Generalization of the Luttinger-Tisza Method

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(Received 3 April 1973)

Using permutation groups the Luttinger-Tisza method for the calculation of the low-temperature ordered state of spin configurations is generalized to treat more than one ion per unit cell. It is found that, for the dipole-dipole interaction, not more than two ions per unit cell can be treated by this method. On the other hand, for the exchange interaction it is possible to treat the case of 2^m ions per unit cell (m is an integer), provided the ions occupy special positions.

I. INTRODUCTION

The Luttinger-Tisza (LT) method[1] for the calculation of low-temperature ordering in crystals containing paramagnetic ions interacting via dipole-dipole (DD) interactions has been successfully applied to the cases of one and two magnetically equivalent ions per unit cell by several workers.[2] The method was originally proposed by Luttinger and Tisza to calculate the low-temperature ordering of ions, occupying the sites of a Bravais lattice and interacting with each other through the DD interaction. Recently, Niemeyer has extended the LT method to include the exchange interaction and has applied it to the case of cerium magnesium nitrate.[3] Felsteiner and Misra have further extended Niemeyer's work to consider cases involving two magnetically equivalent ions per unit cell.[4] Very recently, Niemeyer and Blöte[5] have considered the general problem of more than one ion per unit cell. They use arguments based on the symmetry of a matrix Y [much like the matrix A defined by Eqs. (2a) and (2b) in Sec. II], for the case of two magnetically equivalent ions per unit cell, and show that the eigenvectors of the energy matrix Y for the case of one ion per unit cell can be used to construct the eigenvectors of the corresponding matrix Y for the case of two ions per unit cell. For the cases of more than two magnetically equivalent ions per unit cell, their examination of the corresponding matrix Y leads them to the conclusion that these cases cannot generally be handled by the LT method.

It is the purpose of this paper to investigate the conditions under which it is possible to generalize the LT method to treat more than two magnetically equivalent ions per unit cell. This study is based on the question of the existence of a group of permutations which leave the matrix A invariant.[3] If it is possible to construct such a group for a particular case, then the conditions necessary for the applicability of the LT method are fulfilled. It will be seen that for the exchange interaction it is possible to treat certain special numbers of magnetically equivalent ions per unit cell by the LT method. In Sec. II a brief review of the background theory is presented. Section III deals with the details of the generalization and Sec. IV deals with the situations to which the generalization is applicable. The conclusions are summarized in Sec. V.

II. BRIEF REVIEW OF BACKGROUND THEORY

If the Hamiltonian describing the interaction between the various ions in the crystal can be written

$$\mathcal{H} = \sum_{p,q} \sum_{\alpha,\beta=x,y,z} B_{pq}^{\alpha\beta} S_p^\alpha S_q^\beta \tag{1}$$

(p,q label the ions), then for a regular distribution of ions within the three-dimensional crystal (containing n magnetically equivalent ions per unit cell), one can rearrange Eq. (1) as

$$\mathcal{H} = \sum_{i,j=1}^{8n} \sum_{\alpha,\beta} A_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta S^2 , \tag{2}$$

having divided the crystal into $8n$ sublattices. Here $\vec{S}$ denotes the ionic spin.

For the DD, nearest-neighbor (NN), and next-nearest-neighbor (NNN) exchange interactions,

$$A_{ij}^{\alpha\beta} = \sum_{l\in\{j\}} J_{il}^{\alpha\beta} \quad \text{for } i \neq j ,$$
$$A_{jj}^{\alpha\beta} = \sum_{\substack{l\in\{j\} \\ l\neq j}} J_{jl}^{\alpha\beta} , \tag{2a}$$

$$J_{ij}^{\alpha\beta} = \frac{S^2}{2}\left[\frac{\mu_B^2 g^\alpha g^\beta}{r_{ij}^3}\left(\delta_{\alpha\beta} - \frac{3r_{ij}^\alpha r_{ij}^\beta}{r_{ij}^2}\right) + v_{ij}\Delta_{ij}\delta_{\alpha\beta}\right] \tag{2b}$$

The first term of Eq. (2b) represents the DD interaction, and the second term represents the exchange interaction. In Eq. (2a) $\{j\}$ denotes the set of lattice sites generated by the application of $\vec{\Gamma}^2$ to ion j ($\vec{\Gamma}$ represents the lattice translations $\vec{\Gamma} = l\vec{a} + m\vec{b} + n\vec{c}$; l, m, n are integers and $\vec{a}$, $\vec{b}$, $\vec{c}$ are the unit-cell vectors). Thus the whole lattice is divided into $8n$ sublattices by application of $\vec{\Gamma}^2$ to the vertices of n intercrossing parallelepipeds, each of dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$; one vertex of each of the par-

allelepipeds is located at one of the n ion sites in the unit cell. In Eq. (2b) g^{α} represents the diagonal elements of the g tensor (a coordinate system in which the g tensor is diagonal is chosen), $\vec{r}_{ij}$ is the vector from ion i to ion j; $\Delta_{ij}=1$ if i and j are nearest or next-nearest neighbors, 0 otherwise; and v_{ij} is the exchange-interaction constant between ions i and j.

Using a set of wave functions which are direct products of the one-spin wave functions, the expectation value of $\mathcal{H}$ can be expressed as

$$E=\langle\mathcal{H}\rangle=\sum_{i,j=1}^{8n}\sum_{\alpha,\beta}A_{ij}^{\alpha\beta}\xi_i^{\alpha}\xi_j^{\beta}\ , \tag{3}$$

where

$$\xi_i^{\alpha}=\langle S_i^{\alpha}\rangle/S\ .$$

According to the LT method the minimum value of E is the lowest eigenvalue of the matrix A, provided that the eigenvectors which satisfy the "weak" condition

$$\sum_{i=1}^{8n}\vec{\xi}_i\cdot\vec{\xi}_i=8n \tag{4}$$

also satisfy the "strong" condition

$$\vec{\xi}_i\cdot\vec{\xi}_i=1\quad(\text{all } i)\ . \tag{5}$$

Niemeyer[3] proves a theorem whereby the task of finding the eigenvalues and eigenvectors of the $24n\times24n$ matrix A is reduced to that of finding the eigenvalues and eigenvectors of $8n$ 3×3 matrices, provided that it is possible to construct a group of $8n$ commuting permutations P_t (operating on elements numbered 1, 2, ..., $8n$), having the property

$$A_{P_t iP_t j}^{\alpha\beta}=A_{ij}^{\alpha\beta}\ . \tag{6}$$

It follows that $P_t^2=1$. According to the theorem, then, the eigenvectors of the matrix A can be written as

$$\eta(k,\alpha)=q(k)\varphi_k(\alpha)\quad(k=1,2,\ldots,8n;\ \alpha=x,y,z)\ . \tag{7}$$

In Eq. (7) the $q(k)$ are the common eigenvectors of the group of permutations P_t ($t=1,2,\ldots,8n$.) The $\varphi_k(\alpha)$ are the eigenvectors of the matrix L_k, whose elements are defined as

$$L_k^{\mu\nu}=\sum_j A_{ij}^{\mu\nu}\epsilon_{P(i,j)}(k)\ , \tag{8}$$

where the summation in Eq. (8) can be easily shown to be independent of i, and $\epsilon_{P(i,j)}(k)$ is the eigenvalue of the permutation $P(i,j)$ containing the cycle (i,j), with respect to the eigenvector $q(k)$. When the required conditions are satisfied so that the eigenvectors can be expressed in the form given by Eq. (7), it is seen that the "weak" condition does indeed imply the "strong" condition. The lowest eigenvalue of the matrix A corresponding to the eigenvectors (7) is then the lowest value of energy given by Eq. (3).

III. DETAILS OF THE GENERALIZATION

We first review the conditions that must be satisfied in order that the eigenvectors may be written in the form of Eq. (7): (a) It must be possible to construct a group of $8n$ permutations satisfying Eq. (6), acting on the $8n$ ions situated at the vertices of parallelepipeds. One of these permutations is the identity permutation, and the others are broken into cycles of two elements each, with each element appearing only once in each permutation. (b) All P_t's must commute with each other so they have a common set of eigenvectors. (c) The eigenvalues ϵ_{P_t} must be such that $\epsilon_{P_t}^2=1$.

It is also noted that expressing the eigenvectors of A in the form given by Eq. (7) is equivalent to first diagonalizing the matrix A in the subspace (i,j) (corresponding to the various ions $1,2,\ldots,8n$) and then diagonalizing in the subspace (α,β) (corresponding to the individual spin spaces). Since the elements of $q(k)$ are either $1/\sqrt{(8n)}$ or $-1/\sqrt{(8n)}$, the eigenvectors $\eta(k,\alpha)$ correspond to ferromagnetic, antiferromagnetic, or layered-antiferromagnetic ordering of spins. Then, normalization of $\eta(k,\alpha)$ (i.e., fulfillment of the "weak" condition) implies the fulfillment of the "strong" condition.

Taking cognizance of the conditions (a), (b), and (c), we now proceed to discuss the cases corresponding to different values of n.

A. Case for $n=1$

This case has been discussed in Ref. 3. The eight sublattices are obtained by application of $\vec{\Gamma}^2$ to the vertices of a parallelepiped of dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$ with one of the vertices situated at the given ion in the unit cell; the vertices are labeled $1,2,\ldots,8$. For DD and exchange interactions, the group of permutations is listed in Table I. For reference purposes, we denote this group as $P^{i,i}$ ($i=1,2,\ldots,8$). The corresponding eigenvectors $q(k)$ ($k=1,2,\ldots,8$), expressed as column vectors, are given in Table II. The eigenvalues of the per-

TABLE I. The group of permutations P_t ($t=1,2,\ldots,8$) for eight elements. It is easily seen that $[P_t, P_{t'}]=0$ for all t, t'.

$P_1=I$ (Identity)				
$P_2=$ (1, 2)	(3, 4)	(5, 6)	(7, 8)	
$P_3=$ (1, 3)	(2, 4)	(5, 7)	(6, 8)	
$P_4=$ (1, 4)	(2, 3)	(5, 8)	(6, 7)	
$P_5=$ (1, 5)	(2, 6)	(3, 7)	(4, 8)	
$P_6=$ (1, 6)	(2, 5)	(3, 8)	(4, 7)	
$P_7=$ (1, 7)	(2, 8)	(3, 5)	(4, 6)	
$P_8=$ (1, 8)	(2, 7)	(3, 6)	(4, 5)	

TABLE II. Eigenvectors $q(k)$ for P_t $(t=1, 2, \ldots, 8)$.

$q(1)$	$q(2)$	$q(3)$	$q(4)$	$q(5)$	$q(6)$	$q(7)$	$q(8)$
$\begin{pmatrix}\alpha\\ \alpha\end{pmatrix}^{a}$	$\begin{pmatrix}\beta\\ -\beta\end{pmatrix}^{b}$	$\begin{pmatrix}\gamma\\ \gamma\end{pmatrix}^{c}$	$\begin{pmatrix}\alpha\\ -\alpha\end{pmatrix}^{a}$	$\begin{pmatrix}\delta\\ \delta\end{pmatrix}^{d}$	$\begin{pmatrix}\gamma\\ -\gamma\end{pmatrix}^{c}$	$\begin{pmatrix}\beta\\ \beta\end{pmatrix}^{b}$	$\begin{pmatrix}\delta\\ -\delta\end{pmatrix}^{d}$

[a]Where $\sqrt{8}\alpha = \begin{pmatrix}1\\1\\1\\1\end{pmatrix}$ [c]Where $\sqrt{8}\gamma = \begin{pmatrix}1\\1\\-1\\-1\end{pmatrix}$.

[b]Where $\sqrt{8}\beta = \begin{pmatrix}1\\-1\\-1\\1\end{pmatrix}$ [d]Where $\sqrt{8}\delta = \begin{pmatrix}1\\-1\\1\\-1\end{pmatrix}$.

mutations are given in Table III. It is easily seen that conditions (a), (b), and (c) are fulfilled for this case.

B. Case for $n=2$

This case has been discussed in Ref. 4. The sixteen sublattices are obtained by application of $\vec{\Gamma}^2$ to the vertices of two intercrossing parallelepipeds of dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$, one vertex of each being situated at one of the two ions in the unit cell. The indices $1, 2, \ldots, 8$ label the vertices of one parallelepiped [as in Sec. III A] and the indices $i+8 = 9, 10 \ldots, 16$ label the vertices of the other parallelepiped. An inspection shows that the corresponding group of sixteen permutations $P^{j,j}$ (given in Table IV) can be expressed as

$$P^{j,j} = \begin{bmatrix} P^{i,i} & P^{i+8,i+8} \\ & P^{i,i+8} \end{bmatrix} \quad (i=1,2,\ldots,8;\ j=1,2,\ldots,16)\,. \tag{9}$$

Here $P^{i+8,i+8}$ refers to the group of permutations obtained from the group $P^{i,i}$ $(i=1,2,\ldots,8)$ by replacing each element i by the element $i+8$. $P^{i,i+8}$ refers to the set of eight permutations, each permutation consisting of eight cycles, each cycle containing one element from i $(=1,2,\ldots,8)$ and one element from $i+8$ $(=9,10,\ldots,16)$ in such a way that any side of the parallelepiped, the vertices of which are labeled $1,2,\ldots,8$, goes over into a parallel side of the parallelepiped with the vertices labeled $9,10,\ldots,16$ under the permutations $P^{i,i+8}$. [In general, the following notation will hereafter be used. Both $P^{i,i}$ and $P^{i+2^m,i+2^m}$ $(i=1,2,\ldots,2^m)$ will refer to sets of 2^m permutations, each permutation containing 2^{m-1} cycles. $P^{i,i+2^m}$ will refer to a set of 2^m permutations, each permutation containing 2^m cycles. The permutations for the case $n=2^m$ are easily derived from the knowledge of the permutations for the case $n=2^{m-1}$ in the same fashion as permutations for the case $n=2$ were derived from those for the case $n=1$, as expressed by Eq. (9) above.]

TABLE III. Eignevalues of the vectors $q(k)$ under the operations P_t $(t=1, 2, \cdots, 8)$.

	P_1	P_2	P_3	P_4	P_5	P_6	P_7	P_8
$q(1)$	1	1	1	1	1	1	1	1
$q(2)$	1	−1	−1	1	−1	1	1	−1
$q(3)$	1	1	−1	−1	1	1	−1	−1
$q(4)$	1	1	1	1	−1	−1	−1	−1
$q(5)$	1	−1	1	−1	1	−1	1	−1
$q(6)$	1	1	−1	−1	−1	−1	1	1
$q(7)$	1	−1	−1	1	1	−1	−1	1
$q(8)$	1	−1	1	−1	−1	1	−1	1

The eigenvectors $Q(j)$ $(j=1,2,\ldots,16)$ can be simply expressed in terms of the eigenvectors $q(k)$ $(k=1,2,\ldots,8)$ for the case $n=1$ as follows:

$$Q(2k-1) = \frac{1}{\sqrt{2}}\begin{pmatrix}q(k)\\ q(k)\end{pmatrix}\,,\quad Q(2k) = \frac{1}{\sqrt{2}}\begin{pmatrix}q(k)\\ -q(k)\end{pmatrix}\,. \tag{10}$$

The eigenvalues of P_t $(t=1,2,\ldots,16)$ with respect to $Q(j)$ as given by Eq. (10) are listed in Table V.

It is easily seen that for this case the conditions (a), (b), and (c) are satisfied.

C. Case for $n=2^m$ $(m>1)$

In this case the whole lattice is divided into 8×2^m sublattices which are obtained by application of $\vec{\Gamma}^2$ to 8×2^m vertices of 2^m intercrossing parallelepipeds, each of dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$; one of the vertices of each parallelepiped being situated at one of the ions in the unit cell. An inspection of the group

TABLE IV. The group of permutations P_t $(t=1, 2, \ldots, 16)$ of sixteen elements. It is easily seen that $[P_t, P_{t'}] = 0$ for all t, t'.

$P_1 = I$ (Identity)							
$P_2 = (1,2)$	$(3,4)$	$(5,6)$	$(7,8)$	$(9,10)$	$(11,12)$	$(13,14)$	$(15,16)$
$P_3 = (1,3)$	$(2,4)$	$(5,7)$	$(6,8)$	$(9,11)$	$(10,12)$	$(13,15)$	$(14,16)$
$P_4 = (1,4)$	$(2,3)$	$(5,8)$	$(6,7)$	$(9,12)$	$(10,11)$	$(13,16)$	$(14,15)$
$P_5 = (1,5)$	$(2,6)$	$(3,7)$	$(4,8)$	$(9,13)$	$(10,14)$	$(11,15)$	$(12,16)$
$P_6 = (1,6)$	$(2,5)$	$(3,8)$	$(4,7)$	$(9,14)$	$(10,13)$	$(11,16)$	$(12,15)$
$P_7 = (1,7)$	$(2,8)$	$(3,5)$	$(4,6)$	$(9,15)$	$(10,16)$	$(11,13)$	$(12,14)$
$P_8 = (1,8)$	$(2,7)$	$(3,6)$	$(4,5)$	$(9,16)$	$(10,15)$	$(11,14)$	$(12,13)$
$P_9 = (1,9)$	$(2,10)$	$(3,11)$	$(4,12)$	$(5,13)$	$(6,14)$	$(7,15)$	$(8,16)$
$P_{10} = (1,10)$	$(2,9)$	$(3,12)$	$(4,11)$	$(5,14)$	$(6,13)$	$(7,16)$	$(8,15)$
$P_{11} = (1,11)$	$(2,12)$	$(3,9)$	$(4,10)$	$(5,15)$	$(6,16)$	$(7,13)$	$(8,14)$
$P_{12} = (1,12)$	$(2,11)$	$(3,10)$	$(4,9)$	$(5,16)$	$(6,15)$	$(7,14)$	$(8,13)$
$P_{13} = (1,13)$	$(2,14)$	$(3,15)$	$(4,16)$	$(5,9)$	$(6,10)$	$(7,11)$	$(8,12)$
$P_{14} = (1,14)$	$(2,13)$	$(3,16)$	$(4,15)$	$(5,10)$	$(6,9)$	$(7,12)$	$(8,11)$
$P_{15} = (1,15)$	$(2,16)$	$(3,13)$	$(4,14)$	$(5,11)$	$(6,12)$	$(7,9)$	$(8,10)$
$P_{16} = (1,16)$	$(2,15)$	$(3,14)$	$(4,13)$	$(5,12)$	$(6,11)$	$(7,10)$	$(8,9)$

TABLE V. Eigenvalues of the vectors $q(k)$ under the operations P_t ($t=1, 2, \ldots, 16$).

	P_1	P_2	P_3	P_4	P_5	P_6	P_7	P_8	P_9	P_{10}	P_{11}	P_{12}	P_{13}	P_{14}	P_{15}	P_{16}
$q(1)$	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
$q(2)$	1	1	1	1	1	1	1	1	−1	−1	−1	−1	−1	−1	−1	−1
$q(3)$	1	−1	−1	1	−1	1	1	−1	1	−1	−1	1	−1	1	1	−1
$q(4)$	1	−1	−1	1	−1	1	1	−1	−1	1	1	−1	1	−1	−1	1
$q(5)$	1	1	−1	−1	1	1	−1	−1	1	1	−1	−1	1	1	−1	−1
$q(6)$	1	1	−1	−1	1	1	−1	−1	−1	−1	1	1	−1	−1	1	1
$q(7)$	1	1	1	1	−1	−1	−1	−1	1	1	1	1	−1	−1	−1	−1
$q(8)$	1	1	1	1	−1	−1	−1	−1	−1	−1	−1	−1	1	1	1	1
$q(9)$	1	−1	1	−1	1	−1	1	−1	1	−1	1	−1	1	−1	1	−1
$q(10)$	1	−1	1	−1	1	−1	1	−1	−1	1	−1	1	−1	1	−1	1
$q(11)$	1	1	−1	−1	−1	−1	1	1	1	1	−1	−1	−1	−1	1	1
$q(12)$	1	1	−1	−1	−1	−1	1	1	−1	−1	1	1	1	1	−1	−1
$q(13)$	1	−1	−1	1	1	−1	−1	1	1	−1	−1	1	1	−1	−1	1
$q(14)$	1	−1	−1	1	1	−1	−1	1	−1	1	1	−1	−1	1	1	−1
$q(15)$	1	−1	1	−1	−1	1	−1	1	1	−1	1	−1	−1	1	−1	1
$q(16)$	1	−1	1	−1	−1	1	−1	1	−1	1	−1	1	1	−1	1	−1

of permutations, eigenvectors, and eigenvalues for the cases of $n=2^0$ and $n=2^1$ ions per unit cell gives the prescription of how to deduce the details for the case of $n=2^{m+1}$ from those for the case of $n=2^m$. For example, for $n=2^2$ one has, using $P^{i,i}$ ($i=1, 2, \ldots, 16$) for the case of $n=2^1$, the group of permutations

$$P^{j,j}=\begin{array}{|c|c|}\hline P^{i,i} & P^{i+8\times 2^1,\,i+8\times 2^1} \\ \hline \multicolumn{2}{|c|}{P^{i,\,i+8\times 2^1}} \\ \hline \end{array}$$

$(j=1, 2, \ldots, 32)$ (11)

and the corresponding eigenvectors

$$Q(2k-1)=\frac{1}{\sqrt{2}}\begin{pmatrix}q(k)\\ q(k)\end{pmatrix},\quad Q(2k)=\frac{1}{\sqrt{2}}\begin{pmatrix}q(k)\\ -q(k)\end{pmatrix}$$

$(k=1, 2, \ldots, 16)$, (12)

where $q(k)$ are the eigenvectors for the case $n=2^0$.

In general then, if $P^{i,i}$ denote the group of permutations for the case of $n=2^m$, the group of permutations for the case $n=2^{m+1}$ can be expressed as

$$P^{j,j}=\begin{array}{|c|c|}\hline P^{i,i} & P^{i+8\times 2^m,\,i+8\times 2^m} \\ \hline \multicolumn{2}{|c|}{P^{i,\,i+8\times 2^m}} \\ \hline \end{array}$$

$(j=1, 2, \ldots, 2^{m+1};\ i=1, 2, \ldots, 2^m)$ (13)

and the corresponding eigenvectors are

$$Q(2k-1)=\frac{1}{\sqrt{2}}\begin{pmatrix}q(k)\\ q(k)\end{pmatrix},\quad Q(2k)=\frac{1}{\sqrt{2}}\begin{pmatrix}q(k)\\ -q(k)\end{pmatrix}$$

$(k=1, 2, \ldots, 2^m)$, (14)

where $q(k)$ are the eigenvectors for the case $n=2^m$. In the form of Eq. (14) the $Q(j)$ constitute an orthonormal set.

D. Case for $n=n_0\times m$

(n_0 = odd interger different than 1, $m=1$ or an even integer). Let us first consider the case $m=1$. It is then not possible to express the permutations as products of cycles, each cycle containing two elements, each element occurring once only in each permutation, since the number of elements is odd. Thus the LT technique using permutation groups is not applicable to the case where n (>1) is odd. Also, to the case where n is an even multiple of an odd number, the LT method using permutation groups is not applicable since, as shown in Sec. III C the solution for the case $n=2n_0$ is obtained from that for the case $n=n_0$.

IV. SCOPE OF APPLICABILITY

The interactions and configuration of ions for which Eq. (6) is satisfied, i.e., to which the LT method using permutation groups is applicable, will be analyzed in this section.

A. Case for $n=2^0$ and 2^1

As discussed in Refs. 3 and 4, the symmetry of the matrix elements of A under the group of permutations P_t as expressed by Eq. (6) is satisfied for both the DD and exchange interactions.

B. Case for $n=2^2$

In order to simplify the discussion, the case of four ions distributed on a two-dimensional (2D) lattice will be presented first. (The results for the 3D lattice and for higher values of n can be easily derived using the same arguments as for the 2D lattice for $n=4$.) Let us designate the four ions within the unit cell by the letters A, B, C, D. In general, $ABCD$ form a quadrangle. In 2D the four ions generate sixteen sublattices by application of $\vec{\Gamma}'^2 = 2l\vec{a} + 2m\vec{b}$ (l, m = integers) to the vertices of four intercrossing parallelograms of dimensions $\vec{a}$, $\vec{b}$, one of the vertices of each parallelogram being located at one of the four ions A, B, C, D (see Fig. 1, which also depicts the labeling of the sixteen ions). The group of permutations relevant to this case is $P^{i,i}$ ($i=1,2,\ldots,16$) and is given in Table IV. It is easily seen from Table IV that, since the DD interaction between two ions i and j depends upon the vector $\vec{r}_{ij}$ joining them, not all the permutations P_t ($t=1,2,\ldots,16$) satisfy Eq. (6). For example, using P_2, we see that

$$A_{P_2 1 P_2 4} = A_{23} \neq A_{14} , \tag{15}$$

since $\vec{r}_{23} \neq \vec{r}_{14}$. (This is true even if $ABCD$ forms a parallelogram.) It is then concluded that for the DD interaction it is not possible to treat the case of $n=4$ for a 2D lattice by the LT method, using the permutation groups. Similarly, for $n=2^m$ ($m>2$), DD interactions cannot be treated by this method in either 2D or 3D.

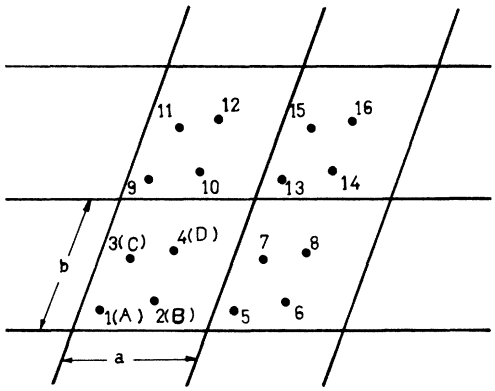

FIG. 1. Case of four ions per unit cell of a two-dimensional Bravais lattice. The configuration and labeling of the ions giving rise to the sixteen sublattice is indicated.

On the other hand, for the NN and NNN exchange interactions only between the ions, which depend on the length of the vector, rather than the vector itself, separating the two ions (the case of NN exchange interaction only is obtained by putting the NNN exchange interaction to zero), it is possible to treat by the LT method the case $n=4$ for a 2D lattice provided that (a) the A, B, C, D form a parallelogram with unequal sides, and (b) the diagonals AC and BD are each of greater length than either of the sides AB and AC. This can be seen by noting that the group of permutations relevant to this case (Table IV) transforms any side of the four parallelograms ($ABCD$ and images) of Fig. 1 into a parallel side (in the same figure), whereas any diagonal of the parallelograms $ABCD$ and its images is transformed into some parallel or nonparallel diagonal of the same four parallelograms. Thus, if in the element $A_{ij}^{\alpha\beta}$, ij represents a side of any of the parallelograms $ABCD$ and its images, then it will correspond to either NN or NNN exchange interaction, and for this element Eq. (6) will be satisfied, since under the group of permutations the length r_{ij} remains invariant. However, if ij represents any diagonal of $ABCD$ or of its images, then $A_{ij}^{\alpha\beta}$ under the operation of permutations goes into $A_{i'j'}^{\alpha\beta}$, where either $\vec{r}_{ij} = \vec{r}_{i'j'}$ (parallel diagonals) or $\vec{r}_{ij} \neq \vec{r}_{i'j'}$ (nonparallel diagonals of $ABCD$ and its images). Fulfillment of Eq. (6) then requires that both the diagonals AC and BD be greater in length than both the sides AB and AC so that $A_{ij}^{\alpha\beta} = A_{i'j'}^{\alpha\beta} = 0$. [In particular, note that for the NN and NNN exchange interaction

$$A_{P_2 1 P_2 4} = A_{23} = 0 = A_{14} , \tag{16}$$

in contrast to Eq. (15).]

It is also seen, using the same arguments and considering only the NN exchange interaction, that the method is applicable to three additional configurations: (i) Sides AB and AC of the parallelogram are equal to each other, but smaller than the diagonals AD and BC. (ii) one of the diagonals, say BC, is smaller in length than one of the sides, say AB, but greater in length than the other side AC. (iii) $ABCD$ is not a parallelogram; however, two opposite sides of it are equal in length, but smaller than both the diagonals and both the remaining sides.

We now consider the case of four ions per unit cell distributed on a 3D lattice, requiring a group of 32 permutations. The four ions per unit cell generate 32 sublattices by application of $\vec{\Gamma}^2$ to the vertices of four intercrossing parallelepipeds of dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$, one of the vertices of each parallelepiped being situated at one of the four ions A, B, C, D. (Note that here $ABCD$, which, in general, constitute the vertices of a tetrahedron, has seven images.) With regard to the NN and NNN exchange interactions between the ions, the LT meth-

od is applicable to the following configurations, taking into account the fact that under the operation of permutations each side s of the tetrahedron is transformed into the side s' of the tetrahedron which does not have a common vertex with s or into the images of s and s'. The four smallest sides (of the six) of the tetrahedron constitute two pairs a, b; each pair consists of two sides (without a common vertex) of equal length. The lengths of the remaining two sides may or may not be the same. (The case when the smallest two sides of the tetrahedron are equal in length and do not have a common vertex and the remaining sides of the tetrahedron are unequal in length can be treated for the NN exchange interaction only.) When the sides of the tetrahedron constitute three pairs a, b, c, the sides of each pair are equal in length and do not have a common vertex, then two of the three pairs could be of the same length, different from that of the third pair. [When a, b, c are of the same length, the case of NN exchange interaction only can be treated. On the other hand, when the lengths of a, b, c are unequal the cases of NN, NNN, and NNNN (next-next-nearest neighbor) can also be treated.]

When the four ions lie in a plane, in a 3D lattice, the configurations (a) and (b) as stated for the 2D case must be satisfied for the NN and NNN exchange interactions only. The method is also applicable, considering only the NN exchange interaction, to the configurations satisfying the conditions (i), (ii), and (iii) for the 2D lattice discussed above. [In the following discussion the term tetrahedron also includes the case of parallelogram (or quadrangle) when the four ions lie in one plane.]

C. Case for $n = 2^3$

For the applicability of the method it is necessary that the eight ions per unit cell lie on the vertices of two tetrahedrons of equal dimensions, not necessarily parallel to each other. The whole lattice is divided into 64 sublattices by the application of $\vec{\Gamma}^2$ to the 64 vertices of 8 intercrossing parallelepipeds each of dimensions $\vec{a}$, $\vec{b}$, $\vec{c}$, one of the vertices of each parallelepiped being situated at one of the eight ions; the relevant group of permutations consists of 64 elements. Depending upon the relative lengths of the various sides of the constituting tetrahedrons the cases of NN-only, NN- and NNN-only, and NN-, NNN-, NNNN-only exchange interactions can be treated in the same way as discussed for the case $n = 4$, provided that the two tetrahedrons are either (i) "far enough" apart (not necessarily parallel to each other), implying that the length of any intertetrahedron side (a line joining any vertex of one tetrahedron with any vertex of the other tetrahedron in the unit cell) is large enough not to contribute to NN, NNN (and NNNN) exchange interactions, or (ii) "sufficiently close" and parallel to each other, where the term "sufficiently close" implies that some intertetrahedron sides correspond to NN or NNN (or NNNN) exchange interaction such that the resulting elements of A satisfy Eq. (16).

Special cases occur when the eight ions occupy the vertices of a parallelepiped or a prism having a trapezoidal cross section. For the case of the parallelepiped, the various conditions are (a) the body and face diagonals are larger than the sides. The following situations can then be treated. When the three sets, consisting of four equal sides of the parallelepiped each, are of unequal lengths, all NN, NNN, and NNNN exchange interactions can be treated; when two of the sets are of equal length, different from the length of the third set, only the NN and NNN exchange interactions can be treated; and when all the three sets are of equal length, only the NN exchange interaction can be treated. (b) When there is one set t smaller in length than any of the other two sets, and the two faces f of the parallelepiped containing t are such that one of their diagonals is larger in length than the length of t but smaller than those sides of f which are different from t, then only the NN exchange interaction can be treated. (c) For the case of the prism, with a trapezoidal cross section, the conditions are as follows: (i) the NN and NNN exchange interactions can be treated when any opposite sides s_1 of the trapezoid are equal, s_1 being smaller than the other two sides s_2 and the lengths of both the sides s_2 are greater than the height of the prism. (ii) Only the NN exchange interaction can be treated when all the conditions remain the same as in (i), except that here the prism height is greater than the lengths of both the sides s_2. [In deducing (a), (b), and (c), use has been made of the fact that under the operation of the group of permutations, any side of the parallelepiped transforms into a parallel side of the parallelepiped and any face diagonal transforms into the other diagonal of the same face or into any of the two diagonals of the opposite face. Similar considerations apply to the transformation of the prism under the group of permutations.]

D. Case for $n = 2^4$

The method becomes applicable when the sixteen ions lie on the vertices of four tetrahedrons identical in dimensions (128 sublattices, 128 elements in the group of permutations) which are either "far enough" apart from each other, or obtained by a "far enough" displacement, not necessarily parallel, of a unit of two parallel "sufficiently close" tetrahedrons, or if the two units are not "far enough" apart, they are parallel and "sufficiently close" to each other. (The various exchange interactions

can be considered, depending upon the relative lengths of sides of the constituting tetrahedrons, in the same way as discussed for the case $n = 4$.)

Special cases occur when the sixteen ions per unit cell are located at the vertices of two separated identical parallelepipeds or two separated identical prisms having trapezoidal cross sections. The various conditions regarding the distances between the ions can be deduced in the same way as for $n = 2^3$.

E. Case for $n = 2^m$ ($m \geq 5$)

Generalizing in the same fashion as for the cases in Secs. IV A–IV D, it is seen that the method is applicable under appropriate conditions regarding the relative lengths of the sides of the constituting tetrahedrons when the ions are located at the vertices of 2^{m-2} separated tetrahedrons of identical dimensions, which may or may not be parallel to each other, depending upon the intertetrahedron distances. (There are 2^{m+3} sublattices and 2^{m+3} elements in the group of permutations.)

Special cases occur when the ions in the unit cell are located at the vertices of 2^{m-2} separated parallelepipeds or prisms with trapezoidal cross sections. The conditions regarding the distances between the various ions in the unit cell can be deduced in the same way as discussed in the preceding examples.

V. CONCLUDING REMARKS

The main points of the investigation presented in this paper regarding the general applicability of the LT method using permutation groups can be summarized as follows.

(i) It is possible to treat the cases of one and two magnetically equivalent ions per unit cell for both the DD, NN, and NNN exchange interactions.

(ii) It is also possible to treat, for the NN and NNN exchange interactions, under appropriate conditions regarding the configuration of the ions in the unit cell as discussed in Sec. IV, those cases of magnetically equivalent ions where the number n of ions per unit cell can be expressed in the form $n = 2^m$ ($m > 1$).

The conclusion (i) above has also been arrived at by Niemeyer and Blöte [5] using the properties of their matrix Y. Their method, however, fails to bring out the conclusion stated in (ii) above.

It should be remarked that there may be cases of more complicated configurations than discussed in this paper for $n = 2^m$ ($m \geq 3$) where the method is applicable. Nevertheless, the configurations of ions in a unit cell discussed here do indeed amply demonstrate the general applicability of the LT method using permutation groups.

ACKNOWLEDGMENTS

The author is grateful to the National Research Council of Canada for financial support and to Dr. B. Frank for a critical reading of the manuscript and useful comments.

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[1] J. M. Luttinger and L. Tisza, Phys. Rev. 70, 954 (1946).
[2] J. M. Daniels and J. Felsteiner, Can. J. Phys. 42, 1469 (1964), application to cerium magnesium nitrate (CMN); J. Felsteiner and A. Rabinovitch, Solid State Commun. 7, 1649 (1969), application to CMN, neodymium ethyl sulfate (NdES), and DyES; J. Felsteiner, J. Chem. Phys. 52, 2784 (1970), application to $Tb(OH)_3$, $Dy(OH)_3$, $Ho(OH)_3$, $Nd(OH)_3$, $Gd(OH)_3$, and $Er(OH)_3$; J. Felsteiner, J. Phys. C 3, L174 (1970), application to $ErCl_3 \cdot 6H_2O$; E. Lagendijk, H. W. Blöte, and W. J. Huiskamp, Physica (Utr.) 61, 220 (1972), application to CMN, DyES, ErEs, and $ErCl_3 \cdot 6H_2O$; see also Ref. 3 for an application to CMN, and Ref. 4 for application to $GdCl_3$, $Gd(OH)_3$, $Tb(OH)_3$, $Ho(OH)_3$, $Nd(OH)_3$, and $Er(OH)_3$.
[3] Th. Niemeyer, Physica (Utr.) 57, 281 (1972).
[4] J. Felsteiner and S. K. Misra, Phys. Rev. B (to be published); also see S. K. Misra and J. Felsteiner, Phys. Rev. B (to be published) for application to $GdCl_3 \cdot 6H_2O$.
[5] Th. Niemeyer and H. W. J. Blöte, Physica (Utr.) (to be published).