Approximate Theory of Ferrimagnetic Spin Waves*

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The wave functions and the energy spectrum for the spin-wave problem in a normal spinel are found by means of a straightforward extension of Anderson's approach to antiferromagnetism. Only $A-B$ exchange is assumed to exist, and the calculation is carried to second order in the magnitude of the propagation vector, $\mathbf{k}$. Five distinct energy surfaces (E *vs* k) are found, two of which are identical, in the classical limit, to the ones previously reported by H. Kaplan. Although the energy surfaces are all spherically symmetric, the wave functions depend in general on the direction of $\mathbf{k}$. The error in a calculation by Vonsovski and Seidov, which led to a linear energy *vs* k relation for the lowest branch (as opposed to the quadratic relation found by H. Kaplan), is pointed out.

I. INTRODUCTION

CURRENT experiments being performed by Brockhouse[1] on the inelastic scattering of neutrons by ferrites have stimulated interest in the theory of ferrimagnetic spin waves. The theoretical situation has been somewhat confused since there are in the literature two papers which give contradictory results. In one of these, H. Kaplan[2] applies the semiclassical spin-wave theory in the form presented for example by Herring and Kittel,[3] to the normal spinel structure. By assuming that the spin-wave amplitudes for the tetrahedral (A) sites are equal and those for the octahedral (B) sites are equal, he finds two branches in the energy (E) *vs* propagation vector ($\mathbf{k}$) relation. The lower one, for small k, is quadratic in k. In the other paper, Vonsovski and Seidov[4] use the approach of Holstein and Primakoff[5] or Anderson.[6,7] Again assuming equal spin-wave amplitudes for the A sites and for the B sites, they obtain two branches in the E *vs* k curve. However, when the $A-A$ and $B-B$ interactions are put equal to zero in their result, their lower branch is linear in k for small k, in serious contradiction to the result of H. Kaplan. Because of experimental uncertainty, Brockhouse[1] was not able to decide definitely between the two relations; however, his data tended to favor H. Kaplan's result.

The situation is resolved when one notes an elementary, but essential error in the calculation of Vonsovski and Seidov. Namely, their assumption of equal spin-wave amplitudes on the A sites and on the B sites is incorrectly made in the Hamiltonian (whereas H. Kaplan does it in the equations of motion). The fact that the two results differ is simply a matter, speaking classically, of whether one differentiates the Hamiltonian (to obtain the equations of motion) before or after equality between various independent variables is assumed.[8] Stated alternatively, one sees in their transformation of variables given by Eq. (1,8), in the translation[4] of their work, where they assume the equality of the various spin wave amplitudes, and which they then use in the Hamiltonian, the total number of independent variables is reduced.

It is felt that the calculation of the full energy spectrum and the wave functions (with the removal of the *a priori* assumption as to the equality of any spin wave amplitudes) will be of interest. Knowledge of the wave functions, for example, will enable one to calculate the neutron scattering cross section, which should be useful in connection with current experiments. Consequently we carry out this calculation in the present paper. Our method is a straightforward extension of that used by Anderson[6] for the antiferromagnetic case; our model is a normal spinel in which only $A-B$ exchange is assumed to exist. The results are obtained to second order in k.

Five energy surfaces (E *vs* k) are found, two of which in the classical limit of large spin quantum numbers, are, as expected, identical to those found by H. Kaplan (when the $A-A$ and $B-B$ interaction terms are neglected in the latter). Although all the energy surfaces are spherically symmetric, the normal coordinates (and therefore the wave functions), with the exception of those corresponding to H. Kaplan's surfaces, depend on the direction, $\hat{k}$, of $\mathbf{k}$. This is physically reasonable since the properties of the lattice depend on $\hat{k}$. For example, the densities of B sites in adjacent planes of constant $\mathbf{k}\cdot\mathbf{r}$ will be in the ratio 3:1 when $\mathbf{k}$ is in the (1,1,1) direction, while this ratio will be 1:1 for $\mathbf{k}$ in a

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[1] B. N. Brockhouse, Phys. Rev. **106**, 859 (1957).
[2] H. Kaplan, Phys. Rev. **86**, 121 (1952). Although an arbitrary distribution of two types of paramagnetic ions among the A and B sites was considered, the sites in a given sublattice were assumed to be mathematically equivalent, this being realized by an averaging process. This is the same as assuming that one is dealing with a normal spinel structure and that furthermore one is considering only those normal modes in which the spin wave amplitudes are the same within a given sublattice.
[3] C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).
[4] S. V. Vonsovski and Y. M. Seidov, Izvest. Acad. Nauk S.S.S.R. **18**, 319 (1954). Translation available through Columbia Technical Translations.
[5] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).
[6] P. W. Anderson, Phys. Rev. **86**, 694 (1952).
[7] Ryogo Kubo [Phys. Rev. **87**, 568 (1952)] has shown the equivalence of these approaches.
[8] This may be seen in terms of the formalism of the present paper. The "before" and "after" methods indeed lead to quadratic and linear relations, respectively, when the spin quantum numbers for all sites are equal (which is the case considered by Vonsovski and Seidel).

cubic direction. In connection with the higher energy surfaces, it is noted that, from energy considerations alone, neutrons with wavelength of the order of 1 to 2 A have sufficient energy to excite them. (See Sec. IV.)

II. THE HAMILTONIAN

We begin with the Heisenberg exchange Hamiltonian,

$$H=J\sum_{\langle i,j\rangle}\mathbf{S}_i^A\cdot\mathbf{S}_j^B, \tag{1}$$

which is appropriate to a normal spinel structure assuming interaction only between nearest neighbor $A-B$ pairs. $\mathbf{S}_i^A$ and $\mathbf{S}_j^B$ are the spin operators (in units of $\hbar$) for the tetrahedral and the octahedral atoms, respectively. The summation is over the nearest neighbors j of i, followed by the summation of i over the A sites. We make the approximation (following Anderson)[6]

$$S_i^{Az'}\cong S_A,\quad S_i^{Bz'}\cong -S_B, \tag{2}$$

where S_A and S_B are the spin quantum numbers associated, respectively, with the A and B sites, and z' refers to an arbitrary direction. Then, for large S_A and S_B,[9]

$$\begin{aligned} S_i^{Az'}&\cong S_c^A-\frac{(S_i^{Ax'})^2+(S_i^{Ay'})^2}{2S_c^A},\\ S_j^{Bz'}&\cong -S_c^B+\frac{(S_j^{Bx'})^2+(S_j^{By'})^2}{2S_c^B}, \end{aligned} \tag{3}$$

where

$$S_c^A=[S_A(S_A+1)]^{\frac{1}{2}},\quad S_c^B=[S_B(S_B+1)]^{\frac{1}{2}}. \tag{4}$$

Equation (1) becomes, dropping a constant term and neglecting higher order terms in the x' and y' components,

$$\begin{aligned} H=&\tfrac{1}{2}JZ_A\left(\frac{S_c^B}{S_c^A}\right)\sum_i[(S_i^{Ax'})^2+(S_i^{Ay'})^2]\\ &+\tfrac{1}{2}JZ_B\left(\frac{S_c^A}{S_c^B}\right)\sum_i[(S_j^{Bx'})^2+(S_j^{By'})^2]\\ &+J\sum_{\langle i,j\rangle}(S_i^{Ax'}S_j^{Bx'}+S_i^{Ay'}S_j^{By'}), \end{aligned} \tag{5}$$

where Z_A and Z_B are 12 and 6, the numbers of nearest neighbors "seen" by the A and the B sites, respectively. Note that (5) reduces to Anderson's Eq. (6) for the antiferromagnetic structures which he considers.

The six magnetic atoms in a primitive unit cell for the spinel structure[10] are shown in Fig. 1. The primitive

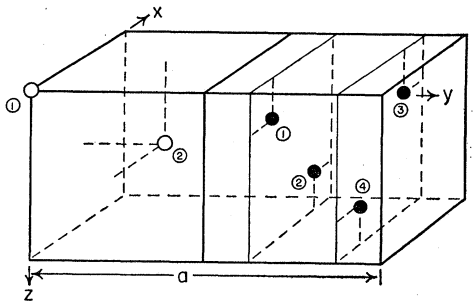

FIG. 1. The sites in a primitive unit cell of the spinel structure. ○ $=A$ sites; ● $=B$ sites. (Oxygen not shown.)

translations may be taken as

$$\mathbf{a}_1=\tfrac{1}{2}a(1,1,0),\quad \mathbf{a}_2=\tfrac{1}{2}a(0,1,1),\quad \mathbf{a}_3=\tfrac{1}{2}a(1,0,1), \tag{6}$$

where the numbers in parentheses refer to the x, y, and z components, respectively. The atoms are located at

$$\begin{aligned} &\varrho_1^A=0, && \varrho_2^A=\tfrac{1}{4}a(1,1,1),\\ &\varrho_1^B=\tfrac{1}{8}a(1,5,1), && \varrho_2^B=\tfrac{1}{8}a(3,5,3),\\ &\varrho_3^B=\tfrac{1}{8}a(3,7,1), && \varrho_4^B=\tfrac{1}{8}a(1,7,3). \end{aligned} \tag{7}$$

Each of the two cubes shown in Fig. 1 are octants of the usual cubic unit cell. Any two such octants sharing an edge are identical, any two sharing a face are different.

The general location of the sites is

$$\begin{aligned} \mathbf{r}_\alpha^A(n)&=\mathbf{R}_n+\varrho_\alpha^A,\quad \alpha=1,2,\\ \mathbf{r}_\beta^B(n)&=\mathbf{R}_n+\varrho_\beta^B,\quad \beta=1,2,3,4, \end{aligned} \tag{8}$$

where the lattice translations are

$$\mathbf{R}_n=n_1\mathbf{a}_1+n_2\mathbf{a}_2+n_3\mathbf{a}_3,\quad n_i=1,2,\cdots,N_i, \tag{9}$$

and

$$N=N_1N_2N_3=\text{total No. of unit cells.} \tag{10}$$

We now make the transformation to the Fourier transforms (spin-wave amplitudes) of the spin variables:

$$\begin{aligned} S^{Ay'}[\mathbf{r}_\alpha^A(n)]&=(S_A/N)^{\frac{1}{2}}\textstyle\sum_{\mathbf{k}}\exp[i\mathbf{k}\cdot\mathbf{r}_\alpha^A(n)]P_\alpha^A(\mathbf{k}),\\ S^{Ax'}[\mathbf{r}_\alpha^A(n)]&=(S_A/N)^{\frac{1}{2}}\textstyle\sum_{\mathbf{k}}\exp[-i\mathbf{k}\cdot\mathbf{r}_\alpha^A(n)]Q_\alpha^A(\mathbf{k}),\\ S^{By'}[\mathbf{r}_\beta^B(n)]&=-(S_B/N)^{\frac{1}{2}}\textstyle\sum_{\mathbf{k}}\exp[-i\mathbf{k}\cdot\mathbf{r}_\beta^B(n)]P_\beta^B(\mathbf{k}),\\ S^{Bx'}[\mathbf{r}_\beta^B(n)]&=(S_B/N)^{\frac{1}{2}}\textstyle\sum_{\mathbf{k}}\exp[i\mathbf{k}\cdot\mathbf{r}_\beta^B(n)]Q_\beta^B(\mathbf{k}), \end{aligned} \tag{11}$$

where

$$\begin{aligned} &\mathbf{k}=2\pi\sum_{i=1}^{3}\left(\frac{m_i}{N_i}\right)\mathbf{b}^i,\\ &\mathbf{a}_i\cdot\mathbf{b}^j=\delta_{ij},\\ &m_i=0,\pm1,\pm2,\cdots,\pm(\tfrac{1}{2}N_i-1),\tfrac{1}{2}N_i. \end{aligned} \tag{12}$$

The inverses follow from

$$\textstyle\sum_n\exp[i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_n]=N\delta_{\mathbf{k}\mathbf{k}'}. \tag{13}$$

[9] Equation (3) is the central approximation of the theory. Anderson's justification for the antiferromagnetic ground state, as well as F. J. Dyson's recent work [Phys. Rev. **102**, 1217 (1956)], which justifies the spin-wave picture for long wavelengths and small excitation in the ferromagnetic case, indicates that the approximation is probably valid for the present case.

[10] For a detailed discussion of the spinel structure see, for example, E. W. Gorter, Philips Research Repts. **9**, 295 (1954).

Using (2), we find for the commutators

$$[P_\alpha{}^A(\mathbf{k}),Q_{\alpha'}{}^A(\mathbf{k}')]=[P_\alpha{}^B(\mathbf{k}),Q_{\alpha'}{}^B(\mathbf{k}')] =-i\delta_{\alpha\alpha'}\delta_{\mathbf{k}\mathbf{k}'}, \quad (14)$$

with all other pairs of variables commuting.

Note that we have N values of $\mathbf{k}$ and therefore $6N$ Q's and $6N$ P's, which corresponds correctly to the $12N$ variables $S_i{}^{Ax}$, $S_i{}^{Ay}$, $S_i{}^{Bx}$, $S_i{}^{By}$. Equations (11) are now to be substituted in (5). Sample terms are

$$\sum_i (S_i{}^{Ax'})^2=S_A\sum_{\mathbf{k}}\sum_\alpha Q_\alpha{}^A(\mathbf{k})Q_\alpha{}^A(-\mathbf{k}), \quad (15)$$

and

$$\sum_{\langle i,j\rangle} S_i{}^{Ax'}S_j{}^{Bx'}=\frac{1}{N}(S_AS_B)^{\frac{1}{2}}\sum_{\langle n,\alpha;n',\beta\rangle}\sum_{\mathbf{k},\mathbf{k}'}\exp[-i\mathbf{k}\cdot\mathbf{r}_\alpha{}^A(n) +i\mathbf{k}'\cdot\mathbf{r}_\beta{}^B(n')]Q_\alpha{}^A(\mathbf{k})Q_\beta{}^B(\mathbf{k}'). \quad (16)$$

To treat the latter equation, write

$$\mathbf{r}_\beta{}^B(n')=\mathbf{r}_\alpha{}^A(n)+\boldsymbol{\tau}_{\alpha\beta}, \quad (17)$$

where $\boldsymbol{\tau}_{\alpha\beta}$ is independent of n. Then

$$\sum_{\langle i,j\rangle} S_i{}^{Ax'}S_j{}^{Bx'}=(S_AS_B)^{\frac{1}{2}}\sum_{\mathbf{k}}\sum_{\langle\alpha,\beta\rangle} Q_\alpha{}^A(\mathbf{k})Q_\beta{}^B(\mathbf{k}) \times\exp(i\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}), \quad (18)$$

where

$$\sum_{\langle\alpha,\beta\rangle}=\text{sum over the nearest neighbors }\beta\text{ of }\alpha\text{ and sum }\alpha \text{ over the unit cell} =\sum_{\langle\beta,\alpha\rangle}.$$

Referring to Fig. 1, we see that

$$\sum_{\langle\alpha,\beta\rangle} Q_\alpha{}^A(\mathbf{k})Q_\beta{}^B(\mathbf{k})\exp(i\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}) =\sum_{\alpha=1}^{2}\sum_{\beta=1}^{4}\zeta_{\alpha\beta}(\mathbf{k})Q_\alpha{}^A(\mathbf{k})Q_\beta{}^B(\mathbf{k}), \quad (19)$$

where

$$\zeta_{\alpha\beta}(\mathbf{k})=\sum_{u=1}^{3}\exp(i\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}{}^u), \quad (20)$$

and $\boldsymbol{\tau}_{\alpha\beta}{}^u$, $u=1, 2, 3$, are the vectors connecting the αth A site to the three nearest neighbor B sites characterized by a given $\boldsymbol{\rho}_\beta{}^B$. The $\boldsymbol{\tau}_{\alpha\beta}{}^u$ are given explicitly in Eq. (26).

Thus (5) becomes

$$H=\sum_{\mathbf{k}}\Big\{\frac{A}{2}\sum_{\alpha=1}^{2}[P_\alpha{}^A(\mathbf{k})P_\alpha{}^A(-\mathbf{k})+Q_\alpha{}^A(\mathbf{k})Q_\alpha{}^A(-\mathbf{k})] +\frac{B}{2}\sum_{\beta=1}^{4}[P_\beta{}^B(\mathbf{k})P_\beta{}^B(-\mathbf{k})+Q_\beta{}^B(\mathbf{k})Q_\beta{}^B(-\mathbf{k})] +\gamma\sum_{\alpha=1}^{2}\sum_{\beta=1}^{4}[\zeta_{\alpha\beta}(\mathbf{k})Q_\alpha{}^A(\mathbf{k})Q_\beta{}^B(\mathbf{k}) -\zeta_{\alpha\beta}(-\mathbf{k})P_\alpha{}^A(\mathbf{k})P_\beta{}^B(\mathbf{k})]\Big\}, \quad (21)$$

where

$$A=JZ_AS_c{}^B(S_A/S_c{}^A),\quad B=JZ_BS_c{}^A(S_B/S_c{}^B),\quad \gamma=J(S_AS_B)^{\frac{1}{2}}. \quad (22)$$

We now make the canonical transformation to the sine and cosine transforms,

$$\begin{aligned} P_\alpha{}^{A+}(\mathbf{k})&=2^{-\frac{1}{2}}[P_\alpha{}^A(\mathbf{k})+P_\alpha{}^A(-\mathbf{k})],\\ Q_\alpha{}^{A+}(\mathbf{k})&=2^{-\frac{1}{2}}[Q_\alpha{}^A(\mathbf{k})+Q_\alpha{}^A(-\mathbf{k})],\\ P_\alpha{}^{A-}(\mathbf{k})&=-i2^{-\frac{1}{2}}[P_\alpha{}^A(\mathbf{k})-P_\alpha{}^A(-\mathbf{k})],\\ Q_\alpha{}^{A-}(\mathbf{k})&=i2^{-\frac{1}{2}}[Q_\alpha{}^A(\mathbf{k})-Q_\alpha{}^A(-\mathbf{k})], \end{aligned} \quad (23)$$

with similar definitions for $P_\alpha{}^{B\pm}$ and $Q_\alpha{}^{B\pm}$. (The $+$ and $-$ variables are, respectively, even and odd in $\mathbf{k}$ to $-\mathbf{k}$.) Then

$$\begin{aligned} H=\sum{}'_{\mathbf{k}}\{&\tfrac{1}{2}A\textstyle\sum_\alpha[(P_\alpha{}^{A+})^2+(P_\alpha{}^{A-})^2+(Q_\alpha{}^{A+})^2+(Q_\alpha{}^{A-})^2]\\ &+\tfrac{1}{2}B\textstyle\sum_\beta[(P_\beta{}^{B+})^2+(P_\beta{}^{B-})^2+(Q_\beta{}^{B+})^2+(Q_\beta{}^{B-})^2]\\ &+\gamma\textstyle\sum_{\alpha,\beta}\zeta_{\alpha\beta}{}^e(Q_\alpha{}^{A+}Q_\beta{}^{B+}-P_\alpha{}^{A+}P_\beta{}^{B+}-Q_\alpha{}^{A-}Q_\beta{}^{B-}\\ &+P_\alpha{}^{A-}P_\beta{}^{B-})+\gamma\textstyle\sum_{\alpha,\beta}\zeta_{\alpha\beta}{}^o(Q_\alpha{}^{A-}Q_\beta{}^{B+}\\ &-P_\alpha{}^{A-}P_\beta{}^{B+}+Q_\alpha{}^{A+}Q_\beta{}^{B-}-P_\alpha{}^{A+}P_\beta{}^{B-})\}, \end{aligned} \quad (24)$$

where $\sum'_{\mathbf{k}}$ means to sum over half the Brillouin zone [Eq. (12)], such that, if $\mathbf{k}$ is in the set, $-\mathbf{k}$ is not

$$\zeta_{\alpha\beta}{}^e=\sum_{u=1}^{3}\cos(\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}{}^u),\quad \zeta_{\alpha\beta}{}^o=\sum_{u=1}^{3}\sin(\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}{}^u), \quad (25)$$

and the arguments $(\mathbf{k})$ of all the functions have been omitted in the notation. It is interesting to note that the nonvanishing of $\zeta_{\alpha\beta}{}^o$ connects the sine and cosine transforms. This corresponds to the nonvanishing of the first derivative terms in the Herring-Kittel semiclassical equations, and corresponds physically to the fact that the two A sites in our unit cell are not equivalent lattice points and the four B sites are not equivalent lattice points.

The $\boldsymbol{\tau}_{\alpha\beta}{}^u$, in units of $a/8$, are given explicitly by

$$\begin{aligned} \boldsymbol{\tau}_{11}{}^u&=(1,1,-3),(1,-3,1),(-3,1,1);\\ \boldsymbol{\tau}_{12}{}^u&=(-1,1,3),(-1,-3,-1),(3,1,-1);\\ \boldsymbol{\tau}_{13}{}^u&=(-1,-1,-3),(-1,3,1),(3,-1,1);\\ \boldsymbol{\tau}_{14}{}^u&=(1,-1,3),(1,3,-1),(-3,-1,-1); \end{aligned} \quad (26)$$

with

$$\boldsymbol{\tau}_{1\beta}{}^u=-\boldsymbol{\tau}_{2\beta}{}^u,\quad u=1,2,3;\quad \beta=1,2,3,4. \quad (27)$$

It may be noted that (27) is consistent with the well-known fact that every B site is a center of inversion; i.e., if some B is connected to an A by $\boldsymbol{\tau}$, then $-\boldsymbol{\tau}$ connects that B with another A. The pair of A's, by (27), will always be an A_1 and an A_2. On the other hand, the fact that there is no $\boldsymbol{\tau}_{1\beta}{}^u$ which is $-\boldsymbol{\tau}_{1\beta}{}^{u'}$ is consistent with the fact that the A's are *not* inversion centers.

From Eq. (27) it follows that

$$\zeta_{1\beta}{}^e=\zeta_{2\beta}{}^e, \quad (28)$$

and

$$\zeta_{1\beta}{}^{o}=-\zeta_{2\beta}{}^{o}.$$

We may be interested in the calculation to second order in k. Hence we may put

$$\zeta_{\alpha\beta}{}^{e}=3-\tfrac{1}{2}\sum_{u=1}^{3}(\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}{}^{u})^2;\quad \zeta_{\alpha\beta}{}^{o}=\sum_{u=1}^{3}\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}{}^{u}. \tag{29}$$

Then

$$\begin{aligned}
\zeta_{11}{}^{o}&=-\tfrac{1}{8}a(k_x+k_y+k_z),\\
\zeta_{12}{}^{o}&=\tfrac{1}{8}a(k_x-k_y+k_z),\\
\zeta_{13}{}^{o}&=\tfrac{1}{8}a(k_x+k_y-k_z),\\
\zeta_{14}{}^{o}&=\tfrac{1}{8}a(-k_x+k_y+k_z),
\end{aligned} \tag{30}$$

and

$$\sum_{u=1}^{3}(\mathbf{k}\cdot\boldsymbol{\tau}_{\alpha\beta}{}^{u})^2=\frac{11k^2a^2}{64}+x_{\alpha\beta}, \tag{31}$$

where

$$\begin{aligned}
x_{11}&=-\tfrac{5}{32}a^2(k_xk_y+k_xk_z+k_yk_z),\\
x_{12}&=\tfrac{5}{32}a^2(k_xk_y-k_xk_z+k_yk_z),\\
x_{13}&=\tfrac{5}{32}a^2(-k_xk_y+k_xk_z+k_yk_z),\\
x_{14}&=\tfrac{5}{32}a^2(k_xk_y+k_xk_z-k_yk_z),
\end{aligned} \tag{32}$$

and

$$x_{1\beta}=x_{2\beta}. \tag{33}$$

Hence the coefficients in the Hamiltonian (24) are anisotropic in $\mathbf{k}$.

It follows that

$$\textstyle\sum_\alpha \zeta_{\alpha\beta}{}^{o}=\sum_\beta \zeta_{\alpha\beta}{}^{o}=0, \tag{34}$$

and

$$\textstyle\sum_\beta x_{\alpha\beta}=0. \tag{35}$$

Also

$$\zeta_{\alpha\beta}{}^{e}=3-(11/128)k^2a^2-\tfrac{1}{2}x_{\alpha\beta}\equiv\eta-\tfrac{1}{2}x_{\alpha\beta}. \tag{36}$$

III. THE NORMAL MODES

Referring to the Hamiltonian (24), one sees that the normal coordinates and frequencies are to be found for a 12-variable problem (fixed $\mathbf{k}$). Fortunately there is enough symmetry in the equations to enable one to find these coordinates by inspection when the coefficients are kept only to second order in k. However, for the sake of clarity, we shall first reduce the problem to three 4-variable problems, and then drop higher terms.

The normal coordinates can obviously be found by treating the P's and Q's as classical quantities. The equations of motion, using (24), are

$$\begin{aligned}
\dot{Q}_\alpha{}^{A+}&=AP_\alpha{}^{A+}-\gamma\textstyle\sum_\beta(\zeta_{\alpha\beta}{}^{e}P_\beta{}^{B+}+\zeta_{\alpha\beta}{}^{o}P_\beta{}^{B-}),\\
\dot{Q}_\beta{}^{B+}&=BP_\beta{}^{B+}-\gamma\textstyle\sum_\alpha(\zeta_{\alpha\beta}{}^{e}P_\alpha{}^{A+}+\zeta_{\alpha\beta}{}^{o}P_\alpha{}^{A-}),\\
\dot{Q}_\alpha{}^{A-}&=AP_\alpha{}^{A-}+\gamma\textstyle\sum_\beta(\zeta_{\alpha\beta}{}^{e}P_\beta{}^{B-}-\zeta_{\alpha\beta}{}^{o}P_\beta{}^{B+}),\\
\dot{Q}_\beta{}^{B-}&=BP_\beta{}^{B-}+\gamma\textstyle\sum_\alpha(\zeta_{\alpha\beta}{}^{e}P_\alpha{}^{A-}-\zeta_{\alpha\beta}{}^{o}P_\alpha{}^{A+}),\\
\dot{P}_\alpha{}^{A+}&=-AQ_\alpha{}^{A+}-\gamma\textstyle\sum_\beta(\zeta_{\alpha\beta}{}^{e}Q_\beta{}^{B+}+\zeta_{\alpha\beta}{}^{o}Q_\beta{}^{B-}),\\
\dot{P}_\beta{}^{B+}&=-BQ_\beta{}^{B+}-\gamma\textstyle\sum_\alpha(\zeta_{\alpha\beta}{}^{e}Q_\alpha{}^{A+}+\zeta_{\alpha\beta}{}^{o}Q_\alpha{}^{A-}),\\
\dot{P}_\alpha{}^{A-}&=-AQ_\alpha{}^{A-}+\gamma\textstyle\sum_\beta(\zeta_{\alpha\beta}{}^{e}Q_\beta{}^{B-}-\zeta_{\alpha\beta}{}^{o}Q_\beta{}^{B+}),\\
\dot{P}_\beta{}^{B-}&=-BQ_\beta{}^{B-}+\gamma\textstyle\sum_\alpha(\zeta_{\alpha\beta}{}^{e}Q_\alpha{}^{A-}-\zeta_{\alpha\beta}{}^{o}Q_\alpha{}^{A+}).
\end{aligned} \tag{37}$$

By considering the constructs $\sigma_1{}^{\pm}=\sum_\alpha Q_\alpha{}^{A\pm}$, $\sigma_2{}^{\pm}=\sum_\beta \zeta_{1\beta}{}^{e}Q_\beta{}^{B\pm}$, $\sigma_3{}^{\pm}=Q_1{}^{A\mp}-Q_2{}^{A\mp}$, $\sigma_4{}^{\pm}=\sum_\beta \zeta_{1\beta}{}^{o}Q_\beta{}^{B\pm}$, using the exact properties (28) and Eqs. (37), it is easy to see that

$$\begin{aligned}
q^{\pm}&=\sum_{i=1}^{4}a_i{}^{\pm}\sigma_i{}^{\pm},\\
p^{\pm}&=a_1{}^{\pm}\tau_1{}^{\pm}-a_2{}^{\pm}\tau_2{}^{\pm}+a_3{}^{\pm}\tau_3{}^{\pm}-a_4{}^{\pm}\tau_4{}^{\pm}
\end{aligned} \tag{38}$$

are normal coordinates and their conjugate momenta. Here, $\tau_i{}^{\pm}$ are obtained from $\sigma_i{}^{\pm}$ by replacing Q by P; the $a_i{}^{\pm}$ are determined by

$$\begin{pmatrix} A & \mp\gamma\xi_e & 0 & \mp\gamma\xi' \\ \pm 2\gamma & -B & 0 & 0 \\ 0 & -\gamma\xi' & A & -\gamma\xi_o \\ 0 & 0 & 2\gamma & -B \end{pmatrix} \begin{pmatrix} a_1{}^{\pm} \\ a_2{}^{\pm} \\ a_3{}^{\pm} \\ a_4{}^{\pm} \end{pmatrix} = \omega \begin{pmatrix} a_1{}^{\pm} \\ a_2{}^{\pm} \\ a_3{}^{\pm} \\ a_4{}^{\pm} \end{pmatrix}, \tag{39}$$

where

$$\xi_e=\sum_{\beta=1}^{4}(\zeta_{1\beta}{}^{e})^2,\quad \xi_o=\sum_{\beta=1}^{4}(\zeta_{1\beta}{}^{o})^2,\quad \xi'=\sum_{\beta=1}^{4}\zeta_{1\beta}{}^{e}\zeta_{1\beta}{}^{o}, \tag{40}$$

and the values of ω are the roots of the quartic

$$\begin{aligned}
&[(\omega-A)(\omega+B)+2\gamma^2\xi_e][(\omega-A)(\omega+B)+2\gamma^2\xi_o]\\
&\qquad-4\gamma^4\xi'^2=0.
\end{aligned} \tag{41}$$

The exact roots may be found by first neglecting ξ' [see Eq. (48)]. Considering well-known rules governing various sums of products of the roots, it then becomes apparent that the exact solutions are

$$\begin{aligned}
2\omega_1&=(A-B)-[(A+B)^2-8\gamma^2\xi_e+x]^{\frac{1}{2}},\\
2\omega_2&=(A-B)+[(A+B)^2-8\gamma^2\xi_e+x]^{\frac{1}{2}},\\
2\omega_3&=(A-B)-[(A+B)^2-8\gamma^2\xi_o-x]^{\frac{1}{2}},\\
2\omega_4&=(A-B)+[(A+B)^2-8\gamma^2\xi_o-x]^{\frac{1}{2}},
\end{aligned} \tag{41'}$$

with

$$x=4\gamma^2(\xi_e-\xi_o)\{1-[1+\xi'^2/(\xi_e-\xi_o)^2]^{\frac{1}{2}}\}.$$

The conjugate pairs satisfy

$$\dot{q}=\omega p,\quad \dot{p}=-\omega q, \tag{42}$$

and

$$[q,p]=i, \tag{43}$$

the latter assuming the commutation relations (14). [Equations (43) with (39) completely determines the a's.] This provides 8 of the 12 normal coordinates; we shall denote them by $q_j{}^{\pm}$, $p_j{}^{\pm}$, $j=1, 2, 3, 4$.

The remaining four normal coordinates and momenta are readily found to be

$$q_i{}^{\pm}=\sum_{\beta=1}^{4}e_{i\beta}Q_\beta{}^{B\pm},\quad p_i{}^{\pm}=\sum_{\beta=1}^{4}e_{i\beta}P_\beta{}^{B\pm},\quad i=5, 6, \tag{44}$$

the $e_{i\beta}$ being determined by

$$\textstyle\sum_\beta e_{i\beta}\zeta_{1\beta}{}^{e}=\sum_\beta e_{i\beta}\zeta_{1\beta}{}^{o}=0,\quad \sum e_{i\beta}e_{j\beta}=\delta_{ij}. \tag{45}$$

These coordinates are degenerate with the frequency

$$\omega=B. \tag{46}$$

We now note that ξ' is of order k^3 whereas ξ_e and ξ_o are, respectively, of order k^0 and k^2. Hence, in the calculation to second order, we may neglect terms in ξ' in Eqs. (39) and (41). The resulting solution is immediately found to be

$$\begin{aligned}
q_i^{\pm}&=d_i(\textstyle\sum_\alpha Q_\alpha{}^{A\pm}\pm c_i\sum_\beta \zeta_{1\beta}{}^e Q_\beta{}^{B\pm}), && i=1,2\\
p_i^{\pm}&=d_i(\textstyle\sum_\alpha P_\alpha{}^{A\pm}\mp c_i\sum_\beta \zeta_{1\beta}{}^e P_\beta{}^{B\pm}), && i=1,2\\
q_j^{\pm}&=d_j(Q_1{}^{A\mp}-Q_2{}^{A\mp}+c_j\textstyle\sum_\beta \zeta_{1\beta}{}^o Q_\beta{}^{B\pm}), && j=3,4\\
p_j^{\pm}&=d_j(P_1{}^{A\mp}-P_2{}^{A\mp}-c_j\textstyle\sum_\beta \zeta_{1\beta}{}^o P_\beta{}^{B\pm}), && j=3,4\\
2\omega_1&=A-B-[(A+B)^2-8\gamma^2\xi_e]^{\frac{1}{2}},\\
2\omega_2&=A-B+[(A+B)^2-8\gamma^2\xi_e]^{\frac{1}{2}},\\
2\omega_3&=A-B-[(A+B)^2-8\gamma^2\xi_o]^{\frac{1}{2}},\\
2\omega_4&=A-B+[(A+B)^2-8\gamma^2\xi_o]^{\frac{1}{2}},
\end{aligned} \tag{48}$$

where

$$\begin{aligned}
c_i&=(A-\omega_i)/\gamma\xi_e=2\gamma/(B+\omega_i), && i=1,2\\
d_i&=(2-\xi_e c_i{}^2)^{-\frac{1}{2}}, && i=1,2\\
c_j&=(A-\omega_j)/\gamma\xi_o=2\gamma/(B+\omega_j), && j=3,4\\
d_j&=(2-\xi_o c_j{}^2)^{-\frac{1}{2}}, && j=3,4.
\end{aligned} \tag{49}$$

Explicit calculation gives, to second order,

$$\xi_e=4\eta^2=36(1-\epsilon)^2, \tag{50}$$

where

$$\epsilon=11k^2a^2/384, \quad \xi_o=k^2a^2/16. \tag{51}$$

Using (22), we see that

$$\gamma^2=AB/Z_AZ_B, \tag{52}$$

so that we may rewrite ω_1 and ω_2:

$$\begin{aligned}
2\omega_1&=A-B-[(A+B)^2-4AB(1-\epsilon)^2]^{\frac{1}{2}},\\
2\omega_2&=A-B+[(A+B)^2-4AB(1-\epsilon)^2]^{\frac{1}{2}}.
\end{aligned} \tag{53}$$

These branches are identical to those found by H. Kaplan[11] if we go to the classical limit $S_c{}^{A,B}\cong S_{A,B}$.

To second order, assuming that $AB\epsilon/(A-B)^2\ll 1$, we have

$$\begin{aligned}
\omega_1&=-2AB\epsilon/(A-B),\\
\omega_2&=A-B+2AB\epsilon/(A-B),\\
\omega_3&=-B+2AB\epsilon/33(A+B),\\
\omega_4&=A-2AB\epsilon/33(A+B).
\end{aligned} \tag{54}$$

We see that some of the frequencies are definitely negative. This is not accidental (since the ω's to second order are roots of equations quadratic in ω), but has the following significance. Whenever $\omega<0$, the corresponding coefficient d is imaginary, so that the corresponding operators are not Hermitian. However, the simple canonical transformation $q'=iq$, $p'=-ip$ relieves us of this difficulty. Thus, dropping the primes in these cases, the Hamiltonian (24) is

$$H=\tfrac{1}{2}\textstyle\sum'_k\{\sum|\omega|(p^2+q^2)\}, \tag{55}$$

[11] Note: H. Kaplan's J_{AB} is $J/2$ and his a is one-half our a.

where the second $\sum$ means to sum over the 12 normal modes. (ω is in units of $\hbar$.) Since the various q's and p's satisfy $[q_j,p_k]\cong i\delta_{jk}$ [approximately, even considering (14) to be exact, since ξ' was neglected], the wave functions follow immediately in terms of the solutions to the harmonic oscillator problem.

IV. DISCUSSION

In Fig. 2 is shown a sketch of the five branches found for the spectrum. The case $B<A<2B$ is shown, so that ω_2 is lower than ω_3 or ω_5. However, for $A>2B$, the branches ω_3 and ω_5 will lie below ω_2. (Note that when $A<2B$, $S_A>S_B$.) When $A=B$, ω_1 and ω_2 coalesce and are linear in k for small k; ω_3 and ω_4 equal $\omega_5=A$ for $k=0$, deviating from this value quadratically in k. To the order of our approximation (3), $A=JZ_AS_B$, $B=JZ_BS_A$, so that $A=B$ implies $S_A=2S_B$ or antiferromagnetism (i.e., zero net moment in the ground state).

It is interesting to see how the zero-point excitation enters into the calculation of the z component of the total spin to give the expected value $2N(S_A-2S_B)$ instead of $2N(S_c{}^A-2S_c{}^B)$. Using (3) and (11) we find, to order $1/S_{A,B}$,

$$\begin{aligned}
S_{z'}&\equiv\textstyle\sum S_i{}^{Az'}+\sum S_j{}^{Bz'}\\
&=2N(S_c{}^A-2S_c{}^B)-\tfrac{1}{2}\textstyle\sum'_k\sum_\sigma\{\sum_\alpha[(P_\alpha{}^{A\sigma})^2\\
&\qquad+(Q_\alpha{}^{A\sigma})^2]-\textstyle\sum_\beta[(P_\beta{}^{B\sigma})^2+(Q_\beta{}^{B\sigma})^2]\},
\end{aligned} \tag{56}$$

where $\sigma=+$ and $-$. Write Eqs. (38) and (44) as

$$q_i=\textstyle\sum \bar{C}_{ij}Q_j, \quad p_i=\sum \bar{D}_{ij}P_j. \tag{57}$$

Since we are assured by normal mode theory [and Eq. (43)] that $[q_j,p_k]=i\delta_{jk}$ [for the exact normal coordinates and assuming (14) is exact], the inverse, C, of $\bar{C}$ is simply the transpose, $\bar{D}^T$, of $\bar{D}$; also $(\bar{D})^{-1}=\bar{C}^T$. We thus find

$$\begin{aligned}
S_{z'}&=2N(S_c{}^A-2S_c{}^B)-\tfrac{1}{2}\textstyle\sum'_k\sum_\sigma\{-(p_1{}^\sigma)^2-(q_1{}^\sigma)^2\\
&\quad+(p_2{}^\sigma)^2+(q_2{}^\sigma)^2-(p_3{}^\sigma)^2-(q_3{}^\sigma)^2+(p_4{}^\sigma)^2+(q_4{}^\sigma)^2\\
&\qquad-[(p_5{}^\sigma)^2+(q_5{}^\sigma)^2+(p_6{}^\sigma)^2+(q_6{}^\sigma)^2]\},
\end{aligned} \tag{58}$$

for $A\gtrless B$. For $A<B$, interchange 1 and 2. [The p's and q's here are the Hermitean operators used in (55).] Hence the eigenstates of (55) are also eigenstates of $S_{z'}$; using the values $(p_i{}^\sigma)^2+(q_i{}^\sigma)^2=2n_i{}^\sigma+1$, $n_i{}^\sigma=1,2,\cdots$, it is seen the zero-point excitation terms in modes 5

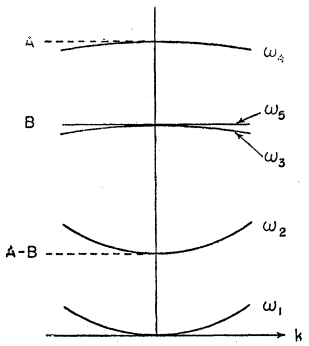

FIG. 2. The spectrum (drawn for the case $B<A<2B$).

and 6 add up to N, thus canceling the terms in $S_c{}^A$ and $S_c{}^B$ of order $1/S_{A,B}$.[12] Hence

$$S_{z'}=2N(S_A-2S_B)-\sum'_{\mathbf{k}}\sum_\sigma(n_2{}^\sigma-n_1{}^\sigma+n_4{}^\sigma -n_3{}^\sigma-n_5{}^\sigma-n_6{}^\sigma), \quad A>B. \quad (59)$$

Note that the excitation of modes 1 and 2 contributes to $S_{z'}$ in the same way as found by Keffer[13] for antiferromagnetic structures in the presence of a magnetic field parallel to the spin axis. This is reasonable: for S_A slightly different from $2S_B$, the energy eigenstates are approximately those linear combinations of the degenerate states corresponding to $\omega_1=\omega_2$ which are eigenstates of $S_{z'}$ (as they must be since $S_{z'}$ is a good quantum number) and hence of the Hamiltonian including the energy due to a magnetic field in the z' direction.

Perhaps some insight into the dependence of the normal coordinates on the direction of $\mathbf{k}$ may be gained by the following simple intuitive discussion. Consider the example $\mathbf{k}=k(0,1,0)$. Referring to Fig. 1, it is seen that atoms B_1 and B_2 are in one plane of constant $\mathbf{k}\cdot\mathbf{r}$ while B_3 and B_4 are in another such plane. Hence one might expect B_1 and B_2 to be equivalent and similarly for B_3 and B_4, each pair then behaving analogously to the example of Fig. 3(a). This is borne out in all the normal modes [Eqs. (44) and (47)], in the sense that, say, $Q_1{}^{B+}$ and $Q_2{}^{B+}$ are either equal or π out of phase; similarly for any pair of variables corresponding to B_1 and B_2 or B_3 and B_4. [This follows since $\zeta_{1\beta}{}^e=\zeta_{11}{}^e(1,1,1,1)$, $\zeta_{1\beta}{}^o=\zeta_{11}{}^o(1,1,-1,-1)$, $e_{1\beta}=(1/2)(1,-1,1,-1)$, $e_{2\beta}=(1/2)(1,-1,-1,1)$.] As a second example we take the case $\mathbf{k}=(k/\sqrt{3})(1,1,1)$. By the same reasoning B_2, B_3, and B_4 would be expected to be equivalent and hence to behave analogously to the example of Fig. 3(b). This is again borne out in all the normal coordinates: $\zeta_{12}{}^e=\zeta_{13}{}^e=\zeta_{14}{}^e$, $\zeta_{12}{}^o=\zeta_{13}{}^o=\zeta_{14}{}^o(=-\zeta_{11}{}^o/3)$, $e_{1\beta}=(1/\sqrt{2})(0,0,1,-1)$, $e_{2\beta}=(1/\sqrt{6})(0,2,-1,-1)$. For the example of Fig. 3(b), the normal coordinates are $x_1+x_2+x_3$, x_2-x_3 and $2x_1-x_2-x_3$; furthermore, the two oscillatory modes are degenerate, completing the analogy (as far as we shall carry it).

It is interesting to compare these results with those obtained in a preliminary calculation, in which all terms ($\zeta_{\alpha\beta}{}^o$ and $x_{\alpha\beta}$) anisotropic in $\mathbf{k}$ were neglected. The only difference in the spectrum is that the small deviation of ω_3 from the value B, and ω_4 from A, did not occur. Differences of zero order in k enter into the coordinates associated with ω_3 and ω_5 in the sense that the perturbation requires a particular linear combination (for each direction of $\mathbf{k}$) of the originally degenerate coordinates associated with the frequency B. The differences in the remaining coordinates are of higher order.

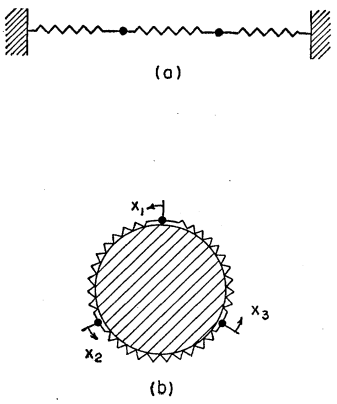

FIG. 3. Examples of coupled oscillators. (Equal masses and equal spring constants; the masses are constrained to move along the lines of the springs.)

Equation (55) provides the starting point for the calculation of the neutron scattering cross section. This will be reported in a future paper with the purpose of comparing the result for scattering involving the lowest mode with current experiments, as well as a step towards answering the question as to the possibility of observing the higher modes. In relation to this question, we should mention that the energies needed to excite the latter are in the slow-neutron range ($\approx 10^{-2}$ ev as estimated by using the value, $J=4\times10^{-3}$ ev found by Brockhouse[1] in connection with the lowest branch, and taking $S_A=2.5$, $S_B=2.25$, characteristic of magnetite).

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Note added in proof.—After the present manuscript was submitted for publication, the author became aware of a paper by J. S. Kouvel [Technical Report 210, Cruft Laboratory, Harvard, February 1, 1955 (unpublished)] in which the semiclassical spin vector method is applied to the ordered magnetite structure; in the region of overlap of his and the present results there is agreement as expected.

[12] It is felt that the alternation of sign from modes 1 to 2 and 3 to 4 (which results in no contribution from the zero-point excitation in these modes), although deduced from expressions (47)–(49), valid only to second order in k, probably occurs for all k. The statement concerning modes 5 and 6 is exact since (44)–(46) are valid for all k.

[13] F. Keffer, thesis, Berkeley, January, 1952 [see Keffer, Kaplan, and Yafet, Am. J. Phys. **21**, 250 (1952)].