ANTIFERROMAGNETISM IN THE SELFCONSISTENT SPIN WAVE APPROXIMATION AND THE EVALUATION OF LATTICE GREEN FUNCTIONS.

A thesis
submitted in partial fulfilment of
the requirements for the degree of
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David James Austen

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ABSTRACT

Part I:

The macroscopic properties of an antiferromagnet for temperatures below the Neel temperature are derived from the spin wave Heisenberg Hamiltonian by a variational method. Using the Dyson-Maleev and Holstein- Primakoff transformations, three solutions valid for arbitrary spin, magnetic field and uniaxial anisotropy are discussed in terms of their theoretical prediction for susceptibility, sublattice magnetization and internal energy. Specifically, for zero anisotropy, we obtain a new result for the parallel susceptibility and derive many properties in the limit of infinite (classical) spin. Also, we compare our three solutions for a finite value of anisotropy.

Part II:

A method is developed whereby lattice Green functions and Watson-like integrals can be readily computed by splitting them into a sum of a power series and an asymptotic series. The method is illustrated by the evaluation of an antiferromagnetic integral and a basic thermodynamic Watson's integral and followed by a treatment of the generalized problem on all the cubic lattices.

CONTENTS

PART II

LATTICE GREEN FUNCTIONS

PART I

ANTIFERROMAGNETISM

CHAPTER 1

INTRODUCTION

The aim of this thesis is to describe a method for obtaining, at low temperatures, the macroscopic properties of an antiferromagnet by using a non-Hermitian approximation to the spin wave Heisenberg Hamiltonian. By invoking the variational theorem, we are able to fit a very general trial Hamiltonian to the exact Hamiltonian and then determine many macroscopic properties (especially susceptibility) from the appropriate derivatives of the free energy. We are interested in a three dimensional Heisenberg Hamiltonian with uniaxial anisotropy and external magnetic field. Since antiferromagnetism is such an active area for research, it is necessary to indicate where this work stands in relation to others; in the following we will consider some of the relevant papers:

In 1952, Kubo, Anderson and Ziman, first considered very low temperature properties using non-interacting spin wave theory. Kubo (1952) and Ziman (1952) looked at some of the thermodynamic properties whereas Anderson (1952) was concerned mostly with zero point deviations. Kanamori and Yosida (1955) and Kanamori and Tachiki (1962) calculated and compared some of the low temperature properties such as magnetization and susceptibility. Oguchi

(1960) derived low temperature expansion for variou properties to order $1/S^2$. Later, M.Bloch (1963 and thesis), who calculated magnetization curves, showed how to use a self-consistent method to obtain formulae which give reasonable results up to 80 or 90% of the Neel temperature T_N. Low (1963), Nagai (1969), Nagai and Tanaka (1969, 1970) and Kanamori and Itoh (1968, 1972) have compared theories to experimental results. These authors have all used the Holstein-Primakoff transformations and thus Hermitian approximations.

In 1956, Dyson (1956) showed how to use a non-Hermitian Hamiltonian to calculate some of the properties of a ferromagnet and obtained the famous T^4 correction term. For an antiferromagnet, two separate non-Hermitian Hamiltonians have been developed (Maleev 1958, Dembinski 1964) and used as well as the one by Holstein-Primakoff. Harris, Szaniecki, Armour and Herbert have made extensive use of the non-Hermitian representations in an attempt to calculate exact low temperature expansions. Harris (1968, 1969) considered $\frac{1}{S}$ and $\frac{1}{Z}$ expansions, Szaniecki (1965, 1968a, 1968b, 1971) has discussed the kinematical interaction and zero point deviations, and Armour (1970, 1971) and Herbert (1970) have also looked at kinematical effects. Recently, Collins and Tondon (1972) have used a non-Hermitian Hamiltonian to calculate the effect of second neighbour interactions.

Liu (1966) attempted to combine a self-consistent

calculation such as Bloch's with the non-unitary transformations introduced by Dyson etc. Unfortunately, he erroneously obtained a Hermitian Hamiltonian (in disagreement, for example, with Szaniecki 1968a) and thus encountered no serious problems in a Green's function solution. As well, he did not obtain agreement with Oguchi (1960) in respect to the low temperature susceptibilities. Flax and Raich (1970) used Liu's method to investigate the anisotropic Heisenberg Hamiltonian.

Our idea is to determine to what extent a non-Hermitian "Hamiltonian" is capable of explaining the properties, particularly the susceptibilities, of an antiferromagnet. For this purpose, three alternate approximations are made, one involving the Holstein-Primakoff transformation (where we consider more terms than previous authors) and two using Dyson-Maleev transformations (where all terms are taken). The general plan then is to determine the best solution to the Hamiltonian. We will show that each of these representations is equivalent to evaluating the antiferromagnetic Hamiltonian in three different approximations. The aim is to use a variational calculation with a very general approximate Hamiltonian and, by minimizing the free energy, determine the best solution.

Unfortunately it soon becomes apparent that non-Hermitian Hamiltonians do not yield approximations which are upper bounds to the free energy. This mathematical criterion usually used to compare approximations becomes

invalid. It is shown however that the procedure used in variational minimization has a useful generalization to the non-Hermitian cases and by such a method we can determine the best parameters for the approximate Hamiltonian in each case. Having obtained the best parameters in three cases, the properties are compared from a practical standpoint.

The above procedure is equivalent to choosing some approximate Hamiltonian and using first order perturbation theory. Second order theory is not carried out for two reasons: first, the second order solution is algebraically complex and second, the results would be expressed in terms of six dimensional integrals which at the present time are difficult to compute either numerically or analytically.

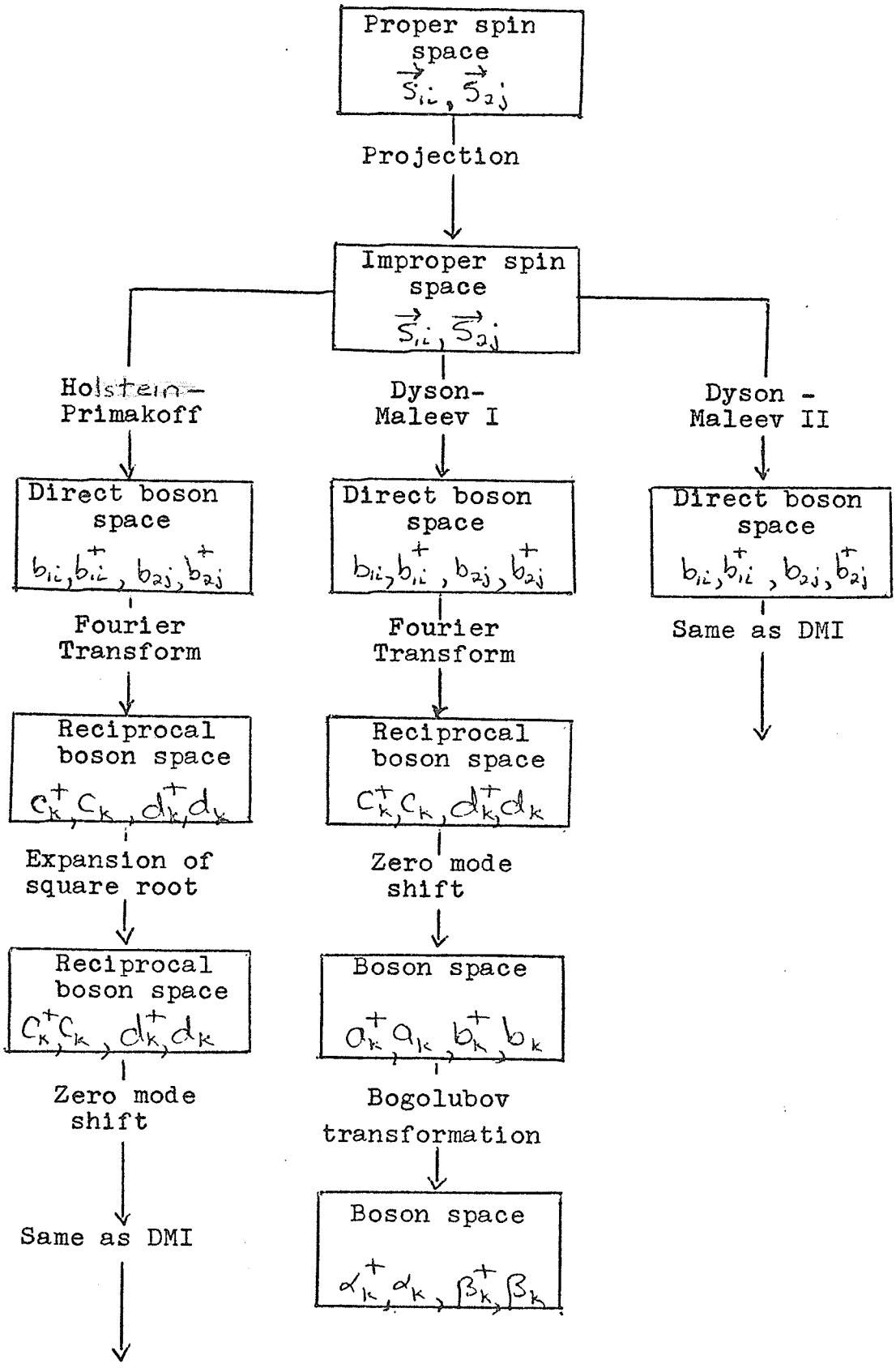
The Heisenberg model assumes that the process responsible for magnetic ordering is the pairwise interaction of spins which are localized at lattice sites. The energy of interaction is proportional to the inner product of the spin vectors, the constant of proportionality being the exchange constant (Löwdin 1962). In most cases the exchange constant between two spins decreases with increasing separation so it is natural to assume that nearest neighbour spins contribute most of the energy. In an antiferromagnet the exchange constant is negative and this tends to make nearest spins antiparallel.

Another form of energy in an antiferromagnet arises from anisotropy (Keffer 1965) there being two forms in particular, dipolar-dipolar and uniaxial. Although these contribute substantially less energy to the system than the exchange energy, as we will see, the antiparallel arrangement cannot be achieved without some stabilizing force. In a ferromagnet, one can use a small field to stabilize the system; however, in the antiferromagnetic case one sees that the spin arrangement makes this impossible. Mathematically, it is convenient to take uniaxial anisotropy into account whereas other forms become very difficult to handle. For this reason we consider only uniaxial forces in a manner similar to Nagai and Tanaka (1970) who increase the role of uniaxial anisotropy to take into account the dipolar-dipolar. This serves to stabilize the system and hopefully will predict those macroscopic properties which do not depend strongly on anisotropy.

Due to the algebraic complexity of this work, which includes many transformations and representations, figure 1 is included to help in guiding the reader through the mathematical details of the later chapters. In this figure the operators used in each representation are summarized and the type of space is specified. The aim is to transform the Hamiltonian from proper spin space to an infinite boson space in which the approximate Hamiltonian is di-

FIGURE 1

Schematic representation of Hamiltonians (in boxes) and transformations used in text.


Proper spin space
$\vec{S}_{1i}, \vec{S}_{2j}$
Projection
Improper spin space
$\vec{S}_{1i}, \vec{S}_{2j}$
Holstein-Primakoff
Dyson-Maleev I
Dyson - Maleev II
Direct boson space
$b_{1i}, b_{1i}^+, b_{2j}, b_{2j}^+$
Direct boson space
$b_{1i}, b_{1i}^+, b_{2j}, b_{2j}^+$
Direct boson space
$b_{1i}, b_{1i}^+, b_{2j}, b_{2j}^+$
Fourier Transform
Fourier Transform
Same as DMI
Reciprocal boson space
$c_k^+ c_k, d_k^+ d_k$
Reciprocal boson space
$c_k^+ c_k, d_k^+ d_k$
Expansion of square root
Zero mode shift
Reciprocal boson space
$c_k^+ c_k, d_k^+ d_k$
Boson space
$a_k^+ a_k, b_k^+ b_k$
Zero mode shift
Bogolubov transformation
Same as DMI
Boson space
$\alpha_k^+ \alpha_k, \beta_k^+ \beta_k$

agonal. Our procedure relating spin and boson spaces is novel in that the proper spin space is projected onto an infinite spin space immediately and then this is mapped in three separate ways onto boson spaces. Most authors map the proper spin space onto a subspace of the boson space and then extend this to the full boson space. The former route is taken as it serves to demonstrate the mathematical equivalence of the various representations of the Hamiltonian - this fact is necessary for a comparitive study. This is done at the risk of confusing the reader who is familiar with the usual treatment of Dyson (1956) etc. However, these methods differ only in philosophy and not in outcome.

In chapter 2, we set up three representations of the Hamiltonian in reciprocal boson space. In chapter 3, an extension of the usual variational theorem is discussed, allowing us to obtain three solutions. In order to obtain these solutions, it is necessary to derive non-unitary generalizations of the Bogolubov and zero-mode shift transformations. By the end of chapter 3, a method is developed whereby three solutions valid for arbitrary uniaxial anisotropy and magnetic field are obtained. In chapter 4, a comparison of these solutions is undertaken. First, we find many thermodynamic properties in the limit of vanishing field and anisotropy so that we can compare with other authors, namely Liu. We find that the parallel susceptibility differs from his result.

The comparison of the three solutions shows that one of the non-Hermitian approximations not only is easiest to use but seems to give reasonable results. Finally in chapter 4, the results of the infinite spin limit are derived.

The second part of this thesis is concerned with the semi-analytic evaluation of integrals appearing in the first part. It was possible however to take the study far beyond the antiferromagnet context and consider Lattice Green Functions which are currently a very active area of research. We give a brief introduction to Lattice Green Functions, reprint a paper we have published on the subject and then show how the necessary antiferromagnetic integrals may be handled.

CHAPTER 2

GENERAL FORMULATION

A. HAMILTONIAN

Let us consider a lattice system composed of N atoms or particles each having localized spin. Further consider only those lattices which are separable into two interpenetrating sublattices of $N/2$ spins each. In such a system each spin is subject to a torque which we will assume is due to three interactions: a Heisenberg exchange interaction taken only between nearest neighbour spins, a Zeeman force due to an external magnetic field and a uniaxial force which is a result of anisotropy.

The antiferromagnetic model considered in this thesis is then characterized by the following Hamiltonian

$$H = J \sum_{i\delta} \vec{S}_{1i} \cdot \vec{S}_{2i+\delta} + J \sum_{j\delta} \vec{S}_{1j+\delta} \cdot \vec{S}_{2j} + g\mu_B \vec{B} \cdot \left[\sum_i \vec{S}_{1i} + \sum_j \vec{S}_{2j} \right] - D \left[\sum_i (S_{1i}^z)^2 + \sum_j (S_{2j}^z)^2 \right] \qquad (2.A.1)$$

where

a) J is the modulus of the Heisenberg exchange constant

b) $\sum_i$ is a sum over sublattice 1

c) $\sum_j$ is a sum over sublattice 2

d) $\sum_{\delta}$ is a sum over z nearest neighbors

e) $\vec{S}_{1i}$ is the spin of the i site of sublattice 1

f) $\vec{S}_{2j}$ is the spin of the j site of sublattice 2

g) g is Lande factor

h) μ_B is Bohr Magneton

i) $\vec{B}$ is the external magnetic field taken to be in the zx plane in order to allow calculation of the parallel and perpendicular susceptibilities

j) D is the coefficient of uniaxial anisotropy.

In order to discuss each part of (2.A.1) separately, we define H_1, V_1, V_2 and V_3 such that

$$H = J H_1 + g\mu_B B_z V_1 + g\mu_B B_x V_2 - D V_3 \,. \qquad (2.A.2)$$

B. PROPERTIES OF LOCALIZED SPIN

A single spin operator satisfies the usual angular momentum commutation relations which can be written in the form

$$[S^z, S^+] = S^+ \,, \quad [S^z, S^-] = -S^- \qquad (2.B.1)$$

$$[S^+, S^-] = 2S^z$$

where

$$S^{\pm} = S^x + i S^y \,. \qquad (2.B.2)$$

The ground state $|0\rangle$ is defined by

$$S^- |0\rangle = 0 \quad , \quad S^z |0\rangle = -S|0\rangle \tag{2.B.3}$$

and the excited states by

$$|n\rangle = \left[(2S)^n n!\right]^{-1/2} (S^+)^n |0\rangle \tag{2.B.4}$$

where the state $|n\rangle$ has n spin deviations from the ground state. These vectors are orthogonal and have norm given by

$$\langle n|m\rangle = \delta_{n,m} F_n \tag{2.B.5}$$

$$F_n = 1 \cdot \left(1 - \tfrac{1}{2S}\right) \cdots\cdots \left(1 - \tfrac{(n-1)}{2S}\right) .$$

One can easily construct an orthonormal basis by dividing $|n\rangle$ by $F_n^{1/2}$. In the orthonormal basis the spin operators have a matrix representation (see figure 2) such that

$$S^z_{k\ell} = -(S-k+1)\,\delta_{k\ell}$$

$$S^+_{k\ell} = \left[1 - \tfrac{(\ell-1)}{2S}\right]^{1/2} \ell^{1/2} (2S)^{1/2}\, \delta_{k,\ell+1} \tag{2.B.6}$$

$$S^-_{k\ell} = \left[1 - \tfrac{(k-1)}{2S}\right]^{1/2} k^{1/2} (2S)^{1/2}\, \delta_{k+1,\ell}$$

$$1 \le k \le (2S+1),$$

$$1 \le \ell \le (2S+1) .$$

FIGURE 2.

Spin matrices in an orthonormal basis.

$$S^z = \begin{bmatrix} -S & & & & \\ & -(S-1) & & 0 & \\ & & -(S-2) & & \\ & & & \ddots & \\ & 0 & & & S \end{bmatrix}$$

$$S^+ = \begin{bmatrix} 0 & & & & \\ (2S)^{1/2} & 0 & & 0 & \\ & (4S)^{1/2}\left(1-\frac{1}{2S}\right)^{1/2} & & & \\ & & \ddots & & \\ & 0 & & (2S)^{1/2} & 0 \end{bmatrix}$$

S^- is the transpose of S^+.

C. PROPER SPIN SPACE and GROUND STATE

A spin operator at a lattice site satisfies the relations given in the preceeding section. We now require that spin operators at different sites commute i.e.

$$[\vec{S}_{1i}, \vec{S}_{1k}] = [\vec{S}_{2i}, \vec{S}_{2k}] = 0 \qquad (2.C.1)$$

so that we are dealing with a representation involving the direct product of N spin operators. A basis for the Hamiltonian (2.A.1) is therefore a direct product of states of the form (2.B.4). The space defined by this basis is $(2S+1)^N$ dimensional and is called the proper spin space. Our use of "proper" and "improper" in this connection follows Dyson (1956).

In the limit of classical spin (see appendix I) it is easily seen that our Hamiltonian will have a ground state such that spins on opposite sublattices are oppositely directed (see as well Herbert 1970). We therefore rotate every spin on sublattice 2 by 180 degrees so that (2.B.3) becomes

$$\begin{aligned} &S_{1i}^z |0_{1i}\rangle = -S|0_{1i}\rangle, \quad S_{2j}^z |0_{2j}\rangle = S|0_{2j}\rangle, \\ &S_{1i}^- |0_{1i}\rangle = 0, \quad S_{2j}^+ |0_{2j}\rangle = 0, \qquad (2.C.2) \\ &S_{1i}^+ |0_{1i}\rangle = (2S)^{1/2} |1\rangle, \quad S_{2j}^- |0_{2j}\rangle = (2S)^{1/2} |1\rangle. \end{aligned}$$

A state of interest can be represented by the direct product

$$|0_{11}\rangle \cdots |0_{1i}\rangle \cdots |0_{1N/2}\rangle |0_{21}\rangle \cdots |0_{2j}\rangle \cdots |0_{2N/2}\rangle$$

or by

$$\{|0_1\rangle |0_2\rangle\} \quad . \tag{2.C.3}$$

This state is not an eigenstate of H for finite spin. As a consequence the ground state has zero point spin wave excitations and adjacent spins are not quite oppositely directed.

It is convenient to rewrite H in terms of S^+ and S^- as follows

$$H_1 = \sum_{i\delta} \left[S^z_{1i} S^z_{2i+\delta} + \frac{1}{2}\left(S^+_{1i} S^-_{2i+\delta} + S^-_{1i} S^+_{2i+\delta} \right) \right]$$

$$+ \sum_{j\delta} \left[S^z_{1j+\delta} S^z_{2j} + \frac{1}{2}\left(S^+_{1j+\delta} S^-_{2j} + S^-_{2j+\delta} S_{2j} \right) \right]$$

$$V_1 = \sum_i S^z_{1i} + \sum_j S^z_{2j}$$

$$V_2 = \frac{1}{2} \sum_i \left(S^+_{1i} + S^-_{1i} \right) + \frac{1}{2} \sum_j \left(S^+_{2j} + S^-_{2j} \right) \tag{2.C.4}$$

$$V_3 = \sum_i \left(S^z_{1i} \right)^2 + \sum_j \left(S^z_{2j} \right)^2 \quad .$$

The raising and lowering operators in H_1 and V_2 are the cause of the state (2.C.3) not being an eigenstate of H. In general the ground state of an antiferromagnet is a

linear combination of spin deviated states. Only in the limit of classical spin does (2.C.3) become the ground state.

The ground state of the antiferromagnet has been studied by many authors - Anderson(1951,1952), Kubo(1952), Oguchi(1960) , Davis(1960), Marshall(1955), Lieb and Mattis (1962), and Pratt(1961) and although still a live issue because of the kinematical effect (Herbert 1970, Szaniecki 1971), is not a particular concern in this work.

D. PROPERTIES OF BOSON OPERATORS

Boson operators are defined by the relations

$$[b, b^+] = 1 \quad , \quad [b^+, b^+] = 0 \qquad (2.D.1)$$

and b^+ is the adjoint of b . Letting the vacuum state of our boson system be $|0\rangle$, it satisfies

$$\langle 0|0\rangle = 1 \quad , \quad b|0\rangle = 0 \quad . \qquad (2.D.2)$$

The excited states are orthonormal if we define them as

$$|n\rangle = (n!)^{-1/2} (b^+)^n |0\rangle \quad , \quad n \geq 0 \qquad (2.D.3)$$

that is $\langle m|n\rangle = \delta_{m,n}$.

It is also convenient to define a number operator $\hat{n} = b^+ b$ and it follows that

$$[b^+, \hat{n}] = -b^+ \quad , \quad [b, \hat{n}] = b \qquad (2.D.4)$$

and $\hat{n}\,|n) = n\,|n)$.

Note that our boson operators are defined on an infinite dimensional space. In order to set up a correspondence between spin space and boson space one may either enlarge the $(2S+1)$ dimensional spin space or contract the boson space; we choose the former course though other authors use the latter(Dyson 1956).

Matrices for boson operators (see figure 3) are

$$\hat{n}_{\ell m} = (m-1)\,\delta_{\ell,m} \quad ,$$

$$b^{+}_{\ell m} = m^{1/2}\,\delta_{\ell,m+1} \quad , \qquad (2.D.5)$$

$$b_{\ell m} = m^{1/2}\,\delta_{\ell+1,m} \quad .$$

E. EXTENDED SPIN SPACE

Consider the spin matrices (see figure 2) and make the natural extension to infinite dimensional space (see figure 4) that is

$$S^{z}_{k\ell} = -(S-k+1)\,\delta_{k\ell}$$

$$S^{+}_{k\ell} = \left[1-\frac{(\ell-1)}{2S}\right]^{1/2} \ell^{1/2}\,(2S)^{1/2}\,\delta_{k,\ell+1} \qquad (2.E.1)$$

$$S^{-}_{k\ell} = \left[1-\frac{(k-1)}{2S}\right]^{1/2} k^{1/2}\,(2S)^{1/2}\,\delta_{k+1,\ell}$$

FIGURE 3.

Boson matrices

$$\hat{n} = \begin{bmatrix} 0 & & & & 0 \\ & 1 & & & \\ & & 2 & & \\ & & & 3 & \\ 0 & & & & \ddots \end{bmatrix}$$

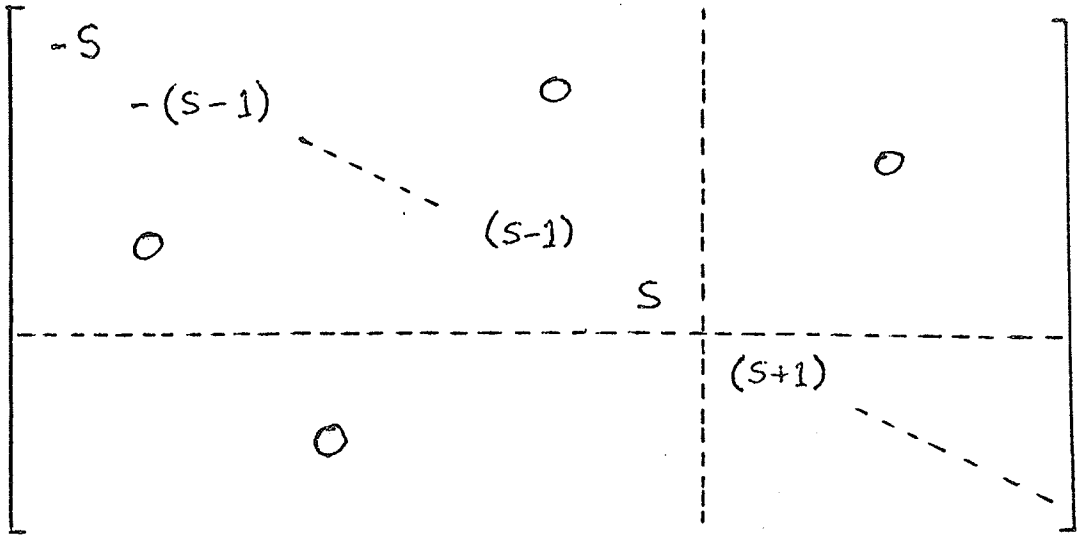
$$b^{+} = \begin{bmatrix} 0 & & & & 0 \\ 1^{1/2} & 0 & & & \\ & 2^{1/2} & 0 & & \\ & & 3^{1/2} & & \\ 0 & & & & \ddots \end{bmatrix}$$

b is the transpose of b^{+}

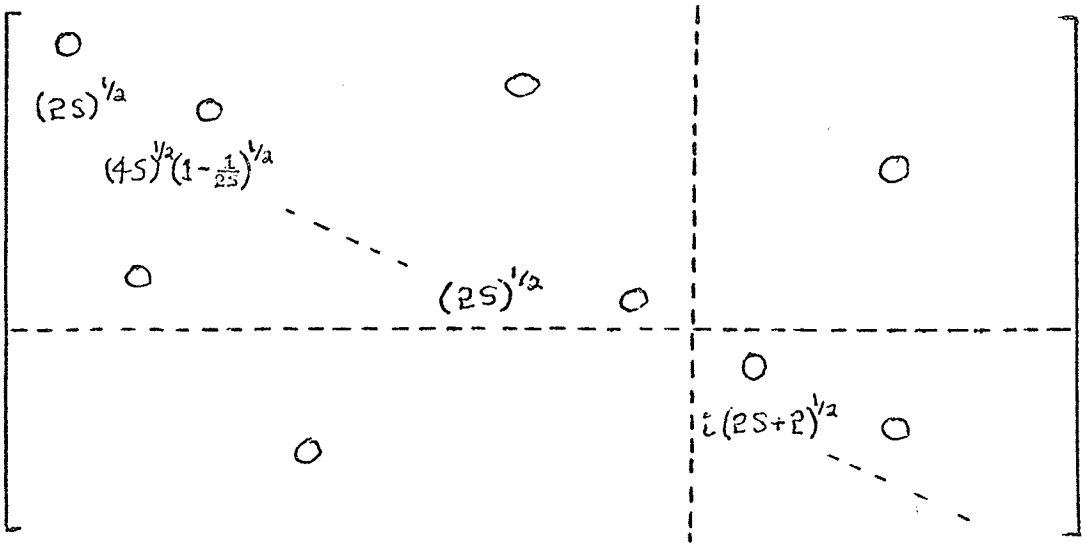
FIGURE 4.

Spin matrices in extended spin space partitioned into proper and improper parts.

$S^z =$



$S^+ =$



S^- is the transpose of S^+

but k and ℓ range from 1 to ∞ . Such matrices satisfy the same commutation relations (2.B.1) as the original spin matrices; however S^+ and S^- are no longer mutually adjoint but satisfy

$$(S^+)^+ = (S^-)^* \qquad (2.E.2)$$

This means that $S^x = S^+ + S^-$ is not a Hermitian operator in extended space. From the matrix representation (figure 4) one can show that S^x is normal (S^x commutes with $(S^x)^+$) and symmetric.

These matrices are defined on a larger space which is formed by adjoining an improper space to the proper spin space. Taking the direct product of N such spaces gives a space for the Hamiltonian H which is made up of a $(2S+1)^N$ dimensional proper spin space and an infinite dimensional improper spin space. H is no longer a Hermitian operator in extended space.

The effect of operating in this extended spin space with operators which originally are defined in the proper space is termed the kinematical effect by Dyson (1956). Since this will lead to an error in calculating the eigenvalues of H it is referred to as the kinematical interaction. We shall investigate the consequences of using this extended spin space in the section (3.A). We can note however that in the limit of infinite spin the proper spin space becomes infinite and the kinematical

interaction is zero. This means that at least there exists a solution which is valid for classical spin. The present work adopts the philosophy inherent in most of the investigations of spin wave problems, that the error introduced in using the extended space should be small for large spin. The consensus of previous investigation suggests that the results so obtained will be useful even for $S = 1/2$.

F. HOLSTEIN-PRIMAKOFF TRANSFORMATION

The Holstein-Primakoff (HP) transformation is a mapping from the extended spin space to a boson space. This mapping is easily constructed by noticing that S^+ (see equation 2.E.5) is a product of two matrices

$$S^+_{km} = \sum_{\ell} A_{k\ell} B_{\ell m} \qquad (2.F.1)$$

where

$$A_{km} = (2S)^{1/2} m^{1/2} \delta_{k,m+1} \qquad (2.F.2)$$

$$B_{\ell m} = [1 - \ell/2S]^{1/2} \delta_{\ell,m} \; . \qquad (2.F.3)$$

$B_{\ell m}$ is a diagonal matrix and has elements which are functions of $\hat{n}$ (see equation 2.D.6). A_{km} is a simple multiple of b^+ (see equation 2.D.5). The conventional method for constructing the HP transformation is somewhat different (see for example Kittel, Quantum Theory of Solids).

The full mapping for each spin on both sublattices is represented in operator notation by (henceforth we let $\hat{n} = n$)

$$S_{1i}^z \rightarrow -S + n_{1i}^+$$

$$S_{1i}^+ \rightarrow (2S)^{1/2} b_{1i}^+ \left(1 - \tfrac{1}{2S} n_{1i}\right)^{1/2}$$

$$S_{1i}^- \rightarrow (2S)^{1/2} \left(1 - \tfrac{1}{2S} n_{1i}\right)^{1/2} b_{1i} \qquad \text{(2.F.4)}$$

$$S_{2j}^z \rightarrow S - n_{2j}$$

$$S_{2j}^+ \rightarrow (2S)^{1/2} \left(1 - \tfrac{1}{2S} n_{2j}\right)^{1/2} b_{2j}$$

$$S_{2j}^- \rightarrow (2S)^{1/2} b_{2j}^+ \left(1 - \tfrac{1}{2S} n_{2j}\right)^{1/2}$$

This transformation is linear, unitary, preserves the spin commutation relation (canonical), and therefore preserves the eigenvalues of H .

In the boson representation the Hamiltonian becomes

$$H_1 = \tfrac{1}{2} \sum_{i\delta} \Big\{ \left(-S^2 + S n_{1i} + S n_{2j+\delta} - n_{1i} n_{2j+\delta}\right)$$

$$+ 2S\, b_{1i}^+ \left(1 - \tfrac{1}{2S} n_{1i}\right)^{1/2} b_{2i+\delta}^+ \left(1 - \tfrac{1}{2S} n_{2j+\delta}\right)^{1/2} \Big\}$$

$$+ \tfrac{1}{2} \sum_{j\delta} \Big\{ \left(-S^2 + S n_{1i+\delta} + S n_{2j} - n_{1j+\delta} n_{2j}\right)$$

$$+ 2S\, b_{1j+\delta}^+ \left(1 - \tfrac{1}{2S} n_{1j+\delta}\right)^{1/2} b_{2j}^+ \left(1 - \tfrac{1}{2S} n_{2j}\right)^{1/2} \Big\}$$

$+$ hermitian conjugate

$$V_1 = \sum_i n_{1i} - \sum_j n_{2j} \qquad (2.F.5)$$

$$V_2 = \frac{(2S)^{1/2}}{2} \sum_i b_{1i}^{+} \left(1 - \frac{1}{2S} n_{1i}\right)^{1/2} + \frac{(2S)^{1/2}}{2} \sum_j b_{2j}^{+} \left(1 - \frac{1}{2S} n_{2j}\right)^{1/2} + \text{h.c.}$$

$$V_3 = \sum_i \left(S^2 - 2S n_{1i} + n_{1i} n_{1i}\right) + \sum_j \left(S^2 - 2S n_{2j} + n_{2j} n_{2j}\right)$$

A major problem of the HP approach is now apparent; in order to proceed with the calculations it is necessary to expand the square root operators found in the above equation (Oguchi 1960). Such an expansion is not valid for $n_i > 2S$ and, even in the proper space, any finite number of terms only approximately represents the Hamiltonian. However, this does have the advantage of making H Hermitian. Retaining three terms in the expansion, which amounts to an expansion in S to order $1/S^2$ (Harris 1968), we get

$$H_1 = \frac{1}{2} \sum_{i\delta} \left[\left(-S^2 + S n_{1i} + S n_{2i+\varepsilon} - n_{1i} n_{2i+\delta}\right) + 2S\, b_{1i}^{+} b_{2i+\varepsilon}^{+} - \frac{1}{2} b_{1i}^{+} b_{2i+\varepsilon}^{+} \left(n_{1i} + n_{2i+\delta}\right) + \frac{1}{8S} b_{1i}^{+} b_{2i+\varepsilon}\, n_{1i}\, n_{2i+\delta} - \frac{1}{16S} b_{1i}^{+} b_{2i+\varepsilon}^{+} \left(n_{1i}\, n_{1i} + n_{2i+\delta}\, n_{2i+\varepsilon}\right)\right]$$

$$+\frac{1}{2}\sum_{j\delta}\Big[(-S^2+Sn_{1j+\delta}+Sn_{2j}+n_{1j+\delta}n_{2j})$$

$$+2S\,b^{+}_{1j+\delta}b_{2j}-\frac{1}{2}b^{+}_{1j+\delta}b^{+}_{2j}(n_{1j+\delta}+n_{2j})$$

$$+\frac{1}{8S}\,b^{+}_{1j+\delta}b^{+}_{2j}\,n_{1j+\delta}\,n_{2j} \qquad (2.F.6)$$

$$-\frac{1}{16S}\,b^{+}_{1j+\delta}b_{2j}\,(n_{1j+\delta}n_{1j+\delta}+n_{2j}n_{2j})\Big]$$

$$+\ \text{h.c.}$$

$$V_2=\frac{(2S)^{1/2}}{2}\sum_i\Big(b^{+}_{1i}-\frac{1}{4S}\,b^{+}_{1i}n_{1i}-\frac{1}{32S^2}b^{+}_{1i}n_{1i}n_{1i}\Big)$$

$$+\frac{(2S)^{1/2}}{2}\sum_j\Big(b^{+}_{2j}-\frac{1}{4S}\,b^{+}_{2j}n_{2j}-\frac{1}{32S^2}\,b^{+}_{2j}n_{2j}n_{2j}\Big)$$

$$+\ \text{h.c.}$$

V_1 and V_3 remain unchanged.

At this stage it is possible to take advantage of the translational invariance of the lattice and for this purpose we introduce Fourier transform operators. Let

$$b_{1i}=\Big(\frac{2}{N}\Big)^{1/2}\sum_k c_k\,e^{i\vec{k}\cdot\vec{R}_{1i}}$$

$$b^{+}_{1i}=\Big(\frac{2}{N}\Big)^{1/2}\sum_k c^{+}_k\,e^{-i\vec{k}\cdot\vec{R}_{1i}}$$

$$b_{2j}=\Big(\frac{2}{N}\Big)^{1/2}\sum_k d_k\,e^{-i\vec{k}\cdot\vec{R}_{2j}} \qquad (2.F.7)$$

$$b^{+}_{2j}=\Big(\frac{2}{N}\Big)^{1/2}\sum_k d_k^{+}\,e^{i\vec{k}\cdot\vec{R}_{2j}}$$

The reciprocal vectors $\vec{k}$ (the vector dependence of subscripts will be omitted) defined by this operation satisfy the usual property

$$\vec{k} \cdot \vec{R}_{ii} = 2\pi \times (\text{integer}) \qquad (2.F.8)$$

where R_{ii} is the position of the ith atom. This transform can be thought of as a further mapping onto reciprocal space that does not change the eigenvalues of H.

With this substitution we obtain

$$H_1 = -NzS^2 + azS \sum_k \left(c_k^+ c_k + d_k^+ d_k \right.$$

$$\left. + \gamma(k)\, c_k^+ d_k^+ + \gamma(k)\, c_k d_k \right) \qquad (2.F.9)$$

$$- \frac{z}{N} \sum_{kk'q}' \left[4\gamma(k-k')\, c_k^+ c_{k'}\, d_{k+q}^+ d_{k'+q} \right.$$

$$+ \gamma(k+q)\, c_{k'}^+ c_k c_{k+q} d_{k+q} \left(1 + \tfrac{1}{8S}\right)$$

$$+ \gamma(k+q)\, c_{k'+q}^+ c_k^+ c_{k'} d_{k+q} \left(1 + \tfrac{1}{8S}\right)$$

$$+ \gamma(k)\, c_k d_{k'}^+ d_{k+q} d_{k'-q} \left(1 + \tfrac{1}{8S}\right)$$

$$\left. + \gamma(k)\, c_k^+ d_{k+q}^+ d_{k'-q} d_{k'} \left(1 + \tfrac{1}{8S}\right) \right]$$

$$- \frac{z}{4SN} \sum_{kk'k''qq'} \Big[c_k d^\dagger_{k'} d^\dagger_{k''} d_{k+q} d_{k'+q'} d_{k''-q'+q} \gamma(k)$$

$$+ c^\dagger_{k'} c^\dagger_{k''} c_{k+q} c_{k'+q'} c_{k''-q'+q} d_k \gamma(k)$$

$$- 2 c^\dagger_k c_{k'} c_{k''} d^\dagger_{k+q} d_{k'+q'} d_{k''+q-q'} \gamma(k-k'-k'')$$

$$+ c^\dagger_k d^\dagger_{k+q} d^\dagger_{k'+q} d^\dagger_{k''-q'+q} d_{k'} d_{k''} \gamma(k)$$

$$+ c^\dagger_{k+q} c^\dagger_{k'+q'} c^\dagger_{k''-q'+q} c_{k'} c_{k''} d_k \gamma(k)$$

$$- 2 c^\dagger_{k'} c^\dagger_{k''} c_k d^\dagger_{k'+q} d^\dagger_{k''+q-q'} d_{k+q} \gamma(k-k'-k'') \Big]$$

$$V_1 = \sum_k (c^\dagger_k c_k - d^\dagger_k d_k)$$

$$V_2 = \frac{(SN)^{1/2}}{2} \Big[c^\dagger_0 + d^\dagger_0$$

$$- (1 + \tfrac{1}{8S}) \frac{1}{2SN} \sum_{kk'} (c^\dagger_k c^\dagger_{k'} c_{k+k'} + d^\dagger_k d^\dagger_{k'} d_{k+k'})$$

$$- \frac{1}{8N^2S^2} \sum_{kk'k''q} (c^\dagger_k c^\dagger_{k'} c^\dagger_{k''} c_q c_{k+k'+k''-q}$$

$$+ d^\dagger_k d^\dagger_{k'} d^\dagger_{k''} d_q d_{k+k'+k''-q}) + \text{h.c.}$$

$$V_3 = NS^2 - 2S \sum_k (c^\dagger_k c_k - d^\dagger_k d_k)(1 - \tfrac{1}{2S})$$

$$+ \frac{2}{N} \sum_{kk'q} (c^\dagger_k c_{k'} c_{k+q} c_{k'-q} + d^\dagger_k d^\dagger_{k'} d_{k+q} d_{k'-q})$$

where

$$\gamma(k) = \frac{1}{z} \sum_{\delta} e^{i\vec{k}\cdot\vec{R}_{\delta}} \qquad (2.F.10)$$

and we have used

$$\delta_{k,k'} = \frac{2}{N} \sum_{i} e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_{i}} \quad . \qquad (2.F.11)$$

This representation is Hermitian and the boson operators have been commuted so that they appear in normal order (c^{+} preceeds c , d^{+} preceeds d and c preceeds d). It is important to note that the eigenvalues of this Hamiltonian are not the same as those of (2.A.1) for two reasons. First (2.A.1) operates in a proper spin space and (2.F.9) operates in an extended space and second, even if we allow (2.A.1) to operate in extended spin space then (2.F.9) is still not equivalent since we have used an expansion for square root operators.

G. DYSON-MALEEV I TRANSFORMATION

In figure 5 we define a diagonal transformation matrix $T_{lm} = t_{l}\,\delta_{lm}$ which has elememts given by the recurrance relation

$$t_{l} = t_{l-1}\left[1 - \frac{(l-2)}{2S}\right]^{-1/2} + \delta_{l,2S+2} \quad , \; l \geq 2 \qquad (2.G.1)$$

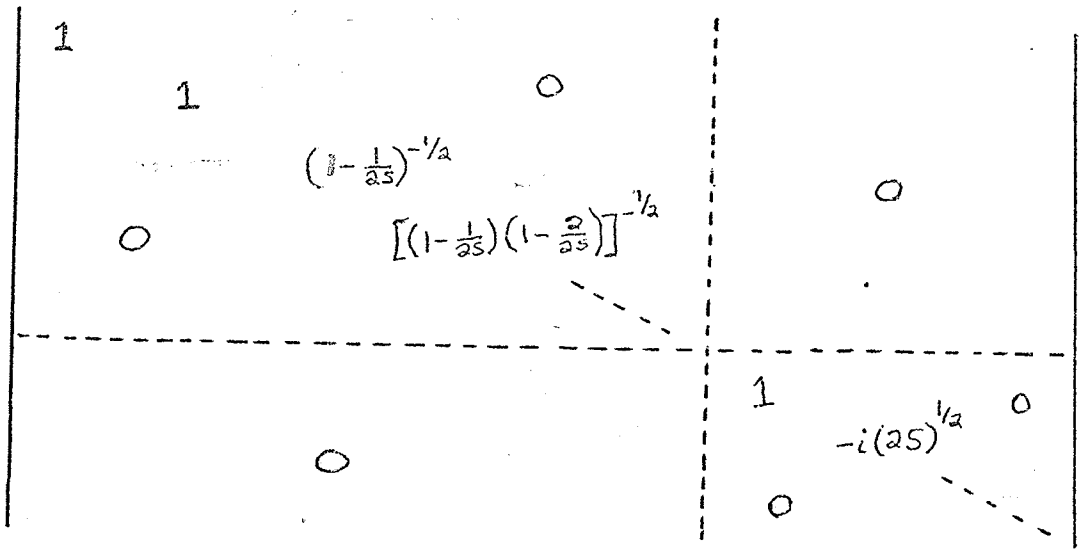
with $t_{1} = 1$. It is easily seen that such a matrix is non-singular and non-unitary. If one transforms S^{z},

FIGURE 5.

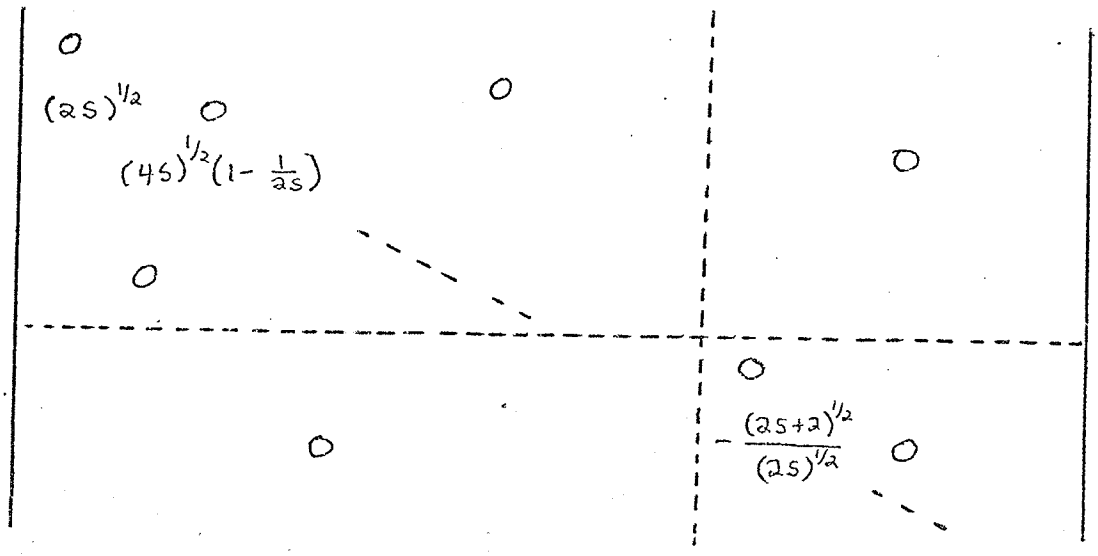
Transformation matrix T corresponding to Dyson-Maleev transformation.

Transformed matrix S^{+}.

$T =$


1
1
$(1-\frac{1}{2s})^{-1/2}$
$[(1-\frac{1}{2s})(1-\frac{2}{2s})]^{-1/2}$
1
$-i(2s)^{1/2}$

$\tilde{S}^{+} =$


$(2s)^{1/2}$
$(4s)^{1/2}(1-\frac{1}{2s})$
$-\frac{(2s+2)^{1/2}}{(2s)^{1/2}}$

S^+ and S^- (see figure 3) with T so that

$$T^{-1} S^z T = \tilde{S}^z ,$$
$$T^{-1} S^+ T = \tilde{S}^+ , \quad T^{-1} S^- T = \tilde{S}^- . \tag{2.G.2}$$

a new representation for the spin matrices is obtained. This representation was introduced by Dyson (1956) and Maleev (1958). The Dyson-Maleev I (DMI) representation uses this transformation on the spin operators of sublattice 2 and transforms sublattice 1 according to the inverse matrix T^{-1}.

The full one-to one mapping (also used by Armour 1971, Liu 1966, Harris 1968) for each spin and both sublattices is represented in operator notation by

$$\begin{aligned}
S_{1i}^z &\rightarrow -S + b_{1i}^+ b_{1i} \\
S_{1i}^+ &\rightarrow (2S)^{1/2} b_{1i}^+ \\
S_{1i}^- &\rightarrow (2S)^{1/2} \left[b_{1i} - (2S)^{-1} b_{1i}^+ b_{1i} b_{1i} \right] \\
S_{2j}^z &\rightarrow S - b_{2j}^+ b_{2j} \\
S_{2j}^+ &\rightarrow (2S)^{1/2} b_{2j} \\
S_{2j}^- &\rightarrow (2S)^{1/2} \left[b_{2j}^+ - (2S)^{-1} b_{2j}^+ b_{2j}^+ b_{2j} \right] .
\end{aligned} \tag{2.G.3}$$

This transformation, as we will see, yields terms in the Hamiltonian involving up to 4 operators. There is an alternate transformation (DMII) which uses the same T for both sublattices and we will investigate this in section H where we will see that the Hamiltonian involves terms up to 6 operators.

The DMI representation has the effect of introducing a "Hamiltonian" which is not Hermitian even in the proper subspace. Although this seems like a disadvantage it enables us to overcome the expansion required by the HP method. In the boson representation H becomes

$$
\begin{aligned}
H_1 = \sum_{i\delta} \Big[& -S^2 + S n_{1i} + S n_{2i+\delta} - n_{1i} n_{2i+\delta} \\
& + S\, b_{1i}^{+} b_{2i+\delta}^{+} - \tfrac{1}{2}\, b_{1i}^{+} b_{2i+\delta} n_{2i+\delta} \\
& + S\, b_{1i} b_{2i+\delta} - \tfrac{1}{2}\, n_{1i}\, b_{1i} b_{2i+\delta} \Big] \\
+ \sum_{j\delta} \Big[& -S^2 + S n_{1j+\delta} + S n_{2j} - n_{1j+\delta} n_{2j} \\
& + S\, b_{1j+\delta}^{+} b_{2j}^{+} - \tfrac{1}{2}\, b_{1j+\delta}^{+} b_{2j}^{+} n_{2j} \\
& + S\, b_{1j+\delta} b_{2j} - \tfrac{1}{2}\, n_{1j+\delta} b_{1j+\delta} b_{2j} \Big]
\end{aligned}
$$

$$
V_1 = \sum_i n_{1i} - \sum_j n_{2j} \qquad (2.G.4)
$$

$$V_2 = \frac{(2S)^{1/2}}{2} \sum_i \left(b_{1i}^+ + b_{1i} - \frac{1}{2S} n_{1i} b_{1i} \right)$$

$$+ \frac{(2S)^{1/2}}{2} \sum_j \left(b_{2j} + b_{2j}^+ - \frac{1}{2S} b_{2j} n_{2j} \right)$$

$$V_3 = \sum_i \left(S^2 - 2S n_{1i} + n_{1i} n_{1i} \right)$$

$$+ \sum_j \left(S^2 - 2S n_{2i} + n_{2i} n_{2i} \right) \quad .$$

Performing a Fourier transform (see 2.F.7) we obtain a representation for H in reciprocal boson space

$$H_1 = - N z S^2 + 2 z S \sum_k \left[c_k^+ c_k + d_k^+ d_k \right.$$

$$\left. + \gamma(k) c_k^+ d_k^+ + \gamma(k) c_k d_k \right]$$

$$- \frac{z}{N} \sum_{kk'q} \left[4\gamma(k-k') c_k^+ c_k d_{k+q}^+ d_{k'+q} \right.$$

$$+ 2\gamma(k+q) c_{k'}^+ c_k c_{k'+q} d_{k+q}$$

$$\left. + 2\gamma(k) c_k^+ d_{k+q}^+ d_{k'-q} d_{k'} \right]$$

$$V_1 = \sum_k \left(c_k^+ c_k - d_k^+ d_k \right) \qquad (2.G.5)$$

$$V_2 = \frac{(SN)^{1/2}}{2} \left[c_o^+ + c_o + d_o^+ + d_o \right.$$

$$\left. - \frac{1}{SN} \sum_{kk'} \left(c_{k+k'}^+ c_k c_{k'} + d_k^+ d_{k'}^+ d_{k+k'} \right) \right]$$

$$V_3 = NS^2 - 2S\sum_k (c_k^+ c_k - d_k^+ d_k)\left(1 - \frac{1}{2S}\right)$$

$$+ \frac{2}{N}\sum_{kk'q} (c_k^+ c_{k'}^+ c_{k+q} c_{k'-q} + d_k^+ d_{k'}^+ d_{k+q} d_{k'-q}) \,.$$

This representation of H now has the following desirable properties; it has a finite number of terms and, in the extended space, it has the same eigenvalues as H .

H. DYSON-MALEEV II TRANSFORMATION

This transformation denoted by DMII (also used by Armour 1971, Herbert 1970, Harris 1968, Collins and Tondon 1972) is characterized by

$$\begin{aligned}
S_{1i}^z &\to -S + b_{1i}^+ b_{1i} \\
S_{1i}^+ &\to (2S)^{1/2} b_{1i}^+ \\
S_{1i}^- &\to (2S)^{1/2}\left[b_{1i} - (2S)^{-1} b_{1i}^+ b_{1i} b_{1i}\right] \\
S_{2j}^z &\to S - b_{2j}^+ b_{2j} \\
S_{2j}^+ &\to (2S)^{1/2}\left[b_{2j} - (2S)^{-1} b_{2j}^+ b_{2j} b_{2j}\right] \\
S_{2j}^- &\to (2S)^{1/2} b_{2j}^+ \,.
\end{aligned} \qquad (2.H.1)$$

It represents a transformation by T^{-1} (see section G) for both sublattices.

Carrying out the same procedures as in section F and

G yields in the boson space

$$H_1 = \sum_{i\delta} \Big(-S^2 + S n_{1i} + S n_{2i+\delta} - n_{1i}\, n_{2i+\delta}$$
$$+ S\, b^+_{1i}\, b^+_{i+\delta} - \frac{1}{2}\, n_{1i}\, b_{1i}\, b_{2i+\delta}$$
$$+ S\, b_{1i}\, b_{2i+\delta} - \frac{1}{2}\, n_{2i+\delta}\, b_{2i+\delta}\, b_{1i}$$
$$+ \frac{1}{4S}\, n_{1i}\, b_{1i}\, n_{2i+\delta}\, b_{2i+\delta} \Big)$$
$$+ \sum_{j\delta} \Big(-S^2 + S n_{1j+\delta} + S n_{2j} - n_{1j+\delta}\, n_{2j}$$
$$+ S\, b^+_{1j+\delta}\, b^+_{2j} - \frac{1}{2}\, n_{2j}\, b_{2j}\, b_{1j+\delta}$$
$$+ S\, b_{1j+\delta}\, b_{2j} - \frac{1}{2}\, n_{1j+\delta}\, b_{1j+\delta}\, b_{2j}$$
$$+ \frac{1}{4S}\, n_{2j}\, b_{2j}\, n_{1j+\delta}\, b_{1j+\delta} \Big)$$

(2.H.2)

$$V_1 = \sum_i n_{1i} - \sum_j n_{2j}$$

$$V_2 = \frac{(2S)^{1/2}}{2} \sum_i \Big(b^+_{1i} + b_{1i} - \frac{1}{2S}\, b^+_{1i}\, b_{1i}\, b_{1i} \Big)$$
$$+ \frac{(2S)^{1/2}}{2} \sum_j \Big(b^+_{2j} + b_{2j} - \frac{1}{2S}\, b^+_{2j}\, b_{2j}\, b_{2j} \Big)$$

$$V_3 = \sum_i (S^2 - 2S n_{1i} + n_{1i}\, n_{1i}) + \sum_j (S^2 - 2S n_{2j} + n_{2j}\, n_{2j}) \,.$$

Using (2.F.7) again transform to reciprocal boson space, we obtain the following expression which is equivalent to (2.H.2):

$$H_1 = -NzS^2 + 2zS\sum_k [c_k^+ c_k + d_k^+ d_k + \gamma(k) c_k^+ d_k^+ + \gamma(k) c_k d_k]$$

$$- \frac{z}{N}\sum_{kk'q} 4\gamma(k-k') c_k^+ c_{k'} d_{k+q}^+ d_{k'+q}$$

$$+ 2\gamma(k) c_k d_{k'}^+ d_{k+q} d_{k'-q}$$

$$+ 2\gamma(k-q) c_{k'}^+ c_k c_{k'+q} d_{k+q}$$

$$+ \frac{2z}{N^2 S}\sum_{kk'k''qq'} c_k^+ c_{k'} c_{k''} d_{k+q}^+ d_{k'+q'} d_{k''+q-q'} \gamma(k-k'-k'')$$

$$V_1 = \sum_k (c_k^+ c_k - d_k^+ d_k) \qquad (2.H.3)$$

$$V_2 = \frac{(SN)^{1/2}}{2}\left[c_o^+ + c_o + d_o^+ + d_o - \frac{1}{SN}\sum_{kk'} (c_{k+k'}^+ c_k c_{k'} + d_{k+k'}^+ d_k d_{k'})\right]$$

$$V_3 = NS^2 - 2S\sum_k (c_k^+ c_k + d_k^+ d_k)\left(1 - \frac{1}{2S}\right)$$

$$+ \frac{2}{N}\sum_{kk'q} (c_k^+ c_{k'}^+ c_{k+q} c_{k'-q} + d_k^+ d_{k'}^+ d_{k+q} d_{k'-q}) .$$

The DMII representation has a different form than DMI but is equivalent and has the same eigenvalues. Note

the 6 operator terms in (2.H.2) and (2.H.3) that arise from the product $\sum S_{i}^{-} S_{2i+\delta}^{+}$ in (2.A.1). There are at least two other equivalent "Hamiltonians" formed by taking the adjoint of DMI and DMII but they do not appear to lead to any differences.

CHAPTER 3

METHODS OF APPROXIMATION

A. VARIATIONAL THEOREM

1) Conventional theory

In order to calculate the thermodynamic properties of H we can make use of the modified Peierls' theorem (Falk 1963, 1964). This theorem states that the exact free energy is always less than the sum of: (a) the free energy associated with an arbitrary trial ensemble $e^{-\beta H_o}$ and (b) the expectation, in the trial ensemble of the difference between the exact Hamiiltonian and the trial Hamiltonian. Mathematically this is expressed as

$$F_e \leq F_o + \langle H - H_o \rangle_o \qquad (3.A.1)$$

where

$$F_o = -\frac{1}{\beta} \ln \left[Tr\, e^{-\beta H_o} \right]$$

$$\langle H - H_o \rangle_o = Tr \left[e^{-\beta H_o} (H - H_o) \right] / Tr\, e^{-\beta H_o}$$

$$\beta = 1 / kT$$

and where

k = Boltzmann's constant

T = Absolute temperature .

Using this theorem a well known method of approximation becomes apparent. One chooses a trial Hamiltonian H_o which depends on one or more parameters α and minimizing expression (3.A.1) yields the "best" value for these parameters.

However, the proof of this theorem implicitly assumes that both H and H_o are Hermitian; if either H or H_o is non-Hermitian the inequality, in general, is not valid. Thus we can use this theory to calculate the properties of H only in the HP representation; in this case we obtain an upper bound for F_e. For the non-Hermitian cases we will need a generalization of this approach.

2) Statement of general variational theorem

In this section we are concerned with the case in which H is Hermitian and $H_o(\alpha)$ is non-Hermitian. Consider a very general statement of the variational theorem (Park, Quantum Theory, p.236):

Let ψ_n be an exact nth eigenfunction, which we will not assume normalized, and suppose that a small error or change is made in it. We require to know the corresponding change in the energy E_n. We write E_n as

$$E_n \int \psi_n^* \psi_n = \int \psi_n^* H \psi_n \qquad (3.A.2)$$

and then we let ψ_n become $\psi_n + \delta\psi_n$ and correspondingly,

E_n becomes $E_n + \delta E_n$. Taking differentials gives

$$\delta E_n \int \psi_n^* \psi_n + E_n \left(\int \delta\psi_n^* \psi_n + \int \psi_n^* \delta\psi_n \right)$$

$$= \int \delta\psi_n^* H \psi_n + \int \psi_n^* H \delta\psi_n \quad . \tag{3.A.2}$$

Using the Hermiticity of H and the fact that ψ_n is an exact eigenfunction gives for the right hand side

$$= E_n \left(\int \delta\psi_n^* \psi_n + \int \psi_n^* \delta\psi_n \right)$$

so that

$$\delta E_n \int \psi_n^* \psi_n = 0$$

or

$$\delta E_n = 0 \quad . \tag{3.A.4}$$

This shows that if one can guess close enough to the correct wave function for any stationary state whatever, then the error in $\langle H \rangle$ will always be a higher order in small quantities than that of the wave function.

If $\psi_n + \delta\psi_n$ and ψ_n are related by a non-unitary transformation the theorem remains valid (however $\delta\psi_n$ and $\delta\psi_n^*$ are no longer simply related). This theorem can be applied to the free energy by replacing H by $e^{-\beta H}$ and E_n by $e^{-\beta E_n}$ and noting that $e^{-\beta H}$ is Hermitian.

This property of the solution has been observed before and it is noted that although the variational functional is stationary at the exact solution, it has in general neither a minimum nor a maximum (Ah-Sam, Jensen and Smith 1971).

On the basis of the modified Peierls theorem and the general variational statement we postulate the following criterion: The "best" value for the parameters of the trial non-Hermitian Hamiltonian satisfies the condition

$$\frac{\partial}{\partial \alpha}\left[F_0 + \langle H - H_0 \rangle_0\right] = 0 \quad ; \qquad (3.A.5)$$

however, the free energy calculated in this way is not necessarily greater than the exact free energy F_e . To show that the lower bound no longer holds is simple- the free energy in general is complex and the ordering relationship is not defined.

Clearly one of the problems in the proof of (3.A.5) is the subjective nature of "best". One notes that any value for α forms an approximation to the solution so that the real problem is which to choose. If the minimum value for (3.A.1) is "best" for a Hermitian approximation then it minimizes the effect of higher order perturbations. In this sense one expects (3.A.5) to represent the "best" solution.

3) Proper spin space

In the following discussion we are concerned only with the proper subspace introduced in chapter 2. In the spin representation H is Hermitian and in the DM boson representation H is non-Hermitian. Choosing some approximation (H_o either Hermitian or non-Hermitian) to H in boson space in general will not be Hermitian when transformed back to spin space. Since the trace is independent of representation we see that any approximation to H in reciprocal boson space amounts to approximating a Hermitian Hamiltonian with one which in general is non-Hermitian.

It would be very fortunate if we could choose an approximate Hamiltonian in boson space which transforms into a Hermitan Hamiltonian in the spin representation. Such approximations are possible but they seem too naive to represent adequately the properties of an antiferromagnet. For example, if one took the Ising terms (z component of spin) these would transform into a Hermitian Hamiltonian in the spin representation but they do not predict spin wave excitations.

4) Kinematical effects

There is another problem associated with this theory. It can be shown that the spin Hamiltonian in extended spin space is not bounded below (Armour 1971)! This implies that any exact solution to the free energy is

meaningless. Fortunately an approximate theory saves us from this dilemma. Consider the following analogy:

We want to calculate the integral

$$\int_0^1 e^{-\beta(x-x^2)}\,dx \qquad (3.A.6)$$

for large values of β. An extension of the domain of x from $(0,1)$ to $(0,\infty)$ clearly indicates that

$$H = x - x^2 \qquad (3.A.7)$$

has no lower bound. If we approximate the integral by

$$\int_0^1 e^{-\beta x}(1+\beta x^2+\cdots)\,dx \qquad (3.A.8)$$

now an extension of the domain introduces only a small error provided we do not take too many terms in the expansion.

The analogy is complete by identifying trace with integration, improper space as domain $(1,\infty)$, and H_0 with x. The important consideration then is not the result of kinematical effect on H but, more important, the kinematical effect on H_0. If H_0 in improper space represents H_0 in proper spin space then we can say that we have an asymptotic series solution for H. We will show that H_0 is bounded below.

B. APPROXIMATE "HAMILTONIAN"

The basic criteria for an approximate Hamiltonian are that it represents the properties of the system and it has an exact solution. Usually this is guaranteed by selecting the dominant part of the full Hamiltonian and treating the remainder as small. For our case, at low temperatures, one expects that those terms containing the fewest number of boson operators will dominate due to the fact that the ground state has been anticipated by (2.C.2). Choosing those terms (see equations 2.F.9, 2.G.5 and 2.H.3) containing only one and two boson operators is a natural candidate for such a trial Hamiltonian.

In order to take into account the higher order terms we introduce eight parameters $\varepsilon_1 \ldots \varepsilon_4, \sigma_1 \ldots \sigma_4$ and obtain

$$H_o = -NJz\,S^2 + 2JzS \sum_k \left[\varepsilon_1 c_k^\dagger c_k + \varepsilon_2 d_k^\dagger d_k \right.$$

$$\left. + \varepsilon_3 \gamma(k) c_k^\dagger d_k^\dagger + \varepsilon_4 \gamma(k) c_k d_k \right] \qquad (3.B.1)$$

$$+ \frac{(SN)}{2} \left(\sigma_1 c_o + \sigma_2 c_o^\dagger + \sigma_3 d_o + \sigma_4 d_o^\dagger \right) .$$

Note that this is a non-Hermitian "Hamiltonian" but we will show that it has an exact solution and has real eigenvalues.

The solution of this operator involves the use of two non-unitary transformations. First make a shift in the zero mode terms represented by

$$
\begin{aligned}
c_k &= a_k + \delta_{k,0}\,(SN)^{1/2}\,\rho_1 \\
c_k^+ &= a_k^+ + \delta_{k,0}\,(SN)^{1/2}\,\rho_2 \\
d_k &= b_k + \delta_{k,0}\,(SN)^{1/2}\,\rho_3 \\
d_k^+ &= b_k^+ + \delta_{k,0}\,(SN)^{1/2}\,\rho_4 \ .
\end{aligned}
\tag{3.B.2}
$$

This transformation is not strictly linear but is an affine transformation and can be made linear only by introducing a larger dimension space. We will not discuss its non-linear aspects but refer the interested reader to Clark and Derrick (Mathematical Methods in Solid and Superfluid Theory). However, a similar transformation has been used before (Kubo 1952, Oguchi 1960) and it does not seem to lead to any difficulties.

Shifting the terms involving two boson operators gives rise to single, zero mode operators with coefficients involving ε_i and ρ_i . These terms can be made to cancel the one boson terms appearing in (3.B.1) by choosing the appropriate $\rho_1 , \ldots \rho_4$. The ρ_i which achieve this cancellation satisfy

$$
\begin{aligned}
\rho_1 &= -\frac{1}{4J_z S}\,(\varepsilon_2\sigma_2 - \varepsilon_3\sigma_3)/(\varepsilon_1\varepsilon_2 - \varepsilon_3\varepsilon_4) \\
\rho_2 &= -\frac{1}{4J_z S}\,(\varepsilon_2\sigma_1 - \varepsilon_4\sigma_4)/(\varepsilon_1\varepsilon_2 - \varepsilon_3\varepsilon_4)
\end{aligned}
\tag{3.B.3}
$$

$$\rho_3 = \frac{1}{2Jz\,S}\,(\varepsilon_1\sigma_4 - \varepsilon_3\sigma_1)\,/\,(\varepsilon_1\varepsilon_2 - \varepsilon_3\varepsilon_4)$$

$$\rho_4 = \frac{1}{2Jz\,S}\,(\varepsilon_1\sigma_3 - \varepsilon_4\sigma_2)\,/\,(\varepsilon_1\varepsilon_2 - \varepsilon_3\varepsilon_4)\ .$$

This leaves

$$H_0 = -NJz\,S^2 + 2Jz\,S\sum_k \left[\varepsilon_1 a_k^+ a_k + \varepsilon_2 b_k^+ b_k\right.$$

$$\left. + \varepsilon_3\gamma(k)\,a_k^+ b_k^+ + \varepsilon_4\gamma(k)\,a_k b_k\right] \qquad (3.B.4)$$

$$- 2Jz\,S^2 N\,(\varepsilon_1\rho_1\rho_2 + \varepsilon_2\rho_3\rho_4 + \varepsilon_3\rho_2\rho_4 + \varepsilon_4\rho_1\rho_3)\ .$$

A second transformation is needed to reduce this to a diagonal representation which can be done by means of a modified Bogulubov transformation (Oguchi 1960):

$$a_k = (\alpha_k\cosh\theta_{1k} - \beta_k^+\sinh\theta_{1k})\,/\,[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}$$

$$a_k^+ = (\alpha_k^+\cosh\theta_{2k} - \beta_k\sinh\theta_{2k})\,/\,[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}$$

$$b_k = (-\alpha_k^+\cosh\theta_{1k} + \beta_k\sinh\theta_{1k})\,/\,[\cosh(\theta_{1k}-\theta_{2k})]^{1/2} \qquad (3.B.5)$$

$$b_k^+ = (-\alpha_k\cosh\theta_{2k} + \beta_k^+\sinh\theta_{2k})\,/\,[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}\ .$$

This non-unitary transformation is canonical (see appendix 2) and therefore defines new boson operators α_k, β_k and α_k^+, β_k^+. Substituting in equation (3.B.4) and setting the coefficients of the terms $\alpha_k\beta_k$ and $\alpha_k^+\beta_k^+$ equal to

zero (see appendix 3) yields conditions on the parameters θ_{1k} and θ_{2k} as functions of $\varepsilon_1 .. \varepsilon_4$. Finally we obtain

$$H_0 = -NJ_z S^2 + 2J_z S \sum_k C\left[\alpha_k^+ \alpha_k - \beta_k^+ \beta_k\right]$$

$$+ 2J_z S \sum_k \left\{ \left[(A+B)^2 - (A^2 - D^2)\gamma(k)^2 \right]^{1/2} (\alpha_k^+ \alpha_k + \beta_k^+ \beta_k + 1) \right.$$

$$\left. - (A+B) \right\} \qquad (3.B.6)$$

$$- 2J_z S^2 N \left(\varepsilon_1 \rho_1 \rho_2 + \varepsilon_2 \rho_3 \rho_4 + \varepsilon_3 \rho_2 \rho_4 + \varepsilon_4 \rho_1 \rho_3 \right)$$

where

$$A = \frac{1}{2}(\varepsilon_3 + \varepsilon_4)$$

$$B = \frac{1}{2}(\varepsilon_1 + \varepsilon_2 - \varepsilon_3 - \varepsilon_4) \qquad (3.B.7)$$

$$C = \frac{1}{2}(\varepsilon_1 - \varepsilon_2)$$

$$D = \frac{1}{2}(\varepsilon_3 - \varepsilon_4) \quad .$$

In this representation the trial Hamiltonian is seen to be diagonal since it depends only on the number operators $n_k = \alpha_k^+ \alpha_k$ and $n_k' = \beta_k^+ \beta_k$; these take on values $0, 1 .. \infty$. For H_0 to have a lower bound places the following condition on its parameters

$$(A+B)^2 - (A^2 - D^2) - C^2 \geq 0 \quad . \qquad (3.B.8)$$

This condition is obtained by noting that $-1 \leq \gamma(k) \leq 1$. We now have an approximate Hamiltonian which is bounded below and has real eigenvalues.

The physical interpretation of (3.B.6) can be made in terms of spin waves. One excites both $\alpha_k^+ \alpha_k$ type waves and $\beta_k^+ \beta_k$ type waves and the difference in energy required to do this is given by $2C$. As we will see, C is directly proportional to an external magnetic field in the z direction so that a magnetic field is said to split the energy spectrum. In this picture the lower bound condition (3.B.8) is equivalent to requiring the dispersion function to be positive.

C. EXPECTATION VALUE OF H

In this section we will show how to calculate the following trace

$$Tr(He^{-\beta H_o}) \Big/ Tr(e^{-\beta H_o}) = \langle H \rangle_o \qquad (3.C.1)$$

where H_o is defined in section (3.B) and H is given in either of sections (2.F), (2.G) or (2.H).

A straight forward method consists of expressing H and H_o in a representation in which H_o is diagonal (i.e. the α, β representation) and then only the diagonal elements of H contribute to $\langle H \rangle_o$. Any term in H which can not be expressed as a function of $n_k = \alpha_k^+ \alpha_k$ and

$n_k' = \beta_k^+ \beta_k$ will be off diagonal and can be omitted. We then make use of the following results which are well known for boson statistics:

$$\langle n_\rho \rangle_0 = \langle n_\rho \rangle_0$$

$$\langle n_\rho n_\sigma \rangle_0 = \langle n_\rho \rangle_0 \langle n_\sigma \rangle_0 \qquad (3.C.2)$$

$$\vdots$$

$$\text{e+c.}$$

This gives an expression for $\langle H \rangle_0$ in terms of the average occupation numbers (of H_0) which are known.

The straight forward approach outlined above is not practical due to the amount of algebra involved; however, it is possible to simplify such a procedure and this will be done in appendix 4. In the following, we will simply motivate and then write down the end results.

If we define the following quantities

$$\alpha_1 = -\frac{1}{SN} \sum_k \langle c_k^+ c_k + d_k^+ d_k + \gamma(k) c_k^+ d_k^+ + \gamma(k) c_k d_k \rangle_0$$

$$\alpha_2 = -\frac{1}{SN} \sum_k \langle c_k^+ c_k + d_k^+ d_k \rangle_0 \qquad (3.C.3)$$

$$\alpha_3 = -\frac{1}{SN} \sum_k \langle c_k^+ c_k - d_k^+ d_k \rangle_0$$

$$\alpha_4 = -\frac{1}{SN} \sum_k \gamma(k) \langle c_k^+ d_k^+ - c_k d_k \rangle$$

then by transforming into the diagonal representation first by shifting according to (3.B.2) to obtain

$$\alpha_1 = -\frac{1}{SN}\sum_k \langle a_k^+ a_k + b_k^+ b_k + \gamma(k) a_k^+ b_k^+ + \gamma(k) a_k b_k \rangle_0 - (\rho_1\rho_2 + \rho_3\rho_4 + \rho_2\rho_4 + \rho_1\rho_3)$$

$$\alpha_2 = -\frac{1}{SN}\sum_k \langle a_k^+ a_k + b_k^+ b_k \rangle_0 - (\rho_1\rho_2 - \rho_3\rho_4)$$

$$\alpha_3 = -\frac{1}{SN}\sum_k \langle a_k^+ a_k - b_k^+ b_k \rangle_0 - (\rho_1\rho_2 - \rho_3\rho_4) \qquad (3.C.4)$$

$$\alpha_4 = -\frac{1}{SN}\sum_k \gamma(k) \langle a_k^+ b_k^+ - a_k b_k \rangle_0 - (\rho_2\rho_4 - \rho_1\rho_3)$$

where now the linear boson terms will have zero expectation and have been omitted. Next the Bogolubov transformation takes us into the α_k, β_k representation in which the approximate Hamiltonian is diagonal and the α_i's become (see appendix 2)

$$\alpha_1 = -\frac{1}{SN}\sum_k \left[(\langle n_k \rangle_0 + \langle n_k' \rangle_0 + 1)\left(\frac{\cosh\theta_k}{\cosh\Delta\theta_k} - \gamma(k)\sinh\theta_k\right) - 1 \right] - (\rho_1\rho_2 + \rho_3\rho_4 + \rho_2\rho_4 + \rho_1\rho_3)$$

$$\alpha_2 = -\frac{1}{SN}\sum_k \left[(\langle n_k \rangle_0 + \langle n_k' \rangle_0 + 1)\left(\frac{\cosh\theta_k}{\cosh\Delta\theta_k}\right) - 1 \right] - (\rho_1\rho_2 + \rho_3\rho_4)$$

$$\alpha_3 = -\frac{1}{SN}\sum_k (\langle n_k \rangle_0 - \langle n_k' \rangle_0) - (\rho_1\rho_2 - \rho_3\rho_4) \qquad (3.C.5)$$

$$\alpha_4 = -\frac{1}{SN}\sum_k \left[\gamma(k)(\langle n_k \rangle_0 + \langle n_k' \rangle_0 + 1)\ \frac{\cosh\theta_k}{\cosh\Delta\theta_k}\sinh\Delta\theta_k - 1 \right] - (\rho_2\rho_4 - \rho_1\rho_3)$$

or in the notation of appendix 2 and 3

$$\alpha_1 = \langle \hat{\alpha}_1 \rangle_0 - (\rho_1\rho_2 + \rho_3\rho_4 + \rho_2\rho_4 + \rho_1\rho_3)$$

$$\alpha_2 = \langle \hat{\alpha}_2 \rangle_0 - (\rho_1\rho_2 + \rho_3\rho_4)$$

$$\alpha_3 = \langle \hat{\alpha}_3 \rangle_0 - (\rho_1\rho_2 - \rho_3\rho_4) \qquad (3.C.6)$$

$$\alpha_4 = \langle \hat{\alpha}_4 \rangle_0 - (\rho_2\rho_4 - \rho_1\rho_3) \ .$$

We can now express $\langle H \rangle_0$, where H was given in terms of the C_k, d_k, by using equation (3.C.3) but, this being a shorthand method, the equations are only valid if α_i is taken in the α_k, β_k representation namely (3.C.5). Examples of the shorthand method are given in appendix 4. The equations become:

1) Holstein-Primakoff

$$\langle H_1 \rangle_0 = -NzS^2 \left[(1+\alpha_1)^2 - \alpha_3^2 - \alpha_4^2 + \frac{1}{45}\alpha_2(\alpha_1 - \alpha_2) \right.$$
$$\left. - \frac{(\alpha_1 - \alpha_2)}{4}(\alpha_2^2 + 5\alpha_3^2 - (\alpha_1 - \alpha_2)^2 + \alpha_4^2) \right]$$

$$\langle V_1 \rangle_0 = -SN\alpha_3 \qquad (3.C.7)$$

$$\langle V_2 \rangle_0 = \frac{(SN)}{2}(\rho_1 + \rho_2)\left[1 + (1 + \frac{1}{8S})\frac{(\alpha_2 + \alpha_3)}{2} - \frac{3}{16}(\alpha_2 + \alpha_3)^2 \right]$$
$$+ \frac{(SN)}{2}(\rho_3 + \rho_4)\left[1 + (1 + \frac{1}{8S})\frac{(\alpha_2 - \alpha_3)}{2} - \frac{3}{16}(\alpha_2 - \alpha_3)^2 \right]$$
$$+ O(\rho^3) .$$

$$\langle V_3\rangle_o = NS^2\left[1 + 2\left(1-\tfrac{1}{2S}\right)\alpha_2 + 2(\alpha_2^2+\alpha_3^2)\right]$$

2) Dyson-Maleev I

$$\langle H_1\rangle_o = -NzS^2[(1+\alpha_1)^2 - (\alpha_3+\alpha_4)^2]$$

$$\langle V_1\rangle_o = -SN\alpha_3 \qquad (3.C.8)$$

$$\langle V_2\rangle_o = \frac{(SN)}{2}(\rho_2+\rho_3) + \frac{(SN)}{2}\rho_1[1+(\alpha_2+\alpha_3)]$$

$$+ \frac{(SN)}{2}\rho_4[1+(\alpha_2-\alpha_3)] + \frac{(SN)}{2}(\rho_1^2\rho_2+\rho_3\rho_4^2)$$

$$\langle V_3\rangle_o = NS^2\left[1 + 2\left(1-\tfrac{1}{2S}\right)\alpha_2 + 2(\alpha_2^2+\alpha_3^2)\right]$$

3) Dyson-Maleev II

$$\langle H_1\rangle_o = -NzS^2[(1+\alpha_1)^2 - \alpha_3^2 - \alpha_4^2 - 2\alpha_2\alpha_4$$

$$+(\alpha_1-\alpha_2-\alpha_4)(\alpha_2^2+\tfrac{1}{2}(\alpha_1-\alpha_2)^2-\alpha_3^2-\tfrac{1}{2}\alpha_4^2)]$$

$$\langle V_1\rangle_o = -SN\alpha_3 \qquad (3.C.9)$$

$$\langle V_2\rangle_o = \frac{(SN)}{2}(\rho_2+\rho_4) + \frac{(SN)}{2}\rho_1[1+(\alpha_2+\alpha_3)]$$

$$+ \frac{(SN)}{2}\rho_3[1+(\alpha_2-\alpha_3)] + \frac{(SN)}{2}(\rho_1^2\rho_2+\rho_3^2\rho_4)$$

$$\langle V_3\rangle_o = NS^2\left[1 + 2\left(1-\tfrac{1}{2S}\right)\alpha_2 + 2(\alpha_2^2+\alpha_3^2)\right] .$$

D. STATIONARY POINT SOLUTION

We are interested in stationary point solutions to the free energy

$$F = F_0 + \langle H - H_0 \rangle_0 + (\text{higher order terms})$$
$$= \langle H \rangle_0 - T S_0 \qquad (3.D.1)$$

where S_0 is the entropy associated with the trial Hamiltonian. The entropy for bosons is well known (Bloch 1963) and can be written

$$S_0 = k \sum_k \left[\langle n_k \rangle_0 \ln \langle n_k \rangle_0 - (\langle n_k \rangle_0 + 1) \ln (\langle n_k \rangle_0 + 1) \right]$$
$$+ k \sum_k \left[\langle n_k' \rangle_0 \ln \langle n_k' \rangle_0 - (\langle n_k' \rangle + 1) \ln (\langle n_k' \rangle_0 + 1) \right] . \qquad (3.D.2)$$

In the previous section we have seen that $\langle H \rangle_0$ is a function of $\alpha_1, \ldots \alpha_4$. However, these are expressed in terms of α_k, β_k by equation (3.C.5) so that F is an explicit function of the eight quantities $\langle n_k \rangle_0$, $\langle n_k' \rangle_0$, Θ_k, $\Delta\Theta_k$, $\rho_1, \ldots, \rho_4$. A specification of these quantities or the original set $\varepsilon_1, \ldots, \varepsilon_4$, $\sigma_1, \ldots, \sigma_4$ completely determines a solution.

Differentiating with respect to Θ_k and $\Delta\Theta_k$ gives

$$\frac{\partial \langle H_0 \rangle_0}{\partial (\Delta\Theta_k)} = \sum_{i=1}^{4} \frac{\partial \langle H \rangle_0}{\partial \alpha_i} \frac{\partial \alpha_i}{\partial (\Delta\Theta_i)} = 0$$

$$= \left[\frac{\partial \langle H \rangle_0}{\partial \alpha_1} + \frac{\partial \langle H \rangle_0}{\partial \alpha_2} \right] \frac{\sinh(\Delta\Theta_k)}{\cosh(\Delta\Theta_k)} - \frac{\partial \langle H \rangle_0}{\partial \alpha_4} \frac{\gamma(k)}{\cosh(\Delta\Theta_k)}$$

$$\frac{\partial \langle H\rangle_o}{\partial \theta_k} = \sum_{i=1}^{4} \frac{\partial \langle H\rangle_o}{\partial \alpha_i}\frac{\partial \alpha_i}{\partial \theta_k} = 0 \qquad (3.D.3)$$

$$= \left[\frac{\partial \langle H\rangle_o}{\partial \alpha_1} + \frac{\partial \langle H\rangle_o}{\partial \alpha_2}\right]\frac{\sinh\theta_k}{\cosh(\Delta\theta_k)} - \frac{\partial \langle H\rangle_o}{\partial \alpha_3}\gamma(k)\cosh\theta_k$$

$$+ \frac{\partial \langle H\rangle_o}{\partial \alpha_4}\gamma(k)\frac{\sinh\theta_k \sinh(\Delta\theta_k)}{\cosh(\Delta\theta_k)}$$

These equations are similar to those (Ap.3.3) so that they can be satisfied by

$$\frac{\partial \langle H\rangle_o}{\partial \alpha_1} = -\frac{1}{2}A \;,\; \frac{\partial \langle H\rangle_o}{\partial \alpha_2} = -\frac{1}{2}B \;, \qquad (3.D.4)$$

$$\frac{\partial \langle H\rangle_o}{\partial \alpha_3} = -\frac{1}{2}C \;,\; \frac{\partial \langle H\rangle_o}{\partial \alpha_4} = -\frac{1}{2}D \;.$$

Differentiating F with respect to $\langle n_k\rangle_o$ and $\langle n_k'\rangle_o$ yields

$$\langle n_k\rangle_o = \frac{1}{\exp(\beta\varepsilon_k)-1} \;,\; \langle n_k'\rangle_o = \frac{1}{\exp(\beta\varepsilon_k')-1} \qquad (3.D.5)$$

where

$$\varepsilon_k = 2Jz\,S\left\{\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2} + C\right\}$$

$$\varepsilon_k' = 2Jz\,S\left\{\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2} - C\right\} .$$

The solution to these equations is obtained self-consistently. Guessing values for $\alpha_1,\ldots,\alpha_4$ and using (3.D.4) gives us values for A, B, C, D. Substituting these in (3.D.5) and using (Ap.3.4) and (3.C.6) gives new values for $\alpha_1,\ldots\alpha_4$. Repeating this procedure hopefully yields

self-consistent values for α, etc.

We have not yet determined values for $\rho_1, \ldots, \rho_4$. These are obtained by differentiating equations (3.C.5), (3.C.7), (3.C.8), and (3.C.9) with respect to ρ_1 etc. giving four equations in the four unknowns $\rho_1, \ldots, \rho_4$. This will be done in chapter 4.

E. DISCUSSION OF CHAPTER 3

We shall delay the calculation of the properties of the antiferromagnet until the next chapter. With the equations developed in this chapter one can calculate the properties of an antiferromagnet for any uniaxial anisotropy and magnetic field. The three approximations all yield slightly different results - we would like to choose the best one. Usually we can distinguish between two solutions by choosing the one with the lowest free energy. However, in the non-Hermitian approximation this no longer serves as a criterion.

We would expect that the Dyson-Maleev solutions are more accurate by recalling that for a ferromagnet cancellation (Oguchi 1960) of terms occurs for HP but not for DM. If this is true, then it is still difficult to choose between DMI and DMII . However, DMI has the fewest number of terms and therefore is easier to use.

It is somewhat encouraging that the differences between DMI and DMII are much less than one might expect from equations (3.C.8) and (3.C.9). This can be seen in

the following way: Consider zero field and zero anisotropy, then using equation (3.D.8) we obtain

1) DMI

$$A = 1+\alpha_1$$

$$B = 0$$

$$C = -(\alpha_3+\alpha_4) \qquad (3.E.1)$$

$$D = -(\alpha_3+\alpha_4)$$

2) DMII

$$A = 1+\alpha_1+\frac{1}{2}\alpha_2^2+\frac{3}{4}(\alpha_1-\alpha_2)^2-\frac{1}{2}\alpha_3^2-\frac{1}{4}\alpha_4^2-\frac{1}{2}\alpha_4(\alpha_1-\alpha_2)$$

$$B = -\frac{1}{2}\alpha_2^2+\frac{3}{4}(\alpha_1-\alpha_2)^2+\frac{1}{2}\alpha_3^2+\frac{1}{4}\alpha_4^2-\frac{1}{2}\alpha_4(\alpha_1-\alpha_2)$$

$$-\alpha_4+\alpha_2(\alpha_1-\alpha_2-\alpha_4) \qquad (3.E.2)$$

$$C = -\alpha_3[1+(\alpha_1-\alpha_2-\alpha_4)]$$

$$D = -\alpha_2-\alpha_4-\frac{1}{2}\alpha_3^2-\frac{1}{4}(\alpha_1-\alpha_2)^2+\frac{1}{2}\alpha_3^2+\frac{1}{4}\alpha_4^2-\frac{1}{2}\alpha_4(\alpha_1-\alpha_2-\alpha_4).$$

Now α_4 can be eliminated from these equations by the relation

$$\alpha_4 = -(\alpha_1-\alpha_2)\frac{D}{A} \qquad (3.E.4)$$

which follows from equations (Ap.3.4)

The properties of interest depend mainly on the dispersion function.

$$\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2} \pm C \qquad (3.E.4)$$

so that the qualities $(A+B)$, (A^2-D^2) and C are most fundamental. For each case we will determine these quantities to order α^2 (we assume α_1 , α_2 , α_3 are of the same order of smallness).

1) DMI

$$\alpha_4 = (\alpha_1-\alpha_2)\alpha_3 + O(\alpha^3)$$

$$A+B = 1+\alpha_1 \qquad (3.E.5)$$

$$A^2-D^2 = (1+\alpha_1)^2 - \alpha_3^2 + O(\alpha^3)$$

$$C = -\alpha_3\left[1+(\alpha_1-\alpha_2)\right] + O(\alpha^3)$$

2) DMII

$$\alpha_4 = (\alpha_1-\alpha_2)\alpha_2 + O(\alpha^3)$$

$$A+B = 1+\alpha_1 + O(\alpha^3) \qquad (3.E.6)$$

$$C = -\alpha_3\left[1+(\alpha_1-\alpha_2)\right] + O(\alpha^3)$$

$$A^2-D^2 = (1+\alpha_1)^2 + \frac{3}{2}(\alpha_1-\alpha_2)^2 - \alpha_3^2 + O(\alpha^3)$$

So we observe the surprising fact that the term in α^2 is the same in both cases for $(A+B)$ and for C. The quantity A^2-D^2 differs in order $O(\alpha^2)$, however, this is of higher order since it appears in conjunction with $\gamma(k)^2$ which ranges from zero to one i.e. the properties are more sensitive to $A+B$ and to C than they are to A^2-D^2.

Therefore we expect only third order differences between DMI and DMII whereas second order differences occur between HP and DM.

CHAPTER 4

CALCULATIONS, RESULTS AND DISCUSSION

A. IDEALIZED ANTIFERROMAGNET USING DMI

There is some difficulty in defining the properties of an ideal antiferromagnet in that for zero anisotropy the system is unstable. However if we determine the properties with a small amount of anisotropy and then allow that to become zero, a unique solution is the result. For calculating properties involving an external field one must let the field tend to zero before the anisotropy thus keeping the system stable at all times. We introduce small parameters θ and $\vec{h}$ so that equation (2.A.2) becomes

$$H = J H_1 + S_z J h_z V_1 + S_z J h_x V_2 - J_z \theta V_3 \qquad (4.A.1)$$

where

$$\theta = D/(Jz) \,,$$

$$\vec{h} = g\mu_B \vec{B}/(sJz) \,.$$

For convenience, we shall first consider only the solution for the DMI representation. We are interested in such properties as internal energy, specific heat, sublattice magnetization, total magnetization and spontaneous susceptibility. These properties are immediately

obtainable from the free energy by well known methods.

1) Internal energy

The internal energy U can be calculated from the free energy F by the formula

$$U = \frac{\partial}{\partial \beta}(\beta F) \qquad (4.A.2)$$

Since in a self-consistent approximation all parameters depend on β one must be careful. However choosing $\langle n_k \rangle_0, \langle n_k' \rangle_0, \theta_k, \Delta\theta_k$ as functions of β

$$U = \frac{\partial(\beta F)}{\partial \langle n_k \rangle_0} \frac{\partial \langle n_k \rangle_0}{\partial \beta} + \cdots + \frac{\partial(\beta F)}{\partial(\Delta\theta_k)} \frac{\partial(\Delta\theta_k)}{\partial \beta} \qquad (4.A.3)$$

$$+ \left. \frac{\partial(\beta F)}{\partial \beta} \right|_{\langle n_k \rangle_0} \cdots$$

The derivatives $\partial(\beta F)/\partial \langle n_k \rangle_0$ ...are zero by the stationary point condition leaving

$$U = \left. \frac{\partial(\beta F)}{\partial \beta} \right|_{\langle n_k \rangle_0} \cdots \qquad (4.A.4)$$

where the partial is taken holding all self-consistent parameters constant. Therefore

$$U = \left. \frac{\partial}{\partial \beta}\left(\beta \langle H \rangle_0 - S_0\right) \right|_{\langle n_k \rangle_0} \cdots \qquad (4.A.5)$$

$$= \langle H \rangle_0$$

as one might expect.

For zero field and zero anisotropy one obtains from (3.C.8) and (3.E.1)

$$U = - JNzS^2 \left[(1+\alpha_1)^2 - (\alpha_3+\alpha_4)^2 \right] \qquad (4.A.6)$$

and

$$A = (1+\alpha_1) \quad , \quad B = 0$$

$$C = -(\alpha_3+\alpha_4) \quad , \quad D = -(\alpha_3+\alpha_4) \, .$$

Solving for α_4 using equation (3.E.3) gives

$$C = D = - \alpha_3 (1+\alpha_1)/(1+\alpha_2) \, . \qquad (4.A.7)$$

For zero external field we expect α_3 (see subsection 2) to be zero so that the solution is

$$U = -JNzS^2 (1+\alpha_1)^2 \qquad (4.A.8)$$

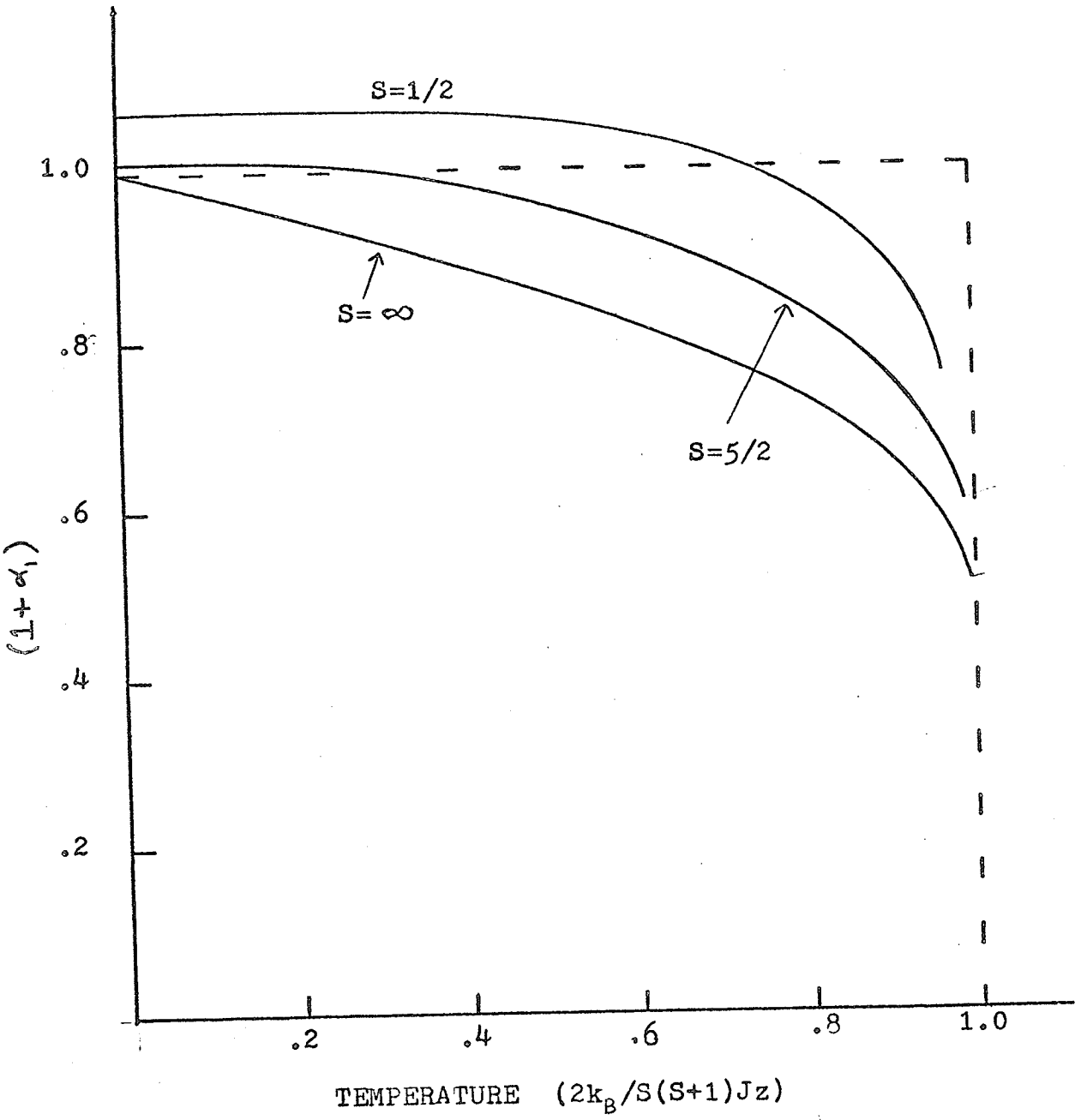
and

$$A = (1+\alpha_1) \quad , \quad B = C = D = 0 \, .$$

The solution for α_1 involves evaluating the sum in equation (Ap.3.4); a method for doing so is illustrated in part II of this thesis. The self-consistent solution for $(1+\alpha_1)$ is plotted in figure 6 for a $CsCl$ type antiferromagnet. The reduced temperature scale which

FIGURE 6

Renormalization parameter $1+\alpha_1$ of an ideal $CsCl$ type antiferromagnet as a function of temperature and spin.


S=1/2
S=∞
S=5/2
1.0
.8
.6
.4
.2
(1+α1)
.2
.4
.6
.8
1.0
TEMPERATURE (2kB/S(S+1)Jz)

was chosen by trial and error, seems to be very convenient for comparison of different spin values. Note that $(1+\alpha_1)$ is strictly decreasing with temperature—this property has already been used in section (3.E.).

2) Total magnetization in z-direction

From equation (4.A.1) with $h_x = 0$ the stationary solution becomes

$$A = (1+\alpha_1)$$

$$B = \left(1 - \frac{1}{2S}\right)\Theta + 2\Theta\alpha_2 \qquad (4.A.9)$$

$$C = \frac{h_z}{2} - \alpha_3 \frac{(1+\alpha_1)}{(1+\alpha_2)} + 2\Theta\alpha_3$$

$$D = -\alpha_3 \frac{(1+\alpha_1)}{(1+\alpha_2)}$$

where we have used equation (2.E.3)

To obtain the magnetization one differentiates the free energy with respect to B_z.

$$m_z = -\frac{\partial F}{\partial B_z} = g\mu_B S N \alpha_3 \qquad (4.A.10)$$

where by an argument similar to that leading to equation (3.A.4) all self consistent parameters can be held constant. However

$$\alpha_3 = -\frac{1}{NS}\sum_k \left(\langle \alpha_k^+ \alpha_k \rangle_0 - \langle \beta_k^+ \beta_k \rangle_0\right) \qquad (4.A.11)$$

$$= -\frac{1}{NS}\sum_k \left[\frac{1}{\exp(\beta\varepsilon_k)-1} - \frac{1}{\exp(\beta\varepsilon_k')-1}\right] .$$

For small C (see equation 3.D.5)

$$\alpha_3 = \frac{C}{2} I$$

where

$$I = \frac{8Jz}{N}\beta \sum_k \frac{e^{\beta\varepsilon_k}}{(e^{\beta\varepsilon_k}-1)^2}\Bigg|_{C=0} . \qquad (4.A.12)$$

Using (4.A.12) and (4.A.9) we can obtain a solution for α_3:

$$\alpha_3 = h_z \frac{I}{4}\left[1 + \frac{I}{2}\frac{(1+\alpha_1)}{(1+\alpha_2)} - \theta I\right]^{-1} \qquad (4.A.13)$$

and therefore

$$m_z = \frac{B_z(g\mu_B)^2 N I}{4Jz}\left[1 + \frac{I}{2}\frac{(1+\alpha_1)}{(1+\alpha_2)}\right] \qquad (4.A.14)$$

for small external field. Allowing B_z to become zero shows that the spontaneous magnetization in the z-direction is zero. This subsection also demonstrates that the parameter α_3 has a physical interpretation being proportional to m_z.

3) Sublattice magnetization

The contribution to the free energy made by introducing, in the z-direction, a field B_{1z} which interacts only with the spins of sublattice 1 is given by

$$= - g\mu_B B_{1z} \sum_{ii'} S^z_{ii'}$$

$$= - g\mu_B B_{1z} SN(1+\alpha_2+\alpha_3) . \qquad (4.A.15)$$

Differentiating

$$- \frac{\partial F}{\partial B_{1z}} = g\mu_B SN(1+\alpha_2+\alpha_3) \qquad (4.A.16)$$

and letting $B_{1z} \to 0$ (implies that $\alpha_3 = 0$ from previous section) the spontaneous sublattice magnetization becomes

$$m_{1z} = g\mu_B SN(1+\alpha_2) \qquad (4.A.17)$$

The graph of $(1+\alpha_2)$ is displayed in figure 7.

4) Parallel Susceptibility

There are two definitions of susceptibility commonly in use:

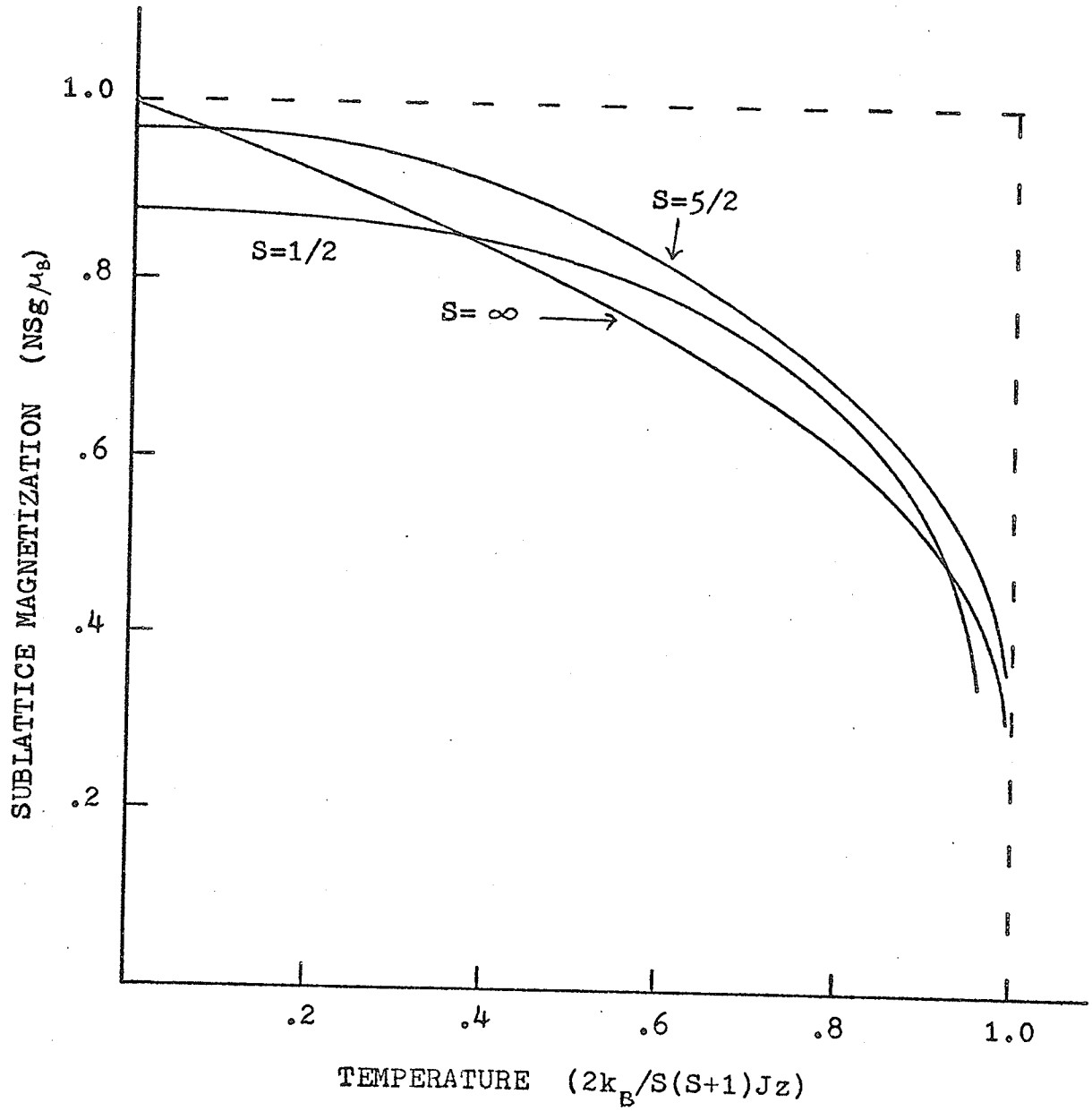
$$a) \quad \bar{\chi}_{11}(B_z) = \frac{\partial m_z}{\partial B_z} \qquad (4.A.18)$$

$$b) \quad \chi_{11}(B_z) = m_z / B_z .$$

In the limit as $B_z \to 0$, $\bar{\chi}_{11}(0) = \chi_{11}(0)$ so that the spontaneous susceptibility has a unique definition. Using definition b), we can obtain the susceptibility immediately from equation (4.A.14);

FIGURE 7

Sublattice magnetization of an ideal $CsCl$ type antiferromagnet as a function of temperature and spin.


1.0
.8
.6
.4
.2
SUBLATTICE MAGNETIZATION (NSgμ_B)
S=5/2
S=1/2
S= ∞
.2
.4
.6
.8
1.0
TEMPERATURE ($2k_B/S(S+1)Jz$)

$$\chi_{11}(0) = \frac{(g\mu_B)^2}{4Jz} NI \left[1 + \frac{I}{2}\frac{(1+\alpha_1)}{(1+\alpha_2)}\right]^{-1} \qquad (4.A.19)$$

where we have already let $\theta = 0$.

An alternate method for obtaining thermodynamic properties involves the use of correlation functions. The exact spontaneous susceptibility is given by

$$\chi_{11}(0) = (g\mu_B)^2 N^2 \int_0^\beta \langle T S_T^z(\tau) S_T^z(0)\rangle \, d\tau \qquad (4.A.20)$$

where

$$S_T^z = \frac{1}{N}\sum_i (S_{1i}^z + S_{2i}^z) = \frac{1}{N}\sum_k (c_k^+ c_k - d_k^+ d_k).$$

This is the Kubo linear response function method (Kubo 1957) and has been derived for the case where the expectation is taken using the exact Hamiltonian H_1. Liu (1966) has used the following method for calculating susceptibility, namely

$$\chi_{11}(0) = (g\mu_B)^2 N^2 \int_0^\beta \langle T S_T^z(\tau) S_T^z(0)\rangle_0 \, d\tau \qquad (4.A.21)$$

where the expectation is calculated using the approximate Hamiltonian H_0. We shall show that this method does not yield the same result as (4.A.19).

Using the fact that $[S_T^z, H_1] = 0$ we get

$$\chi_{11}(0) = (g\mu_B)^2 \beta N^2 \langle S_T^z S_T^z\rangle_0 \qquad (4.A.22)$$

$$= (g\mu_B)^2 \beta \sum_{kk'} \langle (c_k^+ c_k - d_k^+ d_k)(c_{k'}^+ c_{k'} - d_{k'}^+ d_{k'}) \rangle_0 .$$

The best solution for the unperturbed Hamiltonian has $\mathcal{B} = C = D = 0$ so that $n_k = n_{k'}$. Rewriting in normal order one can decouple without further error to find

$$\chi_{\parallel}(0) = (g\mu_B)^2 \beta \sum_k \big(2\langle c_k^+ c_k \rangle_0 \langle d_k^+ d_k \rangle_0 - 2\langle c_k^+ d_k^+ \rangle_0 \langle c_k d_k \rangle_0 + \langle c_k^+ c_k \rangle_0 + \langle d_k^+ d_k \rangle_0 \big) \tag{4.A.23}$$

where

$$\langle c_k^+ c_k \rangle_0 = \langle d_k^+ d_k \rangle_0 = \frac{1}{2} \frac{(2n_k + 1)}{[1 - \gamma(k)^2]^{1/2}} - \frac{1}{2} ,$$

$$\langle c_k^+ d_k^+ \rangle_0 = \langle c_k d_k \rangle_0 = \frac{1}{2} \frac{(2n_k + 1)\gamma(k)}{[1 - \gamma(k)^2]^{1/2}} ,$$

$$n_k = 1 / (e^{\beta \varepsilon_k} - 1) .$$

Therefore

$$\chi_{\parallel}(0) = 2(g\mu_B)^2 \beta \sum_k (n_k^2 + n_k) = 2(g\mu_B)^2 \beta \sum_k \frac{e^{\beta \varepsilon_k}}{(e^{\beta \varepsilon_k} - 1)^2} \tag{4.A.24}$$

and using definition (3.A.12)

$$\chi_{\parallel}(0) = \frac{(g\mu_B)^2 N \quad I}{4JZ} \tag{4.A.25}$$

which is only part of (4.A.19).

This illustrates an important reason for using the free energy as a method for obtaining thermodynamic quantities. Certainly, within the restrictions of the approximate Hamiltonian H_o, if one optimizes the free energy and obtains all properties directly from it one is assured of computing the correct solution. There seems to be no justification for using (4.A.21) instead of (4.A.20) and it is now clear that this does not always yield the best solution.

The result (4.A.19) which is our estimate of $\chi_{\perp}$ and Liu's result are graphed in figure 8. No significance is attached to the tail on the $S=\frac{1}{2}$ result since our approximation is considered poor in this region.

5) Perpendicular susceptibility

Calculating the perpendicular susceptibility involves finding stationary points, with respect to variations of $\rho_1, \ldots, \rho_4$ to the free energy. Placing $h_z = 0$ in equation (4.A.1) we obtain

$$\langle H \rangle_o = -JzNS^2 \left[(1+\alpha_1)^2 - (\alpha_3+\alpha_4)^2 \right.$$

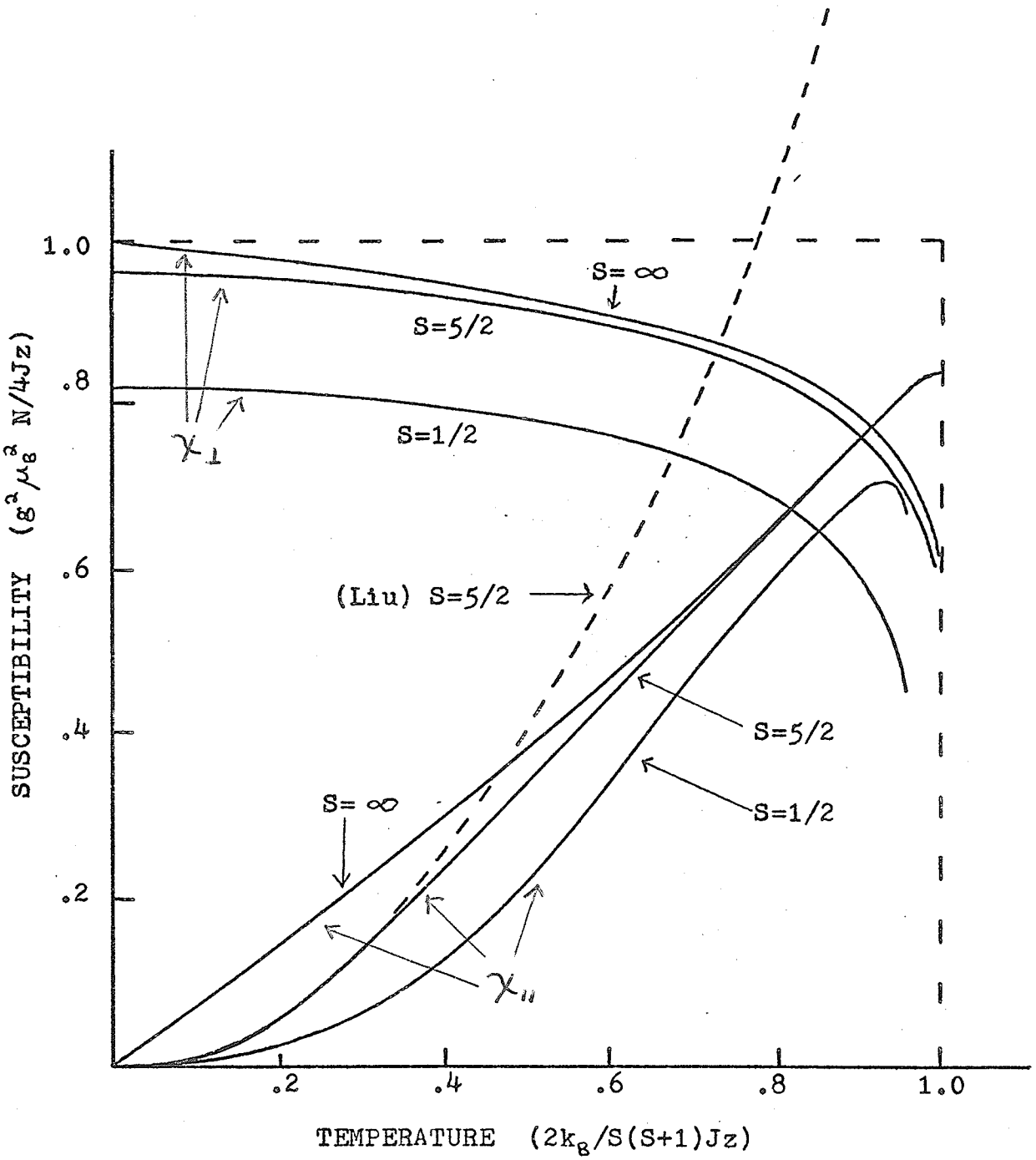
$$+ \theta + 2\theta\left(1-\tfrac{1}{2S}\right)\alpha_2 + 2\theta(\alpha_2^2+\alpha_3^2)$$

$$- \frac{h_x}{2}(\rho_1+\rho_2+\rho_3+\rho_4) \qquad (4.A.26)$$

$$\left. - \frac{h_x}{2}(\rho_1+\rho_4)\alpha_2 - \frac{h_x}{2}(\rho_1-\rho_4)\alpha_3 \right]$$

FIGURE 8

Susceptibilities of ideal CsCl type antiferromagnet in the DMI approximation as a function of temperature and spin.


1.0
.8
.6
.4
.2
SUSCEPTIBILITY ($g^2\mu_B^2$ N/4Jz)
S= ∞
S=5/2
S=1/2
$\chi_\perp$
(Liu) S=5/2
S=5/2
S= ∞
S=1/2
$\chi_{\parallel}$
.2
.4
.6
.8
1.0
TEMPERATURE ($2k_B$/S(S+1)Jz)

The entropy S_0 does not depend on $\rho_1, \ldots, \rho_4$; however, $\alpha_1, \ldots, \alpha_4$ have an explicit dependence as seen by equations (3.C.6). That is

$$\frac{\partial \alpha_1}{\partial \rho_1} = -(\rho_2 + \rho_3) \qquad (3.A.27)$$

$$\vdots$$

$$etc.$$

We can simplify the calculations by discarding all terms in high orders of h_x since eventually we will take the limit as $h_x \to 0$. Note, a priori, that ρ_i is of order h_x whereas α_1 and α_2 are of order h_x^0. We can determine the order of α_3 by calculating C

$$C = -\frac{1}{2}\frac{\partial F}{\partial \alpha_3} \qquad (4.A.28)$$

$$= -(\alpha_3 + \alpha_4) + 2\theta\alpha_3 - \frac{h_x}{4}(\rho_1 - \rho_4)$$

and using (4.A.12)

$$\alpha_3 = -\frac{h_x}{8}(\rho_1 - \rho_4) I \left[1 + \frac{I}{2}\frac{(1+\alpha_1)}{1+\alpha_2} - \theta I\right]^{-1} \qquad (4.A.29)$$

so that α_3 (and α_4) are of order h_x^2.

Differentiating F and retaining only lowest order terms produces the following four equations:

$$2(1+\alpha_1)(\rho_2+\rho_3) + 2\theta\left(1 - \frac{1}{2S}\right)\rho_2 + 4\theta\alpha_2\rho_2 + \frac{h_x}{2}(1+\alpha_2) = 0$$

$$2(1+\alpha_1)(\rho_2+\rho_3) + 2\theta\left(1 - \frac{1}{2S}\right)\rho_3 + 4\theta\alpha_2\rho_3 + \frac{h_x}{2}(1+\alpha_2) = 0$$

$$2(1+\alpha_1)(\rho_1+\rho_4)+2\theta(1-\tfrac{1}{2S})\rho_1+4\theta\alpha_2\rho_1+\frac{h_x}{2}=0 \qquad (4.A.30)$$

$$2(1+\alpha_1)(\rho_1+\rho_4)+2\theta(1-\tfrac{1}{2S})\rho_4+4\theta\alpha_2\rho_4+\frac{h_x}{2}=0$$

which have the solution

$$\rho_2=\rho_3=-\frac{h_x}{8}\,\frac{(1+\alpha_2)}{(1+\alpha_1)+\theta(1-\frac{1}{2S}+2\alpha_2)/2} \qquad (4.A.31)$$

$$\rho_1=\rho_4=-\frac{h_x}{8}\,\frac{1}{(1+\alpha_1)+\theta(1-\frac{1}{2S}+2\alpha_2)/2}$$

From these equations we see that ρ_i is directly related to the transverse field h_x; for zero field in the x direction we do not need to consider linear boson terms in the approximate Hamiltonian. Note that if $\theta=0$ we cannot obtain a solution for ρ_i since the set of equations (4.A.30) is indeterminate; however, in the limit $\theta\to 0$ one obtains a unique solution.

To obtain the susceptibility one uses

$$\chi_\perp(0)=\lim_{B_x\to 0}\left(-\frac{1}{B_x}\frac{\partial F}{\partial B_x}\right)=-\frac{(g\mu_B)^2N}{Jz}\lim_{h_x\to 0}\left(\frac{1}{h_x}\frac{\partial F}{\partial h_x}\right)$$

$$=\frac{(g\mu_B)^2N}{Jz}\,\frac{1+\alpha_2}{4(1+\alpha_1)+2\theta(1-\frac{1}{2S}+2\alpha_2)} \qquad (4.A.32)$$

Now letting $\theta=0$ gives

$$\chi_\perp(0)=\frac{(g\mu_B)^2N}{4Jz}\,\frac{(1+\alpha_2)}{(1+\alpha_1)} \qquad (4.A.33)$$

This result is plotted in figure 8.

6) Specific heat

The definition of specific heat is the following

$$C_v = \partial U / \partial T \qquad (4.A.34)$$

Using equation (4.A.8)

$$C_v = -2 J_z S^2 N (1+\alpha_1) \partial \alpha_1 / \partial T \qquad (4.A.35)$$

since α_1 is a complicated self-consistent function of T it seems best to obtain $\partial \alpha_1 / \partial T$ by numerical differentiation or from the slope of figure 6. This requires a rather accurate calculation of α_1 such as the one carried out in part II.

7) Summary and discussion

First a summary of the properties obtained in this section:

Internal energy

$$U = -JNzS^2 (1+\alpha_1)^2 \qquad (4.A.8)$$

Total magnetization

$$M_z = 0 \qquad (4.A.14)$$

Sublattice magnetization

$$M_{1z} = g\mu_B SN (1+\alpha_2) \qquad (4.A.17)$$

Parallel susceptibility

$$\chi_{\parallel} = \frac{(g\mu_B)^2}{4Jz} NI \left[1 + \frac{I}{2} \frac{(1+\alpha_1)}{(1+\alpha_2)}\right]^{-1} \qquad (4.A.19)$$

Perpendicular susceptibility

$$\chi_{\perp} = \frac{(g\mu_B)^2}{4Jz} N \frac{(1+\alpha_2)}{(1+\alpha_1)} \qquad (4.A.32)$$

Specific heat

$$C_v = -2JzS^2N(1+\alpha_1)\,\partial\alpha_1/\partial T \qquad (4.A.35)$$

where

$$\alpha_1 = -\frac{1}{SN}\sum_k \left[(1-\gamma(k)^2)^{1/2}\coth\left(\frac{\beta\varepsilon_k}{2}\right) - 1\right]$$

$$\alpha_2 = -\frac{1}{SN}\sum_k \left[(1-\gamma(k)^2)^{-1/2}\coth\left(\frac{\beta\varepsilon_k}{2}\right) - 1\right] \qquad (4.A.36)$$

$$I = 2Jz\beta\sum_k \frac{1}{\sinh^2(\beta\varepsilon_k/2)}$$

and

$$\varepsilon_k = 2JzS(1+\alpha_1)(1-\gamma(k)^2)^{1/2}. \qquad (4.A.37)$$

This model has been solved before by Liu (1966). His method involved the use of Green functions and he obtained the same dispersion relation (4.A.37) and he also obtained equations (4.A.36). He however did not consider terms of the form α_3 and α_4 and as we have seen,

both of these terms contribute to the parallel susceptibility. His derivation of $\chi_{\parallel}$ is discussed in subsection 4 earlier in this section. All other results are the same - it is somewhat surprising that the method used here for calculating the perpendicular susceptibility does not lead to a new result.

The low temperature expansions of α_1 , α_2 agree with those obtained by Oguchi (1960) and Liu to order $1/S^2$. Our formula for $\chi_{\parallel}$ (4.A.19) agrees with Oguchi but not with Liu who incorrectly claimed agreement with Oguchi. $\chi_{\perp}$ calculated here however, agrees with Liu and not Oguchi - the $\frac{1}{S^2}$ terms in the perpendicular susceptibility seem to depend on the representation used.

The method of calculation in figures 6,7, and 8 proceeds as follows. One chooses a value for α_1 say 0 and then calculates a new value using equation (4.A.36). This procedure is repeated until convergence is obtained. For reduced temperatures above .8 this method does not lead to convergence. In this region it is necessary to numerically calculate a derivative of the α_1 function and this method (called Newton's method) works very well and leads to fast convergence.

B. IDEAL ANTIFERROMAGNET - HP AND DMII

1) Holstein-Primakoff

The solution to an ideal antiferromagnet using HP

representation is straight-forward; the solutions are obtained in the same manner as for DMI.

Internal energy

$$U = -JNzS^2 \left[(1+\alpha_1)^2 + \frac{1}{4S}\alpha_2(\alpha_1-\alpha_2) - \frac{1}{4}\alpha_2^2(\alpha_1-\alpha_2) + \frac{1}{4}(\alpha_1-\alpha_2)^3 \right] \tag{4.B.1}$$

Total magnetization

$$m_z = 0 \tag{4.B.2}$$

Sublattice magnetization

$$m_{1z} = g\mu_B SN(1+\alpha_2) \tag{4.B.3}$$

Parallel susceptibility

$$\chi_{\parallel} = \frac{(g\mu_B)^2}{4Jz} NI \left[1 + \frac{I}{2}\left(1 + \frac{5}{4}(\alpha_1-\alpha_2)\right) \right]^{-1} \tag{4.B.4}$$

Perpendicular susceptibility

$$\chi_{\perp} = \frac{(g\mu_B)^2}{4Jz} N \frac{\left[1 + (1+\frac{1}{8S})\frac{\alpha_2}{2} - \frac{3}{16}\alpha_2^2\right]}{\left[1 + \alpha_1 + \frac{1}{16S}\alpha_1 + \frac{1}{4}(\alpha_1-\alpha_2)^2 - \frac{1}{16}\alpha_1^2\right]} \tag{4.B.5}$$

where

$$\alpha_1 = -\frac{1}{SN}\sum_k \left[\frac{(A+B) - A\gamma(k)^2}{\left[(A+B)^2 - A^2\gamma(k)^2\right]^{1/2}} \coth\left(\frac{\beta\varepsilon_k}{2}\right) - 1 \right]$$

$$\alpha_2 = -\frac{1}{SN}\sum_k \left[\frac{(A+B)}{[(A+B)^2 - A^2\gamma(k)^2]^{1/2}}\coth\left(\frac{\beta\varepsilon_k}{2}\right) - 1\right]$$

$$I = \frac{2Jz}{N}\beta\sum_k \frac{1}{\sinh^2\left(\frac{\beta\varepsilon_k}{2}\right)} \qquad (4.B.6)$$

$$\varepsilon_k = 2JzS\,[(A+B)^2 - A^2\gamma(k)^2]^{1/2}$$

and

$$A = (1+\alpha_1) + \frac{1}{8S}\alpha_2 - \frac{1}{8}\alpha_2^2 + \frac{3}{8}(\alpha_1-\alpha_2)^2$$

$$B = \frac{1}{8S}(\alpha_1 - 2\alpha_2) + \frac{1}{8}\alpha_2^2 - \frac{3}{8}(\alpha_1-\alpha_2)^2 - \frac{1}{4}\alpha_2(\alpha_1-\alpha_2)\,. \qquad (4.B.7)$$

2) Dyson-Maleev II

Unfortunately, the solution to an ideal antiferromagnet using the DMII representation does not exist. This can be seen by recalling the condition required for the approximate Hamiltonian to have a lower bound i.e.

$$(A+B)^2 - A^2 + D^2 - C^2 \geq 0\,. \qquad (4.B.8)$$

The solutions for DMII with finite anisotropy and zero field are from (3.E.2) and (4.A.1)

$$A = 1 + \alpha_1 + \frac{1}{2}\alpha_2^2 + \frac{3}{4}(\alpha_1-\alpha_2)^2 - \frac{1}{2}\alpha_4(\alpha_1-\alpha_2) - \frac{1}{4}\alpha_4^2$$

$$C = 0$$

$$B = -\tfrac{1}{2}\alpha_2^2 - \tfrac{3}{4}(\alpha_1-\alpha_2)^2 + \tfrac{1}{4}\alpha_4^2 + \tfrac{1}{2}\alpha_4(\alpha_1-\alpha_2)$$

$$- \alpha_4 + \alpha_2(\alpha_1-\alpha_2-\alpha_4) + (1-\tfrac{1}{2S}+2\alpha_2)\Theta \qquad (4.B.9)$$

$$D = -\alpha_2 - \alpha_4 - \tfrac{1}{2}\alpha_2^2 - \tfrac{1}{4}(\alpha_1-\alpha_2)^2 + \tfrac{1}{4}\alpha_4^2 - \tfrac{1}{2}\alpha_4(\alpha_1-\alpha_2-\alpha_4)$$

$$\alpha_4 = -(\alpha_1-\alpha_2)D/A$$

Neglecting terms in Θ^2 for small Θ, we find the condition:

$$\Theta \geq \frac{3}{4}\,\frac{(\alpha_1-\alpha_2)^2}{(1-\frac{1}{2S})} + O(\alpha^3) \quad ; \qquad (4.B.10)$$

the condition for a lower bound is not established. At first this seems like a prohibitive condition; however, the magnitude of Θ predicted by equation (4.B.10) is very small ($\Theta = 10^{-4}$ for $T=0$, $S=\frac{5}{2}$). This amount of anisotropy is probably less than the error incurred in the approximations required by our treatment.

One can easily circumvent this problem by replacing a small amount of anisotropy on the system (Note that for $S=\frac{1}{2}$ this is not possible since $(S^z)^2$ =constant, however, we will not concern ourselves with that problem here). This will be done in the following section.

C. COMPARISON OF HP, DMI AND DMII

As we have seen, in order to have a solution to the

DMII system it is necessary to assume some value for Θ. We will therefore choose $\Theta = .02$ and compute the properties of the three solutions using this value of Θ. To insure that there is no confusion as to what is computed, we will give the form of the solutions again.

For all three cases we have

$$\alpha_1 = -\frac{1}{SN}\sum_k \left\{ (\langle n_k\rangle_0 + \langle n_k'\rangle_0 + 1)\,\frac{(A+B) - A\gamma(k)^2}{[(A+B)^2 - (A^2-D^2)\gamma(k)^2]^{1/2}} - 1 \right\}$$

$$- (\rho_1\rho_2 + \rho_3\rho_4 + \rho_2\rho_4 + \rho_1\rho_3)$$

$$\alpha_2 = -\frac{1}{SN}\sum_k \left\{ (\langle n_k\rangle_0 + \langle n_k'\rangle_0 + 1)\,\frac{(A+B)}{[(A+B)^2 - (A^2-D^2)\gamma(k)^2]^{1/2}} - 1 \right\}$$

$$- (\rho_1\rho_2 + \rho_3\rho_4) \qquad (4.C.1)$$

$$\alpha_3 = -\frac{1}{SN}\sum_k (\langle n_k\rangle_0 - \langle n_k'\rangle_0) - (\rho_1\rho_2 - \rho_3\rho_4)$$

$$\alpha_4 = -\frac{1}{SN}\sum_k \left\{ (\langle n_k\rangle_0 + \langle n_k'\rangle_0 + 1)\,\frac{D\gamma(k)^2}{[(A+B)^2 - (A^2-D^2)\gamma(k)^2]^{1/2}} \right\}$$

$$- (\rho_2\rho_4 - \rho_1\rho_3)$$

$$\langle n_k\rangle_0 = \frac{1}{e^{\beta\varepsilon_k} - 1} \quad , \quad \langle n_k'\rangle_0 = \frac{1}{e^{\beta\varepsilon_k'} - 1}$$

$$\varepsilon_k = 2JzS\left\{[(A+B)^2 - (A^2-D^2)\gamma(k)^2]^{1/2} + C\right\}$$

$$\varepsilon_k' = 2JzS\left\{[(A+B)^2 - (A^2-D^2)\gamma(k)^2]^{1/2} - C\right\}$$

For each case we have:

Note These equations are not the most general; they are valid only if one of h_x and h_z is zero (i.e. ρ_i takes on a different form if $h_z \neq 0$) but usually we need only one of them.

1) Holstein-Primakoff

$$A = (1+\alpha_1) + \frac{1}{8S}\alpha_2 - \frac{1}{8}\alpha_2^2 + \frac{3}{8}(\alpha_1-\alpha_2)^2 - \frac{5}{8}\alpha_3^2 - \frac{1}{8}\alpha_4^2$$

$$B = \frac{1}{8S}(\alpha_1 - 2\alpha_2) + \frac{1}{8}\alpha_2^2 - \frac{3}{8}(\alpha_1-\alpha_2)^2 - \frac{1}{4}\alpha_2(\alpha_1-\alpha_2)$$

$$+ \frac{5}{8}\alpha_3^2 + \frac{1}{8}\alpha_4^2 + \Theta\left(1 - \frac{1}{2S} + 2\alpha_2\right)$$

$$- \frac{h_x \rho}{2}\left(1 + \frac{1}{8S} - \frac{3}{4}\alpha_2\right) \qquad (4.C.2)$$

$$C = h_z/2 - \alpha_3\left[1 + \frac{5}{4}(\alpha_1-\alpha_2) + 2\Theta - \frac{3}{8}\rho h_x\right]$$

$$D = -\alpha_4\left[1 + \frac{(\alpha_1-\alpha_2)}{4}\right]$$

$$\rho = \rho_1 = \rho_2 = \rho_3 = \rho_4$$

$$= -\frac{h_x}{8}\,\frac{1 + (1+\frac{1}{8S})\alpha_2/2 - \frac{3}{16}\alpha_2^2}{1 + \alpha_1 + \frac{1}{16S}\alpha_1 + \frac{1}{4}(\alpha_1-\alpha_2)^2 - \frac{1}{16}\alpha_1^2 + \frac{\Theta}{2}\left(1 - \frac{1}{2S} + 2\alpha_2\right)}$$

2) Dyson-Maleev I

$$A = (1+\alpha_1)$$

$$B = \Theta\left(1 - \frac{1}{2S} + 2\alpha_2\right) - \frac{\rho_1 h_x}{2}$$

$$C = \frac{h_z}{2} - \alpha_3\left(\frac{1+\alpha_1}{1+\alpha_2} + 2\Theta\right)$$

$$D = -\alpha_3 (1+\alpha_1)/(1+\alpha_2) \qquad (4.C.3)$$

$$\rho_1 = \rho_4 = -\frac{h_x}{8} \frac{1}{(1+\alpha_1) + \frac{\Theta}{2}(1 - \frac{1}{2S} + 2\alpha_2)}$$

$$\rho_2 = \rho_3 = -\frac{h_x}{8} \frac{(1+\alpha_2)}{(1+\alpha_1) + \frac{\Theta}{2}(1 - \frac{1}{2S} + 2\alpha_2)}$$

3) Dyson-Maleev II

$$A = (1+\alpha_1) + \frac{1}{2}\alpha_2^2 + \frac{3}{4}(\alpha_1 - \alpha_2)^2 - \frac{1}{2}\alpha_4(\alpha_1 - \alpha_2) - \frac{1}{4}\alpha_2^2$$

$$B = -\frac{1}{2}\alpha_2^2 - \frac{3}{4}(\alpha_1 - \alpha_2)^2 + \frac{1}{4}\alpha_4^2 + \frac{1}{2}\alpha_4(\alpha_1 - \alpha_2)$$

$$- \alpha_4 + \alpha_2(\alpha_1 - \alpha_2 - \alpha_4) + \Theta(1 - \frac{1}{2S} + 2\alpha_2) - \rho_1 h_x/2$$

$$C = \frac{h_z}{2} - \alpha_3(1 + \alpha_1 - \alpha_2 - \alpha_4 + 2\Theta) \qquad (4.C.4)$$

$$D = -\alpha_2 - \alpha_4 - \frac{1}{2}\alpha_2^2 - \frac{1}{4}(\alpha_1 - \alpha_2)^2 + \frac{1}{2}\alpha_3^2 + \frac{1}{4}\alpha_4^2$$

$$- \frac{1}{2}\alpha_4(\alpha_1 - \alpha_2 - \alpha_4)$$

$$\rho_1 = \rho_3 = -\frac{h_x}{8} \frac{1}{1 + \alpha_1 + \Theta(1 - \frac{1}{2S} + 2\alpha_2)/2} + \frac{1}{4}\frac{(\alpha_1 - \alpha_2)^2}{\Theta(1 - \frac{1}{2S})}$$

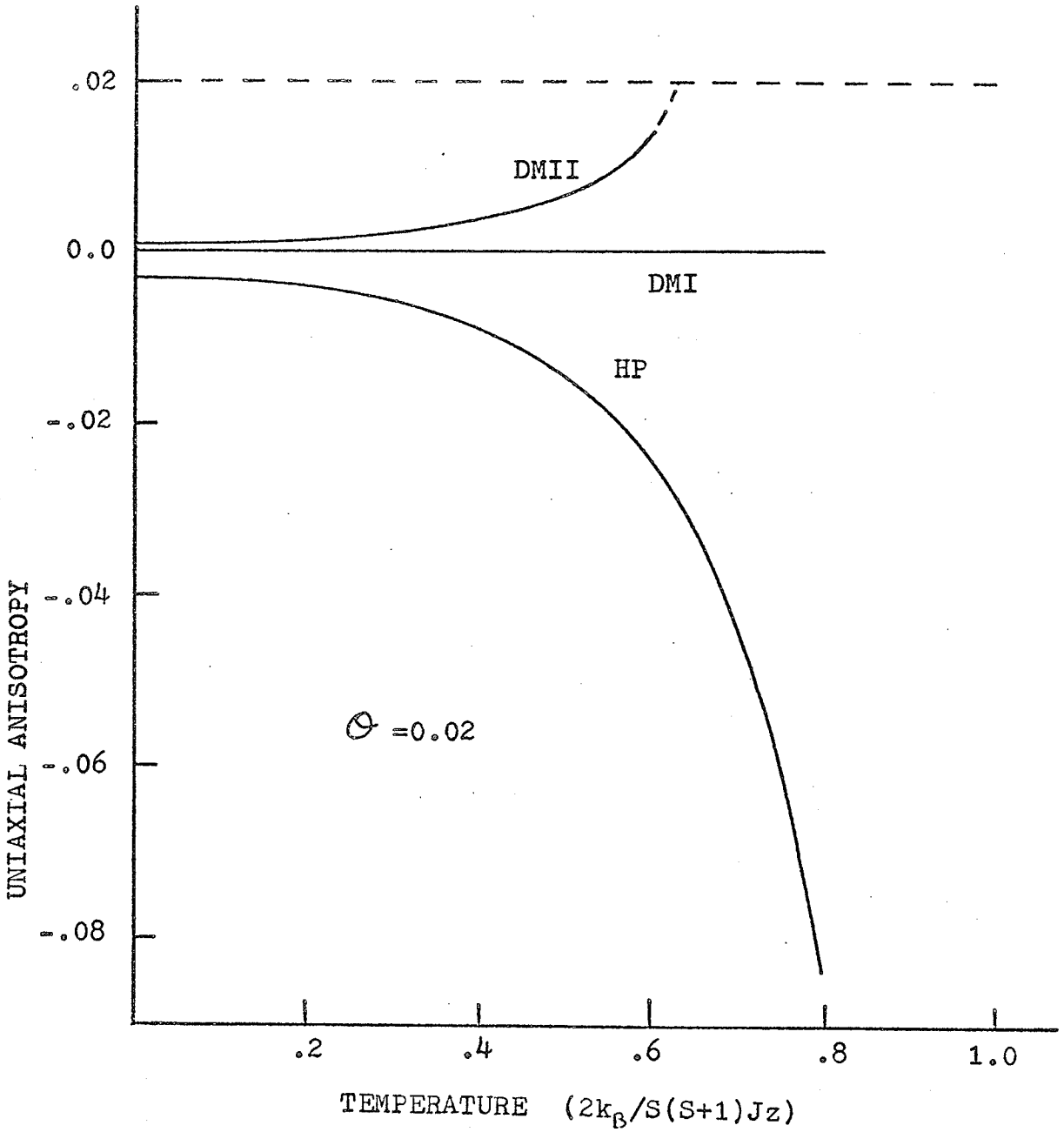
$$\rho_2 = \rho_4 = -\frac{h_x}{8} \frac{1 + \alpha_2}{1 + \alpha_1 + \Theta(1 - \frac{1}{2S} + 2\alpha_2)/2} + \frac{1}{4}\frac{(\alpha_1 - \alpha_2)^2}{\Theta(1 - \frac{1}{2S})}$$

The method of calculation proceeds in an iterative fashion. Choosing an initial value for $\alpha_1, \ldots \alpha_4$ say 0 we can determine the parameters $\rho_1, \ldots, \rho_4$ and then $A, \ldots, D$. Using these gives new values for $\alpha_1, \ldots, \alpha_4$. This procedure is repeated until the desired convergence is obtained. We have already noted in the ideal DMI case that at some value, simple iteration no longer converges. We have circumvented this problem for the ideal case but we will not concern ourselves with this problem here. Although we could find the solution for higher temperatures, the effort does not seem justified.

The first graph, figure 9, where we have taken $h_x = h_z = 0$ and $S = 5/2$ is a plot of the stability of each solution. For each temperature, we have decreased the anisotropy from .02 until the solution disappears. This represents the minimum amount of uniaxial anisotropy required to stabilize the system. Looking at it from the opposite point of view, it represents the amount of (negative) effective anisotropy predicted by the model i.e. HP is very stable compared to DMII. The graph essentially confirms the point brought out in section (3.E); at low temperatures the difference between the DMI and DMII is smaller than between DM and HP. As was discussed in section (4.B) the amounts of effective anisotropy predicted by these models is very small and is probably less than the accuracy of the results. This also shows that there is an inherent uncertainty due to the dynamical interaction

FIGURE 9

Amount of uniaxial anisotropy required to stabilize a spin 5/2, $CsCl$ antiferromagnet as predicted by DMI, DMII and HP.


.02
0.0
-.02
-.04
-.06
-.08
DMII
DMI
HP
Θ =0.02
UNIAXIAL ANISOTROPY
.2
.4
.6
.8
1.0
TEMPERATURE (2k$_B$/S(S+1)Jz)

and even if one knew exactly the anisotropy of a real system, dynamical effects would still cloud the issue. We see that the DMII solution becomes unstable at a reduced temperature of about $.625\,(\pm.25)$ so that although at low temperature DMII has reasonable results, it does not adequately extend to higher temperatures. Finally the strong stability of HP is an unreasonable result since it seems unlikely that the dynamical interaction can lead to an effective stability (this however is uncertain).

In figures 10-13, the properties of the antiferromagnet are compared for $S = 5/2$.

1) Internal energy

All three models predict essentially the same internal energy. The three formulae used are

HP

$$U = -NJz S^2 \left[(1+\alpha_1)^2 - \alpha_3^2 - \alpha_4^2 + \frac{1}{4S}\alpha_2(\alpha_1-\alpha_2) \right.$$
$$\left. - \frac{(\alpha_1-\alpha_2)}{4}\left(\alpha_2^2 + 5\alpha_3^2 - (\alpha_1-\alpha_2)^2 + \alpha_4^2\right)\right]$$
$$+ \Theta\left[1 + 2\left(1-\frac{1}{2S}\right)\alpha_2 + 2(\alpha_2^2+\alpha_3^2)\right]$$

DMI

$$U = -NJz S^2 \left[(1+\alpha_1)^2 - (\alpha_3+\alpha_4)^2\right] \qquad (4.C.5)$$
$$+\Theta\left[1 + 2\left(1-\frac{1}{2S}\right)\alpha_2 + 2(\alpha_2^2+\alpha_3^2)\right]$$

DMII

$$U = -NJ_2S^2\left[(1+\alpha_1)^2-\alpha_3^2-\alpha_4^2-2\alpha_2\alpha_4\right.$$

$$+(\alpha_1-\alpha_2-\alpha_4)\left(\alpha_2^2+\tfrac{1}{2}(\alpha_1-\alpha_2)^2-\alpha_3^2-\tfrac{1}{2}\alpha_4^2\right)$$

$$\left.+\Theta\left[1+2\left(1-\tfrac{1}{2S}\right)\alpha_2+2(\alpha_2^2+\alpha_3^2)\right]\right]$$

with $h_x = h_z = 0$. These are plotted in figure 10.

For HP we see that the dispersion relation is different than DMI and apparently this tends to compensate for the additional terms in the expression above. For DMII which has much the same dispersion relation (as DMI), the term involving α_4 tends to compensate for the extra terms. This leaves a similar energy curve in all three cases.

2) Sublattice magnetization

For this case we plot using $h_x = h_z = 0$ the quantity

$$M_{1z} = g\mu_B NS(1+\alpha_2) \qquad (4.C.6)$$

which is valid for HP, DMI and DMII. The sublattice magnetization plotted in figure II seems to vary much more with representation than the internal energy. This reflects the fact that the sublattice magnetization depends more strongly on the effective anisotropy.

FIGURE 10

Internal energy of a spin 5/2, $CsCl$ type antiferromagnet as predicted by HP, DMI and DMII.

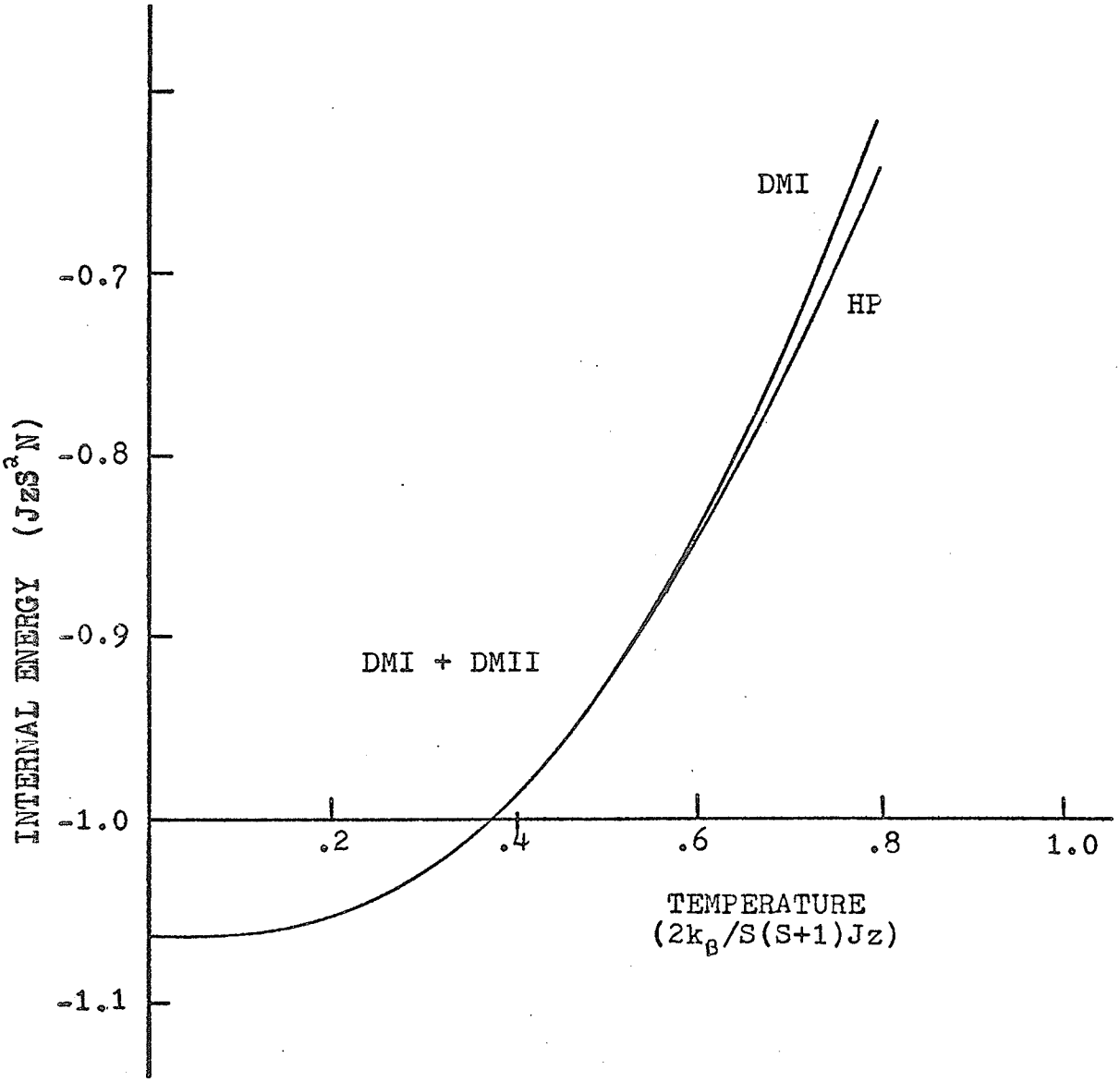
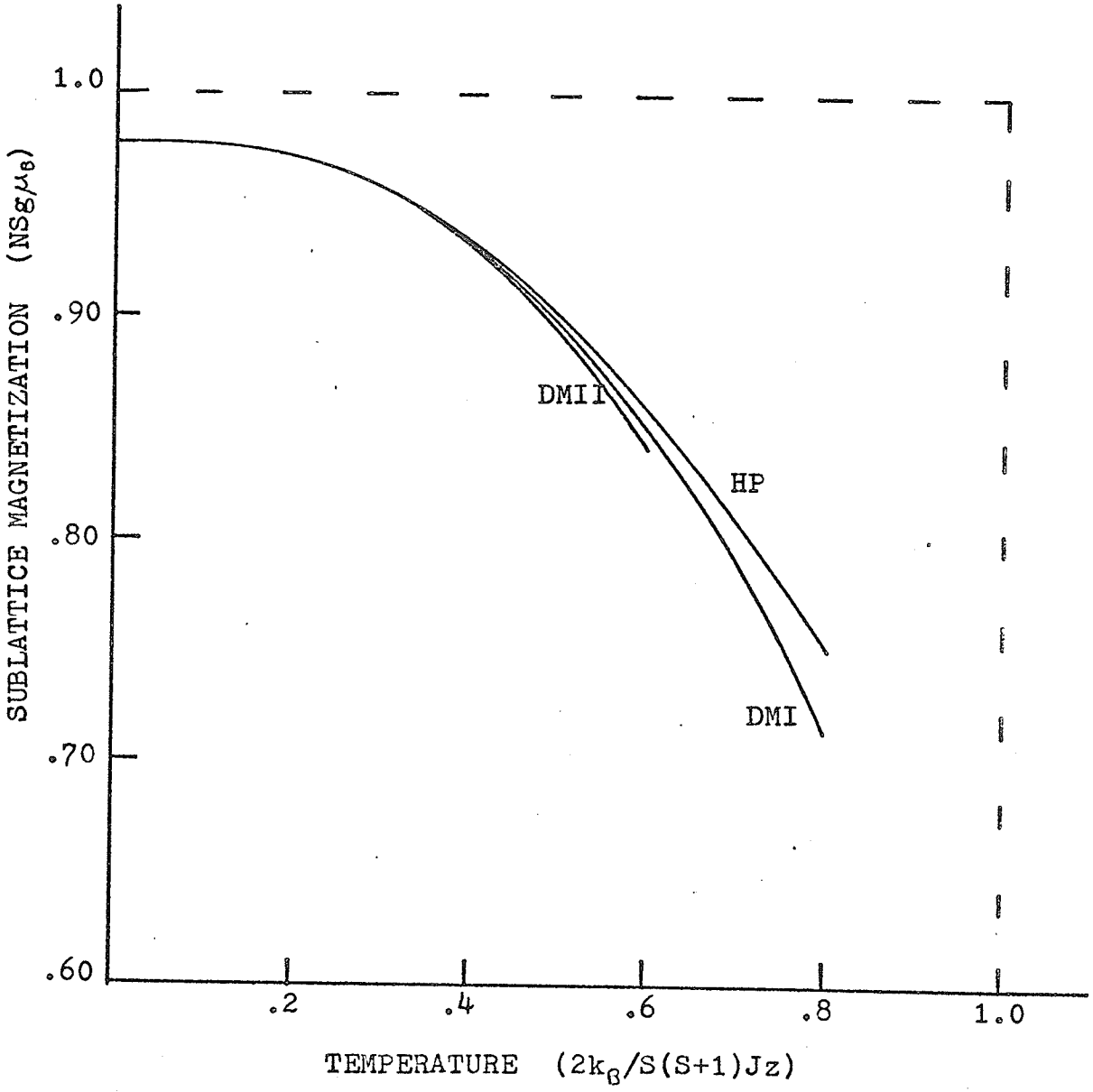

DMI
HP
DMI + DMII
INTERNAL ENERGY (JzS²N)
-0.7
-0.8
-0.9
-1.0
-1.1
.2
.4
.6
.8
1.0
TEMPERATURE
(2k_B/S(S+1)Jz)

FIGURE 11

Sublattice magnetization of a spin 5/2, CsCl type antiferromagnet as predicted by HP, DMI and DMII.


1.0
.90
.80
.70
.60
SUBLATTICE MAGNETIZATION (NSgμ_B)
DMII
HP
DMI
.2
.4
.6
.8
1.0
TEMPERATURE ($2k_B/S(S+1)Jz$)

3) Parallel susceptibility

Placing $h_x = 0$ and $h_z = .0196$ (this corresponds to a 20,000 gauss field) we have solved the same equations. In each case we will choose definition (3.A.18b) and calculate

$$\chi_{11} = \frac{1}{2} M_z / B_z = (g\mu_B)^2 N S \alpha_3 / (2SJz) . \quad (4.C.7)$$

This is not the differential susceptibility but the magnetization per field for a small field.

These results are plotted in figure 12 and have been divided by temperature squared. Along with these results is the zero anisotropy result (zero field) predicted by DMI. This gives us some idea of the anisotropy dependence of χ_{11}.

4) Perpendicular susceptibility

Solving the equations with $h_x = h_z = 0$ and using the following formulae

HP

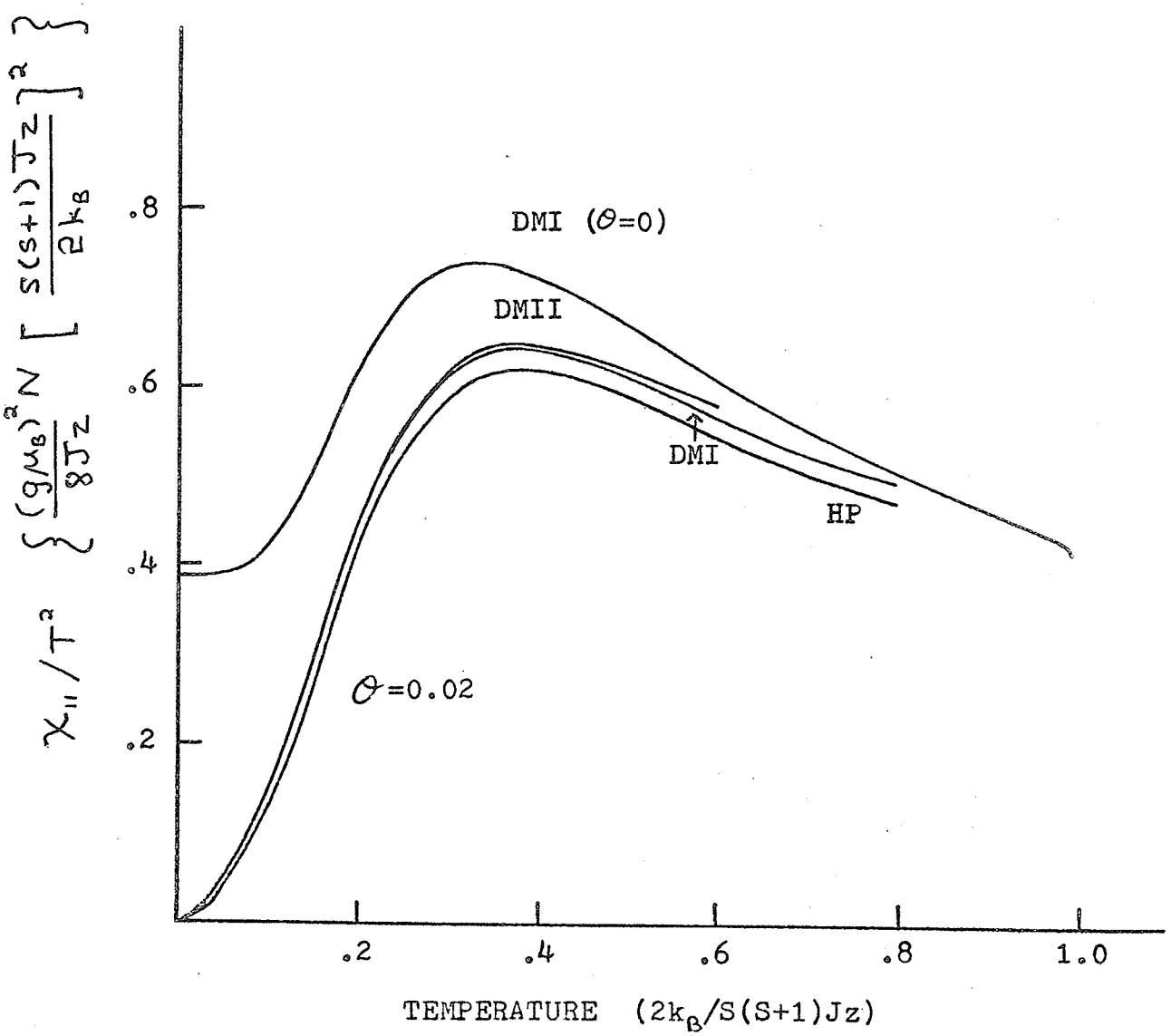
$$\chi_\perp = \frac{(g\mu_B)^2 N}{4Jz} \frac{\left(1 + (1+\frac{1}{8S})\alpha_2/2 - 3/16\,\alpha_2^2\right)^2}{1+\alpha_1+\frac{1}{16S}\alpha_1+\frac{1}{4}(\alpha_1-\alpha_2)^2-\frac{1}{16}\alpha_1^2+\frac{\theta}{2}\left(1-\frac{1}{2S}+2\alpha_2\right)}$$

DMI

$$\chi_\perp = \frac{(g\mu_B)^2 N}{4Jz} \frac{1+\alpha_2}{1+\alpha_1+\frac{\theta}{2}\left(1-\frac{1}{2S}+2\alpha_2\right)}$$

FIGURE 12

Parallel susceptibility of an ideal spin 5/2, $CsCl$ type antiferromagnet as predicted by HP, DMI and DMII for $\theta = .02$ and as predicted by DMI for $\theta = 0$.


$\chi_{\parallel}/T^2 \left\{ \frac{(g\mu_B)^2 N}{8Jz} \left[\frac{S(S+1)Jz}{2k_B} \right]^2 \right\}$
.8
.6
.4
.2
DMI (θ=0)
DMII
DMI
HP
θ=0.02
.2
.4
.6
.8
1.0
TEMPERATURE ($2k_B/S(S+1)Jz$)

DMII

$$\chi_{\perp} = \frac{(g\mu_B)^2 N}{4Jz} \frac{1+\alpha_a}{1+\alpha_1 + \Theta/2\left(1-\frac{1}{2S}+2\alpha_a\right)}$$

we obtain the curves in figure 13. The result for DMII is derived in appendix 4.

5) Free energy

For academic interest, a plot of the free energy is given in figure 14. Note that for an effective temperature of .6

HP $F = -1.1319$

DMI $F = -1.1332$

DMII $F = -1.1335$

We see that DMII has the minimum free energy and that HP has the largest.

6) Conclusion

The instability for DMII and the large effective anisotropy for HP make them less desirable for constituting a solution than the DMI case which is not only the simplest to use but in addition, has desirable effective anisotropy properties.

7.) Discussion

The value of $\Theta = .02$ for the anisotropy chosen in this section has been shown by Nagai and Tanaka (1970) to represent the actual amount of anisotropy in MnF_2 at T=0.

FIGURE 13

Perpendicular susceptibility of a spin 5/2, $CsCl$ type antiferromagnet as predicted by HP, DMI and DMII.

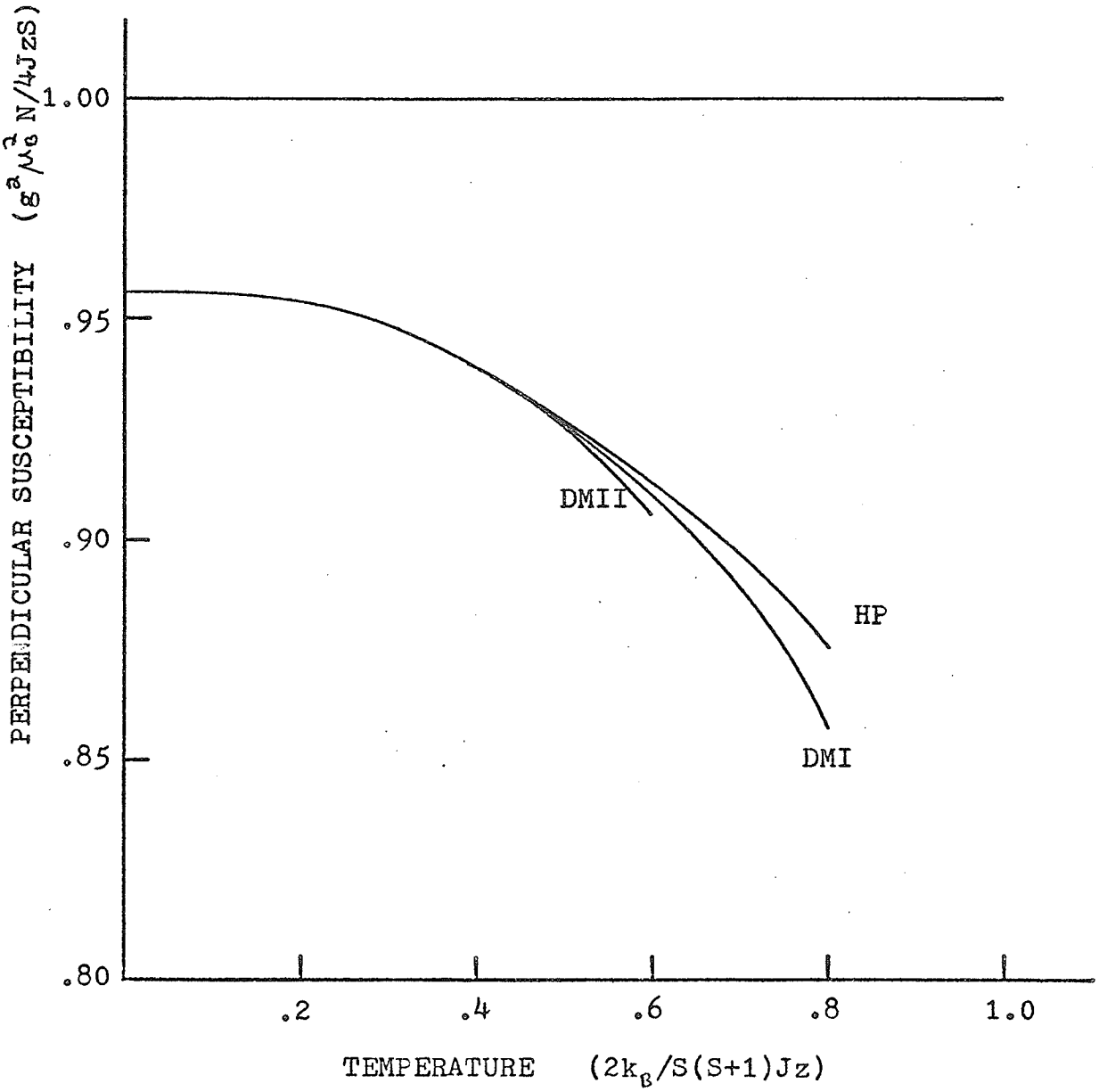
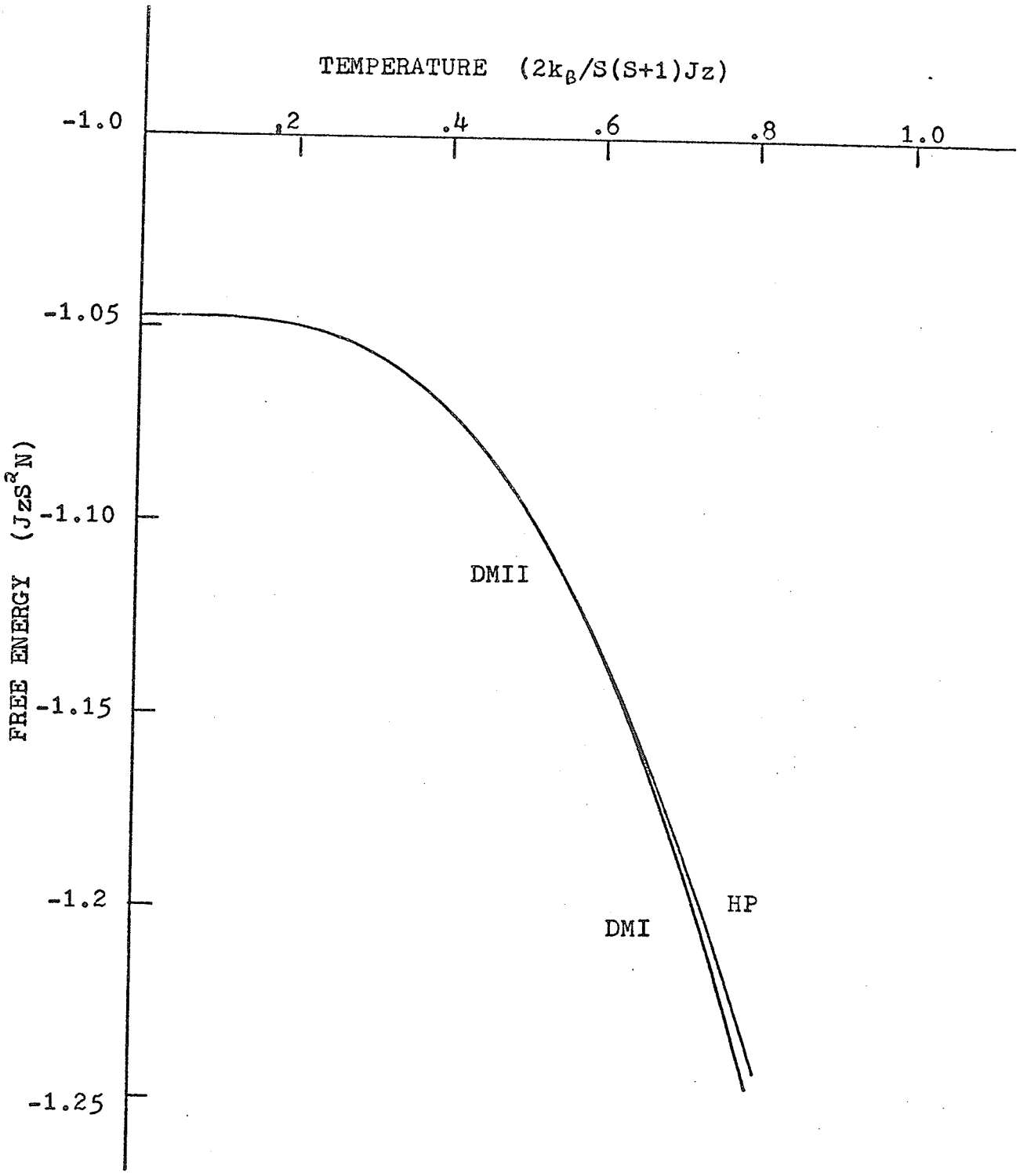

PERPENDICULAR SUSCEPTIBILITY ($g^a \mu_B^2 N/4Jz S$)
1.00
.95
.90
.85
.80
DMII
HP
DMI
.2
.4
.6
.8
1.0
TEMPERATURE ($2k_B/S(S+1)Jz$)

FIGURE 14

Free energy of a spin 5/2,
$CsCl$ type antiferromagnet
as predicted by HP, DMI and DMII.


TEMPERATURE (2k$_B$/S(S+1)Jz)
-1.0
.2
.4
.6
.8
1.0
-1.05
-1.10
-1.15
-1.2
-1.25
FREE ENERGY (JzS^{2}N)
DMII
DMI
HP

However since the anisotropy is 96% dipolar-dipolar, we will have the incorrect temperature dependence. Nevertheless, from equations (3.C.8) we can calculate that the temperature dependence decreases to zero at about a reduced temperature of .9 and therefore has the same general trend as the anisotropy measured in MnF_2. Also representative of MnF_2 is the spin value S=5/2 and lattice structure $C_sC\ell$.

The numerical work done in this section has not been carried out by semi-analytical methods but rather by a method proposed by Loly and Huett (1972). This method uses 10,000 points scattered at random in the irreducible zone of the reciprocal lattice and compresses them to obtain a non-uniform distribution of points weighted to take into account the large contribution of k=0 values. We have not taken the curves past a reduced temperature of .8 due to the breakdown of the simple iteration process. It is felt that they could be extended into this region however, one would use large amounts of computer time.

The semi-analytical and numerical methods have been checked against each other where possible and it is found that the numerical method has an error of less than 2%.

D. INFINITE SPIN LIMIT

In section (3.C) we have argued that the DMI representation is the easiest to use and probably the most accurate. Therefore, we will consider only this approx-

imation and examine the results in the limit of infinite spin, zero field and zero anisotropy. For this case we can simply take the results of section (4.A) and evaluate the infinite spin limit.

There are essentially three reasons for considering this classical case the most important being that the kinematical effect tends to zero and the asymptotic nature of the approximation disappears. Secondly, the solution to the ideal classical antiferromagnet should be the same as the ferromagnet (see appendix 1) thus giving a check on the general method (Loly 1970). Thirdly, the equations take on a much simplified form, making computer calculations unnecessary.

The meaning of infinite spin is the following: let

$$\tau = 2/(\beta J S^2 z) \qquad (4.D.1)$$

so that as $S \to \infty$, $J \to 0$ keeping τ finite. This has the effect of making the spin vector usually of length $\sqrt{S(S+1)}$ into length S, thereby giving rise to the concept of classical spin. This spin is not quantized and can take on any orientation with respect to a fixed co-ordinate system. Thus we have a problem similar to the one in appendix 1.

Consider first the definition of α_1 given by (4.A.36)

$$\alpha_1 = \frac{1}{2S}\left\{1 - \frac{2}{N}\sum_k \left[1-\gamma(k)^2\right]^{1/2} \coth \frac{\beta\varepsilon_k}{2}\right\} \qquad (4.D.2)$$

where

$$\varepsilon_k = 2Jz\,S(1+\alpha_1)\,[1-\gamma(k)^2]^{1/2}$$

$$= \frac{4}{\tau\beta S}(1+\alpha_1)\,[1-\gamma(k)^2]^{1/2} .$$

For large S the argument of the hyperbolic cotangent tends to zero

$$\coth x = \frac{1}{x} + \frac{x}{3} + \cdots \qquad (4.D.3)$$

so that

$$\alpha_1 = \frac{1}{2S}\left\{1 - \frac{\tau S}{2(1+\alpha_1)}\frac{2}{N}\sum_k 1 + O\left(\frac{1}{S}\right)\right\} . \qquad (4.D.4)$$

Putting $S \to \infty$,

$$\alpha_1 = -\tau/4(1+\alpha_1) . \qquad (4.D.5)$$

This has the solution

$$(1+\alpha_1) = \frac{1}{2}\left(1 \pm \sqrt{1-\tau}\right) . \qquad (4.D.6)$$

Choosing the plus sign gives

$$(1+\alpha_1) = \frac{1}{2}\left(1 + \sqrt{1-\tau}\right) \qquad (4.D.7)$$

$$= 1 - \frac{1}{4}\tau - \frac{1}{16}\tau^2 - \frac{1}{32}\tau^3 - \cdots .$$

A similar result holds for α_2. From (4.A.36)

$$\alpha_2 = \frac{1}{2S}\left\{1 - \frac{2}{N}\sum_k [1-\gamma(k)]^{-1/2} \coth \frac{\beta\varepsilon_k}{2}\right\} \qquad (4.D.8)$$

so that

$$\alpha_2 = \frac{1}{2S}\left\{1 - \frac{\tau S}{2(1+\alpha_1)}\frac{2}{N}\sum_k [1-\gamma(k)^2]^{-1}\right\} + O(1/S). \qquad (4.D.9)$$

Make the following definition (for C_sCl $\omega = 1.393204$)

$$\omega = \frac{2}{N}\sum_k \frac{1}{1-\gamma(k)^2} = \frac{2}{N}\sum_k \frac{1}{1-\gamma(k)} \qquad (4.D.10)$$

and we finally obtain in the limit $S \to \infty$

$$\alpha_2 = -\frac{\tau\omega}{4(1+\alpha_1)}$$

and

$$(1+\alpha_2) = 1 - \tau\omega/2(1-\sqrt{1-\tau}) \qquad (4.D.11)$$

$$= 1 - \frac{\tau\omega}{4} - \frac{\tau^2\omega}{16} - \frac{\tau^3\omega}{32} + \cdots .$$

The final quantity of interest is I which was introduced by (4.A.12)

$$I = \frac{8Jz\beta}{N}\sum_k \frac{e^{\beta\varepsilon_k}}{[e^{\beta\varepsilon_k}-1]^2} \qquad (4.D.12)$$

$$= \frac{16}{S^2 N\tau}\sum_k \frac{S^2\tau^2}{(1+\alpha_1)^2\, 16\,[1-\gamma(k)^2]} + O\left(\frac{1}{S}\right).$$

Taking limit as $S \to \infty$

$$= \tau\omega / 2(1+\alpha_1)^2$$

$$= \frac{1}{2}\tau\omega + \frac{1}{4}\tau^2\omega + \cdots \quad . \tag{4.D.13}$$

We are now in a position to write down explicity the properties of an ideal, infinite spin antiferromagnet.

Internal energy

$$U = -JNzS^2(1+\alpha_1)^2$$

$$= -JNzS^2(1+\sqrt{1-\tau})^2/4 \tag{4.D.14}$$

$$= -JNzS^2\left(1 - \tau/2 - \tau^2/16 - \tau^3/32 + \cdots\right)$$

Total magnetization

$$M_z = 0 \tag{4.D.15}$$

Sublattice magnetization

$$M_{1z} = g\mu_B SN(1+\alpha_2)$$

$$= g\mu_B SN\left[1 - \frac{\tau\omega}{2(1+\sqrt{1-\tau})}\right] \tag{4.D.16}$$

$$= g\mu_B SN\left(1 - \tau\omega/4 - \tau^2\omega/16 - \tau^3\omega/32 + \cdots\right)$$

Parallel susceptibility

$$\chi_{\parallel} = \frac{(g\mu_B)^2}{4Jz} N I \left[1 + \frac{I}{2}\frac{(1+\alpha_1)}{(1+\alpha_2)}\right]^{-1} \qquad (4.D.17)$$

$$= \frac{(g\mu_B)^2}{2Jz} \frac{N\tau\omega}{(1+\sqrt{1-\tau})^2} \left[1 + \frac{\tau\omega}{2(1+\sqrt{1-\tau}-\tau\omega/2)}\right]^{-1}$$

$$= \frac{(g\mu_B)^2}{8Jz} N\tau\omega \left[1 - \frac{\tau(\omega-2)}{4} - \frac{\tau^2(\omega-2)}{8} + \cdots\right]$$

Perpendicular susceptibility

$$\chi_{\perp} = \frac{(g\mu_B)^2}{4Jz} N \frac{(1+\alpha_2)}{(1+\alpha_1)}$$

$$= \frac{(g\mu_B)^2}{4Jz} N \left[1 - \frac{\tau\omega}{2(1+\sqrt{1-\tau})}\right] \Big/ (1+\sqrt{1-\tau}) \qquad (4.D.18)$$

$$= \frac{(g\mu_B)^2 N}{4Jz} \left[1 - (\omega-1)\tau/4 - (\omega-1)\tau^2/8 + \cdots\right]$$

Specific heat

$$C_v = -2Jz S^2 N(1+\alpha_1)\frac{\partial \alpha_1}{\partial T}$$

$$= -4Nk_B(1+\alpha_1)\frac{\partial}{\partial \tau}\alpha_1 \qquad (4.D.19)$$

$$= Nk_B \frac{1}{2}\frac{(1+\sqrt{1-\tau})}{\sqrt{1-\tau}}$$

$$= Nk_B\left(1 + \frac{1}{4}\tau + \frac{3}{16}\tau^2 + \cdots\right)$$

CHAPTER 5

SUMMARY

The method described in this thesis provides an example of a way of obtaining an approximate solution by using a non-Hermitian Hamiltonian. Another example of such a method occurs in ferromagnetism where the same Dyson-Maleev transformations are used; however in that case the approximate Hamiltonian H_o is Hermitian in reciprocal boson space. In our case, H_o is not Hermitian in reciprocal boson space and only after the Bogolubov transformation does H_o become Hermitian. The implication of these two examples is the following: one can use a non-unitary transformation to obtain an approximate solution provided there exists a representation in which H_o is Hermitian. In that representation H_o may be a realistic physical solution to the full Hamiltonian H — a drawback being that one cannot compare solutions on the basis of a minimum free energy.

Of relevance to antiferromagnetism are the three approximations obtained here which are valid for arbitrary uniaxial anisotropy and magnetic field. Clearly, investigations of the field dependence and anisotropy dependence of many macroscopic properties can be done with the equations developed. It appears as if DMI is the preferred solution to do this with as it has desir-

able effective anisotropy properties. Of particular consequence is the DMI formula for $\chi_{\parallel}$ for the zero anisotropy limit which is a new result and shows considerable improvement over Liu's (1966) result. As well, the infinite spin results derived in section (4.D) have not been given before.

Preliminary comparisons with experiment for $\chi_{\parallel}$ have been undertaken with the published results for MnF_2. However, Nagai and Tanaka (1970) who used a HP method have obtained excellent agreement with the experimental results. Since we have incorporated anisotropy in the same manner as they have, we expect to obtain a similar agreement - this is due to the fact that the three solutions for $\chi_{\parallel}$ (see figure 12) are very similar and do not depend critically on representation.

One of the shortcomings of this model is the prediction for $\chi_{\perp}$. None of the models presented here are capable of explaining the upturn in the experimental results. It is felt that the Heisenberg model should explain that aspect and in fact Kanamori and Itoh (1972) have shown that second order theories will give an upturn trend. They have used a HP approximation and it now seems that one could use DMI and calculate the second order effects, however, one would be faced with the problem mentioned in the introduction, namely large amounts of algebra and six dimensional integrals.

Finally of mathematical importance is the Bogolubov

transformation itself which, in this work, has been generalized to its non-unitary form.

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APPENDIX 1

CLASSICAL HAMILTONIAN

In the limit of classical spin (Heller and Kramers 1934) we can represent the Heisenberg Hamiltonian (2.A.2) by(using spherical co-ordinates)

$$H_1 = S^2 \sum_{i\delta} \left[\cos\theta_{1i} \cos\theta_{2i+\delta} + \sin\theta_{1i} \sin\theta_{2i+\delta} \cos(\phi_{1i} - \phi_{2i+\delta}) \right]$$

$$+ S^2 \sum_{j\delta} \left[\cos\theta_{2j} \cos\theta_{1j+\delta} + \sin\theta_{2j} \sin\theta_{1j+\delta} \cos(\phi_{1j+\delta} - \phi_{2j}) \right]$$

$$V_1 = S \sum_i \left[\sin\theta_{1i} \cos\phi_{1i} + \sin\theta_{2j} \cos\phi_{2j} \right]$$

$$V_2 = S \sum_i \left[\cos\theta_{1i} + \cos\theta_{2j} \right] \qquad \text{(Ap.1.1)}$$

$$V_3 = S^2 \sum_i \left[\cos^2\theta_{1i} + \cos^2\theta_{2j} \right]$$

We see that a ground state of H_1 occurs for

$$\theta_{1i} = 0 \quad , \quad \theta_{2j} = \pi \qquad \text{(Ap.1.2)}$$

and is the antiferromagnetic ground state.

If we rotate one sublattice by 180 degrees so that $\theta_{1i} \to \theta_{1i} + \pi$ then $\cos\theta_{1i} \to -\cos(\theta_{1i} + \pi)$ and $\sin(\theta_{1i}) \to -\sin(\theta_{1i} + \pi)$ and $H_1 \to -H_1$. By changing the sign of J we see that the ferromagnetic Hamiltonian JH_1 is the same as the antiferromagnetic Hamiltonian. This however is not true for V_1 , V_2 but holds for V_3 . Therefore in the limit of classical spin and zero field the ferro-

magnetic and antiferromagnetic Hamiltonians have the same eigenvalues.

Other references concerning the classical limit are Loly and Mikusik (1970) and Mikusik (thesis).

APPENDIX 2

NON-UNITARY BOGOLUBOV TRANSFORMATION

The relationship between the a_k, b_k and the α_k, β_k used in the text is given by

$$O^{-1}a_k O = (\alpha_k \cosh\theta_{1k} - \beta_k^+ \sinh\theta_{1k})/[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}$$

$$O^{-1}a_k^+ O = (\alpha_k^+ \cosh\theta_{2k} - \beta_k \sinh\theta_{2k})/[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}$$

(Ap.2.1)

$$O^{-1}b_k O = (-\alpha_k^+ \sinh\theta_k + \beta_k \cosh\theta_{1k})/[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}$$

$$O^{-1}b_k^+ O = (-\alpha_k \sinh\theta_{2k} + \beta_k^+ \cosh\theta_{2k})/[\cosh(\theta_{1k}-\theta_{2k})]^{1/2}$$

We have assumed that the operator O exists and has an inverse. O defines a non-unitary transformation except for those cases where $\theta_{1k} = \theta_{2k}$ for which O becomes unitary and the transformation becomes the usual Bogolubov transformation.

The above relations define a canonical transformation. Consider for example

$$[a_k^+, a_k] = a_k^+ a_k - a_k a_k^+$$

$$= O[O^{-1}a_k^+ O O^{-1}a_k O - O^{-1}a_k O O^{-1}a_k^+ O]O^{-1}$$

$$= O[\alpha_k^+ \alpha_k \cosh\theta_{1k}\cosh\theta_{2k} + \text{etc.}$$

$$= O(1)O^{-1} = -1 \qquad \text{(Ap.2.2)}$$

$$[a_k, b_k] = 0 \ .$$

So that if the α_k , β_k operators satisfy boson commutation relations so do the a_k , b_k and visa versa.

Consider the following operators

$$\hat{\alpha}_1 = -\frac{1}{SN}\sum_k (a_k^+ a_k + b_k^+ b_k + \gamma(k) a_k^+ b_k^+ + \gamma(k) a_k b_k)$$

$$\hat{\alpha}_2 = -\frac{1}{SN}\sum_k (a_k^+ a_k + b_k^+ b_k)$$

$$\hat{\alpha}_3 = -\frac{1}{SN}\sum_k (a_k^+ a_k - b_k^+ b_k) \qquad \text{(Ap.2.3)}$$

$$\hat{\alpha}_4 = -\frac{1}{SN}\sum_k \gamma(k)(a_k^+ b_k^+ - a_k b_k)$$

where one hopefully will not be confused with the boson operators α_k and the new operators $\hat{\alpha}_i$. Then in the α_k , β_k representation

$$\hat{\alpha}_1 = -\frac{1}{SN}\sum_k \Big[(\alpha_k^+\alpha_k + \beta_k^+\beta_k + 1)\Big(\frac{\cosh\theta_k}{\cosh\Delta\theta_k} - \gamma(k)\sinh\theta_k\Big)$$

$$- (\alpha_k^+\beta_k^+ + \alpha_k\beta_k)\Big(\frac{\sinh\theta_k}{\cosh\Delta\theta_k} - \gamma(k)\cosh\theta_k\Big)$$

$$- (\alpha_k^+\beta_k^+ - \alpha_k\beta_k)\frac{\sinh\Delta\theta_k}{\cosh\Delta\theta_k} - 1\Big]$$

$$\hat{\alpha}_2 = -\frac{1}{SN}\sum_k \Big[(\alpha_k^+\alpha_k + \beta_k^+\beta_k + 1)\frac{\cosh\theta_k}{\cosh\Delta\theta_k}$$

$$- (\alpha_k^+\beta_k^+ + \alpha_k\beta_k)\frac{\sinh\theta_k}{\cosh\Delta\theta_k} \qquad \text{(Ap.2.4)}$$

$$- (\alpha_k^+ \beta_k^+ - \alpha_k \beta_k) \frac{\sinh \Delta\theta_k}{\cosh \Delta\theta_k} - 1 \Big]$$

$$\hat{\alpha}_3 = - \frac{1}{SN} \sum_k (\alpha_k^+ \alpha_k - \beta_k^+ \beta_k)$$

$$\hat{\alpha}_4 = - \frac{1}{SN} \sum_k \Big[\gamma(k) (\alpha_k^+ \alpha_k + \beta_k^+ \beta_k + 1) \frac{\cosh \theta_k}{\cosh \Delta\theta_k} \sinh \Delta\theta_k$$

$$- \gamma(k) (\alpha_k^+ \beta_k^+ + \alpha_k \beta_k) \frac{\sinh \theta_k}{\cosh \Delta\theta_k} \sinh \Delta\theta_k$$

$$+ \gamma(k) (\alpha_k^+ \beta_k^+ - \alpha_k \beta_k) \frac{1}{\cosh \Delta\theta_k} - 1 \Big]$$

where

$$\theta_k = \theta_{1k} + \theta_{2k}$$

$$\Delta\theta_k = \theta_{1k} - \theta_{2k} \, .$$

APPENDIX 3

APPLICATION OF BOGOLUBOV TRANSFORMATION

We want to diagonalize the operator H_o given by

$$H_o = -NJzS^2 + 2JzS\sum_k (\varepsilon_1 a_k^+ a_k + \varepsilon_2 b_k^+ b_k + \varepsilon_3 \gamma(k) a_k^+ b_k^+ + \varepsilon_4 \gamma(k) a_k b_k) \quad \text{(Ap.3.1)}$$

Using appendix 2 (see Ap.2.3) we can express this as

$$H_o = -NJzS^2 (1 + 2A\hat{\alpha}_1 + 2B\hat{\alpha}_2 + 2C\hat{\alpha}_3 + 2D\hat{\alpha}_4) \quad \text{(Ap.3.2)}$$

where

$$A = \frac{1}{2}(\varepsilon_3 + \varepsilon_4)$$

$$B = \frac{1}{2}(\varepsilon_1 + \varepsilon_2 - \varepsilon_3 - \varepsilon_4)$$

$$C = \frac{1}{2}(\varepsilon_1 - \varepsilon_2)$$

$$D = \frac{1}{2}(\varepsilon_3 - \varepsilon_4) \ .$$

In the α , β representation $\hat{\alpha}_i$ is given by (Ap.2.4). Setting the coefficients of $(\alpha_k^+ \beta_k^+ + \alpha_k \beta_k)$ and $(\alpha_k^+ \beta_k^+ - \alpha_k \beta_k)$ equal to zero yields two equations for Θ_k and $\Delta\Theta_k$.

$$(A+B)\frac{\sinh\Theta_k}{\cosh(\Delta\Theta_k)}\left[1+D\gamma(k)\sinh(\Delta\Theta_k)\right]=A\gamma(k)\cosh\Theta_k$$

$$(A+B)\frac{\sinh(\Delta\Theta_k)}{\cosh\Theta_k}=\frac{D\gamma(k)}{\cosh(\Delta\Theta_k)}\,.$$

The solution is

$$\sinh(\Delta\Theta_k)=D\gamma(k)/(A+B)$$

$$\sinh\Theta_k=\frac{A\gamma(k)}{\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2}}\,. \qquad \text{(Ap.3.3)}$$

Imposing these conditions the operators $\hat{\alpha}_i$ take on the simplified form

$$\hat{\alpha}_1=-\frac{1}{SN}\sum_k\left[(\alpha_k^+\alpha_k+\beta_k^+\beta_k+1)\frac{(A+B)-A\gamma(k)^2}{\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2}}-1\right]$$

$$\hat{\alpha}_2=-\frac{1}{SN}\sum_k\left[(\alpha_k^+\alpha_k+\beta_k^+\beta_k+1)\frac{(A+B)}{\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2}}-1\right]$$

$$\hat{\alpha}_3=-\frac{1}{SN}\sum_k(\alpha_k^+\alpha_k-\beta_k^+\beta_k)$$

$$\hat{\alpha}_4=-\frac{1}{SN}\sum_k\left[(\alpha_k^+\alpha_k+\beta_k^+\beta_k+1)\frac{D\gamma(k)^2}{\left[(A+B)^2-(A^2-D^2)\gamma(k)^2\right]^{1/2}}\right]. \qquad \text{(Ap.3.4)}$$

Finally H_o is diagonal and can be written

$$H_o=-NJ_zS^2+2J_zS\sum_k C(n_k-n_k') \qquad \text{(Ap.3.5)}$$

$$+2J_zS\sum_k\left[(n_k+n_k'+1)\cdot\left\{(A+B)^2-(A^2-D^2)\gamma(k)^2\right\}^{1/2}\right.$$

$$\left.-(A+B)\right]$$

where

$$n_k = \alpha_k^+ \alpha_k$$

$$n_k' = \beta_k^+ \beta_k \quad .$$

APPENDIX 4

ALGEBRA FOR $\langle H \rangle_o$

Simplification of the algebra rests mainly on the observation that if care is taken, one can interchange the order of shifting, rotating (Bogolubov) and taking the expectation. In order to change the order of rotating and taking the expectation, we note that a_k^+ and b_k^+ act as creation operators with respect to $\alpha_k^+ \alpha_k$ (see equation 3.B.4). Similarly a_k and b_k are destruction operators so we know that terms of the form $\langle a_k^+ b_k \rangle_o$ and $\langle a_k b_k^+ \rangle_o$ will be zero whereas terms of the form $\langle a_k^+ b_k^+ \rangle_o$ and $\langle a_k b_k \rangle_o$ will be non zero. Therefore equation (3.C.2) tells us that a term of the form

$$\sum_{kk'q} \langle 4\gamma(k-k') a_k^+ a_{k'} b_{k+q}^+ b_{k'+q} \rangle_o \qquad \text{(Ap.4.1)}$$

decouples (without further approximation) to

$$\sum_{kk'} \left[4 \langle a_k^+ a_k \rangle_o \langle b_{k'}^+ b_{k'} \rangle_o + 4\gamma(k)\gamma(k') \langle a_k^+ b_k^+ \rangle_o \langle a_{k'} b_{k'} \rangle_o \right] . \qquad \text{(Ap.4.2)}$$

The above step requires the property

$$\gamma(k-k') = \gamma(k)\gamma(k') + \gamma'(k)\gamma'(k') \qquad \text{(Ap.4.3)}$$

where γ' is an odd function so that the sum over $\gamma'(k) \times \langle a_k^+ b_k^+ \rangle_o$ cancels identically. This property is true for all the cubic lattices, for which $\gamma(k)$ is a sum or product of cosines. i.e.

$$\cos(k-k') = \cos k \cos k' + \sin k \sin k' .$$

To commute the operations of taking the expectation and shifting one sees that a term of the form

$$\sum_{kk'q} \langle 4\gamma(k-k') c_k^+ c_{k'} d_{k+q}^+ d_{k'+q} \rangle_0 \qquad \text{(Ap.4.4)}$$

when shifted to the a_k, b_k representation will involve only 4-operator terms, 2-operator terms and constant terms. All 2-operator terms which are not creation-annihilation pairs will have zero expectation. The remaining terms are

$$\sum_{kk'} \Big[4\langle a_k^+ a_k \rangle_0 \langle b_{k'}^+ b_{k'} \rangle_0 + 4\gamma(k)\gamma(k') \langle a_k^+ b_k^+ \rangle_c \langle a_{k'} b_{k'} \rangle_c$$

$$+ 4\rho_1\rho_2 \delta_{k,0} \langle b_{k'} b_{k'} \rangle_0 + 4\rho_3\rho_4 \delta_{k',0} \langle a_k^+ a_k \rangle_0 \qquad \text{(Ap.4.5)}$$

$$+ 4\rho_2\rho_4 \delta_{k,0} \gamma(k') \langle a_{k'} b_{k'} \rangle_0 + 4\rho_1\rho_3 \delta_{k',0} \gamma(k) \langle a_k^+ b_k^+ \rangle$$

$$+ 4\rho_1\rho_2\rho_3\rho_4 \delta_{k0} \delta_{k'0} \Big] .$$

These can be regrouped to

$$\sum_{kk'} \Big[4 \big(\langle a_k^+ a_k \rangle_0 + \delta_{k0} \rho_1 \rho_2 \big) (\cdots) \cdots \Big] \qquad \text{(Ap.4.6)}$$

and for instance

$$\sum_k \left(\langle a_k^+ a_k \rangle_0 + \delta_{k0} \rho_1 \rho_2 \right) = \alpha_2 - \alpha_1 \qquad \text{(Ap.4.7)}$$

By this method, all four operator terms are expressible in terms of $\alpha_1 \ldots \alpha_4$.

The odd operator terms can be handled by first shifting one operator to reduce it to an even term for example

$$\sum_{kk'} \langle c_k^+ c_{k'}^+ c_{k+k'} \rangle_0$$

$$= - \sum_{kk'} \langle a_k^+ a_{k'} \rangle_0 \rho_2 - \sum_{kk'} \langle a_k^+ a_k \rangle \rho_2 \qquad \text{(Ap.4.8)}$$

$$- \rho_2^2 \rho_1$$

$$= 2\rho_2 (\alpha_2 - \alpha_3) + \rho_2^2 \rho_1 .$$

Thus, all odd order terms can be expressed in terms of $\alpha_1, \ldots, \alpha_4$ and $\rho_1, \ldots, \rho_4$.

APPENDIX 5

PERPENDICULAR SUSCEPTIBILITY - DMII

Since this result is not easy to obtain, the following outline should be helpful. The free energy is

$$F = -NJz S^2 \Big[(1+\alpha_1)^2 - 2\alpha_2\alpha_4 - \alpha_4^2 + (\alpha_1 - \alpha_2 - \alpha_4)\left(\alpha_2^2 + \tfrac{1}{2}(\alpha_1-\alpha_2)^2 - \tfrac{1}{2}\alpha_4^2\right) - \frac{h_x}{2}(\rho_1+\rho_2+\rho_3+\rho_4) - \frac{h_x}{2}(\rho_1+\rho_3)\alpha_2 + \theta + 2\theta\left(1-\tfrac{1}{2S}\right)\alpha_2 + 2\theta\alpha_2^2 \Big] - kTS_0 \,. \tag{Ap.5.1}$$

Differentiating with respect to ρ_1, ρ_4 and neglecting powers of $O(\alpha^3)$ leaves

$$1)\quad 2(1+\alpha_1)(\rho_2+\rho_3) + 2\alpha_3\rho_3 - 2\alpha_4(\rho_2-\rho_3) + 2\rho_3\left(\alpha_2^2 + \tfrac{1}{2}(\alpha_1-\alpha_2)^2\right) + 2\rho_2\alpha_2(\alpha_1-\alpha_2) + \rho_3(\alpha_1-\alpha_2)^2 + \frac{h_x}{2}(1+\alpha_2) + 2\theta\left(1-\tfrac{1}{2S}+2\alpha_2\right)\rho_2 = 0$$

$$2)\quad 2(1+\alpha_1)(\rho_2+\rho_3) - 2\alpha_2\rho_2 - 2\alpha_4(\rho_3+\rho_2) + 2\rho_3\alpha_2(\alpha_1-\alpha_2) + \rho_2(\alpha_1-\alpha_2)^2 + h_x/2 + 2\theta\left(1-\tfrac{1}{2S}+2\alpha_2\right)\rho_3 = 0 \,. \tag{Ap.5.2}$$

These two equations can be expressed

$$\begin{bmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{bmatrix} \begin{pmatrix} \rho_2 \\ \rho_3 \end{pmatrix} = -\frac{h_x}{2} \begin{pmatrix} 1+\alpha_2 \\ 1 \end{pmatrix} \qquad \text{(Ap.5.3)}$$

where

$$a_{11} = 2(1+\alpha_1) - 2\alpha_4 + 2\alpha_2(\alpha_1-\alpha_2) + 2\theta\left(1-\tfrac{1}{2S}+2\alpha_2\right)$$

$$a_{12} = 2(1+\alpha_1) + 2\alpha_2 + 2\alpha_4 + 2\alpha_2^2 + 2(\alpha_1-\alpha_2)^2$$

$$a_{21} = 2(1+\alpha_1) - 2\alpha_2 - 2\alpha_4 + (\alpha_1-\alpha_2)^2$$

$$a_{22} = 2(1+\alpha_1) - 2\alpha_4 + 2\alpha_2(\alpha_1-\alpha_2) + 2\theta\left(1-\tfrac{1}{2S}+2\alpha_2\right)$$

and the solution is

$$\rho_3 = -\frac{h_x}{8} \; \frac{\theta\left(1-\frac{1}{2S}+2\alpha_2\right) - \frac{1}{2}(\alpha_1-\alpha_2)^2 + O(\alpha^3)}{\theta(1+\alpha_1)\left(1-\frac{1}{2S}+2\alpha_2\right) - \frac{3}{4}(\alpha_1-\alpha_2)^2 + \frac{\theta^2}{2}\left(1-\frac{1}{2S}+2\alpha_2\right)+\cdots}$$

$$\rho_2 = -\frac{h_x}{8} \; \frac{\theta\left(1-\frac{1}{2S}+2\alpha_2\right)(1+\alpha_2) - (\alpha_1-\alpha_2)^2 + O(\alpha^3)}{\theta(1+\alpha_1)\left(1-\frac{1}{2S}+2\alpha_2\right) - \frac{3}{4}(\alpha_1-\alpha_2)^2 + \frac{\theta}{2}\left(1-\frac{1}{2S}+2\alpha_2\right)+\cdots} \qquad \text{(Ap.5.4)}$$

The equations for ρ_1 , ρ_4 are similar to those for ρ_2 , ρ_3 so that $\rho_1 = \rho_3$, $\rho_4 = \rho_2$. Finally we obtain

$$\gamma_\perp \propto (\rho_1+\rho_3)(1+\alpha_2)+(\rho_2+\rho_4)$$

$$= \frac{(1+\alpha_2)\Theta(1-\frac{1}{2s}+2\alpha_2)-\frac{3}{4}(\alpha_1-\alpha_2)^2+O(\alpha^3)}{(1+\alpha_1)\Theta(1-\frac{1}{2s}+2\alpha_2)-\frac{3}{4}(\alpha_1-\alpha_2)^2+\frac{\Theta^2}{2}(1-\frac{1}{2s}+2\alpha_2)+O(\alpha^3)}$$

$$= \frac{(1+\alpha_2)}{1+\alpha_1+\frac{\Theta}{2}(1-\frac{1}{2s}+2\alpha_2)} + \frac{O(\alpha^3)}{\Theta(1-\frac{1}{2s})} .$$

PART II

LATTICE GREEN FUNCTIONS

PART II

A. INTRODUCTION

The aim of this part of the thesis is to determine a semi-analytical method for evaluating integrals found in part I. While doing so, it was discovered that these integrals have applications to other parts of solid state physics, for example, lattice vibrations, and arise from the solution of the Helmholtz equation on a discrete lattice.

The solution of Helmholtz equation is facilitated by the use of Green functions where a delta function drives the differential equation. For a periodic lattice the Green function is given by [1]

$$G(E,r) = \frac{1}{N} \sum_{k} \frac{1}{E-E_k} e^{ik\cdot r}$$

where

$$E_k = 3 - \cos k_x - \cos k_y - \cos k_z \qquad \text{sc.}$$

$$= 4(1 - \cos k_x \cos k_y \cos k_z) \qquad \text{bcc.}$$

$$= 2(3 - \cos k_x \cos k_y - \cos k_y \cos k_z - \cos k_x \cos k_z) \qquad \text{fcc.}$$

In section B of this part, we will reprint a paper published on the evaluation of these integrals, their thermodynamic extension and an integral found in anti-

ferromagnetism known as c^0. In sections C and D, we show how general asymptotic expansions lead to a method of evaluation of the antiferromagnetic integrals needed in part I.

B. LATTICE GREEN FUNCTIONS

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On the Evaluation of Lattice Green Functions and Watson-Like Integrals*

D. J. AUSTEN AND P. D. LOLY

Department of Physics, University of Manitoba, Winnipeg, Manitoba, Canada

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A method is developed whereby lattice Green functions and Watson-like integrals can be readily computed by splitting them into a sum of a power series and an asymptotic series. The method is illustrated by the evaluation of an antiferromagnetic integral and a basic thermodynamic Watson's integral and followed by a treatment of the generalized problem on all the cubic lattices.

1. INTRODUCTION

Recently there has been considerable interest in the evaluation of lattice Green functions (LGF) [1, 2] and thermodynamic extensions of Watson sums (TWS) [3]; the basic Watson sum or integral being a special case of LGF. The essential ingredient of all these three-dimensional integrals is the appearance of the nearest-neighbor dispersion function in a manner which gives a singularity in the integrand. Through investigations to evaluate related integrals arising from the two-sublattice character of an antiferromagnet we have developed a technique which can also be used for the evaluation of LGF and TWS for all the cubic lattices to any desired precision.

These three-dimensional integrals arise as Brillouin zone sums of large lattices that have an essentially continuous distribution of wavevectors, while the Brillouin zone sums are themselves a direct consequence of the lattice periodicity [4]. The dispersion function considered in connection with LGF and TWS is usually restricted to problems that have only nearest-neighbor coupling, though most of the models involved in physical theories become more realistic for longer-ranged interactions.

These integrals may be written in the following manner: Starting with the general LGF,

$$W_i(\eta, l, m, n) = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} dx\, dy\, dz\, \frac{\cos lx \cos my \cos nz}{\eta - \gamma_i(x, y, z)}, \qquad (1)$$

* Research supported by the National Research Council of Canada.

while the general TWS incorporates the Planck or Bose–Einstein distribution in the following manner:

$$T_i(\alpha, \eta, l, m, n) = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} dx\, dy\, dz\, \frac{\cos lx \cos my \cos nz}{\exp\{\alpha[\eta - \gamma_i(x, y, z)]\} - 1}. \quad (2)$$

In (1) and (2) we have already converted the Brillouin zone sum to an integral, simplified the wavevector variables to x, y, and z, and arranged that they range over a cubic domain [5, 3]. Other ingredients of (1) and (2) are i, which distinguishes the lattices, $i = 1$ for sc, $i = 2$ for bcc, $i = 3$ for fcc; α physically represents an inverse temperature giving $0 < \alpha < \infty$, while $\eta \neq 1$ represents an external magnetic field (in the magnetic context) giving the range of interest as $1 < \eta < \infty$. l, m, n are integers which could be zero. The quantity $\eta - \gamma_i$ is known as the dispersion function, while γ_i is a structure factor:

$$i = 1, \quad \text{sc}, \qquad \gamma_1 = \tfrac{1}{3}(\cos x + \cos y + \cos z), \quad (3)$$

$$i = 2, \quad \text{bcc}, \qquad \gamma_2 = \cos x \cos y \cos z, \quad (4)$$

$$i = 3, \quad \text{fcc}, \qquad \gamma_3 = \tfrac{1}{3}(\cos x \cos y + \cos y \cos z + \cos x \cos z). \quad (5)$$

The singularities of the integrands of (1) and (2) occur for $\eta = 1$ at the origin $(x, y, z = 0)$ when the dispersion function vanishes. Furthermore, as $\alpha \to 0$, $T_i \to W_i/\alpha$, giving a connection between TWS and LGF.

Arising from the two-sublattice character of antiferromagnets we find the following integrals:

$$V_i(\delta, l, m, n) = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} dx\, dy\, dz\, \frac{\cos lx \cos my \cos nz}{[\delta^2 - \gamma_i(x, y, z)^2]^{1/2}}, \quad (6)$$

where $\delta > 1$ represents ferrimagnetism, and physically we cannot generate the case for $i = 3$, $i = 1$ being known as the NaCl type, $i = 2$ the CsCl type. There is also some interest in the thermodynamic extension of (6).

Our method is closely related to a technique used in the evaluation of LGF by Maradudin *et al.* [6, 7] where an integral with limits from 0 to ∞ is split to permit the use of both power and asymptotic series. In the present paper we encounter the sum of an infinite number of terms which can be split in an analogous fashion.

Section II shows how a Fourier transform generates an infinite sum for $V_1(1, 0, 0, 0)$ and in Section III a similar result is found for $T_1(\alpha, 1, 0, 0, 0)$, albeit in a different way, and gives a method of evaluating that specific TWS in a region that was previously difficult. Finally, in Section IV, by developing general asymptotic formulas, we consider the problem of calculating the general TWS for all the cubic lattices. In doing so we also find a procedure for the general LGF

(involving the split integral version) which was previously given only for the restricted cases of $W_1(\eta, 0, 0, 0)$ [6] and $W_2(\eta, 0, 0, 0)$ [8].[1]

II. Antiferromagnetic Integrals, c'

The quantity arising in antiferromagnetism is actually $V_i(1, 0, 0, 0) - 1$, known as c' [9] and for the CsCl type ($i = 2$) was definitively evaluated by Davis [10]. Davis' method was to expand the square root and integrate term by term. For the NaCl type ($i = 1$), a larger number of terms arise in the expansion, the convergence being very slow.[2] However, another type of expansion can be used profitably.

Writing $V = V_1(1, 0, 0, 0)$, we have explicitly

$$V = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} \frac{dx\, dy\, dz}{[1 - (1/9)(\cos x + \cos y + \cos z)^2]^{1/2}}. \tag{7}$$

Making use of the Fourier expansion [11],

$$\frac{1}{(1 - t^2)^{1/2}} = \frac{\pi}{2} + \pi \sum_{m=1}^{\infty} J_0(m\pi) \cos(m\pi t), \quad |t| < 1, \tag{8}$$

where $J_0(m\pi)$ is a Bessel function of the first kind, one finds

$$V = \frac{\pi}{2} + \pi \sum_{m=1}^{\infty} \frac{J_0(m\pi)}{(2\pi)^3} \iiint_{-\pi}^{\pi} \cos\left[\frac{m\pi}{3}(\cos x + \cos y + \cos z)\right] dx\, dy\, dx. \tag{9}$$

Expanding the cosine and using the following two identities:

$$\frac{1}{(2\pi)} \int_{-\pi}^{\pi} dx \cos(t \cos x) = J_0(t), \tag{10}$$

$$\frac{1}{(2\pi)} \int_{-\pi}^{\pi} dx \sin(t \cos x) = 0, \tag{11}$$

[1] Using a different procedure, Mannari and Kawabata [Extended Watson Integrals and their Derivatives, Research Notes of Dept. of Physics, Okayama University, No. *15* (1964)] evaluated $W_i(\eta, 0, 0, 0)$ for all the cubic lattices.

[2] This expansion can be rewritten in terms of random walk probabilities

$$V = \sum_{n=0}^{\infty} \frac{(2n)!}{n!\, n!\, 2^{2n}} p_{2n}(0, 0, 0).$$

Although this series is slowly convergent it can be accurately evaluated using the Euler–Maclaurin summation formula. [For details, see C. Domb, *Proc. Camb. Phil. Soc.* **50** (1954).]

we have

$$V = \frac{\pi}{2} + \pi \sum_{m=1}^{\infty} J_0(m\pi)\, J_0^3(m\pi/3). \tag{12}$$

This is evaluated as

$$V = \frac{\pi}{2} + \pi \sum_{m=1}^{N} J_0(m\pi)\, J_0^3(m\pi/3) + \text{asymptotic expansion}. \tag{13}$$

The asymptotic expansion is obtained from the expression [11]

$$J_0(x) \approx (2/\pi x)^{1/2} \left\{\cos(x - \pi/4)\left[1 - \frac{1^2 3^2}{2!\,(8x)^2} + \cdots\right] + \sin(x - \pi/4)\left[\frac{1^2}{1!\,(8x)} - \frac{1^2 3^2 5^2}{3!\,(8x)^3} + \cdots\right]\right\}. \tag{14}$$

We found that ten terms in the expansion of (13) were more than sufficient for seven-figure accuracy. Using $N = 4$ gives $c' = 0.1567154$, the same value being obtained with $N = 3, 5$. The best value previously was 0.156, given by Kubo [9] in 1952.

III. Thermodynamic Integral $T_1(\alpha, 1, 0, 0, 0)$

Letting $T(\alpha) \equiv T_1(\alpha, 1, 0, 0, 0)$, we have explicitly that

$$T(\alpha) = \frac{1}{(2\pi)^3} \int\int\int_{-\pi}^{\pi} \frac{dx\, dy\, dz}{\exp \alpha[1 - (1/3)(\cos x + \cos y + \cos z)] - 1}. \tag{15}$$

For small α (high temperature) we can expand the integrand in a power series about $\alpha = 0$ and integrate term by term with the result

$$T(\alpha) = \frac{W}{\alpha} - \frac{1}{2} + \sum_{m=1}^{\infty} A_m \alpha^{2m-1}, \qquad \alpha < \pi, \tag{16}$$

where W is Watson's integral [5] (= 1.516386...) and

$$A_m = \frac{(-1)^{m-1}}{2m} B_m \sum_{k=0}^{m-1} \frac{1}{(2m - 2k - 1)!\, 6^{2k}} \sum_{l=0}^{k} \frac{(2k)!}{[(l)!]^4 [(k - l)!]^2}, \tag{17}$$

B_m being Bernoulli's numbers. The first few terms of this series have been given before [12].

For large α it is possible to find an asymptotic low temperature expansion [13]. A brief review is appropriate because our method involves a modification of this expansion. Rewriting (15) as a geometrical series and interchanging the order of summation and integration, we get

$$T(\alpha) = \frac{1}{(2\pi)^3} \sum_{m=1}^{\infty} \iiint_{-\pi}^{\pi} dx\, dy\, dz \exp[-m\alpha\{1 - (1/3)(\cos x + \cos y + \cos z)\}]. \tag{18}$$

This can be expressed as a product of modified Bessel functions by using the integral representation [11]

$$I_k(u) = \frac{1}{(2\pi)} \int_{-\pi}^{\pi} \cos kx\, e^{u \cos x}\, dx \tag{19}$$

to obtain[3]

$$T(\alpha) = \sum_{m=1}^{\infty} e^{-m\alpha} I_0^3(m\alpha/3), \qquad 0 < \alpha < \infty. \tag{20}$$

Using the following asymptotic form [11] of $I_0(u)$,

$$I_0(u) \approx [e^u/(2\pi u)^{1/2}][1 + (1/8u) + (9/128u^2) + \cdots] \tag{21}$$

(for u large), (20) may be expressed as

$$T(\alpha) \approx \sum_{m=1}^{\infty} \frac{1}{(2\pi m\alpha/3)^{3/2}} \left[1 + \frac{3}{8(m\alpha/3)} + \cdots\right] \tag{22}$$

$$= \left(\frac{3}{2\pi\alpha}\right)^{3/2} \left[\zeta(3/2) + \frac{9}{8\alpha}\, \zeta(5/2) + \cdots\right] \tag{23}$$

where

$$\zeta(p) = \sum_{m=1}^{\infty} 1/m^p \tag{24}$$

is the Riemann zeta function. This is an asymptotic series so that the results are useful only for large values of α.

[3] This expression appears to have been first developed by Tanaka and Glass (unpublished work) cited in S. H. CHARAP AND E. L. BOYD, *Phys. Rev.* *A*133 (1966), 811.

In order to get definitive results for smaller α, consider the following splitting procedure for (20):

$$T(\alpha) = \sum_{m=1}^{N-1} e^{-m\alpha} I_0^3(m\alpha/3) + (3/2\pi\alpha)^{3/2} [\zeta_N(3/2) + (9/8\alpha)\, \zeta_N(5/2) + \cdots], \qquad 0 < \alpha < \infty, \quad (25)$$

where $\zeta_N(p)$ is a truncated Riemann zeta function

$$\zeta_N(p) = \sum_{m=N}^{\infty} 1/m^p. \quad (26)$$

Due to the fact that

$$\zeta_N(p) \approx \zeta(p)/N^p, \quad (27)$$

for any α we can always find N large enough so that (25) converges in the same manner as (23) does when α is large. By trial and error we find that for six place accuracy N must satisfy the condition

$$N \gtrsim 24/\alpha. \quad (28)$$

With this method one can calculate $T(\alpha)$ for arbitrary α, though for $\alpha \leqslant 1$ it will be more efficient to use the high temperature series (16). For α from 1 to 20

TABLE I

Comparison of three methods for calculating $T(\alpha)$. Ten terms of high temperature series and ten terms of low temperature expansion were used.

α	Low temp. expansion (23)	Extended low temp. expansion (25)	High temp. expansion (16)
0.05		2.98319×10^{1}	2.98319×10^{1}
0.1		1.46722×10^{1}	1.46722×10^{1}
0.5		2.57418	2.57418
1.0		1.09773	1.09773
3.0		2.15227×10^{-1}	2.14803×10^{-1}
5.0		9.13117×10^{-2}	
10.0		2.92608×10^{-2}	
15.0		1.54934×10^{-2}	
20.0	9.94334×10^{-3}	9.94337×10^{-3}	
25.0	7.06705×10^{-3}	7.06705×10^{-3}	
30.0	5.35277×10^{-3}	5.35277×10^{-3}	
60.0	1.87287×10^{-3}	1.87287×10^{-3}	

this method is particularly useful because it fills the gap left by the simple high and low temperature series (see Table I). In this connection we might note that Flax and Raich [3] have proposed a different method that appears to be weak for large α [14] or low temperatures; our present method may be regarded as an extended low temperature series while Flax and Raich [3] have basically extended the high temperature series.

IV. General Method for TWS and LGF for the Three Cubic Lattices

The evaluation of (1) and (2) proceeds in a similar way as for V and $T(\alpha)$. First consider the integral

$$Q_i(\alpha, \eta, l, m, n) = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} \cos lx \cos my \cos nz \exp[-\alpha[\eta - \gamma_i(x, y, z)]]\, dx\, dy\, dz. \quad (29)$$

This integral has an asymptotic (large α) expansion:

$$Q_i(\alpha, \eta, l, m, n) \approx \frac{e^{-\alpha(\eta-1)}}{\alpha^{3/2}} \sum_{k=0}^{K} \frac{a_{k,i}^{(l,m,n)}}{\alpha^k}, \quad (30)$$

where in view of the asymptotic nature of (30) K is chosen according to the precision required [15] (for six-figure accuracy we found 10 usually appropriate), and has a small α expansion.

$$Q_i(\alpha, \eta, l, m, n) = e^{-\alpha\eta} \sum_{k=0}^{\infty} \alpha^k b_{k,i}^{(l,m,n)}, \quad (31)$$

where $a_{k,i}^{(l,m,n)}$ and $b_{k,i}^{(l,m,n)}$ are independent of α and η. The coefficients $a_{k,i}^{(l,m,n)}$ can all be obtained from the following identities:

$$I_k(t) \approx e^t \sum_{l=0}^{K} \frac{C_l^k}{t^{l+1/2}}, \text{ [11]}, \quad (32)$$

$$\frac{1}{x^n} = \frac{1}{\Gamma(n)} \int_0^{\infty} t^{n-1} e^{-xt}\, dt, \quad (33)$$

$$\frac{1}{(x+y)^p} = \frac{1}{x^p} \sum_{k=0}^{\infty} \frac{\Gamma(k+p)}{\Gamma(p)\, k!} (-y/x)^p, \qquad y < x. \quad (34)$$

Making use of (19) and (32) yields for s.c.

$$Q_1(\alpha, \eta, l, m, n) \approx e^{-\alpha(\eta-1)} \sum_{ijk=0}^{K} C_i{}^l C_j{}^m C_k{}^n (3/\alpha)^{i+j+k+3/2}. \tag{35a}$$

Successive applications of (19) and (32)–(34) yield for bcc[4] and fcc

$$\begin{aligned} Q_2(\alpha, \eta, l, m, n) &\approx e^{-\alpha(\eta-1)} \sum_{ijkrs=0}^{K} C_i{}^l C_j{}^m C_k{}^n \\ &\times \frac{\Gamma(r+i+1/2)\,\Gamma(r+j+1/2)\,\Gamma(k+s+1/2)\,\Gamma(i+j+r+s+1)}{r!\,s!\,\Gamma(i+1/2)\,\Gamma(j+1/2)\,\Gamma(k+1/2)\,\Gamma(i+j+r+1)} \\ &\times (1/\alpha)^{i+j+k+r+s+3/2}, \end{aligned} \tag{35b}$$

$$\begin{aligned} Q_3(\alpha, \eta, l, m, n) &\approx e^{-\alpha(\eta-1)} \sum_{ijkrs=0}^{K} \sum_{p=0}^{2} C_i{}^l C_j{}^m C_k{}^n \\ &\times \frac{\Gamma(k+s+1/2)\,\Gamma(j+r+s-p+1/2)\,\Gamma(r+i+p+1/2)}{2^{r+s} p!\,(s-p)!\,\Gamma(i+1/2)\,\Gamma(j+1/2)\,\Gamma(k+1/2)} \\ &\times (3/2\alpha)^{i+j+k+r+s+3/2} \end{aligned} \tag{35c}$$

for α large.

The coefficients $b_{k,i}^{(l,m,n)}$ are obtained in a straightforward manner from (29) by expanding the exponential in a power series followed by term-by-term integration. Having calculated the coefficients $a_{k,i}^{(l,m,n)}$ and $b_{k,i}^{(l,m,n)}$ for a specific integral it is straightforward to use them to calculate this integral for all α and η using the splitting technique of Sections II and III.

In order to evaluate $T_i(\alpha, \eta, l, m, n)$ one makes use of the identity

$$\frac{1}{e^x - 1} = \sum_{j=1}^{\infty} e^{-jx} \tag{36}$$

to express (2) as

$$T_i(\alpha, \eta, l, m, n) = \sum_{j=1}^{\infty} Q_i(j\alpha, \eta, l, m, n). \tag{37}$$

[4] Joyce [8] has developed recurrence relationships for the $a_{k,2}^{(0,0,0)}$ which are much more economical to use than (35b); however, for many problems the general expressions we use (35a–c) are quite adequate.

Splitting this sum in a similar manner to (20) and using (30) and (31), (37) becomes

$$T_i(\alpha, \eta, l, m, n) = \sum_{j=1}^{N-1} e^{-j\alpha\eta} \sum_{k=0}^{\infty} (j\alpha)^k b_{k,i}^{(l,m,n)} + \frac{1}{\alpha^{3/2}} \sum_{k=0}^{K} \frac{a_{k,i}^{(l,m,n)}}{\alpha^k} \zeta_N(k + 3/2, \alpha(\eta - 1)), \tag{38}$$

where

$$\zeta_N(p, x) = \sum_{m=N}^{\infty} \frac{e^{-mx}}{m^p} \tag{39}$$

and N is given by (18) (see Appendix).

$W_i(\eta, l, m, n)$ can be expressed in terms of Q_i in the following manner (see (33)):

$$W_i(\eta, l, m, n) = \int_0^{\infty} Q_i(t, \eta, l, m, n)\, dt. \tag{40}$$

Splitting the domain of integration by introducing a parameter s (analogous to N in (38)) and using (30) and (31), we obtain

$$W_i(\eta, l, m, n) = \sum_{k=0}^{\infty} b_{k,i}^{(l,m,n)} \int_0^s e^{-t\eta} t^k\, dt + \sum_{k=0}^{K} a_{k,i}^{(l,m,n)} \int_s^{\infty} \frac{e^{-t(\eta-1)}}{t^{k+3/2}}\, dt. \tag{41}$$

In (38) and (41) the infinite sum in the first term has good convergence and corresponds to the power series expansion which would be used to evaluate the Bessel functions in the first terms of (13) and (25).

Conclusion

We have developed procedures for the precision evaluation of generalized TWS and generalized LGF for all the cubic lattices. Many applications to physical problems are now possible. Prior to this work there has been very little discussion of TWS, attention having been concentrated on LGF. Moreover, the concentration there has been on the sc lattice for restricted values of l, m, n. In going beyond the integrals discussed in this work one would probably be involved in more complicated dispersion functions such as those resulting from longer-ranged

interactions and then the expansions may become too tedious. For those cases, results to an accuracy better than 1 % may be achieved readily with a more direct numerical integration scheme such as that proposed by Loly and Huett [16].

APPENDIX

To use the method described in the preceding sections it is important to be able to calculate truncated Riemann zeta functions accurately. This may be achieved by the following rearrangements which overcome the slow convergence of (39) for small x.

$$\zeta_N(p, x) = \sum_{m=N}^{\infty} (e^{-mx}/m^p)$$

$$= \sum_{m=1}^{\infty} \sum_{l=1}^{N} \left[\frac{e^{-(2Nm-l)x}}{(2Nm-l)^p} + \frac{e^{-(2Nm+l-1)x}}{(2Nm+l-1)^p} \right]$$

$$= \sum_{m=1}^{\infty} \sum_{l=1}^{N} \sum_{r=0}^{\infty} \frac{e^{-2Nmx}}{(2Nm)^{p+r}} \frac{\Gamma(p+r)}{r!\,\Gamma(p)} [l^r e^{lx} + (1-l)^r e^{(1-l)x}]$$

$$= \frac{1}{N^p} \sum_{r=0}^{\infty} \frac{\zeta(p+r, 2Nx)}{2^{p+r}} \frac{\Gamma(p+r)}{r!\,\Gamma(p)} \left[\sum_{l=1}^{N} \left(\frac{l}{N}\right)^r e^{lx} + \left(\frac{1-l}{N}\right)^r e^{(1-l)x} \right].$$

We have finally a rapidly converging series expansion in terms of the usual Riemann zeta functions.

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C. ANTIFERROMAGNETIC INTEGRALS

From equation (4.A.36) for a C_sCl type lattice, we obtain the following integrals

$$R_1(\alpha) = \frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi}(1-\gamma_2^2)^{1/2}\coth\left[\frac{\alpha}{2}(1-\gamma_2^2)^{1/2}\right]dxdydz$$

$$R_2(\alpha) = \frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi}(1-\gamma_2^2)^{-1/2}\coth\left[\frac{\alpha}{2}(1-\gamma_2^2)^{1/2}\right]dxdydz \qquad (42)$$

$$R_3(\alpha) = \frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi}\frac{\alpha}{2\sinh\left[\frac{\alpha}{2}(1-\gamma_2^2)^{1/2}\right]}dxdydz$$

where γ_2 is given by equation (3).

We can evaluate these integrals by dividing them into two regions. For $\alpha < 2$ we use a high temperature expansion and for $\alpha > 2$ we will use the extended low temperature method developed earlier.

1) High Temperature Expansion

Noting that

$$R_3(\alpha) = -\alpha\frac{d}{d\alpha}R_2(\alpha) \qquad (43)$$

we see that the high temperature expansion for all three integrals follows from

$$\coth x = \frac{1}{x} + \sum_{k=1}^{\infty}\frac{(-1)^{k-1}2^{2k}B_k x^{2k-1}}{(2k-1)!!\,k!\,2^k}, \quad x < \pi$$

$$= \frac{1}{x} + \sum_{k=1}^{\infty} C_{k-1} (-1)^{k-1} x^{2k-1} \tag{44}$$

where B_k = Bernoulli's numbers, and C_k is defined in table 2. We will also need the coefficients defined by the following

$$b_k = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} (1-\gamma_2^2)^{(k-1)/2} \, dx\,dy\,dz \tag{45}$$

where

$$b_{-1} = \omega = 1.393204$$

$$b_0 = 1+c' = 1.118636$$

$$b_1 = 1$$

$$b_2 = 1-c = 1. - .073038 \,.$$

The remaining b_k ($k>2$) can be calculated by expanding the integrand in a power series followed by term by term integration.

The high temperature expansions are

$$R_1(\alpha) = \frac{1}{\alpha} + \frac{1}{2} \sum_{k=1}^{\infty} C_{k-1} b_{2k} (-1)^{k-1} \left(\frac{\alpha}{2}\right)^{2k-1}$$

$$R_2(\alpha) = \frac{\omega}{\alpha} + \frac{1}{2} \sum_{k=1}^{\infty} C_{k-1} b_{2k-2} (-1)^{k-1} \left(\frac{\alpha}{2}\right)^{2k-1} \tag{46}$$

$$R_3(\alpha) = \frac{\omega}{\alpha} - \frac{1}{2}\sum_{k=1}^{\infty} c_{k-1} b_{2k-2} (2k-1)(-1)^{k-1} \left(\frac{\alpha}{2}\right)^{2k-1}.$$

2) Extended Low Temperature Expansion

The low temperature expansion uses a different expression for the hyperbolic cotangent namely

$$\coth x = 1 + 2\sum_{m=1}^{\infty} e^{-2xm} \tag{47}$$

and we also need an integral of the form

$$\Theta(\alpha) = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} (1-\gamma_2^2)^{1/2} e^{-\alpha(1-\gamma_2^2)^{1/2}} dx\,dy\,dz. \tag{48}$$

Using these results we obtain

$$R_1(\alpha) = 1 - c + 2\sum_{m=1}^{\infty} \frac{d^2}{(d\alpha)^2} \Theta(m\alpha) \tag{49}$$

$$R_2(\alpha) = 1 + c' + 2\sum_{m=1}^{\infty} \Theta(m\alpha)$$

$$R_3(\alpha) = -\alpha \frac{d}{d\alpha} R_2(\alpha).$$

It is easily seen that $\Theta(\alpha)$ has a direct expansion in terms of the b_k defined earlier

$$\Theta(\alpha) = \sum_{k=0}^{\infty} \alpha^k b_k (-1)^k / k! \tag{50}$$

and in section D we show that $\Theta(\alpha)$ has the asympotic expansion

$$\Theta(\alpha) \sim \frac{4}{\pi^2 \alpha^2} \sum_{k=0}^{K} a_k / \alpha^{2k} \quad . \tag{51}$$

Dividing the sum in equation (48) into two parts

$$\sum_{m=1}^{\infty} = \sum_{m=1}^{N-1} + \sum_{m=N}^{\infty} \tag{52}$$

and using the direct expansion for small values of m, and the asymptotic expansion for large m yields

$$R_1(\alpha) = 1 - c + 2 \sum_{k=1}^{\infty} \left[\frac{(-1)^k k(k-1) b_k \alpha^{k-2}}{k!} \left(\sum_{m=1}^{N-1} m^{k-2} \right) \right]$$
$$+ \frac{8}{\pi^2 \alpha^4} \sum_{k=0}^{K} \frac{\zeta_N(4+2k)(2k+2)(2k+3) a_k}{\alpha^{2k}}$$

$$R_2(\alpha) = 1 + c' + 2 \sum_{k=0}^{\infty} \left[\frac{(-1)^k \alpha^k b_k}{k!} \left(\sum_{m=1}^{N-1} m^k \right) \right]$$
$$+ \frac{8}{\pi^2 \alpha^2} \sum_{k=0}^{K} \frac{\zeta_N(2+2k)\, a_k}{\alpha^{2k}} \tag{53}$$

$$R_3(\alpha) = - 2 \sum_{k=0}^{\infty} \left[\frac{(-1)^k k \alpha^k b_k}{k!} \left(\sum_{m=1}^{N-1} m^k \right) \right]$$
$$+ \frac{8}{\pi^2 \alpha^2} \sum_{k=0}^{K} \frac{\zeta_N(2+2k)(2k+2) a_k}{\alpha^{2k}}$$

where $\zeta_N(k)$ is a truncated Zeta function given by

$$\zeta_N(k) = \sum_{m=N}^{\infty} 1/m^k \quad . \tag{54}$$

In order to obtain a precision of five decimals, the following conditions are necessary. First, we used 66 coefficients in the direct expansion and evaluated all except b_o using double precision (14 figures). Second, we obtained 10 coefficients in the asymptotic expansion ($K = 10$) using single precision. Third, the value of N was determined by

$$N = 19/\alpha + 1 \tag{55}$$

i.e. we assigned the integer closest to, but less than $19/\alpha + 1$. Under these conditions the method worked well for $\alpha \geq 2$.

D. ASYMPTOTIC EXPANSION OF $Q(\alpha)$

We are interested in an asymptotic expansion for the integral

$$Q(\alpha) = \frac{1}{(2\pi)^3} \iiint_{-\pi}^{\pi} dx\,dy\,dz\,(1-\gamma_2^2)^{-1/2}\, e^{-\alpha(1-\gamma_2^2)^{1/2}} \tag{56}$$

where

$$\gamma_2 = \cos x \cos y \cos z \quad .$$

Starting from the transform

$$\frac{e^{-a\sqrt{s}}}{\sqrt{s}} = \frac{2}{\sqrt{\pi}} \int_o^{\infty} dt\, e^{-a^2/4t^2}\, e^{-st^2}, \qquad s > 0 \tag{57}$$

and substituting

$$a = 1$$

$$s = (1-\gamma_2^2)\alpha^2$$

we obtain

$$Q(\alpha) = \frac{2\alpha}{\sqrt{\pi}} \iiint_{-\pi}^{\pi} \frac{dx\,dy\,dz}{(2\pi)^3} \int_0^\infty dt\, e^{-1/4t^2} e^{-\alpha^2(1-\gamma_2^2)t^2} \quad (58)$$

Next using another transform of the form

$$e^{-b^2/4c^2} = \frac{2c}{\sqrt{\pi}} \int_0^\infty du\, e^{-c^2u^2} \cosh(bu) \quad (59)$$

with

$$b = \gamma_2 \alpha t$$

$$c = 1/2$$

we get

$$Q(\alpha) = \frac{2\alpha}{\pi} \iiint_{-\pi}^{\pi} \frac{dx\,dy\,dz}{(2\pi)^3} \int_0^\infty dt \int_0^\infty du \quad (60)$$

$$\times e^{-1/4t^2} e^{-\alpha^2 t^2} e^{-u^2/4} \cosh(\gamma_2 \alpha t u).$$

Since γ_2 ranges over positive and negative values we obtain the identity

$$\frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi} dx\,dy\,dz\,\cosh(\gamma_2\alpha tu) \tag{61}$$

$$= \frac{1}{(2\pi)^3}\iiint_{-\pi}^{\pi} dx\,dy\,dz\,e^{-\gamma_2\alpha tu} = \Theta_2(\alpha tu,0,0,0,0)$$

where $\Theta_2(\alpha,\eta,\ell,m,n)$ has been introduced in section B. Finally we obtain the integral representation

$$\Theta(\alpha) = \frac{2\alpha}{\pi}\int_0^\infty dt\int_0^\infty du\;\Theta(\alpha tu,0,0,0,0) \tag{62}$$

$$\times\; e^{-1/4t^2}\,e^{-\alpha^2t^2}\,e^{-u^2/4}\,.$$

The major contribution to the integrand occurs for $t=1$ and $u=2\alpha$ so that for large α the asymptotic representation of $\Theta_2(\alpha tu,0,0,0,0)$ is appropriate. From equation (30)

$$\Theta_2(\alpha tu,0,0,0,0) \sim \frac{e^{\alpha tu}}{(\alpha tu)^{3/2}}\sum_{k=0}^{K}\frac{a_{k,2}^{(0,0,0)}}{(\alpha tu)^k} \tag{63}$$

we get

$$Q(\alpha) = \frac{2\alpha}{\pi}\int_0^\infty dt \int_0^\infty du\; e^{-1/4t^2} e^{-\alpha t^2} e^{-u^2/4} e^{\alpha t u}$$

$$\times \sum_{k=0}^{K} \frac{a_{2,k}^{(0,0,0)}}{(\alpha t u)^{3/2+k}} \qquad (64)$$

Defining new variables

$$z = \frac{u}{2} - \alpha t$$

so that

$$-\alpha t < z < \infty$$

leaves

$$Q(\alpha) = \frac{2\alpha}{\pi}\int_0^\infty dt \int_{-\alpha t}^\infty dz\; e^{-1/4t^2} e^{-z^2}$$

$$\times \sum_{k=0}^{K} \frac{a_{k,2}^{(0,0,0)}}{(\alpha t)^{k+3/2}(2z+2\alpha t)^{k+3/2}} \qquad (65)$$

The major contribution occurs for $z=0$ so that equation (34) is appropriate. We have

$$\frac{1}{(z+\alpha t)^{k+3/2}} = \frac{1}{(\alpha t)^{k+3/2}} \qquad (66)$$

$$\times \sum_{m=0}^{\infty} \frac{\Gamma(m+k+3/2)}{\Gamma(k+3/2)\, m!}\left(-\frac{z}{\alpha t}\right)^m, \quad z<\alpha t$$

and neglecting an exponentially small amount gives

$$\Theta(\alpha) = \frac{2\alpha}{\pi} \sum_{k=0}^{K} \sum_{m=0}^{\infty} \frac{a_{k,2}^{(0,0,0)} \Gamma(m+k+3/2)}{\Gamma(k+3/2)\, m!\, \alpha^{2k+m+3}\, 2^{k+3/2}} \tag{67}$$

$$\times \int_0^{\infty} dt\, \frac{e^{-1/4t^2}}{t^{m+2k+3}} \int_{-\alpha t}^{\alpha t} dz\, e^{-z^2} (-z)^m .$$

For large α we can effectively replace the limits of z by ∞ and $-\infty$. In this form the integrals are now separable and have closed form solutions.

Performing the integrations finally leads us to the result

$$\Theta(\alpha) \sim \sum_{m=0}^{\infty} \sum_{k=0}^{K} \frac{2^{m+k+7/2}\, \Gamma(2m+k+3/2)\, (2m-1)!!\, (m+k)!\, a_{k,2}^{(0,0,0)}}{(\pi)^{1/2}\, \alpha^{2+2m+2k}\, \Gamma(k+3/2)\, (2m)!}$$

$$= \frac{4}{\pi^2 \alpha^2} \sum_{k=0}^{K} a_k / \alpha^{2k} \tag{68}$$

$$= \frac{4}{\pi^2 \alpha^2} \left[1 + 6/\alpha^2 + 116/\alpha^4 + \cdots \right]$$

where we have used

$$a_{k,2}^{(0,0,0)} = \frac{1}{(2\pi)^{3/2}} , \qquad k = 1$$

$$= \frac{1}{(2\pi)^{3/2}} \frac{9}{8} , \qquad k = 2$$

$$= \frac{1}{(2\pi)^{3/2}} \frac{281}{128} , \qquad k = 3 .$$

The above expression allows us to calculate 10 coefficients in the asymptotic series for (56). In this form, the expression can be generalized to other lattices and integrals (those involving products of cosines) by replacing $a_{k,2}^{(0,0,0)}$ by $a_{k,i}^{(l,m,n)}$.

The coefficients given here agree with those calculated by Oguchi (1960), who calculated three terms, for both the $CsCl$ and $NaCl$ type lattices.

TABLE 2

Some of the coefficients used to calculate $R_i(\alpha)$.

k	a_k	b_k	c_k
0	1.0	1.118635	.333333
1	6.0	1.0	$.222222\ 10^{-1}$
2	116.0	.926962	$.211640\ 10^{-2}$
3	$4.70399\ 10^{3}$	.875	$.211640\ 10^{-3}$
4	$3.27888\ 10^{5}$	.835026	$.213778\ 10^{-4}$
5	$3.50222\ 10^{7}$	.802734	$.216440\ 10^{-5}$
6	$5.31937\ 10^{9}$	.775764	$.219260\ 10^{-6}$
7	$1.09012\ 10^{12}$	.752686	$.222146\ 10^{-7}$
8	$2.89928\ 10^{14}$	.732570	$.225079\ 10^{-8}$
9	$9.71168\ 10^{16}$	.714780	$.228052\ 10^{-9}$

$$Q(\alpha) \sim \frac{4}{\pi^2 \alpha^2} \sum_{k=0}^{K} a_k / \alpha^{2k}$$

$$Q(\alpha) = \sum_{k=0}^{\infty} \frac{(-\alpha)^k b_k}{k!}$$

$$c_k = \frac{2^{k+1} B_{k+1}}{(2k+1)!!\,(k+1)!}$$