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A three dimensional ferromagnetic Ising 'fluid' model

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Abstract. The three dimensional ferromagnetic spin-half Ising model with an arbitrary external magnetic field is considered in the spatial continuum limit and under a certain tempering condition to be imposed on the pair-wise spin-spin interaction. An expression for the partition function has been obtained for a tempered RKKY type interaction. The solution predicts the classical mean-field behaviour above a critical temperature below which the spontaneous magnetization jumps discontinuously from zero to the saturation value.

Keywords. Ising fluid model; order-disorder transition; ferromagnetism.

1. Introduction

The theoretical problem of order-disorder transitions in general and of phase transition in ferromagnetic systems in particular has often been treated in terms of the statistical mechanics of Ising Model (Ising 1925) which is, in a certain sense, the simplest idealization of the underlying physical situation (Newell and Montroll 1953). Exact solutions of the classical spin-half Ising model with nearest neighbour interactions are however, known only for the one-dimensional (non-zero field) and the two-dimensional (zero field) lattices (Newell and Montroll 1953, Onsager 1944, Yang 1952). From these exact solutions as also from some very general statistical mechanical considerations, it follows that there exists an important relationship between the occurrence of the order-disorder phase transition and the dimensionality and the range of interaction for the system in question. Thus, the one-dimensional Ising system with a *finite range of interaction* shows no transition to the ordered phase because of the offsetting effect of statistical fluctuations. The two- and the three-dimensional Ising systems, on the other hand, do exhibit an order-disorder transition (second order phase transition in the absence of external fields). The question of phase transitions in a one-dimensional system of spins interacting via a long range potential, decreasing exponentially with distance, has been treated intensively in the recent past (Kac 1959, Baker 1961; 1962, Kac and Helfand 1963). The partition function for such a one-dimensional model has been evaluated exactly by Baker (1961, 1962) following the formalism developed by Kac (1959) for the case of a one-dimensional gas of molecules interacting via an exponentially decreasing inter-molecular potential. In this formalism the partition function is expressed as an average over a set of stochastic fields acting on each molecule as a result of inter-molecular interactions, and advantage is taken of the fact that the correlation function for a stationary, Gaussian, Markoff process is an exponentially decreasing function. The one-dimensional system considered by Baker indeed shows a second-order phase transition, akin to the lambdo transition of liquid He^4 in the limit of *infinite range of interaction.* Extension of the above programme to higher dimensionality is limited by the fact that the Markoff process is intrinsically one dimensional. The two-dimensional problem could, however, be treated by assuming the inter particle potential to be factorisable row- and columnwise (Kac and Helfand 1963). This is of course, highly unphysical. Thus, the three-dimensional classical Ising problem has remained unsolved.

Attempts have been made to modify the three-dimensional Ising model so as to make it mathematically tractable and presumably more physical. Thus, in the exactly solvable spherical model of Berlin and Kac (1952), one replaces the dichotomic nature of the spin variables μ_l $(=\pm 1)$ by the sphericalisation condition $\sum_l \mu_l^2 = \mathcal{N}$. In the Gaussian model (Berlin and Kac, 1952) on the other hand, one introduces a Gaussian probability measure for the spin space. In the present paper, we report a treatment of the Ising model in the fluid (continuum) limit, wherein the dichotomic nature of the spin variables is retained, but the spatial discreteness (lattice) is replaced by a continuum. We shall refer to this as the Ising fluid. An essential simplification follows from a certain tempering condition to be imposed on the pair-wise spin-spin 'exchange' interaction, namely, that its spatial Fourier transform should have a finite support in the reciprocal space. This tempering condition is, of course, much more restrictive than is necessary. It holds for the RKKY type long-range interactions but obviously rules out the nearest neighbour (derivative of the delta function type) interactions. We believe that inasmuch as the discreteness of the spin variables is more essential than the discreteness of the spatial lattice, and that the RKKY type interactions do obtain in magnetic alloys and magnetic semiconductors, the solution obtained here should be of considerable interest. The solution is asymptotically exact (in the thermodynamic limit) above a critical temperature T_c. Below T_c, however, no such exactness is claimed.

2. The model

We will consider a system of $\mathcal{N}(=n^3)$ spins denoted by the dichotomic variables $\mu_l = \pm 1$, distributed over the vertices $\mathbf{R}_l$ of a simple cubic lattice of lattice constant a and interacting through the pair-wise potential $\mathcal{J}_{lm} = \mathcal{J}(|\mathbf{R}_l - \mathbf{R}_m|)$. The system is placed in an external magnetic field of strength H (measured in the units of energy). The partition function $\mathcal{Z}(\mathcal{N}, T, H)$ can be written as

$$\mathcal{Z}(\mathcal{N}, T, H) \equiv \mathrm{Tr}\exp(-\beta\mathcal{H})$$
$$= \int_{-\infty}^{+\infty} \cdots \int \exp\Big(\tfrac{1}{2}\beta \sum_{l \neq m} \mathcal{J}_{lm}\mu_l \mu_m + \beta H \sum_l \mu_l\Big) \times \prod_l p(\mu_l)\,\mathrm{d}\mu_l \tag{1}$$

Here $p(\mu)$ is the probability measure in the spin (μ) space given by $p(\mu) = 2\delta(\mu^2 - 1)$, and other symbols have their usual significance. The delta function ensures the dichotomic nature of the spins.

The bilinear exponent occurring in eq. (1) can now be diagonalized by the following unitary transformation (in analogy with the diagonalization procedure of the tight-binding scheme with one Wannier orbital per site):

$$X_{\kappa}^{\mathcal{N}} = (2/\mathcal{N})^{1/2} \sum_l \mu_l \cos(\boldsymbol{\kappa} \cdot \mathbf{R}_l), \quad Y_{\kappa}^{\mathcal{N}} = (2/\mathcal{N})^{1/2} \sum_l \mu_l \sin(\boldsymbol{\kappa} \cdot \mathbf{R}_l) \tag{2}$$

for $\kappa \neq 0$, and

$$X^N_{\kappa=0} = N^{-1/2} \sum_l \mu_l$$

along with the allowed values of the reciprocal vector $\kappa (\equiv \kappa_1, \kappa_2, \kappa_3)$ given by the cyclic boundary condition, i.e.,

$$\kappa_1, \kappa_2, \kappa_3 = (2\pi/na)\, m_1, m_2, m_3,$$

with $m_1, m_2 = 0, \pm 1, \pm 2, \ldots, \pm (n-1)/2$, and

$$m_3 = 0, 1, 2, \ldots, (n-1)/2.$$

This corresponds to κ lying in the upper half of the first Brillouin zone. Thus, in terms of these orthonormalized real variables we get

$$Z(N, T, H) = \int_{-\infty}^{+\infty} \cdots \int \exp\left(\sum_\kappa \tfrac{1}{2} J^N_\kappa \beta (X^N_\kappa)^2 + (Y^N_\kappa)^2 + \beta H \sqrt{N}\, X^N_0\right) \times \times \tilde{p}\left(\left\{X^N_\kappa, Y^N_\kappa\right\}\right) \prod_\kappa dX^N_\kappa\, dY^N_\kappa, \tag{3}$$

where

$$J^N_\kappa = \sum_{(l-m)} J(|\mathbf{R}_l - \mathbf{R}_m|) \cos \kappa \cdot (\mathbf{R}_l - \mathbf{R}_m)$$

Here $\tilde{p}\left(\left\{X^N_\kappa, Y^N_\kappa\right\}\right)$ is the transformed joint probability measure in the $\left\{X^N_\kappa, Y^N_\kappa\right\}$ space. The Jacobian of the transformation is unity.

We shall now consider the continuum limit of the above expression in the asymptotic sense. The continuum limit, i.e., $a \to 0$, $Na^3 = \Omega_N$ (=the volume), and the thermodynamic limit ($\Omega_N \to \infty$) can, however, be combined in an essential way as suggested by the theory of lattice spaces (*Gitterraum*) of Bopp (1958). Accordingly, we write for the direct lattice

$$a \sim n^{-1/2} L, \quad A \sim n^{1/2} L, \quad \sum_l \sim n^{3/2} L^{-3} \int d^3 r \tag{4}$$

and for the reciprocal lattice

$$b \sim 2\pi n^{-1/2} L^{-1}, \quad B \sim 2\pi n^{1/2} L^{-1}, \quad \text{and} \sum_\kappa \sim \frac{n^{\frac{3}{2}} L^3}{(2\pi)^3} \int d^3 \kappa \tag{5}$$

such that the number of lattice points in the direct as well as in the reciprocal lattice is $(A/a)^3 = (B/b)^3 = N$. Here L is some characteristic length of the system. Also,

$$J_{lm} \sim n^{-3/2} J^N(\lambda r), \quad \text{and } J^N_\kappa \sim \left(\frac{1}{L}\right)^3 \int J^N(\lambda r)\, e^{i\kappa \cdot r}\, d^3 r \tag{6}$$

where the factor $n^{-3/2}$ has been introduced so as to retain the proper intensive and the extensive nature of the physical quantities involved. Here $J^N(\lambda r)$ is the interaction function, and λ has the dimension of reciprocal of length. Inasmuch as there is no other characteristic length in the fluid limit, the scale is set by λ^{-1}. We can, therefore, take $L = \lambda^{-1}$ without loss of generality.

Now, we turn to the tempering condition to be imposed on $J^N(\lambda r)$. We require it to have a finite support (to within an additive constant) in the reciprocal space, i.e., $J^N_\kappa = 0$ (or a constant) for $\kappa > \kappa_c$, where κ_c is independent of N, or grows sufficiently slower than N as $N \to \infty$. More specifically, we take an RKKY type interaction:

$$J^N(\lambda r) \sim n^{-3/2}\, I\, j_1(\lambda r) \left(\frac{1}{\lambda r}\right)^3$$

for which

$$\tilde{J}_{\kappa}^{N} \sim I\pi^2\left(1-\frac{\kappa^2}{3\lambda^2}\right)+IC \text{ for } |\kappa| \leqslant \lambda \quad (7)$$
$$\sim IC \text{ for } |\kappa| > \lambda,$$

where C is a constant. Here I is an interaction constant and $j_1(x)$ is the spherical Bessel function of order one. This interaction is of a shorter range than the true RKKY interaction [notice the factor $(\lambda r)^{-3}$] and satisfies the tempering condition stated above.

Now, the exact solvability follows from the following considerations. We note that the set of dichotomic variables μ_l is a set of identically distributed statistically independent variables with mean zero and variance unity. X_{κ}^{N}, Y_{κ}^{N} are linearly independent (*but not statistically independent*) combinations of μ_l, and can be arranged according to some assigned ordinality (e.g., according as $\tilde{J}_{\kappa_0}^{N} > \tilde{J}_{\kappa_1}^{N} > \tilde{J}_{\kappa_2}^{N} > \ldots > \tilde{J}_{\kappa_N}^{N}$). It follows from the well-known extension of the central limit theorem (Feller 1972) that the set of variables X_{κ}^{N}, Y_{κ}^{N} forms the so-called triangular array having the property that for a given $\kappa=\kappa_c$, the truncated set $\left\{X_{\kappa}^{N}, Y_{\kappa}^{N}\right\}_{\kappa \leqslant \kappa_c}$ tends asymptotically to a set of statistically independent Gaussian variables, with mean zero and variance unity, in the limit $N \to \infty$. This asymptotic statistical independence can be expressed mathematically in the present case as

$$\tilde{p}\left(\left\{X_{\kappa}^{N}, Y_{\kappa}^{N}\right\}\right) \sim \prod_{\kappa \leqslant \kappa_c} p_0(X_{\kappa}^{N})\, p_0(Y_{\kappa}^{N})\; \tilde{\tilde{p}}\left(\left\{X_{\kappa}^{N}, Y_{\kappa}^{N}\right\}_{\kappa > \kappa_c}\right) \quad (8)$$

where $p_0(x)$ is the normalized Gaussian distribution function, and $\tilde{\tilde{p}}\left(\left\{X_{\kappa}^{N}, Y_{\kappa}^{N}\right\}_{\kappa > \kappa_c}\right)$ is the joint probability measure of the remaining variables normalised to unity.

Now, all we have to do is to combine the tempering condition (Eq. (7)) and the asymptotic statistical independence (Eq. (8)) by setting $\kappa_c=\lambda$, and substituting them in Eq. (3). We get

$$Z(N, T, H) \sim e^{1/2 N\beta IC} \int_{-\infty}^{+\infty} \ldots \int \exp\left[\sum_{\kappa \leqslant \lambda} (\Gamma_{\kappa}^{N} - \tfrac{1}{2}) \left((X_{\kappa}^{N})^2 + (Y_{\kappa}^{N})^2\right) + \sqrt{N}\beta H X_0^N\right] \times$$
$$\times \prod_{\kappa \leqslant \lambda} \left(\frac{dx_{\kappa}^{N}}{\sqrt{2\pi}}\right)\left(\frac{dy_{\kappa}^{N}}{\sqrt{2\pi}}\right) \quad (9)$$

With $\quad \Gamma_{\kappa}^{N} = \frac{1}{2}\beta\pi^2 I\left(1-\frac{\kappa^2}{3\lambda^2}\right)$

Thus the problem is reduced to evaluating a set of Gaussian integrals. We note at once that for $(\Gamma_{\kappa}^{N} - \frac{1}{2}) < 0$ for all κ, or $T > T_c = \pi^2 I/k_B$, the Gaussian integrals can at once be performed. For $T \leqslant T_c$, we get divergence and the partition function is ordinarily not well defined. We shall presently identify T_c with the critical temperature of phase transition, and then extend the treatment to temperatures below T_c as well.

3. Results and discussion

Case I $\quad T > T_c = I\pi^2/k_B \quad$ (paramagnetic region)

Using the relation $\int_{-\infty}^{+\infty} e^{ax^2+bx}\, dx = \sqrt{\frac{\pi}{-a}}\; e^{-b^2/4a}$ for Re $a<0$,

we get directly the Gibbs thermodynamic potential as

$$G(N, T, H) = -k_B T \ln Z(N, T, H) = \left(\frac{N^{\frac{1}{2}} k_B T}{4\pi^2}\right) \int_0^\infty \ln\left(\epsilon - (1-\epsilon)\frac{x^2}{3}\right) x^2 dx - \frac{NH^2}{4k_B(T-T_c)} + \text{constant} \tag{10}$$

where $\epsilon = 1 - T_c/T$.

From this, one gets the isothermal magnetic susceptibility $\chi(T)$ (per spin basis) as

$$\chi(T) = \mathscr{L}t_{N\to\infty} - \frac{1}{N}\left(\frac{\partial^2 G}{\partial H^2}\right)_T \sim \frac{1}{2k_B(T-T_c)} \tag{11}$$

the well-known classical Curie-Weiss law.

Also the zero-field specific heat $C_{H=0}$ (per unit volume basis) is seen to behave as

$$C_{H=0} = \mathscr{L}t_{N\to\infty} - \frac{1}{\Omega_N}\left(\frac{\partial^2 G}{\partial T^2}\right)_{H=0} \sim (T-T_c)^{-\frac{1}{2}} \text{ for } T \to T_c^+ \tag{12}$$

This confirms that T_c is to be identified with the critical temperature. Spontaneous magnetisation is, of course, identically zero for $T > T_c$.

Case II $\quad T \leqslant T_c = I\pi^2/k_B$ (Ferromagnetic region)

Here we have to note that the integrand in eq. (9) diverges as exp (x^2) as $x \to \pm\infty$. However, the limits of integration can no longer be set as infinity. In point of fact, these must be replaced by the maximum and the minimum values of the variables involved, i.e., by $(X_\kappa^N)_{\substack{\max\\ \min}} = (Y_\kappa^N)_{\substack{\max\\ \min}} = \pm a\sqrt{N}$ for $\kappa \neq 0$, and $(x_{\kappa=0}^N)_{\substack{\max\\ \min}} = \pm\sqrt{N}$, where $0 < a < 1$. The explicit numerical value of a is of no interest at the moment. When this is done, we get meaningful expressions for the thermodynamic quantities such as the spontaneous magnetisation per spin and the specific heat per unit volume. We will first consider the spontaneous magnetisation. For this we have to compute the field dependent contribution to the Gibbs thermodynamic potential G_{Field} (N, T, H) given by

$$G_{(\text{Field})}(N, T, H) \sim -k_B T \ln\left(\int_{-\sqrt{N}}^{+\sqrt{N}} e^{|\epsilon|(X_0^N)^2 + \sqrt{N}\beta H X_0^N} dX_0^N / \sqrt{2\pi}\right)$$

or

$$G_{(\text{Field})}(N, T, H) \sim -k_B T \ln\left\{\frac{1}{\sqrt{2\pi}}\left(\frac{e^{|\epsilon| N + \beta H N}}{2|\epsilon|\sqrt{N} + \beta H\sqrt{N}} + \frac{e^{|\epsilon| N - \beta H N}}{2|\epsilon|\sqrt{N} - \beta H\sqrt{N}}\right)\right\} \tag{13}$$

where we have used the asymptotic form for the error function

$$\mathscr{L}t_{|z|\to\infty} \text{ erf }(z) \sim \frac{1}{\sqrt{\pi}}\frac{1}{z}e^{-z^2}, \text{ for } |\arg z| < 3\pi/4.$$

Thus the spontaneous magnetisation per spin (m_s) is given by

$$m_s \sim -\frac{1}{N}\left(\frac{\partial G_{(\text{Field})}}{\partial H}\right)_T \sim 1 \text{ for } T \leqslant T_c \tag{14}$$

This shows that the spontaneous magnetisation jumps discontinuously at $T = T_c$ to its saturation value of one per spin. Such a jump was of course predicted for the one-dimensional Ising ferromagnet by Anderson and Yuval (1969) and was later established more rigorously by Dyson (1971) for interactions varying as R_{lm}^{-a}, where $1<a<2$.

Next, we shall consider the zero-field specific heat $C_{H=0}$ (per unit volume basis). Again, recalling that the limits of integration are $\pm\alpha\sqrt{\mathcal{N}}$, we get,

$$G(\mathcal{N}, T, H=0) \sim -\left(\frac{\mathcal{N}^{\frac{1}{2}} k_B T}{4\pi^2}\right)\int_0^1 \ln\left(\frac{1-exp\left(-2\alpha^2\mathcal{N}\left(\epsilon+(1-\epsilon)\frac{x^2}{3}\right)\right)}{\epsilon+(1-\epsilon)\frac{x^2}{3}}\right)x^2 \mathrm{d}x + \text{constant} \quad (15)$$

From eq. (15), the zero-field specific heat per unit volume $C_{H=0}$ can be directly obtained as

$$C_{H=0} = \mathscr{L}t_{\mathcal{N}\to\infty} - \frac{\mathrm{I}}{\Omega_{\mathcal{N}}}\left(\frac{\partial^2 G}{\partial T^2}\right)_{H=0} = \left(\frac{k_B \lambda^3 T}{4\pi^2}\right)\frac{\partial^2}{\partial T^2}\int_0^1 \ln|\epsilon+(1-\epsilon)\frac{x^2}{3}|\, x^2 \mathrm{d}x \quad (16)$$

Thus, the critical behaviour of the specific heat $C_{H=0}$ (per unit volume) below T_c is given by

$$C_{H=0} \sim (T_c - T)^{-1/2} \text{ for } T \to T_c \quad (17)$$

We find the critical index to be symmetrical about T_c in respect of specific heat $C_{H=0}$

At the end we would like to make a few clarifying remarks. It must be noted that below T_c, the spontaneous magnetisation jumps to the full saturation value and stays constant, whereas the specific heat is still non-zero. This seems paradoxical. The explanation is that the specific heat is computed on the *per unit volume basis* while the magnetisation on the *per spin basis*. The volume $\Omega_{\mathcal{N}}$, however, varies as $\mathcal{N}^{\frac{1}{2}}$ in the present case. Thus a spin fluctuation of the order of $\mathcal{N}^{1/2}$ per spin can give rise to non-zero finite specific heat on the per unit volume basis.

In conclusion, we would like to point out that the tempering condition used here is sufficient but not necessary. Also, the present results have been derived on the basis of the central limit theorem (cf. Eq. 8). It is, however, well known that this theorem breaks down in the tail of the distribution where the statistical independence no longer holds. The latter introduces no error above T_c in the asymptotic limit as $\mathcal{N}\to\infty$. The behaviour below T_c is, however, expected to get modified.

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