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Thermodynamics of an impurity uniaxial ferromagnet below the Curie point

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A four-dimensional Ising model and an easy-axis ferromagnet with dipole interaction ($d = 3$), containing randomly distributed pinned impurities, are considered. The renormalization-group methods yields exact equations of states for both systems. The temperature and field dependences of the susceptibility, the magnetization, and the heat capacity are obtained below T_c and in an external magnetic field.

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In systems having randomly disposed pinned impurities, scattering by impurities leads to an additional interaction of the critical fluctuations of the order parameter, their sign being that of attraction. Despite the appearance of the new type of interaction, a second-order phase transition takes place in such systems as before, and scale invariance is present with critical exponents that differ from the critical exponents of the "pure" system.[1–3] There is no doubt that one of the most interesting is the case of impurity systems with single-component order parameter, examples of which are Ising's 4-dimensional impurity model with short-range exchange forces, or a three-dimensional impurity easy-axis ferromagnet (ferroelectric) with dipole interaction.[3–5] The interest in these systems is due to several factors. First, the renormalization-group equations for them can be solved exactly, so that the singularities of the thermodynamic quantities at the phase-transition point can be determined exactly; second, as shown in[5], these singularities (above T_c in a zero external field) contain besides powers of t and $\ln t$ (here $(t = T_c)/T_c$) also the unusual factor $\exp\{-(D|\ln t|)^{1/2}\}$, where D is a certain number (see below). Finally, the conclusions of the theory can be verified experimentally on impurity easy-axis ferromagnets (ferroelectrics), e.g., $LiTbF_4$,[6] with nonmagnetic atoms as impurities.

In this paper we obtain by the renormalization-group method an exact equation of state for impurity systems, with a single-component order parameter, and the temperature and field dependences of the thermodynamic quantities below T_c and in an external field.

To make the exposition clearer, we describe first the general procedure for the analysis of impurity systems (the effective-Hamiltonian method) and the necessary relations obtained for the region above T_c in[5]; this is followed by a derivation of the equation of state in the spirit of the known paper of Larkin and Khmel'nitskiĭ[7] and an examination of its consequences. In the conclusion we discuss the possibility of an experimental verification.

The Hamiltonian of Ising's 4-dimensional impurity model is

$$H\{\Phi(\mathbf{x}), \psi(\mathbf{x})\} = \frac{1}{2}\int d^4x \left\{ r_0\Phi^2 + (\nabla\Phi)^2 + \frac{v_0}{12}\Phi^4 + 2\Phi^2\psi \right\}, \qquad (1)$$

where $\psi(\mathbf{x}) \sim n(\mathbf{x}) - \langle n(\mathbf{x})\rangle$ is a random variable describing the local fluctuations of the temperature in the average-field approximation, $n(\mathbf{x})$ is the impurity density, and r_0 is a linear function of the temperature.

For a given impurity configuration, the free energy is

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equal to

$$F(\psi)=-\ln\int d\Phi\exp\{-H(\Phi,\psi)\}. \tag{2}$$

The free energy observed in experiment is the configuration mean value over the distribution of the impurities:

$$F=\langle F(\psi)\rangle_\psi,\quad \langle\ldots\rangle_\psi=\int d\psi P(\psi)\ldots, \tag{3}$$

where $P(\psi)$ is the impurity distribution function. It is not convenient to average logarithms of a continual integral directly; the logarithm is therefore usually represented in the form[6]

$$\ln\int d\Phi\exp\{-H(\Phi,\psi)\}=\frac{\partial}{\partial n}\left\{\int d\Phi\exp[-H(\Phi,\psi)]\right\}^n\Big|_{n=0}$$

$$=\frac{\partial}{\partial n}\left\{\int\prod_{i=1}^n d\Phi_i\exp\Big[-\sum_{i=1}^n H(\Phi_i,\psi)\Big]\right\}\Big|_{n=0}. \tag{4}$$

Averaging (4) according to (3), we obtain

$$F=-\frac{\partial}{\partial n}\left\{\int d\sigma\exp[-H_{eff}(\sigma)]\right\}\Big|_{n=0}, \tag{5}$$

$$H_{eff}=\int d^4x\left\{\frac{1}{2}r_0\sigma^2+\frac{1}{2}(\nabla\sigma)^2+\frac{v_0}{4!}\sum_{i=1}^n\sigma_i^4\right\}+G(\sigma), \tag{6}$$

where

$$\sigma=\{\Phi_1,\Phi_2,\ldots,\Phi_n\},\quad G(\sigma)=-\ln\left\langle\exp\left\{-\int d^4x\,\sigma^2\psi\right\}\right\rangle. \tag{7}$$

Near the transition point, the fluctuations of the impurity density can be regarded as δ-correlated and Gaussian, and consequently

$$G(\sigma)=\frac{u_0}{4!}\int d^4x(\sigma^2)^2. \tag{8}$$

The non-Gaussian correlations of the impurity density lead to the appearance of interactions of the type $(\sigma^2)^3$, $(\sigma^2)^4$, and higher orders, which play no role in the 4-dimensional problem.

It is easily seen from (6) and (8) that the Ising impurity model is equivalent to an n-component Heisenberg ferromagnet with cubic anisotropy in the limit as the number of components n tends to zero. The Ising vertex v plays the role of the cubic vertex, while the impurity vertex u plays the role of the isotropic vertex, with $u_0<0$ and $v_0>0$. It is quite important that the Gell-Mann–Low equations for the charges u and v turn out in the parquet approximation, in the limit as $n\to 0$, degenerate and have no zero-charge solutions for the physical initial conditions $u_0<0$ and $v_0>0$. This degeneracy is accidental and takes place only in the parquet approximation. To obtain a nondegenerate system of equations it suffices to calculate the Gell-Mann–Low functions accurate to third order in the amplitudes, as was done in fact in[3–5]. Aharony[5] obtained the asymptotic behavior of the charges u and v:

$$u=-\frac{8\pi^2}{|\ln r|^{1/2}}\sqrt{\frac{6}{53}},\quad v=\frac{32\pi^2}{|\ln r|^{1/2}}\sqrt{\frac{6}{53}}, \tag{9}$$

where r is the reciprocal susceptibility. For r there is an equation[7]

$$dr/dt=T(r), \tag{10}$$

where $T(r)$ is a vertex with two ends and one corner; this vertex satisfies, in turn, the equation (in first order in the effective charges)

$$\frac{d\ln T(r)}{d\ln r}=\frac{v+2u}{32\pi^2}. \tag{11}$$

From (9)–(11) we obtain

$$T(r)=a\exp\{-(^6/_{53}|\ln r|)^{1/2}\}, \tag{12}$$

$$r=at\exp\{-(^6/_{53}|\ln t|)^{1/2}\},\quad a=\text{const}. \tag{13}$$

The singular part of the heat capacity is defined in terms of the polarization operator $\Pi(0)$:

$$\frac{\partial\Pi(0)}{\partial\ln r}\sim T^2(r),\quad \Pi(0)\sim C_{sing}\sim|\ln t|^{1/2}\exp\left\{-2\left(\frac{6}{53}|\ln t|\right)^{1/2}\right\}. \tag{14}$$

The appearance of $|\ln r|^{1/2}$ in (9) in place of $|\ln r|$, and of the factor $\exp\{-(\frac{6}{53}|\ln t|)^{1/2}\}$ in (13) and (14), is due to the accidental degeneracy of the parquet equations. In the ε-expansion method, the random degeneracy leads to expansion of the critical exponents i powers of $\varepsilon^{1/2}$.[3–5]

We have referred above to the Ising impurity model. It can be shown[5] that for an impurity easy-axis ferromagnet with dipole interaction all the results remain in force, but the characteristic numerical factor $D=\frac{6}{53}=0.11\,321$ must be replaced by the close quantity $D'=9/(81\ln\frac{3}{4}+53)=0.11\,795$.

We proceed to study the system below T_c and in an external magnetic field. In an external field it is necessary to add to the Hamiltonian (6) the term

$$-h\sum_{i=1}^n\sigma_i.$$

Then, just as below T_c at $h=0$, a macroscopic magnetic moment m, which can be regarded as a parameter and obtained from the condition for the minimum of the free energy:

$$\partial F/\partial m=\langle\partial H/\partial m\rangle=0. \tag{15}$$

In analogy with[7], it can be shown that

$$\partial^2F/\partial m^2=r, \tag{16}$$

$$\partial^2r/\partial m^2=v(r). \tag{17}$$

We call attention to the fact that (17) contains only the Ising vertex $v(r)$, and the impurity vertex $u(r)$ drops out because of the transition to the limit as $n\to 0$ (this circumstance was noted in[3]). As before, r is defined as a function of t by Eq. (10). It is very important that, in analogy with the impurity-free case,[1,7] the dependence of the amplitudes of T and v on r is the same as above the transition point. Indeed, to find, say, the amplitude of $v(r)$ above the transition point it was necessary to sum the graphs of the principal logarithmic approxima-

tion (i.e., all the graphs of the type $(\alpha \ln r)^n$) and the graphs of the approximation that follows the principal one (the graphs of the $\alpha(\alpha \ln r)^n$ approximation). It is easily seen that the additional graphs that arise below the transition point differ from the graphs of the $\alpha(\alpha \ln r)^n$ approximation by the factor $|\ln r|^{-1/2}$, and can therefore be disregarded in the asymptotic region $|\ln r| \gg 1$.

We recall that in the impurity-free Ising model the amplitude of $v(r)$ is obtained by summing only the graphs of the principal logarithmic approximation, and the contribution of the additional graphs are of the same order as those of the $\alpha(\alpha \ln r)^n$ approximation.

Integrating (10), (16), and (17) with logarithmic accuracy (i.e., assuming the logarithms to be constant) and taking (15) into account, we obtain a system of equations for the quantities m and r (the equation of state):

$$r = tT(r) + {}^1/_2 m^2 v(r), \tag{18}$$
$$h = mtT(r) + {}^1/_6 m^3 v(r). \tag{19}$$

An equation of state of exactly the same type was obtained by Larkin and Khmel'nitskiĭ[7] for the impurity-free Ising model, but with other amplitudes $T(r)$ and $v(r)$, which are determined here by formulas (9) and (12).

Solving the system (18) and (19) below the Curie point ($t<0$) in a zero external field, we get

$$r = -2at \exp\{-({}^6/_{53}|\ln|t||)^{1/3}\}, \tag{20}$$
$$m^2 = -\frac{3at}{16\pi^2({}^6/_{53})^{1/3}}|\ln|t||^{1/3} \exp\left\{-\left(\frac{6}{53}|\ln|t||\right)^{1/3}\right\}. \tag{21}$$

Integrating (19) with logarithmic accuracy, we obtain the singular part of the free energy:

$$F_{sing} = \frac{1}{2}tm^2T(r) + \frac{1}{4!}v(r)m^4 \approx$$
$$\approx -\frac{3a^2t^2}{64\pi^2({}^6/_{53})^{1/3}}|\ln|t||^{1/3} \exp\left\{-2\left(\frac{6}{53}|\ln|t||\right)^{1/3}\right\}. \tag{22}$$

Differentiating F_{sing} twice with respect to t, we determine the singularity of the heat capacity:

$$\frac{\partial^2 F_{sing}}{\partial t^2} = C_{sing} = -\frac{3a^2}{32\pi^2({}^6/_{53})^{1/3}}|\ln|t||^{1/3} \exp\left\{-2\left(\frac{6}{53}|\ln|t||\right)^{1/3}\right\}. \tag{23}$$

At the phase-transition point ($t=0$) and in a nonzero external field we get from (18) and (19)

$$r \sim m^2|\ln m^2|^{-1/3}, \quad h \sim m^3|\ln m^2|^{-1/3}. \tag{24}$$

It is seen from (13), (14), (20), and (23) that the temperature dependences of the susceptibility and of the heat capacity are the same above and below the Curie point; this is the result of the asymptotic smallness of the additional graphs that arise below the transition point. The reciprocal susceptibility below the transition point is double the value above the transition point at the same value of $|t|$ ("the rule of two"), and as the transition point is approached it decreases more rapidly than the reciprocal susceptibility of the "pure" substance (for which $r \sim -t|\ln|t||^{-1/3}$).[7] The magnetization of the ordinary Ising model ($d=4$) vanishes like[7]:

$$m^2 \sim -t|\ln|t||^{2/3},$$

i.e., more slowly than for the model with impurities. The heat capacity of the ordinary Ising model diverges like $|\ln|t||^{1/3}$ near the transition,[7] but is finite for the impurity model. The field dependences are also different. For the "pure" substance the denominators of formulas (24) contain $|\ln m^2|$ in lieu of $|\ln m^2|^{1/2}$, as for the substance with impurities.

Experimental observation of the temperature dependences (13), (14), (20), (21), and (23) is of great interest, and in particular that of the slowly varying (compared with the degree) factor $\exp\{-(D|\ln|t||)^{1/2}\}$. Just as in the "pure" substance (in experiments with $LiTbF_4$,[6] we obtained a singularity of the heat capacity $|\ln|t||^{1/2}$), this is best done by measuring the heat capacity and not the susceptibility or the spontaneous magnetization, where the slowly varying factors can hardly be discerned against the background of the power-law dependences. Another possibility is to determine the temperature dependence of the coefficients in the equation of state in a weak external field:

$$h = m[f_0(t) + f_2(t)m^2 + \ldots].$$

In the Landau theory we have in the critical theory $f_2(t) = \text{const}$, and in the Larkin-Khmel'nitskiĭ theory[7] $f_2(t) \sim |\ln|t||^{-1}$. A recent experiment with triglycin sulfate[10] has confirmed the conclusions of[7]. For an impurity uniaxial ferroelectric (ferromagnet) we have according to (9) and (19) $f_2(t) \sim |\ln|t||^{-1/2}$.

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