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THEORY OF DISORDERED HEISENBERG FERROMAGNETS

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16. Abstract

A Green's function technique is used to calculate the magnetic properties of Heisenberg ferromagnets in which the exchange interactions deviate randomly in strength from the mean interaction. Systems of sc, bcc, and fcc topologies and of general spin values are treated. Disorder produces marked effects in the density of spin wave states - in the form of enhancement of the low-energy density and extension of the energy band to higher values. The spontaneous magnetization and the Curie temperature decrease with increasing disorder. The effects of disorder are shown to be more pronounced in the ferromagnetic than in the paramagnetic phase.

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SUMMARY

The theory of Heisenberg ferromagnets with distributed disorder is developed by a method using double-time, temperature-dependent Green's functions. Disorder is introduced by allowing the exchange interactions between spins to deviate randomly from the mean interaction. The disorder is characterized by a parameter ρ which is proportional to the mean-square deviation of the exchange interactions from the mean interaction. The equations of motion for Callen-type Green's functions are solved by using Tyablikov decoupling, and an ensemble average is performed over systems with similar disorder to provide an ensemble-averaged Green's function. From these disordered Green's functions the densities of spin wave states are derived which, in turn, are used to calculate the magnetic properties of disordered systems. Several specific systems are investigated including disordered simple cubic, body-centered cubic, and face-centered cubic systems with various spin values. The theory is also applied to mixtures of the chalcogenides of europium, which are good examples of Heisenberg ferromagnets. Disorder is shown to have a marked effect on the density of spin wave states. Modest values of the disorder parameter can produce relatively large changes in the state density - in the form of enhancement of the low-energy densities and extension of the energy band to higher values. The increase of the density of low-energy states due to disorder is of the order $(1 - \rho)^{-3/2}$, leading to a corresponding increase in the low-temperature specific heat. The spontaneous magnetization of a disordered ferromagnet decreases with rising temperature more quickly than for a crystal, and the Curie temperature is shown to decrease linearly with disorder as $1 - \rho$. Calculations for the low-temperature region and for the higher temperature paramagnetic phase show that disorder effects are more pronounced in the low-temperature, ferromagnetic phase.

INTRODUCTION

Until recent years, solid-state physics has been a narrower field of study than its name suggests. Perhaps crystal physics would have described the field more accurately since nearly all published literature on the physics of solids, both theoretical and experimental, was confined to systems with translational symmetry. This is not surprising since the mathematical simplifications inherent in lattice symmetries are considerable. Nor did this state of affairs present much of a limitation since most materials of technological importance had a crystalline structure.

In 1965, the first amorphous ferromagnet was discovered (ref. 1) and several more were identified soon afterward (refs. 2 to 5). When they arrived on the scene, there was virtually no theoretical understanding of what to expect when disorder was present in a magnetic system. The fact that ferromagnetism is a cooperative phenomenon makes even more intriguing the question whether disorder would significantly affect a ferromagnet and, if so, in what manner. In a sense the ions in a ferromagnet are more strongly influenced by the other particles in the system than in an uncooperative phase such as a paramagnet. In a ferromagnet the magnetic moments act in concert to produce a more or less parallel alinement, while the moment directions in a paramagnet are random and the net magnetic moment vanishes. One might ask if the introduction of disorder would have drastic results on such cooperative effects. In its present state, the theory of magnetism is not sufficiently developed to give a definitive answer to these questions. It is the intent of this report to rectify some of this incompleteness by expanding and generalizing the theory of mangetism to include the effects of disorder.

In this report the problem of disorder in a ferromagnet is treated by a Green's function theory that incorporates disorder by allowing a randomness in the exchange interactions coupling the magnetic moments. The advantage of the Green's function method is that it gives good results over the entire temperature range (ref. 6). In the interesting regions near $T = 0$ and near $T = T_C$, the Curie temperature, and in the high-temperature paramagnetic region. Green's functions for disordered systems are derived and used to calculate such properties as the density of spin wave states, spontaneous magnetization, Curie temperature, and specific heat and their dependence on disorder. These calculations are applied to systems of various values of spin and various structures.

Some background material for this study is provided in the form of a brief sketch of the present state of the art. It is of interest to know which types of disorder have received attention, what techniques have been used, and which problems have been solved and which have not. The formal theory of double-time, temperature-dependent Green's functions is presented. These are the Green's functions most useful in statistical physics. These Green's functions are then used to treat the problem of a disordered Heisen-

berg ferromagnet. Specific disordered systems are studied; and, finally, general conclusions are drawn from the various systems investigated. The appendixes contain some of the more lengthy derivations and calculations.

EARLIER WORK

The modern theory of magnetism has developed rapidly since 1926 when Dirac (ref. 7) and Heisenberg (ref. 8) independently discovered the concept of exchange. Classical physics had no way to explain the strong interactions between spins needed to account for ferromagnetism. It was shown that exchange was a purely quantum mechanical effect with no classical analog and depended on the overlap of the wave functions of the electrons whose spin accounted for the magnetic moments. The energy of interaction between a spin at site i and a spin at site j was shown to be $-J(ij)\vec{S}i \cdot \vec{S}j$, the so-called Heisenberg energy, where $J(ij)$ is the exchange integral. Depending on how complicated a ferromagnetic system is considered, the Hamiltonian might contain several more terms. But the Heisenberg term is basic and can be derived from the fundamental Coulomb interactions of the electrons together with the Pauli exclusion principle.

Bloch (refs. 9 and 10) analyzed the Heisenberg model of a ferromagnet by using the concept of spin waves. A spin wave consists of a single reversed spin distributed coherently over a large number of otherwise alined atomic spins in a crystal. Dyson (refs. 11 and 12) considered the interactions between these spin waves and derived a low-temperature series expansion for the magnetization in powers of the absolute temperature T. Other temperature regions were investigated by alternate methods. For temperatures near the Curie temperature T_C, molecular field theory (ref. 13) was used; and for high temperatures, perturbation theory (refs. 14 and 15) was employed to give an expansion in $1/T$. In 1959, Tyablikov (refs. 16 and 17) showed that double-time, temperature-dependent Green's functions could be used to describe the Heisenberg ferromagnet over the entire temperature range. His results for a spin $-1/2$ system showed agreement in the main terms with all three methods. The Green's function theory was extended to treat systems of spin greater than $1/2$ by Tahir-Kheli and ter Haar (ref. 18) and by Callen (ref. 19). Several attempts (refs. 20 and 21) to refine their work have been made since, most having to do with improved decoupling procedures.

The first investigation of a noncrystalline ferromagnet to appear in the literature, either theoretical or experimental, was a 1960 paper by Gubanov (ref. 22) in which he used a semiclassical method to demonstrate the possibility of the existence of amorphous ferromagnets. Since that time, there has been a steady growth in the study of various types of disorder in magnetic systems. One of the first problems treated was the case of an ordered Heisenberg ferromagnet in which one spin is replaced by an

impurity spin differing from the host atoms in either spin magnitude or exchange coupling. This system has been examined in detail by Wolfrom and Callaway (ref. 23) and by several others (refs. 24 and 25), all of whom showed the existence of a spin wave mode localized on the impurity, in addition to a modification of the spin wave band of the host lattice. Hone and Vogelsang (ref. 26) demonstrated that weakly coupled impurities produce low-lying spin wave resonances leading to specific-heat anomalies at low temperatures. These results are valid only for low concentrations of substitutional impurities since interactions between impurities are neglected.

Handrich (ref. 27) studied amorphous Heisenberg ferromagnets by using molecular field theory, which predicted a decreased spontaneous magnetization arising from fluctuations in the structure of the system. A shortcoming of this approximation is its failure to show any dependence of the Curie temperature on these structure fluctuations. Later work (refs. 28 and 29) within the same molecular field approximation using computer experiments showed increases in the Curie temperature brought on by randomness in the exchange interactions. This behavior is not that to be expected from other studies (refs. 30 and 31) that show disappearance of ferromagnetism above critical concentrations of defects.

Recently, Montgomery, Krugler, and Stubbs (ref. 32) treated distributed disorder in a spin -1/2 Heisenberg ferromagnet by a Green's function method. This was the first quantitative demonstration that disorder decreased the Curie temperature. Because that work represents the preliminary part of this study, further comments on it will be saved for later sections of this report.

GENERAL THEORY OF TEMPERATURE-DEPENDENT GREEN'S FUNCTIONS

Often one wishes to calculate the expectation value of products of operators, and a powerful and elegant technique that accomplishes this involves the use of Green's functions. We will consider the two kinds of Green's functions which are most convenient in the statistical treatment of magnetic systems: the retarded Green's function $G_r(t, t')$, and the advanced Green's function $G_a(t, t')$. The definitions (ref. 6) of these and the notation used in their regard are

$$G_r(t, t') \equiv \langle\langle A(t); B(t') \rangle\rangle_r = -i\theta(t - t')\langle [A(t), B(t')] \rangle \tag{1}$$

$$G_a(t, t') \equiv \langle\langle A(t); B(t') \rangle\rangle_a = i\theta(t' - t)\langle [A(t), B(t')] \rangle \tag{2}$$

where θ is a step function,

$$\theta(x) = \begin{cases} 1 & \text{when } x > 0 \\ 0 & \text{when } x < 0 \end{cases} \qquad \cdot \qquad (3)$$

and the square brackets are the usual commutator brackets. The angular brackets are the symbol for an average over a grand canonical ensemble,

$$\langle [A(t), B(t')] \rangle \equiv \frac{\text{Tr}\left\{ e^{-\mathfrak{3C}/kT}[A(t), B(t')] \right\}}{\text{Tr}\left(e^{-\mathfrak{3C}/kT} \right)} \qquad (4)$$

where $\mathfrak{3C}$ is the Hamiltonian for the system. The $A(t)$ and $B(t)$ are operators in the Heisenberg representation

$$A(t) = e^{i(\mathfrak{3C}t/\hbar)}A(0)e^{-i(\mathfrak{3C}t/\hbar)} = e^{i(\mathfrak{3C}t/\hbar)}Ae^{-i(\mathfrak{3C}t/\hbar)} \qquad (5)$$

and thereby obey the Heisenberg equation of motion

$$i\hbar \frac{dA(t)}{dt} = [A(t), \mathfrak{3C}] \qquad (6)$$

In general, the operators $A(t)$ and $B(t)$ are particle creation and annihilation operators or products of these.

There are some properties of these Green's functions that are worthy to note before proceeding. First, neither of them is defined when $t = t'$ because of the discontinuous function θ. Secondly, $G_r(t, t')$ and $G_a(t, t')$ are dependent on time only through $t - t'$. This is seen by writing the definitions in explicit form, using equations (4) and (5) and making use of the commutability of operators under the trace sign. And, finally, the averages involved in equations (1) and (2) are not taken over the vacuum state of the system but over the grand canonical ensemble. And it is here that the Green's functions receive their temperature dependence.

The advantage of Green's functions is that, once their solution is found, they can be used to calculate various correlation functions, from which all the properties of interest in a system can be derived. The equations relating correlation functions and Green's functions are developed in a later discussion of spectral representations. Now, we are interested in writing the equation of motion for the Green's function and finding its solution.

The time rate of change of the retarded Green's function is

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$$i\hbar \frac{d}{dt} G_r(t, t') = \hbar \frac{d\theta}{dt}(t - t') \langle [A(t), B(t')] \rangle - i\theta(t - t') \langle [[A(t), \mathcal{H}], B(t')] \rangle \qquad (7)$$

Noting that

$$\frac{d}{dt} \theta(-t) = -\frac{d}{dt} \theta(t) \qquad (8)$$

we see that both the retarded and advanced Green's functions obey the same equation of motion and the subscripts can be dropped. An integral representation for θ involving the Dirac delta function is

$$\theta(t) = \int_{-\infty}^{t} \delta(t') \, dt' \qquad (9)$$

allowing equation (7) to be rewritten as

$$i\hbar \frac{d}{dt} \langle\langle A(t); B(t') \rangle\rangle = \hbar\delta(t - t') \langle [A(t), B(t')] \rangle + \langle\langle [A(t), \mathcal{H}]; B(t') \rangle\rangle \qquad (10)$$

This equation contains two Green's functions plus an inhomogeneous term involving a delta function which recreates the form of the usual classical Green's function equations. Classically, Green's functions are used to obtain the field caused by a point source or a distribution of point sources, and the solution is generally an integral representation involving the Green's function. In these cases the Green's function is a solution to a differential equation having an inhomogeneous delta function term. It is the similarity of these equations with those like equation (10) that brought about the label "Green's function" for the functions used in quantum field theory.

However, the Green's function on the right side of equation (10) has, in general, more terms than the original Green's function; that is, the operator $[A(t), \mathcal{H}]$ has a higher number of terms than does the operator $A(t)$. To solve equation (10) exactly, one needs to know this higher Green's function, $\langle\langle [A(t), \mathcal{H}]; B(t') \rangle\rangle$, which requires the solution of the equation

$$i\hbar \frac{d}{dt} \langle\langle [A(t), \mathcal{H}]; B(t') \rangle\rangle = \hbar\delta(t - t') \langle [[A(t), \mathcal{H}], B(t')] \rangle + \langle\langle [[A(t), \mathcal{H}], \mathcal{H}]; B(t') \rangle\rangle \qquad (11)$$

This introduces still a higher Green's function, $\langle\langle [[A(t), \mathcal{H}], \mathcal{H}]; B(t) \rangle\rangle$, and it is clear that we can generate an infinite set of coupled equations involving a hierarchy of Green's

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functions of the form $\langle\langle[\ldots[[[A(t),\mathcal{H}],\mathcal{H}],\mathcal{H}]\ldots];B(t')\rangle\rangle$. Although the exact solution for $\langle\langle A(t);B(t')\rangle\rangle$ requires solving the chain of equations, one can sometimes make an approximation that decouples the chain and leaves a finite number of equations which can be solved. This is, in fact, the procedure we employ later when we discuss decoupling more fully. For the present, we assume that the chain typified by equations (10) and (11) can be decoupled and a solution found for the Green's function. The question now is how can $G_r(t,t')$ and $G_a(t,t')$ be used to find the quantities of interest in the system under consideration. To calculate these properties from the Green's functions, use is made of correlation functions and spectral representations which we discuss in some detail in the following paragraphs.

When written out explicitly, the equations for $G_r(t,t')$ and $G_a(t,t')$ contain terms like $\langle A(t)B(t')\rangle$, which are averages over the grand canonical ensemble of products of Heisenberg operators. They are called correlation functions and are extremely important in statistical physics. When $t \neq t'$, these averages are the time correlation functions which are useful in the calculation of transport properties. At equilibrium, the time correlation functions depend only on $t - t'$ as do the double-time Green's functions, again because of the commutability of operators under the trace sign. When $t = t'$, then,

$$\langle A(t)B(t)\rangle = \langle A(0)B(0)\rangle = \langle AB\rangle \tag{12}$$

These are the more usual correlation functions which are used to evaluate the average value of the dynamical quantities associated with the operators.

To develop the spectral representations for the correlation functions, we carry out explicitly the operation implied by the angular brackets, $\langle\ldots\rangle$, using the sum over eigenstates of the Hamiltonian of the system. We assume the set of states $|\mu\rangle$, where

$$\mathcal{H}|\mu\rangle = E_\mu|\mu\rangle \tag{13}$$

to be complete and orthonormal so that we can rewrite equation (4) as

$$\langle A(t)B(t')\rangle = Q^{-1}\sum_\mu \langle\mu|A(t)B(t')|\mu\rangle e^{-E_\mu/kT} \tag{14}$$

where Q is the partition function,

$$Q = \text{Tr}(e^{-\mathcal{H}/kT}) \tag{15}$$

The Dirac notation on the right side of equation (14) should cause no confusion with the

angular averaging brackets on the left. We shall expand equation (14) further in the same basis, $\sum_\nu |\nu\rangle\langle\nu|$, and use relation (5).

$$\langle A(t)B(t')\rangle = Q^{-1} \sum_\mu \sum_\nu \langle\mu|A(0)|\nu\rangle\langle\nu|B(0)|\mu\rangle\, e^{-i(E_\mu - E_\nu)(t'-t)/\hbar}\, e^{-E_\mu/kT}$$

(16)

Similarly,

$$\langle B(t')A(t)\rangle = Q^{-1} \sum_\mu \sum_\nu \langle\mu|B(0)|\nu\rangle\langle\nu|A(0)|\mu\rangle\, e^{-i(E_\mu - E_\nu)(t-t')/\hbar}\, e^{-E_\mu/kT}$$

(17)

By defining a spectral density,

$$I_{AB}(\omega) \equiv Q^{-1} \sum_\mu \sum_\nu \langle\mu|A(0)|\nu\rangle\langle\nu|B(0)|\mu\rangle\, e^{-E_\nu/kT}\, \delta\left(\omega - \frac{E_\nu}{\hbar} + \frac{E_\mu}{\hbar}\right)$$

(18)

we can rewrite equations (16) and (17) more compactly as

$$\langle A(t)B(t')\rangle = \int_{-\infty}^{\infty} I_{AB}(\omega)e^{\hbar\omega/kT}\, e^{-i\omega(t-t')}\, d\omega$$

(19)

$$\langle B(t')A(t)\rangle = \int_{-\infty}^{\infty} I_{AB}(\omega)e^{-i\omega(t-t')}\, d\omega$$

(20)

Expressed in this form, the spectral density $I_{AB}(\omega)$ is seen to be the Fourier transform of the time correlation function.

The spectral representations of the time correlation functions, equations (19) and (20), can be used to write the spectral representations of the retarded and advanced Green's functions. First, we define the time Fourier transform of the Green's functions

$$G_r(t - t') = \int_{-\infty}^{\infty} \left(\frac{1}{\hbar}\right) G_r(E) e^{-i(E/\hbar)(t-t')} \, dE \qquad (21)$$

$$G_r(E) = \frac{1}{2\pi} \int G_r(t - t') e^{i(E/\hbar)t} \, dt$$

More explicitly, the time transform of the retarded Green's function is

$$G_r(E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} -i\theta(t)\left[\langle A(t)B(t')\rangle - \langle B(t')A(t)\rangle\right] e^{i(E/\hbar)t} \, dt \qquad (22)$$

or, by equations (19) and (20)

$$G_r(E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \, \theta(t) \int_{-\infty}^{\infty} d\omega I_{AB}(\omega) e^{i\left[(E/\hbar) - \omega\right]t} (e^{\hbar\omega/kT} - 1) \qquad (23)$$

At this point we notice that the step function $\theta(t)$ can be represented in integral form

$$\theta(t) = \lim_{\epsilon \to 0} \frac{-i}{2\pi} \int_{-\infty}^{\infty} d\omega' \frac{e^{-i\omega't}}{\omega' + i\epsilon} \qquad \epsilon > 0 \qquad (24)$$

That this integral representation has the properties of the discontinuous step function can be verified by a contour integration and the theorem of residues.

The t-integration of expression (23) can be carried out with the help of equation (24):

$$\int_{-\infty}^{\infty} dt \, \theta(t) e^{i(E/\hbar - \omega)t} = \frac{-i}{2\pi} \int_{-\infty}^{\infty} d\omega' \int_{-\infty}^{\infty} \frac{e^{i(E/\hbar - \omega - \omega')t}}{\omega' + i\epsilon} \, dt$$

$$= -i \int_{-\infty}^{\infty} d\omega' \frac{\delta\left(\frac{E}{\hbar} - \omega - \omega'\right)}{\omega' + i\epsilon} = \frac{-i}{\frac{E}{\hbar} - \omega + i\epsilon} \qquad (25)$$

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We used here the identity

$$\delta(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-ixy} \, dy \tag{26}$$

Substitution of equation (25) back into equation (23) gives for the retarded Green's function

$$G_r(E) = \frac{-1}{2\pi} \int_{-\infty}^{\infty} d\omega \, \frac{I_{AB}(\omega)(e^{\hbar\omega/kT} - 1)}{\dfrac{E}{\hbar} - \omega + i\epsilon} \tag{27}$$

By the same procedure the advanced Green's function is

$$G_a(E) = \frac{-1}{2\pi} \int_{-\infty}^{\infty} d\omega \, \frac{I_{AB}(\omega)(e^{\hbar\omega/kT} - 1)}{\dfrac{E}{\hbar} - \omega - i\epsilon} \tag{28}$$

These are the spectral representations of the Green's functions.

Now consider E to be complex. Inspection of equations (27) and (28) shows that we can consider $G_r(E)$ and $G_a(E)$ as one function which has the properties of the retarded Green's function when E is in the upper half-plane and the properties of the advanced Green's function when E is in the lower half-plane. That is,

$$G(E) = \frac{-1}{2\pi} \int_{-\infty}^{\infty} d\omega \, \frac{I_{AB}(\omega)(e^{\hbar\omega/kT} - 1)}{\dfrac{E}{\hbar} - \omega} = \begin{cases} G_r(E) & \mathcal{I}m \, E > 0 \\ G_a(E) & \mathcal{I}m \, E < 0 \end{cases} \tag{29}$$

$G(E)$ can be considered as a single analytic function having singularities along the real axis. The retarded Green's function is analytic in the upper half-plane, and the advanced Green's function is analytic in the lower half of the energy plane. This analyticity follows from a theorem (ref. 33) which states that the complex function $G(E)$ has analytic continuation in the upper (lower) half of the complex E plane if its Fourier transform $G(t)$ vanishes at $t < 0$ ($t > 0$). So the functions $\theta(t - t')$ and $\theta(t' - t)$, which

act to cut off the retarded and advanced Green's function when $t < t'$ and $t > t'$, respectively, are the necessary and sufficient conditions for the analytic continuations in the complex E plane.

It is interesting to see the information that can be gained in crossing the cut along the real axis. That is, by taking the difference in the values of $G(E)$ just above and below the cut, we can evaluate the spectral density, which in turn allows us to calculate the correlation functions. From equations (27) and (28)

$$G(E + i\epsilon) - G(E - i\epsilon) = \frac{-1}{2\pi} \int_{-\infty}^{\infty} d\omega I_{AB}(\omega)(e^{\hbar\omega/kT} - 1)\left(\frac{1}{\frac{E}{\hbar} - \omega + i\epsilon} - \frac{1}{\frac{E}{\hbar} - \omega - i\epsilon}\right) \tag{30}$$

Noting that the Dirac delta function can be written

$$\delta(x) = \lim_{\epsilon \to 0} \frac{i}{2\pi}\left(\frac{1}{x + i\epsilon} - \frac{1}{x - i\epsilon}\right) \tag{31}$$

equation (30) becomes

$$G(E + i\epsilon) - G(E - i\epsilon) = i I_{AB}\left(\frac{E}{\hbar}\right)(e^{E/kT} - 1) \tag{32}$$

Equation (32) is an important relation. If the equations of motion of $G(E)$ can be solved, we can use equation (32) to obtain the spectral density $I_{AB}(E/\hbar)$, which can be used in equations (19) and (20) to produce

$$\langle A(t)B(0)\rangle = i \int_{-\infty}^{\infty} \frac{G(\hbar\omega + i\epsilon) - G(\hbar\omega - i\epsilon)}{e^{\hbar\omega/kT} - 1} e^{\hbar\omega/kT} e^{-i\omega t} d\omega \tag{33}$$

$$\langle B(0)A(t)\rangle = i \int_{-\infty}^{\infty} \frac{G(\hbar\omega + i\epsilon) - G(\hbar\omega - i\epsilon)}{e^{\hbar\omega/kT} - 1} e^{-i\omega t} d\omega \tag{34}$$

We have, finally, in equations (33) and (34) the relations by which we can calculate time correlation functions from the Fourier transforms of the Green's functions. In the case of the simpler correlation function $\langle BA\rangle$, equation (34) is

$$\langle BA \rangle = i \int_{-\infty}^{\infty} \frac{G(\hbar\omega + i\epsilon) - G(\hbar\omega - i\epsilon)}{e^{\hbar\omega/kT} - 1} d\omega \qquad (35)$$

The decision of how to define the Green's function for a particular physical problem will depend in large measure on these last three equations. One wishes the correlation function $\langle BA \rangle$ to be a meaningful, important property of the system under study; and this will dictate the choice of operators B and A, from which the Green's function $\langle\langle A(t); B(t') \rangle\rangle$ is defined.

DISORDERED HEISENBERG FERROMAGNETS

Equation of Motion

Ferromagnetism is a cooperative phenomenon, and it is this fact that makes a ferromagnet so interesting physically and statistically. The direction of the spin of a magnetic moment is influenced by the field of other spins of the system, and the field from that spin in turn affects the other spins. As discussed in the section EARLIER WORK, the fundamental interaction responsible for ferromagnetism is the exchange interaction of the form $J(f,g)\vec{S}_f \cdot \vec{S}_g$. In this expression, $\vec{S}_f$ and $\vec{S}_g$ are the spin operators associated with sites f and g, and $J(f,g)$ is the exchange integral between these sites. For a ferromagnetic interaction, $J(f,g)$ is a positive quantity. The Heisenberg model is a collection of magnetic moments, every pair interacting by exchange. The Hamiltonian for the Heisenberg model is the sum of these interactions.

$$\mathcal{H} = -\frac{1}{2} \sum_f \sum_g J(f,g)\vec{S}_f \cdot \vec{S}_g - Hg\mu_B \sum_f S_f^z \qquad (36)$$

Since each pair of sites is counted twice in the double sum, the factor 1/2 is present. The second term of the Hamiltonian is the Zeeman term with the external magnetic field H assumed to be parallel to the z-axis, g is the Lande g-factor, and μ_B the Bohr magneton. Representing the spin operators in terms of their components

$$S_f^{\pm} = S_f^x \pm i S_f^y \qquad (37)$$

puts the Hamiltonian into the form

$$\mathcal{H} = -\frac{1}{2} \sum_f \sum_g J(f,g) \left[\frac{1}{2} \left(S_f^+ S_g^- + S_g^+ S_f^- \right) + S_f^z S_g^z \right] - Hg\mu_B \sum_f S_f^z \tag{38}$$

These components obey the commutation relations

$$\left. \begin{array}{c} \left[S_f^+, S_g^- \right] = 2\hbar S_f^z \delta_{fg} \\[2mm] \left[S_f^z, S_g^\pm \right] = \pm \hbar S_f^\pm \delta_{fg} \end{array} \right\} \tag{39}$$

For a system of spin-1/2 particles where each spin is in one of two possible configurations, $S^z = \hbar/2$ or $S^z = -\hbar/2$, a very suitable Green's function is one of the type used by Montgomery, Krugler, and Stubbs (ref. 32), $\langle\langle S_f^+(t); S_g^-(t') \rangle\rangle$. From this choice of Green's function the correlation function $\langle S_f^- S_f^+ \rangle$ can be calculated by equation (35). Since, from elementary quantum mechanics,

$$S^- S^+ = S(S+1)\hbar^2 - \left(S^z \right)^2 - \hbar S^z \tag{40}$$

for the spin-1/2 case,

$$S^- S^+ = \frac{\hbar^2}{2} - \hbar S^z \tag{41}$$

That is, $\langle S_f^- S_f^+ \rangle$ is directly related to the magnetization. For spins higher than 1/2, there is ambiguity in the term $\left(S^z \right)^2$ of equation (40) which makes $\langle\langle S_f^+(t); S_g^-(t') \rangle\rangle$ an unsuitable Green's function for spins greater than 1/2.

The Green's function we have chosen to develop the theory of disordered ferromagnets of any spin value are those used by Callen (ref. 19) in his extension of the Green's function theory of crystalline ferromagnets to general spin:

$$G_{fg}^a(t) \equiv \left\langle\!\!\left\langle S_f^+(t); e^{aS_g^z} S_g^- \right\rangle\!\!\right\rangle \tag{42}$$

The t' associated with S_g^z and S_g^- has been set equal to zero. The superscript a is a parameter which will become useful only when generalizing the theory to systems of spin S greater than 1/2. The special case $a = 0$ will be the most important one when using the Green's function to calculate properties of the system.

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To study the time development of this function in the Heisenberg model, we write its equation of motion

$$i\hbar \frac{d}{dt} G_{fg}^a(t) = \hbar\delta(t)\delta_{fg} \left\langle \left[S_f^+, e^{aS_f^z} S_f^- \right] \right\rangle + \left\langle\left\langle \left[S_f^+(t), \mathcal{H} \right]; e^{aS_g^z} S_g^- \right\rangle\right\rangle \tag{43}$$

The averaged value of the commutator we label $\Theta(a)$.

$$\left\langle \left[S_f^+, e^{aS_f^z} S_f^- \right] \right\rangle \equiv \Theta(a) \tag{44}$$

Note, that for $a = 0$, Θ is related to the magnetization $\sigma = \langle S^z \rangle / S$.

$$\Theta(0) = \left\langle \left[S_f^+, S_f^- \right] \right\rangle = 2\hbar \, S_f^z \tag{45}$$

Commuting $S_f^+(t)$ with the Hamiltonian allows equation (43) to be written

$$i\hbar \frac{d}{dt} G_{fg}^a(t) - Hg\mu_B \hbar G_{fg}^a(t) + \hbar \sum_h J(f,h) G_{f,\,hg}^a(t) - \hbar \sum_h J(f,h) G_{h,\,fg}^a(t) = \hbar\delta(t)\delta_{fg}\Theta(a) \tag{46}$$

where

$$G_{f,\,hg}^a(t) = \left\langle\left\langle S_f^z(t) S_h^+(t); e^{aS_g^z} S_g^- \right\rangle\right\rangle \tag{47}$$

The appearance of higher order Green's functions $G_{f,\,hg}^a(t)$ in the equation of motion for $G_{fg}^a(t)$ is an expected development after the discussion of the general theory. We could, of course, write equations of motion for these higher Green's functions and these would introduce still higher order functions. Before talking about ways of decoupling this chain of equations we introduce the Fourier transforms of the functions in equation (46):

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$$G^a_{f,g}(t) = \int_{-\infty}^{\infty} G^a_{fg}(E)e^{-i(E/\hbar)t}\, d\left(\frac{E}{\hbar}\right)$$

$$G^a_{f,hg}(t) = \int_{-\infty}^{\infty} G^a_{f,hg}(E)e^{-i(E/\hbar)t}\, d\left(\frac{E}{\hbar}\right) \Bigg\}$$

$$\delta(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i(E/\hbar)t}\, d\left(\frac{E}{\hbar}\right)$$

$$(48)$$

Substitution of equations (48) into equation (46) allows us to express the equation of motion in terms of Fourier components of the Green's function:

$$E G^a_{fg}(E) = Hg\mu_B\hbar G^a_{fg}(E) - \hbar \sum_h J(f,h)\left[G^a_{f,hg}(E) - G^a_{h,fg}(E)\right] + \frac{\hbar}{2\pi}\delta_{fg}\Theta(a) \qquad (49)$$

The obvious way to decouple the hierarchy of equations for the Green's functions is to find a way of representing higher functions in terms of lower order functions. This procedure presents a finite set of equations which can generally be handled. In most instances (refs. 16 to 21) the set of equations is cut off after the first equation by a decoupling approximation that represents a second Green's function (i.e., one with three indices) in terms of the first, doubly indexed, functions. One gets the impression that some of the decoupling approximations which have been used were not developed a priori from considerations of the physics of the system but that the first criterion was to produce an approximation that would make the mathematics tractable. Plausibility arguments can sometimes be offered as to why the approximation might be expected to be valid under certain conditions (of temperature, e.g.) and the results of these calculations do indeed compare favorably with exact results in many circumstances. But, the statement (ref. 18) of a decade ago that "the philosophy and justification of the decoupling procedure is still far from being well understood" is only slightly less true today.

We decouple in the manner first employed by Tyablikov (refs. 16 and 17) by making the approximation

$$G^a_{f,hg}(E) \simeq \langle S^z \rangle G^a_{hg}(E) \qquad \text{for } f \neq h \qquad (50)$$

which is equivalent to the statement

$$\left\langle\!\!\left\langle S_f^Z(t)S_h^+(t); e^{aS_g^Z}S_g^- \right\rangle\!\!\right\rangle \simeq \left\langle S_f^Z \right\rangle \left\langle\!\!\left\langle S_h^+(t); e^{aS_g^Z}S_g^- \right\rangle\!\!\right\rangle \qquad \text{for } f \neq h \tag{51}$$

This approximation replaces the average of a product by the product of averages. For example, if $f \neq g$, then S_f^Z will commute with the operator $e^{aS_g^Z}S_g^-$ and

$$G_{f,hg}^a(t) = -i\theta(t)\left\langle \left[S_f^Z(t)S_h^+(t), e^{aS_g^Z}S_g^- \right] \right\rangle = -i\theta(t)\left\langle S_f^Z(t)\left[S_h^+(t), e^{aS_g^Z}S_g^- \right] \right\rangle \tag{52}$$

In this case, equation (51) replaces the average of the product of the operators $S_f^Z(t)$ and $\left[S_h^+(t), e^{aS_g^Z}S_g^- \right]$ by the product of the averages of these operators. This is sometimes referred to as ignoring the fluctuations in S_f^Z, but this is not literally true. What the Tyablikov type of decoupling ignores is any spin correlations between sites f and h and between sites f and g, while uncorrelated fluctuations in S_f^Z are treated correctly.

The effect of equation (50) is to reduce to one the types of Green's functions in the equation of motion

$$\left[E - Hg\mu_B\hbar - \hbar\langle S^Z \rangle \sum_h J(f,h) \right] G_{fg}^a(E) + \hbar\langle S^Z \rangle \sum_h J(f,h)G_{hg}^a(E) = \frac{\hbar}{2\pi}\delta_{fg}\Theta(a) \tag{53}$$

Having purged the equation of motion of higher Green's functions, we can simplify equation (53), conceptually at least, by considering it a matrix equation

$$\hat{A}\hat{G} = \hat{1} \tag{54}$$

where

$$A_{fh} \equiv \delta_{fh}\left\{ 2\pi\left[\frac{\frac{E}{\hbar} - Hg\mu_B}{\Theta(a)} - \frac{\langle S^Z \rangle}{\Theta(a)} \sum_j J(f,j) \right] \right\} + \frac{2\pi\langle S^Z \rangle}{\Theta(a)} J(f,h) \tag{55}$$

and $\hat{1}$ is the unit matrix.

The indices in these equations are lattice site labels; and so for a perfect crystal the property of translational invariance would be exploited to solve equation (53), which

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we can do quickly. If we call the Green's function for a perfect crystal Γ, we would write its spatial Fourier transform as

$$\left. \begin{aligned} \Gamma^a_{gf}(E) &= \frac{1}{N} \sum_{\vec{k}} e^{i(f-g) \cdot \vec{k}} \, \Gamma^a_{\vec{k}}(E) \\[1em] J(f,g) &= \frac{1}{N} \sum_{\vec{k}} e^{i(f-g) \cdot \vec{k}} \, \mathcal{J}(\vec{k}) \\[1em] \delta_{fg} &= \frac{1}{N} \sum_{\vec{k}} e^{i(f-g) \cdot \vec{k}} \end{aligned} \right\} \tag{56}$$

where $\vec{k}$ is a reciprocal lattice vector. When substituted into equation (53), these relations (56) yield

$$\Gamma^a_{\vec{k}}(E) = \frac{\hbar \Theta(a)}{2\pi} \frac{1}{E - E_{\vec{k}}} \tag{57}$$

where

$$E_{\vec{k}} = H g \mu_B \hbar + \hbar \langle S^Z \rangle \left[\mathcal{J}(0) - \mathcal{J}(\vec{k}) \right] \tag{58}$$

and

$$\mathcal{J}(\vec{k}) = \sum_{f-g} e^{i(f-g) \cdot \vec{k}} \, J(f,g) \tag{59}$$

The Green's function $\Gamma^a_{\vec{k}}(E)$ has poles on the real axis at $E = E_{\vec{k}}$. These are the eigenvalues of the Hamiltonian, the energies of the elementary excitations of the system. From equation (32) the spectral density $I_{AB}(\omega)$ will have δ-function singularities if the Green's function has poles only on the real axis. In this case, from equation (19) the correlation function will oscillate only at the frequencies $E_{\vec{k}}/\hbar$. In our case the corre-

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lation function for $a = 0$ is $\langle S_g^- S_g^+(t) \rangle$ and its frequency of oscillation is that of the corresponding spin wave.

In a disordered ferromagnet, translational symmetry is absent and so the technique of expressing things in reciprocal lattice space, or $\vec{k}$-space, is not available to us. The solution for the disordered case which follows uses the method of reference 32 and is expressed in terms of $\Gamma_{\vec{k}}^a(E)$.

Introduction of Disorder

Before proceeding, it would be beneficial to define more precisely what is meant by the term disorder in our context. We have stated that a Fourier transform into k-space would be disallowed for equation (53) if translational symmetry were absent. To be more complete, we should say that even in a symmetric lattice the Fourier transform that yielded $\Gamma_{\vec{k}}^a(E)$ in equation (57) would not be valid if the exchange interactions were site dependent, that is, if $J(f, g)$ depended on both indices and not just on $\vec{r}_f - \vec{r}_g$. So, another type of disorder which can be discussed is a randomness in the value of exchange interactions. Certainly, it is difficult to conceive of a system with positional disorder that did not also have disorder in the J's since the exchange integral is very sensitive to the interatomic distance, but the converse is quite conceivable. Substitutional impurities in a perfect-crystal lattice would preserve positional symmetry while producing disorder in the interactions. The disorder to which we will be directing our attention is a randomness in the value of the exchange integrals with or without positional disorder. The requirement on the position of the spin sites is that their ensemble-averaged positions (the ensemble average performed over systems with similar disorder) possess lattice symmetry. This requirement implies that the members of the ensemble must be topologically equivalent. Topological equivalence means not only that a one-to-one correspondence of spin sites be preserved throughout the ensemble, but also that the number of near neighbors of each spin of each ensemble member remain the same. Specifically, this study investigates the cases where the ensemble-averaged spin sites have simple cubic (sc), body-centered cubic (bcc), or face-centered cubic (fcc) symmetries.

What does it mean to take ensemble averages over systems with similar disorder? A way to depict the disorder in a system graphically is shown in figure 1, where the number of exchange interactions per unit energy of strength J is plotted against J. The case of a perfectly ordered ferromagnet with nearest-neighbor interactions of strength J^0 would be represented by the delta function spike, $(1/2)ZN\delta(J - J^0)$, where Z is the number of nearest neighbors, or coordination number, and N is the number of spins in the crystal. The dashed curve might represent a disordered ferromagnet
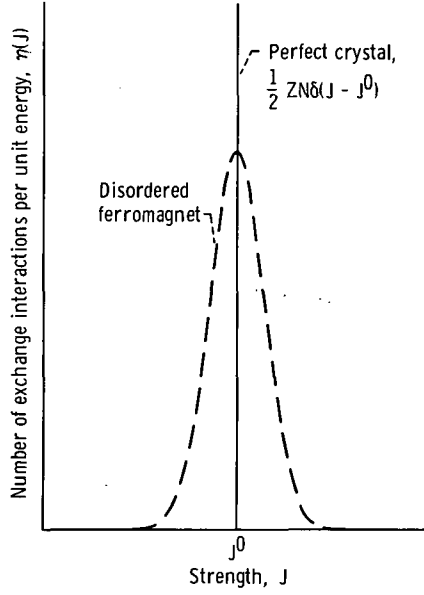
Figure 1. - Distribution of exchange interactions.

whose exchange interactions are distributed randomly about a mean interaction, say J^0. Those systems whose exchange distributions match this curve would be said to have similar disorder, and an ensemble whose members have identical curves is the kind of ensemble we refer to when performing our averages. We assume, also, that the disorder is distributed throughout the ferromagnet and that deviations are not correlated over finite distances.

It is not necessary to know the exact distribution function, $\eta(J)$ of figure 1, to be able to calculate quantitatively the effects of disorder in a magnetic system. This work parameterizes the distribution of J-values by two quantities, a mean value of the exchange integral $J^0(f, g)$ and the mean-square deviation from this mean. We shall characterize the amount of disorder in a system by a disorder parameter ρ which is related to the ratio of the mean-square deviation and the mean:

$$\rho \equiv \frac{2}{Z} \frac{\left\langle \left(J - J^0 \right)^2 \right\rangle}{J^{0^2}} \tag{60}$$

With respect to figure 1, ρ might be thought of as a measure of the width-height ratio of the distribution function curves. Having defined disorder, the task now is the solution of the equation of motion for a disordered ferromagnet (eq. (53)).

Equation (54) is the equation of motion reduced to matrix form. For the case of a perfect crystal this equation is

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$$\hat{A}^0 \hat{\Gamma} = \hat{1} \tag{61}$$

where the superscript on the linear operator $\hat{A}$ indicates perfect order. We define a matrix

$$\hat{\Delta} \equiv \hat{A}^0 - \hat{A} \tag{62}$$

Simple manipulation of equations (54), (61), and (62) leaves

$$\hat{A}^0 \hat{G} - \hat{\Delta} \hat{G} = \hat{1} \tag{63}$$

Since $\hat{\Gamma}$ commutes with its inverse $\hat{A}^0$, operating from the left with $\hat{\Gamma}$ changes equation (63) to a Dyson equation

$$\hat{G} = \hat{\Gamma} + \hat{\Gamma} \hat{\Delta} \hat{G} \tag{64}$$

or

$$\hat{G} = \hat{\Gamma} + \hat{\Gamma} \hat{\Delta} \hat{\Gamma} + \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} + \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} + \ldots \tag{65}$$

We wish to obtain an ensemble-averaged Green's function $\langle G_{fg}^a(E) \rangle$, which is done by performing an ensemble average over systems with similar disorder. Since $\Gamma_{\tilde{k}}^a(E)$ is the Green's function for the perfect-crystal (or zero disorder) case, $\langle \hat{\Gamma} \rangle = \hat{\Gamma}$; and so the ensemble average of equation (64) is

$$\langle \hat{G} \rangle = \hat{\Gamma} + \hat{\Gamma} \langle \hat{\Delta} \hat{G} \rangle \tag{66}$$

which we can put in a Dyson form

$$\langle \hat{G} \rangle = \hat{\Gamma} + \hat{\Gamma} \hat{\Sigma} \langle \hat{G} \rangle \tag{67}$$

where $\hat{\Sigma}$, the "self-energy" is

$$\hat{\Sigma} = \langle \hat{\Delta} \hat{G} \rangle \langle \hat{G} \rangle^{-1} \tag{68}$$

This allows us to express $\langle \hat{G} \rangle$ in terms of $\hat{\Gamma}$ and the self-energy:

$$\langle \hat{G} \rangle = (\hat{1} - \hat{\Gamma} \hat{\Sigma})^{-1} \hat{\Gamma} \tag{69}$$

The quantities $\langle \hat{G} \rangle$ and $\hat{\Sigma}$ are translationally invariant because they are ensemble-averaged quantities and, as such, are diagonal in $\vec{k}$-space. The Fourier space transformation that effects this diagonalization yields

$$\langle G^a(E) \rangle_{\vec{k}} = \left\{ \left[\Gamma^a_{\vec{k}}(E) \right]^{-1} - \Sigma^a_{\vec{k}}(E) \right\}^{-1} \tag{70}$$

Evaluation of $\Sigma^a_{\vec{k}}(E)$ will allow us the solution to the ensemble-averaged Green's function $\langle G^a(E) \rangle_{\vec{k}}$

Upon ensemble averaging, equation (65) becomes

$$\langle \hat{G} \rangle = \hat{\Gamma} + \hat{\Gamma} \langle \hat{\Delta} \rangle \hat{\Gamma} + \hat{\Gamma} \langle \hat{\Delta} \hat{\Gamma} \hat{\Delta} \rangle \hat{\Gamma} + \hat{\Gamma} \langle \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \rangle \hat{\Gamma} + \ldots \tag{71}$$

where Δ involves the differences in the values of the exchange integrals between the perfect and disordered systems. If we assume the deviations of the J_{fg}'s from the mean to be symmetric, all ensemble-averaged quantities in equation (71) involving odd powers of Δ will vanish.

$$\left. \begin{array}{c} \langle \hat{\Delta} \rangle = 0 \\[2mm] \langle \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \rangle = 0 \\[2mm] \langle \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \hat{\Gamma} \hat{\Delta} \rangle = 0 \\[2mm] \vdots \\ \vdots \\ \vdots \end{array} \right\} \tag{72}$$

This assumption defines Γ as the Green's function for a perfect crystal whose exchange interactions are the mean interactions of the disordered ferromagnet, which we call the corresponding perfect crystal.

Now, we make the approximation

$$\hat{\Sigma} \simeq \hat{\Sigma}^0 \equiv \langle \hat{\Delta} \hat{\Gamma} \hat{\Delta} \rangle \tag{73}$$

which in equation (71) is equivalent to replacing $\langle \hat{\Delta}^{2n} \rangle$ by $\langle \hat{\Delta}^2 \rangle^n$. We have carried out the ensemble averaging of equation (73) in appendix B. In the difference matrix $\hat{\Delta}$, we designate the deviations from the mean exchange interaction as

$$j(f, g) \equiv J(f, g) - J^0(f, g) \tag{74}$$

and the ensemble averaging involves use of the relation

$$\langle j(f, g) j(h, i) \rangle = j^2(f, g)\left[\delta_{fh}\delta_{gi} + \delta_{fi}\delta_{gh}\right] \tag{75}$$

Here $j^2(f, g)$ is the mean square deviation for the exchange interaction between sites f and g and is proportional to the disorder parameter ρ (eq. (60)). Using relation (75) in equation (73) gives

$$\left(\hat{\Sigma}^0\right)^a_{gj} = \left(\frac{2\pi\langle S^z\rangle}{\Theta(a)}\right)^2 \left\{ \delta_{gj}\left[\Gamma_{gj} \sum_h j^2(g, h) + \sum_h j^2(g, h)(\Gamma_{hh} - 2\Gamma_{gh}) \right] \right.$$

$$\left. + 2j^2(g, j)(\Gamma_{gj} - \Gamma_{gg}) \right\} \tag{76}$$

The diagonalization of $\hat{\Sigma}^0$ has also been performed in appendix B. In reciprocal lattice space the self-energy becomes

$$\left(\hat{\Sigma}^0\right)^a_{\vec{k}} \equiv \Sigma^a_{\vec{k}} = \left(\frac{2\pi\langle S^z\rangle}{\Theta(a)}\right)^2 \frac{1}{N} \sum_{\vec{k}'} \Gamma_{\vec{k}'}\left[2\mathscr{J}_2(0) - 2\mathscr{J}_2(\vec{k}) - 2\mathscr{J}_2(\vec{k}') + \mathscr{J}_2(\vec{k} + \vec{k}') + \mathscr{J}_2(\vec{k} - \vec{k}')\right] \tag{77}$$

As shown in appendix B, when applied to the three cubic systems with nearest-neighbor interactions, equation (77) simplifies to

$$\Sigma^a_{\vec{k}}(E) = \frac{2\pi\rho}{\hbar\Theta(a)} E_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{E_{\vec{k}'}}{E - E_{\vec{k}'}} \tag{78}$$

We can now write the Green's function for the disordered system explicitly in terms of the eigen energies of the corresponding perfect crystal. Using equation (78) in equation (70) gives

$$\langle G^a(E)\rangle_{\vec{k}} = \frac{\hbar\Theta(a)}{2\pi} \frac{1}{E - E_{\vec{k}} - \rho E_{\vec{k}} \dfrac{1}{N} \displaystyle\sum_{\vec{k}'} \dfrac{E_{\vec{k}}}{E - E_{\vec{k}'}}} \tag{79}$$

This is the Green's function which we will now employ to study the effects of disorder on a ferromagnet.

Density of States

Having finally solved the Green's function for a disordered ferromagnet in equation (79), we can immediately use it to calculate the density of spin wave states g(E). This is a quantity of central importance in the calculations of the magnetic properties because it allows sums over k-states, triple integrals, to be replaced by a single integration over the energy of these states.   For example, we will later wish to calculate the magnetization σ, which involves summing the quantity $\left[\exp(E_{\vec{k}}/kT) - 1\right]^{-1}$ over all $\vec{k}$'s. This is generally done by an integration over the first Brillouin zone,

$$\sum_{\vec{k}} \frac{1}{\exp\left(\dfrac{E_{\vec{k}}}{kT}\right) - 1} \rightarrow \frac{v}{(2\pi)^3} \int\int\int_0^{2\pi/a} dk_x\, dk_y\, dk_z \frac{1}{\exp\left(\dfrac{E_{\vec{k}}}{kT}\right) - 1} \tag{80}$$

where v is the volume of the unit cell. Alternatively, this sum can be performed with a significant saving of time and effort by

$$\sum_{\vec{k}} \frac{1}{\exp\left(\dfrac{E_{\vec{k}}}{kT}\right) - 1} \rightarrow \int_0^\infty \frac{g(E)}{\exp\left(\dfrac{E}{kT}\right) - 1}\, dE \tag{81}$$

The density of states here is the fraction of the total number of spin wave states per unit energy that have energy E.

For a given crystal structure the energy of a spin wave in zero field depends on $\vec{k}$ and the temperature T. The T dependence comes in by virtue of the presence of $\langle S^Z \rangle$ in the expression for the energy, and $\langle S^Z \rangle$ decreases with increasing temperature. Dividing the spin wave energy by $\langle S^Z \rangle$ produces a temperature-independent "energy" that depends only on $\vec{k}$. There are computational advantages to normalizing the spin wave energies in this fashion since we shall be dealing with systems of various geometries and spin values. We define a dimensionless energy

$$\left. \begin{array}{c} x_{\vec{k}} = \dfrac{E_{\vec{k}}}{\left(E_{\vec{k}} \right)_{max}} = \dfrac{E_{\vec{k}}}{\hbar \langle S^Z \rangle J'} \\[3em] x = \dfrac{E}{\hbar \langle S^Z \rangle J'} \end{array} \right\} \qquad (82)$$

For a specific symmetry, g(x) will be unique, whereas a g(E)-against-E curve would be valid only for a particular S and T. J' has the same units as an exchange integral, (Energy) $\times$ (Angular momentum)$^{-2}$, and is dependent on the system under consideration. In systems with nearest-neighbor interactions only, for example, J' has the following values for simple cubic, body-centered cubic, and face-centered cubic geometries. From appendix A,

$$\left. \begin{array}{c} J'_{sc} = 12J \\[1.5em] J'_{bcc} = 16J \\[1.5em] J'_{fcc} = 16J \end{array} \right\} \qquad (83)$$

where J is the nearest-neighbor exchange interaction.

The Green's function $\langle G^0(E)_{\vec{k}} \rangle$ is a natural function from which to derive g(x) since it is also independent of temperature. To show this, we can substitute equations (45), (58), and (82) into equation (79). This gives for the zero-field Green's function,

$$\langle G^0(E)\rangle_{\vec{k}} = \frac{\hbar}{\pi J'} \frac{1}{x - x_{\vec{k}} - \rho x_{\vec{k}} \frac{1}{N} \sum\limits_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}}} = \langle G^0(x)\rangle_{\vec{k}} \tag{84}$$

a function which depends only on $\vec{k}$ and x for a given system and not on T or S.

To derive $g(x)$ formally, we go back to the Green's function of a specific disordered system and recast the diagonalized form of equation (54) as

$$G^0_{\alpha\beta}(E) = \frac{1}{2\pi} \frac{\delta_{\alpha\beta}}{\frac{E}{2\hbar^2\langle S^z\rangle} - B_{\alpha\alpha}} = \frac{1}{2\pi} \frac{\delta_{\alpha\beta}}{\frac{J'}{2\hbar}x - B_\alpha} = G^0_{\alpha\beta}(x) \tag{85}$$

where $B_{\alpha\beta}$ are the elements of the matrix

$$B_{fh} \equiv \frac{1}{2\hbar}\left[\delta_{fh}\sum_j J(f,j) - J(f,h)\right] \tag{86}$$

after diagonalization. Calling the discontinuity in G at the real axis in the energy plane the "imaginary part" of G, that is,

$$\mathcal{Im}\, G(E) \equiv \lim_{\epsilon\to 0} i\left[G(E + i\epsilon) - G(E - i\epsilon)\right] \tag{87}$$

we note that

$$\mathcal{Im}\, G^0_{\alpha\beta}(x) = \delta_{\alpha\beta}\delta\left(\frac{J'}{2\hbar}x - B_\alpha\right) = \frac{2\hbar}{J'}\delta_{\alpha\beta}\delta\left(x - \frac{2\hbar}{J'}B_\alpha\right) \tag{88}$$

The trace of the imaginary part of the Green's function, then, is the density of spin wave states

$$\frac{J'}{2\hbar}\frac{1}{N}\,\mathrm{Tr}\,\mathcal{Im}\, G^0_{\alpha\beta}(x) = \frac{1}{N}\sum_\alpha \delta\left(x - \frac{2\hbar}{J'}B_\alpha\right) \equiv g(x) \tag{89}$$

Obviously, $g(x)$ is normalized, since there are N eigenvalues B_α.

$$\int g(x)\, dx = \frac{1}{N} \sum_{\alpha} \int \delta\left(x - \frac{2}{J'} B_\alpha\right) dx = 1 \tag{90}$$

When the perfect-crystal Green's function (eq. (57)) is substituted in equation (89), the density of states for the ordered system becomes

$$g_0(x) = \frac{J'}{2\hbar} \frac{1}{N} \operatorname{Tr} \mathcal{I}m \, \Gamma^0_{\vec{k}}(E) = \frac{1}{N} \sum_{\vec{k}} \delta\left(x - x_{\vec{k}}\right) \tag{91}$$

Equation (91) is evaluated for lattices of sc, bcc, and fcc symmetries in appendix A. The evaluation involves replacing the sum over $\vec{k}$ states by a threefold integration over the first Brillouin zone.

Finding the density of states for a disordered system $g_\rho(x)$ is more complicated since $\langle G^0(E) \rangle_{\vec{k}}$ is more complicated than $\Gamma^0_{\vec{k}}(E)$. Using equation (79) in equation (89) gives

$$g_\rho(x) = \lim_{\epsilon \to +0} \frac{i}{2\pi} \frac{1}{N} \sum_{\vec{k}} \left(\frac{1}{x + i\epsilon - x_{\vec{k}} - \rho x_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x + i\epsilon - x_{\vec{k}'}}} \right.$$

$$\left. - \frac{1}{x - i\epsilon - x_{\vec{k}} - \rho x_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - i\epsilon - x_{\vec{k}'}}} \right) \tag{92}$$

The self-energy term that appears in the disordered Green's function adds complexity to the problem in that $g_\rho(x)$ cannot be written as a sum of delta functions. Before summing the two terms inside the parentheses of equation (92), it is advisable to rewrite the sum in the self-energy term in a different form. Using the perfect crystal $g_0(x)$ just calculated,

$$\frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x \pm i\epsilon - x_{\vec{k}'}} = \int \frac{x' g_0(x')}{x \pm i\epsilon - x'} \, dx' \tag{93}$$

From the theory of functions of a complex variable (ref. 34), we know that

$$\int \frac{x' g_0(x')}{x - x' \pm i\epsilon} \, dx' = P \int \frac{x' g_0(x')}{x - x'} \, dx' \mp i\pi x g_0(x) \tag{94}$$

where the P before the integral sign indicates the Cauchy principle value. Now, equation (92) can be written

$$g_\rho(x) = \begin{cases} \rho x g_0(x) \dfrac{1}{N} \displaystyle\sum_{\vec{k}'} \dfrac{x_{\vec{k}}}{R\left(x, x_{\vec{k}}\right)^2 + I\left(x, x_{\vec{k}}\right)^2} & \text{for } 0 \le x \le 1 \\[4mm] \dfrac{1}{N} \displaystyle\sum_{\vec{k}} \delta\left[R\left(x, x_{\vec{k}}\right)\right] & \text{for } 1 \le x \end{cases} \tag{95}$$

where

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$$R\left(x, x_{\vec{k}}\right) \equiv x - x_{\vec{k}} - \rho x_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}}$$

$$I\left(x, x_{\vec{k}}\right) \equiv \pi \rho x_{\vec{k}} x g_0(x) \tag{96}$$

Replacing the sum over the first Brillouin zone in equation (95) by an energy integration involving the density of states puts $g_\rho(x)$ into a form more convenient for computational purposes:

$$g_\rho(x) = \begin{cases} \rho x g_0(x) \displaystyle\int \frac{x' g_0(x')}{R(x,x')^2 + I(x,x')^2} \, dx' & \text{for } 0 \leq x \leq 1 \\[4ex] \displaystyle\int g_0(x') \delta\left[R(x,x')\right] dx' & \text{for } 1 < x \end{cases} \tag{97}$$

where the vector sum in $R(x, x')$ has also been replaced by an integral:

$$R(x, x') = x - x' - \rho x' P \int \frac{x'' g_0(x'')}{x - x''} \, dx'' \tag{98}$$

Equation (97) is the density of spin wave states for a disordered ferromagnet, the quantity that will allow us to calculate the effect of disorder on magnetic and thermodynamic properties. But $g_\rho(x)$ is important and interesting in its own right since it shows the manner in which the energy states are redistributed when disorder is introduced in the lattice. The physical insights gained from studying density-of-states curves are helpful in determining when disorder will and will not be important.

Although we discuss in detail the density-of-states curves of the three cubic structures with and without disorder in the section APPLICATION TO SPECIFIC SYSTEMS, there are general remarks and observations which can be made at this stage. For example, the shape of the density-of-states curve at low energies is important in the calculation of low-temperature properties since in Boson systems it is only these low-energy states that are occupied when $T \to 0$. We can get information concerning the

low-energy states in the disordered system by investigating $\langle G^0(x) \rangle_k$ in the limit of small x. Since

$$\lim_{x \to 0} \frac{1}{N} \sum_{\vec{k}} \frac{x_{\vec{k}}}{x - x_{\vec{k}}} = -1 \tag{99}$$

the Green's function for small x can be written

$$\langle G^0(x) \rangle_{\vec{k}} \underset{x \to 0}{\simeq} \frac{\hbar}{\pi J'} \frac{1}{x - x_{\vec{k}}(1 - \rho)} \tag{100}$$

The poles occur when $x = (1 - \rho)x_k$. Another way of stating this is that the introduction of disorder has scaled the energy of the lowest eigenstates downward by the factor $1 - \rho$. There are more spin waves, then, at lower energies in the disordered ferromagnet, and we would expect $g_\rho(x)$ to be accordingly larger than $g_0(x)$ for x near zero. We can be more quantitative about this effect without becoming too complicated.

At low energies the dispersion relation for any crystal must be quadratic

$$x_{\vec{k}} \simeq C_{st} \vec{k}^2 \qquad \text{for small } \vec{k} \tag{101}$$

where C_{st} is some constant which depends on the lattice structure. In this low-energy domain we can calculate the density of states by integrating over a small sphere of radius k_1:

$$g_0(x) \underset{x \to 0}{\simeq} \frac{1}{\frac{4}{3}\pi k_1^3} \int_0^{k_1} 4\pi k^2 \left(x - C_{st}\vec{k}^2 \right) dk = \frac{3}{2C_{st}^{3/2}k_1^3} \sqrt{x} \tag{102}$$

The quadratic dispersion relation results in square-root behavior for the density of states at low energies. If these lowest states had their energies scaled downward by the factor $1 - \rho$,

$$x_k' = (1 - \rho)C_{st}\vec{k}^2 \quad . \tag{103}$$

what would be the effect on the density of states? Going through the procedures of equation (102) again yields

$$g_\rho(x) \underset{x \to 0}{\simeq} (1 - \rho)^{-3/2} g_0(x) \qquad (104)$$

The effect of disorder on the low-energy density of states is to increase the density by $(1 - \rho)^{-3/2}$. This is an important result of the theory, and this factor will reappear when discussing low-temperature properties such as magnetization and specific heat. This increased number of low-energy states is a reflection of the decrease in the exchange coupling between some pairs of spins that comes about with the introduction of disorder and the corresponding decrease in the spin wave energy.

There will be pairs of spins in a disordered ferromagnet that are coupled more strongly than any in the corresponding perfect crystal. We would expect, therefore, that there are spin wave states in the disordered system with energies higher than any in the ordered system. Inspection of equation (97) does indeed show that $g_\rho(x)$ can be finite above $x = 1$; whereas, $g_0(x) = 0$ for $x > 1$.

We can make additional general remarks concerning the effects of disorder on the density of states in regard to the moments of $g_\rho(x)$, where the nth moment of $g_\rho(x)$ is defined

$$\int x^n g_\rho(x)\, dx \qquad (105)$$

Obviously, the zeroth moment is unaffected by ρ since $g_\rho(x)$ is always normalized by the way the density of states is defined. The same is true of the first moment; that is, the average spin wave energy does not change with disorder. We can show this with the help of equation (89)

$$\int xg(x)\, dx = \int x \frac{1}{N} \sum_\alpha \delta\left(x - \frac{2\hbar}{J'} B_\alpha\right) dx = \frac{2\hbar}{J'} \frac{1}{N} \sum_\alpha B_\alpha \qquad (106)$$

The average spin wave energy is seen to depend on the trace of matrix B defined in equation (86), a quantity depending only on the average exchange coupling which is independent of the disorder parameter ρ.

The negative first moment, however, does depend on the amount of disorder. We will show later that this quantity is proportional to the reciprocal of the Curie temperature. In the same manner as above and using equation (85),

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$$\int \frac{g(x)}{x} \, dx = \frac{J'}{2\hbar} \frac{1}{N} \sum_{\alpha} \frac{1}{B_\alpha} = \frac{-J'}{2\hbar} 2\pi \, \text{Tr} \, G^0(0) \tag{107}$$

The trace of the Green's function at zero energy, then, is proportional to the negative first moment. If we evaluate this for the ensemble-averaged Green's function (eq. (84)), we get

$$\int \frac{g_\rho(x)}{x} \, dx = \frac{1}{(1-\rho)N} \sum_{\vec{k}} \frac{1}{x_{\vec{k}}} \tag{108}$$

That is, the minus first moment varies with the amount of disorder as $(1-\rho)^{-1}$.

To summarize, then, even before investigating the density of states of specific systems and their relation to disorder, we have been able to deduce several general features:

(1) We expect the low-energy portion of the $g_\rho(x)$-against-x curve to get larger with increasing disorder as $(1-\rho)^{-3/2}$.

(2) We expect $g_\rho(x)$ to extend above the energy band of the perfect crystal. With these changes of shape:

(3) The area under the curve will remain unchanged.

(4) The average energy will remain unchanged.

(5) The negative first moment of the curve will vary with disorder as $(1-\rho)^{-1}$.

It is interesting to see how these observations are verified later in the detailed density-of-state calculations.

Correlation Functions for Spin Greater than 1/2

Having found the Green's function for a disordered ferromagnet, the central problem becomes the derivation of the corresponding correlation function from which we hope to calculate the magnetization, or $\langle S^z \rangle$. The correlation function that equation (34) allows us to calculate from the Green's function (79) is the spatial Fourier transform of $\left\langle e^{aS^z_g} S^-_g S^+_f(t) \right\rangle$, which we label $\psi(\vec{k}, a)$:

$$\psi(\vec{k}, a) \equiv \sum_f \left\langle e^{aS_g^z} S_g^- S_f^+(t) \right\rangle e^{-i(f-g)\cdot\vec{k}}$$

$$= \lim_{\epsilon \to 0} \frac{i}{\hbar} \int_{-\infty}^{\infty} \frac{\left\langle G^a(E + i\epsilon) \right\rangle_{\vec{k}} - \left\langle G^a(E - i\epsilon) \right\rangle_{\vec{k}}}{\exp\left(\frac{E}{kT}\right) - 1} e^{-i(E/\hbar)t} \, dE \qquad (109)$$

Substitution of equation (79) into this equation would allow the straightforward calculation of $\psi(\vec{k}, a)$, but this correlation function is useful for the $S = 1/2$ case only. For example, for $a = 0$

$$\frac{1}{N} \sum_{\vec{k}} \psi(\vec{k}, 0) e^{i(f-g)\cdot\vec{k}} = \left\langle S_g^- S_f^+(t) \right\rangle \qquad (110)$$

Setting $t = 0$ and $f = g$ gives, for the spin-1/2 system, the magnetization as discussed in equations (40) and (41):

$$\hbar \langle S_f^z \rangle = \frac{\hbar^2}{2} - \left\langle S_f^-(0) S_f^+(0) \right\rangle \qquad (111)$$

For systems with $S > 1/2$, however, the magnetization cannot be calculated from $\psi(\vec{k}, a)$ as it stands. The added complexity inherent in higher spin systems results from the ambiguity in the $\langle (S^z)^2 \rangle$ term in equation (40), which we repeat here for convenience:

$$\langle S_f^- S_f^+ \rangle = S(S + 1)\hbar^2 - \left\langle \left(S_f^z\right)^2 \right\rangle - \hbar \langle S_f^z \rangle \qquad (112)$$

This term is not a variable when $S = 1/2$ since it has the same value for the two possible spin configurations. Callen (ref. 19) has developed a technique for crystalline systems that enables one to relate $\langle S_f^- S_f^+ \rangle$ and $\Theta(a)$ to $\langle S_f^z \rangle$ for any spin value. We can use some of his results to extend the theory of disordered ferromagnetic systems to higher spins. The technique involves utilizing the dependence of $\psi(\vec{k}, a)$ and $\Theta(a)$ on the pa-

rameter a and the solution of an auxiliary differential equation in a. Only the essentials of the Callen method are presented here.

The correlation function that is of main interest is the Fourier transform of equation (109), specifically the case where the site labels are the same and $t = 0$,

$$\left\langle e^{aS_f^z} S_f^- S_f^+ \right\rangle .$$

$$\left\langle e^{aS_f^z} S_f^- S_f^+ \right\rangle = \frac{1}{N} \sum_{\vec{k}} \psi(\vec{k}, a) \tag{113}$$

The dependence of the right side of equation (113) on a is contained in $\Theta(a)$, which can be factored out to leave

$$\left\langle e^{aS_f^z} S_f^- S_f^+ \right\rangle = \Theta(a) \Phi \tag{114}$$

where

$$\Phi = \frac{1}{N} \frac{i}{\hbar} \lim_{\epsilon \to 0} \sum_{\vec{k}} \int_{-\infty}^{\infty} \frac{\hbar}{2\pi} \left[\frac{1}{E + i\epsilon - E_{\vec{k}} - \rho E_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x + i\epsilon - x_{\vec{k}'}}} - \frac{1}{E - i\epsilon - E_{\vec{k}} - \rho E_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - i\epsilon - x_{\vec{k}'}}} \right] dE \tag{115}$$

The technique is to represent equation (114) as a differential equation by writing both $\left\langle e^{aS_f^z} S_f^- S_f^+ \right\rangle$ and $\Theta(a)$ in terms of derivatives of $\left\langle e^{aS_f^z} \right\rangle$ with respect to a. This differential equation is

$$\frac{d^2}{da^2} \left\langle e^{aS_f^Z} \right\rangle + \frac{\hbar \left[(1 + \Phi)e^{a\hbar} + \Phi\right]}{(1 + \Phi)e^{a\hbar} - \Phi} \frac{d}{da} \left\langle e^{aS_f^Z} \right\rangle - S(S + 1)\hbar^2 \left\langle e^{aS_f^Z} \right\rangle = 0 \qquad (116)$$

which has for its solution

$$\left\langle e^{aS_f^Z} \right\rangle = \frac{\Phi^{2S+1}e^{-a\hbar S} - (1 + \Phi)^{2S+1}e^{a\hbar(S+1)}}{\left[\Phi^{2S+1} - (1 + \Phi)^{2S+1}\right]\left[(1 + \Phi)e^{a\hbar} - \Phi\right]} \qquad (117)$$

Finally, $\langle S^Z \rangle$ comes from equation (117) by differentiation.

$$\langle S^Z \rangle = \frac{d}{da} \left\langle e^{aS^Z} \right\rangle \Bigg|_{a=0} = \hbar \frac{(S - \Phi)(1 + \Phi)^{2S+1} + (S + 1 + \Phi)\Phi^{2S+1}}{(1 + \Phi)^{2S+1} - \Phi^{2S+1}} \qquad (118)$$

By this equation one can solve for $\langle S^Z \rangle$ self-consistently for any spin.

The Φ's in equation (118) are the analogous quantities in a disordered system to Callen's perfect-crystal Φ's, which we call Φ^0. We define Φ^0 as the sum in k-space of the occupation numbers $\left[\exp(E_{\vec{k}}/kT) - 1\right]^{-1}$ for spin waves with reciprocal lattice vectors $\vec{k}$. This can be expressed in terms of the density of states as

$$\Phi^0 = \frac{1}{N} \sum_{\vec{k}} \frac{1}{\exp\left(\dfrac{E_k}{kT}\right) - 1} = \int \frac{g_0(x) \, dx}{\exp\left(\dfrac{\hbar J'\langle S^Z \rangle x}{kT}\right) - 1} \qquad (119)$$

where $g_0(x)$ is the crystal density of states. Equation (115) defining Φ for a disordered system can easily be put into this form with the help of equation (92):

$$\Phi = \int \frac{g_\rho(x) \, dx}{\exp\left(\dfrac{\hbar J'\langle S^Z \rangle x}{kT}\right) - 1} = \Phi(\rho, T) \qquad (120)$$

APPLICATION TO SPECIFIC SYSTEMS

In the preceding main section the general theory for disordered ferromagnets was developed. This included the derivation of a Green's function and the resulting density of states for disordered systems. These quantities allowed the self-consistent evaluation of the magnetization. The theory has been developed to a stage where we can now study in more detail the effect of disorder on specific systems. Which properties and which structures are affected most by disorder and in what temperature range? Is the spin value important and what does the theory predict in the case of substitutional disorder as in the mixed chalcogenides of europium? These and other questions are treated in this section.

Cubic Systems

Density of states. - From equation (97) the density of spin wave states for a disordered ferromagnet can be calculated provided the density of states of the corresponding perfect crystal is known. We have calculated the density of states for sc, bcc, and fcc crystals, $g_{sc}(x, 0)$, $g_{bcc}(x, 0)$ and $g_{fcc}(x, 0)$, respectively, and shall summarize these calculations here. A more detailed version of these calculations is presented in appendix A. The technique of Bowers and Rosenstock (ref. 35) was employed in these calculations in which the density of states is written as

$$g(x) = \frac{1}{N} \sum_{\vec{k}} \delta\left(x - x_{\vec{k}}\right) \tag{121}$$

The dispersion relation, the energy's dependence on the wave vector $\vec{k}$, is substituted in the argument of the delta function; and the sum over k-states is replaced by three integrations in the usual way. The dispersion relations for the normalized, dimensionless energy defined in equation (82) are, for nearest-neighbor interactions,

$$x_{\vec{k}} = \frac{1}{2} - \frac{1}{6}\left(\cos ak_x + \cos ak_y + \cos ak_z\right) \quad \text{for sc crystals}$$

$$x_{\vec{k}} = \frac{1}{2} - \frac{1}{2}\left(\cos \frac{a}{2}k_x \cos \frac{a}{2}k_y \cos \frac{a}{2}k_z\right) \quad \text{for bcc crystals}$$

$$x_{\vec{k}} = \frac{3}{4} - \frac{1}{4}\left(\cos \frac{a}{2}k_x \cos \frac{a}{2}k_y + \cos \frac{a}{2}k_y \cos \frac{a}{2}k_z + \cos \frac{a}{2}k_z \cos \frac{a}{2}k_x\right)$$

$$\text{for fcc crystals}$$

$$(122)$$

where a is the edge length of the unit cube.

Performing the first of the integrations implied in equation (121) results in an integrand for the second integration that can be put into a standard form of a complete elliptic integral (ref. 36) of the first kind. The final integration was performed numerically on an IBM 7044-7094 using Gaussian quadrature and, where functions were smoother, Simpson's method.

The densities of states for perfect-crystal sc, bcc, and fcc ferromagnets are shown in figure 2. Notice that singularities appear in all three spectra. For both the bcc and fcc cases the density of states has logarithmic singularities, and there are discontinuities of slope in the sc and fcc spectra. The slope discontinuities at $x = 1/3$ and $x = 2/3$ on the sc curve and at $x = 3/4$ on the fcc curve all occur where the density of states has square-root behavior with slope approaching infinity. These are the familiar van Hove (ref. 37) singularities that appear because of the periodicity of the lattice.

When disorder is introduced in a ferromagnet, the density of states undergoes a more or less continuous change of shape, so that, at small values of the disorder parameter, $g_\rho(x)$ is very similar in appearance to $g_0(x)$. Exceptions to this continuous change of $g_\rho(x)$ with ρ occur where there are infinities in $g_0(x)$. These infinities become finite when ρ becomes nonzero. The reasons for this will be discussed shortly. The most noticeable general effect of disorder on the density of states is to enlarge the number of spin wave states at lower energies, to decrease the number in the upper portion of the perfect-crystal energy band, and then to extend the maximum energy to higher values. These features can be observed in the energy spectra of disordered systems, figure 3, where $g_{sc}(x,\rho)$, $g_{bcc}(x,\rho)$ and $g_{fcc}(x,\rho)$ are plotted with respect to x for various values of the disorder parameter ρ. All the shape changes that depend on disorder are caused by the self-energy term, from which all ρ-dependence originates. A closer look at this term would be useful.

We recall that ρ came into the disordered Green's function (79) through a term which mixed the energy variable x and the eigenvalue x_k, the so-called self-energy,
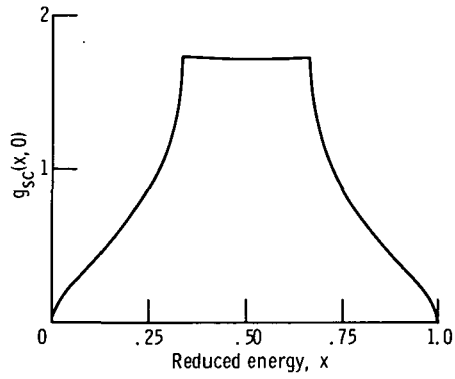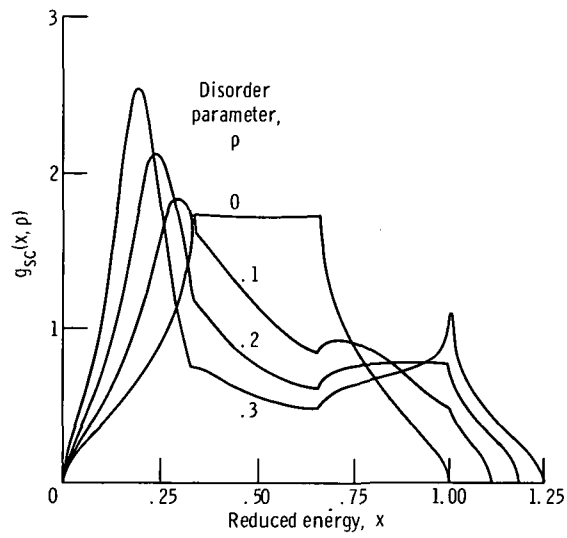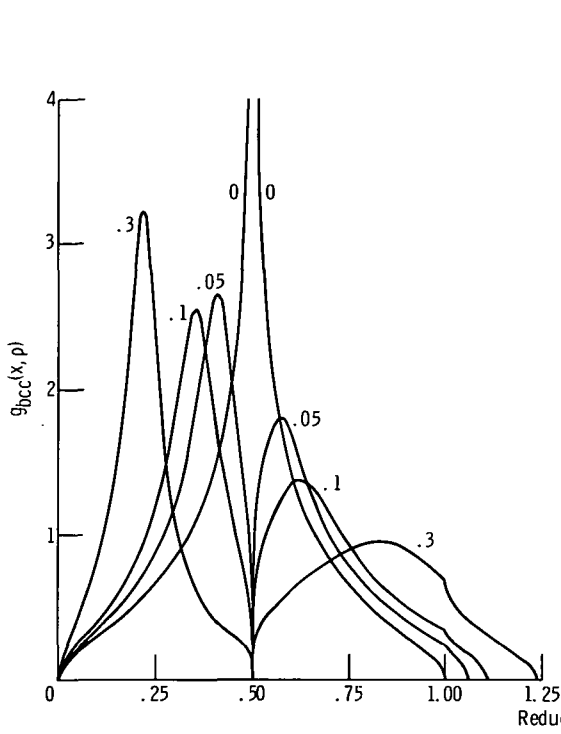
(a) Simple cubic.
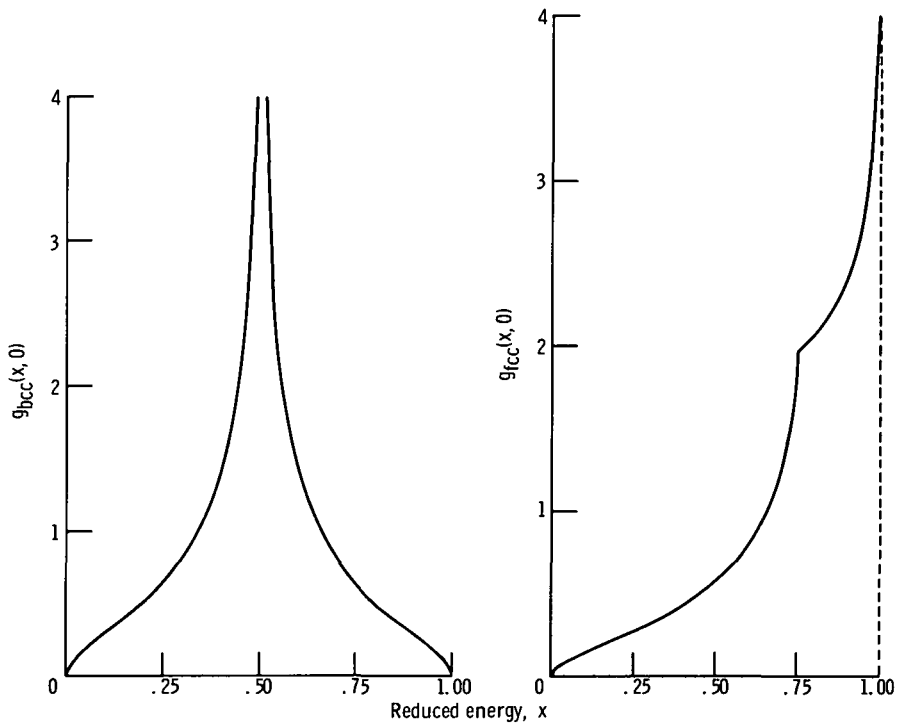
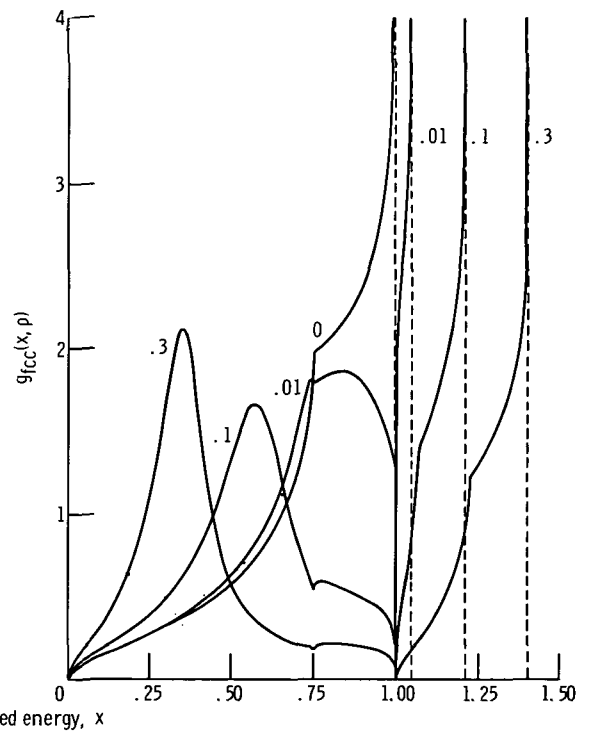(b) Body-centered cubic.

(c) Face-centered cubic.

Figure 2. - Density of spin wave states for perfect ferromagnets as function of reduced energy.

(a) Simple cubic.

(b) Body-centered cubic.

(c) Face-centered cubic.

Figure 3. – Density of spin wave states for disordered ferromagnets as function of reduced energy.

$$\rho x_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}} \tag{123}$$

With its linear dependence on the disorder parameter, the self-energy vanishes with disorder, reducing the disordered Green's function to the perfect-crystal function. Replacing the sum by an integration allows us to rewrite the self-energy as

$$\rho x_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}} = \rho x_{\vec{k}} \Big[-1 + xA(x)\Big] \tag{124}$$

where

$$A(x) \equiv \sum_{\vec{k}'} \frac{1}{x - x_{\vec{k}'}} = P \int_0^1 \frac{g_0(x')}{x - x'} dx' \tag{125}$$

The function $A(x)$ is the sum over all the perfect-crystal eigenstates of the reciprocal of the difference in energy of the variable x and the eigenenergy x_k. We would expect, then, $A(x)$ to be negative for x near the low end of the energy band; to be positive for x near the high end of the band; and to fall off to zero like x^{-1} as x became larger than $x = 1$, the perfect-crystal energy maximum. Figure 4 shows these features of $A(x)$ for the cubic lattices. The function $-1 + A(x)$, which contains all the x-dependence of the self-energy, is also plotted in the same figure.

From these plots it can be seen that $A(x)$ has discontinuities in slope at the same energy values where slope discontinuities appear in $g_0(x)$. Notice also that $A(x)$ and $g_0(x)$ have infinities at the same energy value. By writing the expression for the disordered density of states (eq. (97)) in a way that mentions $A(x)$ explicitly, it is easier to see the effect of the critical points of the self-energy on $g_\rho(x)$.
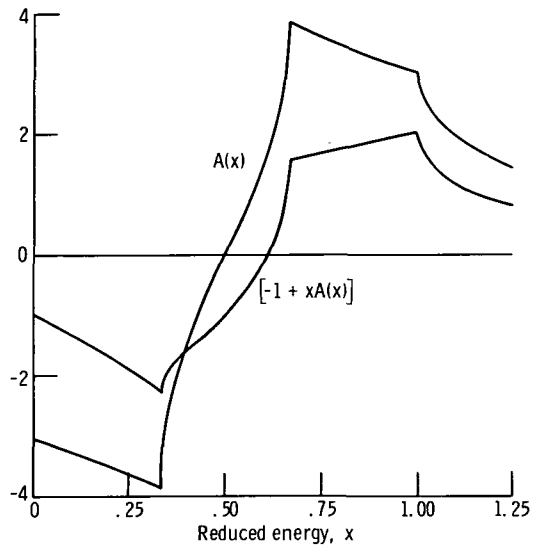
(a) Simple cubic.
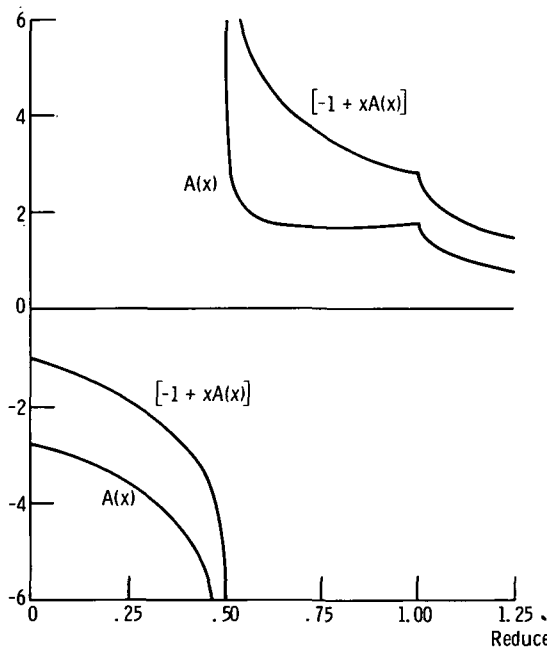
(b) Body-centered cubic.
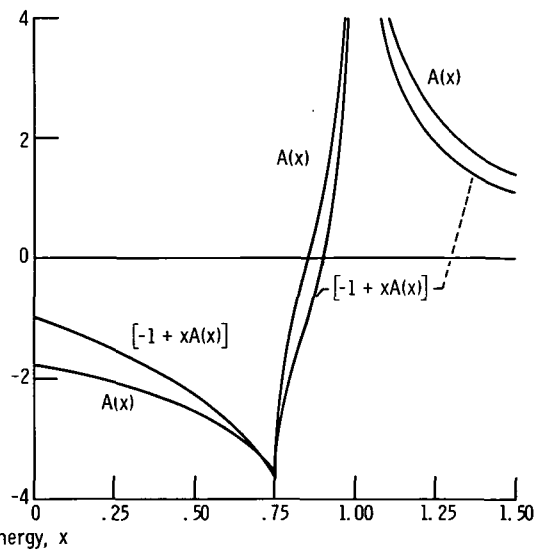
(c) Face-centered cubic.

Figure 4. - A(x) and $[-1 + xA(x)]$ as function of reduced energy for various cubic systems.

$$
g_\rho(x) = \begin{cases} g_0(x)\rho x \displaystyle\int_0^1 \frac{x'g_0(x')\,dx'}{\left(x - x'\{1 + \rho[-1 + xA(x)]\}\right)^2 + \left[\pi\rho x g_0(x)x'\right]^2} & 0 \le x \le 1 \\[20pt] \{1 + \rho[-1 + xA(x)]\}^{-1} g_0\left(\dfrac{1}{1 + \rho[-1 + xA(x)]}\right) & 1 < x \end{cases}
$$

$$(126)$$

In the region $0 \le x \le 1$, $g_\rho(x)$ appears as $g_0(x)$ modulated by an integral function of x. Since slope discontinuities will appear in the integrand only at energies where they appear in $g_0(x)$, $g_\rho(x)$ will exhibit discontinuities in slope at the same values of x but at an altered value for the state density. In this same region, the perfect-crystal energy band, a more dramatic change occurs where $g_0(x)$ has infinities, that is, at $x = 1/2$ in the bcc system and at $x = 1$ in the fcc system. Since $A(x)$ exhibits the same type of singularities at these points, inspection of equation (126) shows that the integrand, with the square of $A(x)$ and $g_0(x)$ in the denominator, becomes vanishingly small. This results in a depression of $g_\rho(x)$ near these values of x at finite values of ρ. That is, the introduction of the smallest amount of disorder causes the density of states near these critical points to change from logarithmically infinite behavior to a decay to zero. Figures 3(b) and (c) show that the energy range over which this depression in $g_\rho(x)$ occurs is small for minor disorder and grows with ρ.

We have observed that the maximum allowable spin wave energy becomes larger as disorder is introduced. The behavior of $g_\rho(x)$ at energies above the perfect-crystal band (i.e., at $x > 1$) is related to the factor $\{1 + \rho[-1 + xA(x)]\}^{-1}$, which modulates both the function g_0 and the argument of g_0. There are two patterns of behavior for $g_\rho(x)$ for $x > 1$, one for the sc and bcc systems and the other for the fcc system. In the former case, $A(x)$ is finite at $x = 1$ and falls monotonically towards zero for increasing x such that the aforementioned modulating factor goes from a positive finite value to unity in the same range. The overall effect on the sc and bcc state density is to extend the upper portion of the spectra preserving the shape, which is of square-root nature. In this energy range the effect of disorder is to produce a high-energy tail, with the spectra showing a slight discontinuity in slope at $x = 1$.

The behavior of the state density in this higher energy region is not the same for the fcc system. Because $A(x)$ falls off from an infinite value at $x = 1$, the factor $\{1 + \rho[-1 + xA(x)]\}^{-1}$ is zero at this value of x and climbs asymptotically to unity. This has two effects. First, $g_{fcc}(x,\rho)$ is depressed immediately above $x = 1$ because of this factor's appearance as a coefficient of g_0. Secondly, the argument of g_0 in the right side of equation (126) is swept through the entire energy range as x goes from 1

to the new energy maximum. The result for the fcc case is that not just the high-energy end of the crystal spectrum is reporduced in a modulated way but the entire crystal-state density reappears in the region $1 < x < x_{max}$, although rescaled in a nonlinear fashion. That is to say, in the energy region above the crystal band, $g_{fcc}(x, \rho)$ begins at zero, exhibits a slope discontinuity, and finally has a logarithmic infinity at the top of the disordered energy band.

An important part of the density of states is the low-energy end. Since spin waves act as bosons, at low temperatures only the low-energy spin wave states are occupied. In calculating magnetic properties at low temperatures, then, only the shape of the density-of-states curve at low energies is important. As has been mentioned in the previous chapter, disorder increases the state density by the factor $(1 - \rho)^{-3/2}$ while preserving the square-root behavior. Since the limiting behavior of the perfect-crystal state densities has been evaluated in appendix A, we are able to write the low-energy part of the disordered density of states:

$$\left.\begin{array}{l} g_{sc}(x, \rho) \underset{x \to 0}{\simeq} \dfrac{6\sqrt{3}}{(1 - \rho)^{3/2}\pi^2} \sqrt{x} \\[20pt] g_{bcc}(x, \rho) \underset{x \to 0}{\simeq} \dfrac{8}{(1 - \rho)^{3/2}\pi^2} \sqrt{x} \\[20pt] g_{fcc}(x, \rho) \underset{x \to 0}{\simeq} \dfrac{4}{(1 - \rho)^{3/2}\pi^2} \sqrt{x} \end{array}\right\} \tag{127}$$

Magnetization and the Curie temperature. - The cooperative nature of ferromagnetism becomes very evident when we study the spontaneous magnetization of a ferromagnet. The dependence of the relative magnetization on temperature for a Heisenberg ferromagnet is shown in figure 5, where we have plotted $\langle S^z \rangle /S = \sigma$ for a simple cubic crystal of spin 1/2. The behavior near the Curie point T_C is of particular interest. Just below T_C the magnetization drops steeply and, in fact, the slope of the curve becomes negatively infinite at T_C. A small increase in the amount of thermal fluctuations in this region produces a relatively large decrease in the magnetization to zero, an abrupt change from a state where the spins act in consort to the uncooperative paramagnetic phase. This precipitates an intriguing question. Since the spontaneous magnetization is very sensitive to thermal fluctuations near the Curie temperature, would the introduction of disorder have a significant effect on the magnetic behavior in this region? The questions of whether the Curie temperature would be altered by disorder and whether the slope discontinuity in the magnetization curve at T_C remains are among those answer-
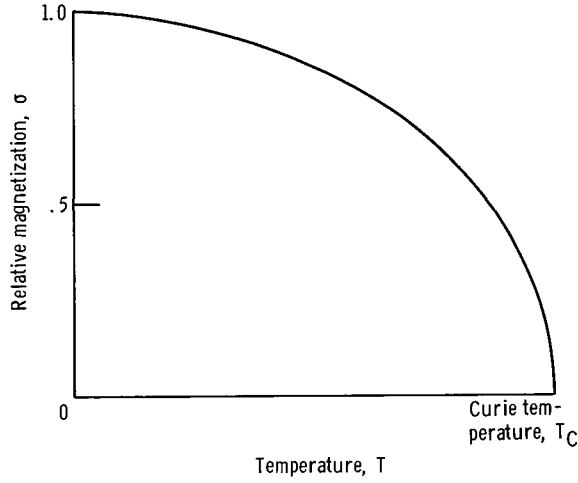
Figure 5. - Relative magnetization as function of temperature for simple cubic (sc) systems of spin 1/2.

ed in this section, which treats the effects of disorder on magnetization over the entire temperature range.

In the preceding main section the expression for the averaged z-component of spin was given for Heisenberg systems of any spin value. For the sake of convenience we shall rewrite it in the form of the relative magnetization,

$$\sigma = \frac{\langle S^z \rangle}{S} = \frac{1}{S} \frac{(S - \Phi)(1 + \Phi)^{2S+1} + (S + 1 + \Phi)\Phi^{2S+1}}{(1 + \Phi)^{2S+1} - \Phi^{2S+1}} \tag{128}$$

where Φ in turn is a function of T, S^z, and ρ,

$$\Phi = \int \frac{g_\rho(x)}{\exp\left(\frac{\hbar J' \langle S^z \rangle x}{kT}\right) - 1} \, dx \tag{129}$$

so that equation (128) is a self-consistent expression for the magnetization.

Upon expansion, equation (128) has the form of the quotient of two power series,

$$\sigma = \frac{1 + a_1 \Phi + a_2 \Phi^2 + \ldots + a_{2S-1} \Phi^{2S-1}}{1 + b_1 \Phi + b_2 \Phi^2 + \ldots + b_{2S} \Phi^{2S}} \tag{130}$$

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where the constants a_i, $(i = 1, 2, \ldots, 2S - 1)$, and b_j, $(j = 1, 2, \ldots, 2S)$, depend on the value of S. Before presenting the results of equation (130) for systems of various spin, structure, and disorder, we are interested in the behavior of σ for two limiting cases: T approaching zero and T approaching T_C from below (i.e., $T \to T_C-$). For low temperatures it will be important to know the coefficients of the lowest power of Φ, a_1 and b_1; and for T near T_C the coefficients of the higher powers of Φ will be needed. These are

$$\left. \begin{array}{l} a_1 = \dfrac{2S^2 + S - 1}{S} \\[4mm] b_1 = 2S + 1 \end{array} \right\} \tag{131}$$

and

$$\left. \begin{array}{ll} a_{2S-1} = \dfrac{1}{3}(2S^2 + 3S + 1) & b_{2S} = 2S + 1 \\[4mm] a_{2S-2} = \dfrac{1}{6}(4S^3 + 4S^2 - S - 1) & b_{2S-1} = S(2S + 1) \\[4mm] a_{2S-3} = \dfrac{1}{10}(4S^4 - 5S^2 + 1) & b_{2S-2} = \dfrac{1}{3}S(2S + 1)(2S - 1) \end{array} \right\} \tag{132}$$

Low temperatures: When the temperature approaches absolute zero we expect all ferromagnets, no matter how weak or strong the exchange interactions, to have all their spins alined parallel. In the absence of thermal fluctuations there will be no spin reversals, and the ground state of a disordered ferromagnet will exhibit the identical magnetization as the corresponding perfect crystal. If, in the manner of Dyson (refs. 11 and 12) we were to expand σ in a power series in T, the leading term would be the same for both ordered and disordered ferromagnets, namely unity. The question which is of interest is where in this series does the effect of disorder first appear and how does the disorder parameter enter. It will be shown that all subsequent terms are affected by disorder.

At low temperatures, Φ is a small quantity because of the rapid exponential decay of the integrand as the energy increases. The factor $\left[\exp\left(E_k/kT\right) - 1\right]^{-1}$ in the integrand of Φ has an infinite, integrable peak at the zero of energy which, at low temperatures, serves to weight the density-of-states function heavily at the low-energy end.

Because the low-temperature magnetization is insensitive to the shape of the high-energy portion of $g(x)$, we are justified in using the low-energy form of $g_\rho(x)$ in the evaluation Φ in this region. Thus, for $H = 0$,

$$\Phi \underset{T \to 0}{\simeq} \int_0^\infty \frac{(1 - \rho)^{-3/2} C_{le} \sqrt{x}}{\exp\left(\frac{\hbar J' \langle S^z \rangle x}{kT}\right) - 1} \, dx \qquad (133)$$

where C_{le} is a constant whose value for various structures can be found from the low-energy forms of the state density in equations (127). The right side of equation (133) can be evaluated by recognizing that it can be put into an integral form of the Riemann zeta-function $\zeta(s)$ by a change of variable:

$$\zeta(s) = \frac{1}{\Gamma(s)} \int_0^\infty \frac{y^{s-1}}{e^y - 1} \, dy \qquad (134)$$

This gives for the low-temperature form of Φ

$$\Phi \underset{T \to 0}{\simeq} \frac{\pi^{1/2} C_{le} \zeta(3/2)}{2(1 - \rho)^{3/2}} \left(\frac{kT}{\hbar J' \langle S^z \rangle}\right)^{3/2} \qquad (135)$$

That is, for T near zero, Φ goes as $T^{3/2}$ and $(1 - \rho)^{-3/2}$.

Equation (130) in the limit of low T becomes

$$\sigma \underset{T \to 0}{\simeq} 1 + (a_1 - b_1)\Phi = 1 - \frac{1}{S}\Phi \qquad (136)$$

Substitution of equation (135) allows σ to be written to a first approximation as

$$\sigma \underset{T \to 0}{\simeq} 1 - \frac{\pi^{1/2} C_{le} \zeta(3/2)}{2 S^{5/2} (1 - \rho)^{3/2}} \left(\frac{kT}{\hbar^2 J'}\right)^{3/2} \qquad (137)$$

which is Bloch's well-known $T^{3/2}$ law revised to include the effects of disorder. Higher terms will also have dependence on ρ, but near $T = 0$ the effect of disorder is to increase the $T^{3/2}$ term by $(1 - \rho)^{-3/2}$, which means that the magnetization of a disordered ferromagnet will decrease more quickly than the corresponding perfect crystal in this temperature range.

Curie temperature: The energy of a spin wave is not uniquely determined by its reciprocal lattice vector but depends also on the magnetization. As the temperature approaches the Curie point from below and the magnetization drops to zero, there is a corresponding decrease in the spin wave energy. To evaluate Φ in this limit, we shall expand the exponential term of the integrand in a power series. If we set the constant quantities of the exponent equal to W,

$$W = \frac{\hbar^2 S J'}{k} \qquad . \tag{138}$$

then

$$\Phi = \int \frac{g_\rho(x)}{\dfrac{W\sigma x}{T} + \dfrac{1}{2!}\left(\dfrac{W\sigma x}{T}\right)^2 + \dfrac{1}{3!}\left(\dfrac{W\sigma x}{T}\right)^3 + \ldots} \, dx$$

$$= \frac{T}{W\sigma} \int \frac{g_\rho(x)}{x} \left[\frac{1}{1 + \dfrac{1}{2}\dfrac{W\sigma x}{T} + \dfrac{1}{6}\left(\dfrac{W\sigma x}{T}\right)^2 + \ldots} \right] dx \tag{139}$$

Because the quantity $W\sigma x/T$ is small near the Curie temperature, the term in the brackets in equation (139) can be rewritten as a power series that gives

$$\Phi \underset{T \to T_C^-}{\simeq} \frac{T}{W\sigma} \int \frac{g_\rho(x)}{x} \, dx - \frac{1}{2} \int g_\rho(x) \, dx + \frac{1}{12} \frac{W\sigma}{T} \int x g_\rho(x) \, dx + \ldots \tag{140}$$

We shall write an expression for $1/\sigma$, the inverse of equation (130); and since Φ is large near the Curie temperature, only its higher powers will be kept:

$$\frac{1}{\sigma}\underset{T\to T_C^-}{\simeq}\frac{b_{2S}\Phi^{2S} + b_{2S-1}\Phi^{2S-1} + b_{2S-2}\Phi^{2S-2}}{a_{2S-1}\Phi^{2S-1} + a_{2S-2}\Phi^{2S-2} + a_{2S-3}\Phi^{2S-3}} \qquad (141)$$

The quotient on the right side of equation (141) is a power series in σ/T containing only odd integer powers:

$$\frac{1}{\sigma}\underset{T\to T_C^-}{\simeq}\frac{3}{(S+1)W}\left[\int\frac{g_\rho(x)}{x}dx\right]\frac{T}{\sigma} + B_1\frac{\sigma}{T} + O\left(\frac{\sigma^3}{T^3}\right) \qquad (142)$$

More than the leading terms were kept in both the numerator and the denominator of equation (141) in order to verify the absence of even powers of σ/T and in order to be able to evaluate the constant B_1 in equation (142), which is

$$B_1 = \frac{(2S-1)(2S+3)W}{20(S+1)}\left[\int\frac{g_\rho(x)}{x}dx\right]^{-1} - \frac{(2S^2 - 2S - 1)W}{4(2S+1)(S+1)}\int xg_\rho(x)\,dx \qquad (143)$$

The Curie temperature can be evaluated from equation (142). Rearranging terms gives

$$\frac{1}{\sigma}\left[1 - \frac{3T}{(S+1)W}\int\frac{g_\rho(x)}{x}dx\right]\underset{T\to T_C^-}{\simeq} B_1\frac{\sigma}{T} \qquad (144)$$

Note that this equation has no solution for arbitrarily large T. For finite σ, the right side of the equation remains finite as $T \to \infty$, while the left side does not. It is an equation valid only to the temperature where σ vanishes. As σ, and therefore the right side of the equation, approaches zero, the left side will become infinite unless the expression in the brackets becomes zero. This condition determines T_C:

$$T_C = \frac{(S+1)W}{3}\left[\int \frac{g_\rho(x)}{x}\, dx\right]^{-1} \tag{145}$$

To put kT_C in terms of the exchange interaction energy,

$$\frac{kT_C}{\hbar^2 J'} = \frac{S(S+1)}{3}\left[\int \frac{g_\rho(x)}{x}\, dx\right]^{-1} \tag{146}$$

As mentioned in the previous main section the Curie temperature is inversely proportional to the negative first moment of the density of states. And it was shown in equations (107) and (108) that

$$\int \frac{g_\rho(x)}{x}\, dx = \frac{1}{(1-\rho)}\int \frac{g_0(x)}{x}\, dx \tag{147}$$

This implies that a disordered ferromagnet has a Curie temperature that is smaller than the T_C for the corresponding perfect crystal by the factor $1 - \rho$.

$$\left(T_C\right)_\rho = (1-\rho)\left(T_C\right)_{\rho=0} \tag{148}$$

The Curie temperature, then, is linear with the mean-square deviation of the exchange integrals.

The calculation of T_C is not complete until the integral of equation (27) is evaluated. Machine calculations of this integral have been performed, using the three state densities derived earlier. Our results agree with the work of Watson (ref. 38), who calculated similar integrals analytically. Thus, we have

$$
\left. \begin{array}{l}
\displaystyle\int \frac{g_{sc}(x, 0)}{x}\, dx = 3.033 \\[20pt]
\displaystyle\int \frac{g_{bcc}(x, 0)}{x}\, dx = 2.786 \\[20pt]
\displaystyle\int \frac{g_{fcc}(x, 0)}{x}\, dx = 1.793
\end{array} \right\} \tag{149}
$$

Finally, substituting for the structure-dependent factor J' by means of equation (83), and using equations (147) and (149), the Curie temperatures for disordered sc, bcc, and fcc structures of any spin value can be written in units of the nearest-neighbor exchange energy:

$$
\frac{kT_C}{\hbar^2 J} = \left\{ \begin{array}{ll}
\dfrac{4S(S + 1)(1 - \rho)}{3.033} = 1.319\, S(S + 1)(1 - \rho) & \text{for sc structures} \\[18pt]
\dfrac{16S(S + 1)(1 - \rho)}{3(2.786)} = 1.914\, S(S + 1)(1 - \rho) & \text{for bcc structures} \\[18pt]
\dfrac{16S(S + 1)(1 - \rho)}{3(1.793)} = 2.975\, S(S + 1)(1 - \rho) & \text{for fcc structures}
\end{array} \right. \tag{150}
$$

Although disorder has a significant effect on the Curie temperature, the shapes of the magnetization-against-temperature curves will be similar near T_C. From equation (144) it can be seen that the magnetization of a disordered ferromagnet will have the same square-root behavior as the perfect crystal with a negatively infinite slope at T_C. To first order, equation (144) can be written

$$
\sigma \underset{T \to T_C^-}{\simeq} \sqrt{\frac{T_C}{B_1}\left(1 - \frac{T}{T_C}\right)} \tag{151}
$$

Intermediate temperatures: Having investigated the spontaneous magnetization of disordered ferromagnets at low temperatures and near the Curie point, we shall present in this section the results of calculations of σ over the entire temperature range of the

ferromagnetic phase. These calculations have been carried out for various values of spin and disorder and for the three cubic structures.

The method used to generate the various σ-against-T curves is as follows. The Φ corresponding to some σ/T ratio was evaluated by a machine integration of equation (129). Equation (130) was then used to solve for σ for any desired spin system. To find the temperature corresponding to the value of σ, we divide by the original σ/T ratio. To show the effect of spin in the theory, these calculations were carried out for spins of 1/2, 1, and 7/2. The S = 7/2 case was included for two reasons. First, it provides an example of behavior for a system of spin significantly higher than the commonly studied S-values. Second, the europium chalcogenides, probably the best examples of Heisenberg ferromagnets, have spin values of 7/2 associated with the europium ions; and it is desirable to have calculations that are applicable to real systems. The appropriate equations for these values of S are

$$\sigma = \frac{1}{1 + 2\Phi} \quad \text{for } S = 1/2$$

$$\sigma = \frac{1 + 2\Phi}{1 + 3\Phi + 3\Phi^2} \quad \text{for } S = 1$$

$$\sigma = \frac{1 + \frac{54}{7}\Phi + \frac{180}{7}\Phi^2 + 48\Phi^3 + 54\Phi^4 + 36\Phi^5 + 12\Phi^6}{1 + 8\Phi + 28\Phi^2 + 56\Phi^3 + 70\Phi^4 + 56\Phi^5 + 28\Phi^6 + 8\Phi^7} \quad \text{for } S = 7/2$$

(152)

Figure 6 shows the magnetization curves for three perfect-crystal ferromagnets of
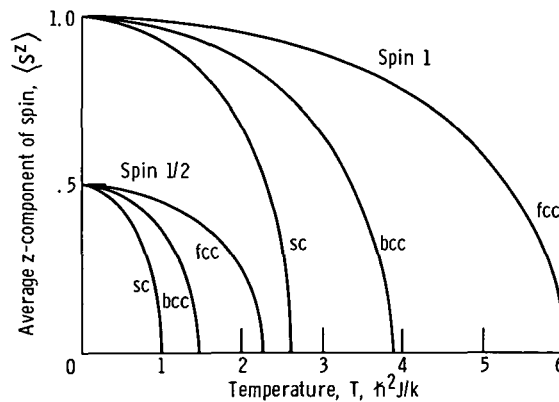

Figure 6. - Average z-component of spin $\langle S^z \rangle$ as function of temperature for simple cubic (sc), body-centered cubic (bcc), and face-centered cubic (fcc) systems for spins of 1/2 and 1.

sc, bcc, and fcc symmetries. Two values of spin, $S = 1/2$ and $S = 1$, are presented for each structure. In this figure the average z-component of spin is the ordinate rather than the relative magnetization σ. For the same value of spin the Curie temperature is seen to be highest for the fcc crystal, intermediate for the bcc, and lowest for the sc. This might be expected, since a spin in a fcc lattice interacts more strongly with the rest of the system, being involved with 12 nearest neighbors. The magnetic moments associated with ions in bcc and sc lattices interact with eight and six neighbors, respectively, and have correspondingly lower Curie temperatures.

The effect of the value of S can also be seen in figure 6. For a given structure the Curie temperature increases with S as $S(S + 1)$. Since the exchange interaction between two spins $(J_{ij}\vec{S}_i \cdot \vec{S}_j)$ increases as S^2, this increase in T_C is understandable.

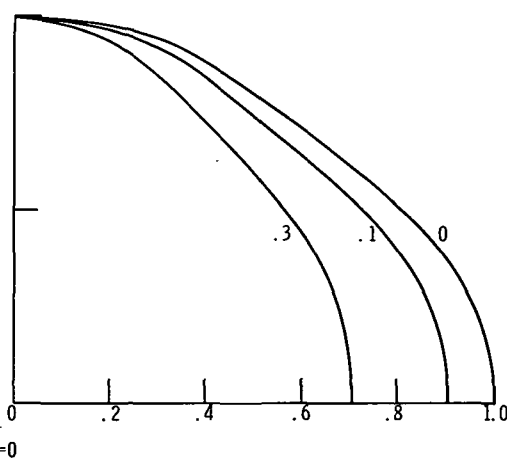The effect of disorder on the spontaneous magnetization is shown in figure 7 for a sc system of $S = 1/2$, a bcc system of $S = 1$, and a fcc system of $S = 7/2$. The be-
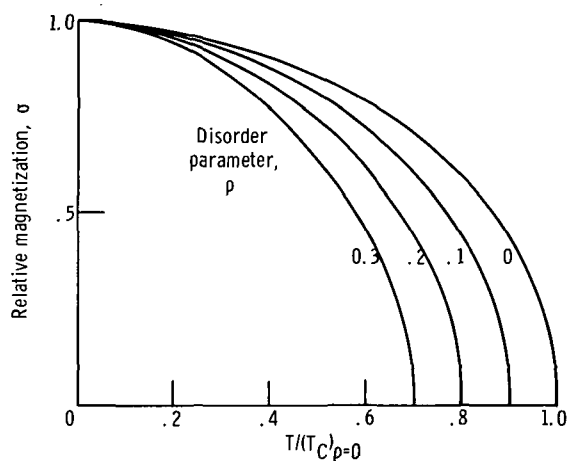
(a) Simple cubic systems of spin 1/2.
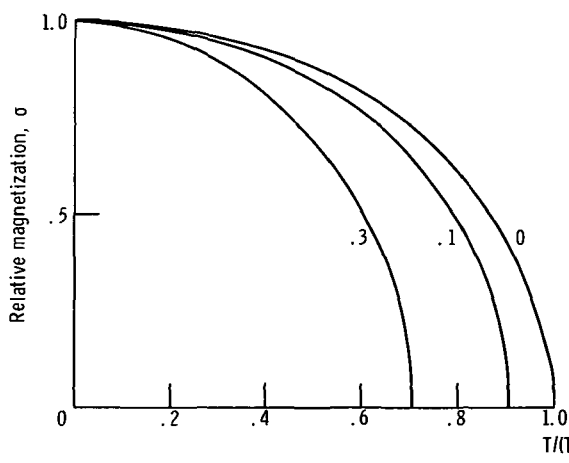
(b) Body-centered cubic systems of spin 1.

(c) Face-centered cubic systems of spin 7/2.

Figure 7. – Relative magnetization as function of temperature for disordered cubic systems of various spins.

havior in each case is qualitatively the same. At any finite temperature, the magnetization of a disordered ferromagnet is smaller than for the less disordered system; and the Curie point in each case decreases linearly with $1 - \rho$.

The σ-against-T curves for all the systems considered, independent of the structure or of the value of S or ρ, all start from a value of unity for σ at $T = 0$ and decrease monotonically to zero where there is a slope discontinuity at $T = T_C$. The behavior of these curves near $T = 0$ and $T = T_C$ is similar (although the displacement of T_C due to disorder is significant), having $T^{3/2}$ and $T^{1/2}$ dependence, respectively, with the coefficients of these terms being functions of ρ. Any changes in shape of these curves due to disorder, then, are subtle changes not easily discernible from the plots as they have been presented. Changes in shape due to ρ would be more easily studied by plotting σ against T for a system with various amounts of disorder in such a way that the curves intersect at $T = 0$ and $T = T_C$. That is, the temperature scale would be altered for each value of ρ to allow the T_C's to be superimposed. This procedure has been followed in figure 8(a), where bcc systems with $S = 1/2$ and with various amounts of disorder are presented. Displayed in this fashion, it can be seen that the effect of disorder is to flatten the curve shape. The change in behavior of the magnetization curves brought on by disorder are not just changes in scaling. This flattening effect of disorder is less strong at higher spin values, as can be seen from figure 8(b), where the same type of plot is done for a bcc system of $S = 7/2$. Recently, Sharon and Tsuei (ref. 39) have confirmed this flattening phenomenon in measurements on amorphous ferromagnetic Fe-Pd-P alloys.

Specific heat. - To calculate the magnetic or spin contribution to the specific heat
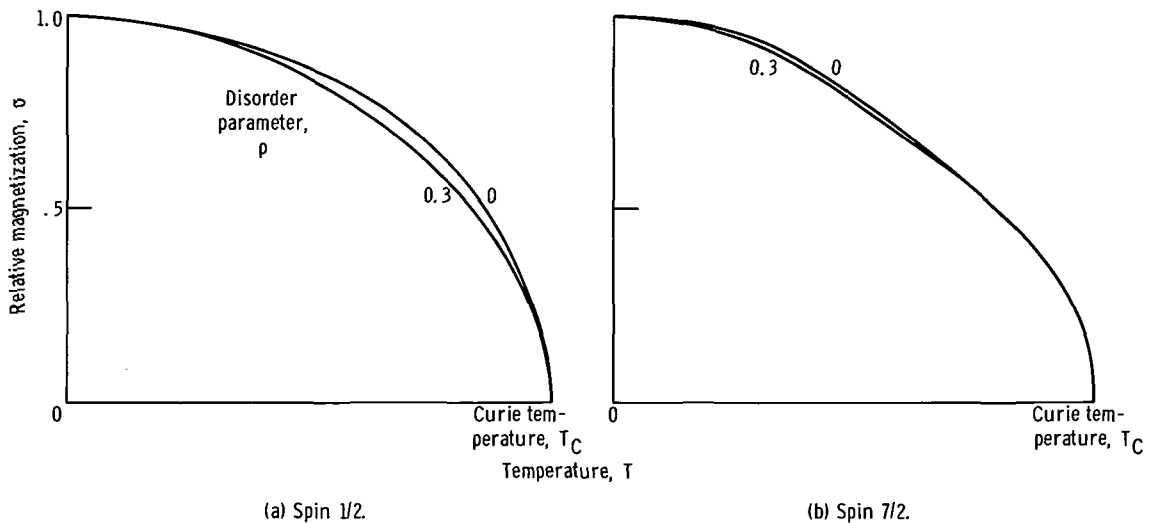

(a) Spin 1/2.   (b) Spin 7/2.

Figure 8. - Relative magnetization as function of rescaled temperature for disordered body-centered cubic systems of spin 1/2 and 7/2.

C_s, one needs an expression for the thermal average of the Hamiltonian. Mills (ref. 40) has derived such an expression exactly from spectral functions of the type discussed in connection with the general theory. Cooke (ref. 41), by an alternate method, has also derived a thermal average $\langle \mathcal{H} \rangle$ which although not exact for $S > 1/2$ is simpler in form than the Mills result. The higher order correlation functions that Cooke ignored, however, do not contribute to the leading terms of $\langle \mathcal{H} \rangle$ at low temperatures. Thus, his result is a good approximation for any spin in this temperature region. Cooke's expression is

$$\frac{\langle \mathcal{H} \rangle}{N} = E_0 + \frac{1}{\hbar} \langle S^z \rangle \sum_{\vec{k}} \frac{E^0(\vec{k}) + \frac{1}{2}\Sigma(\vec{k}, T)}{\exp\left(\dfrac{E_{\vec{k}}}{kT}\right) - 1} \tag{153}$$

where E_0 is some temperature-independent constant that does not affect the specific heat, $E^0(\vec{k})$ is the spin wave energy of a magnon at $T = 0$,

$$E^0(\vec{k}) = \hbar^2 S \left[J(0) - J(\vec{k}) \right] \tag{154}$$

and $\Sigma(\vec{k}, T)$ contains the temperature dependence

$$\Sigma(\vec{k}, T) = \hbar \left[\langle S^z \rangle - \hbar S \right] \left[\mathscr{J}(0) - \mathscr{J}(\vec{k}) \right] \tag{155}$$

Changing the summation in equation (153) to an integration gives

$$\frac{\langle \mathcal{H} \rangle}{N} = E_0 + \frac{1}{2\hbar} \left[\langle S^z \rangle + \hbar S \right] \sum_{\vec{k}} \frac{E_k}{\exp\left(\dfrac{E_{\vec{k}}}{kT}\right) - 1}$$

$$= E_0 + \frac{J'}{2} \left[\langle S^z \rangle^2 + \hbar S \langle S^z \rangle \right] \int \frac{x g_\rho(x)}{\exp\left(\hbar \langle S^z \rangle J' \dfrac{x}{kT}\right) - 1} \, dx \tag{156}$$

At low temperatures, the integral in this expression is easily computed by a change of variable and use of equation (134)

$$\frac{\langle \mathcal{H} \rangle}{N} = E_0 + \frac{J'}{2} \left[\langle S^z \rangle^2 + \hbar S \langle S^z \rangle \right] \frac{C_{1e}}{(1-\rho)^{3/2}} \left(\frac{3\pi^{1/2}}{4} \right) \zeta \left(\frac{S}{2} \right) \left(\frac{kT}{\hbar \langle S^z \rangle J'} \right)^{5/2} \qquad (157)$$

For T near zero, equation (157) indicates that the leading temperature term for $\langle \mathcal{H} \rangle$ is of order $T^{5/2}$ and has dependence on disorder as $(1 - \rho)^{-3/2}$. Since the specific heat is given by

$$C_s = \frac{d \langle \mathcal{H} \rangle}{dT} \qquad (158)$$

it will behave as $(1 - \rho)^{-3/2} T^{3/2}$ at low temperatures. Disorder, then, will have important effects on low-temperature thermodynamic properties of ferromagnets, effects that should be easily detected experimentally. For example, a value of 0.1 for ρ produces a 17 percent increase in the low-temperature specific heat over the value of the corresponding perfect crystal.

Paramagnetic phase. - The cooperative effects of ferromagnetism disappear above the Curie temperature. In this region the system of permanent magnetic moments goes into the paramagnetic phase, where the ordering tendency of the exchange interaction is subdued by the competing influence of thermal fluctuations and the net magnetic moment is zero in the absence of external fields. To investigate how disorder affects the paramagnetic phase of a Heisenberg system, an expression for $1/\sigma$ such as equation (141), is needed. For the sake of computational simplicity, the $S = 1/2$ case is treated and the general results are shown to be valid for any S. For $S = 1/2$, equation (141) becomes

$$\frac{1}{\sigma} = 1 + 2\Phi \qquad (159)$$

The right side of equation (159) can be put in the form of a hyperbolic function:

$$1 + 2 \left[\exp \left(\frac{Hg\mu_B \hbar + \frac{1}{2} \hbar^2 \sigma J'x}{kT} \right) - 1 \right]^{-1} = \coth \left(\frac{Hg\mu_B \hbar + \frac{1}{2} \hbar^2 \sigma J'x}{2kT} \right) \qquad (160)$$

If we write the part of spin wave energy with external field dependence as h and the other energy term as $y(x)$, the right side of this equation can, by the use of a mathematical identity, be rewritten so that

$$\frac{1}{\sigma} = \int \left[\frac{1 + \tanh \frac{h}{2kT} \tanh \frac{y(x)}{2kT}}{\tanh \frac{h}{2kT} \tanh \frac{y(x)}{2kT}} \right] g_\rho(x)\, dx \qquad (161)$$

where $h = Hg\mu_B\hbar$ and $y(x) = (\hbar^2/2)\sigma J'x$. Setting

$$\left. \begin{array}{l} t_0 = \tanh \dfrac{h}{2kT} \\[3mm] t_1 = \tanh \dfrac{y(x)}{2kT} \end{array} \right\} \qquad (162)$$

and bearing in mind that only t_1 has dependence on x,

$$\frac{1}{\sigma} = \frac{1}{t_0} \left[1 + \left(1 - t_0^2\right) \sum_{n=1}^{\infty} \frac{(-1)^n}{t_0^n} \int t_1^n g_\rho(x)\, dx \right] \qquad (163)$$

By use of the series expansion for the hyperbolic tangent, t_1 can be written as a power series in reciprocal temperature:

$$\frac{1}{\sigma} = \frac{1}{t_0} \left[1 - \frac{1 - t_0^2}{t_0} C_1 \left(\frac{\hbar^2 \sigma J'}{4kT} \right) + \frac{1 - t_0^2}{t_0^2} C_2 \left(\frac{\hbar^2 \sigma J'}{4kT} \right)^2 - \cdots \right] \qquad (164)$$

where C_n is the nth moment of the state density

$$C_n = \int x^n g_\rho(x)\, dx \qquad (165)$$

Equation (164) can be used to solve for σ iteratively to any order in T^{-1}. For ex-

ample, where $\sigma = t_0$ is used as a zeroth approximation, the second approximation gives

$$\sigma = t_0 + t_0\left(1 - t_0^2\right)C_1\left(\frac{\hbar^2 J'}{4kT}\right) + t_0\left(1 - t_0^2\right)\left[2\left(1 - t_0^2\right)C_1^2 - C_2\right]\left(\frac{\hbar^2 J'}{4kT}\right)^2 + \ldots \qquad (166)$$

In this equation, it can be seen that the effect of disorder does not appear until the third term, where C_2 appears in the coefficient of T^{-2}. We have shown previously that C_1, the first moment of the density of states, is related to the average energy of spin waves and is unaffected by disorder. The second moment C_2 and higher moments of $g_\rho(x)$ will have ρ dependence, but they do not appear in the leading terms of the series.

The paramagnetic Curie temperature θ is a parameter of experimental interest and it is defined by the Curie-Weiss law,

$$\chi = \frac{C}{T - \theta} \qquad (167)$$

where C is the Curie constant and χ is the magnetic susceptibility. The paramagnetic Curie temperature is found empirically by extrapolating the linear high-temperature region of a $(\chi)^{-1}$-against-T curve to the $(\chi)^{-1} = 0$ axis. Generally, θ is appreciably greater than the actual ferromagnetic transition temperature T_C. Since the susceptibility is proportional to σ/H, we can find θ by multiplying equation (164) by H, keeping only lower order terms in T^{-1}:

$$[\chi]^{-1} \propto \frac{H}{\sigma} \simeq (g\mu_B \hbar)^{-1} \frac{h}{t_0}\left[1 - \frac{C_1}{t_0}\left(\frac{\hbar^2 \sigma J'}{4kT}\right)\right] \qquad (168)$$

For high temperatures, $t_0 \simeq h/2kT$ and so

$$[\chi]^{-1} \propto (g\mu_B \hbar)^{-1}\left(2kT - \frac{C_1 \hbar^2 J'}{2}\right) \qquad (169)$$

when $[\chi]^{-1} = 0$, then

$$T = \frac{C_1 \hbar^2 J'}{4k} = \theta \qquad (170)$$

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The paramagnetic Curie temperature for a system with disorder is identical to the perfect-crystal result.

The conclusion to be drawn from this section is that the paramagnetic phase of a Heisenberg system is less sensitive to disorder than the ferromagnetic phase. We might have intuitively expected this, since in the paramagnetic region the thermal fluctuations have become sufficiently large to destroy the cooperative effects. With this much "thermal disorder" already present in the system, we might have suspected that the additional disorder of randomness in exchange interactions would be less significant than in the lower temperature phase.

Europium Chalcogenide Mixtures

Divalent europium can be made to react with the chalcogenide series, O, S, Se, and Te to form crystals of the cubic NaCl structure. In 1961 (ref. 42), EuO was discovered to be ferromagnetic; and soon afterwards, ferromagnetism was found in EuS and EuSe, while EuTe was found to be antiferromagnetic (refs. 43 and 44). These compounds are insulators and are nearly ideal Heisenberg ferromagnets. Only the Eu^{++} ion is magnetically active in these compounds, and it has a spin of 7/2. As the size of the nonmagnetic ion and the lattice parameter increase, the ferromagnetic coupling decreases and so the Curie temperature decreases from 69 K for EuO to 16.5 K for EuS to 7 K for EuSe. The antiferromagnetic EuTe has a Neel temperature of 7.8 K.

Mixtures of these chalcogenides of europium will be topologically equivalent, but the exchange integral between two Eu^{++} ions will vary according to which chalcogenide ions are nearby. A system disordered in this fashion should be able to be treated by the theory developed in earlier sections. We shall consider the $EuX_\eta Z_{(1-\eta)}$ system where X and Z represent any of the chalcogens - O, S, Se, or Te. Our convention will be to let η be a measure of the concentration of the "weaker" specie, that is, $\left(T_C\right)_{EuZ} > \left(T_C\right)_{EuX}$. The assumption will be made that the X and Z ions are randomly distributed in the lattice. In the NaCl structure the Eu ions form a fcc sublattice and the nonmagnetic ions form another fcc sublattice. Each Eu ion has six nonmagnetic nearest neighbors and, in its own sub lattice, 12 Eu "nearest neighbors." We assume that exchange interactions exist only among nearest-neighbor Eu ions, and we pose the following problem. What is the Curie temperature for such a system, and how does T_C depend on the relative concentration η?

To answer this question we propose a simple model. There are two nonmagnetic ions that lie closest to a line joining two nearest-neighbor Eu ions, and the size of these two ions will be most important in determining the strength of the exchange integral. We assume, then, that the exchange integrals will have one of three values, depending

on which types of ions occupy the two sites closest to the Eu-Eu bond:

$$J = J_1 \quad \text{when two } X \text{ ions occupy these sites}$$

$$J = J_2 \quad \text{when two } Z \text{ ions occupy these sites}$$

$$J = \frac{1}{2}(J_1 + J_2) \quad \text{when an } X \text{ ion and a } Z \text{ ion occupy these sites} \tag{171}$$

In order to calculate the disorder parameter ρ, which will be a function of η, we will need to know J_0, the average exchange interaction or the exchange integral of the corresponding perfect crystal. If N is the number of Eu ions, the total number of exchange interactions is $6N$ and

$$\text{Number of interactions of strength } J_1 = 6N\eta^2$$

$$\text{Number of interactions of strength } J_2 = 6N(1 - \eta)^2$$

$$\text{Number of interactions of strength } \frac{1}{2}(J_1 + J_2) = 6N2\eta(1 - \eta) \tag{172}$$

The average exchange integral is, therefore,

$$J_0 = \eta^2 J_1 + (1 - \eta)^2 J_2 + 2\eta(1 - \eta)\frac{1}{2}(J_1 + J_2) \tag{173}$$

$$= \eta J_1 + (1 - \eta)J_2$$

The disorder parameter for this system is

$$\rho(\eta) = \frac{1}{6}\frac{\langle (J - J_0)^2 \rangle}{J_0^2}$$

$$= \frac{1}{12}d^2\frac{\eta(1 - \eta)}{(d\eta - 1)^2} \tag{174}$$

where d is the ratio

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$$d \equiv \frac{J_2 - J_1}{J_2} \tag{175}$$

From equation (150) the Curie temperature is proportional to the product of $1 - \rho$ and J_0. Therefore, the Curie temperature for $EuX_\eta Z_{(1-\eta)}$, that is $T_C(\eta)$, in units of the T_C for pure EuZ, is

$$\frac{T_C(\eta)}{T_C(0)} = [1 - \rho(\eta)] \frac{J_0}{J_2}$$

$$= \left[1 - \frac{d^2 \eta (1 - \eta)}{12(d\eta - 1)^2}\right](1 - d\eta) \tag{176}$$

This relation has been used to calculate the Curie temperatures of the three systems - $EuS_\eta O_{(1-\eta)}$, $EuSe_\eta S_{(1-\eta)}$ and $EuSe_\eta O_{(1-\eta)}$. The results are shown in figure 9.
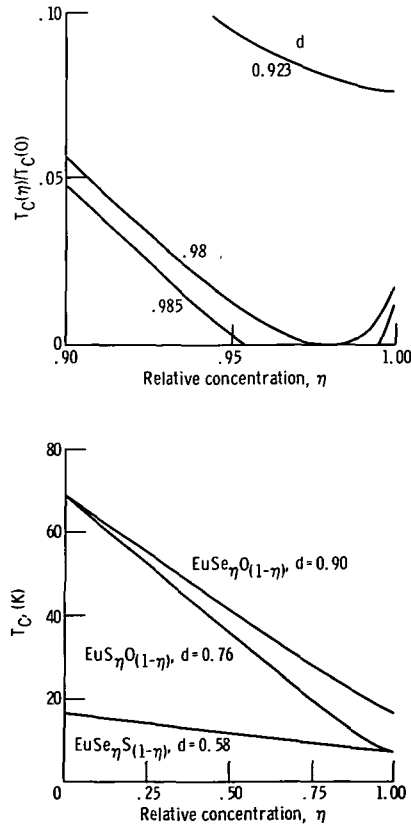

Figure 9. - Curie point as function of relative concentration for europium chalcogenide mixtures.

When the value of d is relatively small, that is, when the Curie temperatures for pure EuX and pure EuZ are relatively close, the change in $T_C(\eta)$ with η is almost linear. This is so because $\rho(\eta)$ never gets very large when d is small, in which case the change in $T_C(\eta)$ is caused almost entirely by the change in $J_0(\eta)$, which is linear. If d becomes sufficiently large, the $[1 - \rho(\eta)]$ term assumes more importance, producing a departure from linearity. In fact, for $d > 0.923$ a minimum occurs in the $T_C(\eta)$-against-η curve. In this case, replacing the X ions of EuX with Z ions would initially cause a reduction in T_C even though EuZ is a stronger ferromagnet with a higher T_C than EuX. For $d > 0.98$ the ferromagnetic phase is destroyed completely by small concentrations of Z (i.e., $\eta \lesssim 1$). For the $EuSe_\eta O_{(1-\eta)}$ system, d = 0.90, a value sufficiently large to warrant an experimental investigation of the region of low oxygen concentration to see if any of these surprising predictions associated with high d-values are observable in this system.

DISCUSSION AND CONCLUSIONS

Several general statements can be made now, in answer to the central question posed in the INTRODUCTION. There it was asked whether disorder would have a significant effect on a cooperative phenomenon like ferromagnetism and if so, how? The Green's function theory developed herein to study distributed disorder in a Heisenberg ferromagnet gives us an affirmative answer, and the disorder has been shown to manifest itself in several ways.

Disorder, introduced by allowing a randomness in the strength of the exchange couplings, has a marked effect on the density of spin wave states. Modest values of the disorder parameter ρ can produce relatively large changes in the state density $g_\rho(x)$ in the form of enhancement of the low-energy densities and extension of the energy band to higher values. Physically, this can be explained as an effect of the addition of greater and lesser exchange interactions than are found in the corresponding ordered system. Some spins in the disordered system will find themselves more strongly coupled to their neighbors, while other spins will be less strongly coupled than in the perfect crystal. Since the energy of a spin wave is proportional to this exchange coupling, there will be more very low-energy magnons and also higher energy magnons than the case where all spins are equally coupled. This is analogous to the situation in lattice dynamics (ref. 45) where the substitution of light impurities into a crystal produces localized modes in the phonon density of states outside the perfect-lattice continuum, while heavy defects produce an enhancement in the lower part of the spectrum. Both of these effects are present in the spin wave state density, where allowing a randomness in the values of exchange interactions corresponds to adding heavy and light impurities to a phonon system.

The increase in the density of low-energy states is of the order $(1 - \rho)^{-3/2}$, and this manifests itself in the low-temperature properties such as the specific heat. When T is near zero, all but the lowest spin wave states are unoccupied. As the disordered ferromagnet is heated slightly, the spin waves are allowed to occupy the states to a higher energy according to the Bose distribution, but there are more of these states than in the ordered system. Thus, more energy must be put into the system to produce the equilibrium distribution associated with a given temperature, and the specific heat is correspondingly larger.

The spontaneous magnetization of a disordered ferromagnet decreases with temperature more quickly than for a crystal and the Curie point was shown to diminish linearly with disorder as $1 - \rho$. The transition from the ferromagnetic to the paramagnetic phase is no less abrupt for a disordered system, however, in that the spontaneous magnetization exhibits the same negatively infinite slope at T_C.

In the preceding main section, the effects of disorder on systems of various structure and spin were presented. Without repeating specific results, there are general conclusions to be drawn from this work. Probably, the most important statement to be made is that order and disorder are of most significance in cooperative phenomena and of less consequence where cooperative effects are absent. A corollary to this statement might be that the effects of disorder are more noticeable at low temperatures than at high. The disorder parameter is involved in the first term of the low-temperature specific-heat expansion, but it is not present in the high-temperature susceptibility until the third term of the series.

At the conclusion of what has been an initial attempt to include the effects of distributed disorder in the theory of magnetism, there are suggestions that can be made for future work in this area. Certainly, the decoupling approximation used in the section Equations of Motion limits the accuracy of our calculations. Improvement in the low-temperature expansions (ref. 19), for example, are sure to come about by the use of more sophisticated decoupling schemes that might include correlation effects. Another improvement to the present work is likely to be produced by a more complete description of the disorder. The one parameter description used herein, treats all distributions of exchange interactions alike, provided the relative mean-square deviations are equal. However, the shape of such distributions are undoubtedly important for some properties. And, of course, to produce results that agree more closely with experimental measurements, inclusion of more than just nearest-neighbor interactions might well be required.

In conclusion, we have generalized the theory of ferromagnetism to include the effects associated with the disappearance of translational symmetry. It is our hope that, in rectifying some of the incompleteness in our knowledge of magnetic systems, we have provided new insights in our understanding of all cooperative phenomena.

Lewis Research Center,
    National Aeronautics and Space Administration,
        Cleveland, Ohio, November 28, 1972,
            502-01.

APPENDIX A

DENSITY OF SPIN WAVE STATES OF PERFECTLY
ORDERED CUBIC FERROMAGNETS

In calculating the density of spin wave states of a disordered ferromagnet, use is made of the density of states of the corresponding perfectly ordered ferromagnet. The details of the derivation of the density of states for crystals of simple cubic (sc), body-centered cubic (bcc), and face centered cubic (fcc) symmetry are presented here. Similar calculations have been carried out by Jelitto (ref. 46) and others (ref. 47) whose results are in agreement with those presented here.

Simple Cubic

According to Green function calculations, a spin wave of wave vector $\vec{k}$ has an energy

$$E(\vec{k}) = Hg\mu_\beta + \hbar\langle S^Z\rangle\left[\mathscr{J}(0) - \mathscr{J}(\vec{k})\right] \tag{A1}$$

When there is no external field, this dispersion relation becomes

$$E(\vec{k}) = \hbar\langle S^Z\rangle \cdot \sum_{f-g} J_{fg}\left[1 - e^{-i(\vec{f}-\vec{g})\cdot\vec{k}}\right] \tag{A2}$$

where J_{fg} is the exchange interaction between the spins at sites f and g. The vectors $\vec{f}$ and $\vec{g}$ are the position vectors for these sites. Exchange interactions are relatively short ranged and so we make the assumption

$$J_{fg} = \begin{cases} J & \text{if } \vec{f} - \vec{g} = \text{Nearest-neighbor vector} \\ 0 & \text{otherwise} \end{cases} \tag{A3}$$

For a simple cubic lattice, then

$$E(\vec{k}) = \hbar \langle S^z \rangle J \sum_{\vec{\Delta}} \left[1 - e^{-i\vec{\Delta} \cdot \vec{k}} \right] \tag{A4}$$

where $\vec{\Delta}$ represents the six possible nearest-neighbor position vectors $\pm a\hat{x}$, $\pm a\hat{y}$, $\pm a\hat{z}$, where a is the length of the cube edge and $\hat{x}$, $\hat{y}$, and $\hat{z}$ are unit vectors in the x, y, and z directions. Carrying out the summation in equation (A4) gives

$$E(\vec{k}) = 2\hbar \langle S^z \rangle J(3 - \cos ak_x - \cos ak_y - \cos ak_z) \tag{A5}$$

It is convenient to deal with a "reduced energy," which is the energy normalized to range between zero and unity. We define, then,

$$X_{\vec{k}} = \frac{E_{\vec{k}}}{\left(E_{\vec{k}} \right)_{max}} = \frac{1}{2} - \frac{1}{6} (\cos ak_x + \cos ak_y + \cos ak_z) \tag{A6}$$

We wish to derive a spin wave density-of-states function $g_{sc}(x)$ which represents the number of spin wave states of energy x per unit energy, divided by the total number of states. The fraction of states with energy between x_1 and x_2, then, would be

$$\int_{x_1}^{x_2} g_{sc}(x) \, dx \tag{A7}$$

From the way we described $g_{sc}(x)$, it is obvious that $g_{sc}(x)$ is a normalized function

$$\int_0^1 g_{sc}(x) \, dx = 1 \tag{A8}$$

The reciprocal lattice vectors $\vec{k}$ which are associated with the spin waves of energy x_k are uniformly distributed throughout k-space. So the derivation of $g_{sc}(x)$ reduces itself to the calculation of the volume bounded by the two surfaces of constant energy: $x(k_x, k_y, k_z) = x$ and $x(k_x, k_y, k_z) = x + dx$. The fraction of the total volume bounded by these two surfaces is $g_{sc}(x) \, dx$.

Since any spin wave can be represented by one wave vector in the first Brillouin zone (i.e., any spin wave with a wave vector outside the first Brillouin zone is indistinguishable from the spin wave with the corresponding wave vector in the first zone),

we need only concern ourselves with the first Brillouin zone when wishing to sample all $\vec{k}$-space. The volume of the first Brillouin zone is $(\pi/a)^3$. We can write $g_{sc}(x)$, then as

$$g_{sc}(x) = \frac{1}{\left(\frac{\pi}{a}\right)^3} \int_0^{\pi/a} dk_x \int_0^{\pi/a} dk_y \int_0^{\pi/a} dk_z \delta(x - x_{\vec{k}}) \qquad (A9)$$

where δ is the Dirac delta function.

Before calculating $g_{sc}(x)$ from equation (A9), it might be beneficial to say a few words concerning the appearance of the delta function. If one were interested in counting the number of spin wave states in the energy range between two energies x_1 and x_2, one way would be to use the delta function in the following manner:

$$\int_{x_1}^{x_2} \sum_{\vec{k}} \delta(x - x_k)\, dx = \text{Number of states in energy range } x_1 < x < x_2 \qquad (A10)$$

As the sum is made over all possible k-vectors, the delta function acts as a counter, registering unity every time $\vec{k}$ is such that $x_1 < x_{\vec{k}} < x_2$ and registering zero when the wave vector is such that its spin wave energy falls outside this range. Comparing equation (A10) with expression (A7) suggests that

$$g_{sc}(x) \alpha \sum_{\vec{k}} \delta(x - x_k) \qquad (A11)$$

The number of spin wave states is of the order of magnitude of the number of atoms in the crystal. In macroscopic samples, this number is sufficiently large to warrant replacement of the sum over k-states with an integration over the Brillouin zone. Thus,

$$\frac{1}{N} \sum_{\vec{k}} \rightarrow \frac{v}{(2\pi)^3} \int_{B.Z.} d\vec{k} \qquad (A12)$$

With this as the rationale for equation (A9), it remains only to carry out the three

integrations. After substituting equation (A6) into equation (A9) and making use of the identity

$$\delta[f(x)] = \frac{1}{\left|\dfrac{df(x)}{dx}\right|} \delta(x - x_0) \tag{A13}$$

where $f(x_0) = 0$, and making the change of variables

$$\cos(ak_x) = u \qquad \cos(ak_y) = v \qquad \cos(ak_z) = w \tag{A14}$$

we have

$$g_{sc}(x) = \frac{6}{\pi^3} \int_{-1}^{1} du \int_{-1}^{1} dv \int_{-1}^{1} dw \frac{\delta[w - (3 - 6x - u - v)]}{\sqrt{(1 - u^2)(1 - v^2)(1 - w^2)}} \tag{A15}$$

We shall see that after performing the w-integration, the limits of integration in the u-v plane must be redefined. That is, since

$$\int_{-1}^{1} \frac{\delta(w - 3 + 6x + u + v)}{\sqrt{(1 - u^2)(1 - v^2)(1 - w^2)}} dw = \begin{cases} \dfrac{1}{\sqrt{(1 - u^2)(1 - v^2)\,1 - (3 - 6x - u - v)^2}} & \text{for } -1 < (3 - 6x - u - v) < 1 \\ 0 \quad \text{otherwise} \end{cases} \tag{A16}$$

the original limits of the u and v integrations must now be more strictly defined so that not only $-1 < u < 1$ and $-1 < v < 1$ but also

$$u + v > 2 - 6x \qquad \text{and} \qquad u + v < 4 - 6x \tag{A17}$$

After the first integration, then

$$g_{sc}(x) = \frac{6}{\pi^3} \int_{u_L}^{u_U} du \int_{v_L}^{v_U} dv \frac{1}{\sqrt{(1 - u^2)(1 - v^2)\left[1 - (3 - 6x - u - v)^2\right]}} \tag{A18}$$

where for $0 \le x \le 1/3$

$$u_U = 1 \qquad v_U = 1$$

$$u_L = 1 - 6x \qquad v_L = 2 - 6x - u$$

for $1/3 \le x \le 2/3$

$$u_U = 3 - 6x \qquad v_U = 1$$

$$u_L = -1 \qquad v_L = 2 - 6x - u$$

and

$$u_U = 1 \qquad v_U = 4 - 6x - u$$

$$u_L = 3 - 6x \qquad v_L = -1$$

and for $2/3 < x < 1$

$$u_U = 5 - 6x \qquad v_U = 4 - 6x - u$$

$$u_L = -1 \qquad v_L = -1$$

The second integration, involving the inverse square root of a fourth-degree polynomial in v, can be reduced to an elliptic integral (ref. 36) of the first kind. After factoring the integrand of equation (A18), we have

$$\int_{v_L}^{v_U} \frac{dv}{\sqrt{(v - 1)(v + 1)[v - (4 - 6x - u)][v - (2 - 6x - u)]}} \qquad (A19)$$

The factors in the integrand of expression (A19) can be arranged in such a way to look like

$$\left[(v - a)(v - b)(v - c)(v - d)\right]^{-1/2} \qquad (A20)$$

where

$$a > b > c > d$$

The roots a, b, c, and d are, of course, 1, -1, 4 - 6x - u, and 2 - 6x - u arranged to satisfy the inequality (A20). When so arranged expression (A19) takes the form

$$\int_c^b \frac{dv}{\sqrt{(v - a)(v - b)(v - c)(v - d)}} \qquad (A21)$$

Such an integration, between the third and second largest roots of a fourth-degree polynomial, can be reduced to a complete elliptic integral of the first kind

$$K(m) = \int_0^1 \frac{dt}{\sqrt{(1 - t^2)(1 - mt^2)}} \qquad (A22)$$

where the argument m is

$$m = \frac{(a - d)(b - c)}{(a - c)(b - d)} \qquad (A23)$$

and the coefficient of K(m) is

$$\frac{2}{\sqrt{(a - c)(b - d)}} \qquad (A24)$$

Carrying out the second integration in this manner leaves

$$g_{sc}(x) = \frac{6}{\pi^3} \int_{u_L}^{u_U} \frac{K(m)}{\sqrt{(1 - u^2)}} \, du \qquad (A25)$$

where

$$u_U = 1$$
$$u_L = 1 - 6x$$
$$\text{for } 0 \le x \le \frac{1}{3}$$

$$u_U = 1$$
$$u_L = -1$$
$$\text{for } \frac{1}{3} \le x \le \frac{2}{3}$$

$$u_U = 5 - 6x$$
$$u_L = -1$$
$$\text{for } \frac{2}{3} \le x \le 1$$

where

$$m = \frac{1}{4}(5 - 6x - u)(-1 + 6x + u)$$

The integrations involved in equation (A25) were carried out on an IBM 7094-7044 and the results are shown in figure 2(a). As could have been predicted by the symmetry of earlier equations, the density of states is symmetric about $x = 1/2$. There are two discontinuities in the slope of the curve, occurring at $x = 1/3$ and $x = 2/3$. The behavior immediately below $x = 1/3$ and above $x = 2/3$ is of square-root nature, which means that the slopes have infinities of inverse square-root nature. By well-known arguments the low-energy density of states must be proportional to the square root of the energy and the proportionality constant can be calculated by evaluating equation (A25) in the limit of x approaching zero:

$$g_{sc}(x) \underset{x \to 0}{\simeq} \frac{6}{\pi^3} \int_{1-6x}^{1} \frac{\frac{\pi}{2}}{\sqrt{1 - u^2}} \, du = \frac{6\sqrt{3}}{\pi^2} \sqrt{x} \tag{A26}$$

It is convenient to have an analytic expression of the density of states in the low-energy limit because, in the calculation of low-temperature thermodynamic quantities, the low-energy portion of the density-of-states curve is the only part that has importance.

Body-Centered Cubic

The density of spin wave states for a body-centered cubic ferromagnet $g_{bcc}(x)$ is calculated in this section in a manner analogous to the procedure for the simple cubic case. The zero-field dispersion relation is written by carrying out the sum indicated in equation (A2), again considering only nearest-neighbor interactions. After defining a reduced energy, the delta-function formalism introduced for the simple cubic case is used to find the volume in k-space between surfaces of constant energy, a process involving a threefold integration over k-space. A plot of the results of these integrations is presented in the form of $g_{bcc}(x)$ against x, and some comments are made on the shape of this curve.

In the body-centered cubic system, each spin has eight nearest neighbors. The dispersion relation resulting from summing terms like $J_{fg}e^{i(\vec{f}-\vec{g})\cdot\vec{k}}$ over nearest neighbors is

$$E(\vec{k}) = 8\hbar\langle S^z\rangle J\left(1 - \cos\frac{a}{2}k_x \cos\frac{a}{2}k_y \cos\frac{a}{2}k_z\right) \tag{A27}$$

where, as before, J is the exchange integral between nearest neighbors and a is the length of the edge of a body-centered cube. The maximum energy that a spin wave in a bcc system can possess is, then, $16\hbar\langle S^z\rangle J$. Dividing equation (A27) by $[E(\vec{k})]_{max}$ yields a reduced energy that is convenient in that it is a dimensionless quantity which varies between zero and unity and has the added advantage of removing the temperature dependence ($\langle S^z\rangle$ is temperature dependent) from the spin wave energy. For the bcc system, then, the reduced energy is

$$x_{\vec{k}} = \frac{1}{2} - \frac{1}{2}\cos\frac{a}{2}k_x \cos\frac{a}{2}k_y \cos\frac{a}{2}k_z \tag{A28}$$

Setting

$$\cos\frac{a}{2}k_x = u \qquad \cos\frac{a}{2}k_y = v \qquad \cos\frac{a}{2}k_z = w \tag{A29}$$

we have

$$g_{bcc}(x) = \frac{2}{\pi^3} \int_{-1}^{1} du \int_{-1}^{1} dv \int_{-1}^{1} dw \frac{\delta\left(w - \frac{1 - 2x}{uv}\right)}{|uv| \sqrt{(1 - u^2)(1 - v^2)(1 - w^2)}}$$

(A30)

The w-integration will produce nonzero results only when u, v, and x are such that

$$-1 < \frac{1 - 2x}{uv} < 1$$

(A31)

Inequality (A31) together with our knowledge that u and v can only have values between +1 and -1 fixes the limits of the u and v integrations after the w-integration is completed. The first integration leaves

$$g_{bcc}(x) = \frac{8}{\pi^3} \int_{|1-2x|}^{1} du \int_{|1-2x/u|}^{1} dv \frac{1}{\sqrt{(1 - u^2)(1 - v^2)\left[u^2 v^2 - (1 - 2x)^2\right]}}$$

(A32)

Since the integrand is an even function of u and of v, it is sufficient to integrate in one quadrant of the u-v plane only because of the symmetry of the integration limits.

Following the procedure of the simple cubic case, the v-integration can be put into the form

$$g_{bcc}(x) = \frac{8}{\pi^3} \int_{|1-2x|}^{1} \frac{du}{u\sqrt{1 - u^2}} \int_{b}^{a} \frac{dv}{\sqrt{(v - a)(v - b)(v - c)(v - d)}}$$

(A33)

where for $0 \le x \le \frac{1}{2}$

$$a = 1 \quad b = \frac{1 - 2x}{u} \quad c = -\frac{1 - 2x}{u} \quad d = -1$$

and for $\frac{1}{2} \le x \le 1$

$$a = 1 \qquad b = -\frac{1 - 2x}{u} \qquad c = \frac{1 - 2x}{u} \qquad d = -1$$

which gives

$$g_{bcc}(x) = \frac{16}{\pi^3} \int_{|1-2x|}^{1} du \; \frac{K\left[\left(\frac{u - |1 - 2x|}{u + |1 - 2x|}\right)^2\right]}{(u + |1 - 2x|)\sqrt{1 - u^2}} \tag{A34}$$

The density of states generated by equation (A34) is shown in figure 2(b). The interesting portions of the curve, near $x = 0$ and $x = 1/2$, have been calculated analytically; and the other points have been evaluated by numerical machine integration. As in the simple cubic case, the curve is symmetric about $x = 1/2$. However, whereas, $g_{sc}(x)$ was everywhere finite; $g_{bcc}(x)$ has an infinity at $x = 1/2$ of squared logarithmic nature. We can show this by evaluating expression (A34) in the limit of $x \rightarrow 1/2$. We define a parameter f which is two times the displacement of x from the singularity:

$$f = 1 - 2x \tag{A35}$$

As x approaches $1/2$ from below, f approaches zero from above. For $x < 1/2$, then

$$g_{bcc}(x) = \frac{16}{\pi^3} \int_{f}^{1} du \; \frac{K\left[\left(\frac{u - f}{u + f}\right)^2\right]}{(u + f)\sqrt{1 - u^2}} \tag{A36}$$

When f is small, the argument of the complete elliptic integral K is very close to 1 in the range of integration, except at the very low end where u is near f. Since $K(m)$ can be expanded in a power series in $m_1 = 1 - m$,

$$K(m) = a_0 + a_1 m_1 + a_2 m_1^2 + \ldots + \left(b_0 + b_1 m_1 + b_2 m_1^2 + \ldots\right) \ln \frac{1}{m_1}$$

when m is near unity, the approximation

$$K(m) \simeq a_0 + b_0 \ln \frac{1}{m_1} \qquad (A38)$$

becomes a good one. The approximation used in calculating the limiting form of $g_{bcc}(x)$ near the singularity is

$$g_{bcc}(x) \underset{f \to 0}{\simeq} \frac{16}{\pi^3} \int_f^1 \frac{K\left(1 - 4\frac{f}{u}\right)}{u\sqrt{1 - u^2}} du \simeq \frac{16}{\pi^3} \int_f^1 \frac{a_0 - b_0 \ln 4f}{u\sqrt{1 - u^2}} du$$

$$+ \frac{16}{\pi^3} \int_f^1 \frac{b_0 \ln u}{u} du \qquad (A39)$$

where

$$a_0 = 1.38629 \qquad b_0 = \frac{1}{2}$$

Carrying out these integrations leaves

$$g_{bcc}(x) \underset{f \to 0}{\simeq} c_0 + c_1 \ln \cdot f + c_2 (\ln f)^2 \qquad (A40)$$

where

$$c_0 = \frac{16}{\pi^3} (a_0 - b_0 \ln 4) \ln 2 = 0.247926$$

$$c_1 = \frac{-16}{\pi^3} (a_0 - b_0 \ln 2) = 0.536521$$

$$c_2 = \frac{8}{\pi^3} b_0 = 0.129006$$

The relative error introduced by the approximations of equation (A39) can be shown to decrease as f approaches zero because the absolute error remains bounded as x approaches the singularity although $g_{bcc}(x)$ does increase.

Finally, the low-energy form of $g_{bcc}(x)$ is evaluated by noticing that the argument of the elliptic integral in equation (A34) is close to zero in the range of integration when x is small. We write, then,

$$g_{bcc}(x) \underset{x \to 0}{\simeq} \frac{16}{\pi^3} \int_{1-2x}^{1} \frac{\frac{\pi}{2}}{2\sqrt{1-u^2}} \underset{x \to 0}{\simeq} \frac{8}{\pi^2} \sqrt{x} \qquad (A41)$$

Face-Centered Cubic

The dispersion relation for spin waves in a fcc ferromagnet with nearest-neighbor interactions is

$$E(\vec{k}) = 4\hbar \langle S^z \rangle J \left(3 - \cos \frac{a}{2} k_x \cos \frac{a}{2} k_y - \cos \frac{a}{2} k_y \cos \frac{a}{2} k_z - \cos \frac{a}{2} k_z \cos \frac{a}{2} k_x \right)$$
$$(A42)$$

where the symbols are defined earlier in this appendix. Dividing equation (A42) by the maximum spin wave energy gives for the reduced energy

$$x_{\vec{k}} = \frac{3}{4} - \frac{1}{4} \left(\cos \frac{a}{2} k_x \cos \frac{a}{2} k_y + \cos \frac{a}{2} k_y \cos \frac{a}{2} k_z + \cos \frac{a}{2} k_z \cos \frac{a}{2} k_x \right) \qquad (A43)$$

Following the procedures outlined earlier,

$$g_{fcc}(x) = \frac{a^3}{(2\pi)^3} \int_0^{2\pi/a} dk_x \int_0^{2\pi/a} dk_y \int_0^{2\pi/a} dk_z \delta\left(x - x_{\vec{k}} \right)$$

$$= \frac{4}{\pi^3} \int_{-1}^{1} du \int_{-1}^{1} dv \int_{-1}^{1} dw \frac{\delta\left(w - \frac{3 - 4x - uv}{u + v} \right)}{|uv| \sqrt{(1 - u^2)(1 - v^2)(1 - w^2)}} \qquad (A44)$$

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After carrying out the integration over w, the v-integration is accomplished after putting the resulting integrand into a standard form for the complete elliptic integral of the first kind, leaving

$$g_{fcc}(x) = \frac{8}{\pi^3} \int_{1-2x}^{1} du \, \frac{K(m_1)}{\sqrt{(1-u^2)(3-4x+u^2)}} \quad \text{for} \quad 0 < x < 1/2$$

$$g_{fcc}(x) = \frac{4}{\pi^3(1-x)} \int_{1-2x}^{2x-1} du \, \frac{K(m_2)}{\sqrt{1-u^2}}$$

$$+ \frac{8}{\pi^3} \int_{2x-1}^{1} du \, \frac{K(m_1)}{\sqrt{(1-u^2)(3-4x+u^2)}} \quad \text{for} \quad 1/2 < x < 3/4 \qquad \text{(A45)}$$

$$g_{fcc}(x) = \frac{16}{\pi^3} \int_{-\sqrt{4x-3}}^{\sqrt{4x-3}} \frac{K(m_3)}{(1-u^2)\left[(4x-2)^2 - 4u^2\right]} + \frac{8}{\pi^3(1-x)} \int_{\sqrt{4x-3}}^{2x-1} du \, \frac{K(m_2)}{\sqrt{1-u^2}}$$

$$+ \frac{8}{\pi^3} \int_{2x-1}^{1} du \, \frac{K(m_1)}{\sqrt{(1-u^2)(3-4x+u^2)}} \quad \text{for} \quad 3/4 \le x \le 1$$

where

$$m_1 = \frac{u^2 - (2x-1)^2}{u^2 + 3 - 4x}$$

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$$m_2 = \frac{-u^2 + (2x - 1)^2}{4(1 - x)^2}$$

$$m_3 = \frac{4(1 - x)^2}{-u^2 + (2x - 1)^2}$$

Figure 2(c) shows the results of machine calculations of equation (A45). Unlike the sc or bcc cases, the density of spin wave states for the fcc system shows no symmetry about $x = 1/2$. The most striking difference in the fcc curve is the logrithmic singularity in $g(x)$ at the energy maximum. At $x = 3/4$ there is a discontinuity in the slope reminiscent of the behavior of $g_{sc}(x)$ at $x = 1/3$ and $x = 2/3$. That is, the slope of $g_{fcc}(x)$ becomes infinite as x approaches 3/4 from below, and the behavior of $g_{fcc}(x)$ is of square-root nature in this region. The limiting form of $g_{fcc}(x)$ as x approaches zero is easy to calculate after noticing that m_1 is small throughout the range of integration when x is small. Thus,

$$g_{fcc}(x) \underset{x \to 0}{\simeq} \frac{4}{\pi^2} \sqrt{x} \qquad (A46)$$

APPENDIX B

DERIVATION AND APPLICATION OF SELF-ENERGY TERM

Derivation and Diagonalization of the Self-Energy

The self-energy matrix is defined by equation (73),

$$\hat{\Sigma}^a \equiv \langle \hat{\Delta} \hat{\Gamma} \hat{\Delta} \rangle \tag{B1}$$

where $\hat{\Delta}$ is the difference between the A matrices for the disordered ferromagnet and the corresponding perfect crystal,

$$\hat{\Delta} \equiv \hat{A}^0 - \hat{A} \tag{B2}$$

and the angular brackets indicate an ensemble average over systems with similar disorder.

The components of $\hat{\Sigma}^a$ in lattice space can be written

$$\Sigma_{gj}^a = \left\langle \sum_h \sum_i \Delta_{gh} \Gamma_{hi} \Delta_{ij} \right\rangle \tag{B3}$$

where, from equation (55),

$$\Delta_{gh} = \langle S^z \rangle \frac{2\pi}{\Theta(a)} \left[\delta_{gh} \sum_f j(g,f) - j(g,h) \right] \tag{B4}$$

Here $j(g,f)$ is the deviation of the exchange coupling between sites g and f from the mean value:

$$j(g,f) = J(g,f) - J^0(g,f) \tag{B5}$$

The superscript zero indicates that the term is associated with the corresponding perfect crystal. Substitution of equation (B4) into equation (B3) gives

$$\Sigma_{gj}^a = \left(\frac{2\pi \langle S^z \rangle}{\Theta(a)} \right)^2 \left\langle \sum_h \sum_i j(g,h) j(j,i) (\Gamma_{gj} + \Gamma_{hi} - \Gamma_{gi} - \Gamma_{jh}) \right\rangle \tag{B6}$$

We assume deviations of the exchange interactions to be symmetric about the mean value. The ensemble averaging, then, is carried out by use of the relation

$$\langle j(g,h)j(i,j)\rangle = j^2(g,h)(\delta_{gi}\delta_{hj} + \delta_{gj}\delta_{hi}) \tag{B7}$$

where $j^2(g,h)$ is the mean-square deviation of the exchange integral between spins at sites g and h. Since $j^2(g,h)$ is an ensemble-averaged quantity, it is translationally invariant and depends only on the ensemble-averaged $|\vec{r}_g - \vec{r}_j|$. Equation (B7) assumes that there are no three-or-more-site correlations in the deviations. For example, the ensemble average of the product $j(i,j)j(i,h)$ vanishes. Thus, equation (B6) becomes

$$\Sigma_{gj}^a = \left(\frac{2\pi\langle S^z\rangle}{\Theta(a)}\right)^2 \left\{\delta_{gj}\left[\Gamma_{gj}\sum_h j^2(g,h) + \sum_h j^2(g,h)(\Gamma_{hh} - 2\Gamma_{gh})\right] + j^2(g,j)(\Gamma_{gj} - \Gamma_{gg})\right\} \tag{B8}$$

Next, we write equation (B8) in a more symmetric form and employ the Fourier transform of Γ_{gh},

$$\Gamma_{gh} = \frac{1}{N}\sum_{\vec{k}} \Gamma_{\vec{k}} e^{i(\vec{g}-\vec{h})\cdot\vec{k}} \tag{B9}$$

where $\vec{k}$ is a reciprocal lattice vector.

$$\Sigma_{gj}^a = \left(\frac{2\pi\langle S^z\rangle}{\Theta(a)}\right)^2 \frac{1}{N}\sum_{\vec{k}} \Gamma_{\vec{k}} \left[2\delta_{gj}\sum_h j^2(g,h) - 2\delta_{gj}\sum_h j^2(g,h)e^{i(g-h)\cdot\vec{k}}\right.$$
$$\left. + j^2(g,j)\left(e^{i(g-j)\cdot\vec{k}} + e^{i(j-g)\cdot\vec{k}}\right) - 2j^2(g,j)\right] \tag{B10}$$

It will be helpful in deriving the diagonalized form of Σ to study the product of Σ and Γ.

$$(\hat{\Gamma}\hat{\Sigma})_{fg} = \sum_g \Gamma_{fg}\Sigma_{gj} \tag{B11}$$

Since

$$\Gamma_{fg} = \frac{1}{N} \sum_{\vec{k}'} \Gamma_{\vec{k}'} e^{i(f-g)\cdot \vec{k}'} \tag{B12}$$

Equation (B11) becomes

$$(\hat{\Gamma}\hat{\Sigma})_{fg} = \left(\frac{2\pi\langle S^z \rangle}{N\Theta(a)}\right)^2 \sum_g \sum_{\vec{k}'} \sum_{\vec{k}} \Gamma_{\vec{k}'}\Gamma_{\vec{k}} e^{i(f-g)\cdot \vec{k}}$$

$$\times \left\{ 2\delta_{gj} \sum_h j^2(g,h) - 2\delta_{gj} \sum_h j^2(g,h)e^{i(g-h)\cdot \vec{k}} + j^2(g,j)\left[e^{i(g-j)\cdot \vec{k}} \right. \right.$$

$$\left. \left. + e^{i(j-g)\cdot \vec{k}} \right] - 2j^2(g,h) \right\} \tag{B13}$$

An example of how this expression will be treated can be given by looking, say, at the second last term. Forgetting the coefficient for the moment, we have

$$\sum_g \sum_{k'} \sum_k \Gamma_{\vec{k}'}\Gamma_{\vec{k}} j^2(g,j) e^{i(j-g)\cdot \vec{k}} e^{i(f-g)\cdot \vec{k}'} = \sum_{\vec{k}'} \sum_{\vec{k}} \Gamma_{\vec{k}'}\Gamma_{\vec{k}} e^{i(f-j)\vec{k}'} \sum_g j^2(g,j) e^{i(j-g)\cdot(\vec{k}+\vec{k}')} \tag{B14}$$

Since $j^2(g,j)$ is translationally invariant and depends only on $(g - j)$, its Fourier transform is

$$\mathscr{J}_2(\vec{k}) \equiv \sum_g j^2(g,j) e^{-i(g-j)\cdot \vec{k}} \tag{B15}$$

which allows equation (B14) to be written

$$\sum_{\vec{k}'} \sum_{\vec{k}} \Gamma_{\vec{k}'}\Gamma_{\vec{k}} \mathscr{J}_2(\vec{k} + \vec{k}') e^{i(f-j)\cdot \vec{k}'} \tag{B16}$$

Similar manipulation of the remaining terms simplifies equation (B13) to

$$(\hat{\Gamma}\hat{\Sigma})_{fg} = \left(\frac{2\pi\langle S^Z\rangle}{N\Theta(a)}\right)^2 \sum_{\vec{k}'}\sum_{\vec{k}} \Gamma_{\vec{k}'}\Gamma_{\vec{k}} e^{i(f-j)\cdot\vec{k}'}\left[2\mathscr{J}_2(0) - 2\mathscr{J}_2(\vec{k})\right.$$

$$\left. - 2\mathscr{J}_2(\vec{k}') + \mathscr{J}_2(\vec{k}+\vec{k}') + \mathscr{J}_2(\vec{k}-\vec{k}')\right] \quad \text{(B17)}$$

Since the matrix $(\hat{\Gamma}\hat{\Sigma})$ is diagonalized by the Fourier transform

$$(\hat{\Gamma}\hat{\Sigma})_{fj} = \frac{1}{N}\sum_{\vec{k}'}(\hat{\Gamma}\hat{\Sigma})_{\vec{k}'}e^{i(f-j)\cdot\vec{k}'} \quad \text{(B18)}$$

comparison with expression (B17) indicates that

$$(\hat{\Gamma}\hat{\Sigma})_{\vec{k}'} = \left(\frac{2\pi\langle S^Z\rangle}{\Theta(a)}\right)^2 \frac{1}{N}\sum_{\vec{k}} \Gamma_{\vec{k}'}\Gamma_{\vec{k}}\left[2\mathscr{J}_2(0) - 2\mathscr{J}_2(\vec{k}') - 2\mathscr{J}_2(\vec{k}) + \mathscr{J}_2(\vec{k}+\vec{k}') + \mathscr{J}_2(\vec{k}-\vec{k}')\right] \quad \text{(B19)}$$

The $\hat{\Gamma}$ and $\hat{\Sigma}$ are both diagonal in reciprocal lattice space, so that

$$(\hat{\Gamma}\hat{\Sigma})_{\vec{k}'} = \Gamma_{\vec{k}'}\Sigma_{\vec{k}'} \quad \text{(B20)}$$

The self-energy can now be written in diagonal form,

$$\Sigma^a_{\vec{k}'} = \left(\frac{2\pi\langle S^Z\rangle}{\Theta(a)}\right)^2 \frac{1}{N}\sum_{\vec{k}} \Gamma_{\vec{k}}\left[2\mathscr{J}_2(0) - 2\mathscr{J}_2(\vec{k}') - 2\mathscr{J}_2(\vec{k}) + \mathscr{J}_2(\vec{k}+\vec{k}') + \mathscr{J}_2(\vec{k}-\vec{k}')\right] \quad \text{(B21)}$$

Application to Cubic Systems with Nearest-Neighbor Interactions

If we rewrite the sum in equation (B21) as

$$\frac{1}{N} \sum_{\vec{k}} \Gamma_{\vec{k}} \left\{ 2\left[\mathscr{J}_2(0) - \mathscr{J}_2(\vec{k}') \right] + 2\left[\mathscr{J}_2(0) - \mathscr{J}_2(\vec{k}) \right] \right.$$

$$\left. - \left[\mathscr{J}_2(0) - \mathscr{J}_2(\vec{k} + \vec{k}') \right] - \left[\mathscr{J}_2(0) - \mathscr{J}_2(\vec{k} - \vec{k}') \right] \right\} \quad \text{(B22)}$$

we recognize terms similar in form to the zero-field spin wave energies. For example, from equation (58), when $H = 0$

$$E_{\vec{k}} = \hbar \langle S^z \rangle \left[\mathscr{J}(0) - \mathscr{J}(\vec{k}) \right] \quad \text{(B23)}$$

From the definitions of $\mathscr{J}_2(\vec{k})$ and $\mathscr{J}(\vec{k})$, equations (B15) and (59), respectively, we see that when only nearest-neighbor interactions are considered

$$\mathscr{J}_2(0) - \mathscr{J}_2(\vec{k}) = \frac{j^2}{J} \left[\mathscr{J}(0) - \mathscr{J}(\vec{k}) \right] = \frac{j^2 E_{\vec{k}}}{J \hbar \langle S^z \rangle} \quad \text{(B24)}$$

Here j^2 is the mean-square deviation for nearest-neighbor exchange interactions, and J is the nearest-neighbor exchange integral. We can use the results of appendix A, where $E_{\vec{k}}$ is evaluated for the cubic systems, to evaluate equation (B24).

For the face-centered cubic system, for example,

$$\mathscr{J}_2(0) - \mathscr{J}_2(\vec{k}) = 4j^2 \left[\sin^2 \frac{a}{4} (k_x + k_y) + \sin^2 \frac{a}{4} (k_x - k_y) + \sin^2 \frac{a}{4} (k_y + k_z) \right.$$

$$\left. + \sin^2 \frac{a}{4} (k_y - k_z) + \sin^2 \frac{a}{4} (k_z + k_x) \sin^2 \frac{a}{4} (k_z - k_x) \right] \quad \text{(B25)}$$

where an alternative expression to equation (A42) has been employed to represent $E_{\vec{k}}$ in terms of the squares of sine functions. This permits equation (B21) to be written

$$\Sigma_{\vec{k}}^a = \frac{2 \langle S^z \rangle j^2}{\Theta(a) J} \frac{1}{N} \sum_{k'} \left[\frac{1}{x - x_{\vec{k}'}} \left(\sum_{i=1}^{3} \sin^2 k_i^+ \sin^2 k_i'^+ + \sin^2 k_i^- \sin^2 k_i' \right) \right] \quad \text{(B26)}$$

where

$$k_1^\pm = \frac{a}{4}(k_x \pm k_y)$$

$$k_2^\pm = \frac{a}{4}(k_y \pm k_z)$$

$$k_3^\pm = \frac{a}{4}(k_z \pm k_x)$$

(B27)

The sum in equation (B26) can be rewritten

$$\sum_{i=1}^{3}\left(\sin^2 k_i^+ \sum_{\vec{k}'} \frac{\sin^2 k_i'^+}{x - x_{\vec{k}'}} + \sin^2 k_i^- \sum_{\vec{k}'} \frac{\sin^2 k_i'^-}{x - x_{\vec{k}'}}\right)$$

(B28)

Because of the equivalence of the lattice sites in a fcc system

$$\sum_{\vec{k}'} \frac{\sin^2 k_i'^+}{x - x_{\vec{k}'}} = \sum_{\vec{k}'} \frac{\sin^2 k_i'^-}{x - x_{\vec{k}'}}$$

(B29)

allowing expression (B28) to be simplified to

$$\sum_{i=1}^{3}\left(\left[\sin^2 k_i^+ + \sin^2 k_i^-\right] \sum_{\vec{k}'} \frac{\sin^2 k_i'^+}{x - x_{\vec{k}'}}\right)$$

(B30)

But

$$\sum_{\vec{k}'} \frac{\sin^2 k_i'^+}{x - x_{\vec{k}'}} = \frac{1}{3} \sum_{\vec{k}'} \frac{1}{x - x_{\vec{k}'}} \sum_{j=1}^{3} \sin^2 k_j'^+ = \frac{2}{3} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}} \tag{B31}$$

again because of symmetry considerations. This further simplifies expression (B28) to

$$\frac{8}{3} x_{\vec{k}} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}} \tag{B32}$$

which, when substituted back into equation (B26), gives

$$\Sigma_{\vec{k}}^{a} = \frac{\pi j^2}{3\hbar\Theta(a)J^2} E_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{x_{\vec{k}'}}{x - x_{\vec{k}'}} \tag{B33}$$

Since the disorder parameter is

$$\rho = \frac{2}{Z} \frac{j^2}{J^2} \tag{B34}$$

where Z is the number of nearest neighbors

$$\Sigma_{\vec{k}}^{a} = \frac{2\pi\rho}{\hbar\Theta(a)} E_{\vec{k}} \frac{1}{N} \sum_{\vec{k}'} \frac{E_{\vec{k}'}}{E - E_{\vec{k}'}} \tag{B35}$$

The other two cubic systems, sc and bcc, can be handled in the same manner since the symmetry arguments are the same. Doing this verifies that equation (B35) is a general result for the three cubic systems.

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