IC/81/132

INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

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INTERNATIONAL ATOMIC ENERGY AGENCY

UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION

1981 MIRAMARE-TRIESTE

International Atomic Energy Agency
and
United Nations Educational Scientific and Cultural Organization

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ABSTRACT

The low energy spin-wave like excitations in diluted ferromagnets near percolation threshold are studied. For this purpose an explicit use of the fractal model for the backbone of the infinite percolating cluster due to Kirkpatrick is made. Three physical effects are identified, which cause the softening of spin-waves as the percolation point is approached. The importance of fractal effects in the calculation of density of states and the low temperature thermodynamics is pointed out.

MIRAMARE - TRIESTE
August 1981

* To be submitted for publication.

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Randomly diluted magnets with short-ranged interactions have an interesting and complex behaviour near the percolation threshold[1,2]. It is now well recognized that a number of their dynamic and thermostatic properties depend crucially on the geometry of the infinitely connected percolating cluster (IPC), when one is above the percolation threshold. In randomly diluted systems, IPC has a very disordered and ramified structure[3], and in recent years much attention has been paid to develop simplified models for it with a view, to understand magnetic critical properties as well as conductance in inhomogenous mixtures. A significant step in this direction has been the so-called 'node-link' model introduced by Shal and Shklovshii[4] and de Gennes[5]. In this model, which has been exhaustively and critically discussed in recent articles[1,6,7], the biconnected component of IPC, also referred to as backbone is visualised as a series of small clusters set on a superlattice of nodes connected by one-dimensional crooked paths. The typical distance between the nodes is of the order of percolation correlation length $\xi_p \sim (p - p_c)^{-\nu_p}$. The average length, L, of the one-dimensional connecting parth is naturally greater than ξ_p and is assumed to diverge according to $(p - p_c)^{-\zeta}$ with $\zeta > \nu_p$. The upper limit on the path connecting nodes is clearly a self-avoiding walk, which means that $L < \xi_p^{1/\nu}$ SAW or $\zeta < \nu_p/\nu$ SAW.

While the node-link model has served a very useful purpose[5,7,8] in providing a nice intuitive understanding of how the reduced connectivity in the IPC affects the various physical properties, it has many drawbacks especially in two dimensions and to some extent in three dimensions[1,6,7]. Through a detailed study of the backbones of IPC in two dimensions, Kirkpatrick[1,3] has made several important observations, which are briefly: (i) As $p \to p_c+$, the one dimensional links do not grow faster than ξ_p. Thus a more appropriate assumption is that $L \approx \xi_p$. (ii) Though many regions of the backbone do look like one dimensional paths, the intersections usually occur much closer together than ξ_p. Thus ξ_p characterises the longest links in the backbone rather than the typical ones. (iii) Finally according to 'node-link' model the fraction of sites, B(p), in the backbone is $(p - p_c)^{d\nu - \zeta}$, which is much smaller than the values determined in computer simulations. Defining an index β_B such that $B(p) \propto (p - p_c)^{\beta_B}$ it is seen that $d\nu - \zeta \simeq 2\beta_B$. This means that the longest links form a negligible part of the backbone close to p_c.

Kirkpatrick[1,3] has taken this to imply that although the 'node-link' model captures some essential physics at length scales bigger than ξ_p, it completely misses the texture present at scales smaller than ξ_p. He models this texture as a self-similar fractal[9], in the following way. Since the backbone must be homogenous on scales bigger than ξ_p, one can regard it as being made up of statistically identical cells of linear dimension ξ_p. Now a finer texture, say some connections, are introduced at a scale of $\xi_p/2$. A similar texture is next introduced at the scale of $\xi_p/4$ and the process is continued by getting finer and finer until the scale equals a lattice constant. The fractal dimension of the backbone, d_B, is identified by calculating the volume of the material in a cell of side ξ, which is $\xi^{d_B}(p) \propto \xi^{d-\beta_B/\nu_p} = \xi^{d_B}$.

This model is very attractive from the geometrical point of view and is also very successful in giving correct exponents for conductivity and Hall effect in random resistor networks[1]. In this paper we use these considerations to study spin-wave excitations in ferromagnets and the resulting consequences for low temperature thermodynamics.

From general grounds, one knows that at sufficiently long wavelengths, i.e. for $q << \xi_p^{-1}$, the spinwave dispersion relation is[10,11,12] $\varepsilon_q = D(p)q^2$. The stiffness coefficient $D(p)$ has been exactly related[11] to the conductivity, $\Sigma(p)$, of a diluted resistor network through the relation

$$\Sigma(p) = D(p)\, P(p) \qquad (1)$$

where $P(p)$ denotes the fraction of sites in the IPC. Thus $D(p)$ vanishes at p_c like $(p - p_c)^{\mu}$, with $\mu = t - \beta_p$, where t is the exponent for the vanishing of conductivity at p_c and β_p is the usual index associated with the percolation probability. Within node-link model, Ziman[7] has related μ to the index ζ. He argues that since the wave moves along a crooked one-dimensional path, the wave-vector Q along the path, which enters into the formula for energy is not the same as, q, which is related to the real distance. For small q, there should be a linear relationship between the two, which he argues to be $q = (\frac{\xi_p}{L})Q$. Substituting this into the formula for excitations on the chain, $\omega = 2\,SJQ^2$, one obtains

$$D \propto (p - p_c)^{2(\zeta - \nu_p)} \qquad (2)$$

If the paths L are taken to be self-avoiding walks, this relation is in good numerical agreement with the numerical values calculated from relation $\mu = t - \beta$.

But as argued above, the one dimensional links are much shorter and surely the texture at length scales shorter than ξ_p affects the spin-wave stiffness. More over the effect of dead ends is not considered in Eq. (2). So we feel that the numerical agreement does not imply a complete elucidation of the problem. More importantly, the fractal nature of the backbone implies a different density of states for spin-wave excitations and hence a qualitatively different thermodynamic behaviour.

Our calculation of spin wave energies proceeds by identifying, somewhat artificially, three effects of the ramification and fractal nature of the IPC. Assuming with Kirkpatrick, that the backbone can be regarded as being made up of statistically identical cells of linear size ξ_p, we apply the standard formula[13] for spin-wave stiffness, given below in Eq. (3), to this cell.

$$D = \frac{\Sigma_{ij}\, J_{ij}\, S_i\, S_j\, r_{ij}^2}{3 \sum_i S_i} \qquad (3)$$

Since here all J_{ij}'s and S_i's are equal and are non-zero only for the nearest neighbours, this formula simply counts the average number of neighbours in the backbone. Clearly this number, α, lies between 2 and 2d for a d-dimensional hypercubic lattice.

The second step of our calculation consists in including the effect of dead ends. One can obtain this effect from formula (3) only, if one recognises that in applying the formula to IPC, the numerator should include only the sites in the biconnected component, whereas the denominator includes all the sites in IPC. Ziman[7] by considering the excitations in a chain to which dead ends are attached regularly, showed that the stiffness is reduced by a factor equal to the number of dead ends attached per site. We have verified this for more complex situations, in higher dimensions. Physically, we may argue that there is no phase drop (or voltage drop in case of resister network) along dead ends. So if one adds dead ends to the backbone, one does not alter $\Sigma(p)$, while the number of sites or $P(p)$ increases, thus $D(p)$ gets reduced by the number of dead ends per site. The third step of the

calculation is to relate the Q-vector on the fractal to the actual q, which, say, is seen by the neutron.

The backbone fractal[1,3] is constructed by taking a cell of size ξ_p and dividing it into 2^d parts. A fraction $(1-f)$ of these cells is removed (leaving the boundaries), and the remaining cells, $2^d f$, are further divided into 2^d parts each, and from each bigger cell a fraction $(1-f)$ is removed randomly as before. The process is continued till the lattice size is reached. A typical example is shown in Fig. 1. The parameter f is related to d_B according to the following equation

$$- \ell n_2 f = d_B = d - \beta_B/\nu_p \tag{4}$$

Now consider the calculation of the numerator, N, of Eq. (3). The contribution from the longest links is $2d(\xi_p - 2) + 2d$. At the next heirarchy we have $d2^d f$ links of length $\xi_p/2$ and these contribute $d.2^d f[2(\xi/2 - 2) + 2d]$. Thus we have a series

$$N = \frac{2JS^2a^2}{3} d \left\{ \xi_p \left[1 + (2^{d-1}f) + (2^{d-1}f)^2 + \cdots (2^{d-1}f)^n \right] + (d-2)\left[1 + (2^d f) + (2^d f)^2 + \cdots (2^d f)^n \right] \right\} \tag{5}$$

where $n = \ell n_2 \xi$. Since $2^{d-1}f > 1$, the sum becomes

$$N = \frac{2JS^2a^2}{3} d \left[\xi_p \frac{(2^{d-1}f)^n}{2^{d-1}f - 1} + \frac{(d-2)(2^d f)^n}{2^d f - 1} \right]$$

$$= \frac{2JS^2a^2}{3} \frac{d\, \xi_p^{d-\beta_B/\nu}}{2^{d-1}f - 1} \left[1 + (d-2) \frac{2^{d-1}f - 1}{2^d f - 1} \right] \tag{6}$$

Note that $\frac{d\xi_p^{d-\beta_B/\nu}}{(2^{d-1}f-1)}$ is simply equal to the number of bonds in one cell of the backbone. Thus the stiffness of the backbone is

$$D_{\text{Backbone}} = \frac{2JSa^2}{3} \left[1 + (d-2) \frac{2^{d-1}f-1}{2^d f-1} \right] \tag{7}$$

However, as argued above, in order to take into account the dead ends or loose spins, we should divide N by the total number of spins in the backbone, which is $\alpha\, \xi^d\, p(p - p_c)^\beta$. Thus we obtain

$$\tilde{D} = \frac{2JSa^2}{3p} \left[1 + (d-2) \frac{2^{d-1}f-1}{2^d f-1} \right] (p - p_c)^{(\beta_B - \beta)} \tag{8}$$

To give some idea of magnitudes here, we note that for $d = 2$, $\beta_B \sim 0.5$, $\beta = 0.15$ and $f = 3/4$, and for $d = 3$ $\beta_B = 0.9$, $\beta = 0.4$ and $f = 0.5$. Thus for $d = 3$, $D \sim \frac{8JSa^2}{9} (p - p_c)^{0.5}$. Eq. (7) for D_{Backbone} is an underestimate. We have also obtained an upper bound for the square lattice shown in Fig. 1.

We begin with the cell in which all the sites are occupied, and calculate the number of free boundaries created as we remove the subcells at each stage. At first level, we have 4 squares and by removing a fraction (1-f) of these we form boundaries of length $4(1-f)\xi/2$. At the next stage we remove $4^2 f(1-f)$ squares and create new boundaries containing $4^2 f(1-f) \frac{\xi}{4} [4\alpha_i + 3\alpha_e]$ spins, where α_i is the fraction of cubcells removed from the interim so that for such cells 4 new boundaries are created and α_e is fraction removed from the periphery, so that only 3 new edges are created. By actually carrying out this process successively, we determined empirically that after about fourth stage $\alpha_i = 1/5$ and $\alpha_e = 4/5$. Now we can count the total number of spins at the boundaries to be

$$4(1-f)\xi/2 \left(\frac{4}{5} + \frac{12}{5} \right) \left[1 + (2f) + (2f)^2 + \cdots (2f)^n \right]$$

$$= \frac{32}{5} (1-f) \xi \frac{(2f)^n}{2f-1} = \frac{32}{5} (1-f) \xi^{2 - \beta_B/\nu_p} \tag{9}$$

The stiffness $D_{Backbone}$ can be calculated by noting that all the interior spins have 4 neighbours, whereas the ones at the edge have only 3. Thus

$$D_{Backbone} = \frac{4JSa^2}{3}\left[1-\frac{3}{5}\right]\xi_p^{2-\beta_B/\nu_p} \Big/ \xi_p^{2-\beta_B/\nu_p}$$

$$= \frac{4}{5} JSa^2 \qquad (10)$$

Thus we find that the two estimates in Eqs. (9) and (10) do not differ much for $d = 2$.

To proceed with the third step, we write the spinwave energy as $\epsilon_q = \tilde{D}\, Q^2$, where Qa is roughly the average phase difference between neighbouring sites on the IPC along the direction in which the wave is progressing. To fix ideas, consider the propagation of a long wave length spinwave on a self-avoiding walk of L steps, so that the end to end direct distance is $\xi_p \sim L^{\nu_{SAW}}$. Let the phase difference between two ends be $\theta = q\xi_p$. Then the average phase difference between successive sites is θ/L for waves $q \gg \xi_p^{-1}$. This gives $\epsilon_q \simeq 2J(\theta/L)^2 = 2J(\xi_p/L)^2 q^2$, which is what Ziman[3] obtained. Now one would like to apply a similar argument to fractal. Consider a single cell of size ξ_p and fix the phase difference between two opposite edges to $\theta = q\xi_p$. The wave-front on a random fractal is obviously very complicated and the phase drop across the cell would occur along a very tortuous path. This path ℓ, is necessarily bigger than ξ_p, and in a simple theory should be related to the fractal dimension d_B. One such simple length which satisfies the above criterion is $\ell^{d_B} = \xi_p^{d}$. Now $|\vec{Q}| = \theta/\ell a$, so that the spin-wave energy ϵ_q becomes

$$\epsilon_q = \tilde{D}\, \xi_p^{2(1-d/d_B)}\, q^2 \qquad (11)$$

$$= \frac{2JSa^2}{3}\left[1 + (d-2)\frac{2^{d-1}f_{-1}}{2^{d}f_{-1}}\right](p-p_c)^{\mu} q^2 \qquad (12)$$

with

$$u = \beta_B - \beta + 2\beta_B/d_B \qquad (13)$$

In Table 1, we have listed various exponents for $d = 2$ and $d = 3$ and compared index μ calculated according to the above formula (13) with the exact relation $\mu = t - \beta$. The agreement is rather good. For larger values of q, one may employ the dynamical scaling argument given by Stauffer[14], according to which

$$\omega_q = q^Z f(q\xi_p) \qquad (14)$$

where $f(x) \to x^{2-Z}$ as $x \to 0$. Applying this to Eq. (12), we find $Z = 2d/d_B$, and thus the critical modes at shorter wavelengths have dispersion

$$\omega_q \sim q^{2d/d_B} \qquad (15)$$

In order to calculate the low temperature properties, one needs to know the density of states. Hitherto, most authors[7,15] have considered low temperature properties by using the dispersion relation (12) in the usual way to write the density of modes as

$$\rho(\omega) \propto \frac{\omega^{(d-2)/2}}{[D(p)]^{d/2}} \qquad (16)$$

This gives rise to contributions to specific heat and magnetisation which strongly diverge as $p \to p_{c+}$, eg. for $d = 3$

$$C_v \propto (p-p_c)^{-3\mu/2}\, T^{3/2} \qquad T \ll T_c(p) \qquad (17)$$

Here one may argue that since the modes are softening as $p \to p_{c+}$, their occupation probability rises vary strongly with temperature, but we would like to note that since the excitations are on a fractal, the q-space available to them is not as large as implied by Eq. (17).

One can use an illuminating scaling argument due to Stapleton et.al.[16], to determine the low energy density of states. Consider a sample of macroscopic linear size L. Its density of states $\rho_L(\omega)$ is

$$\rho_L(\omega) = \sum_i \delta(\omega - \omega_i(L)) \qquad (18)$$

Let the size of the sample be changed by a factor b: $L \to bL$, then it follows from simple hydrodynamic arguments[12] that $\omega_i \to \omega_i(L)/b^2$. Now

$$\rho_{bL}(\omega) = b^{d_B} \rho_L(\omega)$$
$$= \sum_i \delta(\omega - \omega_i/b^2) = b^2 \rho_L(b^2\omega)$$

Thus the scaling relation for density of states is

$$\rho_L(\omega) = b^{2-d_B} \rho_L(b^2\omega) \tag{19}$$

from which one finds at low ω

$$\rho_L(\omega) \propto \omega^{(d_B-2)/2} \tag{20}$$

Though one may not rule out a concentration dependent factor in Eq. (18)[17], we would like to emphasise that the main effect of dilution on density of states occurs through fractal dimensionality of the backbone supporting the excitations, rather than through factors contained in Eq. (17).

Let us discuss a 3-dimensional isotropic ferromagnet . Though the fractal dimensionality indicated is ~ 2, there is other evidence from series studies[18] on $T_c(p)$ to show that long range order does exist till very close to p_c, which is the true multicritical point. From this we presume that $d_B \sim 2+$. Then in the small temperature range $0 < T \ll T_c(p)$, the thermodynamics is governed by spin-wave excitations. In the presence of a field, H, which may be due to anisotropy and/or application, one has[13]

$$\frac{M(T)}{M(0)} - 1 \propto \left(\frac{kT}{J}\right)^{d_B/2} F\left(\frac{d_B}{2}, \frac{g\mu_B H}{kT}\right) \tag{21}$$

$$C_v/k \propto \left(\frac{kT}{J}\right)^{d_B/2} F\left(\frac{d_B}{2}+1, \frac{g\mu_B H}{kT}\right) \tag{22}$$

where

$$F(\lambda,\alpha) = \frac{1}{\Gamma(\lambda)} \int_0^\infty \frac{x^{\lambda-1}\,dx}{e^{x+\alpha} - 1} \tag{23}$$

For small fields Eqs. (21) and (22) assume the forms[13]

$$\frac{M(T)}{M(0)} - 1 \propto \left(\frac{kT}{J}\right)^{d_B/2} \left[\Gamma\left(1-\frac{d_B}{2}\right)\left(\frac{g\mu_B H}{kT}\right)^{\frac{d_B-2}{2}} + \zeta\left(\frac{d_B}{2}\right) + O(H)\right] \tag{24}$$

$$\frac{C_v}{k} \propto \left(\frac{kT}{J}\right)^{d_B/2} \left[\Gamma\left(-\frac{d_B}{2}\right)\left(\frac{g\mu_B H}{kT}\right)^{\frac{d_B}{2}} + \left(\frac{d_B}{2}+1\right)\zeta\left(\frac{d_B}{2}+1\right) + O(H)\right] \tag{25}$$

where $\zeta(x)$ denotes the Riemann Zeta-function. This shows that the use of Eq. (20) leads to a very different behaviour, both with regard to temperature and field.

Though Eqs. (24) and (25) represent clear-cut differences between diluted and homogenous systems, from the point of view of experimental verification, we note that since $T_c(p)$ itself is small, the temperature range of validity of the above formulae is rather restrictive. Until now we know of dilution studies on only one ferromagnetic compound[19]. On the other hand, many antiferromagnetic compounds have been investigated near percolation point[20], and it would be desirable to extend such considerations for antiferromagnetic excitations.

To summarise, we have identified certain physical mechanism which cause the softening of spin-wave stiffness in dilute ferromagnets, as the percolation concentration is approached. We find that an explicit account of the fractal nature of the infinite percolating cluster is necessary for these considerations. The fractal effects alter the nature of the low energy density of states which reflects significantly on the low temperature thermodynamic properties.

Table 1.

d	β	ν_p	β_B	σ	d_B	$\mu=\sigma-\beta$	$\mu=\beta_B-\beta+2\beta_B/d_B$
2	0.14	1.36	0.5	1.10	1.6	0.96	0.98
3	0.39	.88	0.9	1.62	2.0	1.33	1.41

ACKNOWLEDGEMENTS

The author would like to thank Professor Abdus Salam, the International Atomic Energy Agency and UNESCO for hospitality at the International Centre for Theoretical Physics, Trieste. He also wishes to thank Prof. N. Kumar for helpful discussions.

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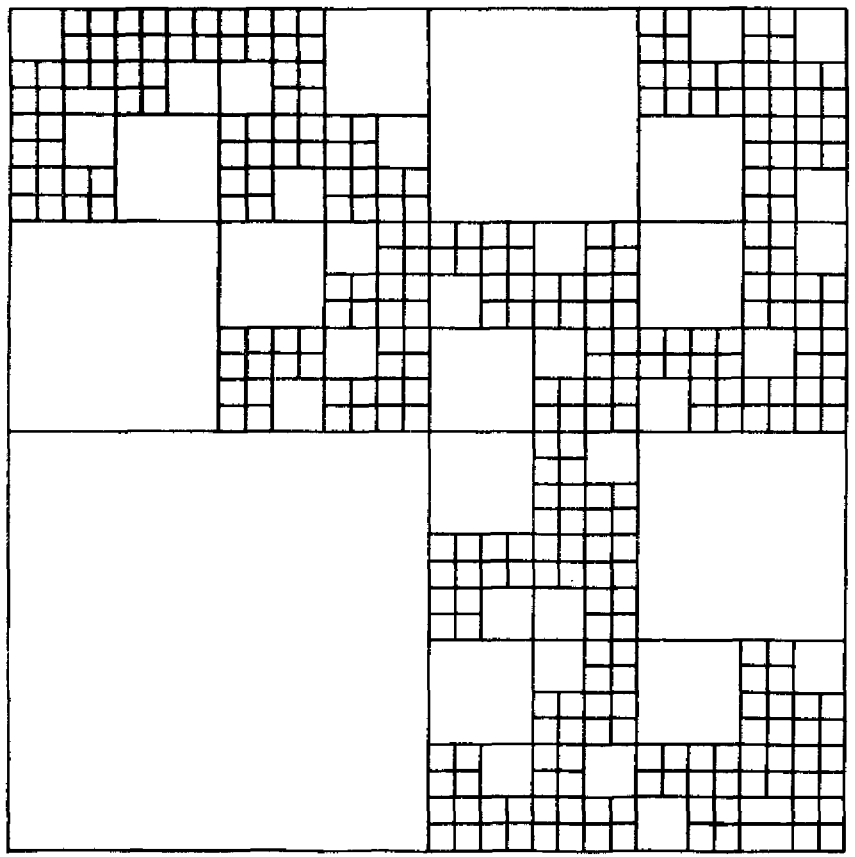

FIG. 1

Kirkpatrick's self-similar fractal model for the single cell of the backbone of an infinite cluster in two dimensions. The linear size of the cell is ξ_p and the backbone is made by joining statistically identical cells of this type. f = 3/4.

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IC/81/89 — Trieste International Symposium on Core Level Excitations in Atoms, Molecules and Solids, 22-26 June 1981 (Extended abstracts).

IC/81/90 INT.REP.* — A. TAWANSI, A.F. BASHA, M.M. MORSI and S.El-KONSOL - Effects of gamma-radiation and rare earth additives on the insulation of sodium silicate glasses.

IC/81/91 INT.REP.* — A. TAWANSI, A.F. BASHA, M.M. MORSI and S. El-KONSOL - Investigation of the electrical conductivity of γ-irradiated sodium silicate glasses containing multivalence Cu ions.