Disordered antiferromagnets with magnetic-atom concentration near the percolation limit

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An isotropic Heisenberg antiferromagnet diluted by nonmagnetic impurities is considered; the concentration of magnetic atoms is assumed to be close to the percolation limit. The density of states of the magnetic excitations, the temperature dependences of the sublattice magnetizations and specific heat, and also the concentration dependence of the Néel temperature, are obtained using scaling theory for the percolation problem.

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1. INTRODUCTION

Recently, there have appeared a number of experimental papers[1] investigating ferri- and antiferromagnets diluted by a sufficiently large quantity of nonmagnetic impurities, distributed randomly in the lattice and taking the place of magnetic atoms. The most interesting property of such alloys is the existence in them of a concentration phase transition: macroscopic magnetic order arises only when the concentration x of magnetic atoms is greater than a certain threshold value x_c, while if $x<x_c$ a magnetically ordered state does not arise even at zero temperature $T=0$. This concentration phase transition is considered in percolation theory.[2,3] The point is that macroscopic magnetic order arises only when the interacting atoms (we shall call them connected) form an infinite cluster. In magnetic insulators, as is well known, it may be assumed that only nearest neighbors in the lattice are connected. Naturally, for small x the connected atoms form only finite clusters, and only at sufficiently high concentrations $x>x_c$ (x_c is called the percolation limit) does an infinite cluster arise.

The concentration phase transition has turned out to be similar in many respects to ordinary second-order phase transitions.[3] In particular, as the critical point is approached the role of spatial fluctuations in the clusters increases and so the geometrical properties of an infinite cluster cannot be understood with the aid of some or other variant of the self-consistent field method. This makes the question of the magnetic properties of such substances especially interesting.

The thermodynamics of ferromagnets with $|x-x_c|/x_c \ll 1$ was investigated theoretically in Refs. 4-6. In view of the fact that all the experiments are carried out on ferri- and antiferromagnets,[1] in the present paper the existing theory[4-6] is generalized to antiferromagnets. The dependence of the density of states on the energy, the temperature dependences of the sublattice magnetizations and specific heat, and also the dependence of the Néel temperature on $x-x_c$, are obtained. It is interesting that the latter dependence turns out to be the same as the dependence of the Curie temperature of a dilute ferromagnet on $x-x_c$.[4,5] Thus, the threshold behavior of the phase-transition temperature is determined not by the type of magnetic order but solely by the symmetry of the Heisenberg Hamiltonian—a fact that is natural from the standpoint of the modern theory of phase transitions.

2. THE ELEMENTARY EXCITATIONS AT ZERO TEMPERATURE

The Hamiltonian of an isotropic Heisenberg antiferromagnet with interaction only between the sublattices has the form

$$\mathcal{H} = -\frac{1}{2}\sum_{i,j} V_{i_1 j_2} \mathbf{S}_{i_1} \mathbf{S}_{j_2}. \tag{1}$$

The subscripts 1 and 2 label the sublattices. The summation is taken only over the sites occupied by magnetic atoms. As already noted, for $x>x_c$ there is an infinite cluster, in which magnetic order is established. At zero temperature, $S^z_{i1}=S$ and $S^z_{i2}=-S$.

The low-frequency excitations in the infinite cluster are long-wavelength, weakly damped spin waves. The small damping of these waves is due to the fact that the Heisenberg Hamiltonian conserves the total spin of the system (for more detail, see the article by Harris and Kirkpatrick).[7] Such hydrodynamic excitations can be described phenomenologically by means of the usual expression for the change in the energy E of the system when fluctuations arise in the transverse components of the sublattice magnetizations $\mathbf{m}_1(\mathbf{r})$ and $\mathbf{m}_2(\mathbf{r})$:

$$E = \int\left\{\frac{A}{2M_0^2}\sum_{\alpha=x,y} |\nabla m_1^\alpha(\mathbf{r}) \nabla m_2^\alpha(\mathbf{r})| + \frac{H_E}{2M_0}|\mathbf{m}_1(\mathbf{r})+\mathbf{m}_2(\mathbf{r})|^2\right\} d^3r. \tag{2}$$

Here, M_0 is the zero-temperature sublattice magnetization, proportional to the concentration $P(x)$ of the atoms in the infinite cluster: $M_0 \sim P(x) \sim (x-x_c)^\beta$, $\beta=0.35$.[3] As regards the coefficients A and H_E, they are studied in detail in the paper by Harris and Kirkpatrick.[7]

The microscopic analysis that they made of the equations of motion for the spin operators showed that the coefficient A is proportional to the conductivity σ of a network of resistances in which the conductance between sites i and j is equal to $\sigma_{ij}=V_{ij}$. The concentration dependence of the conductivity of such a network is now well known in the entire range of concentrations. In particular, near the percolation limit,[3]

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$A \sim \sigma \sim (x-x_c)^t$, $t=1.7$.

The situation with regard to the coefficient H_E, which determines the transverse susceptibility of the antiferromagnet,

$$\chi_\perp = M_0/H_E, \tag{3}$$

is more complicated. For us, the result obtained by numerical calculations on a computer will be sufficient[7]:

$$\chi_\perp \sim (x-x_c)^{-\tau}, \quad \tau = 0.5 \pm 0.2.$$

As is well known, from (2), with the aid of the Landau-Lifshitz equations, we can obtain the linear dispersion law $\omega = C_q$ for the spin waves, their velocity being

$$C = (2A/\chi_\perp)^{1/2}. \tag{4}$$

The density of states $\rho_{inf}(\omega)$ of the magnetic excitations in the infinite cluster at low frequencies is determined by the spin waves and is equal to

$$\rho_{inf}(\omega) = \omega^2/\pi^2 C^3. \tag{5}$$

Up to what frequencies is formula (5) valid?

Hydrodynamic excitations are weakly damped so long as their wavelength is greater than the characteristic length determing the scale of the spatial fluctuations in the system. According to scaling theory for the percolation problem, near the threshold the only characteristic length is the correlation length[3]

$$L \sim (x-x_c)^{\nu}, \quad \nu = 0.90 \pm 0.05.$$

Therefore, the hydrodynamic description of the excitations is possible only for

$$\omega < \omega_0 = CL^{-1} \sim (x-x_c)^{(t+\tau)/2+\nu}.$$

The damping of the waves grows with increase of the frequency, and at $\omega \approx \omega_0$ the damping is of the same order as the excitation energy. Therefore, it is natural to assume that excitations with $\omega \gg \omega_0$ are localized within a correlation length; in the theory of phase transitions it is customary to call such excitations the critical mode.

In order to determine the density of states for $\omega \gg \omega_0$, we shall use the procedure for matching the hydrodynamic mode and the critical mode that was proposed by Halperin and Hohenberg[8] for the description of critical dynamics. Here the chief point for us will be the fact that the properties of the critical mode (the dispersion law, the density of states $\rho_1(\omega)$ per magnetic atom, etc.) do not depend on $x - x_c$. This is explained by the fact that the quantity $x - x_c$ determines the connectedness of the system only at distances greater than L. Within a correlation length we already have connectedness. Thus, $\rho_1(\omega) = f(\omega/V_0)$ (V_0 is the exchange integral between nearest neighbors), and, therefore,

$$\rho_{inf}(\omega) = P(x) f(\omega/V_0), \quad \omega \gg \omega_0. \tag{6}$$

Matching (5) and (6) at $\omega \approx \omega_0$, we obtain

$$f(x) = x^z, \quad z = \frac{4\nu - t - \tau - 2\beta}{2\nu + t + \tau}. \tag{7}$$

Using the index values given above, we obtain $z = 0.35$. Thus, the density of states of the critical mode grows with increase of the frequency, but much more slowly than in the spin-wave region.

To describe the dynamics of the excitations in an infinite cluster it is important to know also the coefficient $\Phi(\mathbf{q})$ connecting the energy density e and the Fourier transform $\mathbf{m}_{q1}$ of the fluctuation of the sublattice magnetization:

$$\Phi(q) = e/|\mathbf{m}_{q1}|^2.$$

To find $\Phi(q)$ in the spin-wave region it is necessary to express $\mathbf{m}_{q2}$ in terms of $\mathbf{m}_{q1}$ using the Landau-Lifshitz equations and substitute into (2). We obtain $\Phi(q) = Aq^2/M_0^2$, and, since $q(\omega) = \omega/C$,

$$\Phi(\omega) = \Phi(q(\omega)) = \omega^2 \chi_\perp M_0^{-2}, \quad \omega \ll \omega_0. \tag{8}$$

Carrying out the matching for $\Phi(\omega)$ just as we did for the density of states, we find

$$\Phi(\omega) \sim \left(\frac{\omega}{V_0}\right)^{2-2(\tau+2\beta)/(t+2\nu+\tau)}. \tag{9}$$

It can be seen from (9) that at high frequencies there is some decrease in the power exponent in $\Phi(\omega)$: it becomes equal to 1.7 – 1.5.

We turn now to the density of states $\rho_{fin}(\omega)$ in finite clusters. As the percolation limit is approached there appear finite clusters of all large sizes up to sizes of the order of the correlation length L. According to the scaling theory, the geometrical properties of finite clusters with linear dimensions L are the same as those of regions of size L in an infinite cluster. Therefore,

$$\rho_{fin}(\omega_0) \approx \rho_{inf}(\omega_0). \tag{10}$$

For $\omega \gg \omega_0$ the quantity $\rho_{fin}(\omega)$ should not depend on $x - x_c$. Since it is normalized to $1 - P(x) \approx 1$, we can state that

$$\rho_{fin}(\omega) = \psi(\omega/V_0), \quad \omega \gg \omega_0, \tag{11}$$

and, matching (10) and (11) at $\omega \approx \omega_0$, we obtain

$$\rho_{fin}(\omega) \sim (\omega/V_0)^{z_1}, \quad z_1 = \frac{4\nu - t - \tau}{2\nu + t + \tau} \approx 0.4. \tag{12}$$

It can be seen from (6), (7), and (12) that, as we should expect, at $\omega > \omega_0$ the density of states in finite clusters is higher than that in an infinite cluster. At low frequencies $\omega \ll \omega_0$ the situation is reversed. Excitations with $\omega \ll \omega_0$ exist only in those finite clusters whose sizes are greater than L. But the number of such clusters is exponentially small[3]; therefore, $\rho_{fin}(\omega)$ is also exponentially small for $\omega \ll \omega_0$, and is considerably smaller than the spin-wave density of states (5).

3. THE THERMODYNAMIC QUANTITIES

The results obtained in the preceding section are valid not only at zero temperature but also for $T \ll T_N$ (T_N is the Néel temperature), when the deviation of the sublattice magnetizations $\langle M \rangle$ from their value at saturation is small, i.e., when $\delta M = M_0 - \langle M \rangle \ll M_0$. Assuming this condition to be fulfilled, we can calculate δM in the usual way:

$$\frac{\delta M}{M_0} = \frac{1}{2M_0^2}\langle |\mathbf{m}_1|^2 \rangle = \frac{1}{2M_0^2}\left\langle \frac{e}{\Phi(q)} \right\rangle = \frac{1}{2M_0^2}\int \frac{\omega \rho_{inf}(\omega)}{(e^{\omega/T}-1)\Phi(\omega)} d\omega. \quad (13)$$

If $T \ll \omega_0$ the integral in (13) is determined by frequencies $\omega \sim T$, and, therefore, δM obeys the usual spin-wave law

$$\delta M/M_0 \sim T^2/AC.$$

It is interesting that even for $T \sim \omega_0$, when all the spin waves are excited, the quantity $\delta M/M_0$ is still small:

$$\left.\frac{\delta M}{M_0}\right|_{T\sim\omega_0} \sim (x-x_c)^{(\nu-t+\tau)/2} \ll 1; \quad (14)$$

this is a consequence of the small number of states in the spin-wave part of the spectrum.

It follows from (14) that, parametrically in the parameter $x - x_c$, the Néel temperature is greater than ω_0. We shall calculate δM in the temperature range $\omega_0 \ll T \ll T_N$. For this we draw attention to the fact that the function $\omega\rho_{inf}(\omega)/\Phi(\omega)$ in the integrand in (13) reaches a maximum at $\omega \sim \omega_0$. This leads to the result that the integral in (13) is determined for $T \gg \omega_0$ by frequencies of the order of ω_0, and we obtain

$$\delta M/M_0 \sim T/(x-x_c)^{t-\nu}. \quad (15)$$

Since at temperatures of the order of the Néel temperature the quantity $\delta M/M_0$ is of order unity, from (15) we obtain

$$T_N \sim V_0 (x-x_c)^{t-\nu}. \quad (16)$$

Since $T_N \gg \omega_0$, the temperature region in which δM depends linearly on T is much broader than the spin-wave region.

The last two results—the concentration dependence (16) for the phase-transition temperature and the existence of a broad temperature region in which the order parameter depends linearly on the temperature—are also valid for dilute ferromagnets[4] and are evidently associated only with the symmetry of the Hamiltonian, and not with the type of magnetic order.

Since the integral (13) for $T \gg \omega_0$ is determined by frequencies $\sim\omega_0$, this means that the magnetic order is destroyed by fluctuations with wavelengths of the order of L. Over lengths shorter than L, short-range magnetic order is preserved even for $T \gg T_N$. For example, in pairs of magnetic atoms the spins are correlated up to temperatures of the order of V_0.[1)]

The specific heat of the magnetic system is

$$C_M = \frac{d}{dT}\int \frac{\rho_{inf}(\omega)+\rho_{fin}(\omega)}{e^{\omega/T}-1}\omega\, d\omega. \quad (17)$$

For $T \ll \omega_0$ the specific heat obeys the spin-wave law $C_M \sim T^3$, while for $T \gg \omega_0$, from (6), (12), and (17), we obtain

$$C_M \sim \left(\frac{T}{V_0}\right)^{1+z_1}. \quad (18)$$

The specific heat (18), like the total density of states for $\omega \gg \omega_0$, is determined by the finite clusters.

The presence of short-range magnetic order in the paramagnetic phase leads to the result that, as can be shown from simple estimates, the formula (18) is valid not only for $T \ll T_N$ but also in the much broader temperature range $\omega_0 \ll T \ll V_0$. The specific heat reaches a maximum at $T \sim V_0$. We have not considered the question of the singularity of the specific heat near the Néel temperature, but, since the contribution of the infinite cluster to the specific heat is small for $T > \omega_0$, it is clear that it is very difficult to distinguish this anomaly experimentally against the background of the specific heat (18).

In a recent paper by Lubensky,[9] the thermodynamics of Heisenberg magnets with $|x - x_c|/x_c \ll 1$ is treated using various heuristic ideas and a hypothesis about the topology of the infinite cluster. The result he obtains[9] for the concentration dependence of the transition temperature coincides with that obtained earlier for ferromagnets[4,5] and with the formula (16) of the present work. However, the result obtained by Lubensky for the temperature dependence of the specific heat is, in our view, incorrect. He uses a model representation of the infinite cluster as a network consisting of one-dimensional parts of length $l \sim |x - x_c|^{-\zeta}$ and assumes that the free energy F depends on the temperature in the following way:

$$F \sim F^+(l/\xi_1(T)), \quad (19)$$

where $\xi_1(T) \sim aV_0/T$ is the correlation length of a one-dimensional Heisenberg chain. Naturally, it only makes sense to introduce $\xi_1(T)$ if $\xi_1(T) < l$, but Lubensky[9] assumes that the free energy also has the form (19) for $\xi_1(T) \gg l$, when the correlation of the spins is in fact determined by the long-wavelength three-dimensional spin waves and not by the excitations within the one-dimensional segments. He does not take into account the spin-wave contribution to the free energy.

We have considered an antiferromagnet with interaction only between atoms on different sublattices. At the same time, the interaction within the sublattices, however small it is, becomes important sufficiently close to the percolation limit. For example, for $x < x_c$ the system can become connected again if the interaction within the sublattices is taken into account. We note that, if the intra-sublattice exchange is antiferromagnetic, then, because of the competition of the interactions within and between the sublattices, near the percolation limit the formation of a state of the spin-glass type is possible.

The results that we have obtained are valid so long as the intra-sublattice exchange energy, and also the magnetic-anisotropy energy, which we have not taken

into account, are smaller than the Néel temperature (16), i.e., our analysis is valid at concentrations not too close to the percolation limit.

[1)]It can be shown that in the infinite cluster there is short-range magnetic order over lengths

$$r \leqslant r(T) = a\left(\frac{V_0}{T}\right)^{\nu/(t-\nu)},$$

where a is the lattice constant.

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Topological instability of singularities at small distances in nematics

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At short distances, the order parameter in a nematic is degenerate on the sphere S^4; this leads to topological removability of the singularities. As a result, the disclinations may have a nonsingular core.

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According to experimental data on measurement of the heat of transition, critical scattering, etc.,[1,2] the transition from isotropic liquid to nematic is a weak transition of the first kind, nearly of the second. Furthermore, the temperature range of existence of the nematic phase is much smaller than the transition temperature. Therefore Landau's theory may be a reasonable approximation for description of a nematic over its whole range of existence.

The order parameter in a nematic is a traceless, symmetric, real second-rank tensor $\hat{Q}=Q_{\alpha\beta}$. In general it has five independent components. This tensor can be represented in the following form:

$$Q_{\alpha\beta}=\sqrt{2}Q_0\{\sin(\varphi+{}^1/_3\pi)(n_\alpha n_\beta-{}^1/_3\delta_{\alpha\beta})+\sin\varphi(l_\alpha l_\beta-{}^1/_3\delta_{\alpha\beta})\}, \quad (1)$$

where Q_0 is the modulus of the order parameter, $\mathrm{Sp}\,\hat{Q}^2=Q_0^2$, and $\mathbf{n}$ and $\mathbf{l}$ are mutually perpendicular unit vectors. The angle φ describes the degree of biaxiality of the tensor $Q_{\alpha\beta}$. When $\varphi=0$, $Q_{\alpha\beta}$ is uniaxial, and $\mathbf{n}$ is the director. The tensor $Q_{\alpha\beta}$ must not change sign upon change of sign of $\mathbf{n}$ or $\mathbf{l}$. Therefore $Q_{\alpha\beta}$ contains no terms of the form $n_\alpha l_\beta$.

There are only two independent invariants of the rotation group constructed from the components of $Q_{\alpha\beta}$, for example $\mathrm{Sp}\hat{Q}^2$ and $\mathrm{Sp}\hat{Q}^3$. Therefore the Landau expansion in powers of Q_0 can be represented in the form[1)]

$$F={}^1/_2A\,\mathrm{Sp}\,\hat{Q}^2-{}^1/_3\sqrt{3}/_2B\,\mathrm{Sp}\,\hat{Q}^3+{}^1/_4C(\mathrm{Sp}\,\hat{Q}^2)^2, \quad (2)$$

where $A=a(T-T^*)$; T^* is a fictitious Curie temperature. On substituting (1) in (2), we get

$$F={}^1/_2AQ_0^2-{}^1/_3B\cos 3\varphi Q_0^3+{}^1/_4CQ_0^4. \quad (3)$$

Minimization of (3) with respect to φ gives $\varphi=0$, provided $B\neq 0$. This means that when $B\neq 0$, only the uniaxial state is stable. If $B=0$, the free energy (2) is a function only of $\mathrm{Sp}\hat{Q}^2$, and the symmetry of the order parameter is higher than in the uniaxial case. The only constraint is

$$\mathrm{Sp}\,\hat{Q}^2=Q_0^2, \quad (4)$$

where Q_0 can be found by minimization of the free energy (3) with $B=0$. It is easy to show that (4) is the equation of a four-dimensional sphere S^4 in the five-dimensional space of the components of the matrix $Q_{\alpha\beta}$.

Now let B be nonzero but small. The biaxial perturbation corresponds to certain motions on the sphere S^4. Such motions have an energy gap $\Delta\sim BQ_0^3$. The corresponding correlation radius is $R_B\sim kB^{-1}Q_0^{-3}$, where k is a quantity of the order of the Frank constants. The criterion for smallness of B is the condition $R_B\gg R_0$, where R_0 is the correlation radius of fluctuations of the modulus Q_0 of the order parameter. In the distance range $R_0<R<R_B$, the order parameter is degenerate on the sphere S^4. When $R>R_B$, the degeneracy parameter will be the ordinary nematic director $\mathbf{n}$. Its domain of variation is the sphere S^2, on which diametrically opposite points are equivalent (since $\mathbf{n}$ and $-\mathbf{n}$ are

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