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Critical dynamic response of the dilute antiferromagnetic chain

A C Maggs and R B Stinchcombe

Department of Theoretical Physics, 1 Keble Road, Oxford, OX1 3NP, UK

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Abstract. The transverse dynamic response of the dilute classical Heisenberg antiferromagnetic chain is calculated near the percolation threshold as $k \rightarrow 0$ and $k \rightarrow \pi$. In the critical limit the universal dynamic structure function, which is calculated in closed form, exhibits dynamic scaling with exponents $z = 1$, $\eta_T = 1$. This function displays the crossover from spin wave $(k - \pi)\xi_p \gg 1$ to hydrodynamic $(k - \pi)\xi_p \ll 1$ response. The relevance of the results to the experimental situation is discussed.

1. Introduction

We give an exact calculation of the transverse contribution to the critical dynamic response of the dilute classical antiferromagnetic Heisenberg chain in the limit of diverging percolative correlation length ξ_p (Essam 1980) and zero temperature. Static dilution-induced critical phenomena have long been of interest but there are relatively few exact results in the field of critical dynamics of dilute systems (Korenblit and Shender 1978, Harris and Kirkpatrick 1977, Kirkpatrick 1979, Stinchcombe 1983a).

This paper obtains the response by carrying out the configurational average over individual chain segments (Stinchcombe and Harris 1983, Harris 1984, Ogadaki and Lax 1980). Each contribution to the average is calculated using the standard linear spin wave approximation (Anderson 1952). Numerical calculations on the same system have been performed by McGurn and Thorpe (1983) while exact results are available for instantaneous correlations (Fisher 1968, Thorpe 1975). The results of our calculation exhibit dynamic scaling (Halperin and Hohenberg 1969, Hohenberg and Halperin 1977) and are in broad agreement with recent experiments (Boucher *et al* 1978 and Endoh *et al* 1979, 1981).

The method of this paper is to solve the equations of motion for individual chain segments with free boundary conditions in a manner similar to that used for the simpler ferromagnetic case by Stinchcombe and Harris (1983). The responses of individual segments are summed with appropriate weights to give the configurationally averaged Green function in closed form to leading order in ξ_p^{-1} . Critical behaviour occurs near the percolation threshold in the limit $k \rightarrow \pi$ in contrast to the ferromagnet where criticality occurs at the centre of the Brillouin zone.

In the remainder of § 1 we set up the model and make some comments on its validity. In § 2 the model is solved in the continuum limit. A discussion follows in § 3.

We start with the Hamiltonian for the bond diluted antiferromagnetic chain in the presence of a site dependent transverse field \mathbf{h}

$$H = \frac{1}{2} \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i \mathbf{h}_i \cdot \mathbf{S}_i \quad (1.1)$$

The exchange coupling J_{ij} is a random variable with the probability distribution

$$P(J_{ij}) = (1-p)\delta(J_{ij}) + p\delta(J_{ij}-J) \quad (1.2)$$

where p is the bond concentration.

Even in one dimension, for large spin magnitude S , linearised equations of motion can be used provided the temperature is so low that the thermal correlation length ξ_T (proportional to temperature T) satisfies $\xi_T \gg \xi_p$, $(k-\pi)^{-1}$. Although thermal and quantum fluctuations destroy long range order there are long regions of correlated spins in which a local z axis can be defined about which spin wave precessional dynamics occur for $(k-\pi)^{-1} \ll \xi_T$.

In the linearised formalism, the dispersion curve in the pure limit is given by

$$\omega = 2J|\sin k|. \quad (1.3)$$

As we intend to sum over an ensemble of chain segments we need the probability of a given site belonging to an n bond chain:

$$P(n) = (1-p)^2 p^n. \quad (1.4)$$

The percolative correlation length ξ_p is the characteristic length controlling the behaviour of the dilute system and is given by

$$\xi_p = |\ln p|^{-1}. \quad (1.5)$$

Using standard methods the equations of motion derived from (1.1) are

$$(SJ_{l,l+1} + SJ_{l,l-1} + (-1)^{\alpha_l} \omega) S_l^+ = -Sh_l^+ - SJ_{l,l+1} S_{l+1}^+ - SJ_{l,l-1} S_{l-1}^+ \quad (1.6)$$

where α_l specifies the sublattice of the l th spin and can equal 1 or 2. S_l^+ is the usual combination ($S_l^+ = S_l^x + iS_l^y$) of the transverse spin components at the l th site.

We define the Green functions $G_{l,l'}^{(n)}$ on an n bond chain segment by

$$S_l^+ = \sum_{l'} G_{l,l'}^{(n)} h_{l'}^+ \quad (1.7)$$

It follows that the average transverse dynamic structure function and hence the neutron cross section is given by

$$\chi(k, \omega, p) = -\text{Im} G(k, \omega + i\epsilon, p) \quad (1.8)$$

where

$$G(k, \omega, p) = \left\langle \sum_{l,l'} G_{l,l'}^{(n)}(\omega) \exp(ik(l-l')) \right\rangle \quad (1.9)$$

(\dots) here denotes a configurational average over all bond configurations.

From dynamic scaling arguments (Halperin and Hohenberg 1969) and the result for the ferromagnet (Stinchcombe and Harris 1983) we expect the result to display the dynamic scaling form

$$\chi(k, \omega, p) = q^{-(2-\eta_{\tau+z})} F(q\xi_p, \omega q^{-z}) \quad (1.10)$$

(where $q = k - \pi$) in the critical limit $\xi_p \rightarrow \infty$ with $q \rightarrow 0$, $\omega \rightarrow 0$.

This is confirmed in the present calculation which gives the functional form of $F(q\xi_p, \omega q^{-z})$ and the critical exponents z and η_T . The relevance of the result to neutron scattering experiments is mentioned. The details of the theory are given in the next section which begins with an outline of the sequence of steps in the development.

2. Theory

We now solve the model which was introduced in § 1. Starting from the equations of motion (1.6), we find the equation obeyed by the Green functions (1.7) which are solved, in the limit appropriate to criticality, by going to a continuum formalism. These Green functions can then be averaged to give the response in closed form (2.26).

To calculate the Green functions, G of (1.7) we decimate the equations of motion (Marland 1978, Goncalves da Silva and Koiller 1981, Stinchcombe 1983b, Stinchcombe and Harris 1983) so that the motion of a spin on a sublattice is given in terms of other spins on the same sublattice.

By considering the equation of motion of a linear combination of the $G_{ll'}$ it will be seen that terms augment or nearly cancel each other in the two limits $k \rightarrow \pi$ (criticality) and $k \rightarrow 0$ (where the field becomes an irrelevant variable and ceases to couple to the chain). In calculating configurational averages we freely discard Green functions, from the ends of chains with an odd number of atoms, which do not fit into our grouping of terms. In the limit of diverging ξ_p these contribute a vanishing proportion of the response.

We begin by considering a particular segment of the diluted chain consisting of n bonds joining sites l with $0 \leq l \leq n$.

Rewriting (1.1) for an atom on the interior of such a chain segment we obtain

$$(2 + (-1)^{\alpha_l \bar{\omega}}) S_l^+ = -\hbar_l^+ - S_{l-1}^+ - S_{l+1}^+ \quad (2.1)$$

where

$$\bar{\omega} = \omega / JS \quad \text{and} \quad \hbar^+ = h^+ / J.$$

The corresponding equation for the spin at the left-hand end site $l=0$ of the segment is

$$(1 + (-1)^{\alpha_0 \bar{\omega}}) S_0^+ = -\hbar_0^+ - S_1^+. \quad (2.2)$$

An analogous equation applies at the other end of the chain.

Performing the decimation by eliminating S_{l-1}^+ and S_{l+1}^+ from (2.1) gives the equations of motion for interior sites:

$$(2 - \bar{\omega}^2) S_l^+ = -(2 - (-1)^{\alpha_l \bar{\omega}}) \hbar_l^+ + \hbar_{l-1}^+ + \hbar_{l+1}^+ + S_{l-2}^+ + S_{l+2}^+. \quad (2.3)$$

These equations have to be solved subject to boundary conditions for the end spins of the decimated system which can be obtained as follows from (2.2): we eliminate S_1^+ from (2.2) to give one form of boundary condition:

$$(1 + (-1)^{\alpha_0 \bar{\omega}}) S_0^+ = -(2 - (-1)^{\alpha_0 \bar{\omega}}) \hbar_0^+ + \hbar_1^+ + S_2^+ + O(\omega^2). \quad (2.4)$$

Another form is obtained by eliminating S_0^+ from (2.2)

$$(1 + (-1)^{\alpha_0 \bar{\omega}}) S_1^+ = (2 - (-1)^{\alpha_0 \bar{\omega}}) \hbar_0^+ - \hbar_1^+ (2 + (-1)^{\alpha_0 \bar{\omega}}) + \hbar_2^+ + S_3^+ + O(\omega^2). \quad (2.5)$$

For chains with odd numbers of atoms we also find the result, obtained in a similar manner,

$$(1 + (-1)^{\alpha_0} \bar{\omega}) S_2^+ = -(2 - 3(-1)^{\alpha_0} \bar{\omega}) \hbar_0^+ + (2 - (-1)^{\alpha_0} \bar{\omega}) \hbar_1^+ - (2 - (-1)^{\alpha_0} \bar{\omega}) \hbar_2^+ + \hbar_3^+ + S_4^+ + O(\omega^2). \tag{2.6}$$

The linear combination of Green functions used in the calculation will be designated $H_{l,l+m}^{(n)}$ and is defined by

$$H_{l,l+m}^{(n)} \equiv (G_{l,l+m}^{(n)} + e^{-ik} G_{l,l+m+1}^{(n)}) + e^{ik} (G_{l+1,l+m}^{(n)} + e^{-ik} G_{l+1,l+m+1}^{(n)}). \tag{2.7}$$

This makes it possible to rewrite (1.9) as,

$$G(k, \omega, p) = \sum_{n=0}^{\infty} (1-p)^2 p^n \sum_{l,l'} G_{ll'}^{(n)} \exp(ik(l-l')). \tag{2.8}$$

$$= \sum_{n=0}^{\infty} (1-p)^2 p^n \frac{1}{2} \sum_{l,l'} (G_{ll'}^{(n)} + G_{ll'}^{(n+1)}) \exp(ik(l-l')) (1 + O(\xi_p^{-1})) \tag{2.9}$$

$$= \sum_{n=0}^{\infty} \frac{1}{2} (1-p)^2 p^n \sum_{j=n}^{n+1} \sum_{m \text{ even}} \langle H_{l,l+m}^{(j)} \rangle \exp(-ikm) \tag{2.10}$$

where

$$\begin{aligned} \langle H_{l,l+m}^{(n)} \rangle &= \sum_{l \text{ even}}^{n-m} H_{l,l+m}^{(n)} \quad \text{for } n \text{ even} \\ &= \sum_{l \text{ even}}^{n-m-1} H_{l,l+m}^{(n)} \quad \text{for } n \text{ odd.} \end{aligned} \tag{2.11}$$

In writing (2.11) we have discarded a vanishing proportion of Green functions as mentioned above.

The next step in the development will be to find the equations of motion of the $H_{l,l'}^{(n)}$ then to go to a continuum limit by replacing difference equations (to be obtained from (2.3)–(2.6)) by differential equations, and summations (2.10)–(2.11) by integrals.

We now suppose the field to which the response is required is periodic in space with wavevector k ($h_l^+ = \exp(-ikl) h_k^+$). For an n -bond segment in such a field we find from (1.7), (2.7),

$$S_l^+ = \sum_{l' \text{ even}} (G_{l,l'}^{(n)} + e^{-ik} G_{l,l'+1}^{(n)}) h_k^+ e^{-ikl'}. \tag{2.12}$$

This implies

$$T_{2l} \equiv S_{2l}^+ + e^{ik} S_{2l+1}^+ = \sum_{l' \text{ even}} H_{2l,l'}^{(n)} e^{-ikl'} h_k^+. \tag{2.13}$$

The natural length of the system is ξ_p defined in (1.5), which we now consider to be large. Then it is convenient to define the space variables x, y, L and reduced frequency and wave vector labels $\tilde{\omega}, \tilde{k}, \tilde{k}$ according to

$$\begin{aligned} y = l / \xi_p & \quad \tilde{k} = (k - \pi) \xi_p & \text{as } k \rightarrow \pi \\ x = (l + m) / \xi_p & \quad \tilde{k} = k \xi_p & \text{as } k \rightarrow 0 \\ L = n / \xi_p & \quad \tilde{\omega} = \frac{1}{2} \tilde{\omega} \xi_p. \end{aligned} \tag{2.14}$$

For each of the limiting cases k near π and k near zero a continuum description (x, y, L continuous) can then be obtained by taking appropriate limits in which $\xi_p \rightarrow \infty$ with \tilde{k} or \tilde{k} held finite, and $\omega \rightarrow 0$ to keep $\tilde{\omega}$ finite.

Correct to leading order in ξ_p^{-1} , we have

$$T_{l-2} + T_{l+2} - 2T_l \rightarrow (4/\xi_p^2)(\partial^2/\partial y^2)(T(y)), \tag{2.15}$$

so that (2.3) becomes

$$\begin{aligned} (\tilde{\omega}^2 + (4/\xi_p^2)(\partial^2/\partial y^2))T(y) &= 4(1 - \cos k) e^{-i\tilde{k}y} \tilde{h}_k^+ & k \rightarrow \pi \\ &= 4(1 - \cos k) e^{-i\tilde{k}y} \tilde{h}_k^+ & k \rightarrow 0. \end{aligned} \tag{2.16}$$

In the two limits $k \rightarrow \pi, 0$, (2.16) together with (2.13) then lead to

$$(\tilde{\omega}^2 + \partial^2/\partial y^2)(H^{(L)}/\xi_p)(y, x; k \rightarrow \pi) = (4/J)\delta(y - x) \tag{2.17}$$

$$(\tilde{\omega}^2 + \partial^2/\partial y^2)(H^{(L)}/\xi_p)(y, x; k \rightarrow 0) = -(1/\xi_p^2)(\partial^2/\partial y^2)\delta(y - x)/J. \tag{2.18}$$

By considering the same continuum limit for the boundary conditions derived from (2.4)–(2.6) it can be shown that for both limits $k \rightarrow \pi, 0$ the field terms acting on the end of the chain contribute a vanishing proportion of the response as ξ_p diverges. Hence to leading order

$$\partial H^{(L)}/\partial y = \pm \tilde{\omega} H^{(L)} \quad \text{when } y = 0, L$$

which we rewrite as

$$\begin{aligned} \partial H^{(L)}/\partial y &= \nu \tilde{\omega} H^{(L)} & y = 0 \\ &= \eta \tilde{\omega} H^{(L)} & y = L \end{aligned} \tag{2.19}$$

where η, ν take on the values ± 1 as required. (Depending on whether the segment ends on an up or a down sublattice.)

With these boundary conditions, (2.17) and (2.18) have the solutions

$$\begin{aligned} \frac{1}{\xi_p} H_{\nu, \eta}^{(L)}(x, y; k \rightarrow \pi) &= \frac{2(\eta i \exp(i\tilde{\omega}(x - 2L)) + \exp(-i\tilde{\omega}x))(\nu i \exp(i\tilde{\omega}y) + \exp(-i\tilde{\omega}y))}{J(\nu - \eta \exp(-2i\tilde{\omega}L))\tilde{\omega}} & (y < x) \\ &= \frac{2(\eta i \exp(i\tilde{\omega}(y - 2L)) + \exp(-i\tilde{\omega}y))(\nu i \exp(i\tilde{\omega}x) \exp(-i\tilde{\omega}x))}{J(\nu - \eta \exp(-2i\tilde{\omega}L))\tilde{\omega}} & (y > x) \end{aligned} \tag{2.20}$$

$$\frac{1}{\xi_p} H_{\nu, \eta}^{(L)}(y, x; k \rightarrow 0) = \frac{\tilde{\omega}^2}{4\xi_p^3} H_{\nu, \eta}^{(L)}(y, x; k \rightarrow \pi). \tag{2.21}$$

In the experimental situation there will always be some coupling between neighbouring chains so that at low temperatures although the dynamics can be considered as a one-dimensional problem there will be a three-dimensional ordering of the system and individual chain segments will be correlated. In such a system and also in an ensemble of uncorrelated segments it is necessary to average over ν and η which independently take on the values ± 1 . This leads us to define

$$\bar{H}^{(L)} = \frac{1}{2} \sum_{\nu, \eta} H_{\nu, \eta}^{(L)} \tag{2.22}$$

which takes into account chains with n both odd and even arising in the sum over j in (2.10). The result is

$$\frac{1}{\xi_p} \bar{H}^{(L)}(x, y; k \rightarrow \pi) = \frac{2}{\tilde{\omega}J} \left(\frac{\cos(L+y-x)\tilde{\omega}}{\sin \tilde{\omega}L} - \frac{\sin(L+y-x)\tilde{\omega}}{\cos \tilde{\omega}L} \right) \quad (y < x). \quad (2.23)$$

Inserting this into the continuum limit of (2.10) and using (1.9) we find the following expression for the average transverse response

$$\chi = -\text{Im } G(k, \omega, p) = -\text{Im } \xi_p/4 \int_0^\infty dL e^{-L} \int_0^L dz \cos \tilde{k}z \int_0^{L-z} \bar{H}^{(L)}(y, y+z) dy \quad (2.24)$$

$$= \frac{\text{Im}}{2J} \xi_p^2 \left[\frac{2}{(\tilde{k}^2 - \tilde{\omega}^2)} + \frac{1}{\tilde{\omega}} \left(\frac{1}{(\tilde{\omega} - \tilde{k})^2} \int_0^\infty e^{-L} \frac{\cos 2\tilde{\omega}L - \cos(\tilde{\omega} + \tilde{k})L}{\sin 2\tilde{\omega}L} dL \right. \right. \\ \left. \left. + \frac{1}{(\tilde{\omega} + \tilde{k})^2} \int_0^\infty e^{-L} \frac{\cos 2\tilde{\omega}L - \cos(\tilde{\omega} - \tilde{k})L}{\sin 2\tilde{\omega}L} dL \right) \right]. \quad (2.25)$$

Completing the integrals we arrive at the following result for the average response in the two limits $k \rightarrow \pi, k \rightarrow 0$:

$$\chi(\tilde{k}, \tilde{\omega}, \xi_p; k \rightarrow \pi)$$

$$= \frac{\pi \xi_p^2}{8J\tilde{\omega}^2} \left(\frac{\coth \pi/4\tilde{\omega}(1 - \sin \tilde{k}\pi/2\tilde{\omega})}{(\tilde{\omega} - \tilde{k})^2(\cosh \pi/2\tilde{\omega} - \sin \tilde{k}\pi/2\tilde{\omega})} \right. \\ \left. + \frac{\coth \pi/4\tilde{\omega}(1 + \sin \tilde{k}\pi/2\tilde{\omega})}{(\tilde{\omega} + \tilde{k})^2(\cosh \pi/2\tilde{\omega} + \sin \tilde{k}\pi/2\tilde{\omega})} \right) \quad (2.26)$$

$$\chi(\tilde{k}, \tilde{\omega}, \xi_p; k \rightarrow 0) = \frac{\tilde{\omega}^2}{4\xi_p^2} \chi(\tilde{k}, \tilde{\omega}, \xi_p; k \rightarrow \pi). \quad (2.27)$$

In the critical limit where ξ_p diverges and where $k - \pi$ and $\tilde{\omega}$ vanish but where \tilde{k} and $\tilde{\omega}$ remain finite the result (2.26) displays the dynamic scaling form (1.10) with $z = 1, \eta_T = 1$, and with the following explicit form for the universal scaling function

$$F(a, b) = \frac{\pi}{2a^2b^2J} \coth \pi J/2ab \left(\frac{1 - \sin \pi J/b}{(b/2J - 1)^2(\cosh \pi J/ab - \sin \pi J/b)} \right. \\ \left. + \frac{1 + \sin \pi J/b}{(1 + b/2J)^2(\cosh \pi J/ab + \sin \pi J/b)} \right) \quad (2.28)$$

where $a = (k - \pi)\xi_p, b = \omega(k - \pi)^{-1}$.

As \tilde{k} varies from 0 to ∞ this function describes the crossover from a broad hydrodynamic response to a sharp spin wave peak at an energy given by the pure dispersion curve (1.3), as will be discussed in the next section.

3. Discussion

The evolution of the scaling function $F(a, b)$ as $(k - \pi)\xi_p$ varies is shown in figure 1. For $(k - \pi)\xi_p \ll 1$ the scattering is dominated by the broad hydrodynamic response which moves down in reduced frequency $(\omega/(k - \pi)^2)$ as $(k - \pi)\xi_p$ increases. As

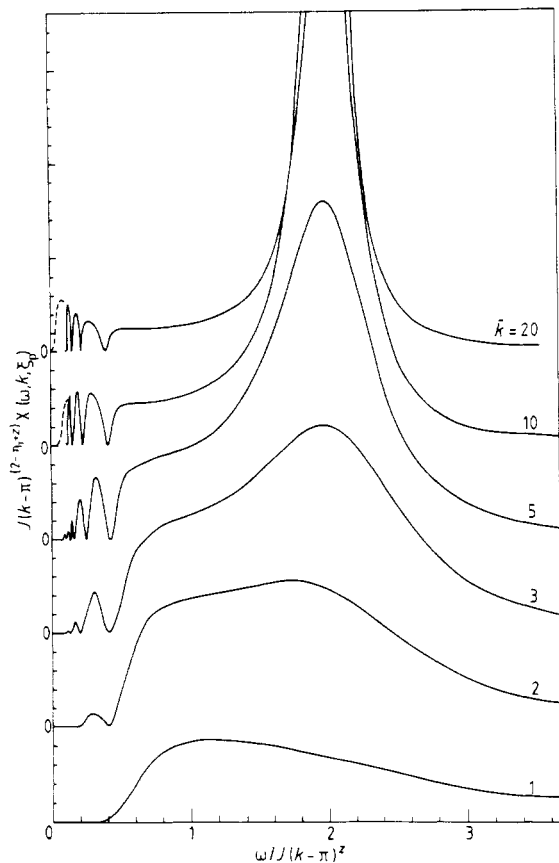


Figure 1. Plot of the universal scaling function $F((k - \pi)\xi_p, \omega(k - \pi)^{-z})$ for the transverse response of the dilute antiferromagnetic classical chain. Here $\tilde{k} = (k - \pi)\xi_p$, $z = 1$, $\eta_T = 1$. For large \tilde{k} the response is dominated by the spin wave peak at the same energy as is found in the pure system. For $\tilde{k} \leq 1$ the response is dominated by the broad hydrodynamic contribution.

$(k - \pi)\xi_p$ increases further the spin wave response becomes dominant with the hydrodynamic contribution continuing to move to lower reduced frequencies. The spin wave peak is predicted to remain at the same energy as the response from a pure sample though it is broadened by the finite magnon lifetime.

For intermediate $(k - \pi)\xi_p$ we see oscillations in the scattering with minima when

$$\tilde{k}/\tilde{\omega} = (4n + 1) \quad n \neq 0. \tag{3.1}$$

This can be explained by considering the oscillations set up in the chain segments by the probing field h_k^+ . For these particular frequencies the ‘overlap’ (defined in some appropriate manner) between the external field and the internal wavefunction will vanish forcing the response of the system also to vanish at these points.

The principal features of the results make contact with the experiments by Boucher *et al* (1978) and Endoh *et al* (1979, 1981) who measured the inelastic scattering of neutrons from the quasi-one-dimensional antiferromagnet TMMC diluted with copper ions. They only found a spin wave peak present when $k\xi_p \geq 2$. It was also found that

the position of the peak did not move when the concentration of copper ions was varied. This is in agreement with what is predicted by the present calculation.

In figure 2 we reproduce some results and computer simulations from the paper of Endoh *et al* (1981). The experimental results show the decrease in intensity as $(k - \pi)\xi_p$ is reduced which has already been noted above for our calculation. The computer simulations for $(k - \pi)\xi_p = 2.2$ and $(k - \pi)\xi_p = 3$ display the same double hump seen in our calculations of figure 1.

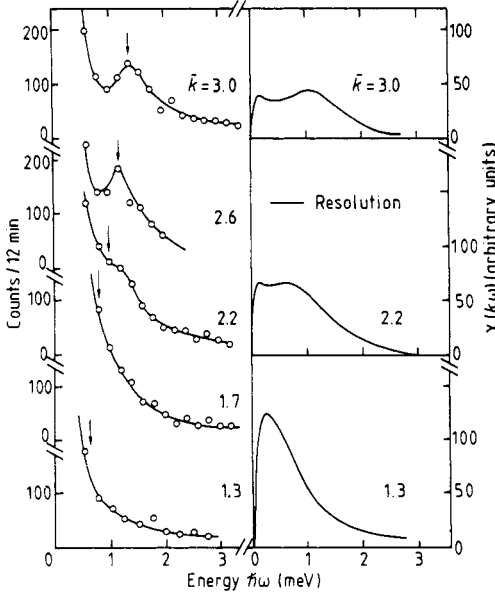


Figure 2. Experimental results for the site-diluted quasi one-dimensional antiferromagnet TMMC (left-hand side; $T = 0.36$) and computer simulations (right-hand side) (taken from the paper of Endoh *et al* 1981). The experimental results clearly show a loss in the intensity of the spin wave peak as \bar{k} decreases. The computer simulations give curves in broad agreement with our exact calculation.

Observation of oscillations predicted in (2.26) is hampered by the strong peak centred at $\omega = 0$, (see figure 2). This is largely due to incoherent nuclear scattering. We also expect a contribution to this peak from longitudinal two-magnon scattering broadened by thermal fluctuations. This response has been considered and can be calculated by an extension of the methods used here.

Of course, our calculation has treated the bond diluted system, while the experiments are on site-diluted chains. We are therefore led to consider possible differences in the leading-order contribution to the response in bond-diluted and site-diluted systems.

Certainly in more than one dimension we expect site-diluted and bond-diluted systems to behave differently as the percolation threshold and the local environment differ. In one dimension, however, clusters are independent chain segments with a probability distribution of chain lengths identical to leading order ξ_p^{-1} . In the case considered in this paper where ξ_p diverges the average response will be the same for both the site- and bond-diluted problem.

It is to be noted that the exponent η_T is not the same for the ferromagnetic and antiferromagnetic systems. We find $\eta_T^{\text{AF}} = 1$. Stinchcombe and Harris (1983) give $\eta_T^{\text{F}} = 2$. These exponents characterise the spatial decay of instantaneous transverse correlations. As is well known, the ferromagnetic and antiferromagnetic systems are in the same static universality class hence the exponent describing the decay of the correlation function $\langle \mathbf{S}_i \cdot \mathbf{S}_i' \rangle$ is the same. This correlation function is dominated by the longitudinal contribution $\langle S_i^z S_i'^z \rangle$ to which the transverse fluctuation is a less singular correction.

It should perhaps be pointed out that the antiferromagnet is also amenable to treatment by a real space scaling method as used by Stinchcombe and Harris (1983) and Stinchcombe (1983b). This would probably be the best way to generalise the results to higher dimensions. Results for the ferromagnetic system using this method are already available (Harris and Stinchcombe 1983).

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