

## Transverse dynamic susceptibility for an Ising spin system: An exact eigenvalue equation for the resonance frequencies

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The dynamics of an Ising system is studied assuming that a microwave radiation field is coupled to the magnetic system through the transverse spin components. The transverse dynamic susceptibility is calculated with the use of the Green-function formalism in an exact manner, without the use of any decoupling scheme. The method yields an exact eigenvalue equation for the resonance frequencies, valid for any dimension and lattice, and a physical meaning of these resonances can be given in terms of the low-lying excitations for an Ising system. Those excitations correspond to spin-flip transitions in elementary clusters which represent all the possible spin arrangements of nearest neighbors. The present method does not allow for a calculation of line intensities without the help of statistical mechanics to evaluate correlation functions.

### I. INTRODUCTION

A system described by a pure-Ising Hamiltonian has no dynamic properties since all the spin operators commute with the total Hamiltonian. On the other hand, an Ising system can be considered as an extremely anisotropic Heisenberg system, where all the three spatial components of the spin are present but spin-spin interactions are only realized along a given direction. Following the above idea the transverse spin component can be coupled with an external perturbation, e.g., microwave radiation as in a typical electron-spin-resonance (ESR) experiment. Proceeding in this way we obtain a dynamic model whose low-lying excitations are in the form of localized spin flips, in contrast to a truly Heisenberg system where the response is monitored by spin waves encompassing the whole system.

The transverse dynamic susceptibility of such a modified Ising model can be calculated through the fluctuation-dissipation theorem using the Green-function formalism developed by Zubarev.<sup>1</sup> Poles of the susceptibility are identified as the resonant frequencies of the system, since they contribute with  $\delta$ -function singularities to the power absorption. In this paper we want to show that an exact eigenvalue equation for the resonance frequencies can be formulated, valid for the general case, which does not depend on the dimension or lattice, but just on the coordination number when only nearest-neighbor interactions are allowed. From the set of Green-function expressions the eigenvalue equation can be isolated in the form of a finite continuous fraction. In this paper we also give an algorithm to generate the characteristic polynomials for any coordination number.

Although this result seems to have been suggested before by calculations on a particular problem,<sup>2-4</sup> the au-

thors have not been able to find a result of such a generality, as the one presented here in the published literature, even when a calculation of this sort is an obvious one to carry out. It is worth noting that, since the eigenvalues of the Ising problem are known for any dimension (but not the partition function), an exact account of the resonance frequencies solves, in principle, the dynamical problem.

The fact that resonance frequencies can be exactly calculated derives from peculiar characteristics of the Green-function equations for an Ising system. The chain of coupled equations is finite and closed, the order of such a chain being equal to the number of nearest neighbors when only nearest-neighbor interactions are taken into account.<sup>2</sup>

The line intensities can only be calculated through the knowledge of several correlation functions. When the number of nearest neighbors increases (for higher-dimensional lattices) higher-order correlation functions appear in the formalism. As noted in Ref. 2, the number of such correlations is larger than the independent equations obtained for the different Green functions, so the method does not allow for a solution concerning the line intensities, and an approximate scheme should be devised, or alternatively, correlations have to be taken from statistical mechanics calculations.

When an external magnetic field is applied, the only correlation functions which are known in exact analytical form are those corresponding to the one-dimensional Ising problem (the results are presented in the Appendix). The authors have proposed in a recent work an approximate approach based on the cluster-variation method<sup>4</sup> to calculate the temperature dependence of the different line intensities. In the present calculation we have found that resonances can be interpreted as spin-flip transitions embedded in different cluster configurations, and in agree-

ment with the results of Ref. 4, it can be realized that processes occurring in clusters of nearest neighbors are the only ones contributing to the macroscopic response.

In what follows we only deal with the case of ferromagnetic exchange coupling when an external static magnetic field is applied in the longitudinal direction, which we shall choose as the  $z$  axis. The incident microwave radiation is polarized in the  $x$ - $y$  plane, thus reproducing the typical geometry of an ESR experiment. The Ising anti-ferromagnet can be treated analogously, the resonance frequencies being the same than those of the corresponding ferromagnetic problem. Temperature dependence of line intensities are of course different in both cases.<sup>4</sup>

## II. GREEN-FUNCTION CALCULATION

The unperturbed Ising Hamiltonian including the Zeeman contribution is written as

$$\hat{H}_0 = -J \sum_{(i,j)} \sigma_i^z \sigma_j^z - \hbar\omega_0 \sum_j \sigma_j^z, \quad (1)$$

where  $J > 0$  is the ferromagnetic exchange constant and  $\hbar\omega_0$  is the Zeeman energy. The parentheses  $(i,j)$  means summation over nearest neighbors. The total spin components are defined by

$$S^z \equiv \sum_j \sigma_j^z, \quad (2)$$

$$S^\pm \equiv \sum_j \sigma_j^\pm = \sum_j (\sigma_j^x \pm i\sigma_j^y). \quad (3)$$

For absorption of microwave radiation polarized in the  $x$ - $y$  plane, the transverse dynamic susceptibility is proportional to the Fourier transform of the retarded Green function given by

$$\begin{aligned} \langle\langle S^+(t); S^- \rangle\rangle_R &\equiv -i\Theta(t) \langle [S^+(t), S^-] \rangle, \\ &= \sum_{k,j} \langle\langle \sigma_k^+(t); \sigma_j^- \rangle\rangle_R, \end{aligned} \quad (4)$$

where the notation of Ref. 1 has been used and the symbol  $[S^+, S^-]$  stands for the commutator

$$(\hbar\omega - 2\hbar\omega_0) {}_m G_{kj}(\omega) = \frac{\hbar}{2\pi} (4\delta_{kj} \langle \sigma_m^z \sigma_k^z \rangle - 2\delta_{mj} \langle \sigma_m^- \sigma_k^+ \rangle) + 2J G_{kj}(\omega) + 2J \sum_l' \langle\langle \sigma_l^z \sigma_m^z \sigma_k^+; \sigma_j^- \rangle\rangle_\omega. \quad (8)$$

In relation (8) the sum  $\sum_l'$  is performed over all the nearest neighbors of site  $k$  different from  $m$ . It is apparent from the general structure of Eqs. (6) and (8), that the relevant quantities to be calculated are

$$\begin{aligned} [1]_{kj} &\equiv \sum_{C_1} m_1 G_{kj}(\omega), \\ [2]_{kj} &\equiv \sum_{C_2} m_1 m_2 G_{kj}(\omega), \\ &\dots, \\ [n-1]_{kj} &\equiv \sum_{C_{n-1}} m_1 m_2 \dots m_{n-1} G_{kj}(\omega), \\ [n]_{kj} &\equiv m_1 m_2 \dots m_n G_{kj}(\omega), \end{aligned} \quad (9)$$

$$[S^+(t), S^-] \equiv S^+(t)S^- - S^-S^+(t).$$

From relation (4) we note that the relevant Green function to be calculated is

$$G_{kj}^{(R)}(t) \equiv \langle\langle \sigma_k^+(t); \sigma_j^- \rangle\rangle_R, \quad (5)$$

or, equivalently, its Fourier transform

$$G_{kj}(\omega) \equiv \langle\langle \sigma_k^+; \sigma_j^- \rangle\rangle_\omega.$$

Using the standard commutation relations among the spin operators we obtain the following equation of motion for  $G_{kj}(\omega)$ :

$$(\hbar\omega - 2\hbar\omega_0) G_{kj}(\omega) = \frac{2\hbar}{\pi} \delta_{kj} \langle \sigma_k^z \rangle + 2J \sum_{(m)} \langle\langle \sigma_m^z \sigma_k^+; \sigma_j^- \rangle\rangle_\omega, \quad (6)$$

where the sum over  $m$  is performed over all the nearest neighbors of site  $k$ .

All the new generated Green function have operators  $\sigma^z$  inserted in the left part of the bracket. So we define, in a short-hand notation,

$$\begin{aligned} {}_m G_{kj}(\omega) &\equiv \langle\langle \sigma_m^z \sigma_k^+; \sigma_j^- \rangle\rangle_\omega, \\ {}_{m_1} G_{kj}(\omega) &\equiv \langle\langle \sigma_{m_1}^z \sigma_k^+; \sigma_j^- \rangle\rangle_\omega, \\ &\dots, \\ {}_{m_1, \dots, m_n} G_{kj}(\omega) &\equiv \langle\langle \sigma_{m_1}^z \dots \sigma_{m_n}^z \sigma_k^+; \sigma_j^- \rangle\rangle_\omega, \end{aligned} \quad (7)$$

$n$  being the coordination number or number of nearest neighbors. It will be noted that no higher-order Green functions are needed, since the system of coupled equations closes for the functions listed above.

New correlation functions are also generated once the equations of motion of the whole chain are written down, but the calculation of those correlations are not relevant for obtaining the resonance frequencies.

As an example let us show the equation of motion coupled to Eq. (6),  $m$  being a nearest neighbor of  $k$  as follows:

where the  $C_r$  are all the different combinations of nearest neighbors of site  $k$ , taken  $r$  at a time. Of course we have

$$\binom{n}{r} = \frac{n!}{(n-r)!r!}$$

of such combinations. With the use of the adimensional variable

$$E \equiv \frac{\hbar(\omega - 2\omega_0)}{2J}, \quad (10)$$

the equations of motion of quantities defined by (9) can be written as

$$\begin{aligned}
 EG_{kj}(E) &= R_{kj}^{(0)} + [1]_{kj} , \\
 E[1]_{kj} &= R_{kj}^{(1)} + 2[2]_{kj} + nG_{kj}(E) , \\
 E[2]_{kj} &= R_{kj}^{(2)} + 3[3]_{kj} + (n-1)[1]_{kj} , \\
 \dots , \\
 E[n-1]_{kj} &= R_{kj}^{(n-1)} + n[n]_{kj} + 2[n-2]_{kj} , \\
 E[n]_{kj} &= R_{kj}^{(n)} + [n-1]_{kj} ,
 \end{aligned}
 \tag{11}$$

where the quantities  $R_{kj}^{(m)}$  are the remainders containing the correlation functions. For instance, we get from (6) and (8)

$$R_{kj}^{(0)} = \frac{\hbar}{\pi J} \delta_{kj} \langle \sigma_k^z \rangle \tag{12}$$

and

$$R_{kj}^{(1)} = \frac{\hbar}{4\pi J} \left[ 4\delta_{kj} \sum_{(m)} \langle \sigma_m^z \sigma_k^z \rangle - 2 \sum_{(m)} \delta_{mj} \langle \sigma_m^- \sigma_k^+ \rangle \right] . \tag{13}$$

The set of Eqs. (11) can be written in the following compact form:

$$E[v]_{kj} = R_{kj}^{(v)} + (v+1)[v+1]_{kj} + (n-v+1)[v-1]_{kj} \tag{14}$$

for  $v=0, 1, \dots, n$ , where the convention

$$\begin{aligned}
 [0]_{kj} &\equiv G_{kj}(E) , \\
 [n+1]_{kj} &= [-1]_{kj} \equiv 0 ,
 \end{aligned}
 \tag{15}$$

has been adopted. The general form of Eq. (14) displays the fact that spin-spin interactions are included up to nearest neighbors only.

The solution of system (11) or (14) is straightforward and can be done in a recursive form: several continued fractions are generated in this process. The general solution can be obtained in a compact form if we introduce

$$f_v^{(n)}(E)[n-v] = (v+1)[n-v-1] + R^{(n-v)} + (n-v+1) \frac{R^{(n-v+1)}}{f_{v-1}^{(n)}(E)} + \dots + \frac{(n-v+1)(n-v+2) \dots (n-1)n}{f_{v-1}^{(n)}(E)f_{v-2}^{(n)}(E) \dots f_0^{(n)}(E)} R^{(n)} , \tag{18}$$

where we have omitted the site indices ( $kj$ ). Relation (18) can be considered valid for  $v=0, 1, 2, \dots, n$  provided that convention (15) is adopted again. In particular, for  $v=n$  we obtain the solution for our Green function  $G_{kj}(E)$  in the form

$$G_{kj}(E) = \sum_{v=0}^n \frac{v!}{f_v^{(n)}(E)f_{v-1}^{(n)}(E) \dots f_{n-v}^{(n)}(E)} R_{kj}^{(v)} . \tag{19}$$

Facing the above result we would like to note the following.

(i) Dependence on site indexes ( $kj$ ) only enters through the remainders  $R_{kj}^{(v)}$ .

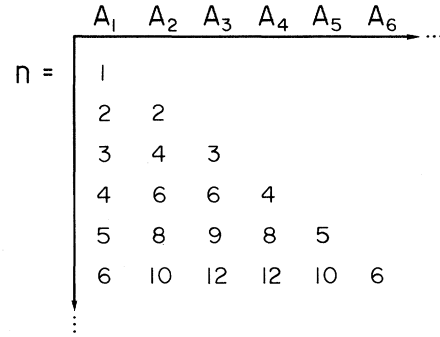


FIG. 1. Coefficients for the continued fraction appearing in the theory can be arranged in a “magic” triangle where columns are multiples of natural numbers, the first column being the coordination number. Relation (17) implies, for a given row, a mirror symmetry.

the following set of continued fractions:

$$\begin{aligned}
 f_0^{(n)}(E) &\equiv E , \\
 f_v^{(n)}(E) &\equiv E - \frac{A_v^{(n)}}{f_{v-1}^{(n)}(E)} , \quad v=1, 2, \dots, n
 \end{aligned}
 \tag{16}$$

where the coefficients  $A_v^{(n)}$  are given by

$$A_v^{(n)} = A_{n-v+1}^{(n)} = v(n-v+1) . \tag{17}$$

Different sets of  $A$  numbers for different coordinations are shown in Fig. 1. The recursive process now proceeds in the following form:

$$\begin{aligned}
 f_0^{(n)}(E)[n] &= R^{(n)} + [n-1] , \\
 f_1^{(n)}(E)[n-1] &= 2[n-2] + R^{(n-1)} + \frac{nR^{(n)}}{f_0^{(n)}(E)} ,
 \end{aligned}$$

yielding the general expression

(ii) Since resonance frequencies are poles of the susceptibility, the only task left is the reduction of the continued fractions  $f_v^{(n)}(E)$  into rational fractions.

(iii) For obtaining the line intensities a partial fraction decomposition must in turn be done and the remainders  $\sum_{k,j} R_{kj}^{(v)}$  have to be calculated. The present method offers no solution to this latter problem.

Fortunately, concerning point (ii) a well-known algorithm can be devised to solve that problem. Writing the continued fractions in the form

$$f_v^{(n)}(E) \equiv \frac{Q_{v+1}(E)}{Q_v(E)} , \tag{20}$$

for a given coordination number  $n$ , the  $Q_v(E)$  polynomials

can be generated by the following recursion relation:

$$Q_\nu(E) = EQ_{\nu-1}(E) - A_{\nu-1}^{(n)} Q_{\nu-2}(E), \quad (21)$$

for  $\nu=2, 3, \dots, (n+1)$ , with the initial values

$$Q_0(E) = 1, \quad Q_1(E) = E. \quad (22)$$

This algorithm is of common use in numerical methods and follows from recursion relation (16). Sets of these  $Q_\nu$  polynomials for different coordination numbers are shown in Fig. 2. Coefficients in relation (19) can now be written as rational fractions. The following result is then easily obtained:

$$G_{kj}(E) = \sum_{\nu=0}^n \frac{Q_{n-\nu}(E)}{Q_{n+1}(E)} \nu! R_{kj}^{(\nu)}, \quad (23)$$

which shows that resonance frequencies are solutions of the equation

$$Q_{n+1}(E) = 0. \quad (24)$$

In Fig. 2 we display the corresponding roots of the characteristic polynomials for several coordination numbers. All the solutions are integers associated to the local field of the different elementary clusters of nearest-neighbor spins. Figure 3 is an illustration of this fact for coordination number  $n=4$ .

Following the procedure of Ref. 2, spectral theorems allow us to find some exact relations between several correlation functions. Unfortunately, the number of these equations is less than the number of correlation functions we have to solve. Ultimately we have to rely on approximate method to estimate the line intensities. The linear chain is an exception which can be determined in exact analytical form with the help of statistical mechanics (see the Appendix).

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COORDINATION NUMBER	CHARACTERISTIC POLYNOMIAL	ROOTS
2	$E(E^2 - 4)$	$0, \pm 2$
3	$E^4 - 10E^2 + 9$	$\pm 1, \pm 3$
4	$E(E^4 - 20E^2 + 64)$	$0, \pm 2, \pm 4$
6	$E(E^6 - 56E^4 + 784E^2 - 2304)$	$0, \pm 2, \pm 4, \pm 6$

FIG. 2. Characteristic polynomials for different coordination numbers, along with the corresponding resonance frequencies.

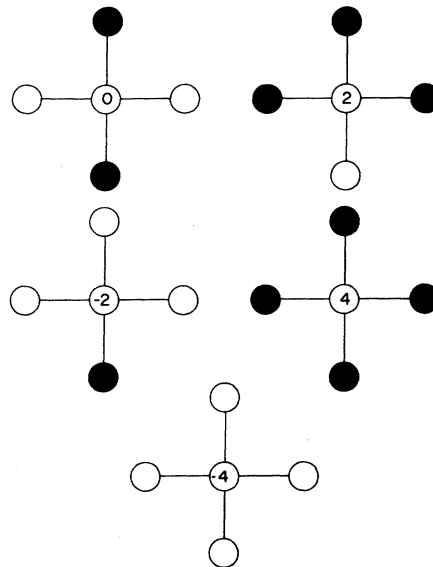


FIG. 3. Elementary clusters for coordination  $n=4$ . Spin up is represented by a closed circle while spin down by an open circle. The local effective field induced at the central site is also indicated. In an ESR experiment the static applied field must be added to the effective field shown here. The above configurations (some of them degenerate) give rise to five resonance lines, whose resonance frequencies are  $2\omega_0$ ,  $2\omega_0 \pm 4J/\hbar$ ,  $2\omega_0 \pm 8J/\hbar$ , respectively.

#### APPENDIX: SOLUTION FOR THE LINEAR CHAIN

For an infinite linear chain (or finite with periodic boundary conditions) with ferromagnetic exchange coupling, the following correlations do not depend on site indices:

$$\langle \sigma_k^z \rangle \equiv M \quad (A1)$$

for magnetization,

$$\langle \sigma_{k-1}^z \sigma_k^z \rangle \equiv \langle \sigma_{k+1}^z \sigma_k^z \rangle \equiv \eta \quad (A2)$$

for short-range—order parameter, and

$$\langle \sigma_{k-1}^z \sigma_k^z \sigma_{k+1}^z \rangle \equiv t \quad (A3)$$

for three-spin correlation.

The remainders appearing in formula (11) can be written as

$$\begin{aligned} R_{kj}^{(0)} &= \frac{\hbar M}{\pi J} \delta_{kj}, \\ R_{kj}^{(1)} &= \frac{2\hbar \eta}{\pi J} \delta_{kj}, \\ R_{kj}^{(2)} &= \frac{\hbar t}{\pi J} \delta_{kj}, \end{aligned} \quad (A4)$$

whereas the resonance frequencies are given by  $E=0, \pm 2$ . The  $Q$  polynomials can be readily generated yielding

$$\begin{aligned}
Q_0(E) &= 1 \\
Q_1(E) &= E, \\
Q_2(E) &= E^2 - 2, \\
Q_3(E) &= E(E^2 - 4),
\end{aligned} \tag{A5}$$

and the solution for the Green function is given by [see Eq. (23) in the main text]

$$\begin{aligned}
G_{kj}(E) &= \left[ \frac{E^2 - 2}{E(E^2 - 4)} \left[ \frac{\hbar M}{\pi J} \right] + \frac{E}{E(E^2 - 4)} \right. \\
&\quad \left. \times \left[ \frac{2\hbar\eta}{\pi J} \right] + \frac{2}{E(E^2 - 4)} \left[ \frac{\hbar t}{\pi J} \right] \right] \delta_{kj}, \tag{A6}
\end{aligned}$$

and through partial fraction decomposition we obtain

$$\begin{aligned}
G_{kk}(E) &= \langle\langle \sigma_k^+; \sigma_k^- \rangle\rangle_E \\
&= \left[ \frac{\hbar}{2\pi J} \right] \frac{M-t}{E} + \left[ \frac{\hbar}{4\pi J} \right] \frac{M+2\eta+t}{E-2} \\
&\quad + \left[ \frac{\hbar}{4\pi J} \right] \frac{M-2\eta+t}{E+2}. \tag{A7}
\end{aligned}$$

The correlation and the corresponding Green function are related through the spectral intensity<sup>1</sup> by a well-known spectral representation. Using this relation for the Green function (A7) we obtain (with  $\beta \equiv 1/k_B T$ )

$$\begin{aligned}
1+M &= \frac{M-t}{1-e^{-\beta\hbar\omega_0}} + \frac{\eta + \frac{1}{2}(M+t)}{1-e^{-\beta\hbar\omega_0} e^{-4\beta J}} \\
&\quad + \frac{\frac{1}{2}(M+t) - \eta}{1-e^{-\beta\hbar\omega_0} e^{4\beta J}}, \tag{A8}
\end{aligned}$$

which allows us to calculate  $t$  if  $M$  and  $\eta$  are known. The magnetization and the short-range parameters can be taken from the exact one-dimensional solution. Those formulas are quoted in Ref. 4, where the following reduced variables are used

$$\tau \equiv \frac{1}{\beta J}, \quad \lambda = \frac{\hbar\omega_0}{J}.$$

An error has been detected in expression (A2) of Ref. 4. The correct relation is given by

$$\begin{aligned}
\eta &= 1 + \frac{2F^{1/2}}{G^2 - 1} \left[ F^{1/2} - G \cosh(\lambda/\tau) \right] \\
&\quad \times \left[ \frac{1 - 2G^{-1} \sinh(2/\tau)}{\sinh^2(\lambda/\tau)} \right] M^2, \tag{A9}
\end{aligned}$$

where

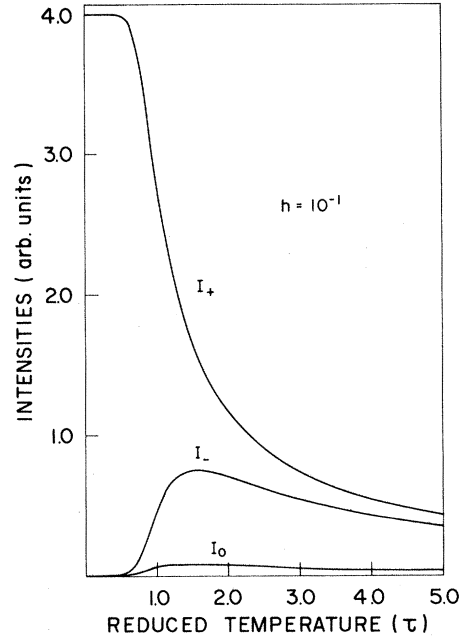


FIG. 4. Line intensities for the Ising ferromagnetic chain as functions of temperature. The applied magnetic field has been chosen as  $\lambda = \hbar\omega_0/J = 0.1$ , and the different lines are identified by their resonance frequency with the convention  $I_0 = I(2\omega_0)$ ,  $I_+ = I[2\omega_0 + (4J/\hbar)]$ , and  $I_- = I[2\omega_0 - (4J/\hbar)]$ .

$$F = G^2 \cosh^2(\lambda/\tau) + 1 - G^2,$$

$$G \equiv e^{2/\tau},$$

and the magnetization is given by

$$M = \frac{\sinh(\lambda/\tau)}{[\cosh^2(\lambda/\tau) - 2e^{-2/\tau} \sinh(2/\tau)]^{1/2}}.$$

The line intensities which follow from relation (A7), when written in arbitrary units, are

$$I_0 \equiv I[E=0] = 2|M-t|,$$

$$I_+ \equiv I[E=2] = M + 2\eta + t, \tag{A10}$$

$$I_- \equiv I[E=-2] = |M - 2\eta + t|,$$

and they are depicted in Fig. 4 as functions of the reduced temperature for a particular value of the static magnetic field.

As a general feature we note that the intensity which dominates at low temperature is the one associated with the cluster where all the spins are parallel to the applied field. Intensities for all the other lines vanish in the limit  $\tau \rightarrow 0$ . This is in complete agreement with the results presented previously by the authors in Ref. 4.

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