Low-temperature excitations in random two-dimensional antiferromagnets†

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Abstract. A numerical method involving the time integration of equations of motion is developed for calculating response functions of antiferromagnets. The method is particularly useful in calculating dynamic response functions $S(k, \omega)$ and can also be used to obtain densities of states. The technique is applied to the two-dimensional random antiferromagnets Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$ and Rb$_2$Mn$_{0.5}$Zn$_{0.5}$F$_4$. In the former case, we find good agreement with the recently measured $S(k, \omega)$ and in the latter we find some interesting clustering effects that should be accessible experimentally.

1. Introduction

The problem of excitations in alloys has been extensively studied in recent years and considerable progress has been made in studying model systems. These efforts have been in two principal directions. The first centres around the coherent potential approximation (CPA) (Elliott et al 1974). This was originally a simple analytic technique for dealing with diagonal disorder in a single-site approximation. Subsequent efforts to extend the CPA to include off-diagonal disorder and to treat clusters of sites, have largely removed the algebraic simplicity. These extensions of the CPA involve solving a set of self-consistent equations numerically and it is difficult to obtain the same kind of insight that was possible with the original CPA.

Other efforts have been entirely numerical and essentially involve techniques for handling large matrices. Some of the earliest and most extensive work to date has been done by Bell (1972) and Dean (1972) who exploit the ‘negative eigenvalue theorem’. This allows the number of eigenstates with an energy less than some specified energy to be found. By changing the specified energy, a histogram can be constructed. The method is very attractive for banded matrices as found in one-dimensional systems. It can also be used in higher dimensions and indeed has been used by Huber (1974) and by Kirkpatrick and Harris (1975) to calculate the density of states in Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$. The method can also give sample eigenvectors but cannot yield, in any obvious way, dynamical response functions like the neutron scattering law $S(k, \omega)$.

In this paper we describe a numerical method involving the time integration of the equations of motion for the system. The method is extremely efficient and easy to apply

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and allows quantities like $S(k, \omega)$ to be obtained easily and naturally. The method can also be used to obtain densities of states but is somewhat less efficient for reasons that are explained in the next section.

Although these methods are in no way restricted or more efficient in lower dimensions, we have nevertheless decided to apply them to two-dimensional antiferromagnets. Magnetic insulators are ideal alloy systems to study because the interactions can be described by simple spin Hamiltonians with very short-range interactions (De Jongh and Miedema 1974, Cowley and Buyers 1972). This is particularly true of the transition-metal compounds like MnF$_2$, KNiF$_3$, Rb$_2$MnF$_4$, etc. We have chosen to study the two-dimensional magnetic alloys Rb$_2$Mn$_{1-c}$Ni$_c$F$_4$ and Rb$_2$Mn$_{1-c}$Zn$_c$F$_4$. The neutron scattering law $S(k, \omega)$ has recently been determined for the former system (with $c = 0.5$) which has two kinds of magnetic ions (Birgeneau et al 1975, Als-Nielson et al 1975). The other system was chosen because it is a dilute antiferromagnet (the Zn$^{2+}$ ion is non-magnetic) and because there is a possibility that some experimental results will be available in the near future (Birgeneau 1975, private communication). Two-dimensional systems were chosen because we expect the effects of clustering to be more apparent in the spectral functions. It is clear from the work of Bell (1972) and Dean (1972) that clustering effects are more pronounced in lower dimensional systems and we elaborate on this in §4.

The layout of the paper is as follows. In the next section, we discuss the equation of motion method and how it can be used to determine dynamic response functions and densities of states. In §3 we apply this to Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$ and in §4 to Rb$_2$Mn$_{0.5}$Zn$_{0.5}$F$_4$. Our results (Alben and Thorpe 1975a (a preliminary account of this work with application to Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$) and b (an account of this work applied to Rb$_2$Mn$_{0.5}$Zn$_{0.5}$F$_4$)) show very good agreement with experiment in the case of Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$ where $S(k, \omega)$ has been determined. We also find spectral features that can be attributed to specific clusters of atoms. However, the experimental energy resolution would have to be increased by a factor of about 3 in order to see these effects. We find that these effects are much more pronounced in the dilute antiferromagnet Rb$_2$Mn$_{0.5}$Zn$_{0.5}$F$_4$ and it should be possible to see them experimentally. Whereas the spectrum of Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$ is always continuous, the spectrum of Rb$_2$Mn$_{0.5}$Zn$_{0.5}$F$_4$ has some rather prominent spikes that arise from isolated islands of Mn spins.

2. The equation of motion method

We consider a simple two-sublattice antiferromagnet with nearest-neighbour exchange $J$ and uniaxial anisotropy $H^A$ such that the system is described by a Hamiltonian $H$ where

$$ H = \sum_{\langle i,j \rangle} J_{ij} S_i \cdot S_j - \sum_{i} (g_i \mu_B H^A_i) \sigma_i S_i^z $$

(1)

and $\sigma_i$ is a variable that is +1 for the up sublattice and −1 for the down sublattice. The factors $g_i$ and $\mu_B$ (the Bohr magneton) have been included in the anisotropy so that $g_i \mu_B H^A_i$ is the anisotropy energy if $H^A_i$ is in magnetic field units. In the general case we have two kinds of magnetic ions distributed at random on the two sublattices, one having a concentration $c$ and the other $1 - c$. The nearest-neighbour exchange can thus take on three values and we shall allow the anisotropy to take on two values appropriate to the two kinds of magnetic sites. This is justified if the anisotropy is mainly of a single-
ion kind—it is not justified if it arises from long-range dipolar interactions. Being a numerical technique, the equation of motion method is not restricted to such a simple Hamiltonian as (1). However, as this Hamiltonian is adequate for the systems we are concerned with in this paper, we will stay with it.

It is convenient to define a Green function $G_\mu(t)$ (Alben and Thorpe 1975a) by

$$G_\mu(t) = \frac{\langle 0| S_i^+ (t) S_i^- (0) - S_i^- (0) S_i^+ (t) |0 \rangle}{2(S_i S_i)^{1/2}}.$$  

(2)

The magnitude of the spin $S_i$ is positive and can take on two values, depending on which magnetic species occupies the site $i$. The factor $2(S_i S_i)^{1/2}$ is included so that at time $t = 0$ we have (with $h = 1$)

$$G_\mu(t = 0) = \frac{\langle 0| S_i^0 |0 \rangle}{S_i} \delta_{\mu \nu} = \sigma_i \delta_{\mu \nu}$$

(3)

where the expectation value is taken in the Néel state since the ground state of the antiferromagnet is not known and $\sigma_i = \pm 1$ depending on whether site $i$ belongs to the up or down sublattice. This approximation corresponds to that of linear spin-wave theory and is good for large values of the spin (Keller 1966). This same approximation is made in linearizing the equations of motion and replacing quantities like $S_i^+ S_j^+$ by $S_i^+ S_j \sigma_j$ where $i$ and $j$ are on different sites, i.e.:

$$i\partial G_\mu(t)/\partial t = \sum_j J_{ij} S_i \sigma_i G_\mu(t) - \sum_j J_{ij} \sigma_i G_\mu(t) + (g_\mu \mu_B H_0^i) \sigma_i G_\mu(t).$$

(4)

Equation (4) together with the initial conditions (3) may be integrated forward in time starting at $t = 0$ if the derivative is taken numerically, i.e.

$$\partial G_\mu(t)/\partial t \approx [G_\mu(t + \Delta/2) - G_\mu(t - \Delta/2)]/\Delta.$$  

(5)

Thus, knowing the Green function at $t - \Delta/2$ and $t$ it can be computed at $t + \Delta/2$ and so on. Strictly speaking, it is only necessary to know the Green function at one previous time, but it is more symmetrical to write it in the form (5) which only involves local truncation errors $O(\Delta^2)$ rather than $O(\Delta)$. The problem of having a sufficient number of points at the start is overcome by making a Taylor series expansion for $G$ about $t = 0$; the necessary derivatives being evaluated from equation (4). In our case, we have demanded that the start also be correct to $O(\Delta^2)$ and so have used $G(t = 0)$ and $\partial G(t = 0)/\partial t$ to evaluate $G(\Delta)$ with error $O(\Delta^2)$.

More elaborate numerical integration schemes could be used, but we have found no problems with the one outlined here. The spectra that we are interested in are bounded and so there is a maximum frequency $\omega_{\text{max}}$. This determines the smallest time period of the normal modes $(2\pi/\omega_{\text{max}})$ and so we chosen $\Delta = 1/n(2\pi/\omega_{\text{max}})$ where $n = 15$ gave sufficient accuracy for most purposes although $n = 20$ was used in some cases.

Having obtained the $G_\mu(t)$ we are now in a position to compute the quantities of interest that are Fourier transforms of $G_\mu(t)$. The neutron scattering law $S(k, \omega)$ may be defined from Fermi’s golden rule (apart from pre-factors that depend on phase space and the geometry of the experiment (Marshall and Lovesey 1971)) as

$$S(k, \omega) = 2\pi \sum_{i} \sum_{\nu} \exp(i k \cdot R_i) g_{\nu} S_i^\nu |1> |^2 \delta(\omega - \omega_i)$$

(6)

where the neutron polarization is taken to be in the $x$ direction which is any direction perpendicular to the spin-ordering direction $z$. Notice that the neutrons respond to the magnetization rather than the spin. With a little manipulation we can express $S(k, \omega)$ as
Fourier transform of the Green function

\[
S(k, \omega) = \int_{-\infty}^{\infty} \exp(\mathrm{i} \omega t) \mathrm{d}t \sum_{ij} \exp(\mathrm{i} k (R_i - R_j) g_{ij} S_i^+ S_j^0) \langle 0 | S_i^+ (t) S_j^+(0) | 0 \rangle
\]

\[
= \frac{1}{2} \int_{-\infty}^{+\infty} \exp(\mathrm{i} \omega t) \mathrm{d}t \sum_{ij} \exp(\mathrm{i} k (R_i - R_j) g_{ij} S_i^+ (t) S_j^+(0) + S_i^-(t) S_j^+(0)) . \tag{7}
\]

The neutron-scattering law (6) or (7) gives no scattering at negative frequencies. Using this, the result can be expressed in the form we want it:

\[
S(k, \omega) = -2 \int_{0}^{\infty} \sin \omega t \mathrm{d}t \mathrm{Im} \left( \sum_{ij} \exp(\mathrm{i} k (R_i - R_j) g_{ij} S_i S_j) \right)^{1/2} G_{ij}(t) \right). \tag{8}
\]

Note that a misprinted form of this equation was reported in a previous account of this work (Alben and Thorpe 1975a).

The density of states can be expressed in terms of the \( G_{ij}(t) \) in a similar way. The equation of motion for the Green function

\[
G_{ij}(\omega) = \int_{0}^{\infty} \exp(\mathrm{i} \omega t) G_{ij}(t) \mathrm{d}t \tag{9}
\]

is given (using (4)) by

\[
\sum_{\delta_{ij}(\omega - \sum_{m} \sigma_{m} S_{m} - \sigma_{ij} H_{ij}^{\alpha}) + J_{ij} (S_i S_j)^{1/2} \right] G_{ij}(\omega) = i G_{ij}(t = 0) = i \sigma_{ij} \delta_{ij}. \tag{10}
\]

This has the form

\[
M G = i \sigma \tag{11}
\]

where the eigenvalues of the matrix \( M \) are the eigenvalues of the system (ii can be shown that these are real for the Hamiltonian (1) despite the fact that \( M \) is not symmetric). Because the system is an antiferromagnet, one half of these eigenvalues are positive and one half negative. We can define a local density of states as

\[
\rho_{l}(\omega) = -\frac{1}{2\pi} \mathrm{Im}(M^{-1}(\omega) + M^{-1}(-\omega))_{ii} \tag{12}
\]

where the frequency has a small positive imaginary part. From equations (11) and (12) we have

\[
\rho_{l}(\omega) = -\frac{1}{2\pi} \mathrm{Im}(i \sigma_{ij} G_{ij}(\omega) + i \sigma_{ij} G_{ij}(-\omega)) \]

\[
= \frac{1}{2\pi} \mathrm{Re}(\sigma_{ij} G_{ij}(\omega) + \sigma_{ij} G_{ij}(-\omega)) \tag{13}
\]

\[
= \frac{\sigma_{ij}}{\pi} \int_{0}^{\infty} \cos \omega t \mathrm{Re} G_{ij}(t).
\]

The two expressions (8) and (13) are now in an appropriate form for computing if the upper integration limit \( \infty \) is replaced by \( T \) and a damping function is included in the integral. We have used \( \exp(-\lambda t) \) as the damping function. After performing the Fourier transforms as required by either (8) or (13), we get Lorentzian peaks (with some residual termination ripples) centred at each \( \omega = \pm \omega_{e} \) where \( \omega_{e} \) are the normal-mode fre-
Random two-dimensional antiferromagnets

The width of the peak is $\Delta \omega \sim 2\pi/T$. The truncation time $T$ is chosen to give the desired resolution which, for example, in figure 1 is $\Delta \omega \approx (1/20) \omega_{\text{max}}$ so that $T \approx 20 \times 15 \times \Delta \approx 300 \Delta$. It is thus only necessary to integrate for $\sim 300$ time steps before truncating and doing the Fourier transform. This is much more efficient than conventional molecular dynamics that requires many thousands of time steps (Rahman 1964). The reason for the increased efficiency is because of the harmonicity that leads to a bounded spectrum with a maximum frequency $\omega_{\text{max}}$. In molecular dynamics, the system is not harmonic and the frequency spectrum has a high-frequency tail.

The method of computation is now clear. For the neutron-scattering law $S(k, \omega)$ it is:

(i) Excite all atoms with an amplitude $g_i(S_i\sigma_i^{1/2}) \exp(ik \cdot R_i)$.

(ii) Let the system evolve with time via equation (4) and weight the amplitude at time $t$ with a factor $g_i(S_i^{1/2}) \exp(-ik \cdot R_i)$.

(iii) Do the sine Fourier transform on the imaginary part of the amplitude.
These factors are all clear from equation (8)—the $\sigma_i$ in (1) comes from the initial condition on the Green function (3).

The analogous computation for the local density of states (13) is:

(i) Excite atom $i$ only with amplitude 1.
(ii) Let the system evolve with time via equation (4) and weight the amplitude at time $t$ with a factor 1 for the $i$ atom only.
(iii) Do the cosine Fourier transform on the real part of the amplitude.

Note that, in this case, the $\sigma_i$ in equation (13) gets absorbed with the initial condition (4) to give a net amplitude of $+1$.

We see that $S(k, \omega)$ for a single $k$ vector and $\rho_i(\omega)$ for a single atom $i$ can be obtained with one computation of the kind described above. Unfortunately, one usually wants the density of states averaged over a number of sites and this would require a large number of independent computations. This difficulty can be overcome by exciting a large number of degrees of freedom with a random phase and weighing the amplitudes at time $t$ with the same random phase. The cross terms between different random phases should average to zero so that one obtains a density of states as a random weighting over local densities of states of the kind (13).

3. Application to Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$

We have applied the ideas of §2 to Rb$_2$Mn$_{0.5}$Ni$_{0.5}$F$_4$ because experimental results have recently become available (Birgeneau et al 1975, Als-Nielson et al 1975) and the system can be described by a simple Hamiltonian of the form (1). The pure materials Rb$_2$MnF$_4$ and Rb$_2$NiF$_4$ are known to be magnetic insulators whose properties are dominated by near-neighbour Heisenberg interactions within the two-dimensional planes (Birgeneau et al 1970, De Wijn et al 1971, 1973). The magnitude of the nearest-neighbour exchange $J_{1\rho}$ anisotropy field $g_{\mu B}H_1^\rho$ and $g$ factor are known to be 0.655 meV, 0.03 meV and 2 for the Mn salt (spin 5/2) and 8.3 meV, 0.28 meV and 2.24 for the Ni salt (spin 1) (Birgeneau et al 1970, 1975, De Wijn et al 1973). Because the lattice constants of these two compounds differ by only 3% it is not unreasonable to assume that the above parameters do not change appreciably in the mixed crystal. The anisotropy $H_1^\rho$ in Rb$_2$NiF$_4$ originates from local crystal fields and so should not be sensitive to composition. This is less true in Rb$_2$MnF$_4$ where the anisotropy is smaller and of a long-range dipole kind, which will be sensitive to composition. Inasmuch as the anisotropy is small, we ignore this effect—it is only important at the very lowest frequencies.

We assume, after Birgeneau et al (1975) that the Mn–Ni exchange is the geometric mean of the Mn–Mn and Ni–Ni exchanges and is equal to 2.33 meV. This is known to be a good approximation in the cubic perovskites and we will show that it leads to good results here.

The Brillouin zone for the square net is also a square net. If the nearest-neighbour separation is $a$, then the nuclear zone is a square of side $2\pi/a$ and the magnetic zone is a square of side $\sqrt{2}(\pi/a)$, but rotated by 90° with respect to the nuclear zone. We have computed $S(k, \omega)$ along the (1, 0) and (1, 1) directions where (1, 0) goes to the centre of a side and (1, 1) goes to a corner in the magnetic zone. Although $k$ is not a good quantum number for the alloy because of the lack of microscopic translational invariance, nevertheless $S(k, \omega)$ does repeat over the nuclear zone. This is because the wavefunction is only defined at the atomic sites where the spins $S_i$ are localized.
We have computer-generated 40 × 40 square nets of Mn and Ni spins where the probability of any site being occupied by a Mn atom is 0.5. The equations of motion were integrated forward in time and the appropriate Fourier transforms performed as described in the §2. In figures 1 and 2 we show S(k, ω) for k along the (1, 0) and (1, 1) directions where the parameters λ, Δ and T have been chosen to give an energy resolution of 1.8 meV.

![Diagram](image)

Figure 2. As for figure 1 but in the (1, 1) direction with k = (n/a)√2(K, K). (a) K = 0, I = 0.081; (b) K = 0.1, I = 0.198; (c) K = 0.2, I = 0.385; (d) K = 0.3, I = 0.558; (e) K = 0.4, I = 0.777; (f) K = 0.5, I = 0.888; (g) K = 0.6, I = 1.04; (h) K = 0.7, I = 1.12; (i) K = 0.8, I = 1.31; (j) K = 0.9, I = 1.39; (k) K = 1.0, I = 1.42.

The ripples apparent in a few of these spectra are due to the truncation of the Fourier transforms at time T. They are most pronounced in figure 2(c) (K = 0.2). It is a straightforward process to eliminate these by increasing the damping factor and we have done this in some cases (e.g. figure 2(b) (K = 0.1)) when the low-frequency peak becomes very narrow and dominates the spectrum. In figure 3 we show the ‘dispersion relations’ ω against k derived from the spectra in figures 1 and 2 and also the intensity in the two peaks. Because of the large difference between the Mn–Mn and Ni–Ni exchange interactions, the spectra separate into a low-frequency part that is essentially the Mn response and a higher-frequency part that is mainly the Ni response. The Mn branch is quite narrow while the Ni branch is much broader. Similar results have been achieved by
Huber (1974) and by Kirkpatrick and Harris (1975). It would be wrong to interpret this as showing that the Mn modes are more localized (Kirkpatrick and Harris 1975). Although the dispersion relations appear symmetric about (0.5, 0) in figure 3, as we go from a nuclear reciprocal point to a magnetic reciprocal lattice point, the intensity increases continuously as we approach the magnetic reciprocal lattice point. This also happens in the pure system. The upper part of figure 4 shows a comparison between theory and experiment (Birgeneau et al 1975, Als-Nielson 1975) for $K = 0.5$ in figure 1(f).

**Figure 3.** Results of calculations of the neutron scattering for $k$ along two directions in the Brillouin zone calculated for $40 \times 40$ nets on Rb$_3$Mn$_{0.73}$Ni$_{0.27}$F$_4$ at a resolution of 18 MeV (from figures 1 and 2). (a) shows the 'dispersion relations' with the positions of the peaks denoted by: ■ upper peak; ▲ lower peak; ● experimental results; | halfwidths. (b) shows the integrated intensities in the two peaks. The total integrated intensity as a function of $k$ is very similar to that of a pure antiferromagnet.

**Figure 4.** The neutron scattering law $S(k, o)$ for $k$ at the zone boundary in the (1, 0) direction. In (a), the theory (for a $100 \times 100$ net with a resolution of 18 MeV) is compared with experiment (Birgeneau et al 1975, Als-Nielson et al 1975 (broken line) on Rb$_3$Mn$_{0.73}$Ni$_{0.27}$F$_4$ which has a similar resolution. (b) shows the same result with a resolution of 0.6 MeV. The full curve is the average of the results, shown in the insert, for two different random configurations. The vertical lines give predictions from an Ising model for the Ni modes.
This corresponds to a point on the magnetic zone boundary. Note that, whereas the shoulder seen experimentally around 22 meV is not evident in the higher-resolution calculations with 100 × 100 nets, it does appear for the 40 × 40 net in figure 1(f) (K = 0.5).

We obviously have more confidence in our results for larger nets. The shoulder is probably just a variation of the kind to be expected in a small sample and is not a real feature as it did not occur in either of the 100 × 100 nets. This shoulder was not found by Kirkpatrick and Harris (1975) in their calculation of S(κ, ω) which is a numerical method involving matrix inversion. It is useful to compare the theoretical intensities with experiment. The ratio of the weights in the upper and lower peaks in figure 1 for K = 0.2, 0.5, and 0.8 is 0.18, 0.71, 0.45 while Kirkpatrick and Harris (1975) find 0.26, 0.70, 0.44 and experiment gives 0.47, 0.9 ± 0.2 and 0.58 (Als-Nielson et al 1975).

Thus while both theories using simple, linearized spin-wave theory agree, the experimental ratios appear to be significantly higher. This may be due to spin-wave renormalization effects although it would be surprising if they were so large. Nevertheless, we would expect linear spin-wave theory to be better in predicting energies than intensities which involve matrix elements.

Also in figure 4 we show calculations at the magnetic-zone boundary with an energy resolution of 0.6 meV. It can be seen that the Ni peak develops substructure. Results using two different 100 × 100 nets show that this substructure is real. Such behaviour can be partially understood by taking only the terms in equation (1) that involve Sf operators (i.e. an Ising model). This is known to be exact at the maximum frequency in the pure antiferromagnet (which occurs at the zone boundary) and predicts five peaks corresponding to the five different configurations of near-neighbours of a Ni atom in the alloy (Birgeneau et al 1975, Holcomb and Harris 1974). These are shown by solid vertical lines in figure 4. It can be seen that the detailed structure in the upper peak is not completely explained by the Ising model. A similar substructure will presumably appear in the Mn peak at an even higher resolution.

In figure 5 we show calculations of the density of states. The upper calculation is averaged over ten local densities of states. These are for a single Ni atom with 0, 1, 2, 3 or 4 Mn neighbours and a single Mn atom with 0, 1, 2, 3 or 4 Ni neighbours. The resulting ten spectra are given a weight of 1/32, 4/32 or 6/32 in the final spectrum shown in figure 5, according to the probability of occurrence. The lower spectrum in figure 5 is over a single randomly chosen degree of freedom involving finite amplitudes on all the sites. This is much more efficient as only one computation is required rather than ten and the result is less noisy. Although the density-of-states calculations are more noisy than the S(κ, ω), nevertheless the structure in the upper peak is clearly present. This is because even in two dimensions there is more phase space associated with the zone-boundary excitations. The weight in the spectrum below 15 meV was always found to be within 2% of 1/2 showing that the Mn and Ni responses are at distinct energies with very little hybridization. In the local density of states for a Mn or Ni site (not shown) the weight was essentially entirely concentrated within the single appropriate peak.

4. Application to \( \text{Rb}_2\text{Mn}_3\text{Zn}_{1-e}F_4 \)

We have also applied the equation-of-motion technique to \( \text{Rb}_2\text{Mn}_3\text{Zn}_{1-e}F_4 \) (Alben and Thorpe 1975b). The motivation for this was largely to see if the clustering effects that were apparent in \( \text{Rb}_2\text{Mn}_{0.5}\text{Ni}_{0.5}F_4 \) are amplified in the case of a magnetic–nonmagnetic
alloy. Numerical calculations by Holcomb and Harris (1974) using the matrix inversion technique for a dilute three-dimensional antiferromagnet led us to expect large effects due to clusters. The number of subpeaks is \((z + 1)\) where \(z\) is the number of near neighbours and so is more prominent in lower dimensions (Bell 1972, Dean 1972) where the individual peaks overlap less.

We have used the Hamiltonian (1) with a nearest-neighbour exchange 0·655 meV and an anisotropy field \((g\mu_B H^A) = 0·03\) meV. In figure 6 we show the neutron scattering law along the \((1, 0)\) direction (same direction as in figure 1) for \(c = 0·5\). It can be seen that the weight moves away from the origin as \(K\) increases from 0 to 0·5 and back again as it goes from 0·5 to 1·0. However, it is not useful to try to define a dispersion relation in

Figure 5. The density of states for Rb_2Mn_xNi_yF_4 calculated over chosen degrees of freedom (upper graph) and for a single random degree of freedom (lower graph). The vertical scale is arbitrary, but the same for both graphs. The calculations were done on 40 \(\times\) 40 nets with an energy resolution of 1·6 MeV (upper curve) and 0·6 MeV (lower curve). The vertical lines are predictions based on an Ising model.

Figure 6. The neutron scattering law \(S(k, \omega)\) for Rb_2Mn_xZn_1-xF_4 with \(c = 0·5\) along the \((1, 0)\) direction where \(k = \sqrt{2\pi \omega k(0)}\). The calculations are for 60 \(\times\) 60 square nets with an energy resolution of 0·6 MeV. The curves for different \(K\) have been normalized so that the peaks heights are the same:
(a) \(K = 0·9\); (b) \(K = 0·7\); (c) \(K = 0·5\); (d) \(K = 0·3\); (e) \(K = 0·1\).
this case as the response is much too broad. In the pure system \((c = 1)\) there is a single, sharp mode that has a maximum energy of 6.6 meV at the magnetic zone boundary \(K = 0.5\). We see that the overall tendency of the spectrum to be weighted towards higher energies for \(k\) near the magnetic-zone boundary remains true in the dilute case. In figure 7 we show the total integrated intensity obtained from figure 6. The intensity changes by about a factor of 100 in going from the nuclear Bragg point to the magnetic Bragg point and is very similar to the behaviour found in the pure antiferromagnet.

In figure 8 we show the substructure at the zone boundary in more detail. We see from figure 6 that this substructure is most pronounced at the zone boundary. Indeed from the Ising model arguments of the previous section we would expect the five subpeaks to occur at 0, 1.67, 3.31, 4.94 and 6.58 meV. We see that in fact the subpeaks are a little below these energies as we would expect from the effect of the \(S_i^z S_j^z\) terms in second-order perturbation theory (Dietrich et al 1975).

![Figure 7](image1.png)

Figure 7. The total integrated scattering intensity as a function of \(K\) (see figure 6) along the \((1, 0)\) direction for \(c = 0.5\). The calculations are for 60 \(\times\) 60 nets with an energy resolution of 0.6 MeV (see figure 6). The vertical scale is logarithmic.

![Figure 8](image2.png)

Figure 8. The structure in \(S(k, \omega)\) for \(c = 0.5\) at the magnetic zone boundary in the \((1, 0)\) direction \((K = 0.5\) in figures 6(c) and 7). The calculations are for 100 \(\times\) 100 nets with an energy resolution of 0.6 MeV. The curves marked (a) and (b) are for two different nets and the average of the two results is shown by (a) + (b).

Deviations from the linear spin-wave approximation are also expected to increase for the dilute system although we cannot take these into account in our computer simulations. We therefore consider the exact results for two-spin and three-spin clusters. We neglect the small anisotropy term. For the two-spin case the exact energies are given by

\[
E^{(2)} = \frac{1}{2}J[j(j + 1) - 2s(s + 1)]
\]

where the total spin \(j\) varies from 0 to 2s. The ground state is \(j = 0\) and allowed transitions associated with \(S(k, \omega)\) connect only to the state \(j = 1\). The allowed excitation energy is \(\Delta E^{(2)} = J = 0.655\) meV. The linear spin-wave analysis predicts an excitation energy of 0, while the maximum energy of the pure system is \(4JS = 6.55\) meV. For system with a high spin, the spin-wave result for a two-spin cluster is a relatively good approximation.
For three spins, the exact energies are given by

\[ E^{(3)} = \frac{1}{2} J [J(j + 1) - j(j' + 1) - s(s + 1)] \]

where \( 2s \geq j' \geq 0 \) and \( j' + s \geq j \geq |j' - s| \). The allowed transitions are summarized by selection rules \( \Delta j' = 0, \pm 1 \) and \( \Delta j = 0, \pm 1 \). For the present case the transition energies are: \( \Delta E^{(3)} = 0 \) meV, 1.64 meV, 2.29 meV, 3.28 meV, 5.57 meV. The linear spin-wave result gives transitions at 0 and 1.64 meV in agreement with the two lowest allowed excitations.

![Figure 9](image1.png)

**Figure 9.** The density of spin-wave states for three different concentrations calculated by exciting a single random degree of freedom as described in the text. The calculations are for 60 x 60 nets with an energy resolution of 0.6 MeV. The vertical scale is arbitrary; but the area under the three graphs is the same.

![Figure 10](image2.png)

**Figure 10.** The fraction of the weight in the peak at 1.64 MeV is shown as a function of the concentration \( c \). (The total density of states being normalized to 1.) The curve \( (a) \) is the contribution from only isolated triads while \( (b) \) includes contributions from triads at the corners or clusters. The percolation concentration for the site problem (Essam 1972) on a square net is marked \( p_c \).

We show in figure 9 the density of states for three different concentrations computed by exciting random degrees of freedom involving finite amplitudes on all the sites. It can be seen that the weight moves to higher energy as the magnetic concentration increases. The peak at around 1.64 meV is due to triads as discussed in the previous paragraph and would be a delta function if our resolution was infinitely sharp. It is much more prominent in the more dilute cases. Indeed it is easy to work out the weight expected in this peak. The probability of a linear triad is \( 2c^2(1 - c)^2 \) while that of a right-angle triad is \( 4c^2(1 - c)^3 \) giving a total probability of \( 2c^2(1 - c)^2(3 - c) \). Each isolated triad has two
modes at 1-64 meV and so the expected weight is $4c^2(1 - c)^7(3 - c)$. The actual weight is the peak at 1-64 meV and is somewhat greater than this prediction, as seen by comparing figures 9 and 10. Of the two modes at 1-64 meV in the isolated triad, one has finite amplitudes on all three atoms while the other has amplitudes +1 and −1 on the two outer atoms. For this mode it is irrelevant what the other two neighbours of the centre atom are. Thus if we count all isolated triads with weight 2 and all triads at the corners of clusters with weight 1, we find an increased probability of $2c^2(1 - c)^7(3 - c)[2 + 1/(1 - c^2)] = 2c^2(1 - c)^7(3 - c)(3 - 2c + 2c^2)$. This is also plotted in figure 10 and gives a better estimate of the weight in the sharp peak at 1-64 meV. It is still only a lower bound as of course this energy can also occur in larger clusters and involve a finite amplitude on more than two sites. We see from figure 10 that more than half the weight at $c = 0.3$ comes from triads that are not isolated. The percolation concentration for the site problem $p_c$ is also indicated in figure 10 (Essam 1972). Nothing very dramatic happens to the spin-wave spectrum on going through this concentration as evidenced by the numerical results. In reality, for $p < p_c$, the spectrum consists entirely of densely spaced delta functions whereas for $p > p_c$, there is a continuous spectrum as well as the delta functions. Our numerical results show that it is unlikely that experiments will be able to distinguish these two kinds of behaviour.

5. Conclusions

We have shown that the response functions of insulating magnetic alloys can be calculated using rather straightforward numerical integration of the equations of motion for the system. Some care is necessary with the initial conditions because the linearized equations of motion lead to a non-symmetric matrix. The method is extremely efficient for calculating the inelastic neutron scattering law $S(k, \omega)$ and can also be used to obtain densities of spin-wave states $\rho(\omega)$. Our results for Rb₂Mn₀.₄Ni₀.₆F₄ show good agreement with experiment although there are differences in intensities that have yet to be accounted for. Our results for the dilute antiferromagnet show that spectra have rather well defined subpeaks with superimposed spikes from localized modes.

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