

## Scattering of Spin Waves by Magnetic Defects\*

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A previous calculation of the scattering amplitude for the scattering of spin waves by magnetic defects in a simple cubic lattice is simplified and extended to body-centered and face-centered cubic lattices. Expressions are given for the mean free path, and the thermal resistivity due to defect scattering is calculated by a method which takes some account of spin-wave interactions.

### I. SCATTERING THEORY

**I**n a previous study<sup>1</sup> (which will be referred to as I), the general theory of the scattering of spin waves by magnetic defects was discussed, and results were obtained for the cross section and mean free path in simple cubic lattices. In the present note, the cumbersome procedures of that calculation are both simplified and generalized, and the results are extended to include body-centered cubic and face-centered cubic lattices. This is of particular interest since several of the recently discovered ferromagnetic insulators are face-centered cubic. The thermal resistivity produced by defect scattering is calculated by a method which includes some of the effects of spin-wave-spin-wave interactions.

The theory of the scattering of spin waves by magnetic defects is an application of the general theory of the scattering of excitations by imperfections in solids. An account of this theory is being published elsewhere.<sup>2</sup> Therefore, the present discussion will contain only as much of the theory as is required to make the application of those results to a spin-wave system intelligible.

We consider a spin system described by a simple Heisenberg exchange Hamiltonian with nearest-neighbor interaction only:

$$H = \sum_{i,\Delta} J(\mathbf{R}_i, \mathbf{R}_i + \Delta) \mathbf{S}(\mathbf{R}_i) \cdot \mathbf{S}(\mathbf{R}_i + \Delta). \quad (1)$$

Neither external fields nor dipole-dipole interactions are included. The system is said to contain a magnetic defect if at some site  $\mathbf{R}_0$  (which we choose as the origin), there is an atom whose spin  $S'$  is coupled to its neighbors by an exchange integral  $J'$ . For the remainder of the atoms, these quantities are  $S$  and  $J$ , respectively. The quantity  $\Delta$  is a vector connecting a lattice site with one of its nearest neighbors.

The excited states of the system which contain a single spin deviation can be described by a set of functions  $\phi(\mathbf{R})$  such that  $|\phi(\mathbf{R})|^2$  gives the probability of finding the spin deviation on site  $\mathbf{R}$ . It was shown in

Ref. 3 that  $\phi(\mathbf{R})$  satisfies the equation

$$E\phi(\mathbf{R}_i) = 2 \sum_{\Delta} J(\mathbf{R}_i, \mathbf{R}_i + \Delta) [S(\mathbf{R}_i + \Delta)\phi(\mathbf{R}_i) - \{S(\mathbf{R}_i)S(\mathbf{R}_i + \Delta)\}^{1/2}\phi(\mathbf{R}_i + \Delta)]. \quad (2)$$

In this equation  $S(\mathbf{R}_i)$  is the "magnitude" of the spin on the site  $\mathbf{R}_i$  in that the eigenvalue of  $S^2(\mathbf{R}_i)$  in the ground state is  $S(\mathbf{R}_i)[S(\mathbf{R}_i) + 1]$ . Equation (2) can be written as a Schrödinger equation in the form

$$\sum_i (m|E - H_0|l)\phi(\mathbf{R}_i) = \sum_i (m|V|l)\phi(\mathbf{R}_i), \quad (3)$$

in which

$$(m|E - H_0|l) = (E - 2JSz)\delta_{l,m} + 2JS\delta_{l-m,\Delta} \quad (4)$$

and

$$(m|V|l) = \delta_{lm} [2S_z(J' - J)\delta_{l,0} + 2(J'S' - JS)\delta_{l,\Delta}] - 2[J'(S'S)^{1/2} - JS]\delta_{l-m,\Delta}(\delta_{l,0} + \delta_{m,0}). \quad (5)$$

Each atom is assumed to have  $z$  nearest neighbors. The subscript  $l-m$  refers to  $\mathbf{R}_l - \mathbf{R}_m$ .

The solutions of this equation for energies,  $E$ , within the continuous spectrum of  $H_0$  have been shown to have the following asymptotic form:

$$\phi(\mathbf{R}_i) = \frac{\Omega^{1/2}}{(2\pi)^{3/2}} \left[ e^{i\mathbf{k}\cdot\mathbf{R}_i} + f \frac{e^{i\mathbf{k}\cdot\mathbf{R}_i}}{R_i} \right], \quad (6)$$

provided the energy is low enough so that the energy wave vector relation (energy band) for spin waves is spherical:

$$E = \gamma k^2, \quad (7)$$

$$\gamma = 2JSa^2, \quad (8)$$

in which  $a$  is the cubic lattice constant. The quantity  $f$  may be interpreted as the scattering amplitude for spin waves. It may be expressed as a sum of scattering amplitudes for partial waves transforming according to one of the irreducible representations of the crystal point group:

$$f = \sum_{\beta} f_{\beta}, \quad (9)$$

in which  $\beta$  denotes an irreducible representation. The partial-wave scattering amplitudes are

$$f_{\beta} = - \frac{2\pi^2}{\gamma D_{\beta}} \sum_{pnm} V_{\beta,pn} P_{\beta,nm} \sum_{\nu} C_{\beta\nu}^{(0)*}(k', R_p) \times C_{\beta\nu}^{(0)}(k_0, R_m). \quad (10)$$

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<sup>1</sup>J. Callaway, Phys. Rev. **132**, 2003 (1963). References to previous studies of the spin-wave-defect interaction are given there.

<sup>2</sup>J. Callaway, J. Math. Phys. **5**, 784 (1964).

<sup>3</sup>T. Wolfram and J. Callaway, Phys. Rev. **130**, 2207 (1963).

In this equation, the functions  $C^{(0)}(k, R)$  are symmetrized linear combinations of plane waves  $(\Omega/8\pi^3)^{1/2} \times \exp(i\mathbf{k} \cdot \mathbf{R})$  transforming according to the  $\nu$ th row of the  $\beta$ th irreducible representation. These functions are characterized by wave vectors  $\mathbf{k}_0$  and  $\mathbf{k}'$ , in which  $\mathbf{k}_0$  is the wave vector of the incident wave and  $\mathbf{k}'$  is a vector of magnitude  $k_0$  parallel to  $\mathbf{R}_l$  [see Eq. (6)]. We can write

$$C_{\beta, \nu}^{(0)}(k, R_m) = \frac{\Omega^{1/2}}{(2\pi)^{3/2}} \sum_{m'} U(\beta, \nu, \mathbf{R}_m) e^{i\mathbf{k} \cdot \mathbf{R}_m}, \quad (11)$$

in which the quantities  $U(\beta, \nu, \mathbf{R}_m)$  are the matrix elements of a unitary transformation. The prime on the summation in Eq. (11) indicates that the vectors  $\mathbf{R}_m$  which are included are those which can be found from any one of them by rotation with all the operators of the point group. (Only distinct vectors are considered.)

The quantities  $V_{\beta, \nu n}$  are given by

$$V_{\beta, \nu n} = \sum_{p, n'} U(\beta, \nu, \mathbf{R}_p) \langle p | V | n \rangle U^\dagger(\mathbf{R}_n, \beta, \nu), \quad (12)$$

in which  $U^\dagger$  is the adjoint of the matrix  $U$ .  $V_{\beta, \nu n}$  is independent of the row of the representation  $\nu$ . The remaining quantities are found as follows. We consider the Green's function,  $\mathcal{G}(\mathbf{R}_l - \mathbf{R}_n)$ , which satisfies

$$\sum_l \langle m | E - H_0 | l \rangle \mathcal{G}(\mathbf{R}_l - \mathbf{R}_n) = \delta_{mn}, \quad (13)$$

and define

$$\Gamma_{\beta, l n} = \sum_{l n} U(\beta, \nu, \mathbf{R}_l) \mathcal{G}(\mathbf{R}_l - \mathbf{R}_n) U^\dagger(\mathbf{R}_n, \beta, \nu). \quad (14)$$

Then, considering only those values of the indices  $l, m$ , such that  $V_{\beta, l m}$  is not zero, we construct the matrix

$$Q_{\beta, l m} = \delta_{lm} - \sum_n \Gamma_{\beta, l n} V_{\beta, n m}. \quad (15)$$

The summation in (10) and (15) includes one term for each different symmetrized combination of plane waves. The quantities  $P_\beta$  and  $D_\beta$  occurring in Eq. (9) may now be defined by writing the matrix inverse to  $Q$  as

$$[Q^{-1}]_{\beta, l m} = D_\beta^{-1} P_{\beta, l m}, \quad (16)$$

with

$$D_\beta = \det(Q_\beta) = \det[I - \Gamma_\beta V_\beta]. \quad (17)$$

The quantity  $D_\beta$  determines the locations of possible scattering resonances or localized modes. Let  $E'$  (in general, a complex number) be a solution of

$$D_\beta(E') = 0. \quad (18)$$

Put  $E' = E_R - i\Gamma/2$ . Then, if  $\Gamma > 0$ ,  $E_R$  is the energy of a scattering resonance, and  $\Gamma$  is the width of the resonance. If  $\Gamma = 0$  (which occurs only outside the continuum of eigenstates of  $H_0$ ), we have a localized mode.

In the case of spin waves, the perturbation  $V$  extends only to nearest neighbors of the defect. The

irreducible representations which occur in the sum over  $\beta$  in Eq. (7) have been determined for cubic lattices<sup>2,3</sup>: They are  $\Gamma_1, \Gamma_{15}$ , and  $\Gamma_{12}$  for a simple cubic lattice;  $\Gamma_1, \Gamma_{15}, \Gamma_{25'}$ , and  $\Gamma_{2'}$  for a body-centered cubic lattice, and  $\Gamma_1, \Gamma_{15}, \Gamma_{25'}$ ,  $\Gamma_{12}$ , and  $\Gamma_{25}$  for face-centered cubic.<sup>4</sup> The  $k$  dependence of the partial-wave scattering amplitude  $f_\beta$  at low energies can be studied by expanding the symmetrized combinations of plane waves  $C_{\beta, \nu}^{(0)}(k, R)$  in powers of  $k$ . One finds that

$$C_{\beta, \nu}^{(0)}(k, R) \propto (kR)^l K_{\beta, \nu, l}(\theta, \phi), \quad (19)$$

in which  $K_{\beta, \nu, l}$  is a Kubic harmonic belonging to the  $\nu$ th row of the  $\beta$ th irreducible representation, and is a linear combination of spherical harmonics of order  $l$ .<sup>5</sup> For  $\Gamma_1, l=0$ ; for  $\Gamma_{15}, l=1$ ; for  $\Gamma_{12}$  and  $\Gamma_{25'}$ ,  $l=2$ , and for  $\Gamma_{2'}$  and  $\Gamma_{25}$ ,  $l=3$ . We conclude that (since  $|\mathbf{k}_0| = |\mathbf{k}'|$ )

$$f_\beta \propto k^{2l}, \quad (20)$$

unless there are cancellations among the elements  $V, P$  which raise the power of  $k$  in (20). We do not expand  $D_\beta$  since this contains the possible resonances.

For spin waves we find that the  $p$ -wave amplitude ( $\Gamma_{15}$ ) is proportional to  $k^2$ , but cancellations do occur for the  $s$ -wave amplitude ( $\Gamma_1$ ), which then turns out also to be proportional to  $k^2$ . Other ( $d, f$ ) partial waves give terms proportional to  $k^4$  and  $k^6$ . Hence, the total amplitude is proportional to  $k^2$  for small  $k$ .

We will now determine the scattering amplitude to this order for simple cubic, body-centered cubic, and face-centered cubic lattices. The  $p$ -wave portion is simplest because only one term appears in the summation over  $p, m$ , and  $n$  in Eq. (8). From this fact it follows, with the use of Eq. (16), that the only scattering element of  $P$  is unity. Hence Eq. (10) has the form

$$f_p = (-2\pi^2/\gamma D_p) V_{p, 11} \sum_\nu C_{\nu, \nu}^{(0)*}(k', R_1) \times C_{\nu, \nu}^{(0)}(k_0, R_1). \quad (21)$$

The rows of the triply degenerate  $\Gamma_{15}$  representation contain functions transforming as  $x, y$ , or  $z$ . In each of the three lattices we have in the small  $k$  limit

$$C_{p_x} = [\Omega^{1/2}/(2\pi)^{3/2}] \sqrt{2} i k_x a, \quad (22)$$

etc., and

$$V_{p, 11} = -2JS[1 - (J'S'/JS)]. \quad (23)$$

Then

$$f_p = (\Omega/2\pi D_p a^2) [1 - (J'S'/JS)] k^2 a^2 \cos\theta, \quad (24)$$

in which  $\theta$  is the angle between  $k$  and  $k'$ .

It is now necessary to express the determinant  $D_p$  in terms of the Green's functions. If  $(\xi, \eta, \zeta)$  are the rectangular components of  $(\mathbf{R}_l - \mathbf{R}_n)/a$ , we have for

<sup>4</sup> Notation for the irreducible representations of the point group is in accord with L. P. Bouckaert, R. Smoluchowski, and E. Wigner, Phys. Rev. **50**, 58 (1936).

<sup>5</sup> F. C. Von der Lage and H. A. Bethe, Phys. Rev. **71**, 612 (1947).

the three lattices:

Simple cubic,

$$D_p = 1 - \frac{1}{2} [1 - (J'S'/JS)] [\mathcal{G}(\mathbf{0}) - \mathcal{G}(2,0,0)]; \quad (25)$$

body-centered cubic,

$$D_p = 1 - \frac{1}{2} [1 - (J'S'/JS)] \times [\mathcal{G}(\mathbf{0}) + \mathcal{G}(1,0,0) - \mathcal{G}(1,1,0) - \mathcal{G}(1,1,1)]; \quad (26)$$

face-centered cubic;

$$D_p = 1 - \frac{1}{2} [1 - (J'S'/JS)] [\mathcal{G}(\mathbf{0}) + 2\mathcal{G}(\frac{1}{2}, \frac{1}{2}, 0) - 2\mathcal{G}(1, \frac{1}{2}, \frac{1}{2}) - \mathcal{G}(1,1,0)]. \quad (27)$$

We infer from these results that for the same value of the lattice constant  $a$ , the scattering amplitude depends on the lattice structure partly through the cell volume  $\Omega$  which has the value  $a^3$ ,  $a^3/2$ , and  $a^3/4$  in the simple body-centered, and face-centered cubic lattices, respectively. In addition, more complicated dependence on structure is contained in the determinants,  $D_p$ .

We now turn to a consideration of the  $s$ -wave amplitude. This calculation is quite tedious and will not be given in detail. For each lattice, the relevant quantities  $V, P$  are  $2 \times 2$  matrices. The symmetrized linear combinations of plane waves are given by

$$C_s^{(0)}(k, 0) = \Omega^{1/2} (2\pi)^{-3/2}, \quad (28a)$$

$$C_s(k, R_1) = (\Omega/z)^{1/2} (2\pi)^{-3/2} \sum_{\Delta} \exp(i\mathbf{k} \cdot \Delta). \quad (28b)$$

Equation (28b) can be expressed in terms of the spin-wave energy  $E(k)$ , since

$$E(k) = 2JS [z - \sum_{\Delta} \exp(i\mathbf{k} \cdot \Delta)].$$

Hence,

$$C_s^{(0)}(k, R_1) = (\Omega/z)^{1/2} (2\pi)^{-3/2} (z - E/2JS). \quad (29)$$

Equation (29) enables us to eliminate the functions  $C_s^{(0)}$  from the  $s$ -wave amplitude. In addition, it is possible to express all the combinations of Green's functions which enter into  $D_s$  and  $P_s$  in terms of  $\mathcal{G}(\mathbf{0})$ , with the aid of certain identities which were derived in Ref. 3. Since  $\mathcal{G}(\mathbf{0})$  can be expressed in terms of the density of states  $G(E)$ ,

$$\begin{aligned} \mathcal{G}(\mathbf{0}) &= \frac{\Omega}{(2\pi)^3} \int \frac{d^3q}{E - E(q)} = P \int \frac{G(E') dE'}{E - E'} - i\pi G(E) \\ &\equiv -g_0/4JS, \quad (30) \end{aligned}$$

this means that the  $s$ -wave amplitude can be determined from the  $G(E)$  without explicit reference to  $E(k)$ . The relevant identities are

$$\begin{aligned} z^{-1} \sum_i \mathcal{G}(\Delta_i) &= [1 - (E/2JSz)] \mathcal{G}(\mathbf{0}) + (1/2JSz) \\ &\equiv -g_1/4JS, \quad (31) \end{aligned}$$

$$\begin{aligned} \sum_j \mathcal{G}(\Delta_i - \Delta_j) &= [1 - (E/2JSz)] \sum_i \mathcal{G}(\Delta_i) \\ &\equiv -g_2/4JS. \quad (32) \end{aligned}$$

In these equations,  $\Delta_i$  and  $\Delta_j$  indicate sites which are nearest neighbors of the defect site (the origin). Only the particular combinations of Green's functions appearing in (30), (31), and (32) are involved in the  $s$ -wave amplitude.

In addition, we have

$$V_s = -4JS \begin{pmatrix} \epsilon & \eta\sqrt{z} \\ \eta\sqrt{z} & \rho \end{pmatrix}, \quad (33)$$

$$P_s = \begin{pmatrix} 1 - g_2\rho - z\eta g_1 & z^{1/2}(g_0\eta + g_1\rho) \\ z^{1/2}(g_1\epsilon + g_2\eta) & 1 - g_0\epsilon - z\eta g_1 \end{pmatrix}, \quad (34)$$

in which

$$\epsilon = \frac{1}{2}z(1 - J'/J), \quad (35a)$$

$$\eta = \frac{1}{2}[(J'/J)(S'/S)^{1/2} - 1], \quad (35b)$$

$$\rho = \frac{1}{2}(1 - J'S'/JS). \quad (35c)$$

After a rather tedious calculation, we obtain for the  $s$ -wave portion of the scattering amplitude to lowest order in  $k$

$$\begin{aligned} f_s &= \frac{-\Omega}{2\pi D_s a^2} \frac{E}{4JS} \left( \frac{J'}{J} \right) \left( 1 - \frac{S'}{S} \right) \\ &= \frac{-\Omega}{4\pi D_s a^2} \left( \frac{J'}{J} \right) \left( 1 - \frac{S'}{S} \right) k^2 a^2, \quad (36) \end{aligned}$$

in which

$$D_s = 1 + \frac{z}{2}(g_1 - g_0) \left( 1 - \frac{J'}{J} \right) + \frac{Eg_1}{4JS} \left( 1 - \frac{J'S'}{JS} \right). \quad (37)$$

It will be observed that  $f_s$  goes to zero as  $k^2$  at low energies. This is the result of cancellation among some of the terms and of the Green's function identities (32). The expression (37) for  $D_s$  is exact. We do not expand  $D_s$  (or  $D_p$ ) since they contain the possible low-energy resonances.

The total amplitude to order  $k^2$  is the sum of (36) and (24):

$$\begin{aligned} f = f_s + f_p &= \frac{-\Omega}{4\pi a^2} \left[ \frac{1}{D_s} \left( \frac{J'}{J} \right) \left( 1 - \frac{S'}{S} \right) \right. \\ &\quad \left. - \frac{2}{D_p} \left( 1 - \frac{J'S'}{JS} \right) \cos d \right] k^2 a^2. \quad (38) \end{aligned}$$

It will be observed that the amplitude in this order depends on the lattice structure in an obvious manner through the cell volume. There is, however, an equally important structure dependence incorporated in the function  $D_s$  and  $D_p$ . The higher order (in  $k$ ) terms in the amplitude also exhibit a significant dependence on the lattice structure since different representations appear. In general, it does not appear to be possible to express the scattering amplitude solely in terms of the

angle between the incoming and outgoing wave vectors when higher order terms are included.

When the scattering amplitude is considered only to lowest order in  $k$ , so that it depends only on the angle  $\theta$  a simple calculation of the mean free path  $l_D$ , for scattering of a spin wave by a defect is possible. This has been shown to be determined by the momentum transfer cross section  $\sigma_n$  which is given by

$$\sigma_n = \int |f(\theta)|^2 (1 - \cos\theta) d\Omega. \quad (39)$$

Then<sup>6</sup>

$$l_D^{-1} = N_D \sigma_n, \quad (40)$$

where  $N_D$  is the concentration of defects. We obtain

$$\begin{aligned} \sigma_n = \frac{\Omega^2 k^4}{4\pi} & \left\{ \frac{1}{|D_s|^2} \left( \frac{J'}{J} \right)^2 \left( 1 - \frac{S'}{S} \right)^2 \right. \\ & + \frac{4}{3} \left( 1 - \frac{J'S'}{JS} \right) \left[ \frac{1}{|D_p|^2} \left( 1 - \frac{J'S'}{JS} \right) \right. \\ & \left. \left. + \frac{1}{2} \left( \frac{J'}{J} \right) \left( 1 - \frac{S'}{S} \right) \left( \frac{1}{D_s^* D_p} + \frac{1}{D_s D_p^*} \right) \right] \right\}. \quad (41) \end{aligned}$$

If we suppose that the denominators  $D_s$  and  $D_p$  can be set equal to unity, thereby neglecting the possibility of resonant scattering, we obtain

$$\begin{aligned} \sigma_n = \frac{\Omega^2 k^4}{4\pi} & \left[ \left( \frac{J'}{J} \right)^2 \left( 1 - \frac{S'}{S} \right)^2 \right. \\ & \left. + \frac{4}{3} \left( 1 - \frac{J'S'}{JS} \right) \left( 1 + \frac{J'}{J} - \frac{2J'S'}{JS} \right) \right]. \quad (42) \end{aligned}$$

## II. THERMAL CONDUCTIVITY

In this section we consider the thermal conductivity of a system of spin waves in interaction with each other and containing  $N_D$  magnetic defects per unit volume. We hope to consider spin-wave-phonon interactions in subsequent work. Unless explicitly stated, the system will be assumed to be of infinite extent.

We propose here to calculate the thermal conductivity of spin waves by the same techniques which have proved to be reasonably successful in application to lattice thermal conductivity.<sup>7</sup> We will consider each scattering process to be described by a relaxation time, but explicit account will be taken of the conservation of crystal momentum by normal spin-wave-spin-wave scattering processes.

The distinction between normal and umklapp processes in spin-wave interactions is just as vital as it is

in the case of phonon-phonon interactions. Since normal processes conserve the total pseudomomentum of the spin-wave system, they cannot, if acting alone, produce a thermal resistance. This statement is essentially independent of the details of the dispersion relation for the magnons. It is true that the interactions between magnons do not conserve the heat flow at the microscopic level (as is the case for phonons if a linear dispersion law is assumed). A statistical argument shows, however, that a thermal resistance will not appear. An explicit demonstration is given in the Appendix.

Nevertheless, it is not permissible to neglect normal processes in studying the thermal conductivity. Their contribution may be pictured qualitatively as that of converting some of the low-frequency magnons into high-frequency magnons which may easily be scattered by defects. We will show that in the limit of strong normal processes, the thermal conductivity becomes limited by the defect scattering in a fashion similar to that obtained by Ziman<sup>8</sup> for lattice thermal conductivity.

The importance of distinguishing normal and umklapp processes has been stressed here because it has been ignored in other calculations of the spin-wave thermal conductivity.<sup>9</sup> We contend that such work may have yielded an erroneous thermal resistance.

We begin by considering the Boltzmann equation in the form

$$\left( \frac{\partial N}{\partial t} \right)_c - \mathbf{V}_k \cdot \nabla T \frac{dN}{dT} = 0, \quad (43)$$

in which  $\nabla T$  is the temperature gradient,  $N$  is the distribution function, and  $\mathbf{V}_k$  is the group velocity of the spin waves

$$\mathbf{V}_k = \hbar^{-1} \nabla_k E(\mathbf{k}). \quad (44)$$

The first assumption of the present approach is that the collision term  $(\partial N / \partial t)_c$  may be approximated as follows:

$$\left( \frac{\partial N}{\partial t} \right)_c = \frac{N(\lambda) - N}{\tau_n} + \frac{N_0 - N}{\tau_u}, \quad (45)$$

in which  $N(\lambda)$  is a displaced Bose distribution

$$N(\lambda) = \{ \exp[(E - \lambda \cdot \mathbf{k}) / KT] - 1 \}^{-1}. \quad (46)$$

$N_0$  is the usual Bose function;  $\tau_n$  is the relaxation time for a single mode via normal processes, and  $\tau_u$  is the relaxation time for all those processes which do not conserve the crystal momentum.<sup>10</sup> We assume that  $\lambda$  is small so that  $N(\lambda)$  may be expanded, and only first-

<sup>8</sup> J. M. Ziman, *Can. J. Phys.* **34**, 1256 (1956).

<sup>9</sup> A. Quattropani, *Phys. Kondens. Materie* **1**, 125 (1963).

<sup>6</sup> J. M. Ziman, *Electrons and Phonons* (Oxford University Press, Oxford, 1960).

<sup>7</sup> J. Callaway, *Phys. Rev.* **113**, 1046 (1959); J. Callaway and H. C. Von Baeyer, *ibid.* **120**, 1149 (1960).

<sup>10</sup> R. E. Nettleton, *Phys. Rev.* **132**, 2032 (1963), has partially justified this approximation for lattice thermal conductivity. In applying Nettleton's discussion to the present calculation, one should note that in spite of appearances, an explicit expression for the normal process relaxation time,  $\tau_N$ , is not required to obtain our essential results, Eqs. (60) and (67). The quantities which are required are  $\tau_D$  and  $\tau_c$ .

order terms in  $\lambda$  are retained. Put

$$\lambda = -S\nabla T/T, \quad (s \text{ is a constant}) \quad (47a)$$

$$N - N_0 = n_1 = -\nu \mathbf{V}_k \cdot \nabla T \left( \frac{E}{KT} \right)^2 \frac{e^{E/KT}}{(e^{E/KT} - 1)^2}, \quad (47b)$$

$$\tau_c^{-1} = \tau_u^{-1} + \tau_n^{-1}. \quad (47c)$$

(These equations are definitions of  $s$ ,  $\nu$ , and  $\tau_c$ .) We also require the group velocity. We continue to use a quadratic dispersion law

$$E = \gamma k^2 + \epsilon, \quad (48)$$

so that

$$\mathbf{V}_k = 2(\gamma/\hbar)\mathbf{k}. \quad (49)$$

The constant term  $\epsilon$  in (48) does not play an important role in the mechanics of the thermal conductivity calculation and has been ignored in computing scattering amplitudes, but it is important in preventing the quantity  $E - \lambda \cdot \mathbf{k}$  in (46) from becoming negative for low energies. It is therefore important to realize that a nonzero value of  $\epsilon$  is always obtained, since the material is magnetized. One has, approximately,<sup>11</sup>

$$\epsilon = g\beta[(4\pi/3)M + H_{\text{ext}}], \quad (50)$$

where  $M$  is the magnetization and  $H_{\text{ext}}$  is a possible external field.

Equations (45)–(49) are substituted in the Boltzmann equation, which then gives a relation between  $\nu$  and  $s$ . This is

$$\nu = \tau_c [1 + \hbar s / (2\gamma \tau_N E)]. \quad (51)$$

The thermal conductivity of the system is

$$\begin{aligned} \kappa &= [1/(2\pi)^3] \int \mathbf{V}_k^2 \nu(k) \cos^2\theta C_{sw}(k) d^3k, \\ &= (2\gamma^2/3\pi^2\hbar) \int k^4 \nu(k) C_{sw}(k) dk, \\ &= (2\gamma^2/3\pi^2\hbar^2) \int k^4 \tau_c [1 + S\hbar/(2\gamma\tau_n E)] C_{sw}(k) dk. \end{aligned} \quad (52)$$

We have assumed cubic symmetry and also employed (49).  $C_{sw}(k)$  is the contribution to the specific heat from a mode of wave vector  $k$ . Evidently another relation between  $\nu$  and  $s$  is required. This is obtained from the requirement that the normal processes do not change the total wave vector of the system:

$$\int \left( \frac{\partial N}{\partial t} \right)_N \mathbf{k} d^3k = \int \frac{N(\lambda) - N}{\tau_N} \mathbf{k} d^3k = 0. \quad (53)$$

This condition leads to the relation

$$\int \frac{\mathbf{k}}{\tau_N} (\mathbf{k} \cdot \nabla T) \left[ s - \frac{2\gamma\nu E}{\hbar} \right] \frac{e^{E/KT}}{(e^{E/KT} - 1)^2} d^3k = 0. \quad (54)$$

Only the component of  $\mathbf{k}$  parallel to the temperature gradient survives on integration over solid angles. We substitute (51) and solve for  $s$

$$s = \frac{2\gamma}{\hbar} \int \frac{\tau_c}{\tau_N} \frac{Ek^4 e^{E/KT}}{(e^{E/KT} - 1)^2} dk / \left[ \int \frac{1}{\tau_N} \left( 1 - \frac{\tau_c}{\tau_N} \right) \frac{k^4 e^{E/KT}}{(e^{E/KT} - 1)^2} dk \right]. \quad (55)$$

We combine (55) and (53), substitute for the spin-wave specific heat

$$C_{sw} = K \left( \frac{E}{KT} \right)^2 \frac{e^{E/KT}}{(e^{E/KT} - 1)^2},$$

introduce the change of variables

$$x = \gamma k^2 / KT, \quad \delta = \epsilon / KT, \quad (56)$$

replace the relaxation time by mean free paths,

$$l_c = V_k \tau_c,$$

etc., and obtain our final expression for the thermal conductivity:

$$\begin{aligned} \kappa &= \frac{K(KT)^2}{6\pi^2\gamma\hbar} \left\{ \int_0^{x_m} x(x+\delta)^2 l_c \frac{e^{x+\delta}}{(e^{x+\delta} - 1)^2} dx \right. \\ &\quad + \left[ \int_0^{x_m} x^{3/2} \frac{l_c}{l_N} \frac{(x+\delta)e^{x+\delta}}{(e^{x+\delta} - 1)^2} dx \right]^2 / \\ &\quad \left. \left[ \int_0^{x_m} \frac{l_c}{l_n l_u} \frac{x^2 e^{x+\delta}}{(e^{x+\delta} - 1)^2} dx \right] \right\}. \quad (57) \end{aligned}$$

The upper limit  $x_m$  for these integrals is determined by an argument analogous to that establishing the Debye temperature for a crystal lattice and is given by  $x_m = (\gamma/KT)(6\pi^2/\Omega)^{2/3}$ . We will consider here only temperatures low enough so that the upper limit can be made infinite.

The dependence of the thermal conductivity on an external magnetic field can be obtained from (57). A magnetic field decreases the thermal conductivity by reducing the probability of excitation of spin waves. It is evident from (57) that in the limit of large  $\delta$  (low temperatures or strong fields), the thermal conductivity must decrease as  $\delta^n e^{-\delta}$ ; here the exponent  $n$  will depend on the scattering mechanism. Further discussion of the field dependence of the conductivity will be reserved for subsequent work.

<sup>11</sup> T. Holstein and Primakoff, Phys. Rev. **58**, 1098 (1940); S. H. Charap and E. Boyd (to be published). This dispersion relation is only approximate since in the presence of dipole-dipole couplings, the spin-wave energies are anisotropic.

There is a great similarity between Eq. (57) and the corresponding expressions which are obtained in the theory of lattice thermal conductivity. In particular, we observe that if the relaxation time for processes which do not conserve the wave vector becomes very long, the thermal conductivity of the spin-wave system increases without limit. If only normal processes were present, there would be no thermal resistance.

It follows that in the limit in which the mean free path for normal processes is very short compared to that for processes which do not conserve the wave vector, the thermal conductivity will depend sensitively on the latter processes. This is the Ziman limit. To obtain the thermal resistance in this limit, we drop the first term of (57) and neglect the contribution of  $l_u$  to  $l_c$ . Then

$$W = \frac{1}{\kappa} = \frac{6\pi^2\gamma\hbar}{K(KT)^2} \int \frac{x^2}{l_u} \frac{e^{x+\delta}}{(e^{x+\delta}-1)^2} dx \Big/ \left[ \int x^{3/2} \frac{(x+\delta)e^{x+\delta}}{(e^{x+\delta}-1)^2} dx \right]^2. \quad (58)$$

One interesting consequence of (58) is that the thermal resistance in the Ziman limit is the sum of additive contributions from each possible type of "U" process. In particular we consider defect scattering, for which the mean free path is given by Eqs. (40) and (41). We will not consider here the effects of possible scattering resonances which could increase the thermal resistance substantially. We abbreviate the mean free path due to defect scattering as

$$l_D^{-1} = N_D A k^4, \quad (59a)$$

in which

$$A = \frac{\Omega^2}{4\pi^2} \left[ \left( \frac{J'}{J} \right)^2 \left( 1 - \frac{S'}{S} \right)^2 + \frac{4}{3} \left( 1 + \frac{J'}{J} - 2 \frac{J'S'}{JS} \right) \right]. \quad (59b)$$

We can evaluate the thermal resistance due to defects approximately by neglecting  $\delta$  in (58). We get

$$W_D = \frac{6\pi^2\hbar N_D A}{K\gamma} \int_0^\infty \frac{x^4 e^x}{(e^x-1)^2} dx \Big/ \left[ \int_0^\infty \frac{x^{5/2} e^x}{(e^x-1)^2} dx \right]^2 = \frac{6\pi^2 N_D A \hbar}{K\gamma} \frac{\Gamma(5)\zeta(4)}{[\Gamma(7/2)\zeta(5/2)]^2} = 38.6 \frac{N_D A \hbar}{JSK a^2}. \quad (60)$$

In the last step of (60), we have substituted Eq. (8).

The thermal resistance due to the defects is independent of temperature. This expression is valid in the limit in which defect scattering is weak compared to normal magnon-magnon scattering. Our treatment has included the effects of magnon-magnon scattering on the distribution function, and thereby has taken account of these interactions implicitly. Our formula, Eq. (60), does not depend explicitly on the details of the magnon-

magnon interaction. It does differ somewhat from the expression for the defect resistivity if these interactions are neglected. The situation in which defect scattering is weak compared to boundary scattering and normal processes are ignored was examined in I. In that case a thermal resistance  $W_D'$  due to defects was obtained which is independent of temperature, and differs from (60) only in respect to the numerical constant. That result is<sup>12</sup>

$$W_D' = 20\pi^2 \frac{N_D A \hbar}{K\gamma} \frac{\zeta(5)}{[\zeta(3)]^2} = 70.8 \frac{N_D A \hbar}{JSK a^2}. \quad (61)$$

It will be noted that the thermal resistance due to defects given in (61) is larger by a factor of almost 2 than that of Eq. (60). This situation should be contrasted with that which obtains in the theory of lattice thermal conductivity where the contribution to the thermal resistance from defects is, in the Ziman limit, larger by a factor of 25 than when umklapp scattering dominates.

Next, we will discuss the thermal conductivity in the opposite limit in which the defect scattering is strong compared to magnon-magnon scattering. This requires a more detailed knowledge of the combined mean free path  $l_c$ , including normal processes. The interaction of two spin waves can produce bound states,<sup>13</sup> and probably scattering resonances. A complete theory of the scattering of two spin waves has not yet been given, but for an introduction, we can make use of a calculation reported by Dyson.<sup>14</sup> Dyson showed that, to the extent that resonances and such may be neglected, the mean free path for a spin wave of wave vector  $k$  due to spin-wave-spin-wave interactions, may be written as

$$l = BT^{5/2} k^2. \quad (62)$$

This expression is valid at low temperatures. The quantity  $B$  is a rather complicated function of  $J$  and  $S$ , and is given by

$$B = \frac{3\zeta(3/2)a}{z\omega S^2} \left( \frac{3K}{2\pi JSz\mu} \right)^{5/2}, \quad (63)$$

in which

$$\omega = (1, 3 \times 2^{-5/3}, 2^{-1/3}), \quad (64)$$

$$\mu = (1, 3 \times 2^{-4/3}, 2^{1/3}),$$

for the simple cubic, body-centered cubic, and face-centered cubic lattices, respectively.

With the use of (62) and (59), we have a combined mean free path including defect scattering and magnon-magnon scattering

$$l_c^{-1} = BT^{5/2} k^2 + N_D A k^4. \quad (65)$$

<sup>12</sup> A factor of  $(K\zeta(3))^{-1}$  was omitted in the statement of this equation in Ref. 1.

<sup>13</sup> M. Wortis, Phys. Rev. **132**, 85 (1963); J. Hanus, Phys. Rev. Letters **11**, 336 (1963).

<sup>14</sup> F. J. Dyson, Phys. Rev. **102**, 1217 (1956).

In the limit  $l_c \ll 1$  which corresponds to defect scattering strong compared to magnon-magnon scattering, we can drop all but the first term of (57). This follows since if we consider both  $l_c$  and  $l_u$  to be proportional to some small constant  $\alpha$ , the first term of (57) is proportional to  $\alpha$ , whereas the second term is proportional to  $\alpha^2$ . We then substitute (65) into (57), let  $\delta \rightarrow 0$ , and obtain

$$\kappa = \frac{K\gamma}{6\pi^2\hbar N_D A} \int_0^{x_m} \frac{x^2}{x+y} \frac{e^x}{(e^x-1)^2} dx, \quad (66)$$

in which  $y = B\gamma T^{3/2}/N_D A K$ .

We consider this integral at temperatures low enough so that the upper limit may be made infinite. Even so, the integral needs to be approximated if we are to avoid numerical computation. We follow the procedure used under somewhat similar circumstances in I, in which we replace  $x^2 e^x / (e^x - 1)^2$  by 1 for  $x < 1$ , and neglect  $y$  for  $x > 1$ . This is valid only when  $y \ll 1$ , but if this condition is not satisfied, it is not legitimate to neglect the second term in (57). Hence

$$\begin{aligned} \kappa &= \frac{K\gamma}{6\pi^2\hbar N_D A} \left[ \int_0^1 \frac{dx}{x+y} + \int_1^\infty \frac{x e^x}{(e^x-1)^2} dx \right] \\ &= \frac{K\gamma}{6\pi^2\hbar N_D A} \left[ \ln\left(1 + \frac{1}{y}\right) + \sum_{n=1}^\infty e^{-n} \left(1 + \frac{1}{n}\right) \right] \\ &= \frac{KJSa^2}{3\pi^2 N_D A \hbar} \left[ \ln\left(1 + \frac{N_D A K}{B\gamma T^{3/2}}\right) + 1.0407 \right]. \quad (67) \end{aligned}$$

Evidently, in this limit, the temperature dependence of the thermal conductivity is weak.

#### APPENDIX

In this appendix, we give an explicit demonstration that for magnons whose energy is related to the wave vector by Eq. (48), normal scattering processes (N. P.) cannot, by themselves, produce a thermal resistance. We recall that a normal process is one in which the total wave vector of the excitations involved is conserved. It is to be distinguished from an umklapp process, for instance, in which the wave vector changes by a reciprocal lattice vector.

It is fairly easy to see intuitively that if only N. P. operate, the crystal relaxes to equilibrium subject to the condition that the total crystal wave vector be nonzero and constant  $\sum_{\mathbf{k}} \mathbf{k} n_{\mathbf{k}} = \mathbf{K}$  (in which  $n_{\mathbf{k}}$  is the number of particles in a state of wave vector  $\mathbf{k}$ ). There must be a flow of heat in the direction of  $\mathbf{K}$ .

The simplest mathematical justification of this intuition can be found by returning to the derivation of the Fermi-Dirac (F. D.) and Bose-Einstein (B. E.) distribution functions for an ideal gas using the micro-canonical ensemble. A system of weakly interacting excitations is considered.

To obtain the distribution function  $n_{\mathbf{k}}$  we maximize the entropy,  $\sigma$ , subject to the constraints imposed by conservation laws.<sup>15</sup> We do this by multiplying those quantities  $g_j(n_{\mathbf{k}})$  which must be conserved by Lagrange undetermined multipliers  $\lambda_j$ . Then we add their variation with respect to  $n_{\mathbf{k}}$  to the variation of  $\sigma$  and set the whole equal to zero. Thus,

$$\frac{\delta\sigma}{\delta n_{\mathbf{k}}} + \sum \lambda_j \frac{\delta g_j}{\delta n_{\mathbf{k}}} = 0. \quad (A1)$$

Let the upper/lower signs represent B. E./F. D. statistics, respectively. We have

$$\delta\sigma/\delta n_{\mathbf{k}} = \ln(n_{\mathbf{k}}^{-1} \pm 1). \quad (A2)$$

Let us choose the following conserved quantities:

$$\sum_{\mathbf{k}} n_{\mathbf{k}} = N; \quad \sum_{\mathbf{k}} E_{\mathbf{k}} n_{\mathbf{k}} = E; \quad \sum_{\mathbf{k}} \mathbf{k} n_{\mathbf{k}} = \mathbf{K}. \quad (A3)$$

We vary these quantities, multiply by Lagrange multipliers, and substitute together with (2) in (1) to obtain

$$n_{\mathbf{k}} = \left[ \exp\left(\frac{E_{\mathbf{k}} - \mu - \boldsymbol{\lambda} \cdot \mathbf{k}}{KT}\right) \mp 1 \right]^{-1}, \quad (A4)$$

where the multipliers of  $N$ ,  $E$ , and  $\mathbf{K}$  have been written in the conventional manner. (If the number of particles is not conserved, we should set  $\mu = 0$ , and this must be done for magnons and phonons.)

Clearly, since  $E_{\mathbf{k}} = E_{-\mathbf{k}}$ , modes with  $\mathbf{k}$  parallel to  $\boldsymbol{\lambda}$  contain more particles than modes with  $\mathbf{k}$  antiparallel, so we expect a net heat current  $Q$  in the direction of  $\boldsymbol{\lambda}$ .

To demonstrate this we consider the expression for the heat current

$$\mathbf{Q} = \sum_{\mathbf{k}} E_{\mathbf{k}} V_{\mathbf{k}} n_{\mathbf{k}}. \quad (A5)$$

We will evaluate this approximately on the basis of the dispersion relation<sup>16</sup>

$$E_{\mathbf{k}} = \gamma k^n + \epsilon. \quad (A6)$$

In this equation the relevant values of the exponent  $n$  are 1 and 2. If  $n = 1$ , we have a situation approximately describing low-energy phonons (also  $\epsilon = 0$  in this case). The case  $n = 2$  characterizes spin waves, but now  $\epsilon \neq 0$ ; a nonzero value of  $E$  is provided by the magnetization ( $M$ ) plus any external field ( $H_{\text{ext}}$ ), as was described in the main text [see Eq. (50)]. With the use of (6), we obtain for  $Q$

$$\mathbf{Q} = \frac{n\gamma}{(2\pi)^3 \hbar} \int \frac{(\gamma k^n + \epsilon) \mathbf{k} k^{n-2}}{\{\exp\{[\gamma k^n + \epsilon - \boldsymbol{\lambda} \cdot \mathbf{k}]/KT\} - 1\}} d^3 k. \quad (A7)$$

<sup>15</sup> K. Huang, *Statistical Mechanics* (John Wiley & Sons, Inc., New York, 1963), p. 192.

<sup>16</sup> The importance of including  $\epsilon$  is that if it is not present, the argument of the exponential in (A4) will become negative for small  $k$  when  $n = 2$ .

The total wave vector  $K$  is

$$\mathbf{K} = \frac{1}{(2\pi)^3} \int \frac{\mathbf{k}}{\{\exp\{[\gamma k^n + \epsilon - \boldsymbol{\lambda} \cdot \mathbf{k}]/KT\} - 1\}} d^3k. \quad (\text{A8})$$

Neither of these integrals is zero. If  $n=1$ , ( $\epsilon=0$ ) it is easy to see that

$$\mathbf{Q} = (\gamma/\hbar)\mathbf{K}. \quad (\text{A9})$$

For  $n=2$ , ( $\epsilon \neq 0$ ) the situation is somewhat more complex. The substitution  $\mathbf{q} = \mathbf{k} - \boldsymbol{\lambda}/2\gamma$  enables the integrals to be transformed to a form in which the portions which vanish on integration over angle can be readily separated. The results are

$$\mathbf{Q} = \frac{\boldsymbol{\lambda}}{4\pi^2\hbar} \left(\frac{KT}{\gamma}\right)^{3/2} \left[ \frac{5KT}{3} I(x_0, \frac{3}{2}) + \left(\epsilon + \frac{\lambda^2}{4\gamma}\right) I(x_0, \frac{1}{2}) \right], \quad (\text{A10a})$$

$$\mathbf{K} = \frac{\boldsymbol{\lambda}}{8\pi^2\gamma} \left(\frac{KT}{\gamma}\right)^{3/2} I(x_0, 1/2), \quad (\text{A10b})$$

in which

$$I(x_0, \nu) = \Gamma(\nu+1) \sum_{n=1}^{\infty} \frac{e^{-nx_0}}{n^{\nu+1}} \quad (\text{A11})$$

and  $KTx_0 = \epsilon - \lambda^2/4$ .

Equation (A10b) may be used to eliminate  $\boldsymbol{\lambda}$  from the expression for  $\mathbf{Q}$ . In the limit in which  $\epsilon$  and  $\lambda$  are both small, we obtain a simple result

$$\mathbf{Q} = C(KT/\hbar)\gamma\mathbf{K}, \quad (\text{A12})$$

in which  $C$  is a numerical constant.

We have shown that in the absence of processes which do not conserve the total wave vector, there is a constant nonzero heat current in the presence of nonzero total crystal wave vector. A state with a nonvanishing  $\mathbf{K}$ , once established, could not be changed by collisions. Since no thermal gradient was assumed, this means that in the absence of umklapp processes there is no thermal resistivity. There is no conflict with the result of the analogous classical problem of the thermal conductivity of gas in a cylinder, since in that case a state of nonzero momentum is not established.