# Spin-wave spectrum of an amorphous ferromagnet\*

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The spin-wave spectrum of an amorphous Heisenberg ferromagnet is calculated by a diagrammatic expansion making use of a transformation suggested by Taylor and Wu. The upper limit of the spectrum is found to occur at frequencies below those of the corresponding crystalline system, while the low-frequency part of the spectrum is enhanced. Internal van Hove singularities are absent in the spin-wave spectrum of the amorphous ferromagnet.

## I. INTRODUCTION

The problem of excitations in disordered systems remains to date one of the greatest challenges in the physics of condensed matter. Nontrivial exact calculations are confined for the most part to one-dimensional systems, <sup>1</sup> and even there the excitation spectra must be found as the solution of functional equations by numerical computations<sup>2</sup> that do not always yield great physical insight into the problem. In three dimensions the only exact results not confined to pathological models are in the form of theorems predicting band gaps under certain circumstances.<sup>3</sup>

The difficulties encountered in the search for exact results in disordered systems have led to a large number of attempts to find approximation schemes that will predict the gross features of the excitation spectrum. Unfortunately, the perceptive remarks of Lifshitz,<sup>4</sup> since confirmed by direct computations,<sup>2</sup> indicate that in some systems the spectrum exhibits essential singularities at the edges of bands and that a great wealth of fine structure exists in the spectrum or its derivatives. One can thus not hope that the results of any of the simpler approximation schemes will always be an accurate indication of the true nature of the spectrum. However, one does expect that as the approximations become more detailed, more and more of the general features of the true spectrum will begin to appear. In a perturbation-theory approach in particular, successive renormalizations and inclusions of higher-order diagrams will be expected to improve quantitatively the accuracy of the approximate spectrum, even though one might not expect to produce such qualitative features as essential singularities.

Perturbation expansions have been particularly widely employed in the theory of alloys, <sup>5</sup> for there it is usually possible to take some perfectly periodic crystal as the unperturbed system and to apply the disordering elements as a perturbation. The theory then proceeds in terms of summations over wave numbers restricted to lie within a single Brillouin zone of the perfect lattice. Perturbation theory is also readily applicable to the problem of electrons moving in the potential due to an amorphous array of scatterers; here the unperturbed Hamiltonian may be taken as the kinetic energy of the free electrons and the perturbation is the entire potential of the scatterers.

The most difficult challenge to the perturbationtheory approach appears in the problem of the spinwave spectrum of an amorphous ferromagnet at low temperatures and in its counterpart, the vibrational spectrum of a glassy solid. Of these two the ferromagnet is the simpler problem in practice and is the one that will be attempted in the present paper. The vibrational system has added complications that can be traced to the fact that there are three polarization directions associated with the phonons in a Bravais lattice, and the Green's-function expansions must consequently deal with tensor quantities. From a more practical point of view it also appears more apposite to consider the amorphous ferromagnet in that experimental results on some such systems are beginning to become available.<sup>6</sup>

While many workers have attempted theoretical investigations of noncrystalline ferromagnets, a large proportion of these have employed some type of mean-field approximation and have thus not been able to discuss the form of the spin-wave spectrum but only the form of the magnetization M(T) and the corresponding Curie temperature,  $T_c$ . Handrich,<sup>7</sup> for example, used a mean-field model in which he assumed that structural fluctuations in the amorphous material would produce fluctuations in the exchange interaction. The Brillouin function was expanded in terms of small deviations from an average exchange, the root mean square of this deviation characterizing the degree of disorder. In subsequent calculations<sup>8</sup> more sophisticated types of mean-field approach were used in an at-

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tempt to compensate for the absence of any effects of short-range order in the earlier work. In some of these theories<sup>7,8</sup> M(T) and  $T_C$  were found to be reduced by the presence of disorder, while in others<sup>9,10</sup> they were increased.

The application of perturbation theory to disordered ferromagnets through a Green's-function formalism has, as far as we know, been limited to disordered alloys, or at least to systems in which a Bravais lattice exists, as in the case of the model chosen by Montgomery et al.<sup>11</sup> These authors allowed the exchange interaction to fluctuate about a mean value and made approximations sufficient to allow summation of a selected set of diagrams. While their results did indicate a reduction of M(T) and  $T_c$  by the disorder, the calculation must unfortunately be considered suspect; grounds for this suspicion include the fact that the van Hove singularities in the spectrum are not predicted to be removed, but on the contrary are increased in number and that, in addition, the lowfrequency part of the spectrum does not tend to the correct continuum limit, but deviates from it by a considerable amount. In summary, then, there appears to exist no previous calculation of the spin-wave spectrum of an amorphous Heisenberg ferromagnet, and those calculations of latticebased models that attempt to mimic the amorphous situation fail in some important respect. A fresh approach is thus called for.

In Sec. II the formalism is presented whereby a transformation due to Taylor and Wu<sup>12</sup> (hereafter referred to as the TW transformation) is adapted to produce a diagrammatic expansion for the Green's function of an amorphous Heisenberg ferromagnet at low temperatures. In Sec. III these methods are applied to some model amorphous systems and results are given for the spin-wave spectra and for a rough estimate of the depression of the Curie temperature by the disorder. In Sec. IV these results are discussed in comparison with previous work.

#### **II. FORMALISM**

In the Heisenberg model of a spin- $\frac{1}{2}$  ferromagnet<sup>13</sup> the Hamiltonian is written as

$$H = -\sum_{\mathbf{1},\mathbf{1}'} J(\mathbf{\overline{1}},\mathbf{\overline{1}'}) \, \mathbf{\overline{s}}(\mathbf{\overline{1}}) \cdot \mathbf{\overline{s}}(\mathbf{\overline{1}'}) \,, \qquad (1)$$

with  $\vec{s}(\vec{1})$  the spin operator for the atom whose equilibrium position is described by the vector  $\vec{1}$ , and  $J(\vec{1},\vec{1}')$  the exchange interaction between spins at  $\vec{1}$  and  $\vec{1}'$ . For a perfect crystal  $J(\vec{1},\vec{1}')$  would only depend on the vector  $\vec{1} - \vec{1}'$  joining the pair of sites, but in an amorphous system this simplification is no longer possible. We may, however, choose  $J(\vec{1},\vec{1}')$  to be equal to  $J(\vec{1}',\vec{1})$ . With the definition<sup>13</sup> of the customary spin-raising and -lowering operators

$$s^* = s_x + is_y$$
;  $s^- = s_x - is_y$ 

and the Holstein-Primakoff boson-creation and -destruction operators  $a_1^{\dagger}$  and  $a_1^{\dagger}$  through the equations

$$s^{+}(1) = \hbar (1 - n_{\bar{1}})^{1/2} a_{\bar{1}};$$
  

$$s^{-}(\bar{1}) = \hbar a_{\bar{1}}^{\dagger} (1 - n_{\bar{1}})^{1/2},$$

with

$$n_{\bar{1}} = a_{\bar{1}} a_{\bar{1}} a_{\bar{1}}$$
,

one finds

$$H = -\sum_{\vec{1},\vec{1}'} J(\vec{1},\vec{1}') \hbar^2 [\frac{1}{2} (1 - n_{\vec{1}})^{1/2} a_{\vec{1}} a_{\vec{1}}^{\dagger} (1 - n_{\vec{1}'})^{1/2} + \frac{1}{2} a_{\vec{1}}^{\dagger} (1 - n_{\vec{1}})^{1/2} (1 - n_{\vec{1}'})^{1/2} a_{\vec{1}'} + (\frac{1}{2} - n_{\vec{1}}) (\frac{1}{2} - n_{\vec{1}'})] .$$
(2)

The very complicated equations of motion for the  $a_{\bar{1}}^{\dagger}$  that result from this Hamiltonian reduce to a simple form at the lowest of temperatures, where the factors of  $(1 - n_{\bar{1}})^{1/2}$  and  $(1 - n_{\bar{1}})^{1/2}$  may be replaced by unity. One then finds

$$[H, a_{\tilde{1}}^{\dagger}] = \hbar^{2} \sum_{\tilde{1}'} J(\tilde{1}, \tilde{1}') (a_{\tilde{1}}^{\dagger} - a_{\tilde{1}'}^{\dagger}) , \qquad (3)$$

from which the excitation energies  $\hbar \omega$  for the spin waves are seen to be given by the eigenvalue equations

$$\sum_{\vec{1}'} \left[ \delta_{\vec{1} \vec{1}'} \left( \hbar \omega - \hbar^2 \sum_{\vec{1}''} J(\vec{1}, \vec{1}'') \right) + \hbar^2 J(\vec{1}, \vec{1}') \right] a_{\vec{1}'}^{\dagger} = 0 , \quad (4)$$

which we abbreviate as

$$\hbar \sum_{i'} D^{-1}(\bar{1},\bar{1}',\omega) a_{\bar{1}'}^{\dagger} = 0 .$$

The frequency density of spin-wave modes is then given by

$$g(\omega) = -\pi^{-1} \operatorname{Im} \sum_{i} D(\bar{1}, \bar{1}, \omega + i0) ,$$

with the Green's function  $D(\vec{1},\vec{1}',\omega)$  defined by

$$\sum_{\mathbf{i}'} D^{-1}(\mathbf{i},\mathbf{i}',\omega) D(\mathbf{i}',\mathbf{i}'',\omega) = \delta_{\mathbf{i}\mathbf{i}}.$$

We rewrite Eq. (4) as

$$\sum_{\vec{1}'} [\omega \, \delta_{\vec{1} \, \vec{1}'} - V(\vec{1}, \vec{1}')] a_{\vec{1}'}^{\dagger} = 0 , \qquad (5)$$

where

$$V(\vec{1},\vec{1}') = -\hbar \left[ J(\vec{1},\vec{1}') - \delta_{\vec{1}\vec{1}'} \sum_{\vec{1}''} J(\vec{1},\vec{1}'') \right] \quad .$$

One notes the presence of disorder in both the diagonal and off-diagonal elements of V(1, 1').

In this notation the Green's function may be expressed in the familiar form 3830

(6)

$$D = D_0 + D_0 VD ,$$

with

 $D_0 = (\omega + i0)^{-1}I$ 

and I the unit matrix. The difficulty that is encountered when one attempts to solve Eq. (6) by iteration then becomes obvious, as convergence will clearly be at best painfully slow when  $\omega$  is small. In physical terms one may say that at the lowest of frequencies the eigenstates of the system will be well approximated by travelling spin waves of large wavelength because the disorder becomes negligible when averaged over distances much larger than the average interatomic spacing. In order to derive a frequency spectrum that approaches this continuum limit at low frequencies one clearly needs to include diagrams of very high order in a perturbation theory based directly on Eq. (5).

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Had we been dealing with a perfectly periodic system, the problem of long-range correlations in the low-frequency excitations would have been simply solved by a transformation to a wave-number representation by means of the transformation matrix S having elements

$$S_{\vec{1}\vec{q}} = N^{-1/2} e^{-i\vec{q}\cdot\vec{1}}$$

with the N vectors  $\vec{q}$  defined to lie within the first Brillouin zone. The unitary matrix S has an inverse E with elements

$$E_{\pi\pi} = N^{-1/2} e^{i \bar{q} \cdot \bar{1}}$$

In an amorphous material E is no longer the inverse of S for any choice of the N vectors  $\mathbf{q}$  and one instead finds

$$ES = I + R$$
,

with

$$R_{\vec{q}\,\vec{q}'} = N^{-1} \sum_{\vec{i}} e^{i(\vec{q}-\vec{q}')\cdot\vec{i}}; \quad (\vec{q}\neq\vec{q}'),$$

and

$$R_{\bar{q}\bar{q}'} = 0; (\bar{q} = \bar{q}').$$

An important step in the TW formalism occurs when it is noted that one may write

$$S^{-1} = (I+R)^{-1}E$$

and so Eq. (6) may be transformed to a wave-number representation in which

$$\tilde{D} = D_0 + D_0 (S^{-1} VS) \tilde{D} , \qquad (7)$$

where

$$\tilde{D} = S^{-1} DS$$

and the matrix elements of  $S^{-1}VS$  are given by

$$(S^{-1}VS)_{\vec{\mathfrak{q}}\vec{\mathfrak{q}}'} = N^{-1} \sum_{\vec{\mathfrak{q}}'', \vec{\mathfrak{l}}, \vec{\mathfrak{l}}'} [(I+R)^{-1}]_{\vec{\mathfrak{q}}\vec{\mathfrak{q}}''}$$

$$\times e^{i\vec{\mathfrak{q}}^{\prime\prime}\cdot\vec{\mathfrak{l}}} V(\vec{\mathfrak{l}},\vec{\mathfrak{l}}^{\prime}) e^{-i\vec{\mathfrak{q}}^{\prime}\cdot\vec{\mathfrak{l}}^{\prime}}$$

$$= N^{-1} \sum_{\vec{\mathfrak{q}}^{\prime\prime}} [(I+R)^{-1}]_{\vec{\mathfrak{q}}\vec{\mathfrak{q}}^{\prime\prime}} \sum_{\vec{\mathfrak{l}}} e^{i(\vec{\mathfrak{q}}^{\prime\prime}-\vec{\mathfrak{q}}^{\prime})\cdot\vec{\mathfrak{l}}}$$

$$\times \sum_{\vec{\mathfrak{l}}(\vec{\mathfrak{l}})} V(\vec{\mathfrak{l}},\vec{\mathfrak{l}}+\vec{\mathfrak{L}}) e^{-i\vec{\mathfrak{q}}^{\prime\prime}\cdot\vec{\mathfrak{L}}(\vec{\mathfrak{l}})} .$$

$$(8)$$

Here  $\vec{L}(\vec{1})$  is written for the displacement  $\vec{1}' - \vec{1}$ . The set of vectors  $\{\vec{L}\}$  represents the configuration of atomic sites as seen from the site at  $\vec{l}$ ; whereas in a Bravais lattice  $\{\vec{L}\}$  would be independent of  $\vec{1}$ the amorphous system does not possess this property.

It is now convenient to decompose the sum over  $\vec{L}$  in Eq. (8) into two parts, an average  $V_0(\vec{q}')$  over all sites  $\vec{l}$  and the site-dependent deviation from that average. We write

$$\sum_{\vec{L}} V(\vec{1}, \vec{1} + \vec{L}) e^{-i\vec{q}' \cdot \vec{L}} = V_0(\vec{q}') + U(\vec{1}, \vec{q}') , \qquad (9)$$
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$$\sum_{\vec{\tau}} U(\vec{1}, \vec{q}) = 0 .$$

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One then finds

$$(S^{-1} VS)_{\vec{a} \vec{a}'} = V_0(\vec{q}) \delta_{\vec{a} \vec{a}'} + W(\vec{q}, \vec{q}') , \qquad (10)$$

with

$$W(\mathbf{\ddot{q}},\mathbf{\ddot{q}}') = \sum_{\mathbf{\ddot{q}}'} \left[ (I+R)^{-1} \right]_{\mathbf{\ddot{q}} \mathbf{\ddot{q}}'} \sum_{\mathbf{\vec{q}}} T(\mathbf{\ddot{1}},\mathbf{\ddot{q}}'',\mathbf{\ddot{q}}')$$

and

$$T(\mathbf{\bar{l}},\mathbf{\bar{q}}'',\mathbf{\bar{q}}') = N^{-1}e^{i(\mathbf{\bar{q}}''-\mathbf{\bar{q}}')\cdot\mathbf{\bar{l}}}U(\mathbf{\bar{l}},\mathbf{\bar{q}}')$$

A new propagator  $G_0$  that includes all possible interactions involving  $V_0(\vec{q})$  is then defined by the equation

$$G_0 = (D_0^{-1} - V_0)^{-1} .$$

Equation (7) then becomes

$$\tilde{D} = (G_0^{-1} - M)^{-1} , \qquad (11)$$

where M is the proper self-energy appropriate to the interaction W and the propagator  $G_0$ . The first few terms in the sum that forms M are shown in Fig. 1. The open circles represent factors of



FIG. 1. Each diagram represents a sum of terms in the perturbation expansion of the proper self-energy M for an amorphous system.

- R while the horizontal lines are the renormalized propagators  $G_0$ . The vertical dashed lines represent factors of T, the terminal cross denoting the particular site  $\tilde{I}$  to which T refers.

With the perturbation expansion expressed in this way some separation of the structural disorder of the system from the dynamical disorder has been achieved, in that the factors of R relate only to the amorphous structure of the material and are in no way dependent on the presence of any disorder in the magnitude of the interaction between adjacent spins.

# **III. THE MODEL**

In order to evaluate the various terms in the diagrammatic expansion it is necessary to specialize to some explicit model for the functions  $V_0(\vec{q})$  and  $U(\vec{1}, \vec{q})$  defined by Eq. (9). From the formulation of the general linearized Heisenberg problem leading to Eq. (5) one has

$$V_0(\vec{\mathbf{q}}) = \frac{\hbar}{N} \sum_{\vec{\mathbf{l}}, \vec{\mathbf{L}}} J(\vec{\mathbf{l}}, \vec{\mathbf{l}} + \vec{\mathbf{L}}) (\mathbf{1} - e^{-i\vec{\mathbf{q}} \cdot \vec{\mathbf{L}}})$$
(12)

and hence

$$U(\vec{1},\vec{q}) = \sum_{\vec{L}(\vec{1})} J(\vec{1},\vec{1}+\vec{L}) (1-e^{-i\vec{q}\cdot\vec{L}}) - V_0(\vec{q}) .$$
(13)

The vectors  $\vec{L}(\vec{1})$  are the positions of the various neighboring atoms relative to the position of the atom at  $\vec{1}$  under consideration.

While it will be desirable to make use of a model that is as simple as possible, there are a number of conditions that must be imposed if some connection with real physical systems is to be maintained. First, we stipulate that a well-defined coordination number z exists and is the same for each atom, a condition that corresponds to the idea that the atoms are linked by bonds. This is equivalent to saying that there is an unambiguous distinction between an atom's nearest neighbors and all other neighbors. We shall take an extreme case of such a bonding picture and suppose that a unique distance a separates each atom from its z nearest neighbors and that the exchange coupling is the same constant J for all such nearestneighbor bonds and zero otherwise.

While the lengths of such bonds may reasonably be assumed uniform, there must necessarily be some variation in the angles between them if a truly amorphous structure is to be constructed. This variation, however, need by no means be large and we shall assume it negligible for the purposes of computation. Each atom is thus assumed to be surrounded by z nearest neighbors to form a cluster identical to that which would be found in a crystalline solid. The difference between the amorphous and crystalline materials lies in the fact that the orientations of these clusters will vary from site to site in a random way in the amorhous case and will not repeat periodically as in a crystal. Equations (12) and (13) thus become

$$V_{0}(\vec{q}) = \hbar z J [1 - j_{0}(qa)]$$
(14)

and

$$U(\vec{1},\vec{q}) = \hbar J \left[ z \, j_0(qa) - \sum_{\vec{a}(\vec{1})} e^{-i\vec{q}\cdot\vec{a}(\vec{1})} \right], \qquad (15)$$

with  $j_0$  the spherical Bessel function of zero order and  $\vec{a}(\vec{l})$  the set of vectors that define the nearestneighbor positions relative to the atom at  $\vec{l}$ . This set of vectors is uniquely defined by three numbers which may be taken as the Euler angles for the rotation of the cluster from some reference orientation.

The rapid convergence of the perturbation series derived in the TW formalism is obtained at the expense of working in the mixed representation in which the various quantities involved are functions of both position 1 and wavenumber  $\overline{q}$  and it now becomes necessary to specify the allowed values of **d**. We impose periodic boundary conditions over some large volume  $\Omega$  to define a density  $\Omega/8\pi^3$  of allowed points in q space and then arbitrarily restrict  $\vec{q}$  to lie inside a sphere of radius  $q_p$  $=(6\pi^2 N/\Omega)^{1/3}$ . While we could equally well have chosen in place of this sphere the first Brillouin zone of some Bravais lattice, the sphere reflects more directly the isotropy of the amorphous system. It also provides a more stringent test of the formalism than any other choice in that it leads to an unperturbed density of states having a large discontinuity that must be removed by further diagrammatic summations.

In the present calculation we shall content ourselves with the evaluation of only the lowest-order terms in the expansion; this will suffice to show that our method can yield physically reasonable results with minimal labor. Accordingly we look first at Fig. 2(a) which contributes to the selfenergy an amount



FIG. 2. In the simplest nontrivial calculation only these two diagrams are summed.

$$\begin{split} M_{a}(\vec{\mathbf{q}}) &= -\sum_{\vec{\mathbf{l}}} R_{\vec{\mathbf{q}} \vec{\mathbf{q}}'} T(\vec{\mathbf{l}}, \vec{\mathbf{q}}', \vec{\mathbf{q}}) \\ &= -N^{-2} \sum_{\vec{\mathbf{l}}, \vec{\mathbf{L}}(\vec{\mathbf{l}}), \vec{\mathbf{q}}'} e^{i(\vec{\mathbf{q}} \cdot \vec{\mathbf{q}}') \cdot \vec{\mathbf{L}}} U(\vec{\mathbf{l}}, \vec{\mathbf{q}}) , \qquad (16) \end{split}$$

where  $\vec{L}(\vec{1}) = \vec{1}' - \vec{1}$ . Because the amorphous array is macroscopically isotropic,  $M_a(\vec{q})$  must be independent of the direction of  $\vec{q}$ , and we can replace  $e^{i\vec{q}\cdot\vec{L}}U(\vec{1},\vec{q})$  in this particular term by its average over all directions of  $\vec{q}$ . We then find

$$M_a = M_a^{(1)} + M_a^{(2)} ,$$

with

$$M_{a}^{(1)} = -\frac{1}{4\pi N^{2}} \sum_{\vec{l},\vec{a},\vec{q}'} e^{-i\vec{q}'\cdot\vec{a}} \int U(\vec{l},\vec{q}) e^{i\vec{q}\cdot\vec{a}} d\Omega_{\vec{q}}, \quad (17)$$

$$M_{a}^{(2)} = -\frac{1}{4\pi N^{2}} \sum_{\vec{l},\vec{q}'} \sum_{\vec{L}}' e^{-i\vec{q}'\cdot\vec{L}} \int U(\vec{l},\vec{q}) e^{i\vec{q}\cdot\vec{L}} d\Omega_{\vec{q}}, \quad (18)$$

where  $d\Omega_{\vec{q}}$  is an element of solid angle in the  $\vec{q}$  direction and the prime on the sum over  $\vec{L}$  indicates that the nearest-neighbor terms  $\vec{L} = \vec{a}$  have been excluded from the summation. Now

$$\sum_{\vec{q}'} e^{-i\vec{q}'\cdot\vec{a}} = N \mathcal{G}(q_D a) \quad ,$$

where

$$G(x) = 3x^{-3}(\sin x - x\cos x).$$

Thus, because the magnitudes of the vectors  $\vec{a}(\vec{l})$ in expression (15) are independent of  $\vec{l}$ , the summation over  $\vec{q}'$  and the integration over  $d\Omega_{\vec{q}}$  are independent, and substitution from Eq. (15) yields

$$M_{a}^{(1)} = -z\hbar J \left( S\left(q_{D}a\right) \left( zj_{0}^{2}(qa) - \sum_{\vec{a}'} j_{0}(q | \vec{a} - \vec{a}' | ) \right).$$
(19)

The remaining contribution  $M_a^{(2)}$  to the diagram of Fig. 2(a) can be immediately seen from Eq. (18) to be small; because the average over directions of  $\vec{q}$  of  $U(\vec{1},\vec{q})$  vanishes, it is necessary for there to be correlations in the directions of  $\vec{L}$  and of the nearest neighbors  $\vec{a}(\vec{1})$  in order for this term to be nonvanishing. Explicitly one has

$$M_{a}^{(2)} = -\left(\frac{\hbar J}{N}\right) \sum_{\mathbf{I}} \sum_{\mathbf{L}'} \mathcal{G}\left(q_{D}L\right) \\ \times \left(zj_{0}(qa) |j_{0}(qL) - \sum_{\mathbf{i}'} j_{0}(q | \mathbf{i}' - \mathbf{L}|)\right), \quad (20)$$

which indeed vanishes when three-particle correlations are ignored; then  $\vec{a}'$  and  $\vec{L}$  may be taken to be randomly oriented and since

$$j_{0}(q | \mathbf{\dot{a}}' - \mathbf{\dot{L}} |) = \sum_{I,m} 4\pi j_{I}(qa) \times j_{I}(qL) Y_{Im}^{*}(\hat{a}') Y_{Im}(\hat{L}) , \qquad (21)$$

the second term in the large parentheses in Eq. (20) cancels the first.

The general pattern of analysis just completed may be repeated in the evaluation of the more comlicated diagrams in the perturbation series. For the purposes of the present calculation it will suffice to consider only the one further term  $M_b$  that is depicted in Fig. 2(b). This yields a contribution to the proper self-energy of

$$\begin{split} M_b(\vec{\mathbf{q}}) = N^{-2} \sum_{\vec{\mathbf{l}}\,,\,\vec{\mathbf{l}}\,',\,\vec{\mathbf{q}}\,'} e^{i(\vec{\mathbf{q}}-\vec{\mathbf{q}}\,')\cdot\vec{\mathbf{l}}} U(\vec{\mathbf{l}}\,,\vec{\mathbf{q}}\,') \\ \times G_0(\vec{\mathbf{q}}\,') e^{i(\vec{\mathbf{q}}\,'-\vec{\mathbf{q}}\,)\cdot\vec{\mathbf{l}}\,'} U(\vec{\mathbf{l}}\,',\vec{\mathbf{q}}\,) \,, \end{split}$$

with

$$G_0(\vec{\mathbf{q}}) = \{\omega + i\mathbf{0} - \hbar z J [1 - j_0(qa)]\}^{-1} .$$
 (22)

It is again convenient to split this summation into two sets of terms, so that

$$M_{b} = M_{b}^{(1)} + M_{b}^{(2)}$$

with

$$M_{b}^{(1)} = N^{-2} \sum_{\vec{1}, \vec{a}, \vec{q}'} U(\vec{1}, \vec{q}') G_{0}(q')$$
$$\times e^{i\vec{q}' \cdot \vec{a}} U(\vec{1} + \vec{a}, \vec{q}) e^{-i\vec{q} \cdot \vec{a}}$$
(23)

and

$$M_{b}^{(2)} = N^{-2} \sum_{\vec{l},\vec{q}'} \sum_{\vec{L}}' U(\vec{l},\vec{q}') G_{0}(q')$$
$$\times e^{i\vec{q}'\cdot\vec{L}} U(\vec{l}+\vec{L},\vec{q}) e^{-i\vec{q}\cdot\vec{L}} , \qquad (24)$$

where again  $\bar{a}$  are the nearest-neighbor vectors of  $\bar{l}$  and the prime on the summation over  $\bar{L} = \bar{l}' - \bar{l}$  indicates that the terms for which  $\bar{L} = \bar{a}$  are excluded. Because  $M_b$  must also be isotropic, Eqs. (23) and (24) may be replaced by their averages over all directions of  $\bar{q}$ , which then also allows the angular part of the summation over  $\bar{q}'$  to be performed. One then finds

$$M_{b}^{(1)} = \frac{3z \hbar^{2} J^{2}}{q_{D}^{3}} \left( z j_{0}^{2}(qa) - \sum_{\vec{a}'} j_{0}(q | \vec{a} + \vec{a}'' |) \right) \\ \times \int_{0}^{q_{D}} G_{0}(q') q'^{2} dq' \\ \times \left( z j_{0}^{2}(q'a) - \sum_{\vec{a}'} j_{0}(q' | \vec{a} - \vec{a}' |) \right) .$$
(25)

In this expression the vectors  $\mathbf{a}'$  are again the nearest-neighbor vectors of  $\mathbf{l}$ , but the vectors  $\mathbf{a}''$ connect the site  $\mathbf{l} + \mathbf{a}$  with its nearest neighbors. The significant point to note here is that although the quantities  $|\mathbf{a} + \mathbf{a}''|$  and  $|\mathbf{a} - \mathbf{a}'|$  may represent the lengths of vectors joining sites that are not nearest neighbors, one does not need any complicated statistical information about the amorphous structure to attach a value to them; in the present model, for instance, they will depend only on the fixed magnitude *a* of the nearest-neighbor distance and on the *bond angle* between such vectors, which in our case will be constant. Under these circumstances

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TABLE I. The parameters  $z_n$  and  $\gamma_n^2$  listed for various types of amorphous structure.

2	<b>x</b> _0^3	Crystalline analog	n	zn	$\gamma_n^2$
4	$9\pi^2 3^{1/2}/4$	diamond	1	3	83
6	$6\pi^2$	simple cubic	1 2	4 1	2 4
8	$9\pi^2 3^{1/2}/2$	body-centered cubic	1 2 3	3 3 1	4 3 8 3 8 3 4
12	$6\pi^2 2^{1/2}$	face-centered cubic	1 2 3 4	4 2 4 1	1 2 3 4

$$\sum_{\mathbf{a'}'} j_0(q \, \big| \, \mathbf{\ddot{a}} + \mathbf{\ddot{a}'}' \big| \,) = \sum_{\mathbf{\ddot{a}'}} j_0(q \, \big| \, \mathbf{\ddot{a}} - \mathbf{\ddot{a}'} \big| \,) \,,$$

which simplifies Eq. (25).

With similar arguments to those used in discussing  $M_a^{(2)}$  one can show  $M_b^{(2)}$  to be small. We in fact find

$$M_{b}^{(2)} = \frac{3\hbar^{2}J^{2}}{Nq_{D}^{3}} \sum_{\vec{1},\vec{L}} \left( zj_{0}(qa)j_{0}(qL) - \sum_{\vec{a}''} j_{0}(q | \vec{L} + \vec{a}'' |) \right) \int_{0}^{q_{D}} G_{0}(q')q'^{2} dq' \\ \times \left( j_{0}(q'a)j_{0}(q'L) - \sum_{\vec{a}'} j_{0}(q' | \vec{L} + \vec{a}'' |) \right), \quad (26)$$

which vanishes when we make use of Eq. (21) and make the reasonable assumption that the pair distribution function is reasonably constant for distances greater than 2*a*. While no further diagrams will be evaluated directly in this first calculation of the spectrum we shall introduce an element of self-consistency by replacing  $G_0(q')$  in Eq. (25) by



FIG. 3. The dots show the spin-wave spectrum of a model amorphous ferromagnet with coordination number z=6. The continuous line is the spectrum of the crystal-line analog, while the dashed line shows the continuum limit to which the spectra tend at low frequencies. The frequency is measured in units of  $\hbar J$ .



FIG. 4. This figure differs from Fig. 3 in showing the case where z=8.

the complete Green's function  $\tilde{D}(q')$ . We then have as our approximation for the proper self-energy

$$\begin{split} M(q,\omega) &= -z\hbar JS(qa) \left( \Im(q_D a) - (\Im \hbar J/q_D^3) \right) \\ &\times \int_0^{q_D} \tilde{D}(q') S(q'a) q'^2 dq' \right) \;, \end{split}$$

with

$$S(qa) = zj_0^2(qa) - \sum_{\vec{a}'} j_0(q | \vec{a} - \vec{a}' |)$$
$$= zj_0^2(qa) - 1 - \sum z_n j_0(\gamma_n qa)$$

and with  $z_n$  and  $\gamma_n$  parameters listed in Table I for each coordination number and symmetry. The magnitude of the product  $x_0 = q_D a$  is determined by our assumed condition that the density of the amorphous system is equal to that of the crystalline material having the same z. Writing x for qa we then have the equations

$$M(x, \omega) = -z\hbar JS(x) \left( \Im(x) - (\Im\hbar J/x_0^3) \times \int_0^{x_0} \tilde{D}(y, \omega)S(y) y^2 dy \right) , \qquad (27)$$

$$\tilde{D}(x,\omega) = [\omega + i0 - V_0(x) - M(x,\omega)]^{-1}, \qquad (28)$$

$$N^{-1}g(\omega) = -(3/\pi x_0^3) \int_0^{x_0} \tilde{D}(x,\,\omega) \, x^2 \, dx \, . \tag{29}$$



FIG. 5. This figure differs from Figs. 3 and 4 in showing the case where z = 12.

It may at first sight appear surprising that physically reasonable results emerge from a treatment in which the perturbation series is truncated at its lowest nontrivial terms. One must recall, however, that for the high-frequency part of the spectrum correlations will only occur in the motions of near neighbors in an amorphous system, while the long-range correlations that exist in the low-frequency modes are well described by the wave-number representation in which we are working.

## **IV. RESULTS**

Numerical calculations of the frequency density  $g(\omega)$  of spin-wave states were performed for z = 6, 8, and 12 (the cases for which the corresponding crystalline spectrum is available for comparison). These results are shown in Figs. 3-5, in which the dots are points calculated for the amorphous material from Eqs. (27)-(29). The continuous lines are the spectra of the simplest crystal having the same coordination number z and are taken from the work of Jelitto, <sup>14</sup> while the dashed lines are the continuum limit. It is reassuring to note that the present results approach this continuum limit at low frequencies and that the area under each curve is unity to within numerical error.

The upper limit of frequency of the spectrum in the amorphous material is in each case reduced below that of its crystalline analog. This is to be expected, since the highest-frequency modes in the crystal occur when precisely half of the spins

- \*Work supported by the U. S. Atomic Energy Commission.
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- <sup>1</sup>J. E. Gubernatis and P. L. Taylor, Solid State Commun. <u>12</u>, 309 (1973); J. Phys. C <u>6</u>, 1889 (1973) and references therein.
- <sup>2</sup>J. E. Gubernatis and P. L. Taylor, J. Phys. C <u>4</u>, L94 (1971).
- <sup>3</sup>P. L. Taylor, Proc. Phys. Soc. <u>88</u>, 753 (1966); <u>90</u>, 233 (1967).
- <sup>4</sup>I. M. Lifshitz, Adv. Phys. <u>16</u>, 49 (1967).
- <sup>5</sup>P. Soven, Phys. Rev. <u>156</u>, 1017 (1967); D. E. Thornton, Phys. Rev. B <u>4</u>, 3371 (1971).
- <sup>6</sup>K. Tanura and H. Endo, Phys. Lett. A <u>29</u>, 52 (1969);
   P. L. Maitrepierre, J. Appl. Phys. <u>40</u>, 4826 (1969);
   T. E. Sharon and C. C. Tsuei, Phys. Rev. B <u>5</u>, 104 (1972).
- <sup>7</sup>K. Handrich, Phys. Status Solidi B 32, K55 (1969).

are precessing in complete antiphase with the other half; such long-range coherence will be absent in the amorphous system.<sup>15</sup> One also notes that the internal van Hove singularities of the crystalline spectrum are absent in the amorphous material. This is perhaps also to be expected, since the various modes may no longer be identified with motions having well-defined wave numbers. It is to be contrasted with the results of the pseudo-crystal model of Montgomery *et al.*,<sup>11</sup> where an increase in the number of singularities was predicted.

The temperature dependence of the magnetization M(T) must be expected to be influenced by the disorder. At the lowest temperatures the deviation of the magnetization from its saturation value will exhibit the usual Bloch  $T^{3/2}$  behavior with a coefficient identical to that of the crystalline analog. At higher temperatures, however, the depletion of the high-frequency end of the spectrum will cause M(T) for the amorphous system to fall below that of the crystal. A very rough estimate of the consequent reduction in the Curie temperature  $T_{C}$ may be obtained from the spin-wave spectrum by use of an approximation suggested by Bogoliubov and Tyablikov, <sup>16</sup> in which both the exchange constant J and the total number of spin-wave modes are themselves assumed proportional to M. Reductions in  $T_c$  of the amorphous ferromagnet below that of the crystalline analog are found to be 3.0, 4.8, and 6.7% for the cases where the coordination number z is 6, 8, and 12, respectively.

- <sup>8</sup>S. Kobe and K. Handrich, Phys. Status Solidi B <u>44</u>, K33 (1971).
- <sup>9</sup>S. Kobe and K. Handrich, Fiz. Tverd. Tela. <u>13</u>, 887 (1971) [Sov. Phys.-Solid State <u>13</u>, 734 (1971)].
- <sup>10</sup>S. Kobe, Phys. Status Solidi B <u>41</u>, K13 (1970).
- <sup>11</sup>C. G. Montgomery, J. I. Krugler, and R. M. Stubbs, Phys. Rev. Lett. 25, 669 (1970).
- <sup>12</sup>P. L. Taylor and S.-Y. Wu, Phys. Rev. B <u>2</u>, 1752 (1970).
- <sup>13</sup>P. L. Taylor, A Quantum Approach to the Solid State (Prentice-Hall, Englewood Cliffs, N.J., 1970), Chap. 3.
- <sup>14</sup>R. J. Jelitto, J. Phys. Chem. Solids <u>30</u>, 609 (1969).
- <sup>15</sup>Such a mode would represent a solution of Eq. (3) with  $a_{1}^{\dagger} = -a_{1}$ , for all neighboring sites. The amorphous material, however, will contain rings of both even and odd numbers of atoms, and this mode thus cannot exist. <sup>16</sup>N. N. Bogoliubov and S. V. Tyablikov, Dokl. Akad.
- Nauk SSSR <u>126</u>, 53 (1959) [Sov. Phys.-Dokl. <u>4</u>, 589 (1959)].

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