

## THE LUTTINGER–TISZA METHOD

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### Synopsis

The Luttinger–Tisza method and generalizations of this method for determining the minimum energy spin arrangement in a crystal subject to certain strong conditions are reviewed. It is shown that one can always replace the strong conditions by a set of additional conditions which in the simplest cases is identical with the single weak condition introduced in the Luttinger–Tisza method. A general method of calculating the minimum energy spin arrangement, based on the space group symmetry of the magnetic atom arrangements, is given, and the method of Niemeyer, based on permutation groups, is shown to be equivalent to this method. It is also shown that the Luttinger–Tisza conjecture concerning the periodicity of the minimum energy spin arrangement is equivalent to a sufficient, but not necessary, condition for a minimum of the energy.

1. *Introduction.* In this paper we discuss the Luttinger–Tisza method<sup>1)</sup> and generalizations of this method<sup>2–8)</sup> for determining the minimum energy arrangement of classical spins defined on a given atom arrangement and interacting *via* a given interaction. We consider the most general case of a spin arrangement defined on an arbitrary atom arrangement and an arbitrary interaction.

The energy to be minimized is given as a quadratic function of the spins and additional so-called strong conditions are imposed which determine the magnitude of each spin. These strong conditions are replaced by a single so-called weak condition in the Luttinger–Tisza method, and in generalizations of this method by a single weak condition and a set of additional parameters<sup>2)</sup> or by a set of weak conditions<sup>3)</sup>. In section 2, after a formal presentation of the problem, we derive a set of additional conditions which can always be used in place of the strong conditions. In the case of one or two equivalent atoms per primitive unit cell of the atom arrangement, we derive a single additional condition which is identical to the Luttinger and Tisza weak condition.

Using these additional conditions, in section 3 we outline a method of simplifying the calculation of the minimum energy spin arrangement. The method outlined is a well-known group-theoretical method<sup>9)</sup> based on the space group symmetry of the atom arrangement on which the spin arrangement is defined. The method of Niemeyer<sup>5,7,8)</sup> to simplify this calculation, based on permutation

groups, is reviewed and it is shown that this latter method is equivalent to that based on the space group symmetry of the atom arrangement.

In the Luttinger–Tisza method, an assumption is made, the so-called Luttinger–Tisza conjecture, concerning the periodicity of the lowest energy spin arrangement. In section 4 it is shown that this conjecture is equivalent to a sufficient, but not necessary, condition for the energy to be minimum.

*2. The Luttinger–Tisza method.* The elements of a space group  $F$  are denoted by  $F = (R | v(R) + t)$ , where  $R$  is a rotation matrix and  $v(R)$  and  $t$  are column matrices representing, respectively, the non-primitive translation associated with  $R$  and a primitive translation. The set of all translations  $(E | t)$  of a space group  $F$  constitute a subgroup  $T$  of  $F$ , and assuming the usual cyclic boundary conditions,  $T$  is a subgroup of  $F$  of order  $N$ . The atoms of the atom arrangement of a crystal whose space group symmetry is  $F$  can be partitioned into *simple crystals* consisting of those atoms whose positions can be obtained by applying all elements of the space group  $F$  to any one atom position  $r^{(10)}$ . A simple crystal is said to be generated by  $F$  from  $r$ . No two simple crystals have atoms in common, and the elements of  $F$  permute the atoms of each simple crystal among themselves. An atom arrangement consisting of  $M$  simple crystals generated by  $F$  from positions  $r_1, r_2, \dots, r_M$  will be denoted by  $[F; r_1, r_2, \dots, r_M]$ .

The position of an arbitrary atom in an atom arrangement  $[F; r_1, r_2, \dots, r_M]$  will be denoted by  $r_{ipn} = r_{ip} + t_n$ .  $t_n$  is a translation of  $T$ , and  $r_{ip}$  denotes the position of the  $p$ th atom of the  $i$ th simple crystal in the primitive unit cell defined by  $T$ . The indices take the values  $n = 1, 2, \dots, N$ ,  $i = 1, 2, \dots, M$ , and  $p = 1, 2, \dots, P(i)$ , where  $P(i)$  denotes the number of atoms in the primitive unit cell belonging to the  $i$ th simple crystal. The total number of atoms in the atom arrangement is then  $N \sum_i P(i)$ .

Let  $S(r)$  denote a spin arrangement defined on the atom arrangement  $[F; r_1, r_2, \dots, r_M]$ . To every atom of the atom arrangement, at positions  $r_{ipn}$ , we associate a spin vector  $S(r_{ipn})$  which for simplicity we denote as  $S_{ipn}$ . The set of all spins which constitutes the spin arrangement  $S(r)$  will be denoted by  $\{S_{ipn}\}$ .

The atom arrangement defined by  $[F; r_1, r_2, \dots, r_M]$  is the atom arrangement of the magnetic atoms in the magnetic phase of the crystal. This is distinguished from the atom arrangement of the magnetic atoms in the non-magnetic phase of the crystal, which is not necessarily invariant under the space group  $F$ . The space group  $F$  has been referred to as the “non-magnetic space group of the magnetic crystal”, the space group of the magnetic atoms determined by X ray when the crystal is in the magnetic phase<sup>11</sup>).

The problem which we are concerned with is the determination of the spin arrangement  $\{S_{ipn}\}$  which minimizes the energy

$$E = \sum_{\substack{\alpha \beta \\ i p n \\ j q m}} J_{ipn, jqm}^{\alpha \beta} S_{ipn}^{\alpha} S_{jqm}^{\beta} \quad (1)$$

under the  $N \sum_i P(i)$  so-called *strong conditions*

$$\mathbf{S}_{ipn} \cdot \mathbf{S}_{ipn} - S_i^2 = 0 \quad (2)$$

for all values of  $i, p$ , and  $n$ .  $S_{ipn}^\alpha$  is the  $\alpha$ th component of the spin  $\mathbf{S}_{ipn}$ , and the coefficients  $J_{ipn, jqm}^{\alpha\beta}$  are the interaction parameters. In eq. (2) the magnitude of the spins are dependent only on the index  $i$ , i.e. on the simple crystal to which the atom at position  $\mathbf{r}_{ipn}$  belongs.

To find the spin arrangement which minimizes the energy, eq. (1), and satisfies the strong conditions, eq. (2), one can attempt to use the Lagrange multiplier method. One introduces a set of  $N \sum_i P(i)$  Lagrange multipliers  $\{\lambda_{ipn}\}$  and defines the function  $G$ :

$$G = \sum_{\substack{\alpha \text{ } ipn \\ \beta \text{ } jqm}} J_{ipn, jqm}^{\alpha\beta} S_{ipn}^\alpha S_{jqm}^\beta - \sum_{ipn} \lambda_{ipn} \left[ \sum_{\alpha} S_{ipn}^\alpha S_{ipn}^\alpha - S_i^2 \right]. \quad (3)$$

This function is a function of both the components of the spins and the Lagrange multipliers. The minimum of the function  $G$  with respect to the set of variables  $\{\mathbf{S}_{ipn} \lambda_{ipn}\}$  is the minimum of the energy  $E$ , eq. (1), with respect to the variables  $\{\mathbf{S}_{ipn}\}$  which satisfy the strong conditions.

To find the solution  $\{\mathbf{S}_{ipn} \lambda_{ipn}\}$  which minimizes  $G$ , we differentiate eq. (3) with respect to these variables and set the derivatives equal to zero:

$$\frac{1}{2} \frac{\partial G}{\partial S_{ipn}^\alpha} = \sum_{\beta \text{ } jqm} J_{ipn, jqm}^{\alpha\beta} S_{jqm}^\beta - \lambda_{ipn} S_{ipn}^\alpha = 0, \quad (4)$$

$$\frac{\partial G}{\partial \lambda_{ipn}} = \sum_{\alpha} S_{ipn}^\alpha S_{ipn}^\alpha - S_i^2 = 0. \quad (5)$$

Equation (4), for all  $\alpha, i, p$  and  $n$ , can be written in the following concise matrix notation

$$(\mathbf{J} - \lambda) \mathbf{S} = 0, \quad (6)$$

where

$$\begin{aligned} J_{(\alpha \text{ } ipn) (\beta \text{ } jqm)} &= J_{ipn, jqm}^{\alpha\beta}, \\ \lambda_{(\alpha \text{ } ipn) (\beta \text{ } jqm)} &= \lambda_{ipn} \delta_{\alpha\beta} \delta_{ij} \delta_{pq} \delta_{nm}, \\ S_{(\alpha \text{ } ipn)} &= S_{ipn}^\alpha. \end{aligned} \quad (7)$$

The Lagrange multipliers  $\{\lambda_{ipn}\}$  are in theory calculated from the condition that a solution  $\{\mathbf{S}_{ipn}\}$  exists to eq. (6), that is:

$$\det (\mathbf{J} - \lambda) = 0. \quad (8)$$

However, since there are  $N \sum_i P(i)$  unknown Lagrange multipliers, one cannot in practice calculate them from eq. (8).

The method of Luttinger and Tisza<sup>1)</sup> to solve this problem is to replace the  $N \sum_i P(i)$  strong conditions, eq. (3), by the single so-called *weak condition*

$$\sum_{ipn} [\mathbf{S}_{ipn} \cdot \mathbf{S}_{ipn} - S_i^2] = 0 \quad (9)$$

and to assume that the spin arrangement  $\mathbf{S}(\mathbf{r})$  corresponding to the lowest energy of eq. (1) is such that

$$\mathbf{S}(\mathbf{r} + 2\mathbf{t}) = \mathbf{S}(\mathbf{r}) \quad (10)$$

for every translation  $\mathbf{t}$  of the subgroup  $T$  of  $F$ . The subgroup of  $T$  consisting of all translations of the form  $(E | 2\mathbf{t})$  will be denoted by  $T^2$ . Equation (10), known as the Luttinger–Tisza conjecture, will be discussed in detail in section 4.

The weak condition, eq. (9), is a necessary but not sufficient condition that the spin arrangement satisfies the strong conditions. Luttinger and Tisza use the Lagrange multiplier method with the single weak condition, define the function  $\bar{G}$

$$\bar{G} = \sum_{\substack{\alpha \beta \\ ipn \\ \beta jqm}} J_{ipn, jqm}^{\alpha\beta} S_{ipn}^{\alpha} S_{jqm}^{\beta} - \lambda \sum_{\alpha ipn} [S_{ipn}^{\alpha} S_{ipn}^{\alpha} - S_i^2] \quad (11)$$

and minimize  $\bar{G}$  with respect to the set of variables  $\{\mathbf{S}_{ipn}, \lambda\}$ . Comparing eqs. (3) and (11) one sees that replacing the strong conditions by a single weak condition is equivalent to assuming that the solution  $\{\mathbf{S}_{ipn}, \lambda_{ipn}\}$  of eqs. (4) and (5) is such that the Lagrange multipliers  $\lambda_{ipn}$  are independent of the indices  $i, p$  and  $n$ , i.e.  $\lambda_{ipn} = \lambda$ .

If the solution  $\{\mathbf{S}_{ipn}, \lambda\}$  which minimizes  $\bar{G}$  is such that the spin arrangement  $\{\mathbf{S}_{ipn}\}$  also satisfies the strong conditions, then one has found the solution to the original problem, i.e. the spin arrangement which minimizes the energy  $E$  and satisfies the strong conditions. If not, then no progress has been made in solving the original problem.

Freiser<sup>3)</sup> has introduced instead of a single weak condition the set of  $\sum_i P(i)$  weak conditions

$$\sum_n [\mathbf{S}_{ipn} \cdot \mathbf{S}_{ipn} - S_i^2] = 0 \quad (12)$$

for all values of the indices  $i$  and  $p$ . These are necessary but not sufficient conditions that the spin arrangement satisfies the strong conditions and is equivalent to assuming that the solution  $\{\mathbf{S}_{ipn}, \lambda_{ipn}\}$  which minimizes the function  $G$  given by eq. (3) is such that the lagrange multipliers  $\lambda_{ipn}$  are independent of the index  $n$ , i.e.  $\lambda_{ipn} = \lambda_{ip}$ .

Lyons and Kaplan<sup>2</sup>) instead of using the single weak condition of Luttinger and Tisza, eq. (9), introduce the single generalized weak condition

$$\sum_{ipn} A_{ipn}^2 [\mathbf{S}_{ipn} \cdot \mathbf{S}_{ipn} - S_i^2] = 0, \quad (13)$$

where the  $\{A_{ipn}\}$  are a set of real, non-zero parameters. This generalized weak condition is, again, a necessary but not sufficient condition that the spin arrangement satisfies the strong conditions. The Lagrange multiplier method is used with a single multiplier, and a solution is found which is a function of the set of parameters  $\{A_{ipn}\}$ . The parameters are then varied to find the set of parameters such that the solution satisfies the strong conditions.

The use of the single generalized weak condition is equivalent to replacing the Lagrange multipliers  $\lambda_{ipn}$  in eq. (3) by  $\lambda A_{ipn}^2$ , where  $\lambda$  is the Lagrange multiplier introduced with the single generalized weak condition and the  $\{A_{ipn}\}$  is a set of parameters. The strength of this method is the possibility that in varying the set of parameters one "might be lucky and have to consider only one set<sup>12)</sup>". If one was to consider all possible sets of values of the parameters, then in fact one is attempting to solve the original problem with the strong conditions. In practice one considers only a subclass of all possible values of the parameters  $\{A_{ipn}\}$ , taking the parameters as independent of the index  $n^2$ ).

Lastly, Bertaut<sup>4</sup>) has used the method of Luttinger and Tisza with the assumption that the solution  $\{\mathbf{S}_{ipn}, \lambda_{ipn}\}$ , which minimizes the function  $G$  given in eq. (3), is such that the Lagrange multipliers  $\lambda_{ipn}$  are independent of the indices  $p$  and  $n$ . In other words, considering the set of Lagrange multipliers as a scalar arrangement defined on the atom arrangement  $[F; \mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_M]$ , the scalar arrangement is assumed to be invariant under the space group  $F$ . We will show below that Bertaut's assumption is correct.

In what follows we will not restrict ourselves to the problem where the energy  $E$ , eq. (1), is due to Heisenberg exchange and dipole-dipole interactions, but consider the case of a general interaction between the spins. The interaction parameters  $J_{ipn, jqm}^{\alpha\beta}$  are not arbitrary but must satisfy the following relations

$$J_{ipn, jqm}^{\alpha\beta} = \sum_{\gamma\delta} R_{\gamma\alpha} R_{\delta\beta} J_{F(i)F(p)F(n), F(j)F(q)F(m)}^{\gamma\delta} \quad (14)$$

for every rotation matrix  $R$  of an element  $F = (R | v(R) + t)$  of the space group  $F$  of the atom arrangement on which the spin arrangement is defined; and where the indices  $F(i)$ ,  $F(p)$  and  $F(n)$  are defined by:

$$\mathbf{r}_{F(i)F(p)F(n)} = (R | v(R) + t) \mathbf{r}_{ipn}.$$

The restrictions on the interaction parameters follow from the space group symmetry of the atom arrangement  $[F; \mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_M]$  and are identical with the restrictions on the atomic force constants found in the theory of lattice dynamics<sup>9</sup>).

We will now prove the following theorem: The solution  $\{S_{ipn}, \lambda_{ipn}\}$  of eqs. (4) and (5) which minimizes the function  $G$  defined in eq. (3), where the interaction parameters satisfy relations (14), is such that the Lagrange multipliers  $\{\lambda_{ipn}\}$  are dependent only on the index  $i$ , i.e.  $\lambda_{ipn} = \lambda_i$ . The number of distinct Lagrange multipliers in the solution of eqs. (4) and (5) is then at most equal to  $M$ , the number of simple crystal which constitute the atom arrangement on which the spin arrangement is defined.

*Proof:* Relations (14) can be rewritten in matrix form as

$$\mathbf{J} = \mathbf{D}(F)^{-1} \mathbf{J} \mathbf{D}(F), \quad (15)$$

where  $\mathbf{J}$  is defined in eq. (7), and

$$\mathbf{D}(F)_{(\alpha ipn)(\beta jqm)} = \varepsilon_R R_{\alpha\beta} \delta_{i, F(j)} \delta_{p, F(q)} \delta_{n, F(m)}, \quad (16)$$

where  $\varepsilon_R = \det R$ . The matrices  $\mathbf{D}(F)$  represent the transformational properties of the components of  $\mathbf{S}$ , see eq. (6), under elements  $F = (R | v(R) + \mathbf{t})$  of the space group  $F$ , that is:

$$(R | v(R) + \mathbf{t}) \mathbf{S} = \mathbf{D}(F) \mathbf{S}. \quad (17)$$

Using eq. (15), we rewrite eq. (6) in the form

$$(\mathbf{J} - \lambda(F)) \mathbf{D}(F)^{-1} \mathbf{S} = 0, \quad (18)$$

where

$$\lambda(F) = \mathbf{D}(F)^{-1} \lambda \mathbf{D}(F) \quad (19)$$

is a diagonal matrix, whose diagonal entries are a permutation of the Lagrange multipliers  $\{\lambda_{ipn}\}$  appearing in the diagonal matrix  $\lambda$  defined in eq. (7):

$$\lambda(F)_{(\alpha ipn)(\beta jqm)} = \lambda_{F(i) F(p) F(n)} \delta_{\alpha\beta} \delta_{ij} \delta_{pq} \delta_{nm}.$$

The Lagrange multipliers can be calculated from the condition that a solution exists to eq. (18):

$$\det(\mathbf{J} - \lambda(F)) = 0. \quad (20)$$

Comparing the characteristic equations corresponding to eqs. (8) and (20), one has that  $\lambda = \lambda(F)$ , that is

$$\lambda_{ipn} = \lambda_{F(i) F(p) F(n)} \quad (21)$$

for every element  $F$  of the space group  $F$ .

The index  $i$  denotes to which simple crystal the atom at position  $r_{ipn}$  belongs, and since the elements of the space group  $F$  permute atoms of each simple crystal among themselves, we have that  $F(i) = i$ . In addition, since all atom positions belonging to a specific simple crystal are generated by  $F$  from any one atom position of the simple crystal, one obtains from eq. (21) that  $\lambda_{ipn} = \lambda_i$ . The Lagrange multipliers of the solution  $\{S_{ipn}, \lambda_{ipn}\}$  of eqs. (4) and (5) are dependent only on the index  $i$ .

This means that the  $N \sum_i P(i)$  strong conditions

$$S_{ipn} \cdot S_{ipn} - S_i^2 = 0 \quad (2)$$

can always be replaced by the  $M$  conditions

$$\sum_{pn} [S_{ipn} \cdot S_{ipn} - S_i^2] = 0, \quad (22)$$

where  $i = 1, 2, \dots, M$ , and where  $M$  is the number of simple crystals which constitute the atom arrangement on which the spin arrangement is defined.

If the atom arrangement on which the spin arrangement is defined consists of a single simple crystal, *i.e.*  $M = 1$ , the condition given by eq. (22) is identical with the Luttinger and Tisza single weak condition, eq. (9). This includes the cases of one atom per primitive unit cell, the case originally considered by Luttinger and Tisza<sup>1)</sup>, and all cases of two equivalent atoms per primitive unit cell<sup>6-8,13)</sup>. In the latter cases inversion is always an element of the space group of the atom arrangement and all atoms belong to a single simple crystal. In general, if the atoms in the primitive unit cell are identical and the coordinates of their positions constitute a set of "coordinates of equivalent positions" in the International Tables of X-Ray Crystallography<sup>14)</sup> with respect to the space group  $F$  (which is equivalent to the statement that the atom arrangement consists of a single simple crystal) then the strong conditions, eq. (2), can be replaced by Luttinger and Tisza's single weak condition, eq. (9).

Comparing eqs. (22) and (12), one has that Freiser's set of weak conditions can be used in place of the strong conditions given in eq. (2). However, unless there is only one atom per primitive unit cell from each simple crystal of the atom arrangement, the number  $\sum_i P(i)$  of unknown Lagrange multipliers which must be introduced using Freiser's set of weak conditions, eq. (12), is larger than  $M$ , the number of Lagrange multipliers which must be introduced when using the conditions given in eq. (22).

Comparing eq. (22) with the use of the single generalized weak condition of Lyons and Kaplan, eq. (13), one has that considering the subclass of sets of parameters  $\{A_{ipn}\}$  where the parameters are independent of the index  $n$  is sufficient in solving the original problem. In fact one can be more restrictive as it is sufficient

to consider the subclass of sets of parameters where the parameters are independent of both the indices  $p$  and  $n$ .

Work by Kaplan and coworkers<sup>2,15-16</sup>) have found in the case of cubic spinals, solutions which satisfy the strong conditions and are such that the parameters  $\{A_{ipn}\}$  are dependent on the index  $p$ . Comparing their theoretical results and the experimental results of Hastings and Corliss<sup>17</sup>) on the magnetic structure of  $\text{MnCr}_2\text{O}_4$  with the above results, one concludes that in the magnetic phase of  $\text{MnCr}_2\text{O}_4$  below 18 K there is an orthorhombic deformation of the atom arrangement on which the spin arrangement is defined<sup>4</sup>).

3. *Calculation of the minimum energy spin arrangement.* In this section we outline a general method of simplifying the calculation of the solution of eqs. (4) and (5) taking into account the results of section 2. The method used is a well-known group theoretical method based on the space group symmetry of the atom arrangement on which the spin arrangement is defined, and used for example in the theory of lattice dynamics<sup>9</sup>). The purpose of this section is to present a general method which is not dependent on any assumptions on the translational invariance of the solution, as the Luttinger-Tisza conjecture, eq. (10). In addition, the method to calculate the solution of eqs. (4) and (5) based on permutation groups<sup>5-8</sup>) and applied only in cases where the Luttinger-Tisza conjecture is assumed to be valid, is shown to be equivalent to this general method.

In matrix notation, the problem is to find the eigenfunction  $S$  of eq. (6)

$$JS = \lambda S \quad (6)$$

which minimizes the energy  $E$  given in eq. (1). The matrices  $J$  and  $S$  are defined in eq. (7) and  $\lambda$  is the matrix:

$$\lambda_{(\alpha ipn)(\beta jqm)} = \lambda_i \delta_{\alpha\beta} \delta_{ij} \delta_{pq} \delta_{nm}.$$

The energy  $E$ , using eqs. (1), (4) and (5), can be written as

$$E = N \sum_i P(i) \lambda_i S_i^2, \quad (23)$$

where  $P(i)$  is the number of atoms in the primitive unit cell belonging to the  $i$ th simple crystal, and the sum is over all simple crystals.

We must then find the set of Lagrange multipliers  $\{\lambda_i\}$   $i = 1, 2, \dots, M$ , the components of the matrix  $\lambda$ , which are solutions of  $\det(J - \lambda) = 0$  and which minimize eq. (23); and the corresponding eigenfunction  $S$  of eq. (6). To simplify this calculation we note that from eqs. (15), (19) and (21)

$$JD(F)S = \lambda D(F)S,$$



where  $D(F)$  is the matrix defined in eq. (16). Consequently, the eigenfunction of eq. (6) are simultaneously basis functions of irreducible representations of the space group  $F$ , in particular, of irreducible representations of  $F$  contained in the representation  $D(F)$  afforded by the set of matrices  $D(F)$ .

We first consider only the subgroup  $T$  of primitive translations  $(E | \mathbf{t}_n)$  of the space group  $F$ , and determine the linear combinations of the components of  $S$  which are basis functions of irreducible representations of  $T$  contained in the representation  $D(T)$  of  $T$ . We calculate the unitary matrix  $U$  such that

$$UD(T)U^{-1} = D_r(T),$$

where  $D_r(T)$  is the reduced form of the representation  $D(T)$ .  $D_r(T)$  is the direct sum of all irreducible representations of the group  $T$ , each irreducible representation appearing  $3 \sum_i P(i)$  times. The matrices of the representation  $D_r(T)$  are diagonal matrices of the form

$$D_r(\mathbf{t})_{(\alpha i p k)(\beta j q k')} = e^{-i\mathbf{k} \cdot \mathbf{t}} \delta_{\alpha\beta} \delta_{ij} \delta_{pq} \delta_{kk'},$$

where  $\mathbf{k}$  and  $\mathbf{k}'$  are vectors of the first Brillouin zone of the reciprocal lattice defined by  $T$ , and take  $N$  possible values. The matrix  $U$  is found to be:

$$U_{(\alpha i p k)(\beta j q n)} = (1/\sqrt{N}) e^{i\mathbf{k} \cdot \mathbf{t}_n} \delta_{\alpha\beta} \delta_{ij} \delta_{pq}.$$

The matrix  $U$  is used to calculate the linear combinations of the components of  $S$ , denoted by  $Q_{ip}^\alpha(\mathbf{k})$ , which transform under translations of  $T$  as basis functions for the  $k$ th irreducible representation of  $T$ . Since the irreducible representations of  $T$  are one-dimensional, the  $Q_{ip}^\alpha(\mathbf{k})$  are eigenfunctions of the translations of  $T$ . Defining the components of  $Q$  as  $Q_{(\alpha i p k)} = Q_{ip}^\alpha(\mathbf{k})$ , we have

$$Q = US \quad \text{and} \quad (E | \mathbf{t}) Q = D_r(\mathbf{t}) Q.$$

That is

$$Q_{ip}^\alpha(\mathbf{k}) = (1/\sqrt{N}) \sum_n e^{i\mathbf{k} \cdot \mathbf{t}_n} S_{ipn}^\alpha \quad (24)$$

and

$$(E | \mathbf{t}) Q_{ip}^\alpha(\mathbf{k}) = e^{-i\mathbf{k} \cdot \mathbf{t}} Q_{ip}^\alpha(\mathbf{k}). \quad (25)$$

From eq. (25) we have that the  $Q_{ip}^\alpha(\mathbf{k})$  are Fourier transforms of the components of  $S^{4,18}$ .

In terms of  $Q$ , using the relation  $S = U^{-1}Q$ , eq. (6) becomes

$$UJU^{-1}Q = \lambda Q$$

which splits into  $N$  independent equations

$$L(k) Q(k) = \lambda(k) Q(k) \quad (26)$$

for the  $N$  values of  $k$ , and where the  $3 \sum_i P(i)$  dimensional matrices  $L(k)$  and  $\lambda(k)$  are defined by

$$L(k)_{(\alpha ip)(\beta jq)} = \sum_m J_{(ipn)(jqm)}^{\alpha\beta} e^{ik \cdot (t_n - t_m)},$$

$$\lambda(k)_{(\alpha ip)(\beta jq)} = \lambda_i \delta_{\alpha\beta} \delta_{ij} \delta_{pq}$$

and where

$$Q(k)_{(\alpha ip)} = Q_{ip}^{\alpha}(k).$$

A necessary and sufficient condition that a set  $\{\lambda_i\}$  is a solution of  $\det(J - \lambda) = 0$ , eq. (8), is that the set  $\{\lambda_i\}$  be a solution of

$$\det(L(k) - \lambda(k)) = 0 \quad (27)$$

for some value of  $k$ . One therefore determines the solutions  $\{\lambda_i\}$  of eq. (27) for each value of  $k$ , and then from among these, the set  $\{\lambda_i\}$  which minimizes the energy given in eq. (23). The corresponding eigenfunction is determined from eq. (26).

The Luttinger-Tisza conjecture, eq. (10), means that one determines the set  $\{\lambda_i\}$  which minimizes eq. (22) from that subset of solutions of eq. (27) corresponding to values of  $k$  for which  $2k = K$ , where  $K$  is a reciprocal lattice vector. For such values of  $k$ , the eigenfunctions of eq. (26), as can be seen from eq. (24), are invariant under all translations of  $T^2$ .

In general, simplifications of eq. (26) can be made by noting that the matrices  $\bar{D}(F)$  defined by

$$\bar{D}(F)_{(\alpha ip)(\beta jq)} = \sum_n D(F)_{(\alpha ipn)(\beta jqm)} \quad (28)$$

commute with the matrices  $L(k)$  and  $\lambda(k)$  for those elements of  $F$  belonging to  $F_k$ , the space group of the vector  $k$ , i.e. for all elements  $F = (R | v(R) + t)$  such that  $Rk = k + K$ . The eigenfunctions of eq. (26) are therefore basis functions of the irreducible representations of the group  $F_k$  contained in the representation  $D(F_k)$ . One can then proceed to simplify the calculations of eq. (26) in a manner similar to the simplification of the calculation of eq. (6)<sup>9</sup>.

Recently, Niemeyer and others<sup>5-8,13</sup> have used a method based on permutation groups to simplify the calculations of the solution of eqs. (4) and (5). They

consider the case of a spin arrangement defined on a single simple crystal with one or two atoms per primitive unit cell and assume the validity of the Luttinger-Tisza conjecture.

In the case of one atom per unit cell, their problem is then to find the lowest energy eigenfunction  $\eta$  of the equation

$$(A - \lambda) \eta = 0, \quad (29)$$

where  $A$  and  $\lambda$  are the twenty-four dimensional matrices

$$A_{(\alpha a)(\beta b)} = \sum_u J_{(\alpha a)(\beta b+u)}, \quad (30)$$

$$\lambda_{(\alpha a)(\beta b)} = \lambda \delta_{\alpha\beta} \delta_{ab}$$

and

$$\eta_{(\alpha a)} = S_a^\alpha,$$

$\alpha = 1, 2, 3$ ,  $a = 1, 2, \dots, 8$ , and  $S_a^\alpha$  is the  $\alpha$ th component of the spin at  $r_a$ , the position of one of the eight atoms in the primitive unit cell defined by  $T^2$ . The energy of the eigenfunction is proportional to  $\lambda$  and the index  $u$  indexes the translations ( $E | t_u$ ) of the group  $T^2$ .

A group of eight matrices  $A(P_i)$ ,  $i = 1, 2, \dots, 8$  is found which commute with each other and the matrix  $A$  of eq. (29). These matrices give the transformational properties of the components of  $\eta$  under a group of permutations of the values of the index  $a$

$$P_i \eta_{(\alpha a)} = \eta_{(\alpha P_i a)} = \sum_{\beta b} A(P_i)_{(\alpha a)(\beta b)} \eta_{(\beta b)}, \quad (31)$$

where

$$A(P_i)_{(\alpha a)(\beta b)} = \delta_{\alpha\beta} \delta_{bP_i a}.$$

These matrices commute with the matrix  $A$

$$A = A(P_i)^{-1} A A(P_i) \quad (32)$$

and hence

$$A_{(\alpha a)(\beta b)} = A_{(\alpha P_i a)(\beta P_i b)}. \quad (33)$$

The simplification of the calculation of the eigenfunctions of eq. (29) are based on that the eigenfunctions of eq. (29) are simultaneously basis functions of irreducible representations of the group  $P$  of permutations  $P_i$ . Since the permutation

group  $P$  in question is abelian, the irreducible representations are one-dimensional and the eigenfunctions of eq. (29) are also eigenfunctions of the permutations of  $P$ .

We point out that this method based on the theory of representations of permutations groups is identical with the analysis given above based on the space group symmetry of the matrix  $J$  of eq. (6). In fact the matrices  $A(P_i)$  defined in eq. (31) are identical to the matrices which give the transformation properties of the components  $S_a^\alpha$  of  $\eta$ , see eq. (17), under the eight translations of  $T$  not contained in  $T^2$  when eq. (10) is taken into account. Relation (33) follows directly from eqs. (15) and (30) when  $P_i$  is replaced by the appropriate translation of  $T$  not contained in  $T^2$ .

For the case of one atom per unit cell, the method used by Niemeyer<sup>5</sup> is then in fact identical with the general method outlined in the beginning of this section, but in the guise and nomenclature of permutation groups. The simplifications derived in ref. 5 consequently are identical with eqs. (25)–(27) in the case of vectors  $k$  where  $2k = K$ .

In the case of two atoms per primitive unit cell of a single simple crystal<sup>6–8,13</sup>) eq. (29) is used, where  $\eta$  is now a forty-eight dimensional vector with components  $\eta_{(\alpha a)}$ ,  $a = 1, 2, \dots, 16$  corresponding to the three components of the sixteen spins in the primitive unit cell of  $T^2$ . The sixteen permutation matrices, see eqs. (31)–(33), derived in refs. 6 and 8, in this case correspond to the eight translations of  $T$  not in  $T^2$  and to these translations combined with inversion, which in the case of two equivalent atoms per primitive cell, is always a symmetry element of the space group  $F$  of the atom arrangement. Inversion is also an element of  $F_k$  for values of  $k$  such that  $2k = K$ , and the procedure given after eq. (28) can be used. One finds directly the result that the eigenfunctions of eq. (29) are eigenfunctions not only of the translations of  $T$  but also of inversion.

The method based on permutation groups to simplify the solution of eqs. (4) and (5) has not been applied to problems other than that of atom arrangements consisting of a single simple crystal with one or two atoms per primitive unit cell. In fact, the impossibility of extending this method in general to additional cases has been concluded<sup>8,19</sup>). The advantage of simplifying the calculation of the solution of eqs. (4) and (5) using eqs. (26) and (27) is that it provides a straightforward method, based on the space group symmetry of the atom arrangement, applicable to all possible cases.

4. *The Luttinger–Tisza conjecture.* The Luttinger–Tisza conjecture, eq. (10), is the assumption that the lowest energy spin arrangement is invariant under the subgroup  $T^2$  of the space group  $F$  of the atom arrangement. In the context of the method of section 3, this conjecture means that one determines the set  $\{\lambda_i\}$  which minimizes eq. (22) from among the subset of solutions of eq. (27) corresponding to values of  $k$  for which  $2k = K$ . One considers then only eight values of the

vector  $\mathbf{k}$ :  $(0\ 0\ 0)$ ,  $(\frac{1}{2}\ 0\ 0)$ ,  $(0\ \frac{1}{2}\ 0)$ ,  $(0\ 0\ \frac{1}{2})$ ,  $(0\ \frac{1}{2}\ \frac{1}{2})$ ,  $(\frac{1}{2}\ 0\ \frac{1}{2})$ ,  $(\frac{1}{2}\ \frac{1}{2}\ 0)$  and  $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$  where the components of  $\mathbf{k}$  have been given in fractions of the three generating translations of the reciprocal lattice.

In substances with predominantly dipole-dipole interactions, the Luttinger-Tisza conjecture so far has been found to be valid<sup>20</sup>), but it is known that it does not hold for cases with predominantly Heisenberg exchange interactions as can be seen by the work of Yoshimura<sup>21</sup>) and Kaplan and co-workers<sup>2,15,16</sup>). A proof of the Luttinger-Tisza conjecture has been given by Karl<sup>22</sup>), but only for the case of a single simple crystal with one atom per primitive unit cell and with restrictive conditions on the interaction parameters, *e.g.* nearest-neighbor interactions only. We will show below that the Luttinger-Tisza conjecture is equivalent to a sufficient, but not necessary, condition for the minimum of the energy  $E$  defined in eq. (23).

We denote by  $E(\mathbf{k})$  the energy corresponding to the set  $\{\lambda_i\}$  calculated from eq. (27) for a specific value of  $\mathbf{k}$ . We first show that the energy  $E(\mathbf{k})$  as a function of  $\mathbf{k}$  has local extremal values at precisely those values of  $\mathbf{k}$  predicted by the Luttinger-Tisza conjecture, *i.e.* at those values of  $\mathbf{k}$  for which  $2\mathbf{k} = \mathbf{K}$ .

From eq. (27) we have<sup>9</sup>)

$$E(\mathbf{k}) = E(\mathbf{k} + \mathbf{K}) \quad (34)$$

and

$$E(\mathbf{k}) = E(-\mathbf{k}). \quad (35)$$

We expand  $E(\mathbf{k})$  in a power series about  $E(\mathbf{k}_0)$ , and using eqs. (34) and (35) we have:

$$\begin{aligned} E(\mathbf{k}_0 + \Delta\mathbf{k}) &= E(\mathbf{k}_0) + (\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=\mathbf{k}_0} \Delta\mathbf{k} + \dots \\ &= E(-\mathbf{k}_0) - (\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=-\mathbf{k}_0} \Delta\mathbf{k} + \dots \\ &= E(\mathbf{k}_0) - (\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=-\mathbf{k}_0+\mathbf{K}} \Delta\mathbf{k} + \dots \end{aligned}$$

From the first and third of these relations we have

$$(\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=\mathbf{k}_0} = -(\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=-\mathbf{k}_0+\mathbf{K}}$$

and consequently  $(\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=\mathbf{k}_0} = 0$  and  $E(\mathbf{k})$  has a local extremal value for those values of  $\mathbf{k}$  for which  $2\mathbf{k} = \mathbf{K}$ , precisely those values of  $\mathbf{k}$  predicted by the Luttinger-Tisza conjecture.

One can show in a similar manner that from  $E(\mathbf{k}) = E(R\mathbf{k})$ , where  $R$  is the rotation matrix of an element  $F = (R | v(R) + \mathbf{t})$  of the space group  $F$ , that:

$$(\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=R\mathbf{k}_0+\mathbf{K}} = R (\nabla_{\mathbf{k}} E(\mathbf{k}))_{\mathbf{k}=\mathbf{k}_0}.$$

The gradient of  $E(\mathbf{k})$  at a specific value of  $\mathbf{k}$  is then invariant under all rotations  $R$  of elements of  $F_{\mathbf{k}}$ , the space group of the vector  $\mathbf{k}$ . The group of all such rotations is referred to as the point group of the vector  $\mathbf{k}$ . If the point group of the vector  $\mathbf{k}$  does not allow an invariant vector, *i.e.* the point group  $C_i$ ,  $C_{2h}$ ,  $D_2$ ,  $D_{2h}$ ,  $S_4$ ,  $C_{4h}$ ,  $D_4$ ,  $D_{2d}$ ,  $D_{4h}$ ,  $C_{3i}$ ,  $D_3$ ,  $D_{3d}$ ,  $C_{3h}$ ,  $C_{6h}$ ,  $D_6$ ,  $D_{3h}$ ,  $D_{6h}$ ,  $T$ ,  $T_h$ ,  $O$ ,  $T_d$  or  $O_h$ , then the gradient of  $E(\mathbf{k})$  is zero and  $E(\mathbf{k})$  takes on a local extremal value for this value of  $\mathbf{k}$ . We note that such vectors  $\mathbf{k}$  include cases where  $2\mathbf{k} \neq \mathbf{K}$ , for example, the point  $R$  on the surface of the Brillouin Zone of the space group  $F = D_{2h}^{28}$  where the point group of the vector  $\mathbf{k}$  is  $D_2^{23}$ .

Consequently, one anticipates local extremal values, possibly local minima, of the energy  $E(\mathbf{k})$  at those values of  $\mathbf{k}$  for which one or both of the following conditions are satisfied:

- 1)  $2\mathbf{k} = \mathbf{K}$ ;
  - 2) the point group of the vector  $\mathbf{k}$  does not allow an invariant vector.
- (36)

However, these conditions are only sufficient and not necessary conditions for local extremal values of  $E(\mathbf{k})$ , and the above argument based on the symmetry properties of the function  $E(\mathbf{k})$  does not preclude the possibility of additional values of  $\mathbf{k}$  for which  $E(\mathbf{k})$  has local extremal values.

We note that the above results, which include the Luttinger–Tisza conjecture, are identical with conditions used in the Landau theory of second-order phase transitions<sup>24</sup>). These conditions arise in the same manner as above as conditions for the minimum of a function which is dependent on the vector  $\mathbf{k}$ <sup>25</sup>). As in the case of second-order phase transitions we can distinguish between local minima of  $E(\mathbf{k})$  due to symmetry, with corresponding values of  $\mathbf{k}$  which satisfy one or both of the conditions (36), and local minima of  $E(\mathbf{k})$  which are not due to symmetry but depend on the specific values of the interaction parameters which enter into eq. (1)<sup>5,13,14,21</sup>).

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