ON THE TEMPERATURE DEPENDENCE OF THE PARAMAGNETIC RESONANCE LINE SHAPE FUNCTION IN THE CASE OF THE ISING MODEL

by

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ABSTRACT

The adequacy of the approximation method used by McMillan and Opechowski (1960) in their theoretical study of the temperature dependence of the paramagnetic resonance line shape function is very difficult to ascertain for the case of a typical paramagnetic crystal. For this reason the approximation method has been investigated, in this thesis, for the very simple case of the one-dimensional Ising model. Exact expressions for the line shape function of the model are compared with expressions obtained by the above mentioned approximation method. The agreement between the two expressions is found to be very good for all temperatures, and in particular, for extremely low temperatures.

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SECTION I

Introduction

Magnetic Resonance and Line Shape Function

Let us consider a paramagnetic ion to the extent that it is characterized by a total angular momentum vector $\vec{S}$ and a magnetic moment vector $\vec{\mu}$ satisfying the relation

$$\vec{\mu} = -g\beta\vec{S} \quad , \quad \beta = |e|\hbar/2mc$$

where the various constants take their usual meaning. We shall call such a quantum mechanical model, with arbitrarily fixed values of g and S, a "spin". We shall call a system containing an arbitrarily large number N of identical spins, i.e., spins with the same values of g and S, a "spin-system" or simply a "system".

When such a spin-system is placed in a constant external magnetic field $\vec{H}$, in the absence of interactions among the spins, the energy of the system will then be quantized into a finite number of distinct Zeeman energy levels. The number of such levels depends on the angular momentum quantum number S as well as the number N of spins present in the system. These levels, except for the highest and the lowest, are highly degenerate. The degeneracy is in general different for different energy levels, and depends again on the values of S and N.

When a weak, sinusoidally varying magnetic field $\vec{H}_1$, frequency ν, is superimposed perpendicularly onto the constant magnetic field $\vec{H}$, and is so weak that its influence on the energy levels of the system is entirely negligible, transitions will take place among those levels if and only if the condition

$$\nu = \nu_0 \qquad \text{where} \quad h\nu_0 \equiv |E' - E|$$

is satisfied, where E and E' are the energy levels of the system among which transition is induced.

In the case where there is no interaction among the spins, we have

$$E' - E = g\beta H (M' - M)$$

where M' and M are the quantum numbers corresponding to E' and E respectively with the selection rule

$$|M' - M| = 1$$

Thus $\nu_0 = g\beta H/h$

Hence an absorption line will be observed at the frequency ν_0 and we shall call this process "Magnetic Resonance".

Were it not for the fact that the occupation numbers are different for different energy levels, there would be neither net absorption nor net emission of energy. Since according to a general result of quantum mechanics, the transition probability between two levels is the same both ways. The average absorption and emission of energy in unit time thus cancel. But in the actual case, the system of paramagnetic ions is in a more or less well-defined thermodynamic equilibrium state, to which let us assign an absolute temperature T. The occupation number of the energy level E of a system of spins will be proportional to the Boltzmann factor namely $\exp\left(-\frac{E}{kT}\right)$, where k is the usual Boltzmann constant.

When $E > E'$

then $\exp\left(-\frac{E'}{kT}\right) > \exp\left(-\frac{E}{kT}\right)$

so there will be more spins at lower energy levels than at higher ones. Consequently there will be more transitions from lower states into higher ones resulting in the net absorption of energy from the oscillating magnetic field $\vec{H}_1$.

When weak exchange as well as dipole interactions exist among the spins, each energy level of the system will be decomposed into a large number of closely spaced sublevels, each of which may or may not be degenerate. As a result of this, the magnetic resonance will take place at a large number of frequencies in the neighbourhood of ν_0 . Instead of a single monochromatic absorption line at ν_0 , we have a large number of absorption lines with different intensities distributed in the close neighbourhood of ν_0 forming an absorption band. Each component line of this band, say a line at frequency ν_p *, is characterized by an intensity function $I(\nu_p)$ ($p = 1,2, \ldots \theta$, the number of component lines in the band). The number θ of component lines present in the band depends on the type and the magnitude of the interactions as well as on the geometrical properties of the system.

We shall define a function $f(\nu_p)$ in terms of the intensity function as follows

$$f(\nu_p) = \frac{I(\nu_p)}{\sum_{p'=1}^{\theta} I(\nu_{p'})} \qquad (p = 1,2,\ldots\theta) \qquad (1\text{-}1)$$

We shall call $f(\nu_p)$ the normalized discrete frequency function at frequency ν_p corresponding to a component

---

* See note at the end of this section.

line in the band.

When the interaction is very weak, the band is very narrow. If, in addition, the interactions are such that the number of component lines present in the band is very large, we can approximate the discrete distribution of intensity function with a continuous one, i.e.,

$$\sum_{p=1}^{\theta} I(\nu_p) \approx \int I(\nu)\, d\nu \tag{1-2}$$

On the other hand, when the absorption band is very narrow, we can still call it a line but it now has a "shape". The "shape" of the line is then characterized by a normalized continuous frequency function known as "shape function" defined as follows

$$f(\nu) = \frac{I(\nu)}{\int I(\nu)\, d\nu} \tag{1-3}$$

<u>Note</u>

We shall use the following notations consistently:

ν_0, $\mathcal{E}_0$ correspond to Zeeman transition frequency and transition energy ,

ν_p, $\mathcal{E}_p$ correspond to a particular component in the absorption band ,

ν, $\mathcal{E}$ correspond to any component in the absorption band without specification; also used when we want to speak about the frequency distribution as a whole.

SECTION II

Statements of the Problem in General Terms and the Description of One-Dimensional Ising Model

Let us describe more explicitly the spin-system which is discussed in McMillan and Opechowski's paper (McMillan and Opechowski 1960).

The spin-system consists of an arbitrarily large number N of identical spins forming a three-dimensional rigid lattice. The properties of each spin are determined by a spin-Hamiltonian $\mathcal{H}_i^o$, which incorporates the effect of the external constant magnetic field $\vec{H}$ and of the crystalline electrostatic field.

The total Hamiltonian of the system is then given by

$$\mathcal{H} = \mathcal{H}^o + \mathcal{H}' + \mathcal{H}'' \tag{2-1}$$

where

$$\mathcal{H}^o = \sum_{i=1}^{N} \mathcal{H}_i^o \tag{2-2}$$

The Hamiltonian $\mathcal{H}'$ describes the spin-spin interactions which include magnetic dipole-dipole interactions as well as exchange interactions. $\mathcal{H}''$ describes the effect of the high frequency oscillating magnetic field $\vec{H}_1$ on the system.

We shall assume that the spin-spin interactions are weak, i.e.,

$$\mathcal{H}^0 \gg \mathcal{H}' \tag{2-3}$$

and also

$$\mathcal{H}' \gg \mathcal{H}'' \tag{2-4}$$

That is, the weak oscillating magnetic field $\vec{H}_1$ only induces transitions, its effect on the energy levels of the system is entirely negligible (see p.2).

Thus the Hamiltonian of the system can be written approximately as

$$\mathcal{H} = \mathcal{H}^0 + \mathcal{H}' \tag{2-5}$$

We shall call $\mathcal{H}^0$ the unperturbed Hamiltonian and $\mathcal{H}'$ the perturbation.

In this case, due to the presence of the weak perturbation $\mathcal{H}'$ and the huge number N of spins in the system, the discrete frequency function (1-1) can be taken, to a very good approximation, as a continuous one (1-3).

The calculation of $f(\nu)$ for this system is a very difficult and still unsolved problem. Since we know from mathematical statistics that a distribution function, such as the line shape function $f(\nu)$ in the present case, is uniquely determined by its moment generating function, instead of trying to obtain directly $f(\nu)$, we calculate its moments which are defined as follows

$$\langle \nu^{\ell} \rangle = \int_0^{\infty} \nu^{\ell} f(\nu)\, d\nu \tag{2-6}$$

where $\ell = 1, 2 \ldots$ give the first, second ... moments of $f(\nu)$. The moments about the frequency ν_0 of Zeeman transition energy $\mathcal{E}_0$ are given by

$$\langle \Delta \nu^{\ell} \rangle = \langle (\nu - \nu_0)^{\ell} \rangle \tag{2-7}$$

The formulae for the first and second moments have been obtained by various authors (see Van Vleck 1948, Pryce and Stevens 1950, Kambe and Usui 1952 and McMillan and Opechowski 1960). They are of the following forms (see McMillan and Opechowski)

$$h \langle \Delta \nu' \rangle = C^{(1)} / B \tag{2-8a}$$

$$h \langle \Delta\nu^2 \rangle = D^{(1)} / B \tag{2-8b}$$

where B, $C^{(1)}$ and $D^{(1)}$ are expressions containing the Boltzmann factor $exp(-\frac{P_\lambda \mathcal{H} P_\lambda}{KT})$ where P_λ is a projection operator defined as follows

$$P_\lambda |\mu, K\rangle = \delta_{\lambda,\mu} |\lambda, K\rangle \tag{2-9}$$

and $|\mu, K\rangle$ satisfies the eigenvalue equation

$$\mathcal{H}^0 |\mu, K\rangle = E_\mu |\mu, K\rangle \tag{2-10}$$

(k = 1,2, ... g_μ, the degeneracy of E_μ)

These moments characterize sufficiently the position and shape of a line shape function $f(\nu)$ of a paramagnetic resonance to make possible a comparison of the theory with experimental data.

In order to evaluate these moments, McMillan and Opechowski employed the following method of approximation. Since $\mathcal{H}^0 \gg \mathcal{H}''$ (2-3), they expanded the Boltzmann factors appearing in (2-8a) and (2-8b) in the following way

$$e^{-\frac{P_\lambda \mathcal{H} P_\lambda}{KT}} = e^{-\frac{P_\lambda \mathcal{H}^0 P_\lambda}{KT}} \left[1 - \frac{P_\lambda \mathcal{H}' P_\lambda}{KT} + \frac{1}{2}\left(\frac{P_\lambda \mathcal{H}' P_\lambda}{KT}\right)^2 - \cdots \right] \qquad (2\text{-}11)$$

We shall call the approximation consisting of keeping only the first term in the square bracket "the first approximation" and so on. In this way the expressions for the first and second moments in both the first and the second approximation have been obtained (see McMillan and Opechowski 1960).

The calculation based on this method of approximation may seem, at first sight, to be entirely incorrect at very low temperature. In equation (2-11), as T tends to zero, $\frac{1}{kT}$ tends to infinity, hence the terms in the square bracket containing this factor goes to infinity and thus the method of approximation seems to be inappropriate at very low temperature. However, the approximation may still be good. The presence of the Boltzmann factors in both the numerators and the denominators of (2-8a) and (2-8b) as well as in (1-1) or (1-3) indicates that at very low temperature, the terms contained in the square bracket in (2-11) in the numerator and denominator may to a good approximation cancel each other.

This thesis is primarily concerned with the investigation of this rather queer, low temperature behaviour of the line shape function $f(\nu)$.

To investigate the low temperature behaviour of $f(\nu)$ for the system discussed by McMillan and Opechowski is by no means easy. For this reason it may be useful to discuss a much simpler system such as a one-dimensional Ising model (Ising 1925) in the hope that we can throw some light on the low temperature behaviour of $f(\nu)$ in the actual problem.

Apart from the simplification of the three-dimensional to one-dimensional lattice of the system of N spins, the employment of a one-dimensional Ising model consists of the following modifications to the model discussed by McMillan and Opechowski (see p. 7 of this thesis):

(a) the angular momentum quantum number S of a spin is taken as S = 1/2 ,

(b) the crystalline electrostatic field is omitted ,

(c) the dipole-dipole interactions are omitted ,

(d) the isotropic exchange interactions J_I

$$J_I = \text{Constant} \cdot \sum_{\ell,m} \sum_{\lambda=x,y,z} S_{\ell,\lambda} S_{m,\lambda}$$

(ℓ ,m labels the spins in the system and $\sum_{\ell,m}$ means summation over all neighbouring pairs of spins)

are replaced by highly anisotropic exchange interaction J_A

$$J_A = \text{Constant} \sum_{\ell,m} S_{\ell,z} S_{m,z}$$

Since we are dealing with one-dimensional Ising model, an adjacent pair of spins can be labelled as ℓ and $\ell + 1$ respectively. Therefore

$$J_A = \text{Constant} \sum_{\ell} S_{\ell,z} S_{\ell+1,z}$$

where $\sum_{\ell}$ means summation over all adjacent pairs of spins.

In order to write down the general expression of the energy levels for this one-dimensional Ising model, we shall use the following terminology: We shall call a spin positive if its magnetic moment vector $\vec{\mu}$ is parallel to the constant external magnetic field $\vec{H}$, negative if it is antiparallel. We shall assume that the exchange energy for an adjacent pair of spins is positive if the spins are antiparallel, and negative if otherwise.

We introduce the following notations:

N = total number of spins in the system

$\vec{H}$ = external constant magnetic field

V_1 = number of positive spins in the system

V_2 = number of negative spins in the system

Q_{+-} = number of adjacent pairs of antiparallel spins

Q = total number of adjacent pairs of spins

J = positive constant of exchange interaction

μ = magnetic moment of individual spins

The general expression for the energy levels of the system is then given by

$$E = \mu H (\nu_2 - \nu_1) + J Q_{+-} - J (Q - Q_{+-})$$

or

$$E = \mu H (2\nu_2 - N) - J (Q - 2Q_{+-}) \qquad (2\text{-}12)$$

where $\mu H (2\nu_2 - N)$ and $-J(Q - 2Q_{+-})$ are respectively the Zeeman and exchange energy of the system.

Due to the frequent appearance of the Boltzmann factor in the following work, we shall consistently use the abbreviations

$$b = e^{-\frac{1}{KT}} \qquad \text{and} \qquad b^{x} = e^{-\frac{x}{KT}}$$

The intensity function at the transition energy $\mathcal{E}_p$ can be written as follows

$$I(\mathcal{E}_p) = \sum_{n,n'} \left(b^{E_{n'}} - b^{E_n}\right) W(n,n') \qquad (2\text{-}13)$$

$$(p = 1 \ldots \theta)$$

where

$W(n,n')$ = number of permissible transitions from the level E_n to the level $E_{n'}$, where the Zeeman energy of level E_n is greater than the Zeeman energy of level $E_{n'}$

and

$\sum_{n,n'}$ = summation over all pairs of n,n' for which $|E_n - E_{n'}| = \mathcal{E}_p > 0$

The frequency function (1-1) at $\nu_p = \mathcal{E}_p / h$ can then be written as

$$f(\mathcal{E}_p) = \frac{I(\mathcal{E}_p)}{\sum_{p'=1}^{\theta} I(\mathcal{E}_{p'})} \qquad (2\text{-}14)$$

$$(p = 1 \cdots \theta)$$

Before proceeding, we must say a few words about a special feature of the one-dimensional Ising model. In this case the only interactions present among the spins are the exchange interactions. The number θ of the component lines in the discrete absorption band is indeed very much limited. The reason for this will be explained in detail in Section IV. It is due to this fact that we must not replace the summation over $\mathcal{E}_{p'}$ in (2-14) with an integration over $\mathcal{E}$ in contrast to what we did at the end of Section I.

The ℓ^{th} moment of $f(\mathcal{E})$ about the Zeeman transition energy $\mathcal{E}_o$ is given by (see (2-6) and (2-7))

$$h^{\ell}\langle \Delta\gamma^{\ell}\rangle = \sum_{p=1}^{\theta} (\mathcal{E}_p - \mathcal{E}_o)^{\ell} f(\mathcal{E}_p) \tag{2-15}$$

SECTION III

One-Dimensional Ising Model (Open-Chain) for N=4

As a sample calculation, we shall take a one-dimensional open-chain Ising model consisting of only four spins and calculate the frequency function $f(\mathcal{E}_p)$, the first and the second moments about $\mathcal{E}_0$, the Zeeman transition energy, in the exact expression, first approximation and second approximation at various temperatures.

In this case, since N is small, we can construct the energy spectrum explicitly as shown in Fig. (3-1). Simply by counting all the permissible transitions among the various energy levels, we see that there exist only five distinct values of transition energy, namely:

$$2\mu H \pm 4J \, , \quad 2\mu H \pm 2J \, , \quad 2\mu H \, . \tag{3-2}$$

The reason for this will be explained in general terms in the next section.

The McMillan and Opechowski's expansion (2-11) now takes the following form:

$$b^{E} = b^{\mu H(2V_2-4)}\left[1 + \frac{J(Q-2Q_{+-})}{KT} + \cdots\right] \tag{3-3}$$

We shall assume $\mu H = 20J$.

By using the equations (2-12), (2-13), (2-14) for $f(\mathcal{E}_p)$ and equation (2-15) for the moments of $f(\mathcal{E})$, the exact values of frequency function $f(\mathcal{E}_p)$ at the transition energies (3-2) and the first and second moments of $f(\mathcal{E})$ about

$$\mathcal{E}_o = 2\mu H \tag{3-4}$$

are calculated at the temperatures

$$T \to \infty,\ T = 20J/K,\ T = 2J/K,\ T \to 0 \tag{3-5}$$

The values in the first and second approximation are also calculated by making use of the equation (3-3).

The results were tabulated in Table (3-6) for comparison. A graph (3-7) was plotted to show the general features of the temperature dependence of the function $f(\mathcal{E})$, in the exact calculation, the first approximation as well as the second approximation.

Our calculation shows that for a one-dimensional open-chain Ising model consisting of four spins, McMillan and Opechowski's method of approximation, in general, holds satisfactorily. At very low temperature, that is, when KT is of the order of exchange constant J, the approximate values become so close to the exact values that the deviation is really entirely negligible.

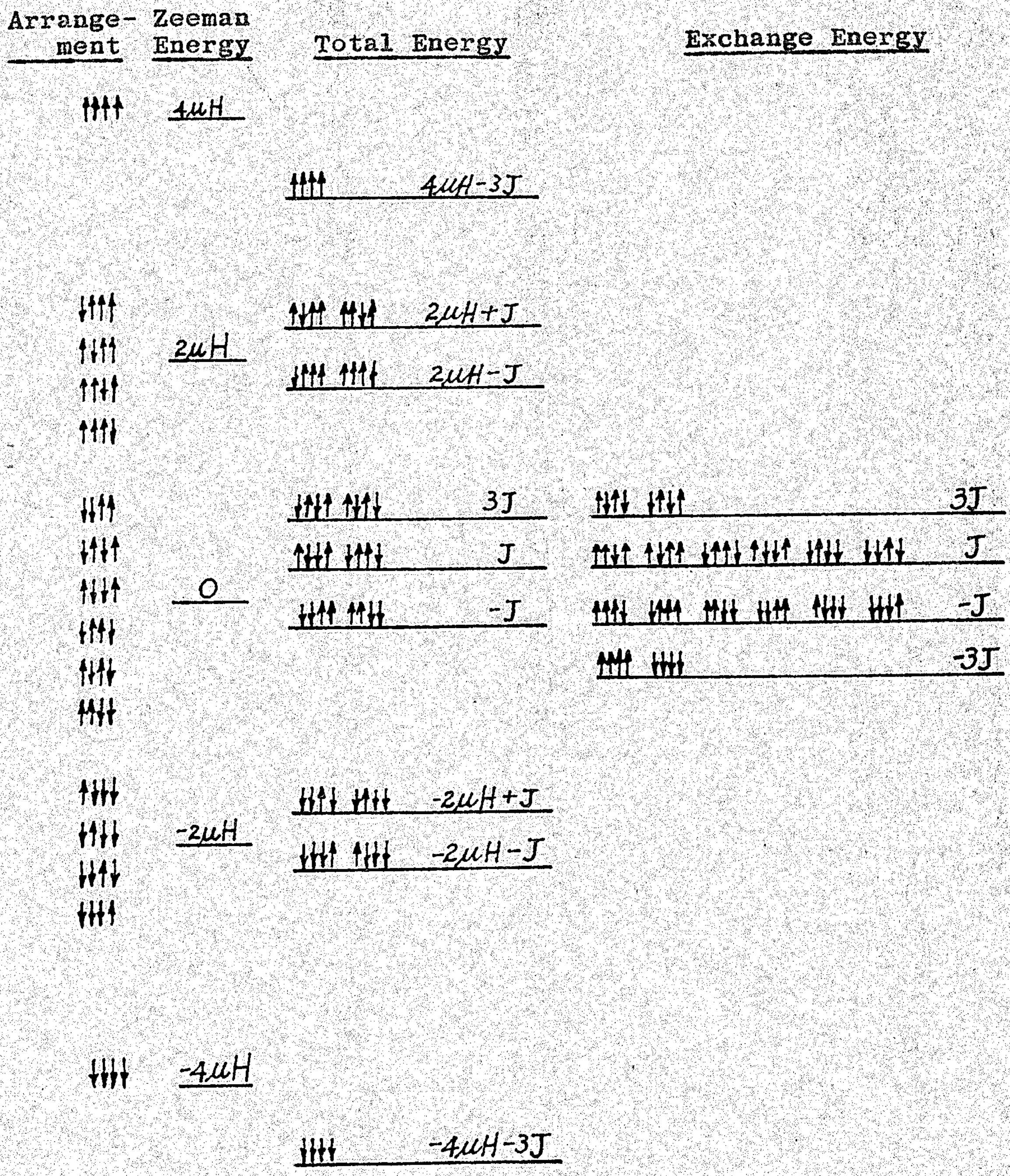

Fig. (3-1) Energy levels and transition scheme of one-dimensional Ising model (open-chain) with 4 spins.

$\mu H = 20J$

	$\mathcal{E}_p$	$T \to \infty$			$T=20J/k$			$T=2J/k$	$T \to 0$
		Exact	1st app'n	2nd app'n	Exact	1st app'n	2nd app'n	Exact 1st & 2nd app'ns*	
$f(\mathcal{E}_p)$	$2\mu H-4J$	9/80	1/8	9/80	.0053	.0071	.0053	0	0
	$2\mu H-2J$	19/80	1/4	19/80	.0490	.0596	.0495	0	0
	$2\mu H$	20/80	1/4	20/80	.0932	.1050	.0939	0	0
	$2\mu H+2J$	21/80	1/4	21/80	.4463	.4404	.4459	1/2	1/2
	$2\mu H+4J$	11/80	1/8	11/80	.4062	.3879	.4055	1/2	1/2
$h\langle\Delta\nu\rangle$		0.15J	0	0.15J	2.3982J	2.2846J	2.3948J	3J	3J
$h^2\langle\Delta\nu^2\rangle$		$6J^2$	$6J^2$	$6J^2$	$8.5652J^2$	$8.3196J^2$	$8.5496J^2$	$10J^2$	$10J^2$

Table (3-6) Results show that in general, McMillan and Opechowski's method of approximation holds satisfactorily at all temperatures.

* At temperatures below say $\frac{2J}{k}$, the differences between approximate and exact values are too small to be shown here.

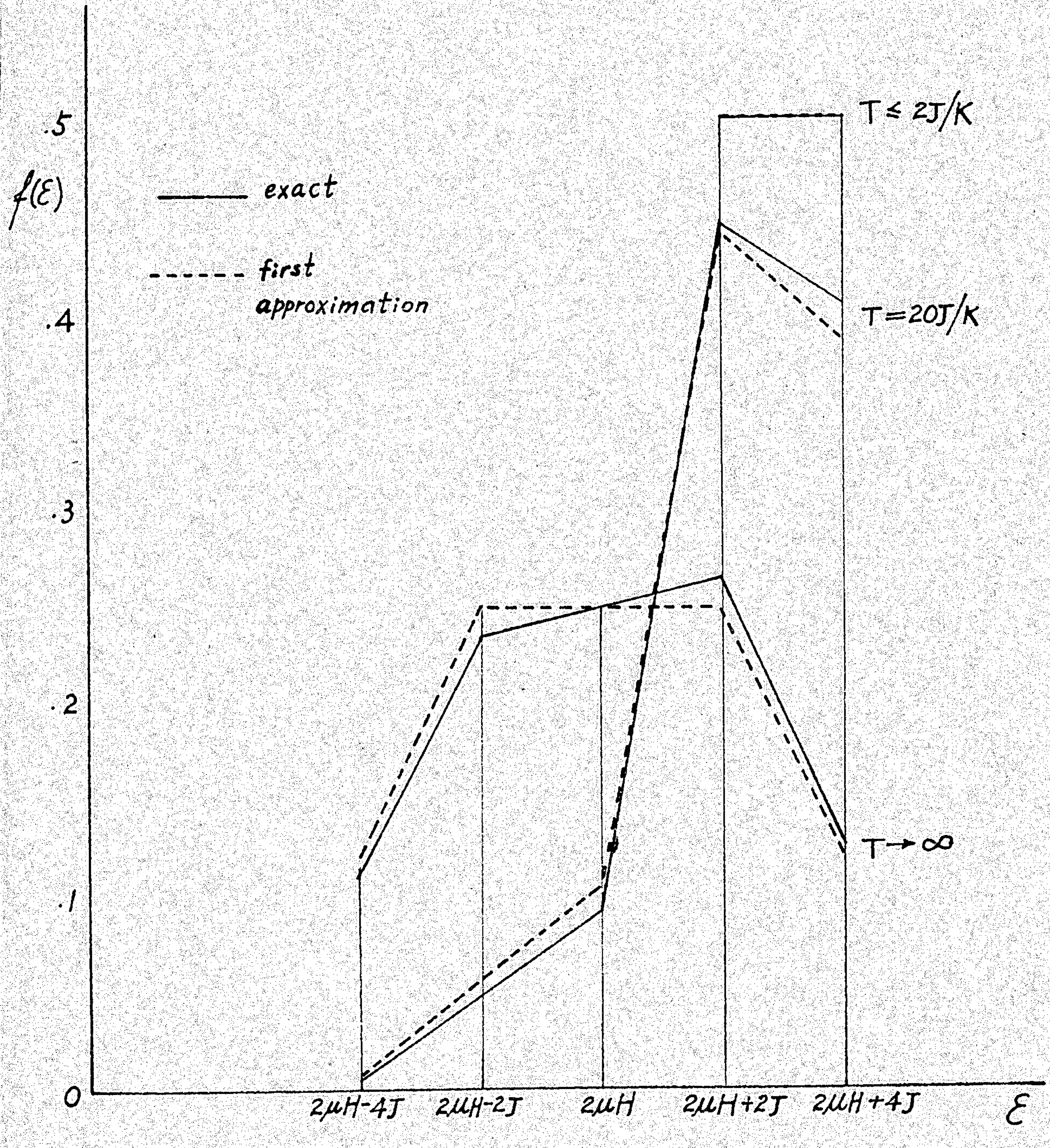

Fig. (3-7) Comparison of frequency function in the exact and first approximation. The 2nd approximation values nearly coincide the exact values and are not shown.

SECTION IV

One-Dimensional Ising Model (Ring) for Arbitrary N

For the model discussed in the previous section, we have seen that McMillan and Opechowski's method of approximation holds sufficiently well at all temperatures, and particularly so at very low temperatures. In order to investigate the low temperature behaviour of the frequency function $f(E)$ in more general terms, we shall take a one-dimensional Ising model consisting of an arbitrarily large number N of spins arranged into a ring. We shall call this model a "ring model" or simply a "ring". When N is sufficiently large, the results obtained for the ring model can be applied to the one-dimensional open-chain Ising model as well, since if N is very large, the end effect associated with an open-chain is indeed negligible.

To calculate the general expression for the frequency function $f(E)$ of a ring model, we shall use the following terminology introduced by Ising: By a "negative (positive) gap", we shall mean a series of negative (positive) spins with positive (negative) spins situated at the two ends.

Since the arrangement is a ring the number of negative gaps is always the same as the number of positive gaps in the same arrangement. The gap is single if it consists of only one spin, multiple otherwise; as shown diagramatically as follows:

```
                                   double positive gap
                             +  +
                                  -
  single negative gap      -              double negative gap
                                  -
                             +  +
                                   double positive gap


                             +  +
single negative gap        -      +       multiple positive gap
                             +  +
```

Let S be the number of negative (positive) gaps in the ring, then (see p. 14)

$$\begin{cases} Q_{+-} = 2S \\ Q = N \end{cases} \tag{4-1}$$

Equation (2-12) then becomes

$$E = \mu H(2\nu_2 - N) - J(N-4S) \tag{4-2}$$

Hence the Zeeman energy of the system is determined by S, the number of negative gaps in the ring.

Let us define

$$\begin{cases} \Delta V_2 \equiv V_2' - V_2 & (4\text{-}3) \\ \Delta S \equiv S' - S & (4\text{-}4) \\ \Delta E \equiv E' - E & (4\text{-}5) \end{cases}$$

then for a transition of the type

$$V_2 \longrightarrow V_2' = V_2 - 1 \qquad (4\text{-}6)$$

$$\text{i.e. } \Delta V_2 = -1 \qquad (4\text{-}7)$$

the change in energy Δ E can be obtained as follows: Since

$$E = \mu H (2V_2 - N) - (N - 4S)J \qquad (4\text{-}2)$$

$$E' = \mu H (2V_2' - N) - (N - 4S')J \qquad (4\text{-}2)'$$

by using (4-4), (4-5) and (4-7), we have

$$\Delta E = -2\mu H + 4(\Delta S)J \qquad (4\text{-}8)$$

Hence for each transition of the type (4-6), the Zeeman energy decreases by a fixed amount of $2\mu H$ while the exchange energy variation depends on the change in S. Thus we can label ΔE as $\Delta E_{\Delta S}$

$$\Delta E_{\Delta S} = -2\mu H + 4(\Delta S) J \tag{4-9}$$

For any transition of the type (4-6), ΔS can have only one of the three values: -1, 0, +1, that is

$$(4\text{-}9) \qquad \Delta S = \begin{cases} -1 & \text{if the transition takes place at a single negative gap;} \\ 0 & \text{if the transition takes place at either end of a multiple negative gap;} \\ +1 & \text{if the transition takes place elsewhere.} \end{cases}$$

This is shown diagramatically as follows:

```
+ + + - - - - -  + + - - + - + +
      ↑ ↑ ↑ ↑ ↑      ↑ ↑   ↑
ΔS =  0,1,1,1,0,     0,0,  -1
```

Here for convenience, we have cut up the ring into an open chain where the two end spins should be taken as adjacent spins in the original ring. The arrows point to where the transition of the type (4-6) is to occur with the value ΔS associated with such a transition.

Combining (4-8) and (4-9), we shall have the following three cases:

$$\begin{cases} \Delta S = -1 \qquad \Delta E_{-1} = -(2\mu H + 4J) & \text{(4-10a)} \\ \Delta S = 0 \qquad \Delta E_{0} = -2\mu H & \text{(4-10b)} \\ \Delta S = +1 \qquad \Delta E_{+1} = -(2\mu H - 4J) & \text{(4-10c)} \end{cases}$$

Note that in the case of a one-dimensional open-chain Ising model, there exist five distinct values of transition energy Δ E, two among which, namely $2\mu H \pm 2J$, arise from the end effect of the open-chain (see Section III). By choosing a ring instead, we have discarded the end effect; thus only three values of transition energy are left, namely: $2\mu H + 4J$, $2\mu H$, $2\mu H - 4J$. In general, for an Ising model with an arbitrary form of lattice, if the number of closest neighbours of a spin is K which is a constant, then the number of transition energy values is K + 1 provided that the number of spins in the system is sufficiently large that we can neglect the effect of the boundaries. A proof of this statement will be found in Appendix I.

From equation (4-2), the energy levels are uniquely defined by giving V_2 and S, therefore we can replace the quantum number "n" and "n'" by v_2,S and v_2',S'. Equation (2-13) thus becomes

$$I(\mathcal{E}_p) = \sum_{V_2', S'; V_2, S} \left(b^{E_{V_2', S'}} - b^{E_{V_2, S}} \right) W(V_2', S'; V_2, S) \tag{4-11}$$

Since a transition is permissible if and only if condition (4-6), namely

$$\Delta V_2 = -1$$

is satisfied, this imposes a relation between V_2' and V_2 in equation (4-11). Moreover (see (4-5), (4-10a), (4-10b) and (4-10c)),

$$\mathcal{E}_p = |\Delta E_{\Delta S}| \tag{4-12}$$

Hence for an intensity function corresponding to a particular component line in the absorption band, the values ΔV_2 and Δ S are fixed. Thus equation (4-11) reduces to the following form

$$I(|\Delta E_{\Delta S}|) = \sum_{V_2, S} \left(b^{E_{V_2-1, S+\Delta S}} - b^{E_{V_2, S}} \right) W_{\Delta S}(V_2, S) \tag{4-13}$$

where $W_{\Delta S}(V_2, S)$ denotes the number of permissible transitions from the level $E_{V_2,S}$ to the level $E_{V_2-1,S+\Delta S}$; and $\sum_{V_2,S}$ means summation over all V_2 and S such that

$$E_{V_2-1,\,S+\Delta S} - E_{V_2,S} = \Delta E_{\Delta S} \tag{4-14}$$

The subscript ΔS appearing in both $\Delta E_{\Delta S}$ and $W_{\Delta S}(V_2, S)$ specifies the particular component line we are interested in in the absorption band.

In order to calculate $W_{\Delta S}(V_2, S)$ in (4-13), we have first to know the following quantities:

A) $G(V_2,S)$ = the number of different arrangements of spins for given values of V_2 and S, i.e., the degeneracy of a given energy level $E_{V_2,S}$ (4-14)

B) $D(V_2,S)$ = the total number of single negative gaps appearing in all possible arrangements for given values of V_2 and S (4-15)

To find $G(V_2,S)$ we essentially follow Ising's method (Ising 1925). In order to have S negative gaps, we need at least S positive and S negative spins arranged into an alternating series in the form of a ring. The remaining V_1-S positive spins and V_2-S negative spins are to be distributed arbitrarily among the S positive and S negative gaps respectively. This leads to the following expression for $G(V_2,S)$

$$\begin{cases} G(V_2,S) = \frac{N}{S}\binom{V_1-1}{S-1}\binom{V_2-1}{S-1} & [V_1,V_2] \neq 0 \quad S=1\cdots[V_1,V_2] \qquad \text{(4-16a)} \\ G \qquad = 1 & [V_1,V_2] = 0 \quad S=0 \qquad \text{(4-16b)} \end{cases}$$

where the notation $[a,b]$ is defined as follows

$$[a,b] = \begin{cases} a & \text{if } a \leq b \\ b & \text{if } a > b \end{cases} \qquad \text{(4-17)}$$

The factor $\frac{N}{S}$ arises from the cyclic symmetry property of the arrangement.

To find $D(V_2,S)$, we proceed as follows:
Let "i" be the number of multiple negative gaps in a given arrangement of spins, let d_i be the number of arrangements belonging to $G(V_2S)$, each of these d_i arrangements has "i" multiple negative gaps. Then it is clear that

$$D(V_2,S) = \sum_i (S-i)\, d_i \qquad \text{(4-18)}$$

where $\sum_i$ means summation over all values of i present in the $G(V_2,S)$ arrangements.

To evaluate d_i, we must distinguish the following cases which give the relations among V_2, S and i:

(a) $V_2 > S \geqslant 2$ implies $i \geqslant 1$

Since there are more negative spins than negative gaps, some of the negative gaps must contain more than one negative spin.

(b) $V_2 > S = 1$ implies $i = 1$

Since there is only one negative gap, all the V_2 negative spins must be contained in this gap.

(c) $V_2 = S$ implies $i = 0$

This means that all the negative gaps are single. We shall label the three cases of D as D_a, D_b and D_c and shall deal with each of them as follows:

Case (a)

Since $\binom{V_1-1}{S-1}$ = number of ways to form S negative gaps with V_1 positive spins; see (4-16a)

$\binom{V_2-S-1}{i-1}$ = number of ways to form S negative gaps with V_2 negative spins, S-i gaps being single; see (4-16a)

$\binom{S}{i}$ = number of ways to choose i multiple negative gaps out of S negative gaps,

hence we have the expression for d_i

$$d_i = \frac{N}{S}\binom{S}{i}\binom{V_1-1}{S-1}\binom{V_2-S-1}{i-1} \qquad (4\text{-}19)$$

where again, the factor $\frac{N}{S}$ arises from the cyclic symmetry property of the arrangement.

Substituting (4-19) into (4-18), we obtain

$$D_a(V_2,S) = \sum_i (S-i)\frac{N}{S}\binom{S}{i}\binom{V_1-1}{S-1}\binom{V_2-S-1}{i-1} \tag{4-20}$$

where the lower limit of summation is certainly i = 1 as required by the condition $V_2 > S \geqslant 2$ which implies $i \geqslant 1$; the upper limit is $[V_2-S,\ S-1]$ as can be seen from the equation (4-20) remembering that $S \leqslant [V_1, V_2]$ (see 4-16a). Hence we have

$$D_a(V_2,S) = \sum_{i=1}^{[V_2-S,S-1]} (S-i)\frac{N}{S}\binom{S}{i}\binom{V_1-1}{S-1}\binom{V_2-S-1}{i-1} \tag{4-21a}$$

<u>Case (b)</u>

Since there exist no single negative gaps,

$$D_b(V_2,S) = 0 \tag{4-21b}$$

<u>Case (c)</u>

Since all the negative gaps are single, it is clear from the discussion of case (a) that:

$$D_c(V_2,S) = N\binom{V_1-1}{S-1} \tag{4-21c}$$

With the help of equations (4-9)(410a)(4-10b) and (4-10c), the factor $W_{\Delta S}(V_2,S)$ in the equation (4-13) can be expressed as follows:

(I) $\Delta S=-1$ $\Delta E_{-1}=-(2\mu H+4J)$

Since this type of transition takes place only at single negative gaps,

$$W_{-1}(V_2,S)=D(V_2,S) \tag{4-22a}$$

(II) $\Delta S=0$ $\Delta E_0=-2\mu H$

Since this type of transition takes place only at either end of a multiple negative gap,

$$W_0(V_2,S)=2(SG-D) \tag{4-22b}$$

where $SG(V_2,S)$ is the total number of negative gaps and $SG(V_2,S)-D(V_2,S)$ is the total number of multiple negative gaps for given V_2 and S.

(III) $\Delta S=+1$ $\Delta E_{+1}=-(2\mu H-4J)$

Since this type of transition takes place "elsewhere" and there are altogether $V_2G(V_2,S)$ possible transitions for given V_2 and S,

$$W_{+1}(V_2, S) = V_2 G - W_{-1} - W_0 = (V_2 - 2S)G + D \qquad (4\text{-}22c)$$

where

$$D(V_2,S) = \begin{cases} D_a(V_2,S) & \text{if} \quad V_2 > S \geqslant 2 \\ D_b(V_2,S) & \text{if} \quad V_2 > S = 1 \\ D_c(V_2,S) & \text{if} \quad V_2 = S \end{cases} \qquad (4\text{-}23)$$

Combining (4-10a,b,c)(4-13)(4-14) and (4-22a,b,c) we have the following set of expressions for the intensity function:

$$\Delta E_{-1} = -(2\mu H + 4J)$$

$$I(2\mu H + 4J) = \sum_{V_2,S}\left[b^{\mu H(2V_2-2-N)-(N-4S+4)J} - b^{\mu H(2V_2-N)-(N-4S)J}\right] D \qquad (4\text{-}24a)$$

$$\Delta E_0 = -2\mu H$$

$$I(2\mu H) = \sum_{V_2,S}\left[b^{\mu H(2V_2-2-N)-(N-4S)J} - b^{\mu H(2V_2-N)-(N-4S)J}\right] \times$$

$$\times 2 \times (SG - D) \qquad (4\text{-}24b)$$

$$\Delta E_{+1} = -(2\mu H - 4J)$$

$$I(|2\mu H - 4J|) = \sum_{V_2,S}\left[b^{\mu H(2V_2-2-N)-(N-4S-4)J} - b^{\mu H(2V_2-N)-(N-4S)J}\right] \times$$

$$\times\left[(V_2 - 2S)G + D\right] \qquad (4\text{-}24c)$$

Since $S \leq [V_1, V_2] \equiv [N - V_2, V_2]$, the upper limits of summation over S in the above set of equations depend on whether $V_2 > \frac{N}{2}$ or $V_2 < \frac{N}{2}$. If N is even, there is a case where $V_2 = \frac{N}{2}$, hence the problem is slightly more complicated than in the case of N odd. For sufficiently large N, the intensity function will not depend on whether N is even or odd. Therefore we will assume, in the remainder of this thesis, that N is odd.

By substituting equations (4-23)(4-21a)(4-21b) and (4-21c) into equations (4-22a)(4-22b) and (4-22c) remembering that N is odd, we obtain the following set of expressions:

$$I(2\mu H + 4J) = (1 - b^{2\mu H + 4J}) \times b^{-(N+2)\mu H - (N+4)J} \times N \times$$

$$\times \left\{ \sum_{V_2 = \frac{N+1}{2}}^{N-2} \sum_{S=2}^{N-V_2} b^{2V_2\mu H + 4SJ} \frac{1}{S} \binom{N-V_2-1}{S-1} \sum_{i=1}^{[V_2-S,\, S-1]} (S-i) \binom{S}{i} \binom{V_2-S-1}{i-1} \right.$$

$$+ \sum_{V_2=3}^{\frac{N-1}{2}} \sum_{S=2}^{V_2-1} b^{2V_2\mu H + 4SJ} \frac{1}{S} \binom{N-V_2-1}{S-1} \sum_{i=1}^{[V_2-S,\, S-1]} (S-i) \binom{S}{i} \binom{V_2-S-1}{i-1}$$

$$\left. + \sum_{V_2=1}^{\frac{N-1}{2}} b^{2V_2\mu H + 4V_2 J} \binom{N-V_2-1}{V_2-1} \right\}$$

(4-25a)

$$
\begin{aligned}
I(2\mu H) = {} & \left(1 - b^{2\mu H}\right) \times b^{-(N+2)\mu H - NJ} \times 2N \times \\
\times \Bigg\{ & \sum_{V_2=\frac{N+1}{2}}^{N-2} \sum_{S=2}^{N-V_2} b^{2V_2\mu H + 4SJ} \binom{N-V_2-1}{S-1} \left[\binom{V_2-1}{S-1} - \frac{1}{S} \sum_{i=1}^{[V_2-S,S-1]} (S-i) \binom{S}{i} \binom{V_2-S-1}{i-1} \right] \\
& + \sum_{V_2=\frac{N+1}{2}}^{N-1} b^{2V_2\mu H + 4J} \\
& + \sum_{V_2=3}^{\frac{N-1}{2}} \sum_{S=2}^{V_2-1} b^{2V_2\mu H + 4SJ} \binom{N-V_2-1}{S-1} \left[\binom{V_2-1}{S-1} - \frac{1}{S} \sum_{i=1}^{[V_2-S,S-1]} (S-i) \binom{S}{i} \binom{V_2-S-1}{i-1} \right] \\
& \sum_{V_2=2}^{\frac{N-1}{2}} b^{2V_2\mu H + 4J} \Bigg\}
\end{aligned}
\tag{4-25b}
$$

$$
\begin{aligned}
I(|2\mu H - 4J|) = {} & \left(1 - b^{2\mu H - 4J}\right) \times N \times b^{-(N+2)\mu H - (N-4)J} \times \\
\times \Bigg\{ & \sum_{V_2=\frac{N+1}{2}}^{N-2} \sum_{S=2}^{N-V_2} b^{2V_2\mu H + 4SJ} \frac{1}{S} \binom{N-V_2-1}{S-1} \left[(V_2-2S) \binom{V_2-1}{S-1} + \sum_{i=1}^{[V_2-S,S-1]} (S-i) \binom{S}{i} \binom{V_2-S-1}{i-1} \right] \\
& + \sum_{V_2=\frac{N+1}{2}}^{N-1} b^{2V_2\mu H + 4J} (V_2-2) \\
& + b^{2N\mu H} \\
& + \sum_{V_2=3}^{\frac{N-1}{2}} \sum_{S=2}^{V_2-1} b^{2V_2\mu H + 4SJ} \frac{1}{S} \binom{N-V_2-1}{S-1} \left[(V_2-2S) \binom{V_2-1}{S-1} + \sum_{i=1}^{[V_2-S,S-1]} (S-i) \binom{S}{i} \binom{V_2-S-1}{i-1} \right] \\
& + \sum_{V_2=3}^{\frac{N-1}{2}} b^{2V_2\mu H + 4J} (V_2-2) \Bigg\}
\end{aligned}
\tag{4-25C}
$$

In order to determine the upper limit of summation over i, we proceed as follows:

Let $V_2 \equiv 2j$ for even V_2, then

$$\begin{cases} 2j-S < S-1 & \text{if} \quad S \geq \frac{V_2+2}{2} \equiv j+1 \\ 2j-S > S-1 & \text{if} \quad S \leq \frac{V_2}{2} \equiv j \end{cases} \tag{4-26a}$$

and $V_2 \equiv 2j+1$ for odd V_2, then

$$\begin{cases} 2j-S+1 < S-1 & \text{if} \quad S \geq \frac{V_2+3}{2} \equiv j+2 \\ 2j-S+1 = S-1 & \text{if} \quad S = \frac{V_2+1}{2} \equiv j+1 \\ 2j-S+1 > S-1 & \text{if} \quad S \leq \frac{V_2-1}{2} \equiv j \end{cases} \tag{4-26b}$$

No confusion will arise by using the same letter "j" to express both even and odd V_2, since we shall replace the summation over V_2 by a summation over even V_2 and a summation over odd V_2. This is necessary because the upper limits of summation over i are different in the two cases as seen from equations (2-26a) and (2-26b). In carrying out this replacement, however, we have to specify whether $V_2 = \frac{N+1}{2}$, the lower limit of summation $\sum\limits_{V_2=\frac{N+1}{2}}^{N-2}$, as well as $V_2 = \frac{N-1}{2}$, the upper limit of summation $\sum\limits_{V_2=3}^{\frac{N-1}{2}}$, is even or odd. For this reason we shall assume that N has the following particular form

$$N \equiv 4p-1 \quad (p=1,2\cdots) \tag{4-27}$$

This particular choice of N will certainly not affect the generality of our results thus obtained provided that N is sufficiently large.

This choice of N leads to the following set of simple yet important relationships:

$$\begin{cases} \frac{N+1}{2} \equiv 2p \text{ even} & V_2 \equiv 2j \quad \therefore \; V_2 = \frac{N+1}{2} \Rightarrow j=p \\ \frac{N+3}{2} \equiv 2p+1 \text{ odd} & V_2 \equiv 2j+1 \quad V_2 = \frac{N+3}{2} \Rightarrow j=p \end{cases} \tag{4-28a}$$

$$\begin{cases} \frac{N-1}{2} \equiv 2p-1 \text{ odd} & V_2 \equiv 2j+1 \quad V_2 = \frac{N-1}{2} \Rightarrow j=p-1 \\ \frac{N-3}{2} \equiv 2p-2 \text{ even} & V_2 \equiv 2j \quad V_2 = \frac{N-3}{2} \Rightarrow j=p-1 \end{cases} \tag{4-28b}$$

By using (4-26a)(4-26b)(4-28a) and (4-28b) we obtain the following set of replacements: whenever we have in a formula a sum

$$\sum_{V_2=\frac{N+1}{2}}^{N-2} \sum_{s=2}^{N-V_2} \sum_{i=1}^{[V_2-s,\, s-1]}$$

We shall replace it by

$$\underbrace{\sum_{j=p}^{2p-2}\left(\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{s=j+1}^{4p-2j-1}\sum_{i=1}^{2j-s}\right)}_{V_2 \text{ even}}+\underbrace{\sum_{j=p}^{2p-2}\left(\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{s=j+2}^{4p-2j-2}\sum_{i=1}^{2j-s-1}+\sum_{s=j+1}\sum_{i=1}^{j}\right)}_{V_2 \text{ odd}}$$

(4-29a)

Similarly we shall replace

$$\sum_{V_2=3}^{\frac{N-1}{2}}\sum_{s=2}^{V_2-1}\sum_{i=1}^{[V_2-s,\,s-1]}$$

by

$$\underbrace{\sum_{j=2}^{p-1}\left(\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{s=j+1}^{2j-1}\sum_{i=1}^{2j-s}\right)}_{V_2 \text{ even}}+\underbrace{\sum_{j=2}^{p-1}\left(\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{s=j+2}^{2j}\sum_{i=1}^{2j-s-1}\right)+\sum_{j=1}^{p-1}\sum_{s=2j+1}\sum_{i=1}^{j}}_{V_2 \text{ odd}}$$

(4-29b)

After making use of expressions (4-29a) and (4-29b) equations (4-25a,b,c) become:

$$I(2\mu H+4J)=\left(1-b^{2\mu H+4J}\right)\times b^{-(N+2)\mu H-(N+4)J}\times N\times$$

$$\times\Bigg\{\sum_{j=2}^{2P-2}\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{j=P}^{2P-2}\sum_{s=j+1}^{4P-2j-1}\sum_{i=1}^{2j-s}+\sum_{j=2}^{P-1}\sum_{s=j+1}^{2j-1}\sum_{i=1}^{2j-s} b^{4j\mu H+4sJ}\frac{1}{s}\binom{4P-2j-2}{s-1}(s-i)\binom{s}{i}\binom{2j-s-1}{i-1}$$

$$+\sum_{j=2}^{2P-2}\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{j=P}^{2P-2}\sum_{s=j+2}^{4P-2j-2}\sum_{i=1}^{2j-s+1}+\sum_{j=2}^{P-1}\sum_{s=j+2}^{2j}\sum_{i=1}^{2j-s+1} b^{(4j+2)\mu H+4sJ}\frac{1}{s}\binom{4P-2j-3}{s-1}(s-i)\binom{s}{i}\binom{2j-s}{i-1}$$

$$+\sum_{j=1}^{2P-2}\sum_{i=1}^{j} b^{(4j+2)\mu H+4(j+1)J}\frac{1}{j+1}\binom{4P-2j-3}{j}(j-i+1)\binom{j+1}{i}\binom{j-1}{i-1}$$

$$+\sum_{V_2=1}^{2P-1} b^{2V_2\mu H+4V_2J}\binom{4P-V_2-2}{V_2-1}\Bigg\} \qquad (4\text{-}30a)$$

$$I(2\mu H)=\left(1-b^{2\mu H}\right)\times b^{-(N+2)\mu H-NJ}\times 2N\times$$

$$\times\Bigg\{\sum_{j=2}^{2P-2}\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{j=P}^{2P-2}\sum_{s=j+1}^{4P-2j-1}\sum_{i=1}^{2j-s}+\sum_{j=2}^{P-1}\sum_{s=j+1}^{2j-1}\sum_{i=1}^{2j-s} b^{4j\mu H+4sJ}\binom{4P-2j-2}{s-1}\left[\binom{2j-1}{s-1}-\frac{1}{s}(s-i)\binom{s}{i}\binom{2j-s-1}{i-1}\right]$$

$$+\sum_{j=2}^{2P-2}\sum_{s=2}^{j}\sum_{i=1}^{s-1}+\sum_{j=P}^{2P-2}\sum_{s=j+2}^{4P-2j-2}\sum_{i=1}^{2j-s+1}+\sum_{j=2}^{P-1}\sum_{s=j+2}^{2j}\sum_{i=1}^{2j-s-1} b^{(4j+2)\mu H+4sJ}\binom{4P-2j-3}{s-1}\left[\binom{2j}{s-1}-\frac{1}{s}(s-i)\binom{s}{i}\binom{2j-s}{i-1}\right]$$

$$+\sum_{j=1}^{2P-2}\sum_{i=1}^{j} b^{(4j+2)\mu H+4(j+1)J}\binom{4P-2j-3}{j}\left[\binom{2j}{j}-\frac{j-i+1}{j+1}\binom{j+1}{i}\binom{j-1}{i-1}\right]$$

$$+\sum_{V_2=2}^{4P-2} b^{2V_2\mu H+4J}\Bigg\} \qquad (4\text{-}30b)$$

$$I(|2\mu H-4J|) = (1-b^{2\mu H-4J}) \times N \times b^{-(N+2)\mu H-(N-4)J} \times$$

$$\times \Bigg\{ \sum_{j=2}^{2P-2} \sum_{S=2}^{j} \sum_{i=1}^{S-1} + \sum_{j=P}^{2P-2} \sum_{S=j+1}^{4P-2j-1} \sum_{i=1}^{2j-S} + \sum_{j=2}^{P-1} \sum_{S=j+1}^{2j-1} \sum_{i=1}^{2j-S} b^{4j\mu H+4SJ} \frac{1}{S} \binom{4P-2j-2}{S-1} \left[(2j-2S)\binom{2j-1}{S-1} + (S-i)\binom{S}{i}\binom{2j-S-1}{i-1} \right]$$

$$+ \sum_{j=2}^{2P-2} \sum_{S=2}^{j} \sum_{i=1}^{S-1} + \sum_{j=P}^{2P-2} \sum_{S=j+2}^{4P-2j-2} \sum_{i=1}^{2j-S+1} \sum_{j=2}^{P-1} \sum_{S=j+2}^{2j} \sum_{i=1}^{2j-S+1} b^{(4j+2)\mu H+4SJ} \frac{1}{S} \binom{4P-2j-3}{S-1} \left[(2j-2S)\binom{2j}{S-1} + (S-i)\binom{S}{i}\binom{2j-S}{i-1} \right]$$

$$+ \sum_{j=2}^{2P-2} \sum_{i=1}^{j} \qquad b^{(4j+2)\mu H+4(j+1)J} \frac{1}{j+1} \binom{4P-2j-3}{j} \left[-\binom{2j}{j} + (j-i+1)\binom{j+1}{i}\binom{j-1}{i-1} \right]$$

$$+ \sum_{V_2=3}^{4P-2} \qquad b^{2V_2\mu H+4J} \quad (V_2-2)$$

$$+ \qquad b^{2N\mu H} \Bigg\} \qquad (4\text{-}30C)$$

The summations over i appearing in the above set of expressions can be reduced by applying combinatorial analysis. The fundamental equalities we are going to use are the following:

$$\binom{n}{r} = \binom{n-1}{r} + \binom{n-1}{r-1} \qquad \textbf{(4-31a)}$$

and

$$\sum_{i=0}^{p} \binom{p}{i}\binom{q}{i} = \binom{p+q}{p} \qquad \textbf{(4-31b)}$$

We observe that:

$$(s-i)\binom{s}{i} = s\binom{s}{i} - s\binom{s-1}{i-1} = s\binom{s-1}{i}$$

then we have, for example:

$$\sum_{i=1}^{s-1}(s-i)\binom{s}{i}\binom{2j-s-1}{i-1} = \sum_{i=1}^{s-1} s\binom{s-1}{i}\left[\binom{2j-s}{i} - \binom{2j-s-1}{i}\right] = s\left[\binom{2j-1}{s-1} - \binom{2j-2}{s-1}\right] = s\binom{2j-2}{s-2}$$

Following the same procedure, all the other summations over i can be performed. After some simple manipulations we arrive at the following forms:

$$I(2\mu H + 4J) = (1 - b^{2\mu H + 4J}) \times b^{-(N+2)\mu H - (N+4)J} \times N \times$$

$$\times \left\{ \sum_{j=2}^{2p-2}\sum_{s=2}^{j} + \sum_{j=p}^{2p-2}\sum_{s=j+1}^{4p-2j-1} + \sum_{j=2}^{p-1}\sum_{s=j+1}^{2j-1} b^{4j\mu H + 4sJ}\binom{4p-2j-2}{s-1}\binom{2j-2}{s-2} \right.$$

$$+ \sum_{j=2}^{2p-2}\sum_{s=2}^{j} + \sum_{j=p}^{2p-2}\sum_{s=j+2}^{4p-2j-2} + \sum_{j=2}^{p-1}\sum_{s=j+2}^{2j} b^{(4j+2)\mu H + 4sJ}\binom{4p-2j-3}{s-1}\binom{2j-1}{s-2}$$

$$+ \sum_{j=1}^{2p-2} b^{(4j+2)\mu H + 4(j+1)J}\binom{4p-2j-3}{j}\binom{2j-1}{j-1}$$

$$\left. + \sum_{V_2=1}^{2p-1} b^{2V_2\mu H + 4V_2 J}\binom{4p-V_2-2}{V_2-1} \right\}$$

(4-32a)

$$I(2\mu H) = (1-b^{2\mu H}) \times b^{-(N+2)\mu H - NJ} \times 2N \times$$

$$\times \Bigg\{ \sum_{j=2}^{2p-2} \sum_{s=2}^{j} + \sum_{j=p}^{2p-2} \sum_{s=j+1}^{4p-2j-1} + \sum_{j=2}^{p-1} \sum_{s=j+1}^{2j-1} b^{4j\mu H + 4sJ} \binom{4p-2j-2}{s-1} \binom{2j-2}{s-1}$$

$$+ \sum_{j=2}^{2p-2} \sum_{s=2}^{j} + \sum_{j=p}^{2p-2} \sum_{s=j+2}^{4p-2j-2} + \sum_{j=2}^{p-1} \sum_{s=j+2}^{2j} b^{(4j+2)\mu H + 4sJ} \binom{4p-2j-3}{s-1} \binom{2j-1}{s-1}$$

$$+ \sum_{j=1}^{2p-2} b^{(4j-2)\mu H + 4(j+1)J} \binom{4p-2j-3}{j} \binom{2j-1}{j}$$

$$+ \sum_{v_2=2}^{4p-2} b^{2v_2 \mu H + 4J} \Bigg\}$$

(4-32b)

$$I(|2\mu H - 4J|) = (1-b^{2\mu H - 4J}) \times b^{-(N+2)\mu H - (N-4)J} \times N \times$$

$$\times \Bigg\{ \sum_{j=2}^{2p-2} \sum_{s=2}^{j} + \sum_{j=p}^{2p-2} \sum_{s=j+1}^{4p-2j-1} + \sum_{j=2}^{p-1} \sum_{s=j+1}^{2j-2} b^{4j\mu H + 4sJ} \binom{4p-2j-2}{s-1} \binom{2j-2}{s}$$

$$+ \sum_{j=2}^{2p-2} \sum_{s=2}^{j} + \sum_{j=p}^{2p-2} \sum_{s=j+2}^{4p-2j-2} + \sum_{j=2}^{p-1} \sum_{s=j+2}^{2j-1} b^{(4j+2)\mu H + 4sJ} \binom{4p-2j-3}{s-1} \binom{2j-1}{s}$$

$$+ \sum_{j=2}^{2p-2} b^{(4j+2)\mu H + 4(j+1)J} \binom{4p-2j-3}{j} \binom{2j-1}{j+1}$$

$$+ \sum_{v_2=3}^{4p-2} b^{2v_2 \mu H + 4J} (v_2 - 2)$$

$$+ b^{2N\mu H} \Bigg\}$$

(4-32c)

Now by rearranging the summations, applying repeatedly the equalities (4-31a) and (4-31b), and after some algebraic manipulation, it is possible to reduce the above set of expressions to the following forms:

$$I(2\mu H+4J)=(1-b^{2\mu H+4J})\times b^{-(N+2)\mu H-(N+4)J}\times N\times \left\{b^{2\mu H+4J}+\sum_{t=2}^{N-2}\sum_{s=2}^{t} b^{2t\mu H+4sJ}\binom{N-t-1}{s-1}\binom{t-2}{s-2}\right\} \qquad (4\text{-}33a)$$

$$I(2\mu H) = (1-b^{2\mu H})\times b^{-(N+2)\mu H-NJ}\times 2N\times \left\{\sum_{t=2}^{N-1}\sum_{s=1}^{t-1} b^{2t\mu H+4sJ}\binom{N-t-1}{s-1}\binom{t-2}{s-1}\right\} \qquad (4\text{-}33b)$$

$$I(|2\mu H-4J|)=(1-b^{2\mu H-4J})\times b^{-(N+2)\mu H-(N-4)J}\times N\times \left\{\sum_{t=3}^{N-1}\sum_{s=1}^{t-2} b^{2t\mu H+4sJ}\binom{N-t-1}{s-1}\binom{t-2}{s}+b^{2N\mu H}\right\} \qquad (4\text{-}33c)$$

As an independent check of these expressions we have calculated the values of intensity function for the ring model with N = 7 (or p = 2, N = 4P-1) by constructing the energy spectrum explicitly and counting all the permissible transitions among the various energy levels, as we once did for the N = 4 linear-chain model (see Section III). Results are compared with those obtained by using the expressions (4-33a)(4-33b) and (4-33c), and found to be identical.

In order to evaluate the double summations in the expressions (4-33a)(4-33b) and (4-33c), we introduce the following transformation:

$$\begin{cases} T = t - 2 \\ r = S - 1 \\ R = N - 3 \\ A = b^{2\mu H} \\ B = b^{4J} \end{cases} \tag{4-34}$$

then we have,

$$I(2\mu H+4J) = (1-b^{2\mu H+4J}) \times b^{-N(\mu H+J)} \times N \times \{1+b^{2\mu H+2J} Q_1\} \tag{4-35a}$$

$$I(2\mu H) = (1-b^{2\mu H}) \times b^{-N(\mu H+J)} \times 2N \times \{b^{2\mu H+4J} Q_2\} \tag{4-35b}$$

$$I(|2\mu H-4J|) = (1-b^{2\mu H-4J}) \times b^{-N(\mu H+J)} \times N \times \{b^{2\mu H+6J} Q_3 + b^{2(N-1)\mu H+4J}\} \tag{4-35c}$$

where

$$Q_1 = \frac{1}{\sqrt{B}} \sum_{T=0}^{R-1} A^T \sum_{r=1}^{T+1} B^r \binom{R-T}{r}\binom{T}{r-1} \tag{4-36a}$$

$$Q_2 = \sum_{T=0}^{R} A^T \sum_{r=0}^{T} B^r \binom{R-T}{r}\binom{T}{r} \tag{4-36b}$$

$$Q_3 = \sqrt{B} \sum_{T=1}^{R} A^T \sum_{r=0}^{T-1} B^r \binom{R-T}{r}\binom{T}{r+1} \tag{4-36c}$$

To evaluate Q's let us consider a generating function:

$$f_{R,T}(z,B) = (1+z\sqrt{B})^{R-T}\left(1+\frac{1}{z}\sqrt{B}\right)^T \qquad T \leq R \tag{4-37}$$

which is analytic except at the origin.

Using binomial expansion, we have

$$\begin{aligned} f_{R,T}(z,B) &= \sum_{u=0}^{R-T} \binom{R-T}{u}(\sqrt{B})^u z^u \cdot \sum_{v=0}^{T} \binom{T}{v}(\sqrt{B})^v z^{-v} \\ &= \sum_{u=0}^{R-T} \sum_{v=0}^{T} \binom{R-T}{u}\binom{T}{v}(\sqrt{B})^{u+v} z^{u-v} \\ &= \sum_{r=-T}^{R-T} \left(\delta_{u-v,r} \sum_{u=0}^{R-T} \sum_{v=0}^{T} \binom{R-T}{u}\binom{T}{v}(\sqrt{B})^{u+v}\right) z^r \end{aligned} \tag{4-38}$$

Let

$$g_{R,T}(r,B) \equiv \sum_{u=0}^{R-T} \sum_{v=0}^{T} \binom{R-T}{u}\binom{T}{v}(\sqrt{B})^{u+v} \delta_{u-v,r} \tag{4-39}$$

then

$$f_{R,T}(z,B) = \sum_{r=-T}^{R-T} g_{R,T}(r,B)\, z^r \tag{4-40}$$

and

$$r=1\ ,\quad g_{R,T}(1,B) = \sum_{u=0}^{R-T} \binom{R-T}{u}\binom{T}{u-1}(\sqrt{B})^{2u-1} = \frac{1}{\sqrt{B}}\sum_{u=1}^{T+1}\binom{R-T}{u}\binom{T}{u-1}B^u \tag{4-41a}$$

$$r=0,\quad g_{R,T}(0,B) = \sum_{u=0}^{R-T}\binom{R-T}{u}\binom{T}{u}(\sqrt{B})^{2u} = \sum_{u=0}^{T}\binom{R-T}{u}\binom{T}{u}B^u \tag{4-41b}$$

$$r=-1,\quad g_{R,T}(-1,B) = \sum_{u=0}^{R-T}\binom{R-T}{u}\binom{T}{u+1}(\sqrt{B})^{2u+1} = \sqrt{B}\sum_{u=0}^{T-1}\binom{R-T}{u}\binom{T}{u+1}B^u \tag{4-41c}$$

Comparing (4-41a), (4-41b) and (4-41c) with (4-36a), (4-36b) and (4-36c), we have

$$Q_1 = \sum_{T=0}^{R-1} A^T g_{R,T}(1,B) \tag{4-42a}$$

$$Q_2 = \sum_{T=0}^{R} A^T g_{R,T}(0,B) \tag{4-42b}$$

$$Q_3 = \sum_{T=1}^{R} A^T g_{R,T}(-1,B) \tag{4-42c}$$

By applying Cauchy integral theorem to (4-40), we have

$$g_{R,T}(r,B) = \frac{1}{2\pi i}\int_c f_{R,T}(z,B)\,\frac{dz}{z^{r+1}} \tag{4-43}$$

where contour c denotes any closed path of integration around the origin in the counter-clockwise sense (see Appendix 2 for proof). Substituting (4-37) into (4-43), we have

$$g_{R,T}(r,B) = \frac{1}{2\pi i}\int_c (1+z\sqrt{B})^{R-T}\left(1+\frac{1}{z}\sqrt{B}\right)^T \frac{dz}{z^{r+1}} \tag{4-44}$$

Substituting (4-44) into (4-42a), (4-42b) and (4-42c), after interchanging the finite summation with integration, we obtain

$$Q_1 = \frac{1}{2\pi i}\int_c \frac{dz}{z^2}(1+z\sqrt{B})^R \sum_{T=0}^{R-1} A^T\left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)^T \tag{4-45a}$$

$$Q_2 = \frac{1}{2\pi i}\int_c \frac{dz}{z}(1+z\sqrt{B})^R \sum_{T=0}^{R} A^T\left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)^T \tag{4-45b}$$

$$Q_3 = \frac{1}{2\pi i}\int_c dz\,(1+z\sqrt{B})^R \sum_{T=1}^{R} A^T\left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)^T \tag{4-45c}$$

Substituting these expressions for Q's into expressions (4-35a), (4-35b) and (4-35c) respectively, we finally obtain:

$$I(2\mu H+4J) = (1-b^{2\mu H+4J}) \times b^{-N(\mu H+J)} \times N \times$$
$$\times\left\{1 + b^{2\mu H+2J}\frac{1}{2\pi i}\int_c \frac{dz}{z^2}(1+z\sqrt{B})^R \sum_{T=0}^{R-1} A^T\left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)^T\right.$$

(4-46a)

$$I(2\mu H) = (1-b^{2\mu H}) \times b^{-N(\mu H+J)} \times 2N \times$$
$$\times\, b^{2\mu H+4J} \frac{1}{2\pi i}\int_C \frac{dz}{z}(1+z\sqrt{B})^R \sum_{T=0}^{R} A^T \left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)^T$$

(4-46b)

$$I(|2\mu H-4J|) = (1-b^{2\mu H-4J}) \times b^{-N(\mu H+J)} \times N \times$$
$$\times \left\{ b^{2\mu H+6J} \frac{1}{2\pi i}\int_C dz\,(1+z\sqrt{B})^R \sum_{T=1}^{R} A^T \left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)^T + b^{2(N-1)\mu H+4J}\right.$$

(4-46c)

Let us summarize what we have done so far. We have calculated the set of three exact expressions (see pages 27-29) for the intensity function as defined by (4-13) for a ring Ising model consisting of an arbitrary number N of spins, where N is of the form N = 4P-1 (P = 1,2,---). For large N, this restriction on N will be of no importance, so that the above set of expressions can be assumed to hold quite generally for large N. However once we assume that N is very large, we can greatly simplify these expressions. This point will become clear in the next step when we try to derive an explicit expression for the intensity function.

Let us resume our calculation:

Since

$$\sum_{T=0}^{R-1}\left[A\left(\frac{1+\sqrt{B}/z}{1+\sqrt{B}\,z}\right)\right]^T = \frac{1-\left[\frac{A(1+\sqrt{B}/z)}{1+\sqrt{B}\,z}\right]^R}{1-\frac{A(1+\sqrt{B}/z)}{1+\sqrt{B}\,z}}$$

equation (4-45a) can then be written as

$$Q_1 = \frac{1}{2\pi i}\int_c \frac{dz}{z^2}(1+z\sqrt{B})^R \left\{\frac{1-\left[\frac{A(1+\sqrt{B}/z)}{1+\sqrt{B}\,z}\right]^R}{1-\frac{A(1+\sqrt{B}/z)}{1+\sqrt{B}\,z}}\right\}$$

$$= \frac{1}{2\pi i}\int_c \frac{1}{\sqrt{B}}\,\frac{dz}{z}\,\frac{(1+z\sqrt{B})^R - A^R(1+\sqrt{B}/z)^R}{(z-z_2)(z-z_3)}$$

(4-47)

where c denotes any contour around origin, and z_2 and z_3 are the roots of the equation

$$z^2 + \frac{1-A}{\sqrt{B}}\,z - A = 0$$

namely

$$z_2 = \frac{1}{2\sqrt{B}}\left[(-1+A)+\alpha\right] \qquad (4\text{-}48a)$$

$$z_3 = \frac{1}{2\sqrt{B}}\left[(-1+A)-\alpha\right] \qquad (4\text{-}48b)$$

where $\alpha = \sqrt{(1-A)^2 + 4AB}$ (4-49)

The exact evaluation of (4-47) leads to a combinatorial sum (see Appendix 3) which is no easier to use than the equation (4-33a) which we have started from. It is for this reason that we shall assume that N is large, and we shall examine the asymptotic behaviour of the function (4-47) with respect to R (=N-3). Expression (4-47) can be rewritten as follows:

$$Q_1 = \frac{1}{2\pi i}\,\frac{1}{\sqrt{B}}\left\{\int_c \frac{(1+z\sqrt{B})^R}{(z-z_2)(z-z_3)}\,\frac{dz}{z} - \int_c \frac{A^R(1+\sqrt{B}/z)^R}{(z-z_2)(z-z_3)}\,\frac{dz}{z}\right\} \tag{4-50}$$

Note that the integrand in (4-47) has one and only one singularity, namely at the origin; the two integrands in (4-50) however, have each three singularities, namely at $z = 0$, $z = z_2$ and $z = z_3$. This does not lead to any difficulty so long as the two contours in the two integrations in (4-50) are the same; in that case, the contributions due to the singularity $z = z_2$ and to the singularity $z = z_3$ in the two integrations cancel. We can now choose a contour c' such that for R tending to infinity,

$$\left|\frac{A(1-\sqrt{B}/z)}{1-\sqrt{B}z}\right|^R \ll 1 \tag{4-51}$$

Hence the contribution from the second term in (4-50) is arbitrarily small in comparison with that from the first term and is consequently dropped (see appendix 4 for the choice of contour c'). Equation (4-50) can then be written, to a good approximation, as

$$\hat{Q}_1 = \frac{1}{2\pi\sqrt{B}\,i}\int_{c'} \frac{(1+z\sqrt{B})^R}{(z-z_2)(z-z_3)}\,\frac{dz}{z} \tag{4-52}$$

where contour c' and R satisfy the condition (4-51) and the symbol "$\wedge$" denotes the asymptotic value.

Now Cauchy Residue Theorem states that

$$\frac{1}{2\pi i}\int_{c} f(z)\,dz = \sum_i \operatorname{Res}\{f(z), z_i\} \tag{4-53}$$

and

$$\operatorname{Res}\{f(z), z_i\} = \lim_{z\to z_i} (z-z_i)\,f(z) \tag{4-54}$$

for simple poles. By using (4-53) and (4-54), we obtain

$$\hat{Q}_1 = \frac{1}{\sqrt{B}}\left[\frac{1}{z_2 z_3} + \frac{(1+\sqrt{B}\,z_2)^{R+1}}{z_2(z_2-z_3)} + \frac{(1+\sqrt{B}\,z_3)^{R+1}}{z_3(z_3-z_2)}\right] \tag{4-55a}$$

Similarly we obtain

$$\hat{Q}_2 = \frac{1}{\sqrt{B}}\left[\frac{(1+\sqrt{B}\,z_2)^{R+1}}{z_2-z_3} + \frac{(1+\sqrt{B}\,z_3)^{R+1}}{z_3-z_2}\right] \tag{4-55b}$$

$$\hat{Q}_3 = \frac{1}{\sqrt{B}}\left[\frac{z_2(1+\sqrt{B}\,z_2)^{R+1}}{z_2-z_3} + \frac{z_3(1+\sqrt{B}\,z_3)^{R+1}}{z_3-z_2}\right] \tag{4-55c}$$

By using (4-48a) and (4-48b), we have the following set of relationships:

$$\begin{cases} z_2 - z_3 &= \alpha/\sqrt{B} \\ z_2 z_3 &= -A \\ 1+\sqrt{B}\,z_2 &= \frac{1}{2}(1+A+\alpha) \\ 1+\sqrt{B}\,z_3 &= \frac{1}{2}(1+A-\alpha) \end{cases}$$

(4-56)

Combining (4-35a), (4-35b), (4-35c), (4-55a), (4-55b) (4-55c) and (4-56), we arrive at the following set of expressions:

$$\hat{I}(2\mu H+4J) = (1-b^{2\mu H+4J}) \times b^{-N(\mu H+J)} \times N \times$$
$$\times \left\{ -\frac{1}{\alpha}\left(\frac{1}{2}\right)^{R+2}\left[(-1+A-\alpha)(1+A+\alpha)^{R+1} - (-1+A+\alpha)(1+A-\alpha)^{R+1}\right]\right\}$$

(4-57a)

$$\hat{I}(2\mu H) = (1-b^{2\mu H}) \times b^{-N(\mu H+J)+(2\mu H+4J)} \times 2N \times$$
$$\times \left\{ \frac{1}{\alpha}\left(\frac{1}{2}\right)^{R+1}\left[(1+A+\alpha)^{R+1} - (1+A-\alpha)^{R+1}\right]\right\}$$

(4-57b)

$$\hat{I}(|2\mu H-4J|) = (1-b^{2\mu H-4J}) \times b^{-N(\mu H+J)+(2\mu H+4J)} \times N \times$$
$$\times \left\{ \frac{1}{\alpha}\left(\frac{1}{2}\right)^{R+2}\left[(-1+A+\alpha)(1+A+\alpha)^{R+1} - (-1+A-\alpha)(1+A-\alpha)^{R+1}\right]\right\}$$

(4-57c)

If we assume that $B \geqslant A$, i.e., $2\mu H \geqslant 4J$, then we have the following relations (see appendix 5 for proof):

$$\begin{cases} 1 > 1 + A - \alpha \geqslant 0 \\ 4 \geqslant 1 + A + \alpha \geqslant 2 \\ 2 \geqslant |-1 + A - \alpha| > 1 \\ 2 \geqslant -1 + A + \alpha \geqslant 0 \end{cases} \tag{4-58}$$

When R approaches infinity as required by the condition (4-51),

$$(1+A+\alpha)^{R+1} \gg (1+A-\alpha)^{R+1} \tag{4-59}$$

hence we can omit the term consisting of $(1+A-\alpha)^{R+1}$ in each of the expressions (4-57a), (4-57b) and (4-57c). Moreover, since we are interested only in the relative magnitude of these expressions, a common factor Θ can be extracted, where

$$\Theta = b^{-N(\mu H+J)} \times N \times \left(\frac{1}{\alpha}\right)\left(\frac{1}{2}\right)^{N-1}(1+A+\alpha)^{N-2} \tag{4-60}$$

Let $\hat{I} = \hat{I}^{*}\,\Theta$, remembering that $R = N-3$, we finally arrive at the following set of expressions:

$$\hat{I}^*(2\mu H+4J) = (1-b^{2\mu H+4J})(\alpha - A + 1) \qquad (4\text{-}61a)$$

$$\hat{I}^*(2\mu H) \quad = (1-b^{2\mu H})\, 4b^{2\mu H+4J} \qquad (4\text{-}61b)$$

$$\hat{I}^*(|2\mu H-4J|) = (1-b^{2\mu H-4J})\, b^{2\mu H+4J}(\alpha + A - 1) \qquad (4\text{-}61c)$$

where
$$b = e^{-\frac{1}{KT}}$$

$$A = e^{-\frac{2\mu H}{KT}}$$

$$\alpha = \left[(1-e^{-\frac{2\mu H}{KT}})^2 + 4\, e^{-\frac{2\mu H+4J}{KT}}\right]^{\frac{1}{2}}$$

It is clear from the expressions (4-61a), (4-61b) and (4-61c) that if $2\mu H \geqslant 4J$, then the intensity function is independent of N, the number of spins present in the system, provided that the conditions imposed on N are satisfied.

If $2\mu H < 4J$, we have to use the expressions (4-57a), (4-57b) and (4-57c) which hold for any values of μH and J.

Let us summarize what we have done. We have obtained the set of expressions (4-61a), (4-61b) and (4-61c) for the intensity function as defined by (4-13) for a one-dimensional Ising model consisting of an arbitrarily large number N of spins arranged into a ring, where N is of the form N = 4P-1 (P = 1,2,---). The assumption that N is large enters our calculation for the expressions (4-61a), (4-61b) and (4-61c) at the following points:

1) that N must be sufficiently large such that (see (4-51))

$$\left| \frac{A(1-\sqrt{B}/z)}{1-\sqrt{B}z.} \right|^{N-3} \ll 1$$

where z satisfies the condition imposed on the contour c' (see appendix 4),

2) that N must be sufficiently large such that (see 4-59)

$$(1+A+\alpha)^{N-2} \gg (1+A-\alpha)^{N-2}$$

However for large N, the restriction that N takes the particular form N = 4P-1 will be of no importance, so that the expressions (4-61a)(4-61b) and (4-61c) can be regarded as quite generally valid. Moreover, when N is large, these expressions can be used for a one-dimensional open-chain Ising model as well.

SECTION V

Intensity Function in the First Approximation for the Ring Model with Arbitrary N

The first approximation calculation (2-11) amounts to putting J = 0 (i.e. B = 1, see (4-34)) in our previous calculation for the intensity function of the ring model. By so doing, the set of expressions (4-32a), (4-32b), (4-32c); (4-33a), (4-33b), (4-33c); (4-57a), (4-57b), (4-57c) and (4-61a), (4-61b), (4-61c), as well as the expressions (5-a), (5-b) and (5-c) in Appendix 3, are all reduced to the same set of expressions which is as follows:

$$\hat{I}^*(2\mu H+4J)_1 = (1-b^{2\mu H})\,2 \tag{5-1a}$$

$$\hat{I}^*(2\mu H) = (1-b^{2\mu H})\,4\,b^{2\mu H} \tag{5-1b}$$

$$\hat{I}^*(|2\mu H-4J|) = (1-b^{2\mu H})\,2\,b^{4\mu H} \tag{5-1c}$$

and

$$\hat{I}(|\Delta E_{\Delta S}|)_1 = \hat{I}^*(|\Delta E_{\Delta S}|)_1\,\Theta_1 \tag{5-2}$$

where $(H)_1$ is given by (4-60) with J = 0, i.e.,

$$(H)_1 = \frac{1}{2}(1+b^{2\mu H})^{N-3} \times b^{-N\mu H} \times N \tag{5-3}$$

and the subscript "1" at the lower right corner indicates that the various expressions are in the first approximation.

The frequency function in the first approximation can be easily obtained by substituting (5-1a), (5-1b) and (5-1c) into (1-1):

$$\hat{f}(2\mu H+4J)_1 = \frac{1}{(1+b^{2\mu H})^2} \tag{5-4a}$$

$$\hat{f}(2\mu H)_1 = \frac{2b^{2\mu H}}{(1+b^{2\mu H})^2} \tag{5-4b}$$

$$\hat{f}(|2\mu H-4J|)_1 = \frac{b^{4\mu H}}{(1+b^{2\mu H})^2} \tag{5-4c}$$

SECTION VI

Comparison of the Asymptotically Exact Expressions (4-61a),(4-61b),(4-61c) with First Approximation Expressions (5-1a),(5-1b),(5-1c) at Very Low Temperature and Conclusions

$$\hat{I}^*(2\mu H+4J)=(1-b^{2\mu H+4J})(\alpha-A+1) \qquad (4\text{-}61a)$$

$$\hat{I}^*(2\mu H) \quad =(1-b^{2\mu H})\ 4b^{2\mu H+4J} \qquad (4\text{-}61b)$$

$$\hat{I}^*(|2\mu H-4J|)=(1-b^{2\mu H-4J})b^{2\mu H+4J}(\alpha+A-1) \qquad (4\text{-}61c)$$

$$\hat{I}^*(2\mu H+4J)_1=(1-b^{2\mu H})\,2 \qquad (5\text{-}1a)$$

$$\hat{I}^*(2\mu H)_1 \quad =(1-b^{2\mu H})\,4\ b^{2\mu H} \qquad (5\text{-}1b)$$

$$\hat{I}^*(|2\mu H-4J|)_1=(1-b^{2\mu H})\,2\ b^{4\mu H} \qquad (5\text{-}1c)$$

Now let us resume our original question about the goodness of McMillan and Opechowski's method of approximation at very low temperature. Since $(\alpha - A + 1)$ and $(\alpha + A - 1)$ are finite at all temperatures (4-58), it is clear that at very low temperature the factor $b^{2\mu H+4J}$ in (4-61b) and (4-61c), and $b^{2\mu H}$ in (5-1b) and (5-1c), tend to zero; hence the frequency function calculated from both sets of expressions tends to the same limit, namely

$$\begin{aligned}
\hat{f}_o(2\mu H+4J) &= 1 \\
\hat{f}_o(2\mu H) &= 0 \\
\hat{f}_o(|2\mu H-4J|) &= 0
\end{aligned} \tag{6-1}$$

where we have used a subscript "o" to denote the temperature $T = 0°K$.

A graph (6-2) is plotted by putting $\mu H = 20J$ for the frequency function $f(\mathcal{E})$ calculated from both the expressions (4-61a), (4-61b), (4-61c), and the expressions (5-1a), (5-1b), (5-1c) at various temperatures. It is seen from the graph that the method of approximation is quite good in general and extraordinarily good at temperature below, say, $T = \frac{4J}{k}$.

Note that the expressions (4-33a), (4-33b) and (4-33c) can be written as follows:

$$I = I^{\#}\,\Phi \qquad (6\text{-}3)$$

where

$$\Phi = b^{-N(\mu H + J)} \times N \qquad (6\text{-}4)$$

and

$$I^{\#}(2\mu H+4J) = (1-b^{2\mu H+4J}) \times \left\{1 + b^{-(2\mu H+4J)} + \sum_{t=2}^{N-2}\sum_{s=2}^{t} b^{2t\mu H+4sJ}\binom{N-t-1}{s-1}\binom{t-2}{s-2}\right.$$

$$(6\text{-}5a)$$

$$I^{\#}(2\mu H) = (1-b^{2\mu H}) \times b^{-2\mu H} \times 2 \times \left\{\sum_{t=2}^{N-1}\sum_{s=1}^{t-1} b^{2t\mu H+4sJ}\binom{N-t-1}{s-1}\binom{t-2}{s-1}\right.$$

$$(6\text{-}5b)$$

$$I^{\#}(|2\mu H-4J|) = (1-b^{2\mu H-4J}) \times b^{-2\mu H+4J} \times \left\{\sum_{t=3}^{N-1}\sum_{s=1}^{t-2} b^{2t\mu H+4sJ}\binom{N-t-1}{s-1}\binom{t-2}{s} + b^{2N\mu H}\right\}$$

$$(6\text{-}5c)$$

When $T \to 0$

then

$$\begin{cases} b^{2\mu H} \to 0 \\ b^{4J} \to 0 \end{cases} \tag{6-6}$$

and

$$\begin{cases} b^{-2\mu H} \to \infty \\ b^{-4J} \to \infty \end{cases} \tag{6-7}$$

hence

$$\begin{cases} I_o^{\#}(2\mu H + 4J) = 1 \\ I_o^{\#}(2\mu H) = 0 \\ I_o^{\#}(|2\mu H - 4J|) = 0 \end{cases} \tag{6-8}$$

It is easy to see that the frequency function tends to the same limit as (6-1), namely:

$$\begin{cases} f_o(2\mu H + 4J) = 1 \\ f_o(2\mu H) = 0 \\ f_o(|2\mu H - 4J|) = 0 \end{cases} \tag{6-9}$$

This shows that the frequency function calculated from the set of expressions (4-33a), (4-33b) and (4-33c) tends to the same limit (6-9) in the following two processes:

(1) We first let N tend to infinity and then let T tend to zero,

(2) We let T tend to zero and impose no new condition on N.

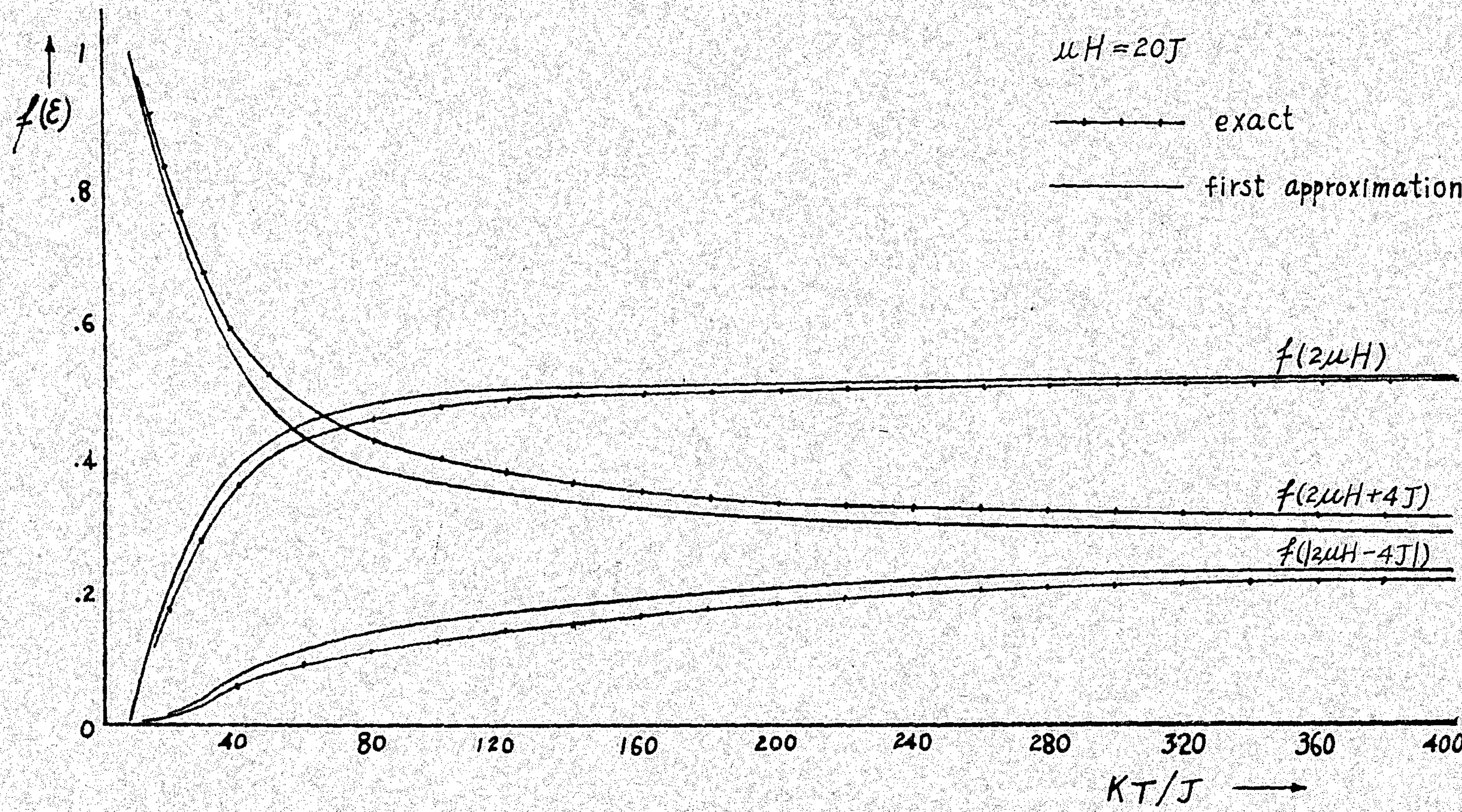

Fig. (6-2) Comparison of the asymptotic values of frequency function in the exact (4-61a,b,c) and first approximation (5-1a,b,c).

APPENDIX I

General Formula of the Number of Component Lines Present in the Absorption Band for an Ising Model with an Arbitrary Form of Lattice (see p.27)

Since the Zeeman transition energy is always the same ($2\mu H$), the number of transition lines in the absorption band is given by the number of different values the transition exchange energy can take

Let K = number of closest neighbouring spins (constant)

V_1 = number of positive spins among the K spins

V_2 = number of negative spins among the K spins

then

$$V_1 + V_2 = K \qquad [V_1, V_2] \geqslant 0 \tag{1}$$

The number of different values that the transition exchange energy can take is equal to the number of different values that V_1 and V_2 can take such that equation (1) is satisfied. It is obvious that this number is K + 1. Hence the number of transition lines in the absorption band is K + 1, provided that we neglect the effect of the boundaries.

APPENDIX 2

(see p.47)

To prove

$$g_{R,T}(r,B) = \frac{1}{2\pi i}\int_c f_{R,T}(z,B)\,\frac{dz}{z^{r+1}} \tag{4-43}$$

where

$$f_{R,T}(z,B) \equiv \sum_{r=-T}^{R-T} g_{R,T}(r,B)\, z^r \tag{4-40}$$

Proof. Substituting (4-40) into R.H.S. of (4-43) we have,

$$\text{R.H.S.} = \frac{1}{2\pi i}\int_c \sum_{r'=-T}^{R-T} g_{R,T}(r',B)\,\frac{dz}{z^{r-r'+1}}$$

$$= \frac{1}{2\pi i}\sum_{r'=-T}^{R-T} g_{R,T}(r',B)\int_c \frac{dz}{z^{r-r'+1}}$$

$$\because \int_c \frac{dz}{z^{r-r'+1}} = \begin{cases} 0 & r' \neq r \\ 2\pi i & r' = r \end{cases}$$

$$\therefore \text{R.H.S.} = g_{R,T}(r,B) \equiv \text{L.H.S.}$$

Q.E.D.

APPENDIX 3

The Exact Evaluation of Q_1, Q_2 and Q_3 (see p.48)

Since

$$Q_1 = \frac{1}{2\pi i}\int_C \frac{1}{\sqrt{B}}\,\frac{dz}{z}\,\frac{(1+z\sqrt{B})^R - A^R(1+\sqrt{B}/z)^R}{(z-z_2)(z-z_3)} \qquad (4\text{-}47)$$

by taking an arbitrary contour around the only singularity of the integrand, namely the origin, and applying Cauchy's Residue Theorem for both simple pole and pole of higher order, we obtain

$$Q_1 = \frac{1}{\sqrt{B}}\left(-\frac{1}{A}\right) - \frac{A^R}{\sqrt{B}}\lim_{z\to 0}\frac{d^R}{dz^R}\,\frac{(1+z\sqrt{B})(z+\sqrt{B})^R}{(z-z_2)(z-z_3)}\,\frac{1}{R!} \qquad (1)$$

In order to calculate the higher order derivative in the above expression, we proceed as follows:

$$\because\ (1+\sqrt{B}\,z)(z+\sqrt{B})^R(z-z_2)^{-1}(z-z_3)^{-1}$$

$$= \frac{1}{z_2 z_3}\,(1+\sqrt{B}\,z)\sum_{r=0}^{R}\binom{R}{r} z^{R-r}(\sqrt{B})^r \sum_{s=0}\left(\frac{z}{z_2}\right)^s \sum_{t=0}\left(\frac{z}{z_3}\right)^t$$

and the coefficient of z^R in the above expression is

$$\frac{1}{z_2 z_3}\sum_{r,s,t}\binom{R}{r}(\sqrt{B})^r\left(\frac{1}{z_2}\right)^s\left(\frac{1}{z_3}\right)^t\ \delta_{R-r+s+t,\,R}$$

$$+\frac{\sqrt{B}}{z_2 z_3}\sum_{r,s,t}\binom{R}{r}(\sqrt{B})^r\left(\frac{1}{z_2}\right)^s\left(\frac{1}{z_3}\right)^t\ \delta_{R-r+s+t,\,R-1}$$

(2)

where $\sum_{r,s,t}$ means summation over all possible combinations of r, s and t.

Since

$$\frac{d^W}{dz^W}\,z^R=\begin{cases}\dfrac{R!}{(R-W)!}\,z^{R-W} & R\geqslant W\\[2ex] 0 & R<W\end{cases} \qquad (3)$$

substituting (2)(3) into (1), we thus have

$$Q_1=\frac{1}{\sqrt{B}}\left(-\frac{1}{A}\right)-\frac{A^R}{\sqrt{B}}\left[\frac{1}{z_2 z_3}\sum_{r,s,t}\binom{R}{r}(\sqrt{B})^r z_2^{-s} z_3^{-t}\right]\left[\delta_{r,\,s+t}+\sqrt{B}\,\delta_{r-1,\,s+t}\right] \qquad (4)$$

Combining (4-35a), (4-48a), (4-48b), (4-56) and (4) we have

$$I(2\mu H+4J)=(1-b^{2\mu H+4J})\times b^{-N(\mu H+J)}\times N\times$$

$$\times\left\{A^R\sum_{r,s,t}\binom{R}{r}(2B)^r(-1+A+\alpha)^{-s}(-1+A-\alpha)^{-t}\left(\delta_{r,s+t}+\frac{1}{2}\delta_{r-1,s+t}\right)\right\} \quad \text{(5-a)}$$

Similarly we obtain

$$I(2\mu H)=(1-b^{2\mu H})\times b^{-N(\mu H+J)+(2\mu H+4J)}\times 2N\times$$

$$\times\left\{A^R\sum_{r,s,t}\binom{R+1}{r}(2B)^{r-1}(-1+A+\alpha)^{-s}(-1+A-\alpha)^{-t}\delta_{s+t,r-1}\right\}$$

$$\text{(5-b)}$$

$$I(|2\mu H-4J|)=(1-b^{2\mu H-4J})\times b^{-N(\mu H+J)+(2\mu H+4J)}\times N\times$$

$$\times\left\{\frac{1}{2}A^R\sum_{r,s,t}\binom{R+1}{r}(2B)^{r-1}(-1+A+\alpha)^{-s}(-1+A-\alpha)^{-t}\delta_{s+t,r-2}\right.$$

$$\left.+b^{2(N-2)\mu H}\right\} \quad \text{(5-c)}$$

APPENDIX 4

To Prove the Existence of the Contour c' (see p.52)

The contour c' in (4-52) must satisfy the condition

$$\left| A(1+\sqrt{B}/z) \right| < \left| 1+\sqrt{B}z \right| \tag{1}$$

where

$$1 \geq A \equiv e^{-\frac{2\mu H}{KT}} \geq 0 \tag{2}$$

$$1 \geq B \equiv e^{-\frac{4I}{KT}} \geq 0 \tag{3}$$

i.e. A and B are positive quantities independent of z but dependent on the absolute temperature T.

Let

$$z \equiv x + iy \tag{4}$$

and

$$\rho \equiv x^2 + y^2 \tag{5}$$

then (1) can be written as follows:

$$A\left|1+\frac{\sqrt{B}}{\rho}(x-iy)\right| < \left|1+\sqrt{B}(x+iy)\right|$$

$$\therefore A^2\left\{\left(1+\frac{\sqrt{B}}{\rho}x\right)^2+\left(\frac{\sqrt{B}}{\rho}y\right)^2\right\} < \left\{\left(1+\sqrt{B}x\right)^2+\left(\sqrt{B}y\right)^2\right\}$$

$$\therefore A^2\left\{1+\frac{2\sqrt{B}}{\rho}x+\frac{B}{\rho^2}(x^2+y^2)\right\} < \left\{1+2\sqrt{B}x+B(x^2+y^2)\right\}$$

$$\therefore (x^2+y^2-A^2)B+2\sqrt{B}\left(1-\frac{A^2}{\rho}\right)x+(1-A^2) > 0$$

(6)

This is the condition that the contour c' must satisfy. Let us choose x and y such that

$$\rho \equiv x^2+y^2 > 1 \qquad (7)$$

If T = 0, we have, by (2) and (3), A = 0 and B = 0, hence condition (6) is satisfied.

If T > 0, by (2) and (3), A > 0 and B > 0, hence by

(7) we always have

$$(x^2+y^2-A^2)B > 0 \tag{8}$$

$$1 - A^2 \geqslant 0 \tag{9}$$

However, we must distinguish the case X > 0 and the case X < 0. If X ⩾ 0, then

$$\sqrt{B}\ \left(1-\frac{A^2}{\rho}\right)X \geqslant 0 \tag{10}$$

hence, by (8), (9) and (10), condition (6) is satisfied.

If X< 0, we can choose y sufficiently large such that

$$(x^2+y^2-A^2)\frac{\sqrt{B}}{2} > |x|$$

hence condition (6) is again satisfied.

Therefore the contour c' can always be found and the approximation (4-52) always valid.

APPENDIX 5

To Prove the Inequalities (4-58) (see p.54)

Given

$$\alpha \equiv \sqrt{(1-A)^2+4AB} \tag{1}$$

$$1 \geqslant B \geqslant A \geqslant 0 \tag{2}$$

first we need to prove the inequality

$$\alpha > A \tag{3}$$

Proof: the inequality (3) can be rewritten as

$$(1-A)^2 + 4AB > A^2$$

or $$1 - 2A(1-2B) > 0 \tag{4}$$

If $\quad B > \frac{1}{2} \quad$ then $\quad 1-2B < 0 \quad \therefore$ (4) holds

If $\quad B = \frac{1}{2} \quad \because \quad 1-2B = 0 \quad \therefore$ (4) holds

If $\quad 0 < B < \frac{1}{2} \quad \because \quad A \leqslant B < \frac{1}{2}$

hence

$$1-2B < 1$$

$$2A < 1 \qquad \therefore \text{ (4) holds}$$

If $\quad B = 0 \quad$ then $\quad A = 0 \quad \therefore$ (4) holds

hence (3) is always true. Q.E.D.

We always need to prove the inequality

$$\alpha \leqslant 2 \tag{5}$$

Proof:

$$\because \quad 0 \leqslant B \leqslant 1$$

$$\therefore \quad \alpha \equiv \sqrt{(1-A)^2 + 4AB}$$

$$\leqslant \sqrt{(1-A)^2 + 4A} = 1 + A \tag{6}$$

but $\quad 0 \leqslant A \leqslant 1$

$\therefore \alpha \leqslant 2 \qquad$ Q.E.D.

Since

$$\alpha \equiv \sqrt{(1-A)^2+4AB} \begin{cases} > 1-A & \text{if } A,B \neq 0 \quad \text{i.e. } T \neq \infty \qquad (7) \\ = 1 & \text{if } A,B = 0 \quad \text{i.e. } T = \infty \qquad (8) \end{cases}$$

We now have the following set of relationships:

by (2) , $2 \geqslant 1+A \geqslant 1$ (9)

(6) , $1+A \geqslant \alpha$ (10)

(3) $\alpha > A$ (11)

(7) $\alpha > 1-A$ if A,B $\neq$ 0 (12)

(8) $\alpha = 1$ if A,B = 0 (13)

It is then easy to see that

by (2), (5) and (11) $4 \geqslant 1+A+\alpha \geqslant 2$

(11) and (10) $1 > 1+A-\alpha \geqslant 0$

(2), (5) and (12) $2 \geqslant -1+A+\alpha \geqslant 0$

(10), (11) $-1 > -1+A-\alpha \geqslant -1+A-(1+A) = -2$ (14)

by (14) $2 \geqslant |-1+A-\alpha| > 1$

Hence the set of inequalities (4-58) is proved.

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