Spin Waves in Complex Lattices*

DUANE C. WALLACE
Sandia Laboratory, Albuquerque, New Mexico
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Consideration is given to a lattice which has an arbitrary number of magnetic atoms, or spins, in each magnetic unit cell. The Hamiltonian for this spin system is taken to include the usual isotropic exchange and magnetic field terms. This Hamiltonian is diagonalized in the spin-wave approximation for the case where the spin-deviation from the + or − z direction is small for each spin. The thermal average values of the z component of each spin are calculated and found in general to be different for spins on inequivalent sites in the unit cell. Insofar as the model Hamiltonian and the spin-wave approximations are valid, it is suggested that nuclear magnetic resonance measurements might be interpreted to give some information about the effective anisotropy fields.

I. INTRODUCTION

THIS paper is concerned with the spin-wave problem for a lattice which has an arbitrary number of magnetic atoms, or spins, per unit cell, and for which the spin deviation from the + or − z direction is small for each spin. The spins are assumed to interact with each other through the usual isotropic exchange, and to interact with the z components of internal and external magnetic fields. On the basis of this model, the Hamiltonian for the spin system is

$$\mathcal{H}=-\sum'_{nn',jj'} J_{nn',jj'}\mathbf{S}_{nj}\cdot\mathbf{S}_{n'j'}-\sum_{n,j} G_j S_{nj}{}^z. \quad (1)$$

Here n labels the unit cell in the lattice ($n=1, 2, \cdots, N$) and j labels the site in the magnetic cell ($j=1, 2, \cdots, T$). $\mathbf{S}_{nj}$ is the spin vector, expressed in units of $\hbar$, which is located at $\mathbf{r}_{nj}$, and $J_{nn',jj'}$ is the exchange constant between the spin vectors $\mathbf{S}_{nj}$ and $\mathbf{S}_{n'j'}$. It is assumed that $J_{nn',jj'}$ depends only on the distance $|\mathbf{r}_{nj}-\mathbf{r}_{n'j'}|$; the prime on the sum in (1) means to omit the terms (nn,jj). The coefficient G_j is linear in the z components of internal and external magnetic fields. For example, with an external field H and an effective anisotropy field H_{aj} at each j site (each field H or H_{aj} is directed along the z axis, being in the $+z$ direction if the corresponding H or H_{aj} is positive),

$$G_j=g_j\beta(H+H_{aj}), \quad (2)$$

where β is the Bohr magneton ($e\hbar/2mc$) and g_j is the spectroscopic splitting factor for the spins on j sites.

Following Van Kranendonk and Van Vleck,[1] the Hamiltonian is transformed to harmonic oscillator coordinates and momenta by the relations

$$\begin{aligned} &S_{nj}{}^x=S_j{}^{1/2}Q_{nj}, \quad S_{nj}{}^y=\epsilon_j S_j{}^{1/2}P_{nj}, \\ &S_{nj}{}^z=\epsilon_j[S_j-\tfrac{1}{2}(P_{nj}{}^2+Q_{nj}{}^2-1)], \end{aligned} \quad (3)$$

where S_j is the magnitude of the spins on j sites. For the present case, the spin vector $\mathbf{S}_{nj}$ is either nearly in the $+z$ direction ($\epsilon_j=+1$) or nearly in the $-z$ direction ($\epsilon_j=-1$), so that the spin deviation $\frac{1}{2}(P_{nj}{}^2+Q_{nj}{}^2-1)$ is small. Thus, following Holstein and Primakoff,[2] terms quadratic in spin deviations are dropped and (1) becomes

$$\mathcal{H}=E_0'+\tfrac{1}{2}\sum_{nn',jj'} K_{nn',jj'} \times[\epsilon_j\epsilon_{j'}P_{nj}P_{n'j'}+Q_{nj}Q_{n'j'}]; \quad (4)$$

$$E_0'=-\sum_{n,j}\epsilon_j G_j(S_j+\tfrac{1}{2}) -\sum'_{nn',jj'}\epsilon_j\epsilon_{j'}J_{nn',jj'}S_j(1+S_{j'}); \quad (5)$$

$$K_{nn',jj'}=K_{n'n,j'j}=-2(S_jS_{j'})^{\frac{1}{2}}J_{nn',jj'} \quad \text{for terms}\neq(nn,jj); \quad (6)$$

$$K_{nn,jj}=\epsilon_j[G_j+2\sum'_{n',j'}J_{nn',jj'}\epsilon_{j'}S_{j'}].$$

The specification of the problem is completed by a set of commutation relations which are good to the same degree of approximation (namely, dropping spin-deviation operators compared to unity):

$$\begin{aligned} &[Q_{nj},Q_{n'j'}]=[P_{nj},P_{n'j'}]=0, \\ &[Q_{nj},P_{n'j'}]=i\delta_{nn'}\delta_{jj'}. \end{aligned} \quad (7)$$

It is not within the scope of the present paper to discuss the approximations which lead to the model Hamiltonian (1), nor those which are involved in transforming to (4)–(7). These latter approximations are discussed qualitatively in references 1 and 2, and quantitatively for a ferromagnetic case by Dyson.[3] It is the purpose of the present paper to give explicitly the transformations which diagonalize (4); this is done in Sec. II. In Sec. III the thermal average value of the z component of spin for spins on j sites is calculated, and an application to nuclear magnetic resonance studies is pointed out. For any case with only two spins per unit cell the problem can be solved explicitly, and this is discussed in Sec. IV. Sáenz[4] has recently found the eigenvalues for the Hamiltonian (4), and the relations between his work and the present are discussed in the Appendix. In the following, the problem is treated in terms of harmonic oscillator formalism for simplicity; a transformation to creation and annihilation operators can be carried out at any point in the

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[1] J. Van Kranendonk and J. H. Van Vleck, Revs. Modern Phys. **30**, 1 (1958).

[2] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

[3] Freeman J. Dyson, Phys. Rev. **102**, 1217, 1230 (1956).

[4] Albert W. Sáenz, Phys. Rev. **125**, 1940 (1962).

development without difficulty. The periodic boundary condition for the lattice is also applied throughout.

II. DIAGONALIZATION OF THE HAMILTONIAN

The first transformation consists of the introduction of traveling waves which have wave vector $\mathbf{k}$ (there are N values of $\mathbf{k}$ distributed uniformly over the first Brillouin zone) and branch index s ($s=1, 2, \cdots, T$). Define first the matrix $L_{\mathbf{k}}$ which has components

$$L_{\mathbf{k},jj'}=\sum_{n'} K_{nn',jj'} \exp[-i\mathbf{k}\cdot(\mathbf{r}_n-\mathbf{r}_{n'})], \quad (8)$$

where $\mathbf{r}_n$ is the vector to some reference point in unit cell n (the definition $\mathbf{r}_{nj}=\mathbf{r}_n+\mathbf{r}_j$ will be followed). Note that $L_{\mathbf{k},jj'}$ does not depend on the index n [$\mathbf{r}_n=0$ can be taken in (8)]. From the symmetry of the lattice and the symmetry of the K coefficients, the following properties of $L_{\mathbf{k}}$ are derived:

$$L_{\mathbf{k},jj'}=L_{-\mathbf{k},j'j}, \quad (9)$$

$$L_{\mathbf{k},jj'}=L_{\mathbf{k},j'j}^* \quad \text{(Hermitian property)}. \quad (10)$$

From (9) it follows that the set of eigenvalues $\lambda_{\mathbf{k}s}$ of $L_{\mathbf{k}}$ is equal to the set $\lambda_{-\mathbf{k}s}$ of $L_{-\mathbf{k}}$, and these can be ordered so that

$$\lambda_{\mathbf{k}s}=\lambda_{-\mathbf{k}s}. \quad (11)$$

Let $V_{\mathbf{k},js}$ be components of the eigenvectors of $L_{\mathbf{k}}$ and write the secular equation

$$\sum_{j'} L_{\mathbf{k},jj'}V_{\mathbf{k},j's}=\lambda_{\mathbf{k}s}V_{\mathbf{k},js}, \quad (j=1, 2, \cdots, T). \quad (12)$$

By taking the complex conjugate of (12), using (9) and (11) and noting that the $\lambda_{\mathbf{k}s}$ are real, it is seen that the eigenvectors can be chosen to obey the relation

$$V_{\mathbf{k},js}^*=V_{-\mathbf{k},js}. \quad (13)$$

Finally, since $L_{\mathbf{k}}$ is Hermitian, the eigenvectors can be chosen to satisfy the orthonormality and completeness relations

$$\sum_j V_{-\mathbf{k},js}V_{\mathbf{k},js'}=\delta_{ss'}, \quad (14)$$

$$\sum_s V_{-\mathbf{k},js}V_{\mathbf{k},j's}=\delta_{jj'}. \quad (15)$$

The traveling wave transformation is now defined in terms of new operators $A_{\mathbf{k}s}$, $B_{\mathbf{k}s}$ as follows:

$$\begin{aligned} P_{nj}&=N^{-1/2}\epsilon_j\sum_{\mathbf{k}s} A_{\mathbf{k}s}V_{\mathbf{k},js}\exp(i\mathbf{k}\cdot\mathbf{r}_n),\\ Q_{nj}&=N^{-1/2}\sum_{\mathbf{k}s} B_{\mathbf{k}s}V_{\mathbf{k},js}\exp(i\mathbf{k}\cdot\mathbf{r}_n). \end{aligned} \quad (16)$$

With the aid of (11), (12), and (14), the Hamiltonian becomes

$$\mathcal{H}=E_0'+\tfrac{1}{2}\sum_{\mathbf{k}s}\lambda_{\mathbf{k}s}(A_{\mathbf{k}s}A_{-\mathbf{k}s}+B_{\mathbf{k}s}B_{-\mathbf{k}s}), \quad (17)$$

where use has also been made of the identity

$$\sum_n \exp[i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}_n]=N\delta(\mathbf{k}+\mathbf{k}'). \quad (18)$$

The transformation (16) can be inverted and the commutators found to be

$$\begin{aligned} &[B_{\mathbf{k}s},B_{\mathbf{k}'s'}]=[A_{\mathbf{k}s},A_{\mathbf{k}'s'}]=0,\\ &[B_{\mathbf{k}s},A_{\mathbf{k}'s'}]=i\delta(\mathbf{k}+\mathbf{k}')M_{\mathbf{k},ss'}, \end{aligned} \quad (19)$$

where $M_{\mathbf{k}}$ is the Hermitian, involutory matrix whose components are

$$M_{\mathbf{k},ss'}=\sum_j V_{-\mathbf{k},js}\epsilon_j V_{\mathbf{k},js'}. \quad (20)$$

Also, because of the Hermiticity of the P_{nj}, Q_{nj},

$$A_{\mathbf{k}s}^\dagger=A_{-\mathbf{k}s}; \quad B_{\mathbf{k}s}^\dagger=B_{-\mathbf{k}s}. \quad (21)$$

If all $\epsilon_j=+1$ (ferromagnetic case), the problem is solved at this point, since for this case $M_{\mathbf{k},ss'}=\delta_{ss'}$. The ferromagnetic Hamiltonian is then

$$\begin{aligned} \mathcal{H}_f&=E_{0f}+\tfrac{1}{2}\sum_{\mathbf{k}s}\lambda_{\mathbf{k}s}(A_{\mathbf{k}s}A_{-\mathbf{k}s}+B_{\mathbf{k}s}B_{-\mathbf{k}s}-1),\\ E_{0f}&=E_0'+\tfrac{1}{2}\sum_{\mathbf{k}s}\lambda_{\mathbf{k}s}, \end{aligned} \quad (22)$$

where the eigenvalues of $\frac{1}{2}(A_{\mathbf{k}s}A_{-\mathbf{k}s}+B_{\mathbf{k}s}B_{-\mathbf{k}s}-1)$ are $m_{\mathbf{k}s}=0, 1, 2, \cdots$.

It now remains for the general case to transform the operators in the Hermitian form (17) to new operators which have the desired commutation relations. If this is to be accomplished by linear transformations, the eigenvalues of the final Hermitian form will be positive if and only if the $\lambda_{\mathbf{k}s}$ are all positive; henceforth it will be assumed that $L_{\mathbf{k}}$ is positive definite for all $\mathbf{k}$.[5]

Now carry out a second transformation defined by

$$A_{\mathbf{k}s}=\lambda_{\mathbf{k}s}^{-1/2}a_{\mathbf{k}s}, \quad B_{\mathbf{k}s}=\lambda_{\mathbf{k}s}^{-1/2}b_{\mathbf{k}s}, \quad (23)$$

where the positive square root is always taken for $\lambda_{\mathbf{k}s}^{1/2}$. The new commutators are

$$[b_{\mathbf{k}s},a_{\mathbf{k}'s'}]=i\delta(\mathbf{k}+\mathbf{k}')R_{\mathbf{k},ss'}, \quad (24)$$

$$R_{\mathbf{k},ss'}=\lambda_{\mathbf{k}s}^{1/2}M_{\mathbf{k},ss'}\lambda_{\mathbf{k}s'}^{1/2}. \quad (25)$$

Let the eigenvalues of $R_{\mathbf{k}}$ be $\omega_{\mathbf{k}t}$ ($t=1, 2, \cdots, T$) and the components of the eigenvectors be $W_{\mathbf{k},st}$. Then

$$\sum_{s'} R_{\mathbf{k},ss'}W_{\mathbf{k},s't}=\omega_{\mathbf{k}t}W_{\mathbf{k},st}, \quad (s=1, 2, \cdots, T). \quad (26)$$

As in the diagonalization of the matrices $L_{\mathbf{k}}$ above, the following relations concerning the diagonalization of the $R_{\mathbf{k}}$ are observed:

$$R_{\mathbf{k},ss'}=R_{-\mathbf{k},s's}; \quad (27)$$

$$R_{\mathbf{k},ss'}=R_{\mathbf{k},s's}^* \quad \text{(Hermitian property)}; \quad (28)$$

$$\omega_{\mathbf{k}t}=\omega_{-\mathbf{k}t}; \quad (29)$$

$$W_{\mathbf{k},st}^*=W_{-\mathbf{k},st}; \quad (30)$$

$$\sum_s W_{-\mathbf{k},st}W_{\mathbf{k},st'}=\delta_{tt'} \quad \text{(orthonormality)}; \quad (31)$$

$$\sum_t W_{-\mathbf{k},st}W_{\mathbf{k},s't}=\delta_{ss'} \quad \text{(completeness)}. \quad (32)$$

A third transformation is now defined by

$$a_{\mathbf{k}s}=\sum_t W_{\mathbf{k},st}c_{\mathbf{k}t}; \quad b_{\mathbf{k}s}=\sum_t W_{\mathbf{k},st}d_{\mathbf{k}t}. \quad (33)$$

Equations (33) can be inverted with the aid of (31). The Hamiltonian, commutators, and Hermitian con-

[5] This important result has been pointed out by Sáenz, reference 4.

jugate relations become

$$\mathcal{H}=E_0'+\tfrac{1}{2}\sum_{\mathbf{k}t}(c_{\mathbf{k}t}c_{-\mathbf{k}t}+d_{\mathbf{k}t}d_{-\mathbf{k}t}); \quad (34)$$

$$[d_{\mathbf{k}t},d_{\mathbf{k}'t'}]=[c_{\mathbf{k}t},c_{\mathbf{k}'t'}]=0,$$
$$[d_{\mathbf{k}t},c_{\mathbf{k}'t'}]=i\omega_{\mathbf{k}t}\delta(\mathbf{k}+\mathbf{k}')\delta_{tt'}; \quad (35)$$

$$c_{\mathbf{k}t}^{\dagger}=c_{-\mathbf{k}t}; \quad d_{\mathbf{k}t}^{\dagger}=d_{-\mathbf{k}t}. \quad (36)$$

A fourth transformation removes the $\omega_{\mathbf{k}t}$ from the commutation relations (35) and thus completes the diagonalization procedure. Here it must be noted that the $\omega_{\mathbf{k}t}$, although real, are of indefinite sign. Let $\omega_{\mathbf{k}t}/|\omega_{\mathbf{k}t}|=\gamma_t$, where $\gamma_t=+1$ or -1. On the basis of continuity arguments, Sáenz has proved that the numbers γ_t and ϵ_j are equal to within a permutation; an alternate proof of this is given in the Appendix. The final transformation can then be taken as

$$c_{\mathbf{k}t}=\gamma_t|\omega_{\mathbf{k}t}|^{1/2}C_{\mathbf{k}t}, \quad d_{\mathbf{k}t}=|\omega_{\mathbf{k}t}|^{1/2}D_{\mathbf{k}t}, \quad (37)$$

where the positive square root is always taken for $|\omega_{\mathbf{k}t}|^{1/2}$. The statement of the problem in diagonal form is then

$$\mathcal{H}=E_0+\tfrac{1}{2}\sum_{\mathbf{k}t}|\omega_{\mathbf{k}t}|\times(C_{\mathbf{k}t}C_{-\mathbf{k}t}+D_{\mathbf{k}t}D_{-\mathbf{k}t}-1); \quad (38)$$

$$E_0=E_0'+\tfrac{1}{2}\sum_{\mathbf{k}t}|\omega_{\mathbf{k}t}|; \quad (39)$$

$$[D_{\mathbf{k}t},D_{\mathbf{k}'t'}]=[C_{\mathbf{k}t},C_{\mathbf{k}'t'}]=0,$$
$$[D_{\mathbf{k}t},C_{\mathbf{k}'t'}]=i\delta(\mathbf{k}+\mathbf{k}')\delta_{tt'}; \quad (40)$$

$$C_{\mathbf{k}t}^{\dagger}=C_{-\mathbf{k}t}, \quad D_{\mathbf{k}t}^{\dagger}=D_{-\mathbf{k}t}. \quad (41)$$

The eigenvalues of $\frac{1}{2}(C_{\mathbf{k}t}C_{-\mathbf{k}t}+D_{\mathbf{k}t}D_{-\mathbf{k}t}-1)$ are $n_{\mathbf{k}t}=0, 1, 2, \cdots$.

III. APPLICATIONS

Let the statistical average of an operator O be denoted by

$$\langle O\rangle=Z^{-1}\,\mathrm{Tr}[O\exp(-\mathcal{H}/KT)], \quad (42)$$

where Z is the partition function, K is Boltzmann's constant, and T is the temperature. Then

$$\langle m_{\mathbf{k}s}\rangle=[\exp(\lambda_{\mathbf{k}s}/KT)-1]^{-1};$$
$$\langle n_{\mathbf{k}t}\rangle=[\exp(|\omega_{\mathbf{k}t}|/KT)-1]^{-1}. \quad (43)$$

The statistical average of the energy of the spin system is

$$\langle E\rangle_f=E_{0f}+\sum_{\mathbf{k}s}\lambda_{\mathbf{k}s}\langle m_{\mathbf{k}s}\rangle, \quad \text{(ferromagnetic case)} \quad (44)$$

$$\langle E\rangle=E_0+\sum_{\mathbf{k}t}|\omega_{\mathbf{k}t}|\langle n_{\mathbf{k}t}\rangle, \quad \text{(general case)} \quad (45)$$

and the specific heat at constant volume is $C_V=\partial\langle E\rangle/\partial T$.

An interesting application of the development in Sec. II is to the calculation of the statistical average of the z component of spin for spins on j sites. The average $\langle S_{nj}{}^z\rangle$ is found to be independent of n and is denoted by $\langle S_j{}^z\rangle$; for the ferromagnetic and general cases the calculation yields

$$\langle S_j{}^z\rangle_f=S_j-N^{-1}\sum_{\mathbf{k}s}\langle m_{\mathbf{k}s}\rangle V_{\mathbf{k},js}V_{-\mathbf{k},js}; \quad (46)$$

$$\langle S_j{}^z\rangle=\epsilon_j(S_j+\tfrac{1}{2})-N^{-1}\epsilon_j\sum_{\mathbf{k}t}|\omega_{\mathbf{k}t}|(\langle n_{\mathbf{k}t}\rangle+\tfrac{1}{2})\times\sum_{ss'}\lambda_{\mathbf{k}s}{}^{-1/2}\lambda_{\mathbf{k}s'}{}^{-1/2}W_{\mathbf{k},st}W_{-\mathbf{k},s't}V_{\mathbf{k},js}V_{-\mathbf{k},js'}; \quad (47)$$

where use has been made of the fact that in the general case $\langle C_{\mathbf{k}t}C_{\mathbf{k}'t'}\rangle$, $\langle D_{\mathbf{k}t}D_{\mathbf{k}'t'}\rangle$ vanish unless $\mathbf{k}=-\mathbf{k}'$, $t=t'$ (and similarly for $\langle A_{\mathbf{k}s}A_{\mathbf{k}'s'}\rangle$, $\langle B_{\mathbf{k}s}B_{\mathbf{k}'s'}\rangle$ in the ferromagnetic case). To obtain the statistical average of the total z component of spin for the system, $\langle S_j{}^z\rangle$ must be summed over j and multiplied by N. The results are

$$\langle S^z\rangle_f=N\sum_j S_j-\sum_{\mathbf{k}s}\langle m_{\mathbf{k}s}\rangle; \quad (48)$$

$$\langle S^z\rangle=N\sum_j\epsilon_jS_j-\sum_{\mathbf{k}t}\gamma_t\langle n_{\mathbf{k}t}\rangle; \quad (49)$$

the latter formula has been given by Sáenz.[4] (In the equation in footnote 20 of reference 4 there should be a sum over alpha on the left-hand side.) In obtaining (49) use has been made of the fact that $\sum_j\epsilon_j=\sum_t\gamma_t$, and that the eigenvectors $\mathbf{W}_{\mathbf{k}t}$ diagonalize the matrix $R_{\mathbf{k}}^{-1}$, yielding the eigenvalues $\omega_{\mathbf{k}t}^{-1}$.

Since the hyperfine field at a j site is proportional to $\langle S_j{}^z\rangle$ for a magnetic material, the nuclear magnetic resonance (NMR) frequency for a nucleus on a j site is (approximately) proportional to the magnitude of this quantity. It is seen from (46) [or (47)] that the $\langle S_j{}^z\rangle_f$ (or $\langle S_j{}^z\rangle$) are, in general, expected to be different for different j. This difference vanishes as $T\to 0$ for the ferromagnetic case (except perhaps when acoustic modes exist), but not necessarily for the general case. If this difference in NMR frequencies for nuclei on inequivalent sites were observed, it could presumably give some information about the effective anisotropy fields H_{aj}; however, in many cases the effect is probably as small as other effects which have been neglected in the model Hamiltonian (1) (such as dipolar interactions between spins), and the approximations used in treating the model.

IV. EXAMPLES

For any case with only two spins per magnetic unit cell ($T=2$), the problem can be solved explicitly in terms of the elements of the matrices $L_{\mathbf{k}}$ for each $\mathbf{k}$. Let $j=1, 2$, suppress the index $\mathbf{k}$ for abbreviation, and represent the elements of $L_{\mathbf{k}}$ by L_{11}, L_{22}, $L_{12}=L_{21}{}^*$. The results of the first transformation in Sec. II are, with $s=\pm$,

$$\lambda_{\pm}=\tfrac{1}{2}[L_{11}+L_{22}\pm A^{1/2}];$$
$$|V_{1\pm}|^2=\tfrac{1}{2}[1\pm(L_{11}-L_{22})A^{-1/2}];$$
$$|V_{2\pm}|^2=\tfrac{1}{2}[1\mp(L_{11}-L_{22})A^{-1/2}];$$
$$A=(L_{11}-L_{22})^2+4L_{12}L_{21}; \quad (50)$$

where the positive square root is always taken for $A^{1/2}$ and where the phase factor in each of the eigenvectors V_+, V_- is arbitrary. The condition that L be positive

definite ($\lambda_{\pm}>0$) is

$$L_{11}+L_{22}>A^{1/2}. \tag{51}$$

At this point the problem is solved for the ferromagnetic case ($\epsilon_1=\epsilon_2=+1$), the spin-wave energies being given by λ_{k+}, λ_{k-}. An interesting example is the case of $CrCl_3$, which has been shown by Narath[6] to be well approximated by a two-dimensional model. Here the Cr atoms lie at the corners, but not at the centers, of hexagons which form a planar net. These Cr planes are separated from one another by two layers of Cl atoms. The Cr spins ($S_1=S_2=S$; $g_1=g_2=g$) are assumed to be coupled to nearest neighbors only, with an exchange constant J. Since the two Cr sites in each (two-dimensional) unit cell are inequivalent, it is here assumed that $H_{a1}\neq H_{a2}$. If the elements of $L_{\mathbf{k}}$ are expanded for small $|\mathbf{k}|$, and $g\beta H_{a1}$, $g\beta H_{a2}\ll JS$ is taken, there results

$$\lambda_{q\pm}=g\beta(H+H_a)+6JS\pm 6JS[1-\tfrac{1}{4}(qb)^2];$$
$$|V_{1\pm}|^2=\tfrac{1}{2}(1\pm\Gamma),\quad |V_{2\pm}|^2=\tfrac{1}{2}(1\mp\Gamma); \tag{52}$$

where $q=|\mathbf{k}|$, $H_a=\frac{1}{2}(H_{a1}+H_{a2})$, b is the distance between nearest-neighbor Cr sites, $\Gamma=g\beta(H_{a1}-H_{a2})/12JS$, and Γ^2 has been neglected compared to 1. If now, in (46), the sum over s is restricted to the lower lying spin-wave branch (λ_{k-}) and the sum over $\mathbf{k}$ is replaced by an integral over all $\mathbf{k}$ space, the statistical averages of z components of spin on each site become

$$\langle S_1^z\rangle_f(T)=\langle S_1^z\rangle_f(0)[1-(1-\Gamma)F],$$
$$\langle S_2^z\rangle_f(T)=\langle S_2^z\rangle_f(0)[1-(1+\Gamma)F], \tag{53}$$

where

$$F=(\sqrt{3}KT/8\pi JS^2)\sum_{n=1}^{\infty} n^{-1}\exp[-ng\beta(H+H_a)/KT]$$

and where $\langle S_1^z\rangle_f(0)=\langle S_2^z\rangle_f(0)=S$ for this system. Thus, even for this simple model, the resonance frequencies for nuclei on sites 1 and 2 are expected to differ if $H_{a1}\neq H_{a2}$. The average of the two equations (53) gives $\langle S^z\rangle_f(T)=\langle S^z\rangle_f(0)[1-F]$, in agreement with the result of Narath.[6]

If $\epsilon_1=+1$, $\epsilon_2=-1$, and $t=\alpha$, β, the solution of the problem is shown by the following outline (where the phase factors for V_+, V_- are taken to be equal):

$$R_{\pm\pm}=\tfrac{1}{2}(L_{11}-L_{22})[1\pm(L_{11}+L_{22})A^{-1/2}];$$
$$R_{+-}=R_{-+}=\{L_{12}L_{21}[(L_{11}+L_{22})^2A^{-1}-1]\}^{1/2}; \tag{54}$$

$$\omega_{\alpha,\beta}=\tfrac{1}{2}[(L_{11}-L_{22})\pm B^{1/2}]; \tag{55}$$

$$|W_{+\alpha,\beta}|^2=\tfrac{1}{2}[1\pm(1+16C)^{-1/2}];$$
$$|W_{-\alpha,\beta}|^2=\tfrac{1}{2}[1\mp(1+16C)^{-1/2}]; \tag{56}$$

$$\gamma_\alpha=+1,\quad \gamma_\beta=-1; \tag{57}$$

$$B=(L_{11}+L_{22})^2-4L_{12}L_{21}; \tag{58}$$

$$C=L_{12}L_{21}(L_{11}L_{22}-L_{12}L_{21})/(L_{11}^2-L_{22}^2)^2; \tag{59}$$

where (57) follows with the aid of the condition (51), and where in (55) and (56) the + sign corresponds to the index α, the − to β. Finally, after some algebra, (47) reduces to

$$\langle S_j^z\rangle=\epsilon_j\{(S_j+\tfrac{1}{2})-N^{-1}\tfrac{1}{2}\textstyle\sum_{kt}(\langle n_{kt}\rangle+\tfrac{1}{2})$$
$$\times[(L_{11}+L_{22})B^{-1/2}+\gamma_t\epsilon_j]\},\quad j=1,2;\quad t=\alpha,\beta. \tag{60}$$

For antiferro- and ferrimagnetic cases, with only nearest-neighbor interactions and where each site would be crystallographically equivalent in the absence of spin, the energy levels $|\omega_{\alpha,\beta}|$ reduce to those given by Van Kranendonk and Van Vleck in their review article.[1] Again for these simple cases, the difference between the magnitudes of $\langle S_1^z\rangle$ and $\langle S_2^z\rangle$ persists, even at $T=0$.

V. DISCUSSION

The value of the present method lies in the complete specification of the transformations which diagonalize the Hamiltonian. This allows thermal average values, such as (46) and (47), to be computed for the model which has been treated. If the lattice is complicated ($T>2$), then it will generally be necessary to use numerical methods to solve the problem. The present method will then be of value since only Hermitian matrices are diagonalized here.

Sáenz[4] has recently carried out a very clever derivation of the eigenvalues of the present problem. His eigenvalues are identical with the ones obtained above, as is shown in the Appendix, although he did not obtain the transformation matrices. The method of Douglass[7] is very useful for special cases (namely, those where the matrices $\epsilon=[\epsilon_j\delta_{jj'}]$ and $L_{\mathbf{k}}$ commute), but can not be directly extended to the more general case treated here.

It would be of interest to investigate, in crystals which have similar nuclei on inequivalent sites, the expected difference in resonance frequencies for these nuclei, to see if this difference can be observed and meaningfully interpreted.

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APPENDIX

This Appendix discusses the relations between a recent paper by Sáenz[4] and the present work. Sáenz defines a matrix L_κ, where $\boldsymbol{\kappa}$ is the wave vector, which is related to the $L_{\mathbf{k}}$ of the present paper by a unitary transformation

$$L_\kappa(\text{Sáenz})=U_{\mathbf{k}}^{-1}L_{\mathbf{k}}U_{\mathbf{k}}\quad\text{when}\quad\boldsymbol{\kappa}=\mathbf{k}, \tag{A1}$$

[6] A. Narath, Phys. Rev. Letters **7**, 410 (1961).

[7] R. L. Douglass, Phys. Rev. **120**, 1612 (1960).

where $U_{\mathbf{k},jj'}=\epsilon_j \exp(i\mathbf{k}\cdot\mathbf{r}_j)\delta_{jj'}$. Therefore, the eigenvalues of L_κ(Sáenz) are the same as those of the present $L_\mathbf{k}$, and so the remaining discussion will be carried out in terms of the $L_\mathbf{k}$ and other notation of the present paper.

Sáenz used the result that the eigenvalues of $\epsilon L_\mathbf{k}$ ($\epsilon_{jj'}=\epsilon_j\delta_{jj'}$) are real if $L_\mathbf{k}$ is positive definite; it seems worthwhile to include a proof here. If $L_\mathbf{k}$ is Hermitian and positive definite, then

$L_\mathbf{k}^{-1}$ exists, and is Hermitian and positive definite; (A2)

$L_\mathbf{k}^{1/2}$, $L_\mathbf{k}^{-1/2}$ exist, and are Hermitian and positive definite. (A3)

Now, suppressing the index $\mathbf{k}$, let an eigenvalue of ϵL be μ, with a corresponding eigenvector x:

$$\epsilon L x=\mu x. \tag{A4}$$

Let $Lx=y$; $x=L^{-1}y$:

$$\epsilon y=\mu L^{-1}y. \tag{A5}$$

Take the inner product of each side of (A5) with y to get

$$(y,\epsilon y)=\mu(y,L^{-1}y). \tag{A6}$$

Since ϵ and L^{-1} are both Hermitian, the inner products in (A6) are real, and since L^{-1} is positive definite, $(y,L^{-1}y)\neq 0$. Therefore μ must be real. Hence L positive definite is sufficient (but not necessary) to insure μ real.

Sáenz has also proved, by continuity arguments, that the number of positive (negative) eigenvalues μ is equal to the number of positive (negative) elements in the matrix ϵ, provided $\mu\neq 0$. This can also be proved for each matrix L: $\mathrm{sig}(\epsilon L)=\mathrm{sig}\epsilon$ if L is positive definite (where sig$M\equiv$signature M). Let $L^{1/2}x=z$, $x=L^{-1/2}z$, and (A4) becomes

$$\epsilon L^{1/2}z=\mu L^{-1/2}z. \tag{A7}$$

Multiply on the left by $L^{1/2}$ to get

$$L^{1/2}\epsilon L^{1/2}z=\mu z. \tag{A8}$$

Now $\mathrm{sig}(L^{1/2}\epsilon L^{1/2})=\mathrm{sig}(\epsilon L)$, since both matrices have the same eigenvalue spectrum [(A4) and (A8)]. But $\mathrm{sig}(L^{1/2}\epsilon L^{1/2})=\mathrm{sig}\epsilon$, since the signature is invariant under a conjunctive transformation ($L^{1/2\dagger}=L^{1/2}$). Therefore, $\mathrm{sig}(\epsilon L)=\mathrm{sig}\epsilon$.

It finally remains to show that the eigenvalues found by Sáenz, namely, the absolute values of the (real) eigenvalues μ of ϵL, are the same as those found here. If V is the (unitary) matrix which diagonalizes L, as in Sec. II above, then

$$V^{-1}LV=L_0, \quad V^{-1}L^{1/2}V=L_0^{1/2}, \tag{A9}$$

where L_0, $L_0^{1/2}$ are diagonal matrices whose elements are λ_s, $\lambda_s^{1/2}$, respectively. But the matrix of (A8), whose eigenvalues are μ, is related by a unitary transformation, with V, to the matrix R of Sec. II:

$$V^{-1}L^{1/2}\epsilon L^{1/2}V=L_0^{1/2}V^{-1}\epsilon V L_0^{1/2}=R. \tag{A10}$$

Therefore, the eigenvalues of ϵL are the same as those of R, and this completes the proof. It also follows from (A10) that sig$R=$sigϵ, a result which was used in Sec. III.

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Role of Fermi Surface and Crystal Structure in Theory of Magnetic Metals

DANIEL MATTIS AND WILM E. DONATH
Thomas J. Watson Research Center, International Business Machines Corporation, Yorktown Heights, New York

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We have investigated the magnetic ground state of metals using an idealized theory of magnetism based on the Ruderman-Kittel-Yosida indirect exchange interaction. The preliminary, but suggestive, results reported here are for simple cubic structures and spherical Fermi surface. We find that as the number of electrons is increased, ferromagnetism is replaced by two different antiferromagnetic structures before the Néel state is finally obtained. The first transition occurs at $k_Fa\sim 0.5\pi$ (k_F=Fermi wave vector, a=lattice constant) at which value the rapid changeover occurs, from ferromagnetism to an antiferromagnetic structure of planes of uniform magnetization, which point along alternating directions. These planes lie perpendicular to the (1, 0, 0) axis at first, then abruptly change over to the (1, 1, 0) axis at $k_Fa\sim 0.7\pi$. The new configuration remains the ground state until k_Fa is further increased to 0.85π. But, at that value a final transition occurs to the Néel state. The Néel state (where every spin is surrounded by antiparallel nearest neighbors) then becomes increasingly stable and reaches maximum stability when the Fermi surface touches the zone boundary at $k_Fa=\pi$. It appears from our calculations that the above-mentioned states are the only stable spiral configurations in the simple cubic lattice without the introduction of anisotropy or nonlinearity into the theory.

EVERYONE knows that there are many competing theories of magnetic metals. Each must be judged on its merits in predicting not just the occurrence of ferromagnetism, but also in explaining with a minimum of adjustable parameters the various possible antiferromagnetic orderings.