decreased with c, in agreement with (12). Here, U is of the order unity, and J_0 and ϵ scale with c. However, the system gets relatively colder as we increase c so that m increases also. Hence, we get a decrease in M.

Summarizing, we have argued that the competition between ferromagnetic, short-range order, and a long-range interaction of the RKKY type is responsible for the canting transition. In fact, canting does not occur in short-range spin-glasses such as (EuSr)S. Moreover, the present argument allows a conceptually simple, physical explanation of the phenomena related to spin canting and reentrance.

The authors thank Professor K. Binder for a critical reading of the manuscript. Part of this work has been supported by the Deutsche Forschungsgemeinschaft (SFB 123).

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[10]This means that the entropy $s(\rho_{av})$ of an averaged state ρ_{av}, here $\rho_{av}=\frac{1}{2}R_M^+\rho+\frac{1}{2}R_M^-\rho$, equals the average of the entropies of the ergodic constituents of ρ_{av}, here $R_M^+\rho$ and $R_M^-\rho$. The affine property only holds in the thermodynamic limit. It allows one to treat the entropy like other thermodynamic observables such as the energy and the magnetization. For example, $u(\rho_{av}) =\frac{1}{2}u(R_M^+\rho)+\frac{1}{2}u(R_M^-\rho)$. See also A. Wehrl, Rev. Mod. Phys. **50**, 221 (1978), in particular, Sec. II. F. 4 and Eqs. (2.1)–(2.3), or R. B. Israel, Ref. 9, p. 42.

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