# SPIN WAVE PROPERTIES OF THE CLASSICAL
# HEISENBERG ANTIFERROMAGNET

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by

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To Bohunka, My Wife

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ABSTRACT

We have studied the antiferromagnetic sublattice magnetization for large spin region by means of the conventional formulation of spin wave theory. Spin wave interactions have been taken into account within the Micheline Bloch approximation which gives the leading interaction effects. The procedure adopted in the classical limit ($S \to \infty$) follows closely the recent work on the analogous ferromagnetic problem by Loly. The results simplify considerably as in the similar ferromagnetic case and the leading terms have the same temperature dependence for the magnetization, which is in contrast to the results for small spin.

CHAPTER ONE

## Introduction to the Heisenberg Model of Magnetism

We investigate a physical system of interacting spins occupying sites of a cubic crystal lattice.

The strong magnetism of solids is due to spins of partly filled inner electronic shells of atoms. We shall assume that both an interaction between the electrons and orbital moments of the electrons are negligible. The energy of such a system is described by the exchange Heisenberg Hamiltonian [1-3]

$$H = -\sum_{\langle \ell, m \rangle} 2 J_{\ell m} \vec{S}_\ell \cdot \vec{S}_m \, ,$$   (1.1)

where $J_{\ell m}$ is exchange integral (often the definition is also $J_{\ell m} = J'_{\ell m}/2$) between $\vec{n}_\ell$ and $\vec{n}_m$ sites and $\vec{S}_\ell$ and $\vec{S}_m$ are spin operators on these lattice sites. The spin operators of each site are only dynamical variables in (1.1) since the distances between spins are known.

Neel [4] first expressed the possibility that the negative sign of the exchange integral could lead to a state in which different sublattices of spins in a crystal may align themselves antiparallel. Such ordered magnetic arrangement of sublattices, each possessing uniform magnetization, was later called antiferromagnetism.

Here we are interested in the character of energy spectrum near to the ground state of magnetic system. For

simplicity we shall assume that there are only two equivalent
sublattices, one inside the other, representing an isotropic
crystal. Such an antiferromagnetic state was taken as a first
reasonable approximation in Bethe's [5] and later in Hulthen's [6]
calculation of the exact ground state for one dimensional anti-
ferromagnet. However, in the three-dimensional antiferromagnet,
no exact theory exists at $T = 0^{\circ} K$ (ground state).

Fortunately we have spin wave theory, which is one of the
most useful schemes for approximating the character of energy
spectrum near to the ground state. The elementary excitations
of an antiferromagnetic spin system from an ordered array of
alternately pointing spins, which is assumed to be a reasonable
approximation of the ground state, have a wavelike form and
are called spin waves.

The classical dispersion law is satisfactory for spin
waves, because the quantum and classical equations of motion
are identical; as one can prove by means of the commutation
rules for spin operators. One can prove, also, that in
equilibrium the energy of the antiferromagnet should be
minimum. Such antiferromagnetic ground state configurations [7-12]
are governed by the signs and values of the exchange integrals
in various environments and geometries of the crystal lattice.
There are only two possible 2-sublattice arrangements for cubic
crystal lattices, namely the NaCl (simple cubic) and CsCl
(body centred cubic) structures; face centred cubic magnetic

lattice cannot be represented as an assembly of two identical cubic lattices with opposite spins [7,8] . The sublattices of the NaCl and CsCl structure have symmetry of f.c.c. and s.c. lattice,* respectively.

The lowest energy state of an antiferromagnet must be a singlet state [13] , and thus have no directional character, that is, have total $S$ equal to zero. However, the direction of the resulting antiparallel magnetic moments of the two sublattices is arbitrary; so that the ground state is degenerate. This degeneracy cannot be removed by an external field. For a not too large external field, the spins would arrange themselves fully in a plane perpendicular to the external field, but the direction of the spins in the plane would still be arbitrary [6,14] .

In any real antiferromagnetic crystal this degeneracy is removed by the real anisotropy. An anisotropy can be introduced in the form of an effective anisotropy field such that the spins on sublattice 1 are preferentially oriented in the $+z$ direction, say, and those on sublattice 2 in the $-z$ direction by introducing a hypothetical field $h_1$ and $h_2$, respectively.

The Hamiltonian (1.1) then becomes

$$ H_s = 2|J| \sum_{\langle l,m \rangle} \vec{S}_l \cdot \vec{S}_m + h_1 \sum_l S_l^z + h_2 \sum_m S_m^z , \qquad (1.2) $$

where $l$ runs over all the atoms on sublattice 1, and $m$ over those on sublattice 2; $J$ is the negative constant of the isotropic antiferromagnetic interaction.

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* See Appendix 3

For finite values of $h_1$ and $h_2$ the ground state of (1.2) turns out to be only slightly dependent on these values as long as $h_1$ is not smaller than of order $\frac{|J|}{N}$ , where $N$ is the total number of atoms in the crystal [15].

We accordingly define the antiferromagnetic ground state of (1.1) corresponding to the assumed two sublattice structure, as the limiting ground state of (1.2) for first $N$ tending to infinity and then $h_1$ and $h_2$ tending to zero. This does not differ from the completely ordered state obtained by letting $h_1$ and $h_2$ tending to infinity [15]. Such a completely ordered state becomes identical to that of the Ising approximation [16-18] and it is used as the initial state in the spin wave theory of antiferromagnets.

The antiferromagnetic spin waves are introduced to describe the small deviations of the state of the spin system from this completely ordered state. The most fruitful theoretical approaches have involved the use of boson models for spin waves. The first such theory was presented by Holstein and Primakoff [19] in 1940 for the ferro case and applied to antiferromagnetism in 1952 by Kubo [20], who calculated some thermodynamic quantities and ground state for anti-ferromagnetic cubic lattice. Anderson [21], who presented a similar but semiclassical theory of antiferromagnet, carefully examined the zero point energy and showed that ground state lies between the limits $N|J|z S^2$ and $N|J|z S^2 (1 + \frac{1}{zS})$

predicted by his variational method. Davis [22] used
perturbation expansion for the ground state energy where the
off diagonal part of antiferromagnetic exchange Hamiltonian is
treated as perturbation--this was also evaluated using a
diagram technique by Boon [23]. This treatment gives the spin
wave ground state about 93% completely ordered, i.e. the
sublattice $x$-component of spin is 93% of its maximum value.

One of the characteristics of the spin wave approximation
is the fact that at sufficiently low temperatures the influence
of the interaction between the spin deviations may be neglected.
Dyson [24] invented for the ferromagnetic case a general theory
of spin wave interactions. He defines two kinds of interactions:
One is the kinematical interaction which arises from the fact
that more than $2S+1$ units of reversed spin cannot be attached
to the same atom simultaneously, where $S$ is the magnitude of
atomic spin in units of $\hbar$. The other is the dynamical inter-
action which represents the nondiagonal part of Hamiltonian in
its basic set of states.

Since the boson approach replaces the $2S+1$ states of
each spin by a system with an infinite number of states in a
form of quantum harmonic oscillator whose lowest $2S+1$ states
correspond exactly to those of the spin, the rest of the states
are spurious states causing the kinematic interaction. Dyson
has shown that the kinematic effect was negligible for $T \ll T_c$
and gave a low temperature result which is assymptotically

exact as $T \rightarrow 0^{\circ}K$.

Oguchi [25] used the Holstein-Primakoff transformation of spin operators to bosons and his result was in agreement with a significant part of Dyson's result, correct to order $\frac{1}{S}$ as was recently shown [26] in case of magnetization. Oguchi also used similar procedure for antiferromagnet.

The difficulty of estimating the kinematic effect can be removed in a special case investigated just recently by Loly [27] for ferromagnetic problems and furthermore the bound states, whose effect is also difficult to estimate [28], are absent in this case--this is the case of infinite spin or classical Heisenberg model. Loly has improved results of some authors [28,29] for large $S$ case and achieved a considerable extension of the result given by Heller and Kramers [30].

Present work deals with similar problem for antiferromagnetic cubic crystals along the lines of Oguchi's treatment for anti-ferromagnet but using the self consistently renormalised form of spin wave energy [31] for thermodynamic calculation.

## Antiferromagnetic Spin Wave Hamiltonian and Magnetization

After the common redefinition of the spin operator $\vec{S}_j \equiv (S_j^x, S_j^y, S_j^z)$ into the so-called projection form, $S_j \equiv (S_j^+, S_j^-, S_j^z)$, where $S_j^+ = S_j^x + i\, S_j^y$ and $S_j^- = S_j^x - i\, S_j^y$ the spin Hamiltonian (1.2) becomes

$$H_S = 2|J| \sum_{\langle \ell, m \rangle} \left[ \tfrac{1}{2} ( S_\ell^+ S_m^- + S_\ell^- S_m^+ ) + S_\ell^z S_m^z \right] +$$
$$+ h_1 \sum_\ell S_\ell^z + h_2 \sum_m S_m^z . \qquad (2.1)$$

Following Holstein and Primakoff [19], we express the atomic spin operators of the sublattice 1 in the form

$$S_\ell^+ = (2S)^{\frac{1}{2}} f_\ell(S)\, a_\ell \;,\;\; S_\ell^- = (2S)^{\frac{1}{2}} a_\ell^+ f_\ell(S) \;,\;\; S_\ell^z = S - m_\ell \;, \qquad (2.2)$$

where the operator $f_\ell(S)$ is defined as follows $f_\ell(S) = \left( 1 - \frac{m_\ell}{2S} \right)^{\frac{1}{2}}$, $m_\ell = a_\ell^+ a_\ell$ is a spin deviation operator of $S_\ell^z$ component from its maximal value $S$ along the $+z$ axis in units of $\hbar$. $m_\ell$ is the number operator with allowed values $0, 1, 2, \ldots$ .

Let us define correspondingly the spin deviation operator for the sublattice 2, as follows $m_m = S - (-S_m^z)$.

Since we define $m_m$ as a spin deviation operator of $S_m^z$ component from its maximal value $S$ along the $-z$ axis, the projection spin components for the sublattice 2 become

$$S_m^+ = (2S)^{\frac{1}{2}} b_m^+ f_m(S), \qquad S_m^- = (2S)^{\frac{1}{2}} f_m(S) b_m \qquad (2.3)$$

$$S_m^z = -S + n_m ,$$

where similarly as in the case of sublattice 1 the operator $f_m(S)$ is defined by

$$f_m(S) = (1 - \frac{n_m}{2S})^{\frac{1}{2}}, \qquad n_m = b_m^+ b_m .$$

The integers $0, 1, 2, \ldots 2S$ are eigen-values of the spin deviation operators $n_\ell = S - S_\ell^z$ and $n_m = S + S_m^z$ .

Since we are dealing with two sublattices we have formed two independent boson spaces each corresponding to one sublattice.

An alternative approach is the Dyson-Maleev method, which is conceptually difficult, but has certain advantages from more rigorous treatment. As far as we are concerned for the present purpose we will use the Holstein-Primakoff transformation.

The operators $a^+$, $b^+$ and $a, b$ are defined by the commutation rules

$$[a_i, a_\ell^+] = \delta_{i\ell}, \qquad [b_j, b_m^+] = \delta_{jm},$$

$$[a_i, b_m^+] = [a_i, b_m] = [a_i^+, b_m] = [a_i^+, b_m^+] = 0$$

( $\delta_{jm}$ and $\delta_{i\ell}$ are Kronecker's deltas )

and are to be regarded as the creation and annihilation operators of the spin deviation, correspondingly.

By using these Bose operators the Hamiltonian (2.1) can be written as follows

$$H = H^{(0)} + 2z|J|S \left[ (1-h'_1) \sum_{\ell}^{N/2} a^+_\ell a_\ell + (1+h'_2) \sum_{m}^{N/2} b^+_m b_m \right] +$$

$$+ 2|J|S \sum_{\langle \ell, m \rangle} \left[ f_\ell f_m a_\ell b_m + a^+_\ell b^+_m f_\ell f_m - \frac{1}{S} a^+_\ell a_\ell b^+_m b_m \right], \tag{2.5}$$

where

$$H^{(0)} = -Nz|J|S^2(1 - h'_1 + h'_2) , \quad h'_1 = \frac{h_1}{2z|J|S} , \quad h'_2 = \frac{h_2}{2z|J|S} \tag{2.6}$$

and $z$ is number of nearest neighbors. At the next step we use binomial expansion for $f(S)$

$$f(S) = 1 - \frac{1}{4} \frac{m}{S} - \frac{1}{32} \frac{m^2}{S^2} - \frac{1}{128} \frac{m^3}{S^3} - \dots . \tag{2.7}$$

Then the Hamiltonian has the form

$$H = H^{(0)} + H^{(1)} + H^{(2)} \tag{2.8}$$

where terms with 6 and more boson operators are missing or in different words we have picked up all terms to the order $\frac{1}{S}$.

So

$$H^{(1)} = 2z|J|S \left[ \sum_{\ell} (1-h'_1) a^+_\ell a_\ell + \sum_{\ell+g} (1+h'_2) b^+_{\ell+g} b_{\ell+g} + \right.$$

$$\left. + \frac{1}{z} \sum_{\ell, g} (a_\ell b_{\ell+g} + a^+_\ell b^+_{\ell+g}) \right], \tag{2.9}$$

$$H^{(2)} = -\frac{|J|}{2} \sum_{\ell, g} \left( b^+_{\ell+g} b_{\ell+g} a_\ell b_{\ell+g} + a^+_\ell b^+_{\ell+g} b^+_{\ell+g} b_{\ell+g} + \right.$$

$$\left. + a^+_\ell a_\ell a_\ell b_{\ell+g} + a^+_\ell b_{\ell+g} a^+_\ell a_\ell + 4 a^+_\ell a_\ell b^+_{\ell+g} b_{\ell+g} \right), \tag{2.10}$$

$\ell$ and $g$ indicates the position vectors of the sublattice 1, $\vec{n}_\ell$, and all their nearest neighbors on the sublattice 2, $\vec{n}_g$, respectively; so symbolically $\vec{\ell} + \vec{g} = \vec{m}$.

Because of the translational symmetry of the sublattices,

it is convenient to make a Fourier transformation from the operators $a_{\vec{\ell}}^+, a_{\vec{\ell}}, b_{\vec{m}}^+, b_{\vec{m}}$ to the spin wave variables $a_{\vec{k}}^+, a_{\vec{k}}, b_{\vec{k}}^+, b_{\vec{k}}$ defined by

$$a_k^+ = \left(\tfrac{2}{N}\right)^{\frac{1}{2}} \sum_{\ell}^{N/2} e^{-ik\ell} a_\ell^+ \quad , \qquad a_k = \left(\tfrac{2}{N}\right)^{\frac{1}{2}} \sum_{\ell}^{N/2} e^{ik\ell} a_\ell ,$$

$$b_k^+ = \left(\tfrac{2}{N}\right)^{\frac{1}{2}} \sum_{m}^{N/2} e^{ikm} b_m^+ \quad , \qquad b_k = \left(\tfrac{2}{N}\right)^{\frac{1}{2}} \sum_{m}^{N/2} e^{-ikm} b_m , \tag{2.11}$$

where we have left out the vector sign for both lattice vectors $\vec{\ell}, \vec{m}$ and reciprocal lattice vector $\vec{k}$ from the first Brillouin zone.

Commutation rules for these new operators are bosonlike* as follows

$$[a_k, a_{k'}^+] = [b_k, b_{k'}^+] = \delta(k - k')$$

$$[a_k, b_{k'}^+] = [a_k, b_{k'}] = [a_k^+, b_{k'}] = [a_k^+, b_{k'}^+] = 0 . \tag{2.12}$$

Then

$$H^{(1)} = 2z|J|S \sum_k \Big[(1 - h_1') a_k^+ a_k + (1 + h_2') b_k^+ b_k + \\ + \gamma_k (a_k b_k + a_k^+ b_k^+)\Big] \tag{2.13}$$

and

$$H^{(2)} = -\frac{|J|z}{4N} \sum_{k_1 k_2 k_3 k_4} \Big[ \gamma(k_3) \delta(k_2 + k_4 - k_1 - k_3) b_{k_1} b_{k_2} b_{k_3} b_{k_4} + \\ + \gamma(k_1) \delta(k_1 - k_2 - k_3 + k_4) a_{k_1}^+ b_{k_2}^+ b_{k_3}^+ b_{k_4} + \\ + \gamma(k_2) \delta(k_1 - k_2 + k_3 - k_4) a_{k_1}^+ b_{k_2}^+ a_{k_3}^+ a_{k_4} + \\ + \gamma(k_4) \delta(k_1 - k_2 - k_3 + k_4) a_{k_1}^+ a_{k_2} a_{k_3} b_{k_4} + \\ + 4 \gamma(k_1 - k_2) \delta(k_1 - k_2 - k_3 + k_4) a_{k_1}^+ a_{k_2} b_{k_3}^+ b_{k_4} \Big] , \tag{2.14}$$

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*Proof in Appendix 1

where

$$\gamma^\wedge(\kappa) = \gamma_k = \frac{1}{z} \sum_\rho^z e^{ik\rho} \quad .$$

If the crystal in consideration has a center of symmetry, then

$$\gamma(\kappa) = \gamma(-\kappa) \qquad .$$

To carry out the diagonalization of the Hamiltonian, i.e. to get rid of such terms as $a_k b_{k_1}, ..., b_{k_1}^+ b_{k_2} a_{k_3} b_{k_4}^+, ...$ --terms where the two boson subspaces are mixing, we transform to new boson operators $\alpha_k^+, \alpha_k, \beta_k^+, \beta_k$ such that the boson commutation rules remain similar for the new operators, i.e.

$$[\alpha_k, \alpha_{k'}^+] = [\beta_k, \beta_{k'}^+] = \delta(\kappa - \kappa')$$

$$[\alpha_k, \beta_{k'}] = [\alpha_k^+, \beta_{k'}^+] = [\alpha_k, \beta_{k'}^+] = [\alpha_k^+, \beta_{k'}] = 0 \quad . \tag{2.15}$$

The new operators are defined by a transformation often called Bogolyubov [29] transformation:

$$a_k^+ = \alpha_k^+ u - \beta_k v \qquad\qquad a_k = \alpha_k u - \beta_k^+ v$$

$$b_k^+ = -\alpha_k v + \beta_k^+ u \qquad\qquad b_k = -\alpha_k^+ v + \beta_k u \tag{2.16}$$

where $u$ and $v$ are certain functions of the wave vector $\vec{k}$ to be defined*.

Then the Hamiltonian (2.1) can be written* as

$$H = H_o + \sum_k ( A_k^- m_k + A_k^+ m_k' ) +$$

$$+ \sum_{k_1 k_2} [ B^{(1)}( m_1 m_2 + m_1' m_2' ) + B^{(2)} m_1 m_2' ] \quad , \tag{2.17}$$

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*See Appendix 1

where $n_k = \alpha_k^+ \alpha_k$ and $n'_k = \beta_k^+ \beta_k$ are occupation number operators in the first subspace and second subspace, respectively;

$$H_0 = 2z|J|S\left\{-\frac{1}{2}SN(2D-1) - \sum_k [(D^2 - \gamma_k^2)^{\frac{1}{2}} - D] - \right.$$
$$\left. - \frac{1}{2SN}\left[\sum_k \left(\frac{D - \gamma_k^2}{(D^2 - \gamma_k^2)^{\frac{1}{2}}} - 1\right)\right]^2\right\}$$

(2.18)

and

$$D = 1 - \frac{h'_1}{2} + \frac{h'_2}{2} \; ;$$

(2.19)

the coefficients in the free boson part of Hamiltonian are

$$A_k^{\pm} = 2z|J|S\left[(D^2 - \gamma_k^2)^{\frac{1}{2}} \pm \frac{1}{2}(h'_1 + h'_2) - \right.$$
$$\left. - \frac{1}{SN}\frac{D - \gamma_k^2}{(D^2 - \gamma_k^2)^{\frac{1}{2}}}\sum_k \left(\frac{D - \gamma_k^2}{(D^2 - \gamma_k^2)^{\frac{1}{2}}} - 1\right)\right];$$

(2.20)

two body interactions are described by the coefficients $B^{(1)}$ and $B^{(2)}$ where we have taken on account only diagonal elements, which is sufficient for the renormalization of energy*;

$$B^{(1)} = -\frac{z|J|}{N}\left(\frac{D - \gamma_{k_1}^2}{(D^2 - \gamma_{k_1}^2)^{\frac{1}{2}}}\frac{D - \gamma_{k_2}^2}{(D^2 - \gamma_{k_2}^2)^{\frac{1}{2}}} - 1\right),$$

(2.21)

$$B^{(2)} = -\frac{2z|J|}{N}\left(\frac{D - \gamma_{k_1}^2}{(D^2 - \gamma_{k_1}^2)^{\frac{1}{2}}}\frac{D - \gamma_{k_2}^2}{(D^2 - \gamma_{k_2}^2)^{\frac{1}{2}}} + 1\right). \;**$$

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*See next chapter

**In Appendix 1 we illustrate in a more detailed example how one can transform Hamiltonian (2.9) to the form of (2.17).

Using the above results we will derive an expression for the antiferromagnetic reduced sublattice magnetization in the case of $h_1 = h_2 = 0$ , defined as quantum statistical average

$$m(T) = \langle \frac{S^z_{sub}}{N/2\,S} \rangle \qquad (2.22)$$

where spin z-component of the first sublattice is

$$S^z_{sub} \equiv \sum_\ell S^z_\ell = \frac{N}{2} S - \sum_\ell a^+_\ell a_\ell \quad . \qquad (2.23)$$

After transformation of $S^z_{sub}$ into new boson variables and neglecting two nondiagonal terms in which the new boson subspaces are mixing and will vanish under the thermal trace we get for (2.23) formula

$$S^z_{sub} = \frac{N}{2} S - \sum_k \left[ u^2(k)\, \alpha^+_k \alpha_k + v^2(k)\, \beta^+_k \beta_k + v^2(k) \right] \quad . \qquad (2.24)$$

If we substitute into the last equation the expressions $u(k)$ and $v(k)$ derived in Appendix 1, we obtain for the reduced sublattice magnetization

$$m(T) = 1 - \frac{c'}{2S} - \frac{2}{SN} \sum_k \langle n_k \rangle (1 - \gamma^2_k)^{-\frac{1}{2}}, \qquad (2.25)$$

where

$$c' = \frac{2}{N} \sum_k \left[ (1 - \gamma^2_k)^{-\frac{1}{2}} - 1 \right] \quad .$$

The characteristic feature of this theory is the fact that the sublattice magnetic moment at $T = 0^\circ K$ is not equal to the magnitude $\frac{N}{2} S$.

# CHAPTER THREE

## Spin Wave Renormalization in Antiferromagnet

Keeping the Heisenberg Hamiltonian up to the diagonal term of fourth order in spin wave operators, (2.17), the temperature dependence of the spin wave energy, together with the sublattice magnetization, can be studied by a self-consistent treatment.

This approximation has been found very useful in interpreting experimental data [33-35] for those magnetic insulators which seem to be fairly well described by the Heisenberg model. In this formalism the dispersion relation appears to have the same form as for free spin waves, apart from a temperature-dependent renormalization factor $\alpha(T)$. It was found [31,36] that no solution for $\alpha(T)$ exists above a maximum temperature $T_m$ which is close to the Neel temperature (in ferromagnetic case to the Curie temperature). An interesting feature of the $\alpha(T)$ is its independence of $\vec{k}$, at least for the cubic lattices.

In absence of an anisotropy, i.e. if $D=1$, the $\alpha$- and $\beta$-modes are degenerate. Therefore writing $m_k = m'_k$, the Hamiltonian (2.17) reduces to

$$H^\circ = E^\circ + 2\sum_k A^\circ_k m_k + \sum_{kk'} (2B^\circ_1 + B^\circ_2) m_k m_{k'} , \qquad (3.1)$$

where

$$E^0 = -z|J|SN(S + c + \frac{c^2}{4S}), \quad c = \frac{2}{N} \sum_k [1 - (1 - \gamma_k^2)^{\frac{1}{2}}]$$

$$A_k^0 = \epsilon_k (1 + \frac{c}{2S}), \quad \epsilon_k = 2z|J|S(1 - \gamma_k^2)^{\frac{1}{2}}$$

$$B_1^0 = -\frac{z|J|}{N} [(1 - \gamma_k^2)^{\frac{1}{2}} (1 - \gamma_{k'}^2)^{\frac{1}{2}} - 1]$$

$$B_2^0 = -\frac{2z|J|}{N} [(1 - \gamma_k^2)^{\frac{1}{2}} (1 - \gamma_{k'}^2)^{\frac{1}{2}} + 1].$$

Investigation of the Helmholtz free energy of our system determines average occupancies $\langle m_k \rangle$ as follows.

The free energy is given by

$$F = \langle H^0 \rangle - TS, \tag{3.2}$$

where

$$S = -k_B \sum_k [\langle m_k \rangle \ln \langle m_k \rangle - (\langle m_k \rangle + 1) \ln (\langle m_k \rangle + 1)]$$

and $k_B$ is the Boltzman constant. The condition that free energy of the system is an extremum with respect to the average occupancy is $\frac{\partial F}{\partial \langle m_k \rangle} = 0$ so we obtain

$$\tag{3.3}$$
$$\langle m_k \rangle = \{ \exp \beta [(1 + \frac{c}{2S}) \epsilon_k - \frac{1}{z|J|NS^2} \sum_{k'} \epsilon_k \epsilon_{k'} \langle m_{k'} \rangle] - 1 \}^{-1},$$

where we have used equalities

$$2B_1^0 + B_2^0 = -\frac{1}{z|J|NS^2} \epsilon_k \epsilon_{k'}, \quad \frac{1}{k_B T} = \beta$$

and $A_k^0 = \epsilon_k (1 + \frac{c}{2S})$.

This result leads to the conclusion that the occupation number has a Bose-Einstein form with a renormalized temperature dependent energy

$$\epsilon_k(T) = \alpha(T) \epsilon_k, \tag{3.4}$$

where $\alpha(T)$ satisfies the implicit equation

$$\alpha(T) = 1 + \frac{c}{2S} - \frac{2}{NS} \sum_{k'} (1 - \gamma_{k'}^2)^{\frac{1}{2}} \langle m_{k'} \rangle \qquad (3.5)$$

and

$$\langle m_{k'} \rangle = (\exp \beta \, \epsilon_{k'}(T) - 1)^{-1}.$$

Before solving equation (3.5) we have to evaluate the summation in (3.5) and c. Once we are at the stage to calculate such coefficient or those coming from the temperature dependence of magnetization, (2.16), we have to deal with difficult integrals depending on the crystal structure of the material in consideration. Such integrals through first Brillouin zone have considerable influence on the thermodynamic calculation and results.

Let us list them for a convenience:

$$\frac{2}{N} \sum_{k} [1 - (1 - \gamma_k^2)^{\frac{1}{2}}] \equiv c$$

$$\frac{2}{N} \sum_{k} [(1 - \gamma_k^2)^{-\frac{1}{2}} - 1] \equiv c'$$

$$\qquad (3.6)$$

$$\frac{2}{N} \sum_{k} \langle m_k \rangle (1 - \gamma_k^2)^{\frac{1}{2}}$$

$$\frac{2}{N} \sum_{k} \langle m_k \rangle (1 - \gamma_k^2)^{-\frac{1}{2}}.$$

The last two integrals depend on temperature through Bose-Einstein function $\langle m_k \rangle$ and therefore in order to evaluate these two integrals we are usually dealing with temperature expansion of $\langle m_k \rangle$ in certain temperature region.

Let us write down qualitatively the $\alpha(T)$ given in finite spin case by several [28,30,32] authors

$$\alpha = \alpha_0 - \alpha_1 T^4 - \dots \quad , \tag{3.7}$$

where $\alpha_0, \alpha_1$ are constants depending on $S$ and particularly $\alpha_0 = 1 + \dfrac{c}{2S}$ . In Appendix 2 are listed known values of $c$ and $c'$ calculated by different authors for the two cubic lattices.

By using the spin wave renormalization method we obtain the same result as in Oguchi's answer for magnetization

$$m(T) = m_0 - m_1 T^2 - m_2 T^4 - m_3 T^6 - \dots - $$
$$ - n_1 T^6 - \dots \tag{3.8}$$

where $m_1, m_2, m_3, n_1$ are constants depending on the atomic spin and integral $c$. Since the leading temperature term in equation (2.25) has the form $\text{const} \times \left(\dfrac{T}{\alpha}\right)^2$ , substitution of equation (3.7) into this term gives the first term from the spin wave interactions, $n_1 T^6$. For $m_0$ the explicit form is

$$m_0 = 1 - \dfrac{c'}{2S} \cdot \tag{3.9}$$

Then we can conclude that spins in an antiferromagnetic sub-lattice are not, even at $T=0^\circ K$ completely aligned.

# CHAPTER FOUR

## <u>Large Spin and the Classical Limit</u>

Because of the difficulty of estimating the kinematic effect for temperatures that are not negligible compared to $T_c$ the range of validity of Dyson's low temperature result is not known very well. The bound states of two spin waves is also difficult to estimate [28].

In addition to these problems we have further complication of the complexity of the antiferromagnetic ground state. In ferromagnetic case when we eliminate kinematic effect the bound states disappear. In the present work we follow the same philosophy and the kinematic effect obviously is absent, in analogy. Further more it is plausible that the bound state also disappears. In addition we find a simplification of the ground state problem.

The kinematic effect vanishes in the limit $S \to \infty$, since from the topological point of view there is no problem to represent the infinite spin by an infinite system of bosons from Hilbert space completely. So the spurious states will not occur and then the kinematic effect vanishes. The thermodynamic problems may now be calculated much easier.

Let us look at some features of the large $S$ behavior. The spin Hamiltonian (1.2) can be rewritten to the form

$$H = 2|J|S^2 \sum_{\langle \ell, m \rangle} \vec{\Delta}_\ell \cdot \vec{\Delta}_m + h_1 S \sum_\ell \Delta_\ell^z + h_2 S \sum_m \Delta_m^z \tag{4.1}$$

where we have defined dimensionless operator $\vec{\Delta}_\ell$, $\vec{\Delta}_\ell \equiv \dfrac{\vec{S}_\ell}{S}$,
$S$ is the maximum component of $S_\ell^z$ in units of $\hbar$.

As $S \to \infty$, $\vec{\Delta}_\ell$ tends to a classical unit vector since
(i) the magnitude of $\vec{S}_\ell$, $[S(S+1)]^{\frac{1}{2}}$ tends to $S$,
(ii) the eigenvalues of $S_\ell^z$ then tend to infinity resulting in
a continuous set of eigenvalues for $\Delta_\ell^z$ ranging from $-1$ to
$+1$, and

(iii) the $\vec{\Delta}_\ell$ operators then commute in a classical sense, i.e.

$$\tag{4.2}$$

$$[\Delta_k^x, \Delta_\ell^y] = \frac{i}{S} \Delta_\ell^z \delta_{k\ell} .$$

The classical model of an antiferromagnet is based on equation
(4.1) for $S \to \infty$ with $JS^2$ taken constant, or equivalently since
$S$ is measured in units of $\hbar$ by letting $S\hbar \to$ constant.

Let us have a look at the $S$ dependence of $T_N$. If we
take the anisotropy constants equal to zero, dimensional
argument gives $k_B T_N \propto |J|S^2$ for large spin, which is readily
confirmed by the molecular field approximation for which one
has $k_B T_N = \dfrac{2z}{3} |J| S(S+1)$.

For all temperatures obeying the inequality $k_B T \gtrsim |J|S$,
up to and including $T_N$, it is permissible to replace the
Bose-Einstein function by the expansion in powers of $T$ given
by [27,28]

$$n(x) = \frac{1}{x} - \frac{1}{2} + \frac{x}{12} - \frac{x^3}{720} + O(x^5) , \tag{4.3}$$

where $x = \beta E(\vec{R}) \ll 1$ and $E(\vec{R})$ is eigenvalue of the spin Hamiltonian (4.1).

Following the above discussion the criterion $k_B T \gtrsim JS$ may be rearranged as follows

$$T \gtrsim \frac{T_N}{S} \; .$$

(4.4)

This means that the useful temperature range of the conventional result becomes a smaller fraction of $T_N$ as the spin increases. This idea has also been used by Vaks, Larkin and Pikin [29].

To reduce the mathematical inconvenience of the limiting case $S \to \infty$, we introduce dimensionless temperature $\tau$ defined by

$$\tau = \frac{k_B T}{2 |J| z S^2} \; .$$

(4.5)

This becomes clear since temperature enters the problem through the density matrix $e^{-\beta H}$.

The renormalization coefficient and the sublattice magnetization from the previous chapters will be then given for large spin by the self-consistent equation

$$\alpha = 1 + \frac{1}{2S} - \frac{1}{\alpha} \tau - \frac{\alpha}{12\,S^2} \mathbb{I}_1 \frac{1}{\tau} +$$
$$+ \frac{\alpha^3}{1440\, z |J| S^5} \mathbb{I}_{\frac{3}{2}} \frac{1}{\tau^3} + \cdots$$

(4.6)

and

$$m = 1 + \frac{1}{2S} - \frac{1}{\alpha} \mathbb{I}_{-1} \tau - \frac{\alpha}{12\,S^2} \frac{1}{\tau} +$$
$$+ \frac{\alpha^3}{720\, S^4} \mathbb{I}_1 \frac{1}{\tau^3} + \cdots \; ,$$

(4.7)

where integral $\mathbb{I}_m$ is defined as follows

$$\mathbb{I}_m \equiv \frac{2}{N} \sum_{\vec{k}} \left(1 - \gamma_{\vec{k}}^2\right)^m \, , \tag{4.8}$$

$m$ is an integer or a half-integer.

Now taking the limit $S \to \infty$ the above equations simplify to the form

$$\alpha = 1 - \frac{\tau}{\alpha} \tag{4.9}$$

and

$$m = 1 - \mathbb{I}_{-1} \frac{\tau}{\alpha} \, , \tag{4.10}$$

where

$$\mathbb{I}_{-1} = \frac{2}{N} \sum_{\vec{k}} \left(1 - \gamma_{\vec{k}}^2\right)^{-1} \, .$$

Iterative solution for $\alpha$ now gives

$$\alpha = 1 - \tau - \tau^2 - 0\left(\tau^3\right) \tag{4.11}$$

where it follows that

$$m = 1 - \mathbb{I}_{-1} \tau - \mathbb{I}_{-1} \tau^2 - 0\left(\tau^3\right) \, ; \tag{4.12}$$

the quadratic term expresses the spin waves interaction. It turns out that for simple cubic lattice value of $\mathbb{I}_{-1}$ equals Watson's integral*.

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* Communication with Loly

# CONCLUSION

The results for the renormalization coefficient $\alpha$
and relative sublattice magnetization $m$ in the infinite spin
limit are different from those for small spin [compare
equation (4.11), (4.12) and (3.7), (3.8)].

By the comparison of our results with those for the
classical ferromagnet given by Loly we see that implicit
equations for $\alpha$ are identical. In addition the antiferromagnetic
sublattice magnetization has qualitatively the same behavior
as the ferromagnetic magnetization in Loly's work [24], but for
the ferromagnet the coefficient of both the linear and
quadratic terms is Watson's integral instead of the $I_{-1}$ in
our case [equation (4.12)].

The ground state simplifies since in the infinite spin
limit constants in equations (3.1), (3.9), (4.7) of order $\frac{1}{S}$
vanish implying maximum alignment.

Straight forward extension of these results can be done
in case of the antiferromagnetic specific heat and susceptibility.
As a continuation of our work the ferrimagnet and its thermo-
dynamic properties should be calculated.

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## An Example of the Fourier and Bogolyubov Transformation

     Since a Hamiltonian operator represents energy, which is an additive quantity we can transform individual parts of a total Hamiltonian separately. We choose $H^{(1)}$ of our total Hamiltonian. Recalling (2.9) (for brevity $2z|J|S \equiv 1$)

$$H^{(1)} = \sum_{\ell} (1 - h_1') \, a_\ell^+ a_\ell + \sum_{\ell + \varphi} (1 + h_2') \, b_{\ell+\varphi}^+ b_{\ell+\varphi} +$$

$$+ \sum_{\ell, \varphi} \frac{1}{2} (a_\ell b_{\ell+\varphi} + a_\ell^+ b_{\ell+\varphi}^+)$$

and using Fourier transform (2.11) we can write the last part of $H^{(1)}$ as follows

$$\frac{1}{2} \sum_{\ell, \varphi} (a_\ell b_{\ell+\varphi} + a_\ell^+ b_{\ell+\varphi}^+) =$$

$$= \sum_{\varphi} \sum_{k_1, k_2} \delta(k_2 - k_1) \, e^{i k_2 \varphi} \, a_{k_1} b_{k_2} + \tag{1}$$

$$+ \sum_{\varphi} \sum_{k_1, k_2} \delta(k_1 - k_2) \, e^{-i k_2 \varphi} \, a_{k_1}^+ b_{k_2}^+ =$$

$$= \sum_{k} \gamma_k (a_k b_k + a_k^+ b_k^+) \, ,$$

where

$$\delta(k) = \delta(-k) = \frac{2}{N} \sum_{\ell} e^{i k \ell} \quad , \quad N \gg 1 \tag{2}$$

is delta-function and

$$\gamma_k = \frac{1}{z} \sum_{\rho} e^{ik\rho} \tag{3}$$

is function depending on the structure of a given lattice.
Also in a similar manner as (1)

$$\sum_{\ell} a_{\ell}^+ a_{\ell} = \sum_{k} a_{k}^+ a_{k} \quad , \quad \sum_{\ell+\rho} b_{\ell+\rho}^+ b_{\ell+\rho} = \sum_{k} b_{k}^+ b_{k} \quad . \tag{4}$$

It is important to point out that the symbols $\ell$, $\ell+\rho$ represent position vectors $\vec{\pi}_{\ell}$, $\vec{\pi}_{\ell+\rho}$ of the atomic spins and vector $\vec{k}$ goes through the first Brillouin zone. So Fourier transform gives

$$H^{(1)} = \sum_{k} \left[ (1-h_1')\, a_k^+ a_k + (1-h_2')\, b_k^+ b_k + \right.$$
$$\left. + \gamma_k \left( a_k b_k + a_k^+ b_k \right) \right] \tag{5}$$

Commutation relations for the boson spin wave (2.12) are given by the commutation relations of the crystal lattice bosons (2.4) as follows

$$[a_{k'}, a_k^+] = \frac{2}{N} \sum_{\ell,\ell'} e^{i(k'\ell - k\ell')} [a_{\ell'}, a_{\ell}^+] = \delta(\kappa - \kappa') \tag{6}$$

Similarly for the rest of (2.12). Hamiltonian (5) is represented by the nondiagonal quadratic form, which can be transformed by the unitary transformation (2.16) to the diagonal form as

$$H_\chi = \sum_{k} \left[ x_k^- \alpha_k^+ \alpha_k + x_k^+ \beta_k^+ \beta_k \right], \tag{7}$$

From (2.16) follows

$$\dot{a}_k = u\,\dot{\alpha}_k - v\,\dot{\beta}_k^+ \quad , \quad \dot{b}_k = -v\,\dot{\alpha}_k^+ + u\,\dot{\beta}_k \; ,$$

where

$$\dot{\alpha}_k = \frac{i}{\hbar}\left[H_x\,,\,\alpha_k\right] = -\frac{i}{\hbar}\sum_k x_k^-\,\alpha_k$$

$$\dot{\beta}_k = \frac{i}{\hbar}\left[H_x\,,\,\beta_k\right] = \frac{i}{\hbar}\sum_k x_k^+\,\beta_k$$

and then

$$\dot{a}_k = -\frac{i}{\hbar}\sum_k \left(x_k^-\,u\,\alpha_k + x_k^+\,v\,\beta_k^+\right) \; ,$$

$$\dot{b}_k = -\frac{i}{\hbar}\sum_k \left(x_k^-\,v\,\alpha_k^+ + x_k^+\,u\,\beta_k\right) \; . \tag{11}$$

By the equating coefficients $\dot{a}_k$ from (10) and (11) we get two homogeneous equations for the unknown $u$ and $v$ as follows

$$\left[(h_1' - 1) + x_k^-\right] u + \gamma_k v = 0$$

$$\gamma_k u + \left[(h_1' - 1) - x_k^+\right] v = 0 \; , \tag{12}$$

which are soluable only if

$$\det \begin{vmatrix} (h_1' - 1) + x_k^- \; , & \gamma_k \\ \gamma_k \; , & (h_1' - 1) - x_k^+ \end{vmatrix} = 0 \; . \tag{13}$$

From the equations for $\dot{b}_k$ we get similarly

$$\left[(1 + h_2') - x_k^+\right] u - \gamma_k v = 0$$

$$-\gamma_k u + \left[(1 + h_2') + x_k^-\right] v = 0 \tag{14}$$

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## Numerical Quantities for Antiferromagnet

| author (year) | type of structure | | | |
|---|---|---|---|---|
| | NaCl, s.c. | | CsCl, b.c.c. | |
| | c | c′ | c | c′ |
| Anderson (1952) | 0.097 | 0.156 | -- | -- |
| Kubo (1952) | -- | -- | 0.073 | 0.150 |
| Davis (1962) | -- | -- | 0.073038 | 0.118636 |
| M. Bloch (1965) | -- | -- | -- | 0.1185 |

$$c = \frac{2}{N} \sum_{\vec{k}} \left[ 1 - (1 - \gamma_{\vec{k}}^2)^{\frac{1}{2}} \right]$$

$$c' = \frac{2}{N} \sum_{\vec{k}} \left[ (1 - \gamma_{\vec{k}}^2)^{-\frac{1}{2}} - 1 \right]$$
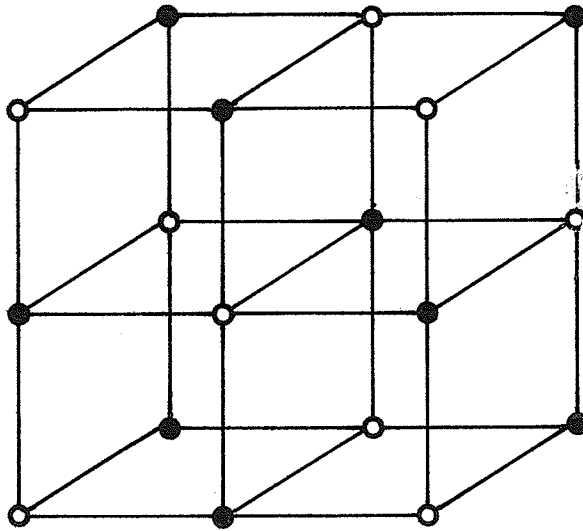
s.c. lattice (f.c.c. sublattices)
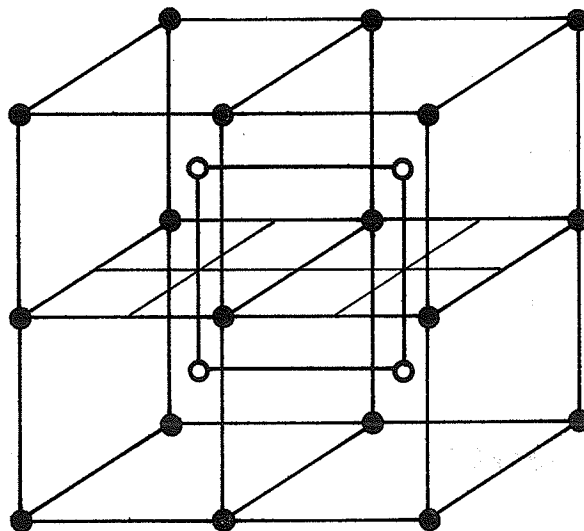


b.c.c. lattice (s.c. sublattices)