

# Maximum Variational Principle for Conduction Problems in a Magnetic Field, and the Theory of Magnon Drag

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A maximum variational principle is shown to be applicable for conduction problems (including "drag" effects) in the presence of a magnetic field. The part of the diagonal tensor element that is even in the magnetic field is maximized. The relation between the "high-field" work of Chambers, Lifshitz, and co-workers and the variational method is pointed out, the latter being applied to accommodate open orbit effects, and to obtain interpolation formulas to span high- and low-field solutions (keeping in view the phonon drag effects). It was found that standard operator expansion techniques are useful for obtaining solutions for the high-field limit. Symmetry considerations are facilitated by use of some of the operators suggested by the variational problem, and it is shown that if no drag effects are present and a time of relaxation permitted, some new relations emerge for the cross coefficients connecting the charge flow and temperature gradient. Finally, because the scattering by spin waves is analogous to the scattering of phonons, (since double-magnon processes are shown to be negligible at low temperatures), the theory of "magnon-drag" follows precisely that of phonon drag, and the effects can be automatically incorporated in all the expressions.

## 1. INTRODUCTION

THE main effort of this paper is to produce a maximum variational principle that applies to conduction problems in a magnetic field.

The work of Ziman,<sup>1,2</sup> Garcia-Moliner and Simons,<sup>3</sup> Tsuji,<sup>4</sup> and Bross,<sup>5</sup> and others has indicated some intrinsic difficulty that prevented a true maximum principle when in the presence of a magnetic field, as for example holds for the electrical conductivity in the absence of a magnetic field.<sup>6</sup> This difficulty has been related by Ziman to a zero contribution to a kind of entropy production. The definition of entropy in the problem was, however, adjusted by Ziman and he was able to arrive at an extremal principle; but still a maximum principle remained elusive. It is not clear to what extent the entropy interpretation is significant; nevertheless the difficulty is a real one, since it is reflected in the mathematical theorems that can be proved. Tsuji, on the other hand, has provided a maximum principle, but what is maximized does not involve the magnetic field directly, and the principle has the curious feature that the Boltzmann equation's exact solution appears in the side condition, this solution kept unaltered during variations, and then (after the variations are taken) set equal to the trial function and solved for. Tsuji's resulting equations and Ziman's are the same (and equal to Kohler's<sup>7</sup> for spherical energy surfaces), and we feel that the setting equal of the exact

solution to the variational trial function after variations are taken in Tsuji's work, is essentially an assumption about convergence that is equivalent to the non-maximum nature of the Ziman formulation of the problem. (Tauber's<sup>8</sup> work resembles Tsuji's in this respect.) Thus it has appeared as if a conspiracy exists against a true maximum principle in the presence of a magnetic field.

Our result is that a maximum principle *does* hold in the presence of the magnetic field, but what is maximized is the part of the diagonal conductivity tensor element that is even in the magnetic field. We should like to acknowledge a conversation with Professor T. Holstein in which he remarked that a true maximum principle might have to do with something even in the magnetic field. We had previously separated the Boltzmann equation into even and odd parts, but had lost interest in the problem until stimulated again by Professor Holstein's remark.

A second effort of the paper has been to relate the variational formulation of the problem with the recent work of Kohler,<sup>9</sup> McClure,<sup>10</sup> Chambers,<sup>11</sup> Lifshitz *et al.*,<sup>12,13</sup> and others who solved the Boltzmann equation with a view to effects in large magnetic fields. This relationship is discussed in Sec. 5, and in fact the variational principle can be used to provide interpolation formulas that accommodate open orbit effects. The problem as we have envisaged it resolves itself into consideration of the inverse operator  $1/(L+M)$ , where  $L$  is the ordinary collision operator (involving drag effects) [Eq. (A14)], and  $M$  the magnetic operator [Eq. (2.3)].

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<sup>1</sup> J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1960), p. 508.

<sup>2</sup> J. M. Ziman, *Can. J. Phys.* **34**, 1256 (1956).

<sup>3</sup> F. Garcia-Moliner and S. Simons, *Proc. Cambridge Phil. Soc.* **53**, 848 (1957).

<sup>4</sup> M. Tsuji, *J. Phys. Soc. Japan* **14**, 618 (1959). See references therein.

<sup>5</sup> H. Bross, *Z. Naturforsch.* **14a**, 560 (1959).

<sup>6</sup> M. Kohler, *Z. Physik* **124**, 772 (1948); **125**, 679 (1949); **126**, 495 (1949). See also A. H. Wilson, *Theory of Metals* (Cambridge University Press, New York, 1953), Chap. X.

<sup>7</sup> M. Kohler, *Ann. Physik.* **5**, 181; **6**, 18 (1949).

<sup>8</sup> G. E. Tauber, *Can. J. Phys.* **36**, 1308 (1958); see also Dorn, *Z. Naturforsch.* **12a**, 739 (1957).

<sup>9</sup> M. Kohler, *Ann. Physik.* **5**, 99 (1949).

<sup>10</sup> J. W. McClure, *Phys. Rev.* **101**, 1642 (1956).

<sup>11</sup> R. G. Chambers, *Proc. Roy. Soc. (London)* **A238**, 344 (1956).

<sup>12</sup> I. M. Lifshitz, M. Y. Azbel', and M. I. Kaganov, *Soviet Phys.—JETP* **4**, 41 (1956).

<sup>13</sup> M. Y. Azbel', M. I. Kaganov, and I. M. Lifshitz, *Soviet Phys.—JETP* **5**, 967 (1957). See also G. E. Zil'berman, *Ibid.* **2**, 650 (1956).

The high-field expansion regards  $M$  greater than  $L$ , but one of the eigenvalues of  $M$  is zero. A method for overcoming the difficulty associated with this is discussed in Sec. 5 and Appendix E for the case when a time of relaxation exists, and the natural extension of the method to the situation when such an assumption is not valid is presented in Appendix F.

In dealing with the maximum principle, operators were found which directed our attention to symmetry considerations. The Onsager relations have long ago been shown by Kohler<sup>14</sup> and Meixner<sup>15</sup> to be satisfied by the solutions of the Boltzmann equation. We present a shortened proof containing drag effects, and, in so doing, uncover some apparently new symmetry relations [Eq. (4.7)], that are valid when a time of relaxation exists and the drag effects can be neglected. Presumably these could be verified by measurements on impurity scattering at temperatures below which drag effects are important.

Finally, it is shown in Sec. 7 that spin waves stimulate magnon-drag effects almost exactly analogous to phonon-drag effects. The analogy with phonons is almost exact, since double-magnon processes are shown in Appendix G to be negligible. The difference between magnon and phonon scattering formally consists then only in that the former alters the electron spin, the latter does not. (Also in the former, the one-electron energy depends on spin.)

The paper is organized as follows: Sec. 2 defines the Boltzmann equation and the conductivity tensor, appealing to Appendix A for some details. The maximum principle is derived in Sec. 3 with the crucial symmetry relation proved in Appendix B. A method for handling integrals involving inverse operators is discussed in Appendix C. The Onsager relations and others are discussed in Sec. 4. For this, the heat conductivity is required, and that is given in Appendix D. The relation between the variational solutions and the recent high-field work is discussed in Sec. 5, with some of the matrix element calculations in Appendixes E and F. The results for the high-field drag effects is tabulated in Sec. 5. The new variational principle is applied in Sec. 6, and interpolation formulas discussed there. The theory of magnon drag is carried out in Sec. 7, with the justification of the neglect of double-magnon processes indicated in Appendix G.

## 2. DEFINITIONS

The distribution function for the electrons is assumed to be of the form

$$f(\mathbf{k}, s) = f_0(E_s(\mathbf{k})) - \frac{\partial f_0}{\partial E} g(\mathbf{k}, s), \quad (2.1)$$

where  $s$  denotes spin and  $\mathbf{k}$  the one-electron state and  $f_0$  is the Fermi function; the Boltzmann equation is set

up in Appendix A, and is<sup>16</sup>

$$(L+M)g = -(\mathbf{A} \cdot \mathbf{X} + \mathbf{B} \cdot \mathbf{Y}), \quad (2.2)$$

where  $L$  [given by (A14)] is the collision operator (containing phonon-drag and magnon-drag effects) acting on  $g$ , and  $M$  is the "magnetic operator"

$$M(g) = -\frac{|e|}{\hbar c} \frac{\partial f_0}{\partial E} \mathbf{v} \times \mathbf{H} \cdot \nabla_{\mathbf{k}} g = \frac{\partial f_0}{\partial E} \frac{d}{dt} g, \quad (2.3)$$

where  $\mathbf{H}$  is the magnetic field,  $\mathbf{v}$  the velocity of the electron, and  $t$  [the  $s(k)$  of McClure<sup>10</sup> and the  $t(\theta)$  of Chambers<sup>11</sup>] representing the time from some point of origin in its orbit as the electron would rotate in  $\mathbf{k}$  space under the influence of the magnetic field alone.  $\mathbf{X}$  and  $\mathbf{Y}$  are the forces

$$\mathbf{X} = \mathbf{E} + |e| \nabla_{\mathbf{r}} \zeta, \quad (2.4a)$$

$$\mathbf{Y} = -T^{-1} \nabla_{\mathbf{r}} T, \quad (2.4b)$$

where  $\mathbf{E}$  is the electric field, and  $\zeta$  the Fermi energy.  $\mathbf{A}$  and  $\mathbf{B}$  are then

$$\mathbf{A} = |e| \mathbf{v} (\partial f_0 / \partial E), \quad (2.5a)$$

$$\mathbf{B} = -(E - \zeta) \mathbf{v} (\partial f_0 / \partial E) + \gamma(\mathbf{k}, s) + \gamma^{(m)}(\mathbf{k}, s), \quad (2.5b)$$

where  $\gamma$  is the phonon-drag term, Eq. (A18) of Appendix A, [equivalent to the  $H_x$  of Eq. (31) of TM1] and  $\gamma^{(m)}$  the corresponding magnon-drag term (see Sec. 7), obtained by solving the phonon and magnon Boltzmann equations.

In demonstrating the maximum principle, we must introduce an operator  $L^{-1}$  which is the inverse of  $L$  defined as follows:

$$L^{-1}L(g) = g. \quad (2.6)$$

When a time of relaxation  $\tau$  exists, we have

$$\begin{aligned} L &\rightarrow (1/\tau)(\partial f_0 / \partial E), \\ L^{-1} &\rightarrow \tau(\partial f_0 / \partial E)^{-1}, \end{aligned} \quad (2.7)$$

The solution to (2.2) is formally

$$g = -(L+M)^{-1}[\mathbf{A} \cdot \mathbf{X} + \mathbf{B} \cdot \mathbf{Y}], \quad (2.8)$$

and hence the charge flow  $\mathbf{J}$  and the energy flow  $\mathbf{W}$  can be written

$$J_i = \sum_j \sigma_{ij} X_j + \sum_j S_{ij} Y_j, \quad (2.9a)$$

$$W_i^{(\text{elec})} = \sum_j \sigma_{ij}' X_j + \sum_j S_{ij}' Y_j, \quad (2.9b)$$

where

$$\sigma_{ij} = -|e| \sum_{\mathbf{k}} v_i (\partial f_0 / \partial E) (L+M)^{-1} A_j, \quad (2.10a)$$

$$S_{ij} = -|e| \sum_{\mathbf{k}} v_i (\partial f_0 / \partial E) (L+M)^{-1} B_j, \quad (2.10b)$$

$$\sigma_{ij}' = \sum_{\mathbf{k}} v_i E (\partial f_0 / \partial E) (L+M)^{-1} A_j, \quad (2.11a)$$

$$S_{ij}' = \sum_{\mathbf{k}} v_i E (\partial f_0 / \partial E) (L+M)^{-1} B_j. \quad (2.11b)$$

<sup>16</sup> Some of the notation and of the derivations in the present paper refer to previous articles, Phys. Rev. **112**, 1587 (1958); **120**, 381 (1960). We refer to these as TM1 and TM2, respectively, in the text. References to the literature on the variational principle can be found there, also.

<sup>14</sup> M. Kohler, Ann. Physik. **40**, 601 (1941).

<sup>15</sup> J. Meixner, Ann. Physik. **38**, 609 (1940).

The effects of the nonequilibrium component of the phonons (and magnons) appear in two places: First in the  $\gamma$ 's in **B**, Eq. (2.5b), which give rise to the "drag" effects in the thermoelectric power, and second in the additional contributions to  $L$  [see Eq. (A14)], which gives rise to "relaxation" effects in all the normal transport coefficients. For a magnetic field, all that alters is that  $L$  has  $M$  added to it, and the conclusions are of the same sort as without a magnetic field.

There are two quantities we shall want expressions for. If a magnetic field is applied in the  $z$  direction, and an electric field is applied in the  $x$  direction, and no current flow is allowed in the  $y$  direction, then a field  $X_y$  is built up. The ratio  $X_x/J_x$  is then

$$\rho_{xx} = X_x/J_x = [\sigma_{xx} - \sigma_{xy}\sigma_{yx}/\sigma_{yy}]^{-1}. \quad (2.12)$$

If a magnetic field is applied in the  $z$  direction, and a thermal gradient in the  $x$  direction, then  $X_x$  and  $X_y$  are both generated, if no electrical currents are allowed to flow ( $J_x=0$ ,  $J_y=0$ ). The latter conditions allow  $X_x$  and  $X_y$  to be determined as functions of  $Y_x$ , and the thermoelectric power is then

$$S = X_x(dT/dy)^{-1} = (\rho_{xx}/T)[S_{xx} - S_{yx}(\sigma_{xy}/\sigma_{yy})]. \quad (2.13)$$

Thus the "drag" effects enter  $S$  linearly through  $S_{xx}$  and  $S_{xy}$ , and the relaxation effects enter in a complex way in all the  $S_{ij}$ 's and  $\sigma_{ij}$ 's.

### 3. THE MAXIMUM PRINCIPLE

Consider  $g(\mathbf{k}, s)$  to be separated into a part  $g^{(e)}$  even, and a part  $g^{(o)}$  odd in the magnetic field. The Boltzmann equation (2.2) then separates into two, one arising from the terms odd in the magnetic field:

$$g^{(o)} = -L^{-1}Mg^{(e)}, \quad (3.1a)$$

the other from terms even in the magnetic field, which when (3.1a) is substituted into it becomes

$$\mathcal{L}(g^{(e)}) = -(\mathbf{A} \cdot \mathbf{X} + \mathbf{B} \cdot \mathbf{Y}), \quad (3.1b)$$

where

$$\mathcal{L} \equiv L - ML^{-1}M. \quad (3.2)$$

Now in Appendix B, it is shown that  $\mathcal{L}$  is *positive definite, and symmetric*, [i.e., satisfies Eq. (B2) with  $\mathcal{L}$  taking the place of the operator  $O$ ]. Hence, (3.1b) which is identical in form with the Boltzmann equation without a magnetic field, except that  $L$  has become  $\mathcal{L}$ , leads to a true maximum principle in the usual way. We shall not repeat the details of the variational procedure; these can be found in the articles mentioned in reference 6. Corresponding to the separation of  $g$  into even and odd parts, we can separate  $\sigma_{ij}$  and  $S_{ij}$  into even and odd parts by means of the identity

$$\frac{1}{L+M} = \frac{1}{\mathcal{L}} \left(1 - M \frac{1}{L}\right) = \left(1 - \frac{1}{L}M\right) \frac{1}{\mathcal{L}}. \quad (3.3)$$

Thus, the even part  $\sigma_{ij}^{(e)}$  and the odd part  $\sigma_{ij}^{(o)}$  are

$$\sigma_{ij}^{(e)} = -|e| \sum_{\mathbf{k}} v_i (\partial f_0 / \partial E) \mathcal{L}^{-1} A_j, \quad (3.4a)$$

$$\sigma_{ij}^{(o)} = -|e| \sum_{\mathbf{k}} v_i (\partial f_0 / \partial E) \mathcal{L}^{-1} (-ML^{-1}) A_j, \quad (3.4b)$$

and similarly for  $S_{ij}$ : The quantity maximized by the new variational principle is then  $\sigma_{ii}^{(e)}$ . This will provide us with  $g^{(e)}$ ; and Eq. (3.1a) will then enable us to get  $g^{(o)}$ .

Thus the major conclusion of this paper is that where symmetry properties are concerned, one can deal with the operator  $\mathcal{L}$  which has all the virtues of  $L$ . In practice, one is however faced with integrals involving the inverse operator  $L^{-1}$  but it is shown in Appendix C that all such integrals reduce to an "internal" Boltzmann equation, which can be solved separately so to speak, off to one side. It is in practice the fact that these internal Boltzmann problems must converge independently that distinguishes our solutions from Ziman's and Tsuji's.

### 4. SYMMETRY PROPERTIES

The basic symmetry properties that the conductivity coefficients satisfy are the Kelvin-Onsager relations. Kohler<sup>14</sup> and Meixner<sup>15</sup> have shown that they are satisfied by the solutions to the Boltzmann equation including a magnetic field. The situation including the nonequilibrium component of the phonons requires that the phonon heat flow be computed, and again the relations have been shown to be satisfied.<sup>4,16</sup> Use of the operator  $\mathcal{L}$  introduced in the last section facilitates these proofs. We find also that when a time of relaxation is assumed to exist, and phonon-drag effects neglected, some new symmetry relations occur.

If the electric and thermal currents are not written as in (2.9) but as<sup>17</sup>

$$J_i = \sum_j S_{ij}^{(1)} X_j^* + \sum_j S_{ij}^{(2)} Y_j, \quad (4.1a)$$

$$W_i = \sum_j S_{ij}^{(3)} X_j^* + \sum_j S_{ij}^{(4)} Y_j, \quad (4.1b)$$

where  $W_i$  includes the electron and phonon energy flows, and where

$$X_j^* = \mathcal{E}_j + \frac{1}{|e|} T \frac{d}{dx_j} \frac{\zeta}{T} = X_j + \frac{\zeta}{|e|} Y_j, \quad (4.2)$$

then the Kelvin-Onsager relations are

$$S_{ij}^{(m)}(H) = S_{ji}^{(m)}(-H) \cdots, \quad m=1, 4, \quad (4.3a)$$

$$S_{ji}^{(2)}(H) = S_{ji}^{(3)}(-H), \quad (4.3b)$$

$$S_{ii}^{(m)}(H) = S_{ii}^{(m')}(-H) \cdots (mm') \\ = (1,1)(2,3)(3,2)(4,4). \quad (4.3c)$$

<sup>17</sup>See. H. B. Callen, Phys. Rev. **73**, 1349 (1948).

The relationship between the coefficients in (4.1a) and (2.9a) are

$$S_{ij}^{(1)} = \sigma_{ij}, \quad (4.4a)$$

$$S_{ij}^{(2)} = S_{ij} - \frac{\zeta}{|e|} \sigma_{ij} \\ = \sum_{\mathbf{k}} A_i \frac{1}{L+M} [v_j E (\partial f_0 / \partial E) - \gamma_j]. \quad (4.4b)$$

To get the relations between the coefficients in (4.1b) and (2.9b) requires computing the energy flow of the phonons: This is done in Appendix D. The result is

$$S_{ij}^{(3)} = \sum_{\mathbf{k}} [v_i E (\partial f_0 / \partial E) - \gamma_i] \frac{1}{L+M} A_j, \quad (4.4c)$$

$$S_{ij}^{(4)} = T \kappa_{ij} + \sum_{\mathbf{k}} [v_i E (\partial f_0 / \partial E) - \gamma_i] \\ \times \frac{1}{L+M} \left( B_j - \frac{\zeta}{|e|} A_j \right), \quad (4.4d)$$

where  $\kappa_{ij}$  is given in (D4). Now in Appendix B it is shown that  $\mathcal{L}^{-1} M L^{-1}$  is "antisymmetric" as defined by Eq. (B3), and that  $\mathcal{L}$  is symmetric. Hence from Eqs. (3.3) and (4.4), the Kelvin-Onsager relations (4.3) are satisfied.

We see moreover from (4.4b) that if phonon-drag effects are neglected

$$S_{ij}^{(2)}(H) = \frac{1}{|e|} \sum_{\mathbf{k}} A_i \frac{1}{L+M} A_j E. \quad (4.6)$$

If now the collisions are elastic, i.e., if a time of relaxation is assumed to exist for  $L$ , then  $E$  commutes with  $L+M$ ; hence  $S_{ij}^{(2)}$  [and consequently  $S_{ij}$ , Eq. (2.10b)] satisfies

$$S_{ij}^{(2)}(H) = S_{ji}^{(2)}(-H), \\ S_{ii}^{(2)}(H) = S_{ii}^{(2)}(-H), \quad (4.7)$$

that is, it satisfies the same relations that  $S_{ij}^{(1)}$  and  $S_{ij}^{(4)}$  do. (This is apparently a new result, as a search has failed to reveal it in the literature.)

It may be possible to verify (4.7) experimentally at very low temperatures, below the phonon-drag region and in which elastic impurity scattering prevails.

## 5. VARIATIONAL AND "HIGH-FIELD" SOLUTIONS

It is possible to correlate the solutions of the Boltzmann equation that have recently been discussed by McClure,<sup>10</sup> Chambers,<sup>11</sup> Lifshitz *et al.*<sup>12,13</sup> (which have led to the open-orbit high-field considerations) with the variational solutions as follows. First a complete set of (not necessarily orthogonal) functions  $\varphi_n(\mathbf{k}, s)$  is introduced. By definition

$$\sum_n \varphi_n(\mathbf{k})^* \varphi_n(\mathbf{k}') = \delta_{\mathbf{k}, \mathbf{k}'}. \quad (5.1)$$

Then we insert in the tensor elements (2.10) factors like (5.1) on each side of  $1/(L+M)$ , or using the result of Sec. 2, around  $1/\mathcal{L}$ . For the purposes of this section it is advantageous to use the former; we shall come back to the new formulation in later sections. Thus

$$\sigma_{ij} = - \sum_{nn'} A_{in} \left( \frac{1}{L+M} \right)_{nn'}^{ij} A_{jn'}, \quad (5.2a)$$

$$S_{ij} = - \sum_{nn'} A_{in} \left( \frac{1}{L+M} \right)_{nn'}^{ij} B_{jn'}, \quad (5.2b)$$

where, say

$$A_{in} = \sum_{\mathbf{k}} A_i(\mathbf{k}) \varphi_{in}(\mathbf{k}), \quad (5.3a)$$

$$\left( \frac{1}{L+M} \right)_{nn'}^{ij} = \sum_{\mathbf{k}} \varphi_{in}(\mathbf{k})^* \frac{1}{L+M} \varphi_{jn'}(\mathbf{k}). \quad (5.3b)$$

To obtain (5.2) and (5.3), we have in fact inserted one factor (5.1),  $\delta_{\mathbf{k}\mathbf{k}'}^{(i)}$ , say, on the left of  $1/(L+M)$ , and another,  $\delta_{\mathbf{k}\mathbf{k}'}^{(j)}$ , say, on the right,  $\delta^{(i)}$  and  $\delta^{(j)}$  differing by containing  $\varphi_{in} = v_i(E - \zeta)^n$  and  $\varphi_{jn} = v_j(E - \zeta)^n$ , respectively. This corresponds to the usual approximation, in which  $\varphi_{in}$  and  $\varphi_{jn}$  are assumed to be selected out in the respective places because of the orthogonality of angular factors (arising specifically in  $A_n$  or  $B_n$ ). Of course, a truly complete set would incorporate both situations, and no distinction would have been made.

Equations (5.2) form the solution to the problem. The *variational expressions* are obtained from this by first approximating the infinite  $n$  sum as a finite one,  $n=0, 1 \dots N$  (and letting  $N$  go to infinity afterward), and employing the relation

$$(O^{-1})_{nn'} = O^{nn'} / \|O\|, \quad (5.4a)$$

where  $O$  represents any operator, and  $O^{nn'}$  is the cofactor of  $O_{nn'}$  in the determinant  $\|O\|$  whose elements are

$$O_{nn'} = \sum_{\mathbf{k}} \varphi_n(\mathbf{k})^* O(\varphi_{n'}). \quad (5.4b)$$

Equations (5.2) become then in one step

$$\sigma_{ij} = - \sum_{nn'} A_{in} \frac{(L+M)_{ij}^{nn'}}{\|L+M\|} A_{jn'} = \frac{D^{ij}(A_i, A_j)}{D^{ij}}, \quad (5.5a)$$

$$S_{ij} = D^{ij}(A_i, B_j) / D^{ij}, \quad (5.5b)$$

where  $D^{ij}$  is a determinant with elements  $L_{ij}^{nn'} + M_{ij}^{nn'}$ , and where  $D^{ij}(A_i, A_j)$  is  $D^{ij}$  bordered with a row of  $A_{in}$  and a column of  $A_{jn}$ , the lower right-hand element being zero. Equations (5.5) are the variational expressions for the transport coefficients.

The *high-field expressions* following from (5.2) arise by by choosing the functions  $\varphi_n$  to be the eigenfunctions of the operator  $M$  [Eq. (2.3)]

$$\varphi_n = e^{i\omega_n t}, \quad (5.6a)$$

$$\omega_n = 2\pi n l / T \sim H^{-1}, \quad (n=0, \pm 1, \dots), \quad (5.6b)$$

where  $T=T(k_z, E)$  is the period of the orbit (see references 10–13). If that is done and if we assume a constant time of relaxation  $\tau$ , and a constant  $T$ , then

$$(L+M)^{-1}\varphi_n = (\partial f_0/\partial E)^{-1}(\tau^{-1}+i\omega_n)^{-1}\varphi_n, \quad (5.7)$$

and (5.2) becomes in one step

$$\sigma_{ij} = |e|^2 \sum_{nn'} v_{in} v_{jn'} \frac{\tau}{1+i\omega_n \tau} \frac{\partial f_0}{\partial E} \varphi_n \varphi_{n'}, \quad (5.8)$$

which is just the result of references 10–13 in the same approximation.

Thus the high-field work and the standard variational procedure differ essentially only in the choice of the complete set  $\varphi_n$ , since (5.8) can be converted to determinants just as in (5.4) (see Sec. 6).

If a time of relaxation is not assumed (or assumed not to be constant), then (5.7) is no longer valid, and it becomes a question of expanding  $1/(L+M)$ . This can be done by standard operator techniques:

$$\frac{1}{L+M} = \frac{1}{L'+M'} = \frac{1}{M'} - \frac{1}{M'} L' \frac{1}{M'} + \dots, \quad (5.9)$$

where we added and subtracted a quantity  $C$  (to be determined):

$$\begin{aligned} L' &= L - C, \\ M' &= M + C, \end{aligned} \quad (5.10)$$

because one of the eigenvalues of  $M$  is zero, which can become embarrassing. It turns out to be advantageous to choose  $C$  such that  $L$  has no diagonal elements, and  $M'$  has *only* diagonal elements, with respect to the eigenfunctions of  $M$ . Such a condition, when a time of relaxation is valid, is satisfied by the choice  $L_{00}$ , where

$$L_{nn'}(k_z, E) = \oint dl \varphi_n^* L(\varphi_{n'}), \quad (5.11)$$

the integral being taken around an orbit (if closed) or a period, in general. Since the eigenfunctions (5.6) are pure exponentials, we have  $L_{nn}=L_{00}$  for any  $n$ . It is then obvious that  $L_{nn'}=0$ , and in fact we get

$$\begin{aligned} (1/M')_{nn'} &= (L_{00}+M_n)^{-1} \delta_{-n,n'} \\ &= (\partial f_0/\partial E)^{-1} (\tau_{00}^{-1}+i\omega_n)^{-1} \delta_{-n,n'}, \end{aligned} \quad (5.12a)$$

$$\begin{aligned} L_{nn'} &= L_{nn'} - \delta_{-n,n'} L_{00} \\ &= (\partial f_0/\partial E) [(\tau^{-1})_{nn'} - (\tau^{-1})_{nn} \delta_{-n,n'}], \end{aligned} \quad (5.12b)$$

where the  $(1/\tau)_{nn'}$ 's are obtained from (2.7). In the limit of large  $H$  we can expand  $1/M_{n'}$ :

$$\lim_{H \rightarrow \infty} \frac{1}{M_{n'}} \sim \text{const}, \quad n=0 \\ \frac{1}{M_{n'}} \sim 1/H, \quad n \neq 0. \quad (5.13)$$

If a time of relaxation cannot be assumed, then the generalization of this procedure can be handled by the

introduction of certain "projection" operators, as shown in Appendix F.

The expansion here, and the method in Appendix F, should be contrasted with the "Fourier method" of Sec. 5 of Lifshitz *et al.*<sup>12</sup> Our results will differ by containing directly the matrix elements of  $M'$  which keeps the diagonal part of  $L+M$  together in one piece. When a time of relaxation may be thought to exist, our results will relate to those of Chambers<sup>11</sup> in a way parallel to how our results in Appendix F relate to those of reference 12.

The matrix elements for the case when a time of relaxation is assumed are discussed in Appendix E, it not being difficult to see from (5.9) that the high-field limit is

$$\begin{aligned} \lim_{H \rightarrow \infty} \left( \frac{1}{L'+M'} \right)_{mn} &= \frac{1}{M_n'} \delta_{nm} \\ &- \frac{1}{M_n'} \left[ L_{mn'} - L_{m0'} \frac{1}{M_{0'}} L_{0n'} \right] \frac{1}{M_n'}, \end{aligned} \quad (5.14)$$

which with (5.13) yields

$$\lim_{H \rightarrow \infty} \left( \frac{1}{L'+M'} \right)_{00} = \frac{1}{M_{0'}} = \frac{1}{L_{00}} \sim \text{const}, \quad (5.15a)$$

$$\begin{aligned} \lim_{H \rightarrow \infty} \left( \frac{1}{L'+M'} \right)_{0n} &= -\frac{1}{M_{0'}} \frac{L_{0n'}}{M_n'} \\ &= -\frac{1}{L_{00}} \frac{L_{0n}}{L_{00}+M_n} \sim \frac{1}{H}, \end{aligned} \quad (5.15b)$$

$$\lim_{H \rightarrow \infty} \left( \frac{1}{L'+M'} \right)_{m0} = -\frac{1}{L_{00}+M_m} \frac{L_{m0}}{L_{00}} \sim \frac{1}{H}, \quad (5.15c)$$

$$\lim_{H \rightarrow \infty} \left( \frac{1}{L'+M'} \right)_{n,-n} = \frac{1}{M_n'} = \frac{1}{L_{00}+M_n} \sim \frac{1}{H}. \quad (5.15d)$$

With these results, we can return to (5.2) and write the limiting forms of  $\sigma_{ij}$  and  $S_{ij}$  for certain assumptions concerning  $v_{i0}$  (i.e.,  $A_{i0}$ ). The drag effects manifest themselves in the fact that  $B_{j0} \neq 0$  in general whether or not open orbits occur, because  $\gamma_{i0} \neq 0$ . [See Eq. (2.5b) and Eq. (A18).] Thus the high-field limits are [for  $S$ , see Eq. (2.13)]:

$$\text{I. } v_{i0} \neq 0; \quad v_{j0} \neq 0; \quad i \neq j;$$

$$\sigma_{ij} \rightarrow -\sum_{\mathbf{k}} A_{i0} \frac{1}{M_{0'}} A_{j0} \sim \text{const}, \quad (5.16a)$$

$$S_{ij} \rightarrow -\sum_{\mathbf{k}} A_{i0} \frac{1}{M_{0'}} B_{j0} \sim \text{const}, \quad (5.16b)$$

$$S = \frac{\rho_{ii}}{T} \left[ S_{ii} - S_{jj} \frac{\sigma_{ij}}{\sigma_{jj}} \right] \sim \text{const}. \quad (5.16c)$$

II.  $v_{i0}=0$ ;  $v_{j0}\neq 0$ ;  $i\neq j$ ;

$$\sigma_{ij} \rightarrow - \sum_{m\neq 0} \sum_{\mathbf{k}} A_{im} \left\{ \left[ \frac{1}{L'+M'} \right]_{m0} A_{j0} + \left[ \frac{1}{L'+M'} \right]_{m,-m} A_{j,-m} \right\} \sim \frac{1}{H}, \quad (5.17a)$$

$$S_{ij} \rightarrow - \sum_{m\neq 0} \sum_{\mathbf{k}} A_{im} \left\{ \left[ \frac{1}{L'+M'} \right]_{m0} B_{j0} + \left[ \frac{1}{L'+M'} \right]_{m,-m} B_{j,-m} \right\} \sim \frac{1}{H}, \quad (5.17b)$$

$$S = \frac{\rho_{ii}}{T} \left[ S_{ii} - S_{ji} \frac{\sigma_{ij}}{\sigma_{jj}} \right] \sim H. \quad (5.17c)$$

III.  $v_{i0}\neq 0$ ;  $v_{j0}=0$ ;  $i\neq j$ ;

$$\sigma_{ij} \rightarrow - \sum_{n\neq 0} \sum_{\mathbf{k}} \left\{ A_{i,0} \left[ \frac{1}{L'+M'} \right]_{0n} + A_{i,-n} \left[ \frac{1}{L'+M'} \right]_{-n,n} A_{jn} \right\} \sim \frac{1}{H}, \quad (5.18a)$$

$$S_{ij} \rightarrow - \sum_{\mathbf{k}} A_{i0} \frac{1}{M'_0} B_{j0} \sim \text{const (drag)}, \quad (5.18b)$$

$$S \rightarrow \text{const.} \quad (5.18c)$$

IV.  $v_{i0}=v_{j0}=0$ ;  $i\neq j$ ;

$$\sigma_{ij} \rightarrow - \sum_{n\neq 0} \sum_{\mathbf{k}} A_{i,-n} \frac{1}{M'_n} A_{jn} \sim \frac{1}{H}, \quad (5.19a)$$

$$S_{ij} \rightarrow - \sum_{m\neq 0} \sum_{\mathbf{k}} A_{im} \left\{ \left[ \frac{1}{L'+M'} \right]_{m0} B_{j0} + \left[ \frac{1}{L'+M'} \right]_{m,-m} B_{j,-m} \right\} \sim \frac{1}{H}, \quad (5.19b)$$

$$S \rightarrow \frac{\rho_{ii}}{T} \left[ -S_{ji} \frac{\sigma_{ij}}{\sigma_{jj}} \right] \sim \text{const.} \quad (5.19c)$$

V.  $v_{i0}\neq 0$ ;  $i=j$ ;

$$\sigma_{ii} \rightarrow - \sum_{\mathbf{k}} A_{i0} \frac{1}{M'_0} A_{i0} \sim \text{const}, \quad (5.20a)$$

$$S_{ii} \rightarrow - \sum_{\mathbf{k}} A_{i0} \frac{1}{M'_0} B_{i0} \sim \text{const.} \quad (5.20b)$$

VI.  $v_{i0}=0$ ;  $i=j$ ;

$$\sigma_{ii} \rightarrow - \sum_{n\neq 0} \sum_{\mathbf{k}} A_{i,-n} \frac{1}{M'_n} A_{in} = - \sum_{n=1}^{\infty} A_{in} A_{i,-n} \left( \frac{1}{M'_n} + \frac{1}{M'_{-n}} \right) \sim \frac{1}{H^2}, \quad (5.21a)$$

$$S_{ij} \rightarrow - \sum_{n\neq 0} \sum_{\mathbf{k}} A_{in} \left\{ \left[ \frac{1}{L'+M'} \right]_{n0} B_{i0} + \left[ \frac{1}{L'+M'} \right]_{n,-n} B_{i,-n} \right\} \sim \frac{1}{H} \text{ (drag)}. \quad (5.21b)$$

The following points are to be noted about these equations. The  $a$  members correspond to well-known results<sup>12,13</sup> (except for use of  $M'_n$  for  $M_n$ ). The  $b$  members all contain drag effects in addition to the "drift" effects. In (5.19b) the drag effects add a new type of term (the first  $m$  sum) which has the same limiting values (with respect to powers of  $H$ ) as the drift term. In (5.18b) however, the drag effect yields a term which approaches a constant in the limit, whereas the drift effect would yield a term going as  $1/H$ . Similarly, in (5.21b), the drag effect yields a term whose limit goes as  $1/H$ , whereas the drift term goes as  $1/H^2$ . Thus the high-field limits are altered by phonon-drag effects.

The  $c$  members of these equations show the thermoelectric power limits as calculated from (2.13), but the notation needs some explaining. In these equations, the temperature gradient is supposedly applied in the  $i$  direction, where  $i$  is perpendicular to  $z$ , the direction of the magnetic field, and  $j$  is mutually perpendicular to  $i$  and  $z$ . Thus in (5.17c),  $S_{ji}$  corresponds to what is calculated in (5.18b).

In one case, (5.17c), we find  $S \sim H$ . However, in device applications what is crucial is not  $S$  but the "figure of merit"  $Z = S^2/\rho\kappa$ , and it will be the case that the limit  $S^2 \sim H^2$  is compensated by the limit  $\rho \sim H^2$ , so that  $Z \sim \text{const}$ . (This supposes that  $\lim_{H \rightarrow \infty} \kappa = \kappa_{\text{phonon}}$ , when  $\rho \sim H^2$ .)

It should be noted that in the thermoelectric powers, the limiting expressions behave as some power of  $1/H$ , and this power is the same whether or not drag quantities are included. One might have anticipated say (5.18b) perhaps to yield a different asymptotic value for some  $S$ , but it is not the case; both terms in (5.17c) have the same limiting power of  $1/H$ .

This completes the formal solution of the Boltzmann equation. The generalization to situations where a relaxation time does not exist is carried out in Appendix F. The main object has been to show the relation between the variational solution (5.5) and the high-field solution (5.8). This relationship is developed further in the next section.

## 6. NEW VARIATIONAL SOLUTIONS AND INTERPOLATION FORMULAS

In the last section we have confined ourselves to the usual approach in which matrix elements of the operator  $L+M$  appear. We saw, however, in Sec. 3 that it is the operator  $\mathfrak{L}$  that has the desirable symmetry properties. In this section, we shall convert to the new variational principle.

First, instead of (5.2), we obtain from (2.10) using (3.3),

$$\sigma_{ij} = - \sum_{nn'} A_{in} \bar{A}_{jn} (\mathfrak{L}^{-1})_{nn'}^{ij}, \quad (6.1a)$$

$$S_{ij} = - \sum_{nn'} A_{in} \bar{B}_{jn} (\mathfrak{L}^{-1})_{nn'}^{ij}, \quad (6.1b)$$

where, for any function  $Q_j$ ,

$$\bar{Q}_{jn} \equiv \sum_{\mathbf{k}} \varphi_{jn}(\mathbf{k}) (1 - ML^{-1}) Q_j(\mathbf{k}). \quad (6.2)$$

Thus instead of (5.5) we obtain

$$\sigma_{ij} = \mathfrak{D}(A_i, \bar{A}_j) / \mathfrak{D}, \quad (6.3a)$$

$$S_{ij} = \mathfrak{D}(A_i, \bar{B}_j) / \mathfrak{D}, \quad (6.3b)$$

where the elements of the determinant  $\mathfrak{D}$  are  $\mathfrak{L}_{nn'}^{ij}$ , and where  $\mathfrak{D}(A_i, \bar{B}_j)$  is the determinant  $\mathfrak{D}$  bordered with a row of  $A_{in}$  and a column of  $\bar{B}_{jn}$ , the lower right-hand element being zero. The results (6.3) are what the new variational principle leads to, and are to be contrasted with (5.5). We repeat that, *in practice*, what distinguishes (6.3) from (5.5) is that in (6.3), integrals involving  $L^{-1}$  occur that are evaluated by separate internal variational principles, and that these integrals must converge independently, whereas on the old principle they appear automatically in an approximation that corresponds to the number of rows and columns in  $D$ .

We should like now to get a variational expression that will include open-orbit effects. This amounts to constructing an appropriate complete set of functions  $\varphi_n$ . In the standard treatments, the choice was

$$\varphi_n^{(x)} = v_x (E - \zeta)^n \cong v \cos \alpha_x (E - \zeta)^n, \quad (6.4)$$

where spherical energy surfaces lay in the background, and  $\alpha_x$  is the angle between  $\mathbf{k}$  and the direction of applied electric field,  $x$ . Corresponding sets of functions  $\varphi_n^{(y)}$  and  $\varphi_n^{(z)}$  would also be necessary as used in (5.2) and (6.1).

Now if a magnetic field is applied in the  $z$  direction, and spherical surfaces still obtain, the standard expressions (6.4) may be written

$$\begin{aligned} v_x &= v \sin \alpha \cos \beta, \\ v_y &= v \sin \alpha \sin \beta = v \sin \alpha \cos(\beta + \pi/2), \\ v_z &= v \cos \alpha, \end{aligned} \quad (6.5)$$

where  $\alpha$  is the angle between  $z$  and  $\mathbf{k}$ , and where  $\beta$  is the azimuthal about the polar  $z$ . A complete set of

angular functions is suggested by each component as follows:

$$\varphi_{n_1 n_2 n_3}^{(x)} = \sin n_1 \alpha \cos n_2 \beta (E - \zeta)^{n_3}, \quad (6.6a)$$

$$\varphi_{n_1 n_2 n_3}^{(y)} = \sin n_1 \alpha \cos [n_2 (\beta + \frac{1}{2} \pi)] (E - \zeta)^{n_3}, \quad (6.6b)$$

$$\varphi_{n_1 n_2 n_3}^{(z)} = \cos n_1 \alpha \cos [(n_2 - 1) \beta] (E - \zeta)^{n_3}, \quad (6.6c)$$

where  $n_i = 0, \pm 1, \dots$ , ( $i = 1, 2, 3$ ). The usual expression is obtained by letting  $n_1 = n_2 = 1$ .

Now if we wish open-orbit effects, we are interested in distorted surfaces, and in particular in the possibility of there being a component  $V_{x0}$  of  $v_x$  which is constant with respect to  $\beta$ . This corresponds roughly to the term  $n_2 = 0$  in (6.6), except that for distorted surfaces, if we go around the orbit in  $k$  space, not only does  $\beta$  alter, but  $\alpha$  will vary also. Therefore we desire in addition, a replacement of  $\alpha$  by a quantity  $\bar{\alpha}$  which will remain constant as the electron describes an orbit. We can do this formally as follows: Consider a solid object with a center. This is to represent the occupied part of a zone. Through the center draw the  $z$  axis, and at some distance  $L$  from the center along  $z$  draw a plane perpendicular to the  $z$  axis. This plane cuts the object in a slice of area with perimeter  $P$ . We now construct a conical surface connecting the perimeter of this slice with the center of the object. The curved area of the conical surface is called  $S$ . If the object is a sphere, then the perimeter is a circle of radius  $R$  ( $P = 2\pi R$ ) and the perpendicular distance from the center to the perimeter is a constant  $D$ , say. Thus,

$$\sin \alpha \equiv R/D = A/S, \quad \text{for spherical surfaces,} \quad (6.7)$$

since  $A = \pi R^2$  and  $S = \pi R D$ . We can now use  $A/S$  in the general definition of  $\bar{\alpha}$  since it has meaning even for distorted surfaces:

$$\bar{\alpha} = \sin^{-1}(A/S), \quad \text{for distorted surfaces.} \quad (6.8)$$

$\bar{\alpha}$  is a continuous variable going from 0 to  $\pi/2$  and then from  $\pi/2$  to 0 as the slice moves from the top of the object to the middle to the bottom. It remains constant as the electron rotates in its orbit. Such a choice for  $\bar{\alpha}$  is not unique, but it shows that at least a consistent definition is possible. A complete set of functions can then be constructed as in (6.6) with  $\alpha$  replaced by  $\bar{\alpha}$ . Open-orbit effects will be accommodated by  $n_2 = 0$  in (6.6b), and the usual results (spherical surfaces) will appear when  $n_1 = n_2 = 1$  and  $\alpha = \bar{\alpha}$ . We are still a long way from detailed computations involving distorted surfaces, but we can at least formally write out the expansion. (As an approximation, one might use any averaged  $\alpha$ .)

Equation (6.6) will lead to a ratio of determinants as in (6.3) where the rows and columns of  $\mathfrak{D}$  are labeled  $(n_1 n_2 n_3) = (000), (001), \dots, (010), (011), \dots, (100), (101), \dots$ , etc.

A "simple" interpolation formula can be obtained for  $\sigma_{xx}$ , ( $x \neq z, x' \neq z$ ) by choosing  $n_1 = 1$ , taking the two

possibilities  $n_3=0, 1$ , and allowing  $n_3=0$ . (The  $n_3$  choice corresponds to the usual electrical conductivity energy dependence assumption.)

$$\sigma_{xx'} = - \frac{\begin{vmatrix} \mathfrak{D} & A_{100}^{(x')} \\ A_{100}^{(x)} A_{110}^{(x)} & 0 \end{vmatrix}}{\begin{vmatrix} L_{100,100}^{(xx')} & L_{110,100}^{(xx')} \\ L_{100,110}^{(xx')} & \mathfrak{L}_{110,110}^{(xx')} \end{vmatrix}}, \quad xx' \neq z. \quad (6.9)$$

Certain simplifications have been made. First  $M$  operating on  $\varphi_{1,0,0}^{(x)}$  is zero whence [see (3.2)]

$$\mathfrak{L}_{n_1 n_2 n_3, n_1' 0 n_3'}^{(xx')} = L_{n_1 n_2 n_3, n_1' 0 n_3'}^{(xx')}, \quad (6.10)$$

and similarly for  $n_2$  and 0 exchanged, for  $\mathfrak{L}$  is symmetric. Also for the same reason  $\bar{A}_{n_1 0 n_3}^{(x)} = A_{n_1 0 n_3}^{(x)}$  [see (6.2)].

In the limit of high-magnetic fields, the terms in  $\mathfrak{L}$  get large as  $H^2$ , and hence will dominate. Thus, multiplying out the determinants in (6.9), we get cancellation, with the result ( $x'=x$ )

$$\lim_{H \rightarrow \infty} \sigma_{xx} = [A_{100}^{(x)}]^2 / L_{100,100}^{(xx)}, \quad xx' \neq z \text{ (open orbits)}, \quad (6.11)$$

which is the variational expression for this limit. If there are no open orbits, then  $A_{100}^{(x)} = 0$ , and this result does not hold. We get instead, from (6.9)

$$\lim_{H \rightarrow \infty} \sigma_{xx} = A_{110}^{(x)} \bar{A}_{110}^{(x)} / \mathfrak{L}_{110,110}^{(xx)}, \quad (6.12)$$

which is the variational expression in this case corresponding results hold for  $x \neq x'$ . We have ignored possible vanishing of terms because of symmetry or other reasons, and have limited the discussion to  $xx' \neq z$ . For intermediate situations ( $H \rightarrow \infty$ ), Eq. (6.9) as it stands should give an indication of how things go. As more and more angular terms are deemed necessary, more and more rows and columns are added to the determinants. For a given number, however, the variational principle provides the "best" expression.

In this way, we can see how the variational principle leads to interpolation formulas. The real difficulty has of course not been avoided; namely, the "hard pounding" (to use Ziman's phrase<sup>18</sup>) that must accompany any detailed calculation.

It should be noted that in these results, we have not assumed that  $L$  could be represented by a time of relaxation. Note also that the matrix element of  $\mathfrak{L}$  in (6.12) involves the inverse operator  $L^{-1}$ . Such integrals may be handled by the method of Appendix C. The results here may be compared with Tsuji's work<sup>4</sup> (which uses the ordinary variational principle and has not considered the large  $H$  limit).

## 7. THEORY OF MAGNON-DRAG

All the results of the preceding sections can be taken over for the situation in which the scattering is by spin waves, and this leads directly to magnon drag in the thermoelectric power. The analogy between magnons and phonons is not, strictly speaking, exact, because (1) double-magnon processes occur in first order in the spin-wave treatment, whereas they do not appear in the corresponding order of electron-phonon interactions; and (2) the electron energy depends on spin for the former, but not for the latter. Double-phonon processes have been considered by Franzak and the author,<sup>19</sup> and the corresponding analysis for magnons can be fashioned by analogy, with the result that double-magnon processes appear to be negligible at low temperatures. Thus the usual treatment of electron-phonon processes and that for magnon-electron processes differ only in that the former do not involve spin flips, whereas the latter do, and that in the latter the one-electron energies depend on spin.<sup>20</sup> We shall develop the analogy in this section.

The electron-phonon interactions  $V_{\text{el-ph}}$  and electron-magnon interactions  $V_{\text{el-mag}}$  arise from the perturbation of the Coulomb and exchange terms in the Hartree-Fock equation, which when converted to second quantized short-hand notation are<sup>21</sup>:

$$V_{\text{el-ph}} = \sum_l [v_{\text{ion}}(\mathbf{r}-\mathbf{R}(l)) - v_{\text{ion}}(\mathbf{r}-\mathbf{R}(l))] \quad (7.1a)$$

$$\rightarrow \sum_{s\mathbf{k}\mathbf{k}'\sigma} c_{\mathbf{k}s} c_{\mathbf{k}'s}^\dagger [a_j(\sigma) V_{\mathbf{k}\mathbf{k}',j\sigma} + a_j(\sigma)^\dagger V_{\mathbf{k}\mathbf{k}',j\sigma}^*] + \sum_{s\mathbf{k}\mathbf{k}'j'\sigma'} c_{\mathbf{k}s} c_{\mathbf{k}'s}^\dagger [a_j(\sigma) a_{j'}(\sigma') V_{\mathbf{k}\mathbf{k}',j\sigma j'\sigma'} + \dots], \quad (7.1b)$$

$$V_{\text{el-mag}} = \sum_l v_{\text{ex}}(\mathbf{r}-\mathbf{R}(l)) = -2 \sum_l \mathfrak{S} \cdot \mathbf{S}_l J(\mathbf{r}-\mathbf{R}(l)) \quad (7.2a)$$

$$\rightarrow \sum_{s\mathbf{k}\mathbf{k}'\kappa} c_{\mathbf{k}s} c_{\mathbf{k}'s}^\dagger [b(\kappa) J_{\mathbf{k}\mathbf{k}',\kappa}^{ss'} + b(\kappa)^\dagger J_{\mathbf{k}\mathbf{k}',\kappa}^{ss'*}] + \sum_{\mathbf{k}\mathbf{k}'\kappa\kappa'} J_{\mathbf{k}\mathbf{k}',\kappa\kappa'} b(\kappa) b(\kappa')^\dagger [c_{\mathbf{k}+} c_{\mathbf{k}'+}^\dagger - c_{\mathbf{k}-} c_{\mathbf{k}'-}^\dagger]. \quad (7.2b)$$

<sup>18</sup> Reference 1, p. 512.

<sup>19</sup> E. Franzak and M. Bailyn, *Bull. Am. Phys. Soc.* **5**, 280 (1960), and see article elsewhere in this issue. [Also an application of a similar procedure is given in M. Bailyn, *Phys. Rev.* **121**, 1336 (1961).]

<sup>20</sup> Kasuya [*Progr. Theoret. Phys. Japan* **22**, 227 (1959)] has also considered the problem of scattering by spin waves. He was not looking for magnon-drag, however.

<sup>21</sup> For spin waves see S. V. Vonsovskii and E. A. Turov, *Zhur. Exptl. i Teoret. Fiz.* **24**, 419 (1953); and A. I. Akhiezer, V. G. Bar'yakhtar, and M. I. Kaganov, *Soviet Phys.—Uspekhi* **3**, 567 (1961).



The  $c_{ks}$ 's are creation operators for the electrons, the  $a$ 's for the phonons and  $b$ 's for the magnons. The wave vectors  $\mathbf{k}$ ,  $\sigma$ ,  $\kappa$  refer to the electrons, phonons, and magnons, respectively. The phonons are also distinguished by a polarization index  $j$  ( $j=1, 2, 3$ ). The double-phonon effects appear in the last member of (7.1b), where the term written out involves the destruction of two phonons, and the dots indicate that there are corresponding expressions for a creation-destruction pair and for two creations. The double-magnon terms in the last member of (7.2b) arise from the  $z$  component of the  $\mathbf{s} \cdot \mathbf{S}$  spin vector dot product. The above expressions are not all-inclusive. For example, we have left out processes in which a magnon and a phonon are emitted simultaneously, and in which three or more of either magnons or phonons are emitted or absorbed.  $\ell$  is the ion position index.

The single-magnon processes are immediately analogous to the single-phonon processes provided the replacement [see (A10)]<sup>21</sup>

$$V_{\mathbf{k}\mathbf{k}'j\sigma} \rightarrow J_{\mathbf{k}\mathbf{k}',\kappa} \left( \frac{2S}{N} \right)^{\frac{1}{2}} \int \psi_{\mathbf{k}'s'}^* J(\mathbf{r}) \psi_{\mathbf{k}s} d^3r, \quad (7.3)$$

is made, and provided the sum over polarization  $j$  is neglected. Thus, all we have to do to convert to magnon effects is to use (7.3) in (A8) and (A9), and allow for spin flips. But the double-magnon processes must be shown to be negligible: This is done in Appendix G. Finally, from the  $z$  component of the  $\mathbf{s} \cdot \mathbf{S}$  product in (7.2), there arises a difference between the one-electron energies of up-spin electrons and those of down-spin electrons (up and down relative to the spin-wave description, i.e., the axis of alignment) which can be written<sup>22</sup>

$$E_{\pm}(\mathbf{k}) = E(\mathbf{k}) - \frac{1}{2}(1 \mp \mu)J_{\mathbf{k}\mathbf{k}}, \quad (7.4)$$

where  $J_{\mathbf{k}\mathbf{k}}$  is the diagonal matrix element, and  $\frac{1}{2}SN\mu$  is the total spin of the ion system (of the  $d$  electrons, that is, which provide the spin waves), where  $\mathbf{S}$  is the spin of one ion, and  $N$  the number of ions. Thus  $\frac{1}{2}N(1-\mu)$  is the number of magnons.

The consequence of the last point is for example that when the variational principle is applied, the expansion functions must take into account the difference in spin. Such a situation has in fact been worked out in a previous work.<sup>23</sup> The result is that for a certain number of rows and columns in the determinant  $D$  [see Eq. (5.5)] corresponding to an approximation in the phonon problem, we must now have twice as many, because of spin. The details are rather involved, and we shall content ourselves with merely making the reference.

We shall complete this section by writing out the magnon-drag contributions  $S^{(\text{mag})}$  to the thermoelectric power. The expression is an example of a quantity

completely analogous to the corresponding phonon-drag one.<sup>24</sup> [The latter can be found for example in Eqs. (H2)–(H7) of TM2.<sup>16</sup>]

$$S^{(\text{mag})} = \frac{1}{T\alpha_0} \frac{m_0}{3\Delta\hbar} \sum_{\kappa} \frac{dN_0}{dz} \mathbf{v}(\kappa) \times \sum_{\mathbf{k}\mathbf{k}'s's'}^{[\kappa]} [\mathbf{v}(\mathbf{k}s) - \mathbf{v}(\mathbf{k}'s')] \alpha(\kappa; \mathbf{k}s\mathbf{k}'s'). \quad (7.5)$$

Here  $N_0(\omega(\kappa))$  is the equilibrium distribution function of the magnons,  $\Delta$  is the crystal volume,  $\mathbf{v}(\kappa)$  the magnon velocity  $\nabla_{\kappa} E(\kappa)$ ,  $z = \hbar\omega(\kappa)/\kappa T$ ,

$$\alpha_0 = -\frac{1}{\Delta} \sum_{\mathbf{k}} \sum_s |e| m \hbar^{-1} [v_x(\mathbf{k}, s)]^2 \frac{\partial f_0}{\partial E} = \frac{|e|}{\hbar\Delta_0}, \quad (7.6)$$

and  $\alpha(\kappa; \mathbf{k}s\mathbf{k}'s')$  is the relative probability that a magnon  $\kappa$  will interact via the electron process  $\mathbf{k}s \rightarrow \mathbf{k}'s'$ . The expression for it is<sup>25</sup>

$$\alpha(\kappa; \mathbf{k}s\mathbf{k}'s') = \frac{N^{-1} D_{\mathbf{k}s\mathbf{k}'s'}^{(m)} \Omega(-) \delta(-)}{-\frac{dN_0}{dz} \frac{1}{\kappa T \tau(\mathbf{K})} + \sum_{\mathbf{k}\mathbf{k}'s's'}^{[\kappa]} N^{-1} D_{\mathbf{k}s\mathbf{k}'s'}^{(m)} \Omega(-) \delta(-)}, \quad (7.7)$$

where

$$D_{\mathbf{k}s\mathbf{k}'s'}^{(m)} = \frac{|J_{\mathbf{k}ss'\mathbf{k}'}|^2 f_0(\epsilon) - f_0(\epsilon')}{\kappa T |e^{-\epsilon} - e^{-\epsilon'}|}, \quad (7.8)$$

$$\epsilon = \frac{E_s(\mathbf{k}) - \zeta}{\kappa T}, \quad (7.9)$$

$$\Omega(-) \rightarrow 2\pi\hbar\delta(E' - E - \hbar\omega(\kappa)), \quad (7.10)$$

$$\delta(-) = \delta(\mathbf{k}' - \mathbf{k} - \kappa - \mathbf{K}), \quad (7.11)$$

and where, in (7.7),  $\tau(\kappa)$  is the relaxation time for the  $\kappa$  magnons involving all processes which do not involve electrons (such as magnon-magnon and magnon-phonon processes). Since the difference in energy in (7.4) is independent of  $\mathbf{k}$ , and since we may expect that the difference at the Fermi level may be accommodated by a change in  $k$  magnitude that will not upset the usual geometry in  $k$  space,<sup>26</sup> we may adapt the free-electron approximation as

$$v_x(\mathbf{k}s) \rightarrow (\hbar/m^*)k_x, \quad (7.12)$$

where  $m^*$  is spin independent. Thus the square bracket in (7.5) is approximately the same for all processes

<sup>24</sup> M. Bailyn, Phil. Mag. **5**, 1059 (1960). See also TM1, reference 16.

<sup>25</sup> The notation on the right-hand side here resembles that of TM1 and TM2 of reference 16. See also reference 24.

<sup>26</sup> Namely that an electron is scattered to and from approximately the same energy surface. This refers to the *geometry* in  $k$  space, and does not imply an elastic collision approximation elsewhere.

<sup>22</sup> See S. V. Vonsovskii and E. A. Turov, reference 21, Eq. (23).

<sup>23</sup> M. Bailyn, "Transport in Metals with Magnetic Impurities," Westinghouse Research Report 029-B000-P1.

involving the same reciprocal lattice vector (for all processes of a given umklapp type, to use the term employed in TM1). As a result, the same preliminary sum over  $kk'$  can be employed that was used in reference 24, and we end up with

$$S^{(\text{mag})} = \frac{1}{T\alpha_0} \frac{1}{3\Delta} \sum_{\kappa} \frac{dN_0}{dz} \frac{m_0}{m^*} \times \sum_{\mathbf{K}} (\mathbf{\kappa} + \mathbf{K}) \frac{\tau_{\mathbf{K}}(\mathbf{\kappa})^{-1}}{\tau(\mathbf{\kappa})^{-1} + \sum_{\mathbf{K}'} \tau_{\mathbf{K}'}(\mathbf{\kappa})^{-1}}, \quad (7.13)$$

where

$$\tau_{\mathbf{K}}(\mathbf{\kappa})^{-1} = (3\pi/2) S [\zeta_0^{-2} \langle |J_{\mathbf{K}\mathbf{K}'}^{ss'}|^2 \rangle_{\kappa}] \times [k_0/|\mathbf{\kappa} + \mathbf{K}|] \omega(\mathbf{\kappa}), \quad (7.14)$$

where  $S$  is the spin of the individual ion,  $\zeta_0$  is given by (A13), and the square brackets are designed to be dimensionless. The angle bracket is an average over  $\mathbf{K}\mathbf{K}'$  for a given  $\mathbf{\kappa}$  and  $\mathbf{K}$ . (Clear distinctions among the magnon wave vector  $\mathbf{\kappa}$ , the reciprocal lattice vector  $\mathbf{K}$ , the electron wave vector  $\mathbf{k}$ , and the Boltzmann constant  $\kappa$  are casualties of the notation.)

This result is identical with that for phonon drag, and will yield for example anomalous signs for the Umklapp processes. Thus to distinguish magnon drag from phonon drag will in general be difficult. Our experience with phonon drag in metals is that at best, the theory can be shown not to be inconsistent with experiment, all predictions being in the realm of guess work since the actual numerical result is a small difference between large terms of different sign, each rather sensitive to impurity content, etc. (The small difference may however be large compared to the diffusion component.) In the case of magnon drag, we merely point out that the magnon-magnon relaxation time has been estimated to be within an order of magnitude of the magnon-electron relaxation time,<sup>27</sup> which indicates the possi-

bility that  $S^{(\text{mag})}$  might actually play a role in the thermoelectric power of some substances.

## 8. SUMMARY

We have attempted in this paper to formulate a maximum variational principle for conduction problems in the presence of a magnetic field, including phonon-drag effects. We have shown that for symmetry considerations the operator  $\mathfrak{L} = L - ML^{-1}M$  is the important one, not  $L + M$ . Methods for the high-magnetic field expansions have been indicated, and the limiting expressions for the drag effects in the thermoelectric power have been discussed. Finally, magnon-drag effects have been shown to be analogous to phonon-drag effects.

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## APPENDIX A. THE BOLTZMANN EQUATION

The method of taking into account the nonequilibrium component of the phonons by solving first the phonon Boltzmann equation and substituting the result into the electron Boltzmann equation has been shown in TM1.<sup>16</sup> The only extension required for this paper is the inclusion of a magnetic field streaming term. However, in order to have expressions which will convert to the spin-wave scattering problem (Sec. 7) with no trouble, it is desirable to rewrite some of the steps in a new notation.

The Boltzmann equations for the electrons and phonons are

$$-\left(\frac{\partial f}{\partial t}\right)_{\text{drift}} = \left(\frac{\partial f}{\partial t}\right)_{\text{coll}} = \sum_j \sum_{\mathbf{k}'s'} \{ [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) + P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] [g(\mathbf{k}'s') - g(\mathbf{k}s)] + \sum_j G_j(\boldsymbol{\sigma}) [P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s) - P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s)] \}, \quad (A1)$$

$$-\left(\frac{\partial N}{\partial t}\right)_{\text{drift}} = \left(\frac{\partial N}{\partial t}\right)_{\text{coll}} = G_j(\boldsymbol{\sigma}) \lambda_j(\boldsymbol{\sigma})^{-1} + \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}'}^{[j\sigma]} [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) - P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] [g(\mathbf{k}'s') - g(\mathbf{k}s)], \quad (A2)$$

where

$$\frac{1}{\lambda_j(\boldsymbol{\sigma})} = -\frac{\partial N_0}{\partial \hbar\omega} \frac{1}{\tau(j\boldsymbol{\sigma})} + \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}'}^{[j\sigma]} [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) + P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] \geq 0, \quad (A3)$$

and where  $N(j\boldsymbol{\sigma}) = N_0 - [\partial N_0 / \partial \hbar\omega] G_j(\boldsymbol{\sigma})$ . The phonons are described by a wave vector  $\boldsymbol{\sigma}$ , a polarization index

<sup>27</sup> E. Abrahams, Phys. Rev. **98**, 587 (1955), has estimated the electron-magnon relaxation time at  $10^{-8}$  sec, and C. Kittel and E. Abrahams, Revs. Modern Phys. **25**, 233 (1953), have estimated magnon-magnon relaxation at  $\sim 10^{-9}$  sec.

$j (= 1, 2, 3)$  and a frequency  $\omega_j(\boldsymbol{\sigma})$ .  $\tau(j\boldsymbol{\sigma})$  is the phonon-relaxation time involving all processes except the electron-phonon ones. The sums as in (A3) are defined in TM1, and mean that all processes are summed over that pertain to a particular type of phonon ( $j\boldsymbol{\sigma}$ ). The

drift terms are

$$\left(\frac{\partial f}{\partial t}\right)_{\text{drift}} = [e|\mathbf{v} \cdot \mathbf{X} - (E - \zeta)\mathbf{v} \cdot \mathbf{Y}] \frac{\partial f_0}{\partial E} + M(g), \quad (\text{A4})$$

$$(\partial N / \partial t)_{\text{drift}} = -(\partial N_0 / \partial \hbar \omega) \hbar \omega \nabla_{\sigma} \omega \cdot \mathbf{Y} \quad (\text{A5})$$

where  $\mathbf{X}$  and  $\mathbf{Y}$  are defined by (2.4). The  $P$ 's are

$$P_j^{(a)}(\mathbf{k}'s; \mathbf{k}s) = (\kappa T)^{-1} W_j^{(a)}(\mathbf{k}s \rightarrow \mathbf{k}'s) f_0(1 - f_0') \times \delta[E' - E - \hbar \omega_j(\sigma)], \quad (\text{A6})$$

$$P_j^{(e)}(\mathbf{k}'s; \mathbf{k}s) = (\kappa T)^{-1} W_j^{(e)}(\mathbf{k}s \rightarrow \mathbf{k}'s) f_0(1 - f_0') \times \delta[E' - E + \hbar \omega_j(\sigma)], \quad (\text{A7})$$

$$W_j^{(a)}(\mathbf{k}s \rightarrow \mathbf{k}'s) = 2\pi \hbar^{-1} |V_{\mathbf{k}\mathbf{k}',j\sigma}|^2 N(j\sigma), \quad (\text{A8})$$

$$W_j^{(e)}(\mathbf{k}s \rightarrow \mathbf{k}'s) = 2\pi \hbar^{-1} |V_{\mathbf{k}\mathbf{k}',j\sigma}|^2 [N(j\sigma) + 1], \quad (\text{A9})$$

$$V_{\mathbf{k}\mathbf{k}',j\sigma} = \delta(\mathbf{k} - \mathbf{k}' + \sigma + \mathbf{K}) [2MN\omega_j(\sigma)/\hbar]^{-\frac{1}{2}} \times \mathbf{e}_j(\sigma) \cdot \int \psi_{\mathbf{k}s} \psi_{\mathbf{k}'s}^* \nabla d^3r. \quad (\text{A10})$$

The  $N(j\sigma)$  of (A8) and (A9) are the phonon-occupation numbers;  $\mathbf{e}_j(\sigma)$  is the unit vector in the direction of polarization of the  $j\sigma$  phonons;  $M$  is ion mass;  $v(r)$  is the potential from one ion at  $\mathbf{r}=0$ ; and  $\mathbf{K}$  is a reciprocal lattice vector or zero. The  $P$ 's satisfy

$$P_j^{(a)}(\mathbf{k}'s; \mathbf{k}s) = P_j^{(e)}(\mathbf{k}s; \mathbf{k}'s). \quad (\text{A11})$$

For order-of-magnitude estimates (which we need later in Appendix G), it is sometimes useful to write the square root in (A10) as

$$\left[\frac{\hbar}{2M\omega_j(\sigma)}\right]^{\frac{1}{2}} = \left[\frac{\zeta_0}{\hbar\omega_j(\sigma)} \frac{m^*}{M}\right]^{\frac{1}{2}} k_0, \quad (\text{A12})$$

where  $\zeta_0$  is the Fermi energy at  $T=0$  and can be written

$$\zeta_0 = \hbar^2 k_0^2 / 2m^*, \quad (\text{A13})$$

$$k_0^3 = 3\pi^2 N / \Delta,$$

where  $\Delta$  is the crystal volume.

When (A2) is solved for  $G$  and substituted into (A1), the equation (2.2) results, where

$$L(g) = L^{\text{ph}}(g) + L^{\text{ph-drag}}(g), \quad (\text{A14})$$

$$L^{\text{ph}}(g) = \sum_{\mathbf{k}'s'j} [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) + P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] [g(\mathbf{k}'s') - g(\mathbf{k}s)], \quad (\text{A15})$$

$$L^{\text{ph-drag}}(g) = \sum_{\mathbf{k}'s'j} [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) - P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] \lambda(j\sigma) \Gamma_{j\sigma}(g), \quad (\text{A16})$$

where in (A16)

$$\Gamma_{j\sigma}(g) = -\frac{1}{2} \sum_{\mathbf{k}\mathbf{k}'s's'}^{[j\sigma]} [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) - P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] \times [g(\mathbf{k}'s') - g(\mathbf{k}s)] = \sum [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) - P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] g(\mathbf{k}'s'). \quad (\text{A17})$$

And also in (2.2), or rather in (2.5b),

$$\gamma(\mathbf{k}s) = - \sum_{\mathbf{k}'s'j} [P_j^{(a)}(\mathbf{k}'s'; \mathbf{k}s) - P_j^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] \times \lambda(j\sigma) \frac{\partial N_0}{\partial \hbar \omega} \hbar \omega \nabla_{\sigma} \omega. \quad (\text{A18})$$

In order to convert to the spin-wave problem (see Appendix G), all that has to be done is to replace the  $V$  of (A10) by the spin wave  $J$ , and to keep this replacement in mind in the  $W$ 's of (A8) and (A9), and then in the  $P$ 's of (A6) and (A7). Then the Boltzmann equation (2.2) reads precisely the same, except that the spin-wave description  $\mathbf{k}$  replaces the phonon description  $\sigma j$ .

## APPENDIX B. SYMMETRY PROPERTIES OF THE OPERATORS

We define integrals

$$(h, g)_O = - \sum_{\mathbf{k}} h(\mathbf{k}) O(g), \quad (\text{B1})$$

of arbitrary functions  $h$  and  $g$ , for the operator  $O$ . The functions  $h$  and  $g$  will all be assumed to be periodic in the repeated zone scheme in  $\mathbf{k}$  space. It is well known that  $L^{\text{ph}}$  satisfies the symmetry relations,<sup>6,16</sup>

$$(h, g)_O = (g, h)_O, \quad (\text{B2a})$$

$$(h, h)_O \geq 0, \quad (\text{B2b})$$

and it was shown in TM1 that  $L(g)$  [see (A14)] also does. It is also known that  $M$  satisfies the antisymmetrical relation,<sup>14,15</sup>

$$(h, g)_M = -(g, h)_M, \quad (\text{B3})$$

which corresponds to changing the sign of the magnetic field on the right-hand side of the equation.

We wish now to prove that the operator  $\mathcal{L}$  of (3.2) satisfies (B2). By successive use of the relations (B2) (valid for  $L$ ) and (B3) (valid for  $M$ ), we get

$$\sum_{\mathbf{k}} g [-ML^{-1}M(h)] = \sum_{\mathbf{k}} [M(g)] L^{-1} [M(h)] = \sum_{\mathbf{k}} [M(h)] L^{-1} [M(g)], \quad (\text{B4})$$

since if  $L$  is symmetric, so is  $L^{-1}$ . But the quantity in (B4) is symmetrical in  $g$  and  $h$  (since  $L^{-1}$  is symmetric) and is positive definite [i.e., satisfies (B2b)] when  $g=h$  (since  $L^{-1}$  is positive definite if  $L$  is). Thus,  $-ML^{-1}M$  satisfies (B2); but  $L$  does also. Hence the sum, which is  $\mathcal{L}$ , must satisfy (B2). This completes the proof. It follows also that  $\mathcal{L}^{-1}$  satisfies (B2).

We wish finally to prove that  $\mathcal{L}^{-1}ML^{-1}$  satisfies (B3). By successive manipulations as in (B4), it is easy to show that

$$\sum_{\mathbf{k}} g \mathcal{L}^{-1} M L^{-1}(h) = -\sum_{\mathbf{k}} h L^{-1} M \mathcal{L}^{-1}(g). \quad (\text{B5})$$

But

$$L^{-1} M \mathcal{L}^{-1} = L^{-1} M (1 - L^{-1} M L^{-1} M)^{-1} L^{-1}, \quad (\text{B6})$$

[using  $(AB)^{-1} = B^{-1}A^{-1}$ ]. But  $L^{-1}M$  commutes with functions of  $L^{-1}M$ . Thus (B6) becomes

$$(1 - L^{-1} M L^{-1} M)^{-1} L^{-1} M L^{-1} = \mathcal{L}^{-1} M L^{-1}, \quad (\text{B7})$$

which when substituted in (B5) completes the proof.

### APPENDIX C. SUMS INVOLVING INVERSE OPERATORS

We consider sums of the form

$$(\phi, q)_{L^{-1}} = -\sum_{\mathbf{k}} \phi(\mathbf{k}) L^{-1}(q). \quad (\text{C1})$$

To know  $L^{-1}$  is to know the solution to a Boltzmann equation. In fact each sum as in (C1) is analogous to a current flow, and can be calculated by a separate variational principle, adaptable to the particular  $\phi$  and  $q$  of (C1).

The corresponding Boltzmann equation is for an unknown, say  $h(k)$ , and reads

$$L(h) = q(\mathbf{k}). \quad (\text{C2})$$

The solution is

$$h = L^{-1}(q), \quad (\text{C3})$$

and the current corresponding to the flux  $\phi$  is then precisely (C1). In fact (C2) is analogous to (2.2) of the text, (C3) is analogous to (2.8), and (C1) is analogous to (2.9) using (2.10). The analogy is complete, and the variational solutions (5.5) are applicable at once. These require a complete set of functions  $\varphi_n$ , which as noted before may be chosen to fit most readily the functions of  $\phi$  and  $q$  of (C1). Thus the sum in (C1) can be written

$$(\phi, q)_{L^{-1}} = -D^{(\varphi)}(\phi, q)/D^{(\varphi)}, \quad (\text{C4})$$

where we have placed an index  $\varphi$  on the  $D$ 's in order to specify that the solution is in terms of the expansion functions  $\varphi_n$ . Of course the complete solution will be independent of which set we choose, but an approximate solution (one or two rows or columns in  $D$ , for example) will depend on which functions we have chosen. The elements of  $D$  above are  $d_{nm}$ , where

$$d_{nm} = -\sum_{\mathbf{k}} \varphi_n(\mathbf{k}) L(\varphi_m) = d_{mn}, \quad (\text{C5a})$$

and the other integrals are

$$\phi_n = \sum_{\mathbf{k}} \varphi_n(\mathbf{k}) \phi(\mathbf{k}). \quad (\text{C5b})$$

As an example of how this works, consider evaluating  $(g, h)_{\mathcal{L}}$

$$\sum_{\mathbf{k}} g \mathcal{L}(h) = \sum_{\mathbf{k}} g L(h) + \sum_{\mathbf{k}} [M(g)] L^{-1}[M(h)], \quad (\text{C6})$$

where we used (B4). The first term involving  $L$  can be evaluated by integration (in principle), but the second

term will be a ratio of determinants of similar integrals:

$$\sum_{\mathbf{k}} [M(g)] L^{-1}[M(h)] = D^{(\varphi)}(M(g), M(h))/D^{(\varphi)}. \quad (\text{C7})$$

The determinant can be expanded in a series, and the series must converge independently of whatever approximation is used that leads to (C6).

### APPENDIX D. CALCULATION OF $\mathbf{W}^{\text{ph}}$

The heat flow of the phonons is, using (A2) for  $G_j(\sigma)$ ,

$$\begin{aligned} \mathbf{W}^{\text{ph}} &= \sum_{j\sigma} \mathbf{v}(j\sigma) \hbar \omega_j(\sigma) \left[ -\frac{\partial N_0}{\partial \hbar \omega} G_j(\sigma) \right] \\ &= \sum_{j\sigma} \mathbf{v}(j\sigma) \hbar \omega_j(\sigma) \left[ -\frac{\partial N_0}{\partial \hbar \omega} \lambda(j\sigma) \right] \left\{ \frac{\partial N_0}{\partial \hbar \omega} \hbar \omega \mathbf{v} \cdot \mathbf{Y} \right. \\ &\quad \left. + \sum_{\mathbf{k}\mathbf{k}'}^{[j\sigma]} [P^{(a)}(\mathbf{k}'s'; \mathbf{k}s) - P^{(e)}(\mathbf{k}'s'; \mathbf{k}s)] g(\mathbf{k}s) \right\}, \quad (\text{D1}) \end{aligned}$$

where  $\mathbf{v}(j\sigma) = \nabla_{\sigma} \omega_j(\sigma)$  is the phonon-group velocity. The first term in square brackets is the drift term in (A2), the second is the electron "reaction" term. The latter can be written in terms of  $\gamma$  of (A18) by employing the relation

$$\sum_{j\sigma} \sum_{\mathbf{k}\mathbf{k}'}^{[j\sigma]} = \sum_{\mathbf{k}\mathbf{k}'j}. \quad (\text{D2})$$

[See Eq. (21) of TM1.] Thus we may write

$$\begin{aligned} \mathbf{W}_i^{\text{ph}} &= \sum_j T_{\kappa ij} Y_j + \sum_{\mathbf{k}s} g(\mathbf{k}s) \gamma_i(\mathbf{k}s) \\ &= \sum_j T_{\kappa ij} Y_j - \sum_{\mathbf{k}s} \gamma_i(\mathbf{k}s) \frac{1}{L+M} [\mathbf{A} \cdot \mathbf{X} + \mathbf{B} \cdot \mathbf{Y}], \quad (\text{D3}) \end{aligned}$$

using (2.8), where

$$T_{\kappa im} = -\sum_{j\sigma} [v(j\sigma)]_i [v(j\sigma)]_m (\hbar \omega)^2 \left( \frac{\partial N_0}{\partial \hbar \omega} \right) \lambda(j\sigma). \quad (\text{D4})$$

### APPENDIX E. MATRIX ELEMENTS OF $1/(L+M)$ , ASSUMING A RELAXATION TIME

From (5.9), we get for the first few terms

$$\begin{aligned} \left[ \frac{1}{L+M} \right]_{mn} &= \frac{1}{M'_n} \delta_{n,-m} - \frac{1}{M'_m} L'_{mn} \frac{1}{M'_n} \\ &\quad + \sum_{n'} \frac{1}{M'_m} L'_{mn'} \frac{1}{M'_{n'}} L'_{n'n} \frac{1}{M'_{n'}} \\ &\quad - \sum_{n'n''} \frac{1}{M'_m} L'_{mn'} \frac{1}{M'_{n'}} L'_{n'n''} \frac{1}{M'_{n''}} L'_{n''n}. \quad (\text{E1}) \end{aligned}$$

We are interested in the limit  $H \rightarrow \infty$ . Therefore, from (5.13), we desire the  $n$  and  $n''$  sums in (E1) to contain as few  $1/M'_n$ ,  $n \neq 0$ , as possible. However, if two succes-

sive  $M_n$ 's in (E1) have  $n=0$ , the sandwiched-in matrix element of  $L'$  must be  $L'_{00}$  which is zero. This simplifies things enormously, and the result is (5.14) for the lowest power of  $1/H$ . From this, we were able to write (5.15) for the largest terms in the limit  $H \rightarrow \infty$ . We list below the next largest terms:

$$\frac{1}{M'_0} \left[ \sum_{n' \neq 0} L'_{0n'} \frac{1}{M'_{n'}} L'_{n'0} - \sum_{n' \neq 0} \sum_{n'' \neq 0} L'_{0n'} \frac{1}{M'_{n'}} L'_{n'n''} \frac{1}{M'_{n''}} L'_{n''0} \right] \frac{1}{M'_0} \sim \frac{1}{H}, \quad (\text{E2a})$$

$$\frac{1}{M'_0} \sum_{n' \neq 0} L'_{0n'} \frac{1}{M'_{n'}} L'_{n'n} \frac{1}{M'_n} \sim \frac{1}{H^2}, \quad (\text{E2b})$$

$$\frac{1}{M'_n} \sum_{n' \neq 0} L'_{nn'} \frac{1}{M'_{n'}} L'_{n'0} \frac{1}{M'_0} \sim \frac{1}{H^2}, \quad (\text{E2c})$$

$$-\frac{1}{M'_n} \left[ L'_{nn} - L'_{n0} \frac{1}{M'_0} L'_{0n} \right] \frac{1}{M'_n} \sim \frac{1}{H^2}. \quad (\text{E2d})$$

Here, (E2a) corresponds to (5.15a), (E2b) to (5.15b), etc.

#### APPENDIX F. MATRIX ELEMENTS OF $1/(L+M)$ , GENERAL

When a time of relaxation does not exist, difficulties arise because  $L$  is not a function, and the  $\varphi_n$  of (5.6) do not form a complete set for all the variables  $t, k_z, E$ . We would like to be able to use the simplification that arose in the previous Appendix, namely that  $L_{00}'=0$ . But if the  $\varphi_n$ 's of (5.6) are used, and  $L$  is an operator acting on  $t, k_z$ , and  $E$ , then the meaning of the matrix element  $L_{nn}'$  defined in (5.11) is rather obscure.

The difficulty can be overcome by introducing projection operators  $P_n$  which operate on any function  $Q(t, k_z, E)$ ,

$$Q(t, k_z, E) = \sum_n Q_n(k_z E) \varphi_n(t), \quad (\text{F1})$$

as follows:

$$P_n Q = Q_n(k_z E) \varphi_n(t). \quad (\text{F2})$$

(These  $P_n$ 's are the operator counterparts of the  $kk'$  indices on  $W_{kk'}$  in Sec. 5 of reference 12.) Clearly

$$\sum_n P_n = 1, \quad (\text{F3})$$

where 1 is the unit operator. Now from (A14), we can write

$$L(Q) = \sum_n L P_n(Q). \quad (\text{F4})$$

Thus in a formal way, we can write the operator  $L$  as

$$L = \sum_{nn'} L_{nn'}, \quad (\text{F5a})$$

$$L_{nn'} = P_n^\dagger L P_n, \quad (\text{F5b})$$

where  $P_n^\dagger$  is the Hermitian conjugate of  $P_n$ . The quantity  $C$  in (5.10) is now chosen to be

$$C = \sum_n L_{nn}. \quad (\text{F6})$$

Thus the operator  $1/M'$  is diagonal with respect to the functions  $\varphi_n(t)$  and has the matrix elements (that are still operators)

$$\oint dt \varphi_m \frac{1}{M'} \varphi_n = \oint dt \varphi_m \frac{1}{\sum_{n'} L_{n'n'} + M} \varphi_n = \delta_{n,-m} \oint dt \varphi_{-n} \frac{1}{L_{nn} + M_n} \varphi_n, \quad (\text{F7a})$$

$$\oint dt \varphi_0 \frac{1}{M'} \varphi_0 = \oint dt \varphi_0 \frac{1}{L_{00}} \varphi_0. \quad (\text{F7b})$$

This corresponds roughly to (5.17) of the text. We also have  $L_{nn}'=0$ . The analogy with the time of relaxation situation is then complete.

To evaluate something like (2.10), we proceed as follows:

$$\sigma_{ij} = \sigma_{ij}^{(1)} + \sigma_{ij}^{(2)} + \dots, \quad (\text{F8})$$

where the superscript  $p$  in  $\sigma_{ij}^{(p)}$  indicates how many times  $1/N$  appears;

$$\sigma_{ij}^{(1)} = - \sum_{\mathbf{k}} A_i \frac{1}{M'} A_j \quad (\text{F9a})$$

$$= - \sum_n \sum_{\mathbf{k}} A_{i,-n} \varphi_{-n} \frac{1}{M_n'} A_{jn} \varphi_n, \quad (\text{F9b})$$

$$\sigma_{ij}^{(2)} = \sum_{\mathbf{k}} A_i \frac{1}{M'} L' \frac{1}{M'} A_j \quad (\text{F10a})$$

$$= \sum_{n \neq 0} \left[ \sum_{\mathbf{k}} A_{in} \varphi_n \frac{1}{M_n'} L_{n0}' \frac{1}{L_{00}} A_{i0} \varphi_0 + \sum_{\mathbf{k}} A_{i0} \varphi_0 \frac{1}{L_{00}} L_{0n}' \frac{1}{M_n'} A_{jn} \varphi_n \right] + \sum_{n, n' \neq 0} \sum_{\mathbf{k}} A_{in} \varphi_n \frac{1}{M_n'} L_{nn'}' \frac{1}{M_{n'}} A_{jn'} \varphi_{n'}. \quad (\text{F10b})$$

The sums here may be evaluated by successive applications of the method of Appendix C. As an example, consider the first sum in the square brackets of (F10b). It can be written

$$\sum_{\mathbf{k}} \left( P_n A_{in} \varphi_n \frac{1}{M_n'} \right) L \left( P_0 \frac{1}{L_{00}} A_{i0} \varphi_0 \right), \quad (\text{F11})$$

because  $L_{n0}' = L_{n0}$  [since  $P_n P_{n'} (n' \neq n) = 0$ ]. The problem is what to use for the quantities enclosed in circular

brackets. Suppose we introduce a complete set of functions  $\psi_\mu(\mathbf{k})$ . Then

$$P_0 \frac{1}{L_{00}} A_{i0} \varphi_0 = P_0 \sum_\mu \psi_\mu(\mathbf{k}) U_\mu, \quad (\text{F12})$$

where

$$U_\mu = \sum_{\mathbf{k}} \psi_\mu(\mathbf{k}) \frac{1}{L_{00}} A_{i0} \varphi_0 = -D^{(x)}(\psi_\mu, A_{i0} \varphi_0) / D^{(x)}, \quad (\text{F13})$$

where to evaluate  $U_\mu$  by means of the method of Appendix C, we have introduced another set of expansion functions  $\chi_\mu(\mathbf{k})$ , which may (or may not) be the same as the  $\psi_\mu$ . The elements of  $D^{(x)}$  are

$$d_{\mu\nu} = \sum_{\mathbf{k}} \chi_\mu(\mathbf{k}) L_{00} \chi_\nu(\mathbf{k}). \quad (\text{F14})$$

Eq. (F11) now reads

$$\sum_\mu U_\mu \sum_{\mathbf{k}} (A_{in} \varphi_n) \frac{1}{M_n} [P_0 L(\psi_\mu)], \quad (\text{F15})$$

and the inverse operator here can be taken care of by another application of the method of Appendix C. It is clear that such computations will very quickly become very tedious. In addition, an attempt to make computations must overcome the difficulty that  $v_i(\mathbf{k})$  is not well known for substances with distorted energy surfaces. Nevertheless the method indicated in the Appendix is the natural generalization of the one in Appendix E to the case where a time of relaxation does not exist.

#### APPENDIX G. NEGLECT OF DOUBLE-MAGNON PROCESSES

The effect of double-phonon processes in first-order perturbation theory has been worked out by Franzak<sup>19</sup> for the electrical resistivity. This was done with the alkali metals in mind, in which a large phonon anisotropy exists. The results were calculated numerically in some detail at high temperatures, with the result that the double-phonon processes contributed at most about 4% to the resistivity for the most anisotropic of the alkalis; but probably 1% would be a more representative figure. In the case of double-magnon processes at low temperatures, we have two differences from the above-mentioned calculation: (1) The limit of low temperatures decreases the effects enormously, and (2) the magnon-electron interaction matrix element (7.2) is much larger than the electron-phonon matrix element (7.1). If it were not for (2), there would be no question of the neglect of the double-magnon processes.

We shall here outline the results of the double-phonon calculation. The geometry of an umklapp double-phonon process is shown in Fig. 1. There is a sum over the two phonon wave vectors that ranges over the overlap volume of the  $\sigma$  and  $\sigma'$  spheres. We shall call this volume  $\eta(u)$  times the total volume of the sphere,

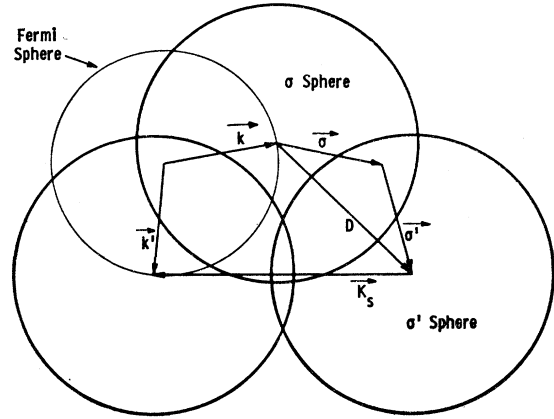


FIG. 1. Two-phonon umklapp transition.

where  $\eta(u)$  is a fraction that depends on  $u = |\mathbf{k} - \mathbf{k}'|/2k_0$ . For simplicity, a free-electron model was used. This enabled the electron part of the matrix elements for the single and double processes to be similar. (A statement of this type of approach has since been made<sup>28</sup> in a more general way.) Thus, the two matrix elements in (7.1b) were taken to be ( $\mathbf{s} = \mathbf{k} - \mathbf{k}'$ )

$$V_{\mathbf{k}\mathbf{k}'j\sigma} = i\mathbf{s} \cdot \boldsymbol{\epsilon}_j(\boldsymbol{\sigma}) [2MN\omega_j(\boldsymbol{\sigma})/\hbar]^{-1/2} V_{\mathbf{k}\mathbf{k}'}, \quad (\text{G1})$$

$$V_{\mathbf{k}\mathbf{k}'j\sigma j'\sigma'} = \frac{1}{2} [i\mathbf{s} \cdot \boldsymbol{\epsilon}_j(\boldsymbol{\sigma})] [i\mathbf{s} \cdot \boldsymbol{\epsilon}_{j'}(\boldsymbol{\sigma}')] \times 2MN[\omega_j(\boldsymbol{\sigma})\omega_{j'}(\boldsymbol{\sigma}')]^{1/2} \hbar^{-1} V_{\mathbf{k}\mathbf{k}'}, \quad (\text{G2})$$

where

$$V_{\mathbf{k}\mathbf{k}'} = \frac{eN}{\Delta} \int e^{-i\mathbf{s} \cdot \boldsymbol{\rho}} v(\boldsymbol{\rho}) d^3\rho. \quad (\text{G3})$$

[See (A10).] In the case of double-magnon processes we have correspondingly  $N^{-1/2} J_{\mathbf{k}\mathbf{k}}$  and  $N^{-1} J_{\mathbf{k}\mathbf{k}}$ . Thus we have order-of-magnitude-wise for the ratio and double-to single-phonon processes:

$$\left| \frac{V_{\mathbf{k}\mathbf{k}'j\sigma j'\sigma'}}{V_{\mathbf{k}\mathbf{k}'j\sigma}} \right|^2 \cong \frac{1}{12} \frac{\hbar^2 s^2}{2M} \frac{1}{\hbar\omega} \frac{1}{N} = \left( \frac{1}{12} \frac{\zeta}{\hbar\omega} \frac{m}{M} \right) \frac{1}{N}, \quad (\text{G4})$$

whereas for the magnons the corresponding ratio is  $1/N$ . The factor in (G4) can be about  $10^{-3}/N$ , so that the double-magnon processes are, all other things being equal, much larger relative to single-magnon processes than are double-phonon processes relative to single.

In the variational principle the electrical resistivity is proportional to the  $d_{00}$  integral:

$$d_{00} = -\sum_{\mathbf{k}} k_x L^{\text{ph}}(k_x). \quad (\text{G5})$$

There will be a single-phonon contribution  $d_{00}^{(1)}$  and a double-phonon contribution  $d_{00}^{(2)}$ , and we shall be interested in the ratio  $d^{(2)}/d^{(1)}$ . Adapting Franzak's

<sup>28</sup> See the second reference in footnote 19.

results,<sup>29</sup> we write this ratio as follows:

$$\frac{d_{00}^{(2)}}{d_{00}^{(1)}} = \frac{\zeta}{\kappa T} \frac{m}{M} \frac{1}{9\pi^5}$$

$$\times \frac{\sum_{ii'} \int du u^7 \eta(u) |V_{\mathbf{k}\mathbf{k}'}|^2 \langle\langle Q_2^{\text{ph}}(jj') \rangle\rangle}{\sum_i \int du u^5 |V_{\mathbf{k}\mathbf{k}'}|^2 \langle Q_1^{\text{ph}}(j) \rangle}, \quad (\text{G6})$$

$$Q_1^{\text{ph}}(j) = \frac{\delta_- + \delta_+}{(e^\gamma - 1)(1 - e^{-\gamma})}, \quad (\text{G7})$$

$$Q_2^{\text{ph}}(jj') = \frac{1}{\gamma\gamma'(e^\gamma - 1)(e^{\gamma'} - 1)} \times \left[ \frac{\gamma + \gamma'}{1 - e^{-\gamma - \gamma'}} (\delta_{++} + \delta_{--}) + \frac{\gamma - \gamma'}{|e^{-\gamma'} - e^{-\gamma}|} (\delta_{+-} + \delta_{-+}) \right], \quad (\text{G8})$$

where

$$\begin{aligned} \gamma &= \hbar\omega_j(\boldsymbol{\sigma})/\kappa T, \quad \gamma' = \hbar\omega_{j'}(\boldsymbol{\sigma}')/\kappa T, \\ \delta_{\pm\pm} &= \delta(\mathbf{k} - \mathbf{k}' \pm \boldsymbol{\sigma} \pm \boldsymbol{\sigma}' + \mathbf{K}_s), \\ \delta_{\pm\mp} &= \delta(\mathbf{k} - \mathbf{k}' \pm \boldsymbol{\sigma} \mp \boldsymbol{\sigma}' + \mathbf{K}_s). \end{aligned} \quad (\text{G9})$$

The four  $\delta$  functions in (G8) correspond to the four possibilities: Two creations, two destructions, a creation and a destruction, and a destruction and a creation. The average over  $Q^{(1)}$  is over all directions of  $\mathbf{k} - \mathbf{k}'$  for a given  $|\mathbf{k} - \mathbf{k}'|$  magnitude. The double-average over  $Q^{(2)}$  is first an average over the overlap region of the  $\boldsymbol{\sigma}$  and  $\boldsymbol{\sigma}'$  spheres in Fig. 1, and second an average over  $\mathbf{k} - \mathbf{k}'$  directions for a given  $|\mathbf{k} - \mathbf{k}'|$  magnitude. The  $\boldsymbol{\sigma}$ 's can approach zero for Umklapp processes here with no trouble.

To convert this to the magnon case, we replace the right-hand side of (G2) by  $N^{-1}J_{\mathbf{k}\mathbf{k}'}$  and the right-hand side of (G1) by  $N^{-\frac{1}{2}}J_{\mathbf{k}\mathbf{k}'}$ . Thus in (G6),  $V_{\mathbf{k}\mathbf{k}'}$  gets re-

placed by  $J_{\mathbf{k}\mathbf{k}'}$  provided  $Q_2$  gets multiplied by

$$\left[ \frac{1}{4} (i\mathbf{s} \cdot \boldsymbol{\epsilon})^2 (i\mathbf{s} \cdot \boldsymbol{\epsilon}')^2 \frac{1}{M^2} \frac{\hbar}{2\omega} \frac{\hbar}{2\omega'} \right]^{-1} \cong 36(\kappa T)^2 \left(\frac{M}{m}\right)^2 \left(\frac{2m}{\hbar^2 s^2}\right)^2 \gamma\gamma', \quad (\text{G10})$$

and  $Q_1$  by

$$\left[ (i\mathbf{s} \cdot \boldsymbol{\epsilon})^2 \frac{1}{M} \frac{\hbar}{2\omega} \right]^{-1} \cong 3\kappa T \frac{M}{m} \left(\frac{2m}{\hbar^2 s^2}\right) \gamma, \quad (\text{G11})$$

whence

$$\frac{d_{00}^{(2)}}{d_{00}^{(1)}} \Big|_{\text{magnon}} \cong \frac{1}{3\pi^5} \frac{\int du u^3 \eta(u) |J_{\mathbf{k}\mathbf{k}'}|^2 \langle\langle Q_2^{(m)} \rangle\rangle}{\int du u^3 |J_{\mathbf{k}\mathbf{k}'}|^2 \langle Q_1^{(m)} \rangle}, \quad (\text{G12})$$

where

$$Q_1^{(m)} = \gamma \frac{\delta_- + \delta_+}{(e^\gamma - 1)(1 - e^{-\gamma})}, \quad (\text{G13})$$

$$Q_2^{(m)} = \frac{1}{(e^\gamma - 1)(e^{\gamma'} - 1)} \left| \frac{\gamma' - \gamma}{e^{-\gamma'} - e^{-\gamma}} \right| (\delta_{+-} + \delta_{-+}), \quad (\text{G14})$$

$$\gamma = \hbar\omega(\boldsymbol{\kappa})/\kappa T, \quad \gamma' = \hbar\omega(\boldsymbol{\kappa}')/\kappa T. \quad (\text{G15})$$

The ratio of integrals in (G12) will at low temperatures not be greater than 1, and the smallness of the effect of double-magnon processes is assured by the  $1/3\pi^5$  factor in (G12).

The reason why the double-phonon processes at high temperatures are as important as even 1% is because of the extreme anisotropy of the phonons. This is seen from the factor  $\gamma^{-2}(\gamma')^{-2}$  in (G8) (in the high-temperature limit), these  $\gamma^{-1}$ 's having extremely large values even for the Umklapp processes. For magnons, however, the  $Q_2$  factor is cut down by a factor  $\gamma\gamma'$  [see (G10)], and at low temperatures, the exponentials cause a drastic numerical alteration.

<sup>29</sup> In reference 19, Eqs. (4.5) and (4.6) give  $d_{00}^{(1)}$  and  $d_{00}^{(2)}$ .