

The second-neighbour Ising chain as a model for spin-phonon interactions: susceptibility

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Abstract. The connection between Ising systems with spin-phonon interactions and Ising systems with second-neighbour interactions is developed, treating the phonon spectrum by the Einstein approximation. The effective spin Hamiltonian has competing interactions of the type that Stephenson has shown lead to a 'disorder point', a temperature at which the character of the spin-spin correlation function changes. The one-dimensional case is taken as an example and Stephenson's susceptibility expression derived by a new method. The relevance of disorder-point phenomena and the associated anomalous susceptibility to spin-phonon systems is discussed.

1. Introduction

Frankel and Rapaport (1970) have recently investigated the properties of an Ising chain with nearest-neighbour and second-neighbour interactions. They derived formulae for the zero-field free energy and for various correlation functions in finite open-ended chains and described an application of their results to the study of binary alloys. Stephenson (1970) also investigated this system as an example of the occurrence of disorder points. A disorder point is a temperature at which the behaviour of the spin-spin correlation functions changes. For a second-neighbour Ising chain the change is from a monotonic decay to an oscillatory decay with temperature dependent wavelength. The present results reproduce Stephenson's expression for the susceptibility, using matrix methods without having to transform the spin variables. Morita and Horiguchi (1972) have calculated the ground states in the presence of an applied field. The present work is motivated by the connection between Ising systems with distant-neighbour interactions and Ising systems with spin-phonon interactions. The general formalism was derived by Bolton and Lee (1970). A simplified derivation for the special case of an Einstein phonon spectrum is presented in § 2. These results can also be derived by assuming the phonon frequency is independent of the wavelength and using the Bolton and Lee formulae directly. Section 3 derives the susceptibility of a second-neighbour Ising chain. The matrix method used can also be applied to chains with third-neighbour interactions. Section 4 discusses the general properties of the susceptibility and possible applications to spin-phonon systems. The implications of disorder-point phenomena for the theory of spin-phonon interactions are also considered.

2. The Hamiltonian

If a system of Ising spins $s(\mathbf{r}_i) = \pm 1$ are located at lattice sites \mathbf{r}_i and nearest-neighbour

spins interact with a strength that varies linearly with distance, then the Hamiltonian can be written :

$$\hat{H} = \frac{1}{2m} \sum_{\langle r_i \rangle} \hat{\mathbf{p}}(r_i) \cdot \hat{\mathbf{p}}(r_i) + \frac{\alpha}{2} \sum_{\langle r_i \rangle} \hat{\mathbf{u}}(r_i) \cdot \hat{\mathbf{u}}(r_i) - \sum_{\langle r_i \rangle} \sum_{\langle a_j \rangle} \{J + \epsilon(\hat{u}(r_i)_j - \hat{u}(r_i + a_j))s(r_i)s(r_i + a_j)\} - \sum_{\langle r_i \rangle} m\mathcal{H}s(r_i) \quad (1)$$

where \mathbf{a}_j are the lattice basis vectors and $\hat{u}(r_i)_j, \hat{p}(r_i)_j$ are components of the position and momentum operators $\hat{\mathbf{u}}(r_i), \hat{\mathbf{p}}(r_i)$, for the spin at r_i . These operators obey the normal commutation relations.

The first terms of the Hamiltonian correspond to treating the phonons by the Einstein model approximation.

If the substitution

$$\hat{u}'(r_i)_j = \hat{u}(r_i)_j - \frac{\epsilon}{\alpha} s(r_i) \{s(r_i + a_j) - s(r_i - a_j)\} \quad (2)$$

is made, then the Hamiltonian can be written :

$$\hat{H} = \frac{1}{2m} \sum_{\langle r_i \rangle} \mathbf{p}(r_i) \cdot \mathbf{p}(r_i) + \frac{\alpha}{2} \sum_{\langle r_i \rangle} \hat{\mathbf{u}}'(r_i) \cdot \hat{\mathbf{u}}'(r_i) - \sum_{\langle r_i \rangle} m\mathcal{H}s(r_i) - J \sum_{\langle r_i \rangle} \sum_j s(r_i)s(r_i + a_j) - \left(\frac{\epsilon}{\alpha}\right)^2 \sum_{\langle r_i \rangle} \sum_{\langle a_j \rangle} \{2 - 2s(r_i + a_j)s(r_i - a_j)\}. \quad (3)$$

Since the commutation relations of the $u'(r_i)_j$ are the same as for the $u(r_i)_j$, the first two terms still represent Einstein model phonons. The remaining terms represent an independent system of Ising spins with first- and second-neighbour interactions. This part of the Hamiltonian can be rewritten for one-dimensional cases as

$$\begin{aligned} \hat{H} &= - \sum_{j=1}^N (J_1 s_j s_{j+1} + J_2 s_j s_{j+2} + m\mathcal{H} s_j) \\ &= - \sum_{i=1}^{\frac{1}{2}N} \{J_1 s_{2i} s_{2i+1} + J_1 s_{2i+1} s_{2i+2} + J_2 s_{2i} s_{2i+2} + J_2 s_{2i+1} s_{2i+3} \\ &\quad + m\mathcal{H}(s_{2i} + s_{2i+1})\}. \end{aligned}$$

The assumed boundary conditions are

$$s_{N+1} = s_1, \quad s_{N+2} = s_2, \quad N \text{ even.}$$

3. Thermodynamics

The thermodynamic properties of the system can be derived from the partition function :

$$Z = \text{Tr}_{\text{spin states}} \exp\left(-\frac{\hat{H}}{kT}\right). \quad (5)$$

Dobson (1969) reviewed in detail how the partition function can be expressed in terms

of a transfer matrix, and showed that the results obtained did not depend on the boundary conditions or the symmetry of the matrix.

Using the variables :

$$A = \exp\left(\frac{2J_1}{kT}\right) \quad (6a)$$

$$B = \exp\left(\frac{2J_2}{kT}\right) \quad (6b)$$

$$h = \exp\left(\frac{2m\mathcal{H}}{kT}\right) \quad (6c)$$

the transfer matrix is

$$V = \begin{bmatrix} hAB & hB^{-1} & h & Ah \\ h^{-1}B^{-1} & h^{-1}AB & h^{-1}A & h^{-1} \\ 1 & A^{-1} & A^{-1}B & B^{-1} \\ A^{-1} & 1 & B^{-1} & A^{-1}B \end{bmatrix}. \quad (7)$$

The partition function is

$$Z = \text{Tr } V^{\frac{1}{2}N} = \lambda_1^{\frac{1}{2}N} + \lambda_2^{\frac{1}{2}N} + \lambda_3^{\frac{1}{2}N} + \lambda_4^{\frac{1}{2}N} \quad (8)$$

where $\lambda_1 \geq \lambda_2 \geq \lambda_3 \geq \lambda_4$ are the four eigenvalues of the matrix V . In the limit $N \rightarrow \infty$, the free energy becomes

$$F = -kT \ln Z \rightarrow -\frac{1}{2}NkT \ln \lambda_1 \quad \text{if } \lambda_1 \neq \lambda_2. \quad (9)$$

The magnetization is

$$M = -\frac{\partial F}{\partial \mathcal{H}} = \frac{1}{2}NkT \frac{\partial \lambda_1}{\partial \mathcal{H}} \frac{1}{\lambda_1}. \quad (10)$$

so that the susceptibility is:

$$\chi = -\frac{\partial^2 F}{\partial \mathcal{H}^2} = \frac{1}{2}NkT \left\{ \lambda_1 \frac{\partial^2 \lambda_1}{\partial \mathcal{H}^2} - \left(\frac{\partial \lambda_1}{\partial \mathcal{H}} \right)^2 \right\} \frac{1}{\lambda_1^2}. \quad (11)$$

The eigenvalues are found from the quartic equation

$$\det(V - \lambda I) = \sum_{i=0}^4 q_i \lambda^i = 0. \quad (12)$$

Although the general solution of a quartic can be expressed in closed form, so that the free energy in arbitrary fields is known, it is easiest to find the susceptibility less directly.

If $y^{(n)} = \partial^n y / \partial \mathcal{H}^n$ then differentiating equation (12) with respect to the \mathcal{H} gives:

$$\sum_{i=0}^4 C_m^n q_i^{(m)} (\lambda^i)^{(n-m)} = 0 \quad (13)$$

with

$$(\lambda^i)^{(n+1)} = i \sum_{m=0}^n (\lambda^{i-1})^{(m)} \lambda^{(n-m+1)} C_m^n. \quad (14)$$

The C_m^n are the binomial coefficients.

From this point on only quantities at zero field are considered. Let

$$\lambda_0 = \lambda_1(\mathcal{H} = 0) = \exp\left(\frac{2F(\mathcal{H} = 0)}{NkT}\right). \quad (15)$$

Equation (27) shows that $q_i^{(m)} = 0$ for all odd m . An inductive proof shows that $(\lambda^i)^{(m)} = 0$ for all i and all odd n . If one has $(\lambda^i)^{(2n+1)} = 0$ for all $n < m$ then equations (13) and (14) give

$$\sum_{i=0}^4 q_i (\lambda^i)^{(2m+1)} = 0 \quad (16a)$$

$$(\lambda^i)^{(2m+1)} = i\lambda^{(2m+1)}\lambda^{i-1}. \quad (16b)$$

Therefore

$$\left(\sum_{i=0}^4 i q_i \lambda^{i-1}\right) \lambda^{(2m+1)} = 0$$

thus

$$\lambda^{(2m+1)} = 0$$

and

$$(\lambda^i)^{(2m+1)} = 0. \quad (17)$$

Since for $m = 0$ equations (16a) and (16b) hold without any initial assumptions, then equation (17) holds for all m .

For the second derivatives

$$\sum_{i=0}^4 \{q_i^{(2)} \lambda^i + q_i (\lambda^i)^{(2)}\} = 0 \quad (18)$$

$$(\lambda^i)^{(2)} = i\lambda^{(2)}(\lambda^{i-1}). \quad (19)$$

Equations (13) and (14) enable one to calculate all field derivatives of the free energy, at zero field, once λ_0 is known. If the transfer matrix is between successive groups of n Ising spins, it will be of dimension $2^n \times 2^n$ so that finding the eigenvalues involves solving a polynomial of degree 2^n . In zero field the degree of the polynomial can be reduced by a change of variable (Frankel and Rapaport 1970) or equivalently by a matrix transformation (Dobson 1969). The equations above enable one to calculate the susceptibility once the full transfer matrix and the zero-field free energy are known.

In the present case a further simplification results from the fact that the characteristic equation, equation (12), can be expressed as:

$$(\lambda^2 - \lambda a + b)^2 - \lambda^2 c^2 = 0 = \sum_{i=0}^4 q_i \lambda^i \quad \text{for } \mathcal{H} = 0. \quad (20)$$

Differentiating with respect to c gives

$$\sum_{i=0}^4 \frac{\partial q_i}{\partial c} \lambda^i = -2c\lambda^2 = -\sum_{i=0}^4 q_i \lambda^{i-1} i \frac{\partial \lambda}{\partial c}. \quad (21)$$

Since equation (27) shows that a, b, c are non-negative the largest eigenvalue is

$$\lambda_0 = \frac{1}{2}(a+c) + \frac{1}{2}\{(a+c)^2 - 4b\}^{1/2} \quad (22)$$

giving

$$\lambda_0^2 = \lambda_0(a+c) - b$$

$$\frac{\partial \lambda}{\partial c} = \frac{\lambda_0}{\{(a+c)^2 - 4b\}^{1/2}} \quad (23)$$

so that

$$\sum_{i=0}^4 q_i \lambda^{i-1} = 2c \lambda_0 \{(a+c)^2 - 4b\}^{1/2}. \quad (24)$$

From equations (11), (18) and (19) the initial susceptibility is:

$$\chi_0 = \frac{NkT}{2\lambda_0} \frac{\partial^2 \lambda}{\partial \mathcal{H}^2} = -\frac{NkT}{2\lambda_0} \left(\sum_{i=0}^4 q_i^{(2)} \lambda^i \right) \left(\sum_{i=0}^4 q_i \lambda^{i-1} \right)^{-1} \quad (25)$$

$$= -\frac{NkT}{2\lambda_0} \frac{\{q_3^{(2)}(a+c) + q_2^{(2)}\}\lambda_0 - q_3^{(2)}b + q_1^{(2)}}{2c\{(a+c)^2 - 4b\}^{1/2}} \quad (26)$$

after using $q_4^{(2)} = q_0^{(2)} = 0$ and equations (22), (23) and (24).

The characteristic equation is actually

$$0 = \lambda^4 - \lambda^3 \{AB(h+h^{-1}) + 2A^{-1}B\} + \lambda^2 \{B^2(A^2 + A^{-2}) - 2B^{-2} + 2(h+h^{-1})(B^2 - 1)\} \\ - \lambda \{(h+h^{-1})BA^{-1}(B - B^{-1})^2 + 2AB(B - B^{-1})^2\} + (B - B^{-1})^4. \quad (27)$$

At zero field it has the form of equation (20) with:

$$a = (A + A^{-1})B \quad (28a)$$

$$b = (B - B^{-1})^2 \quad (28b)$$

$$c = 2B^{-1} \quad (28c)$$

$$q_1^{(2)} = -2BA^{-1}(B - B^{-1})^2 \left(\frac{4m^2}{k^2 T^2} \right) \quad (29a)$$

$$q_2^{(2)} = 4(B^2 - 1) \left(\frac{4m^2}{k^2 T^2} \right) \quad (29b)$$

$$q_3^{(2)} = -2AB \left(\frac{4m^2}{k^2 T^2} \right). \quad (29c)$$

The susceptibility is then

$$\chi_0 = \frac{Nm^2 B \{(2A + 2 + A^2 B^2 - B^2)\lambda_0 - B(B - B^{-1})^2(A - A^{-1})\}}{kT \lambda_0 \{(A - A^{-1})^2 B^2 + 4(A + A^{-1}) + 8\}^{1/2}} \quad (30)$$

with

$$\lambda_0 = \frac{1}{2}(A + A^{-1})B + B^{-1} + \frac{1}{2}\{(A - A^{-1})^2 B^2 + 4(A + A^{-1}) + 8\}^{1/2}. \quad (31)$$

This value of λ_0 is, as required, the square of the value obtained by Frankel and Rapaport and by Stephenson. The susceptibility expression has been shown to reduce to the form obtained by Stephenson.

4. Results and conclusions

Frankel and Rapaport have pointed out that for various ratios of interaction strengths there is a change in the ground state ordering. $J_1 = 0, J_2 > 0$ defines a boundary between ferromagnetic and antiferromagnetic ground states with the exact boundary corresponding to two independent, interpenetrating chains with ferromagnetic ground states. The susceptibilities behave as expected, diverging as $T \rightarrow 0$ in the ferromagnetic case and tending to zero as $T \rightarrow 0$ in the antiferromagnetic case. The boundary $J_1 = -\frac{1}{2}J_2 > 0$ is a similar boundary between ferromagnetic and antiferromagnetic ground states.

At the boundary $J_1 = \frac{1}{2}J_2 < 0$ the change is between two possible antiferromagnetic ground states. Near the boundary, the susceptibility is anomalously large compared to the susceptibility further from this boundary. For $J_1 = \frac{1}{2}J_2 < 0, \chi \rightarrow \infty$ as $T \rightarrow 0$. The qualitative explanation is that the two interactions compete so that the spins are only weakly coupled. This is shown by the spin-spin correlation functions obtained by Rapaport and by Stephenson which show how the spins are comparatively free to align in response to an applied field instead of being locked into a local antiferromagnetic ordering.

Stephenson has shown that disorder points occur for $-|J_1| < \frac{1}{2}J_2 < 0$. The susceptibility varies smoothly through the disorder temperature and does not show any distinctive behaviour except near the $J_1 = \frac{1}{2}J_2 < 0$ boundary.

The transformation from equations (1) to (4) is equivalent to using the particular case of an Einstein phonon spectrum in the effective spin Hamiltonian of Bolton and Lee (1970). The general form of the effective Hamiltonian has the original nearest-neighbour interaction plus additional four-spin interactions. In equation (4) these take the form $s_i s_{i+2} (s_{i+1})^2$ which for spin $\frac{1}{2}$ reduces to a two-spin interaction. If the phonon spectrum corresponding to nearest-neighbour harmonic interactions is used then, in one dimension, the four-spin terms are of the form $(s_i s_{i+1})^2$ which will be constant for spin $\frac{1}{2}$ (Mattis and Schultz 1963, Bolton and Lee 1970).

In three dimensions it can be shown that the Debye approximation for the phonon spectrum also leads to a negative coupling between spins separated by two lattice vectors (Lee and Bolton 1971). There are of course additional four-spin interactions. Although the Einstein phonon spectrum is unrealistic in one dimension, the effective spin Hamiltonian might be expected to give rise to behaviour analogous to the high temperature behaviour of a three-dimensional magnetoelastic system. It seems probable that in an antiferromagnetic system competition between the interactions will lead to an enhanced susceptibility near the Néel point in three dimensions as it does in one dimension.

To extend the prediction of anomalously large susceptibility to a three-dimensional system, the following conditions must hold.

(i) The Einstein phonon must be capable of giving the dominant contribution to the true spin-phonon interaction. This is not true in one dimension but the results of Lee and Bolton (1971) suggest that it may be true in three dimensions, particularly in materials where the Debye temperature is well below the Néel temperature.

(ii) The competing interactions must lead to an enhanced susceptibility in three dimensions. To verify this would involve a study of a fourth-neighbour Ising system.

(iii) The anomalous behaviour must occur in a region that is actually accessible to the system. Lee and Bolton (1971) have shown that a sufficiently strong spin-phonon coupling can cause a second-order transition to become first order. When this type of system is treated by the mean-field approximation the solution formally corresponds to

transforming the critical point on to a re-entrant branch of the spontaneous magnetization curve. The transformed critical region no longer corresponds to actual equilibrium states of the system.

The three factors listed above indicate the need for a study that goes well beyond the tentative arguments suggested here. The importance of this investigation stems from the fact that the susceptibility has been increased by the spin-phonon interactions. In contrast, the formalism of Mattis and Schultz (1963) which is based on homogeneous strains, indicates that the susceptibility should be less than the maximum susceptibility of the incompressible system. This means that a detailed study of the susceptibility may reveal a qualitative difference between theories of magnetoelastic effects based on homogeneous strains and theories based on spin-phonon interactions.

The range of values for which disorder points occur is $-|J_1| < \frac{1}{2}J_2 < 0$. This means that disorder-point phenomena may be expected for all but extremely strong spin-phonon interactions. Stephenson has shown that as well as a disorder temperature T_D , there is a temperature T_E at which the second-neighbour correlation function changes sign. This behaviour severely restricts the range of validity of the work of Bolton *et al* (1972) whose approximate descriptions of the second-neighbour correlation function do not reflect this change of behaviour.

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