Magnetic susceptibility of an antiferromagnet with frustrated bonds

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A model is proposed for an antiferromagnet with frustrations which has undergone a reversible transition into a spin-glass phase. The magnetic susceptibility is calculated for both strong and weak single-ion anisotropies. The frustrated bonds lead to an unusual temperature dependence of the longitudinal and transverse susceptibilities in the antiferromagnetic phase.

The introduction of a spatial disorder in a magnetic crystal frequently gives rise to competing exchange interactions (frustrated bonds) between spins. In a ferromagnet, for example, antiferromagnetic bonds appear, while in an antiferromagnet ferromagnetic bonds appear between spins in different sublattices. While frustrated ferromagnets have been studied quite extensively, both experimentally and theoretically,[1] the research on antiferromagnets with frustrations is in its infancy. Nevertheless, several systems of this type have been identified[2–7]; measurements of the magnetic susceptibility have revealed several anomalies in these substances. In particular, the transverse susceptibility increases quite rapidly with decreasing temperature in the antiferromagnetic phase[4–6]; it does not remain constant or nearly constant as it does in antiferromagnetic crystals with a slight magnetic anisotropy.

In this letter we offer a model for studying the properties of frustrated antiferromagnets and the so-called reversible transition from an antiferromagnetic phase to a spin glass. We show that the presence of frustrated bonds leads to an anomalous temperature dependence of the longitudinal and transverse susceptibilities; in particular, the experimental results of Ref. 4 are explained.

The simplest model system which describes the properties of frustrated antiferromagnets consists of two subsystems of spins with an exchange interaction only between the spins of the different subsystems, S_{i1} and S_{j2} (i, j are site indices), so that the Hamiltonian of the system is

$$\mathcal{H} = \sum_{i,j} J_{ij} \mathbf{S}_{i1} \mathbf{S}_{j2} - D \sum_{\substack{i \\ p=1,2}} (S_{ip}^z)^2 - \sum_{\substack{i \\ p=1,2}} \mathbf{H}\mathbf{S}_{ip}, \tag{1}$$

where $D > 0$ in the single-ion anisotropy constant, and H is the external magnetic field. The exchange interaction energies J_{ij} are assumed (as in the Sherrington-Kirkpatrick "single-sublattice" model[8]) to be independent of i and j and to be distributed normally with a probability density

$$P(J_{ij}) = \frac{N^{1/2}}{\sqrt{2\pi J}} \exp\left(-\frac{(J_{ij} - J_0/N)^2}{2J^2} N\right), \tag{2}$$

and the spins S are assumed to be classical vectors with a length $S=\sqrt{3}$. Here N is the total number of spins in each subsystem.

The free energy can be calculated in our model, following Refs. 8 and 9, by a method of replicas; in the antiferromagnetic phase, we can clearly use the replica-symmetric approximation. As a result, we find the following system of equations for the magnetizations of the sublattices, m_p^μ ($p=1,2$ is the sublattice index; $\mu = 2, y, z$), the Edwards-Anderson parameters q_p^μ, and the quadrupole parameters Q_p^μ:

$$m_p^\mu = \langle\langle S_p^\mu \rangle\rangle_c, \qquad q_p^\mu = \langle\langle S_p^\mu \rangle^2 \rangle_c, \qquad Q_p^\mu = \langle\langle (S_p^\mu)^2 \rangle\rangle_c. \tag{3}$$

Here $\langle\ \rangle$ means a thermodynamic average, calculated in the following way:

$$\langle f(\mathbf{S}_p)\rangle = L^{-1}\int \frac{d\Omega}{4\pi} f(\mathbf{S})\exp\left(\sum_\mu a_k^\mu S^\mu + \beta D(S^z)^2 + \beta\sum_\mu b_k^\mu (S^\mu)^2\right), \tag{4}$$

$$a_k^\mu = \beta(H^\mu - J_0 m_k^\mu + J\sqrt{q_k^\mu}\, z^\mu),$$
$$b_k^\mu = \frac{1}{2}(\beta J)^2 (Q_k^\mu - q_k^\mu),\ \beta = 1/T,\ k \neq p. \tag{5}$$

The partition function L is determined by the same integral as in (4), but without $f(\mathbf{S})$. The integration in (4) is carried out over the solid angle in the space of the vector $\mathbf{S}$. The spatial average $\langle\ \rangle_c$ in (3) reduces to an integration over z^μ with a Gaussian distribution.

It follows from (3) that in a slightly anisotropic ($D \ll J_0$) magnetic material an antiferromagnetic phase arises if $\tilde{J} = J/J_0 < 1$; the Néel temperature at $H=0$ is $T_N = J_0$. Near the Néel point, with $\tau = (T_N - T)/T_N \ll 1$, the parameters m_p^μ and q_p^μ are small, and Eqs. (3) can be expanded in powers of these parameters. Differentiating with respect to H, we then find the longitudinal ($\chi_\parallel$) and transverse ($\chi_\perp$) susceptibilities in a zero field:

$$\chi_{\parallel,\perp} = (2J_0)^{-1}(1 + B_{\parallel,\perp}\tau) \tag{6}$$

$$B_\parallel = \frac{(1-\tilde{J}^2)(-18\tilde{J}^6 - 3\tilde{J}^4 + 70\tilde{J}^2 - 25)}{(1+\tilde{J}^2)(5+3\tilde{J}^2)(5+11\tilde{J}^2-6\tilde{J}^4)}, \tag{7}$$

$$B_\perp = \frac{\tilde{J}^2(8-3\tilde{J}^2)}{5+11\tilde{J}^2-6\tilde{J}^4}. \tag{8}$$

The second set of parentheses in the numerator in (7) and, along with it, the entire coefficient $B_\parallel$, changes sign at $\tilde{J}^2 = 0.37$. Consequently, if $1 > \tilde{J}^2 > 0.37$, i.e., if the number of frustrated bonds is not too small, then $\chi_\parallel$, after undergoing a slope change at the Néel point, increases with decreasing temperature deep in the antiferromagnetic phase, rather than decreasing, as in ordered antiferromagnets. This increase is slower than in the paramagnetic phase. If $\tilde{J}^2 < 0.37$, on the other hand, then $\chi_{\parallel}$ decreases with increasing τ, in the usual way.

We see from (8) that the frustrations always lead to an increase in $\chi_{\perp}$ with increasing τ in slightly anisotropic antiferromagnets, while $\chi_{\perp}$ in crystals is essentially independent of the temperature in the antiferromagnetic phase.

In highly anisotropic ($D \gg J_0$) systems, there is an antiferromagnetic phase if $\tilde{J} < 1$, and the Néel temperature is $T_N = 3J_0$. From (3) we find the following expressions for the susceptibilities:

$$\chi_{\parallel} = \frac{1}{2J_0}\left(1 + \tau \frac{(J_0^2 - J^2)(2J^2 - 9J_0^2)}{(J_0^2 + J^2)(2J^2 + 9J_0^2)}\right), \tag{9}$$

$$\chi_{\perp} = \frac{1}{2D}\left(1 - \tau \frac{J_0}{2D} \frac{3J_0^2 - J^2}{3J_0^2 + 2J^2}\right). \tag{10}$$

It follows from (9) that, as in the case of a slight anisotropy, the frustrations can lead to an increase in $\chi_{\parallel}$ with τ; a necessary condition here is $J_0 > J > (1/\sqrt{2})J_0$. This temperature dependence of $\chi_{\perp}$, despite the frustrations, is qualitatively the same as that in highly anisotropic ordered antiferromagnets.

The increase in $\chi_{\perp}$ with decreasing temperature observed in Ref. 4 in the slightly anisotropic antiferromagnet $Eu_xSr_{1-x}As_3$ agrees with the results of the present study. It should be expected that as x is reduced, and the triple point in the (T,x) phase diagram is approached, we will see a similar behavior of $\chi_{\parallel}$.

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