Dynamics of localised modes in a random one-dimensional diluted antiferromagnet

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Abstract. We study the dynamics of localised modes in a random one-dimensional diluted Heisenberg antiferromagnet with an XY anisotropy and an applied magnetic field perpendicular to the chain, using the double-time retarded and advanced Green functions at zero temperature.

1. Introduction

The study of elementary magnetic excitations on the random one-dimensional diluted antiferromagnet is interesting both theoretically and experimentally (Birgenau and Shirane 1978, Cowley *et al* 1980). Theoretically, the parameters of the model are those of the pure system, which can be defined precisely (Wolfram and Callaway 1963, Cowley and Buyers 1972, Harris *et al* 1974, Holcomb 1974), and we can find analytical solutions for the problem using well known approximation schemes. Experimentally, nature offers us real physical systems that correspond remarkably closely to these idealised models, which allow the necessary test for our theories. Neutron studies of the more notable random one-dimensional diluted antiferromagnet $(CD_3)_4NMn_cCu_{1-c}Cl_3$ (TMMC:Cu) (Endoh *et al* 1979, 1981, Boucher *et al* 1978, Shirane and Birgenau 1977) have contributed to the understanding of the static and dynamic behaviour of the random one-dimensional diluted antiferromagnet.

The effects of randomness are most dramatic in one dimension (Nagler *et al* 1984, Birgenau and Shirane 1978, Endoh *et al* 1981) because the presence of a single vacancy in the spin chain breaks up the long-range correlation and blocks up the free propagation of spin waves. Then, for the dynamics, the random one-dimensional diluted antiferromagnet is constituted by a set of finite chains. As in a chain an end spin has only one nearest neighbour whereas an interior spin has two, a resonance associated with excitations on end spins is expected to occur at one-half the zone boundary frequency. However, as we discuss extensively in this paper, the presence of localised modes associated with excitations on end spins depends on the Hamiltonian of the model.

In this paper we investigate the resonances associated with excitations on end spins of the random one-dimensional diluted Heisenberg antiferromagnet with an XY anisotropy and an applied magnetic field perpendicular to the chain.

We consider a random two-sublattice antiferromagnet chain model, with the impurity–host atom exchange parameter small relative to the host atom–host atom exchange parameter. In the limit of low concentration of impurities and with no tendency for the formation of clusters of impurities, so pairs of impurities are very rare, and at zero temperature, if the spins of the host atoms are sufficiently large, independently of the sign of the impurity–host atom exchange parameter, the chain has long-range order and is in the classical Néel ground state.

Our results can be tested on TMMC : Cu at very low temperatures, in the limit of low concentration of Cu, because the Mn–Cu exchange parameter is small relative to the Mn–Mn exchange parameter, there is no tendency for the formation of clusters of Cu atoms in this compound, and the Mn atom has a spin ($S = 5/2$) sufficiently large.

In the Heisenberg chain the excitations on end spins are not separated from the spin-wave band. This is explained in terms of the singular nature of the density of states in one dimension at the band edges (Endoh *et al* 1981). We show that an XY anisotropy separates the excitations on end spins from the excitations on interior spins. In this respect this work complements the work of Endoh *et al* (1981), who show that an Ising anisotropy separates the excitations on end spins from the excitations on interior spins.

Thermodynamically, an XY anisotropy and an applied magnetic field perpendicular to the chain in the Heisenberg chain are equivalent (Pires and Gouvêa 1984). We show that an applied magnetic field perpendicular to the chain, differently from an XY anisotropy, does not separate the excitations on end spins from the excitations on interior spins.

The Ising chain and the Heisenberg chain with an XY anisotropy and an applied magnetic field perpendicular to the chain with $h = 2[2(1 - b)]^{1/2}$, where h is the scaled applied magnetic field and b is the anisotropy parameter, are thermodynamically equivalent (Pires and Gouvêa 1984). We show that this equivalence is not preserved in the dynamics of these random diluted antiferromagnets.

2. Hamiltonian

The model we treat is a one-dimensional Heisenberg antiferromagnet, with an XY anisotropy and a magnetic field applied perpendicular to the chain, in which a fraction, $1 - c$, of magnetic ions chosen at random are replaced by non-magnetic ions. The Hamiltonian for this model may be written as

$$\mathcal{H} = 2J \sum_n \rho_n \rho_{n+1} (S_n \cdot S_{n+1} - D S_n^z S_{n+1}^z) - \mu g H \sum_n \rho_n S_n^x \tag{2.1}$$

with the z axis along the chain direction. Here J and D are the exchange and anisotropy interaction parameters, μ is the Bohr magneton, g is the Landé factor and H is the applied magnetic field. The ρ_n are random variables having the value 1 or 0 depending on whether or not the site n is occupied by a magnetic ion. We assume the ρ_n are independent: $\overline{\rho_n} = \overline{\rho_n^2} = c$, and $\overline{\rho_m \rho_n} = c^2$ if $n \neq m$. The non-magnetic ions create vacancies randomly distributed in the spin chain.

It is convenient to write the Hamiltonian (2.1) in terms of the spin creation and annihilation operators S_m^+ and S_m^-. At zero temperature and in the low-concentration

limit of non-magnetic ions widely separated, at the Néel ground state, all spins are aligned in the y direction, so we write

$$S_m^+ = S_m^z + iS_m^x \qquad S_m^- = S_m^z - iS_m^x \qquad (2.2)$$

which obey the commutation relations

$$[S_m^+, S_n^-] = 2\delta_{mn}S_m^y \qquad [S_m^y, S_n^+] = \delta_{mn}S_m^+ \qquad [S_m^y, S_n^-] = -\delta_{mn}S_m^-. \qquad (2.3)$$

Equation (2.1) may be expressed in terms of these operators, with the result

$$\mathcal{H} = 2J \sum_n \rho_n\rho_{n+1}[S_n^y S_{n+1}^y + \tfrac{1}{2}(1 - D/2)(S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+)$$

$$- \tfrac{1}{4}D(S_n^+ S_{n+1}^+ + S_n^- S_{n+1}^-)] + \tfrac{1}{2}i\mu gH \sum_n \rho_n(S_n^+ - S_n^-). \qquad (2.4)$$

For convenience we have defined operators that create excitations on vacancy sites as well as on the magnetic sites. The random variables ρ_n ensure that the non-physical excitations on the vacancies are decoupled from the true magnetic excitations of physical interest.

It is convenient to modify the Hamiltonian so that each term does not depend on the product of two random variables, $\rho_n\rho_{n+1}$. So we take the additive form for the Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 - \sum_l q_l \mathcal{H}_l \qquad (2.5)$$

with

$$\mathcal{H}_0 = \sum_n \{2J[S_n^y S_{n+1}^y + \tfrac{1}{2}(1 - D/2)(S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+)$$

$$- \tfrac{1}{4}D(S_n^+ S_{n+1}^+ + S_n^- S_{n+1}^-)] + (i\mu gH/2)(S_n^+ - S_n^-)\} \qquad (2.6)$$

and

$$\mathcal{H}_l = \sum_\Delta \{2J[S_l^y S_{l+\Delta}^y + \tfrac{1}{2}(1 - D/2)(S_l^+ S_{l+\Delta}^- + S_l^- S_{l+\Delta}^+)$$

$$- \tfrac{1}{4}D(S_l^+ S_{l+\Delta}^+ + S_l^- S_{l+\Delta}^-)] + (i\mu gH/2)(S_l^+ - S_l^-)\} \qquad (2.7)$$

where $\Delta = \pm 1$ and the q_l are random variables having the value 1 or 0 depending on whether or not the site l is occupied by a vacancy.

The Hamiltonians (2.4) and (2.5) are equivalent except when two or more vacancies are nearest neighbours of one another. Since magnetic properties depend only on the magnetic ions, the excitations will not be affected if we compute properly only the contributions of the magnetic sites. In our model there is no tendency for the formation of clusters of vacancies, so in the low-concentration limit pairs of vacancies are very rare.

3. Green functions

We study the excitations of the system by using the retarded and advanced Green functions (Zubarev 1960)

$$g_{mn}^r(t) = -i\theta(t)\langle[S_m^\alpha(t), S_n^\beta(0)]\rangle_0 \qquad (3.1a)$$

$$g_{mn}^a(t) = i\theta(-t)\langle[S_m^\alpha(t), S_n^\beta(0)]\rangle_0 \qquad (3.1b)$$

where the angular brackets with the 0 subscript denote the ground-state average.

We introduce the temporal Fourier transform

$$g_{mn}^{\mathrm{r,a}}(\omega) = \int_{-\infty}^{\infty} g_{mn}^{\mathrm{r,a}}(t)\, \mathrm{e}^{\mathrm{i}\omega t}. \tag{3.2}$$

So, if we make a cut along the real axis of the complex variable ω, under very general conditions, we can consider the functions $g_{mn}^{\mathrm{r}}(\omega)$ and $g_{mn}^{\mathrm{a}}(\omega)$ as a single analytic function defined as

$$g_{mn}(\omega) = \langle\!\langle S_m^\alpha : S_n^\beta \rangle\!\rangle_\omega = \begin{cases} g_{mn}^{\mathrm{r}}(\omega) & \operatorname{Im}\omega > 0 \\ g_{mn}^{\mathrm{a}}(\omega) & \operatorname{Im}\omega < 0. \end{cases} \tag{3.3}$$

The dynamic equation for this Green function is

$$\omega\langle\!\langle S_m^\alpha : S_n^\beta \rangle\!\rangle_\omega + \langle\!\langle [S_m^\alpha, \mathcal{H}] : S_n^\beta \rangle\!\rangle_\omega = \langle [S_m^\alpha, S_n^\beta] \rangle \tag{3.4}$$

and in the calculations we assume the standard decoupling of the second term on the left

$$\langle\!\langle S_l^y S_m^\alpha : S_n^\beta \rangle\!\rangle = \langle S_l^y \rangle \langle\!\langle S_m^\alpha : S_n^\beta \rangle\!\rangle \qquad \alpha(\beta) \neq y. \tag{3.5}$$

We also introduce the spatial Fourier transform

$$G(q,\omega) = \frac{1}{N} \sum_{m,n} \mathrm{e}^{\mathrm{i}q(m-n)}\, g_{mn}(\omega) \tag{3.6a}$$

$$G(q,\omega) = \sum_{m-n} \mathrm{e}^{\mathrm{i}q(m-n)} \langle g_{mn}(\omega) \rangle \qquad N \to \infty \tag{3.6b}$$

and its inverse

$$G_{mn}(\omega) = \frac{1}{N} \sum_q \mathrm{e}^{-\mathrm{i}q(m-n)}\, G(q,\omega) = \langle g_{mn}(\omega) \rangle \tag{3.7}$$

with

$$\langle g_{mn}(\omega) \rangle = \frac{1}{N} \sum_l g_{l+m-n,l}(\omega). \tag{3.8}$$

The Green function (3.7) depends only on the difference $(m-n)$, because the averaging process restores the translational symmetry.

The Green functions g and G contain features representing the spin excitations and spurious excitations on the vacancies. We require only the physical response $\rho_m \rho_n g_{mn}(\omega)$ on magnetic sites

$$\bar{G}(q,\omega) = \frac{1}{N} \sum_{m,n} \mathrm{e}^{\mathrm{i}q(m-n)}\, \rho_m \rho_n G_{mn}(\omega) \tag{3.9a}$$

$$\bar{G}(q,\omega) = \sum_{m-n} \mathrm{e}^{\mathrm{i}q(m-n)} \langle \rho_m \rho_n G_{mn}(\omega) \rangle \qquad N \to \infty. \tag{3.9b}$$

However, it is easier to work with $G(q,\omega)$ than with $\bar{G}(q,\omega)$. So we will make all calculations using $G(q,\omega)$ and then relate these to $\bar{G}(q,\omega)$ by the formal equation (Harris *et al* 1974)

$$\bar{G}(q,\omega) = [1 - L(q,\omega)] G(q,\omega) \tag{3.10}$$

where $L(q,\omega)$ is the vacancy projector, which we define after describing the calculation of $G(q,\omega)$.

As we are studying a two-sublattice model, it is convenient to introduce the four Green functions (Lines 1964)

$$g_{mn}^{11}(\omega) = \langle\!\langle S_m^+ : S_n^- \rangle\!\rangle_\omega \qquad g_{mn}^{12}(\omega) = \langle\!\langle S_m^+ : S_n^+ \rangle\!\rangle_\omega$$
$$g_{mn}^{21}(\omega) = \langle\!\langle S_m^- : S_n^- \rangle\!\rangle_\omega \qquad g_{mn}^{22}(\omega) = \langle\!\langle S_m^- : S_n^+ \rangle\!\rangle_\omega. \qquad (3.11)$$

Here $g_{mn}^{11}(\omega)$ and $g_{mn}^{22}(\omega)$ are non-zero only if the sites m and n belong to the same sublattice, while $g_{mn}^{12}(\omega)$ and $g_{mn}^{21}(\omega)$ are non-zero if the sites m and n belong to different sublattices.

4. The pure chain

We introduce the canonical transformation of the spin operators on the down sublattice defined as (Cowley and Buyers 1972)

$$S_m^+ \to S_m^- \qquad S_m^- \to S_m^+ \qquad S_m^y \to -S_m^y \qquad (4.1)$$

where m is a site of the down sublattice.

In the study of the pure chain we introduce the four Green functions

$$P_{mn}^{\alpha\beta}(\omega) = g_{mn}^{\alpha\beta}(\omega) = G_{mn}^{\alpha\beta}(\omega) \qquad (4.2)$$

with $\alpha(\beta) = 1$ and 2. The last equality in (4.2) comes from translational symmetry of the pure crystal.

From the dynamic equation (3.4) and from the standard decoupling procedure (3.5) we find the dynamic equations for the Green functions (4.2) as

$$\sum_k [(\omega - h^2/2\omega)I\delta_{mk} - D_{mk}]P_{kn}(\omega) = \delta_{mn}K \qquad (4.3)$$

where

$$I = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \qquad D = \begin{bmatrix} D^{11} & D^{12} \\ D^{21} & D^{22} \end{bmatrix} \qquad P = \begin{bmatrix} P^{11} & P^{12} \\ P^{21} & P^{22} \end{bmatrix} \qquad K = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \qquad (4.4)$$

and

$$D_{ij}^{11} = 2\delta_{ij} \qquad D_{ij}^{12} = 2(1 - D/2)\delta_{i\pm1,j} \qquad D_{ij}^{21} = -D_{ij}^{12} \qquad D_{ij}^{22} = -D_{ij}^{11} \qquad (4.5)$$

with a frequency scale $2JS = 1$ and $h = \mu gH/2JS$.

The equations (4.3) are solved by using spatial Fourier transform (3.6) to give

$$P^{\alpha\beta}(q, \omega) = \frac{1}{N} \sum_{m,m} e^{iq(m-n)} P_{mn}^{\alpha\beta}(\omega) \qquad (4.6a)$$

$$P^{\alpha\beta}(q, \omega) = \sum_{m-n} e^{iq(m-n)} P_{mn}^{\alpha\beta}(\omega). \qquad (4.6b)$$

So, we find

$$\begin{bmatrix} x(\omega) - 2 & a\cos q \\ -a\cos q & x(\omega) + 2 \end{bmatrix}\begin{bmatrix} P^{11}(q, \omega) & P^{12}(q, \omega) \\ P^{21}(q, \omega) & P^{22}(q, \omega) \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \qquad (4.7)$$

or

$$P(q, \omega) = \frac{1}{x^2(\omega) - 4 + a^2\cos^2 q}\begin{bmatrix} x(\omega) + 2 & -a\cos q \\ -a\cos q & -x(\omega) + 2 \end{bmatrix} \qquad (4.8)$$

with the lattice parameter $\lambda = 1$, and

$$x(\omega) = \omega - h^2/2\omega \qquad a = 2 - D. \tag{4.9}$$

The elementary magnetic excitations of this system are magnons given by the poles of the equation (4.8). So

$$x^2(\omega) - 4 + a^2 \cos^2 q = 0 \tag{4.10}$$

gives the elementary magnetic excitations of the pure chain.

5. The random diluted system

From the dynamic equation (3.4) and using the Hamiltonian (2.5), the canonical operator transformation (4.1) and the standard decoupling procedure (3.5), we derive a Dyson equation for the Green functions of the random diluted system

$$g_{mn}(\omega) = P_{mn}(\omega) + \sum_{k,j} P_{mk}(\omega)V_{kj}(\omega)g_{jn}(\omega) \tag{5.1}$$

with

$$g = \begin{bmatrix} g^{11} & g^{12} \\ g^{21} & g^{22} \end{bmatrix} \qquad V = \begin{bmatrix} V^{11} & V^{12} \\ V^{21} & V^{22} \end{bmatrix} \tag{5.2}$$

and

$$V = \sum_l q_l V_l \tag{5.3}$$

where V_l is the potential associated with a single vacancy on the site l and has the four blocks with non-null elements

$$V_l^{11}(\omega) = - \begin{bmatrix} 1 & 0 & 0 \\ 0 & 2+h^2/2\omega & 0 \\ 0 & 0 & 1 \end{bmatrix} \qquad V_l^{12}(\omega) = -\tfrac{1}{2}a \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix} \tag{5.4}$$

$$V_l^{21}(\omega) = V_l^{12}(\omega) \qquad\qquad V_l^{22}(\omega) = V_l^{11}(-\omega).$$

We introduce the scattering T matrix defined as

$$T_{mn}(\omega) = \sum_k V_{mk}(\omega)M_{kn}^{-1}(\omega) \tag{5.5}$$

with

$$M_{mn}(\omega) = \delta_{mn} - \sum_k P_{mk}(\omega)V_{kn}(\omega) \tag{5.6}$$

and rewrite equation (5.1) as

$$g_{mn}(\omega) = P_{mn}(\omega) + \sum_{k,j} P_{mk}(\omega)T_{kj}(\omega)g_{jn}(\omega). \tag{5.7}$$

We can also write formally a Dyson equation for the Green functions $G_{mn}(\omega)$, i.e.

$$G_{mn}(\omega) = P_{mn}(\omega) + \sum_{k,j} P_{mk}(\omega)\sigma_{kj}(\omega)G_{jn}(\omega) \qquad (5.8)$$

where

$$G = \begin{bmatrix} G^{11} & G^{12} \\ G^{21} & G^{22} \end{bmatrix} \qquad \sigma = \begin{bmatrix} \sigma^{11} & \sigma^{12} \\ \sigma^{21} & \sigma^{22} \end{bmatrix} \qquad (5.9)$$

and $\sigma(\omega)$ is the self-energy.

We see from (5.3) that the problem of a single vacancy localised on the site l is tractable. The scattering T matrix is now given by

$$T_{l;mn}(\omega) = \sum_{k} V_{l;mk}(\omega)M_{l;kn}^{-1}(\omega) \qquad (5.10)$$

with

$$M_{l;mn}(\omega) = \delta_{mn} - \sum_{k} P_{mk}(\omega)V_{l;kn}(\omega). \qquad (5.11)$$

The T_l matrix has non-null elements only in four blocks, similarly to the blocks of V_l defined in (5.4). The presence of g on the right-hand side of equation (5.1) shows that the exact solution includes multiple scattering of the spin excitations by a single defect.

From the configurational average of equation (5.7) and from equation (5.8) we find

$$\sigma = (I + \langle T \rangle P)^{-1}\langle T \rangle \qquad (5.12)$$

where $\langle T \rangle$ is the configurational average of the scattering T matrix.

In order to find the configurational average of the scattering T matrix, we first write

$$T = V(1 + PT) = \sum_{l} Q_l \qquad (5.13)$$

where

$$Q_l = q_l V_l(1 + PT) = T_l\left(1 + P \sum_{m \neq l} Q_m\right). \qquad (5.14)$$

To eliminate the restriction $m \neq l$ in (5.14), we introduce the operator $\bar{P}$ defined as

$$\bar{P}_{mn}^{\alpha\beta} = P_{mn}^{\alpha\beta} \qquad \text{if} \quad m(n) \neq l-1, l, l+1$$
$$\bar{P}_{mn}^{\alpha\beta} = 0 \qquad \text{if} \quad m(n) = l-1, l, l+1 \qquad (5.15)$$

and $\alpha(\beta) = 1.2$. From (5.14) and (5.15) we derive

$$T = \sum_{l} T_l(1 + \bar{P}T). \qquad (5.16)$$

Let us make the approximation (average T-matrix approximation (ATA))

$$\left\langle \sum_{l} T_l T \right\rangle = \left\langle \sum_{l} T_l \right\rangle \langle T \rangle = (1 - c)\langle T_l \rangle\langle T \rangle \qquad (5.17)$$

to find

$$\langle T \rangle = [I - (1 - c)\langle T_l \rangle \bar{P}]^{-1}(1 - c)\langle T_l \rangle. \qquad (5.18)$$

From (5.12) and (5.18) we derive

$$\sigma = [I - (1 - c)\langle T_l \rangle P_l]^{-1}(1 - c)\langle T_l \rangle. \tag{5.19}$$

So in the low-concentration limit $(1 - c) \ll 1$, we have

$$\sigma = (1 - c)\langle T_l \rangle \tag{5.20}$$

where $\langle T_l \rangle$ is the configurational average of the scattering matrix for a single localised vacancy.

The Green functions $G_{mn}(\omega)$ have translational symmetry (3.7), then the spatial Fourier transform of (5.8) is

$$G(q, \omega) = P(q, \omega) + P(q, \omega)\sigma(q, \omega)G(q, \omega) \tag{5.21}$$

where

$$G^{\alpha\beta}(q, \omega) = \frac{1}{N} \sum_{m,n} G^{\alpha\beta}_{mn}(\omega)\, e^{iq(m-n)}$$

$$\sigma^{\alpha\beta}(q, \omega) = (1 - c) \sum_{m,n} T^{\alpha\beta}_{l;mn}(\omega)\, e^{iq(m-n)}. \tag{5.22}$$

$T^{\alpha\beta}_{l;mn}$ has non-null elements for $m(n) = l - 1, l, l + 1$, where l is a site occupied by a vacancy, and $\alpha(\beta) = 1, 2$.

From (5.21) we find

$$G(q, \omega) = [I - P(q, \omega)\sigma(q, \omega)]^{-1}P(q, \omega). \tag{5.23}$$

In order to find the vacancy projector $L(q, \omega)$, we introduce the restricted spatial Fourier transform (Cowley and Buyers 1972)

$$\underline{G}(q, \omega) = \frac{1}{N} \sum_{l,n} q_l g_{ln}(\omega)\, e^{iq(l-n)}$$

$$\underline{V}(q, \omega) = \sum_{k} V_{l;kl}(\omega)\, e^{iq(k-l)}$$

$$\underline{G}_\Delta(q, \omega) = \frac{1}{N} \sum_{l,n} q_l g_{l+\Delta,n}(\omega)\, e^{iq[(l+\Delta)-n]} \tag{5.24}$$

$$\underline{V}_\Delta(q, \omega) = \sum_{k} V_{l;k,l+\Delta}(\omega)\, e^{iq[k-(l+\Delta)]}$$

where $\Delta = \pm 1$. So the spatial Fourier transform of equation (5.1) is

$$G(q, \omega) = P(q, \omega) + P(q, \omega)\underline{V}(q, \omega)\underline{G}(q, \omega)$$

$$+ \sum_\Delta P(q, \omega)\underline{V}_\Delta(q, \omega)\underline{G}_\Delta(q, \omega). \tag{5.25}$$

From equations (5.21) and (5.25) we find

$$\sigma(q, \omega)G(q, \omega) = \underline{V}(q, \omega)\underline{G}(q, \omega) + \sum_\Delta \underline{V}_\Delta(q, \omega)\underline{G}_\Delta(q, \omega) \tag{5.26}$$

from which we derive

$$\underline{G}(q, \omega) = (1 - c)\overline{M}(q, \omega)G(q, \omega) = L(q, \omega)G(q, \omega) \tag{5.27}$$

with

$$\overline{M}(q, \omega) = \sum_{l-n} M^{-1}_{l;ln}(\omega)\, e^{iq(l-n)}.$$

Then the physical Green function is

$$\overline{G}(q, \omega) = G(q, \omega) - \underline{G}(q, \omega) = [I - L(q, \omega)]G(q, \omega). \tag{5.28}$$

The poles of the physical Green function (5.28) give the magnetic elementary excitations of the random diluted system. Equations (5.28), (5.27), (5.24) and (5.11) show that the excitations associated with the defects, created by the vacancies, are the solutions of the determinant equation

$$\det(I - PV_l) = 0. \tag{5.29}$$

We can factorise this equation to find

$$x(\omega) - \tfrac{1}{2}[1 - \tfrac{1}{4}(1 - b)^2]^{1/2}\{[9 - \tfrac{1}{4}(1 - b)^2]^{1/2} - [1 - \tfrac{1}{4}(1 - b)^2]^{1/2}\} = 0 \tag{5.30}$$

with the anisotropy parameter

$$b = 1 - D. \tag{5.31}$$

The physical quantity of experimental interest is the scattering function, which is directly related to the scattering cross section in a scattering experiment. This quantity is related to the Greeen function at zero temperature by (Lovesey 1980, Cowley and Buyers 1972)

$$S(q, \omega) = \text{Im}[\overline{G}^{11}(q, \omega) \pm \overline{G}^{12}(q, \omega) + \overline{G}^{22}(q, \omega) \pm \overline{G}^{21}(q, \omega)] \tag{5.32}$$

where the upper signs refer to nuclear reciprocal lattice points and the lower signs refer to magnetic reciprocal lattice points.

Substituting (3.10) in (5.33) we obtain

$$S(q, \omega) = \text{Im}\{[1 - L^{11}(q, \omega) - L^{21}(q, \omega)][G^{11}(q, \omega) \pm G^{12}(q, \omega)]$$
$$+ [1 - L^{12}(q, \omega) - L^{22}(q, \omega)][G^{22}(q, \omega) \pm G^{21}(q, \omega)]\}, \tag{5.33}$$

Using the well known Dirac formula

$$\frac{1}{X \pm i0^+} = \frac{1}{X} \mp i\pi\delta(X) \tag{5.34}$$

and (5.6), (5.23) and (5.27), we see that (5.33) is a linear combination of delta functions with arguments given by (4.10) and (5.30).

So our theory gives a zero halfwidth at the half-maximum of the scattering function, which is consistent with the approach used in this work (Birgenau and Shirane 1978). The non-zero halfwidths at the half-maximum of the scattering function in the works of Harris *et al* (1974), Holcomb (1974) and Endoh *et al* (1981) are introduced *ad hoc*, considering a non-null small imaginary part in the frequencies of the scattering function.

Consequently, equations (4.10) and (5.30), and equation (5.33), give the same information about the dynamics of the system.

6. Results and discussion

We discuss the roles played by *XY* anisotropy and by applied magnetic field perpendicular to the chain, and the effect of both on the localisation of the resonances
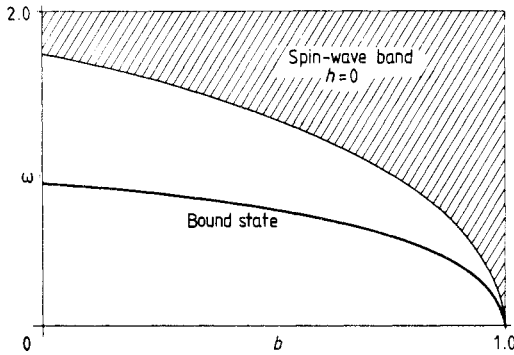
Figure 1. Frequency ω (in units of $2JS$) versus the XY anisotropy parameter b, showing how the resonance associated with the defect is pushed to zero frequency as the model is changed from XY model to isotropic Heisenberg model, as the b parameter is increased from 0 to 1, with no applied magnetic field.
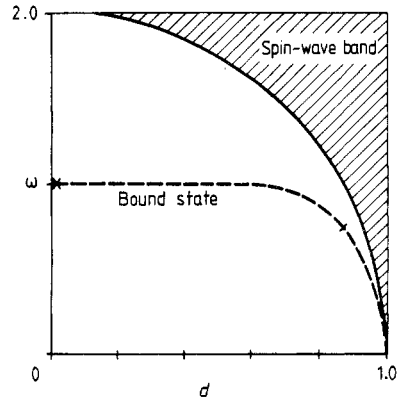
Figure 2. Frequency ω (in units of $2JS$) versus the Ising anisotropy parameter d, with no applied magnetic field, showing how the resonance associated with the defect is pushed to zero frequency as the model is changed from Ising model to isotropic Heisenberg chain, as the d parameter is increased from 0 to 1. (After Endoh *et al* 1981.)

associated with excitations on end spins of the random one-dimensional diluted Heisenberg antiferromagnetic chain.

We show in figure 1 the role played by XY anisotropy. The superior and inferior limits of the energy band are given by equation (4.10) with $h = 0$, and $q = \pi/2$ and $q = 0$, respectively. The excitations on spin chain ends are given by equation (5.30). We see that, in the Heisenberg chain, $b = 1$, there are no separated resonances associated with excitations on end spins. This is explained in terms of the singular nature of the density of states in one dimension at the band edges (Endoh *et al* 1981). When the anisotropy parameter decreases from $b = 1$ to $b = 0$, i.e. the model is changed from Heisenberg to XY model, the resonances associated with excitations on end spins separate from the spin-wave band. At $b = 0$, i.e. XY model, the resonance associated with excitations on end spins occurs at a frequency equal to one-half of the inferior limit of the spin-wave band frequencies.

In figure 2 we show results of a similar study of Endoh *et al* (1981), who show that, when the Ising anisotropy parameter d changes from $d = 1$ to $d = 0$, i.e. the model is changed from Heisenberg to Ising, the frequencies of the resonances associated with excitations on end spins are different from the spin-wave band frequencies.

This part of our work complements the work of Endoh *et al* (1981) and we can conclude that in the Heisenberg model the anisotropy, independent of its type, separates the frequencies of the resonances associated with excitations on spin chain ends from the spin-wave band frequencies.

Thermodynamically, the Heisenberg chains with an applied magnetic field perpendicular to the chain or with an XY anisotropy with $h = 2[2(1 - b)]^{1/2}$, where h is the scaled magnetic field ($h = \mu g H/2JS$) and b is the XY anisotropy parameter ($b = 1 - D$), are equivalent (Pires and Gouvêa 1984). In figure 3 we show plots of the spin-wave band with $b = 1$ (Heisenberg model), and of resonances associated with excitations on end spins, equation (5.30). We see that the resonances associated with excitations on end
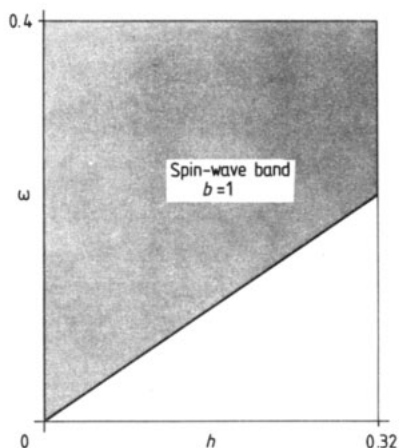
Figure 3. Frequency ω (in units of $2JS$) versus the scaled applied magnetic field h ($h = \mu gH/2JS$), for the isotropic Heisenberg chain.
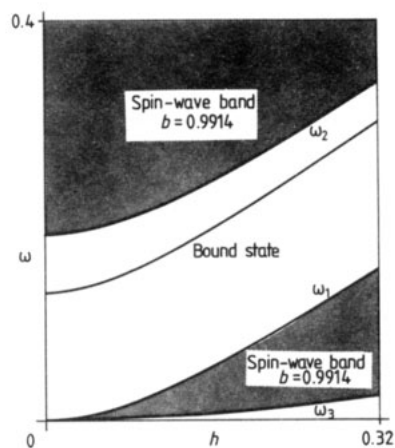

Figure 4. Frequency ω (in units of $2JS$) versus the scaled applied magnetic field h ($h = \mu gh/2JS$), with XY anisotropy parameter $b = 0.9914$.

spins do not occur at frequencies outside the spin-wave band, even for fields as great as 70 kOe ($h \simeq 0.32$). Then, differently from XY anisotropy, an applied magnetic field perpendicular to the chain does not localise the resonances associated with excitations on end spins.

An Ising model and a Heisenberg model with an XY anisotropy and an applied magnetic field perpendicular to the chain, with $h = 2[2(1-b)]^{1/2}$, are thermodynamically equivalent (Pires and Gouvêa 1984). As we are interested particularly in TMMC:Cu, in figure 4 we plot equations (4.10) and (5.30) with $b = 0.9914$, which is the XY anisotropy parameter of TMMC:Cu (Heilmann *et al* 1981). First, we see a split of the spin-wave band, which comes from the out-of-plane spin contributions. Secondly, for $b = 0.9914$ and $h = 0.26$, the Heisenberg model does not have the same behaviour as the Ising model.

Then we may conclude that the thermodynamic equivalence between an XY anisotropy and an applied magnetic field perpendicular to the chain, and between the Ising model and the Heisenberg model with an XY anisotropy and an applied magnetic field perpendicular to the chain, are not preserved on the dynamics of the random one-dimensional diluted antiferromagnets.

Acknowledgments

Appendix

In the calculations using equation (5.30) we have to know the Green functions $P_{mn}^{\alpha\beta}(\omega)$, α (β) = 1, 2, which have translational symmetry, whence $P_{mn}^{\alpha\beta}(\omega) = P_{|m-n|}^{\alpha\beta}(\omega)$, and we need to evaluate $P_0^{\alpha\alpha}(\omega)$, $P_2^{\alpha\alpha}(\omega)$ and $P_1^{\alpha\beta}(\omega)$.

These Green functions are given by equation (3.8) as

$$P_{mn}^{\alpha\beta}(\omega) = \frac{1}{N} \sum_q e^{-iq(m-n)} P^{\alpha\beta}(q, \omega) \tag{A.1}$$

which for an infinite chain can be approximated by the integral over the first Brillouin zone

$$P_{mn}^{\alpha\beta}(\omega) = \frac{1}{2\pi} \int_{-\pi}^{\pi} e^{-iq(m-n)} P^{\alpha\beta}(q, \omega)\, dq. \tag{A.2}$$

In the evaluation of the integrals (A.2) we make use of the residues method to find

$$P_0^{11}(\omega) = x + 2/(A^2 + AB)^{1/2}$$
$$P_2^{11}(\omega) = [x + 2/B(A^2 + AB)^{1/2}][-(2A + B) + 2(A^2 + AB)^{1/2}] \quad \text{(A.3)}$$
$$P_1^{12}(\omega) = [2/a(A^2 + AB)^{1/2}][A - (A^2 + AB)^{1/2}]$$

with

$$A = x^2 - 4 \qquad\qquad B = a^2 \tag{A.4}$$

$$P^{22}(\omega) = P^{11}(-\omega) \qquad P^{21}(\omega) = P^{12}(\omega). \tag{A.5}$$

When A is real and $|A| \le B$ the integrals (A.2) are not well defined; following the standard procedure (Wolfram and Callaway 1963) we add to the frequency a small, positive, imaginary part.

As we are interested only in the localised impurity states, we take in our calculations ω real and $|A| > B$.

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