shell.) The values obtained from this calculation are also given in Table I and agree with the measurements to within 0.5 μ_B/molecule, indicating that the atomic configurations of the rare-earth and iron atoms are not very different from those assumed in the calculations.[15] This conclusion is in agreement with that reached on the basis of the Mössbauer experiments.

The measured Curie temperatures are all sufficiently high so that the magnetic fields determined at 78°K are a good approximation to the 0°K values. This is borne out in the case of the $HoFe_2$ sample where Mössbauer measurements were also made at 4°K yielding a field only 3% higher than that found at 78°K.

CONCLUSIONS

These measurements have shown that the fields at the iron nucleus in the RFe$_2$ compounds (except $CeFe_2$) are all very similar. This indicates a corresponding similarity in the atomic configurations and suggests that the contribution to the field made by the conduction electron polarization is either small or, more likely, dominated by the interaction with the iron d electrons. The isomer shift indicates that the configuration is like that of metallic iron. Measurements of the magnetization confirm that the magnetic properties of the iron atoms in these compounds are similar to those of iron in its natural lattice. In the case of $CeFe_2$ the Mössbauer effect shows the presence of two magnetically non-equivalent types of iron. These are thought to result from the transfer of the cerium $4f$ electron to the iron d band. The isomer shift indicates that these electrons are not localized on particular iron atoms. It is suggested that the two magnetically distinct types of iron arise from a spatial spin-density fluctuation in the d band.

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[15] The agreement between the measured and the calculated values is improved if a somewhat smaller value is used for the magnetic moment of the iron atom.

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Spin Waves in Exchange-Coupled Complex Magnetic Structures and Neutron Scattering

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In this paper, we develop a spin-wave theory of the Holstein-Primakoff type for exchange-coupled crystals with an arbitrary number n of magnetic ions per primitive magnetic unit cell, when the resultant electronic spin vectors of these ions are mutually parallel or antiparallel in a given domain, except for spin-wave fluctuations. A simple and systematic method is presented for finding a complete set of normal spin-wave modes. This method is used to derive a convenient formula for the component of the total electronic spin vector of the magnetic ions in a domain along the axis of spin alignment. We show that there exists at least one "acoustic" branch among the $\leq n$ distinct branches of the spin-wave spectrum when the magnetic anisotropy and external magnetic field contributions vanish. For the case when all the $m\geq 1$ acoustic branches existing in the absence of these contributions are identical, we prove that the acoustic spin-wave energies corresponding to a given wave-number vector $\boldsymbol{\kappa}$ are of $O(|\boldsymbol{\kappa}|^{2/m})$ for $|\boldsymbol{\kappa}|\to 0$. The situation in which a single acoustic branch exists when no external magnetic field or anisotropy effects are present is studied in detail and, under suitable restrictions, an explicit formula is derived for the energies of the magnons of this branch for $|\boldsymbol{\kappa}|\to 0$. We apply this spin-wave theory to obtain general cross-section formulas for the one-magnon zero-phonon scattering of neutrons by the class of exchange-coupled crystals referred to in the first sentence of this abstract, when the magnetic ions in these crystals are completely quenched orbitally. The formulas in question are used to predict a spin-wave phenomenon of wide generality for polarized incident neutrons. This phenomenon is of particular experimental interest in connection with the acoustic spin-wave scattering of such neutrons by crystals of this class having a single acoustic branch and has been qualitatively confirmed by experiments on magnetite. For the last-mentioned crystals, we use an exact limit result of this paper to suggest a simple approximate form of the general cross-section equations pertaining to acoustic spin-wave scattering, when only magnons of sufficiently small $|\boldsymbol{\kappa}|$ are of interest.

I. INTRODUCTION

IN this paper, we employ a formalism which is a natural generalization of that of Holstein and Primakoff[1] to construct a spin-wave theory applicable to exchange-coupled crystals[2] with an arbitrary number of magnetic ions per primitive magnetic unit cell. We apply this theory to the spin-wave scattering of neutrons of any initial polarization by such crystals for the case

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

[2] The term "exchange" is employed in this paper to denote both ordinary exchange and superexchange, it being hoped that no confusion will be caused by this usage.

of complete orbital quenching of the magnetic ions. However, it is believed that the theory in question will be useful in a number of other problems.

A helpful summary of the customary spin-wave developments, especially for the simpler exchange-coupled spin structures, was given by Van Kranendonk and Van Vleck.[3] Several recent theoretical studies have dealt with the spin-wave spectra of various special exchange-coupled crystals with complex magnetic structures. Among those studies, we begin by mentioning the calculations of Kaplan on the spin-wave modes of cubic normal spinels[4] and his classical spin-wave analysis of the stability of such spinels.[5] Attention should be drawn to the investigations of Harris[6] and Douglass[7] on the spin-wave spectrum of yttrium iron garnet. This last paper is also valuable because of its general spin-wave theoretical developments. Finally, the work of Goedkoop[8] on spin waves in complex antiferromagnets should also be mentioned. The reader will find a more complete enumeration of earlier investigations on the spin-wave spectra of complex exchange-coupled crystals in references 3 and 4.

The study of spin waves in exchange-coupled solids by means of neutron scattering has received and is continuing to receive attention, both theoretically and experimentally. The reader is referred to an earlier publication[9] for a survey of the research in this field for ferromagnets and simple antiferromagnets.[10] As to exchange-coupled solids with complex magnetic structures, calculations of the one-magnon zero-phonon scattering of initially unpolarized neutrons by normal cubic spinels and by complex antiferromagnets have been carried out by Kaplan[11] and in reference 8, respectively. The conclusions in these last two papers are in good agreement with the corresponding experimental results on the spin-wave scattering of such unpolarized neutrons obtained by Brockhouse[12] and by Riste, Blinowski, and Janik[13] for magnetite, and by Riste and Wanik[14] for hematite. The experiments of Ferguson and Sáenz[15] on the spin-wave scattering of polarized neutrons incident on magnetite are particularly relevant from the standpoint of the present investigation, as will appear later.

We shall now summarize the main results of the present paper.

In Sec. II, we construct the generalized Holstein-Primakoff type of spin-wave theory alluded to above. For crystals having $n \geq 1$ magnetic ions per primitive magnetic unit cell, the magnetic Hamiltonian of interest is written as the sum of a part involving the exchange interactions and of a part representing the effects of an external magnetic field and of hypothetical anisotropy fields of a conventional type. Our only assumption concerning the long-range magnetic order is that, in a given domain, the resultant electronic spin-vector operators of the magnetic ions are mutually parallel or antiparallel, except for spin-wave fluctuations. Making the usual spin-wave approximations in the above magnetic Hamiltonian, we develop a simple general method for determining a complete set of normal spin-wave modes, under suitable conditions of positive definiteness. This method is employed to derive a convenient equation for the total electronic spin vector of all the magnetic ions in a domain along the pertinent axis of spin alignment. As expected, there are at most n distinct energy branches of the spin-wave spectrum. It is proved that at least one of these branches is of the "acoustic" type when external field and anisotropy effects are absent. For the situation when all the $m \geq 1$ acoustic branches existing in the absence of these effects are identical, we show that the energies of the acoustic magnons with wave-number vectors $\boldsymbol{\kappa}$ are of $O(|\boldsymbol{\kappa}|^{2/m})$ for $|\boldsymbol{\kappa}| \rightarrow 0$. Particular attention is paid in this section to a case which is believed to obtain for typical ferromagnets and ferrimagnets, namely, the case where there is exactly one acoustic branch when the external field and anisotropy contributions vanish. A necessary condition for the existence of such a unique acoustic branch is derived and an explicit limit formula for $|\boldsymbol{\kappa}| \rightarrow 0$ is given for the energies of the spin waves of this branch. This formula is valid under mild restrictions on the exchange-coupling constants and provided that each magnetic ion site has a point symmetry belonging to any one of 22 point groups.

In Sec. III, we employ the spin-wave theory of Sec. II to find general cross-section formulas for the one-magnon zero-phonon scattering of arbitrarily polarized incident neutrons by the class of exchange-coupled solids considered in the latter section, for the situation of complete orbital quenching of the magnetic ions. These formulas lead us to the prediction of interesting and widely occurring spin-wave phenomena for initially

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[9] A. W. Sáenz, Phys. Rev. **119**, 1542 (1960).
[10] Outside of the theoretical work on simple antiferromagnets in reference 9, see the detailed calculations of O. Nagai and A. Yashimori, Progr. Theoret. Phys. (Kyoto) **25**, 595 (1961), on the spin-wave spectrum of MnF_2 and on the scattering of neutrons by spin waves in this substance. The following experimental studies on the spin-wave scattering of initially unpolarized neutrons by ferromagnets, not reported in reference 9, should be noted: the measurements of R. D. Lowde and N. Umanatha, Phys. Rev. Letters **4**, 452 (1960) on iron and those of R. N. Sinclair and B. N. Brockhouse, Phys. Rev. **120**, 1638 (1960) on an fcc Co alloy.
[11] T. A. Kaplan, Bull. Am. Phys. Soc. **4**, 178 (1959).
[12] B. N. Brockhouse, Phys. Rev. **106**, 859 (1957); **111**, 1273 (1958).
[13] T. Riste, K. Blinowski, and J. Janik, J. Phys. Chem. Solids **9**, 153 (1959).
[14] T. Riste and A. Wanic, J. Phys. Chem. Solids **17**, 318 (1961).
[15] G. A. Ferguson, Jr. and A. W. Sáenz, J. Phys. Chem. Solids (to be published).

polarized neutrons. The phenomena in question pertain to the sensitive dependence of the intensity of neutrons scattered by a given spin-wave mode on the direction and magnitude of the initial neutron polarization and on the direction of magnetization of the scattering domains, this dependence being different for neutrons scattered by magnon emission processes than for those scattered by magnon absorption processes. After considering some of the general experimental implications of these phenomena, the latter are discussed in detail for the special case of acoustic spin-wave scattering by exchange-coupled crystals which are of the type treated in Sec. II and which have a single acoustic branch in the absence of anisotropy and external magnetic field contributions (more exactly, for scattering by magnons belonging to the branch in such lattices with energies $\epsilon_{\kappa,1}$ in the sense of Secs. II and III). An exact limit formula of Sec. III, whose proof is based on a result established in the Appendix, is exploited to suggest an essential simplification, of an approximate kind, of the general cross-section formulas of Sec. III pertaining to acoustic spin-wave scattering, for the last-mentioned crystals with a unique acoustic branch and for the case when only magnons with sufficiently small $|\kappa|$ are of interest. The resulting approximate cross-section equations obtainable in this manner are parallel in structure to the corresponding exact equations for $n=1$ (simple ferromagnets).

II. SPIN WAVES IN EXCHANGE-COUPLED LATTICES WITH $n\geq 1$

The indices $i, j, k=1, 2, \cdots, N$ will serve to designate the primitive magnetic unit cells, and the indices $\alpha, \beta, \gamma, \delta=1, 2, \cdots, n$ will be used to denote the magnetic ions in one of these cells.[16] The equilibrium position of the αth ion in the ith magnetic cell is $\mathbf{X}_{i,\alpha}=\mathbf{X}_i+\boldsymbol{\varrho}_\alpha$, where $\mathbf{X}_i$ is a lattice translation vector. The resultant electronic spin quantum number and spin vector operator of this last ion are denoted by S_α and $\mathbf{S}_{i,\alpha}$, respectively, with $S_\alpha\geq\frac{1}{2}$.

The direction of the z axis will be chosen to be collinear with the axis of spin alignment in a domain. It is convenient to introduce the diagonal matrix $\sigma\equiv[\sigma_\alpha\delta_{\alpha\beta}]$, where $\sigma_\alpha\equiv 1$ (-1) when $\mathbf{S}_{i,\alpha}$, except for spin-wave deviations, is parallel (antiparallel) to this z direction.

In the spirit of Holstein and Primakoff,[1] we are led to write for the components of $\mathbf{S}_{i,\alpha}$ with respect to a Cartesian set of axes x, y, z:

$$S_{i,\alpha}^{(x)}\pm iS_{i,\alpha}^{(y)}\cong S_\alpha^{\frac{1}{2}}\{q_{i,\alpha}\pm i\sigma_\alpha p_{i,\alpha}\}, \tag{2.1}$$

$$S_{i,\alpha}^{(z)}=\sigma_\alpha\{S_\alpha-n_{i,\alpha}\}, \tag{2.2}$$

$$n_{i,\alpha}=\tfrac{1}{2}\{p_{i,\alpha}^2+q_{i,\alpha}^2-1\}; \tag{2.3}$$

where $q_{i,\alpha}$ and $p_{i,\alpha}$ are Hermitian operators such that $[q_{i,\alpha},q_{j,\beta}]=[p_{i,\alpha},p_{j,\beta}]=0$ and $[p_{i,\alpha},q_{j,\beta}]=(1/i)\delta_{ij}\delta_{\alpha\beta}$, with $[a,b]\equiv ab-ba$. In this investigation, we shall disregard the restriction that the maximum eigenvalue of $n_{i,\alpha}$ is $2S_\alpha$.

We shall adopt the following model Hamiltonian to describe the pertinent interactions of the magnetic ions:

$$H\equiv-\sum_{i,j,\alpha,\beta} J_{i\alpha,j\beta}(\mathbf{S}_{i,\alpha}\cdot\mathbf{S}_{j,\beta})-\sum_{i,\alpha}\sigma_\alpha A_\alpha S_{i,\alpha}^{(z)}. \tag{2.4}$$

Here, $J_{i\alpha,j\beta}$ will be taken to depend solely on α, β, and $\mathbf{X}_{i,\alpha}-\mathbf{X}_{j,\beta}$, this last dependence being of the customary short-range variety. Moreover, as usual, $J_{j\beta,i\alpha}\equiv J_{i\alpha,j\beta}$ and $J_{i\alpha,i\alpha}\equiv 0$. The quantities A_α are linear and homogeneous in the components of the external magnetic field and in those of a hypothetical anisotropy field of conventional type acting on the αth magnetic ion in a unit cell, it being supposed that the direction of these fields is collinear with that of the z axis.

A. Spin-Wave Hamiltonian

Let us introduce the canonically conjugate variables

$$\begin{aligned} q_{\kappa(l),\alpha}&\equiv N^{-\frac{1}{2}}\textstyle\sum_j \exp[i\kappa(l)\cdot\mathbf{X}_{j,\alpha}]q_{j,\alpha}\\ &=q_{-\kappa(l),\alpha}^\dagger,\\ p_{\kappa(l),\alpha}&\equiv N^{-\frac{1}{2}}\textstyle\sum_j \exp[-i\kappa(l)\cdot\mathbf{X}_{j,\alpha}]p_{j,\alpha}\\ &=p_{-\kappa(l),\alpha}^\dagger; \end{aligned} \tag{2.5}$$

where the vectors $\kappa(l)$ $(l=1, 2, \cdots, N)$ are defined as usual in terms of the positions of the familiar N uniformly distributed points in a primitive unit cell of the reciprocal lattice of the lattice of the $\mathbf{X}_i$; where $B^\dagger$ denotes the adjoint of an operator or $n\times n$ matrix B; and where we have used the Hermiticity of the $q_{i,\alpha}$ and $p_{i,\alpha}$.

Let us also define a matrix L_κ with elements

$$\begin{aligned} L_{\kappa,\alpha\beta}\equiv 2\{\sigma_\alpha\sum_{k,\gamma}J_{k\gamma,0\alpha}\sigma_\gamma S_\gamma+\tfrac{1}{2}A_\alpha\}\delta_{\alpha\beta}-2\sigma_\alpha\sigma_\beta(S_\alpha S_\beta)^{\frac{1}{2}}\\ \times\sum_k J_{k\beta,0\alpha}\exp[i\kappa\cdot(\mathbf{X}_{k,\beta}-\mathbf{X}_{0,\alpha})], \end{aligned} \tag{2.6}$$

$\mathbf{X}_0$ corresponding to some fixed unit cell and κ being a vector with arbitrary real components. In this paper, κ will invariably be supposed to be real in this sense.

Making use of the properties of $J_{i\alpha,j\beta}$ stated previously, one obtains from (2.6):

$$L_\kappa^\dagger=L_\kappa, \tag{2.7}$$

$$L_{-\kappa}=L_\kappa^*; \tag{2.8}$$

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[16] Barring an explicit statement to the contrary, all equations, inequalities, and limits in the present paper involving any free lower case Greek or Roman indices listed in this sentence of the text should be understood to hold in the respective ranges $1, 2, \cdots, n$ and $1, 2, \cdots, N$. All sums and products over any one of the indices just alluded to extend over the full range of n or N values appropriate to the index in question, in the absence of explicit restrictions on such sums and products.

where B^* designates the complex conjugate of a matrix B.

From (2.1) to (2.6), employing the above properties of $J_{i\alpha,j\beta}$ and neglecting terms quadratic in $n_{i,\alpha}$, one obtains:

$$H=C+\tfrac{1}{2}\sum_{l,\alpha,\beta} L_{\kappa(l),\alpha\beta}\{p_{\kappa(l),\alpha}{}^{\dagger}p_{\kappa(l),\beta} +\sigma_\alpha\sigma_\beta q_{\kappa(l),\beta}{}^{\dagger}q_{\kappa(l),\alpha}\}, \quad (2.9)$$

$$C\equiv -N\{\sum_{k,\alpha,\beta}\sigma_\alpha\sigma_\beta(S_\alpha+1)S_\beta J_{k\beta,0\alpha} +\sum_\alpha (S_\alpha+\tfrac{1}{2})A_\alpha\}; \quad (2.10)$$

where the summation over l in (2.9) and all such summations henceforth extend over the N values of l mentioned above. From now on, the symbol H will be reserved exclusively to designate the approximate spin-wave Hamiltonian (2.9).

B. Diagonalization of the Spin-Wave Hamiltonian

For an exchange-coupled solid of the type of interest here, simple physical considerations indicate that the stability of the long-range magnetic ordering of the electronic spins requires that the excited eigenstates of H have energies *greater* than the corresponding ground state energy.[17]. Because of the peculiar structure of (2.9), a necessary and sufficient condition for these excited states to have the property in question is that $L_{\kappa(l)}$ be positive definite for any of the above N distinct vectors $\boldsymbol{\kappa}(l)$. This global condition of positive definiteness cannot be fulfilled when $A_\alpha=0$, since then at least one of the eigenvalues of L_0 vanishes, as we shall prove in subsection C of this section. This fact constituted our principal motivation for introducing the simple nonexchange terms in (2.4), following the current practice in various familiar special cases.

For large enough crystals, it is easily proved that the above positive definiteness condition on $L_{\kappa(l)}$ is equivalent to the mathematically more convenient requirement that L_κ be positive definite for all $\boldsymbol{\kappa}$. This last requirement will play a central role in most of the succeeding developments of this paper.

We shall denote by $L_\kappa{}^{(0)}$ the matrix L_κ when $A_\alpha=0$. It will be invariably supposed that $L_\kappa{}^{(0)}$ has the property that all its eigenvalues are non-negative for all $\boldsymbol{\kappa}$. This property will be called property (*a*) in what follows.

In virtue of (2.6), property (*a*) implies that L_κ is positive definite for all $\boldsymbol{\kappa}$ if $A_\alpha>0$. However, for arbitrary n, this last requirement on the A_α can be shown not to be necessary for L_κ to have this property.

Let the eigenvalues of σL_κ be denoted by $\mu_{\kappa,\alpha}$ and let us define

$$\epsilon_{\kappa,\alpha}\equiv|\mu_{\kappa,\alpha}|. \quad (2.11)$$

[17] For a discussion of this stability criterion for simple ferromagnets and antiferromagnets, see, for example, reference 3, pp. 13–14.

The symbols $\mu_{\kappa,\alpha}{}^{(0)}$ and $\epsilon_{\kappa,\alpha}{}^{(0)}$ will stand, respectively, for $\mu_{\kappa,\alpha}$ and $\epsilon_{\kappa,\alpha}$ for the case when $A_\beta=0$.

Combining the continuity properties of L_κ exhibited in (2.6) with (2.8) and with (2.11), it can be proved that the $\mu_{\kappa,\alpha}$ can be numbered in such a way that, for any fixed α, $\mu_{\kappa,\alpha}$ and $\epsilon_{\kappa,\alpha}$ are jointly continuous functions of the components κ_x, κ_y, and κ_z of $\boldsymbol{\kappa}$ and of all the A_β for all choices of these variables, and such that

$$\mu_{-\kappa,\alpha}=\mu_{\kappa,\alpha}{}^*, \quad (2.12)$$

$$\epsilon_{-\kappa,\alpha}=\epsilon_{\kappa,\alpha}. \quad (2.13)$$

Henceforth, we shall assume that the $\mu_{\kappa,\alpha}$ have been numbered in this fashion.

If L_κ is positive definite for every $\boldsymbol{\kappa}$, then, for fixed α, $\mu_{\kappa,\alpha}$ is either positive for all $\boldsymbol{\kappa}$ or negative for all $\boldsymbol{\kappa}$. Let us denote the sign of $\mu_{\kappa,\alpha}$ by ω_α when L_κ has this global positive definiteness property. *The numbers* $\omega_1, \omega_2, \cdots, \omega_n$ *and* $\sigma_1, \sigma_2, \cdots, \sigma_n$ *are equal to within a permutation.* These properties of the $\mu_{\kappa,\alpha}$ can be established straightforwardly by employing the continuity of the $\mu_{\kappa,\alpha}$ in $\boldsymbol{\kappa}$ and in the A_β, (2.7), and the fact that the Euclidean space whose points are all the ordered n-tuples $(A_1,A_2,\cdots,A_n)$ with values of A_β such that the corresponding L_κ are positive definite for all $\boldsymbol{\kappa}$ has the following properties: it is nonempty [since (a) implies that the points $(A_1,A_2,\cdots,A_n)$ with $A_\beta>0$ are in this space] and it is connected [because of (2.6)]. Using (2.11), we therefore find that

$$\epsilon_{\kappa,\alpha}=\omega_\alpha\mu_{\kappa,\alpha} \quad (2.14)$$

for all $\boldsymbol{\kappa}$ and α when the A_β are such that L_κ is positive definite for every $\boldsymbol{\kappa}$. Equation (2.14) also holds for $A_\beta=0$. To show this, one exploits the fact that this equation is true for $A_\beta>0$ to take the limit $A_\beta\to 0+$ of both sides of the equation in question, and one invokes the continuity of $\mu_{\kappa,\alpha}$ and $\epsilon_{\kappa,\alpha}$ with respect to the A_β.

In order to reduce H to a sum of uncoupled harmonic-oscillator Hamiltonians, let us introduce the matrices T_κ, which obey the simultaneous equations,

$$\sigma L_\kappa T_\kappa=T_\kappa\mu_\kappa, \quad (2.15)$$

$$T_\kappa{}^{\dagger}\sigma T_\kappa=\omega, \quad (2.16)$$

where $\mu_\kappa\equiv[\mu_{\kappa,\alpha}\delta_{\alpha\beta}]$ and $\omega\equiv[\omega_\alpha\delta_{\alpha\beta}]$.

Such matrices T_κ exist for all $\boldsymbol{\kappa}$ when L_κ is positive definite for all $\boldsymbol{\kappa}$. This statement is a consequence of (2.7) and of the equality of the signatures of σ and ω, in virtue of well-known matrix theorems.

When L_κ has this global positive definiteness property, it can be shown by means of (2.8) and (2.12) that one can find for all $\boldsymbol{\kappa}$ matrices T_κ which, outside of satisfying (2.15) and (2.16), also obey the requirement,

$$T_{-\kappa}=T_\kappa{}^*. \quad (2.17)$$

Consider a set of matrices $T_{\kappa(l)}$ satisfying (2.15) to (2.17) for all $\boldsymbol{\kappa}=\boldsymbol{\kappa}(l)$ corresponding to the N values of l stated above. Such matrices $T_{\kappa(l)}$ are nonsingular

because of (2.16). In terms of these $T_{\kappa(l)}$, we define the canonically conjugate variables $Q_{\kappa(l),\alpha}$ and $P_{\kappa(l),\alpha}$ as follows for the N values of l in question:

$$Q_{\kappa(l),\alpha}\pm iP_{\kappa(l),\alpha} \equiv\sum_\beta \{T_{\pm\kappa(l),\beta\alpha}q_{\pm\kappa(l),\beta}\pm iT_{\mp\kappa(l),\alpha\beta}{}^{-1}p_{\mp\kappa(l),\beta}\}; \quad (2.18)$$

where the superscript -1 indicates matrix inversion. Note that the $Q_{\kappa(l),\alpha}$ and $P_{\kappa(l),\alpha}$ are Hermitian because of (2.5) and (2.17).

If L_κ is positive definite for every $\boldsymbol{\kappa}$, one finds from (2.9) and from (2.13) to (2.18) that H can be reduced to the following diagonal form[18,19]:

$$H=H_0+\sum_{l,\alpha}\epsilon_{\kappa(l),\alpha}n_{\kappa(l),\alpha}, \quad (2.19)$$

$$H_0\equiv C+\tfrac{1}{2}\sum_{l,\alpha}\epsilon_{\kappa(l),\alpha}, \quad (2.20)$$

$$n_{\kappa(l),\alpha}\equiv\tfrac{1}{2}[P_{\kappa(l),\alpha}{}^2+Q_{\kappa(l),\alpha}{}^2-1]. \quad (2.21)$$

Since the eigenvalues of any of the $n_{\kappa,\alpha}$ are either zero or positive integers, the $\epsilon_{\kappa,\alpha}$ represent the energies of spin waves with wave-number vectors $\boldsymbol{\kappa}$, all these energies being positive if L_κ satisfies the condition of positive definiteness just stated (recall that then $\mu_{\kappa,\alpha}\neq0$ for any $\boldsymbol{\kappa}$ and α). One speaks of the set of energies $\epsilon_{\kappa,\alpha}$ for all $\boldsymbol{\kappa}$ and fixed α as the αth branch of the spin-wave spectrum. Obviously, there are $\leq n$ distinct branches of this spectrum.

At this point, it is easy to derive the simple formula, alluded to in Sec. I, for the component along the z axis of the total electronic spin-vector operator of the magnetic ions in a domain.

Since the ω_α are a permutation of the σ_α,

$$\sum_\beta\omega_\beta=\sum_\beta\sigma_\beta. \quad (2.22)$$

From (2.2), (2.3), (2.5), (2.16) to (2.18), (2.21), and (2.22), we obtain the desired formula[20]

$$\sum_{i,\alpha}S_{i,\alpha}{}^{(z)}=N\sum_\alpha\sigma_\alpha S_\alpha-\sum_{l,\alpha}\omega_\alpha n_{\kappa(l),\alpha}. \quad (2.23)$$

[18] Note that if all the σ_α have the same sign, i.e., if one is dealing with a ferromagnetic spin arrangement, then H_0 in (2.20) reduces to

$$H_0=-N\{\sum_{k,\alpha,\beta}S_\alpha S_\beta J_{k\beta,0\alpha}+\sum_\alpha S_\alpha A_\alpha\},$$

a result which agrees with the classical value of the ground state energy of such a ferromagnet. One can derive this result with the aid of (2.6), (2.10), (2.20), the circumstance that $J_{i\alpha,i\alpha}\equiv0$, and the identity

$$\sum_\alpha\epsilon_{\kappa,\alpha}=\sum_\alpha L_{\kappa,\alpha\alpha},$$

which is valid for all $\boldsymbol{\kappa}$ when every eigenvalue of L_κ is non-negative for any given $\boldsymbol{\kappa}$.

[19] It is instructive to contrast the present diagonalization method with that in reference 7.

[20] Let $\langle\ \rangle$ denote an average with respect to the canonical density operator $\exp[-\beta H]$ (β has its customary statistical mechanical meaning in this footnote). One finds that $\langle\mathbf{S}_{i,\alpha}\rangle$ is independent of i by a direct calculation based on the spin-wave developments of this section, a result which is also demanded by considerations of translational invariance. Combining this result with (2.23), one obtains:

$$\langle S_{i,\alpha}{}^{(z)}\rangle=\sum_\alpha\sigma_\alpha S_\alpha-\frac{1}{N}\sum_{l,\alpha}\omega_\alpha\langle n_{\kappa(l),\alpha}\rangle,$$

where, as usual, $\langle n_{\kappa,\alpha}\rangle\equiv\{\exp[-\beta\epsilon_{\kappa,\alpha}]-1\}^{-1}$.

C. Acoustic Spin Waves

As usual, a spin-wave branch with energies $\epsilon_{\kappa,\alpha}$ will be said to be an acoustic spin-wave branch if

$$\epsilon_{0,\alpha}=0. \quad (2.24)$$

We shall now prove that there is at least one acoustic spin-wave branch when $A_\beta=0$.

Since the $\mu_{\kappa,\alpha}{}^{(0)}$ are eigenvalues of $\sigma L_\kappa{}^{(0)}$, it is clear that

$$\prod_\beta\mu_{\kappa,\beta}{}^{(0)}=\det[\sigma L_\kappa{}^{(0)}], \quad (2.25)$$

where $\det[B]$ designates the determinant of any $n\times n$ matrix B. Because of (2.11) and (2.25), an acoustic branch exists for $A_\beta=0$ if and only if

$$\det[L_0{}^{(0)}]=0. \quad (2.26)$$

The result (2.26) can be proved immediately by using (2.6), which implies that

$$\sum_\beta L_{0,\alpha\beta}{}^{(0)}S_\beta{}^{\frac{1}{2}}=0, \quad (2.27)$$

so that the existence of at least one acoustic branch when $A_\gamma=0$ has been established.

There is a close connection between (2.27) and the commutativity of the spin-wave Hamiltonian (2.9) with the x and y components of the total spin vector operator $\sum_{i,\alpha}\mathbf{S}_{i,\alpha}$ for $A_\beta=0$, understanding these components in the approximate sense of (2.1). In fact, one finds from (2.1), (2.5), and (2.9), and from the circumstance that $L_0{}^{(0)}$ is a symmetric matrix in virtue of (2.7) and (2.8), that (2.27) guarantees the existence of these commutation properties, and that they would not exist if (2.27) were not true. The commutation properties in question are, of course, merely an expression of the invariance of the exact model Hamiltonian (2.4) with respect to rigid rotations of all the $\mathbf{S}_{i,\alpha}$ when $A_\beta=0$. We thus see that the existence of an acoustic branch for $A_\beta=0$ is intimately related to this last property of rotational invariance.

When not all the A_β vanish, it should be clear that there will not be an acoustic branch, except possibly by accident.

The manner in which the energies of acoustic spin waves vary with $|\boldsymbol{\kappa}|$ as $|\boldsymbol{\kappa}|\to0$ is of interest in a number of problems. We shall determine this $|\boldsymbol{\kappa}|$ dependence for $A_\alpha=0$ in situations when all $m\geq1$ acoustic energy branches are identical, i.e., in cases when

$$\left.\begin{array}{ll}\epsilon_{0,\alpha}{}^{(0)}=0, & 0\leq\alpha\leq m,\\ \epsilon_{0,\alpha}{}^{(0)}\neq0, & m+1\leq\alpha\leq n,\end{array}\right\} \quad (2.28)$$

and when

$$\epsilon_{\kappa,1}{}^{(0)}=\epsilon_{\kappa,2}{}^{(0)}=\cdots=\epsilon_{\kappa,m}{}^{(0)} \quad (2.29)$$

for every $\boldsymbol{\kappa}$.

To discover this dependence, we remark that $\det[L_\kappa{}^{(0)}]$ has the following properties for any $\boldsymbol{\kappa}$: it is real by (2.7), and it is therefore even in $\boldsymbol{\kappa}$ by (2.8), and it is analytic in the components of $\boldsymbol{\kappa}$ by (2.6). It there-

fore follows from (2.26) that

$$\det[L_\kappa^{(0)}]=F(\boldsymbol{\kappa})+O(|\boldsymbol{\kappa}|^4) \tag{2.30}$$

for $|\boldsymbol{\kappa}|\to 0$, where $F(\boldsymbol{\kappa})$ is a homogeneous quadratic function of the components of $\boldsymbol{\kappa}$.

If (2.28) holds and if (2.29) is satisfied for every $\boldsymbol{\kappa}$, then (2.11), (2.25), (2.30), and the continuity of the $\epsilon_{\kappa,\alpha}^{(0)}$ in the components of $\boldsymbol{\kappa}$ yield:

$$\epsilon_{\kappa,1}^{(0)}=|\boldsymbol{\kappa}|^{2/m}G(\boldsymbol{\kappa}) \tag{2.31}$$

for $m\geq 1$ and all $\boldsymbol{\kappa}$, where $G(\boldsymbol{\kappa})$ is a function which is continuous in $\boldsymbol{\kappa}$ in the sense just mentioned for $\boldsymbol{\kappa}\neq\mathbf{0}$ and which approaches a well-defined limit when $|\boldsymbol{\kappa}|\to 0$ for fixed $\boldsymbol{\kappa}/|\boldsymbol{\kappa}|$. We therefore see that the energies of the m coincident acoustic branches of interest are of $O(|\boldsymbol{\kappa}|^{2/m})$ for $|\boldsymbol{\kappa}|\to 0$.

If there is a unique acoustic branch when $A_\alpha=0$, we shall say that $L_\kappa^{(0)}$ has property (b). In the remainder of this subsection, we shall concern ourselves with the situation when this property holds.

Let us notice that (2.7), (2.8), and (2.27) imply that the matrix whose element in the αth row and βth column is $(S_\alpha S_\beta)^{\frac{1}{2}}L_{0,\alpha\beta}^{(0)}$ has the properties: it is a symmetric matrix all of whose elements are real and the sum of all the elements in any of its rows (columns) is zero. Because of these properties, it can be shown that

$$\mathrm{cof}[L_{0,\alpha\beta}^{(0)}]=\Gamma(S_\alpha S_\beta)^{\frac{1}{2}}/\textstyle\prod_\gamma S_\gamma, \tag{2.32}$$

$$\mathrm{cof}[(\sigma L_0^{(0)})_{\alpha\beta}]=\Gamma\sigma_\alpha(S_\alpha S_\beta)^{\frac{1}{2}}/\textstyle\prod_\gamma \sigma_\gamma S_\gamma. \tag{2.33}$$

Here, Γ is a real number which is independent of α and β, and $\mathrm{cof}[B_{\alpha\beta}]$ denotes the cofactor of the element $B_{\alpha\beta}$ of an $n\times n$ matrix B. Equations (2.32) and (2.33), which are readily seen to be equivalent, will be very useful in the following discussions of this subsection.

We proceed to prove that a necessary condition for (b) to hold is that

$$\textstyle\sum_\beta \sigma_\beta S_\beta\neq 0. \tag{2.34}$$

In fact, if (b) is fulfilled, then $\mu_{0,1}^{(0)}=0$ and $\mu_{0,\alpha}^{(0)}\neq 0$ for $\alpha\neq 1$, so that one finds under these circumstances:

$$\prod_{\beta\neq 1}{}' \mu_{0,\beta}^{(0)}=\sum_\gamma \mathrm{cof}[(\sigma L_0^{(0)})_{\gamma\gamma}]\neq 0, \tag{2.35}$$

because of a standard identity concerning sums of products of roots of a secular determinant.

From (2.33) and (2.35),

$$\prod_{\alpha\neq 1}{}' \mu_{0,\alpha}^{(0)}=\Gamma\sum_\beta \sigma_\beta S_\beta/\prod_\gamma \sigma_\gamma S_\gamma\neq 0, \tag{2.36}$$

when (b) obtains, so that (2.34) is evidently a necessary condition for (b) to be true.

If (a) and (b) are satisfied, then one has for the branch whose energies $\epsilon_{\kappa,1}$ reduce to those of the single acoustic branch when $A_\alpha\to 0$:[21]

$$\omega_1=\mathrm{sgn}\{\textstyle\sum_\beta \sigma_\beta S_\beta\}; \tag{2.37}$$

where we employ the usual notation "sgn" to designate the sign the pertinent number inside of the curly brackets. In virtue of (2.23), (2.37) simply expresses the fact that the excitation of spin waves with energies $\epsilon_{\kappa,1}$ decreases the magnitude of the z component of the total electronic spin in a domain, a result which is in accord with one's intuitive expectations.

To prove (2.37) under the above restrictions, we notice that (a) implies that the cofactor of any diagonal element of $L_\kappa^{(0)}$ is non-negative. Combining this result with (2.32) and (2.36), one sees that

$$\Gamma>0, \tag{2.38}$$

when (a) and (b) are satisfied.

Since $\mu_{0,\alpha}^{(0)}\neq 0$ for $\alpha\neq 1$ if (b) holds, we conclude from (2.11), (2.14) [in whose proof (a) was assumed to obtain], (2.36), and (2.38), when (a) and (b) are fulfilled:

$$\prod_{\alpha\neq 1}{}' \omega_\alpha=\mathrm{sgn}\{\sum_\beta \sigma_\beta S_\beta\}\prod_\gamma \sigma_\gamma. \tag{2.39}$$

But, since the ω_α are a permutation of the σ_α,

$$\prod_\beta \omega_\beta=\prod_\beta \sigma_\beta. \tag{2.40}$$

The desired result (2.37) under the conditions of interest is an immediate consequence of (2.39) and (2.40).

We shall conclude this subsection by deriving an explicit formula for $\epsilon_{\kappa,1}^{(0)}$ which should be useful in the study of long-wavelength acoustic spin waves in a number of ferrimagnets.

Using elementary properties of determinants, we can write:

$$\begin{aligned}\det[L_\kappa^{(0)}]=\Big[\Gamma\sum_{\alpha,\beta}(S_\alpha S_\beta)^{\frac{1}{2}}L_{\kappa,\alpha\beta}^{(0)}\\ +\sum_\alpha\Big(\sum_\beta (S_\alpha S_\beta)^{\frac{1}{2}}L_{\kappa,\alpha\beta}^{(0)}\Big)\\ \times\{(S_\alpha S_\delta)^{-\frac{1}{2}}[\textstyle\prod_\gamma S_\gamma]\,\mathrm{cof}[L_{\kappa,\alpha\delta}^{(0)}]-\Gamma\}\Big]/\displaystyle\prod_\gamma S_\gamma;\end{aligned} \tag{2.41}$$

where δ is any fixed integer from 1 to n.

Suppose now that we are dealing with the class of exchange-coupled crystals for which

$$\sum_{k,\beta}\sigma_\beta S_\beta J_{k\beta,0\alpha}(\mathbf{X}_{k,\beta}-\mathbf{X}_{0,\alpha})=\mathbf{0}. \tag{2.42}$$

As follows from a subsequent statement in this subsection, (2.42) is satisfied for a very large number of magnetic structures. For the moment, however, the pertinent point to notice is that (2.42), in conjunction

[21] It should be noted that (2.37) is an immediate consequence of (2.16) and of (A1) of the Appendix. Since this way of proving (2.37) is based on properties of the T_κ and since our definition of the ω_α in subsection A of this section is independent of the T_κ, it is not as direct a proof of (2.37) as that given in the text.

with (2.6), implies:

$$\sum_{\beta} (S_\alpha S_\beta)^{\frac{1}{2}} L_{\kappa,\alpha\beta}{}^{(0)}$$
$$=\sigma_\alpha S_\alpha \sum_{k,\beta} \sigma_\beta S_\beta J_{k\beta,0\alpha}[\boldsymbol{\kappa}\cdot(\mathbf{X}_{k,\beta}-\mathbf{X}_{0,\alpha})]^2+O(|\boldsymbol{\kappa}|^3) \quad (2.43)$$

for $|\boldsymbol{\kappa}|\rightarrow 0$.

From (2.32), (2.43), and the analyticity of the $L_{\kappa,\alpha\beta}{}^{(0)}$ in $\boldsymbol{\kappa}$ mentioned earlier, one deduces that the terms

$$\sum_\alpha \left(\sum_\beta (S_\alpha S_\beta)^{\frac{1}{2}} L_{\kappa,\alpha\beta}{}^{(0)}\right) \times\{(S_\alpha S_\delta)^{-\frac{1}{2}}[\prod_\gamma S_\gamma] \operatorname{cof}[L_{\kappa,\alpha\delta}{}^{(0)}]-\Gamma\}$$

in (2.41) vanish at least as rapidly as $|\boldsymbol{\kappa}|^3$ when $|\boldsymbol{\kappa}|\rightarrow 0$. Employing this fact in conjunction with (2.41) and (2.43), one concludes that

$$\lim_{\boldsymbol{\kappa}\rightarrow 0}\left\{\frac{\det[L_\kappa{}^{(0)}]}{|\boldsymbol{\kappa}|^2}\right\} =\Gamma \sum_{k,\alpha,\beta} \sigma_\alpha\sigma_\beta S_\alpha S_\beta \times J_{k\beta,0\alpha}(\boldsymbol{\xi}\cdot[\mathbf{X}_{k,\beta}-\mathbf{X}_{0,\alpha}])^2/\prod_\gamma S_\gamma, \quad (2.44)$$

when (2.42) holds. In (2.44), $\boldsymbol{\xi}$ is a unit vector in the direction of $\boldsymbol{\kappa}$ and the limit is taken for fixed $\boldsymbol{\xi}$.

Let us suppose that $L_\kappa{}^{(0)}$ has the properties (a) and (b), and that (2.42) is satisfied. Then, employing (2.14), (2.25), (2.36), (2.37), and (2.44), we obtain:

$$\lim_{\boldsymbol{\kappa}\rightarrow 0}\left\{\frac{\epsilon_{\kappa,1}{}^{(0)}}{|\boldsymbol{\kappa}|^2}\right\} = \sum_{k,\alpha,\beta} \sigma_\alpha\sigma_\beta S_\alpha S_\beta \times J_{k\beta,0\alpha}(\boldsymbol{\xi}\cdot[\mathbf{X}_{k,\beta}-\mathbf{X}_{0,\alpha}])^2/|\sum_\gamma \sigma_\gamma S_\gamma|. \quad (2.45)$$

This is the explicit limit formula for $\epsilon_{\kappa,1}{}^{(0)}$ alluded to previously. We should like to emphasize that (a) and (b) are not sufficiently strong to imply (2.45). For instance, one can construct hypothetical examples of exchange-coupled crystals such that (a) and (b), but not (2.42), are fulfilled, and such that (2.45) does not hold.

We shall now exhibit a wide class of magnetic structures for which (2.42) obtains.

Let us consider the usual case when, for fixed α and β, $J_{i\alpha,j\beta}$ depends only on $|\mathbf{X}_{i,\alpha}-\mathbf{X}_{j,\beta}|$, so that $J_{i\alpha,j\beta}=J_{\alpha\beta,n}$, where n refers to the fact that the sites $\mathbf{X}_{i,\alpha}$ and $\mathbf{X}_{j,\beta}$ are nth-nearest neighbors. Let $\sum_k{}^{(\alpha,\beta,n)}$ denote a summation over all sites $\mathbf{X}_{k,\beta}$ which are nth-nearest neighbors of a site $\mathbf{X}_{0,\alpha}$, for prescribed α, β, and n. A sufficient condition for (2.42) to hold for a given choice of the $J_{\alpha\beta,n}$ is that

$$\sum_k{}^{(\alpha,\beta,n)} (\mathbf{X}_{k,\beta}-\mathbf{X}_{0,\alpha})=\mathbf{0} \quad (2.46)$$

for all α, β, and n for which $J_{\alpha\beta,n}\neq 0$.

It can be shown that (2.46) is true for all α, β, and n, i.e., that (2.42) holds for an arbitrary selection of the $J_{\alpha\beta,n}$, when each magnetic ion occupies a site whose point symmetry belongs to one of the following 22 point groups:

$\bar{1}$, $2/m$, 222, mmm, $\bar{3}$, 32, $\bar{3}m$, $\bar{4}$, 422, $4/m$, $\bar{4}2m$, $4/mmm$, $\bar{6}$, 622, $6/m$, $\bar{6}m2$, $6/mmm$, 23, $m3$, 432, $\bar{4}3m$, $m3m$.

If the point symmetry of a magnetic ion site belongs to one of the remaining 10 point groups, then (2.47) cannot hold for all α, β, and n except by accident.[22]

III. SCATTERING OF NEUTRONS BY SPIN WAVES IN EXCHANGE-COUPLED CRYSTALS WITH $n\geq 1$

In treating the magnetic scattering of neutrons by the exchange-coupled crystals considered in Sec. II, we shall assume that the magnetic ions are completely quenched orbitally and that the magnetic electrons move rigidly with the corresponding nuclei, and shall make other familiar hypotheses listed by Van Hove[23] and in reference 9.[24] We shall restrict our attention to magnetic scattering processes involving all possible transitions in which there is a difference of ± 1 between the initial and final occupation numbers of one and only one of the energy levels $\epsilon_{\kappa(l),\alpha}$ in (2.19), and in which the initial and final vibrational states are identical (one-magnon zero-phonon processes).

Let a neutron, with wave-number vector $\mathbf{k}$ and polarization f along an arbitrary unit vector $\boldsymbol{\lambda}$, be scattered by such an exchange-coupled solid. Let the wave-number vector of the scattered neutron be $\mathbf{k}'\equiv\mathbf{k}+\mathbf{q}$, lying inside $d\Omega$, and let ϵ be the change in neutron energy on scattering. Denoting the neutron spin vector operator by $\mathbf{s}$ and an arbitrary unit vector independent of $\boldsymbol{\lambda}$ by $\boldsymbol{\lambda}'$, we designate the eigenvalues of $2(\boldsymbol{\lambda}'\cdot\mathbf{s})$ by ν, so that $\nu=\pm 1$.

The cross section per unit-energy range and per primitive magnetic unit cell for scattering a neutron by all possible one-magnon zero-phonon processes from $\mathbf{k}$ to $\mathbf{k}'$, and from the above initial spin state described by f and $\boldsymbol{\lambda}$ to an eigenstate of $(\boldsymbol{\lambda}'\cdot\mathbf{s})$ corresponding to a given ν is denoted by $d^2\sigma(\nu)/d\epsilon d\Omega$.

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[22] The results quoted in this paragraph were kindly pointed out to the author by Dr. E. Prince.

[23] L. Van Hove, Phys. Rev. **95**, 1374 (1954).

[24] In comparing the results of the present section with those of Sec. IV of reference 9, devoted to parallel questions, one should keep in mind the two observations below. The first of these concerns the presence of some typographical errors in the formulas of the section just cited, which are eliminated by making the following corrections in reference 9: removing the minus sign in the definition of $\psi_1(\mathbf{e};\alpha)$ in Eqs. (4.5); changing the signs of the terms $-2f(\mathbf{e}\cdot\boldsymbol{\lambda})(\mathbf{e}\cdot\boldsymbol{\mu})\mathfrak{N}(\epsilon,\mathbf{q})$ in (4.12), $+2\eta f(\mathbf{e}\cdot\boldsymbol{\lambda})(\mathbf{e}\cdot\boldsymbol{\mu})$ in (4.16a), and $+2\eta f(\mathbf{e}_0\cdot\boldsymbol{\lambda})(\mathbf{e}_0\cdot\boldsymbol{\mu})$ in (4.17); and replacing $d\theta>0(<0)$ in line 14 after Eq. (4.18) by $d\theta<0(>0)$. The second observation is that it has proved convenient in the present paper to use a somewhat different notation from that of Sec. IV of reference 9. The symbols α, $\varphi_1(\mathbf{e};\alpha)$, $\psi_1(\mathbf{e};\alpha)$ (when the above correction is made), $d^2\sigma_{01}(\alpha)/d\epsilon d\Omega$, and $d^2\sigma_{01}/d\epsilon d\Omega$ of this last reference correspond, respectively, to the symbols ν, $\varphi(\mathbf{e};\nu)$, $\psi(\mathbf{e};\nu)$, $d^2\sigma(\nu)/d\epsilon d\Omega$, and $d^2\sigma/d\epsilon d\Omega$ of the present publication.

Define

$$\left.\begin{aligned}\phi(\mathbf{e};\nu)\equiv 1+(\mathbf{e}\cdot\boldsymbol{\mu})^2+2\nu f\{([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\lambda}'])\\ -([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\mu}])([\mathbf{e}\times\boldsymbol{\lambda}']\\ \cdot[\mathbf{e}\times\boldsymbol{\mu}])-\tfrac{1}{2}(\boldsymbol{\lambda}\cdot\boldsymbol{\lambda}')[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2]\},\\ \psi(\mathbf{e};\nu)\equiv 2(\mathbf{e}\cdot\boldsymbol{\mu})(\mathbf{e}\cdot\{f\boldsymbol{\lambda}-\nu\boldsymbol{\lambda}'\});\end{aligned}\right\} \quad (3.1)$$

where $\mathbf{e}\equiv|\mathbf{q}|^{-1}\mathbf{q}$ and $\boldsymbol{\mu}$ is a unit vector parallel to a z direction chosen as mentioned in Sec. II.

To an accuracy consistent with our spin-wave approximations in Sec. II, we obtain the following result for the one-magnon zero-phonon scattering by large enough crystals, provided that L_κ is positive definite for all $\boldsymbol{\kappa}$:

$$\frac{d^2\sigma(\nu)}{d\epsilon d\Omega}=\sum_\alpha \frac{d^2\sigma_\alpha(\nu)}{d\epsilon d\Omega}, \quad (3.2)$$

$$\frac{d^2\sigma_\alpha(\nu)}{d\epsilon d\Omega}\equiv\tfrac{1}{2}\sum_{\eta=\pm1}\{\phi(\mathbf{e};\nu)+\eta\omega_\alpha\psi(\mathbf{e};\nu)\}\times U(\eta\epsilon)\mathcal{P}_\alpha(\epsilon,\mathbf{q}), \quad (3.3)$$

$$\mathcal{P}_\alpha(\epsilon,\mathbf{q})\equiv\tfrac{1}{2}(k'/k)\sum_{\eta'=\pm1}\sum_\tau\int d\boldsymbol{\kappa}\,\delta(\mathbf{q}-\boldsymbol{\kappa}-2\pi\boldsymbol{\tau})\times \mathcal{S}_{\kappa,\alpha}{}^\tau[\langle n_{\kappa,\alpha}\rangle+\tfrac{1}{2}(1-\eta')]\delta(\epsilon-\eta'\epsilon_{\kappa,\alpha}), \quad (3.4)$$

$$\mathcal{S}_{\kappa,\alpha}{}^\tau\equiv|\textstyle\sum_\beta \sigma_\beta S_\beta{}^{\frac{1}{2}}f_\beta(|\boldsymbol{\kappa}+2\pi\boldsymbol{\tau}|)\times\exp[-2\pi i\boldsymbol{\tau}\cdot\boldsymbol{\rho}_\beta]T_{\kappa,\beta\alpha}|^2; \quad (3.5)$$

where $U(\zeta)\equiv1(0)$ when $\zeta>0$ (<0); the $\boldsymbol{\kappa}$ integration runs over a primitive unit cell of the reciprocal lattice of the lattice of the $\mathbf{X}_i$; the $\boldsymbol{\tau}$ summation runs over all lattice vectors $2\pi\boldsymbol{\tau}$ of this reciprocal lattice; $\langle n_{\kappa,\alpha}\rangle$, given by the customary formula in footnote 20, is the average number of spin waves of the αth branch with wave-number vector $\boldsymbol{\kappa}$ for a certain temperature; $S_\beta f_\beta(|\mathbf{q}|)$ is the coherent magnetic scattering amplitude of the βth ion in a unit cell times the Debye-Waller exponential $\exp[-W_\beta]$; and the matrix elements $T_{\kappa,\beta\alpha}$ pertain to matrices T_κ which are solutions of (2.15) and (2.16), but not necessarily of (2.17). It should be clear that the value $1(-1)$ of η and η' corresponds to one-magnon absorption (emission) processes.

It is a straightforward matter to prove that the result for $d^2\sigma(\nu)/d\epsilon d\Omega$ expressed by (3.2) to (3.5) is correct to within the stated accuracy when one requires that the matrix elements of T_κ in (3.5) satisfy (2.15) to (2.17). Leaving the details to the reader, we mention that this result can be established either by employing the time-dependent approach of reference 23, as in a parallel calculation in Sec. IV of reference 9, or by using stationary-state methods. It is not immediately obvious that the result in question for $d^2\sigma(\nu)/d\epsilon d\Omega$ holds when it is only required that the $T_{\kappa,\beta\alpha}$ in (3.5) satisfy (2.15) and (2.16). To eliminate the condition that these matrix elements should obey (2.17), we proceed as follows. Let T_κ be a solution of (2.15) to (2.17), and let T_κ' be a solution of (2.15) and (2.16), but not necessarily of (2.17). Then the structure of (2.15) and (2.16) implies that $T_\kappa'=T_\kappa U_\kappa$, where U_κ is a unitary matrix which commutes with μ_κ. This relation between T_κ and T_κ' can be exploited to prove that the right-hand side of (3.5) remains unaltered in value when one replaces $T_{\kappa,\beta\alpha}$ by $T_{\kappa,\beta\alpha}'$ therein. Our last conclusion implies that it is unnecessary for the matrix elements in (3.5) to be solutions of (2.17), as far as concerns the validity of the cross-section equations of interest.

Carrying out a summation over the final neutron spin states, we obtain from (3.1) to (3.3):

$$\frac{d^2\sigma}{d\epsilon d\Omega}\equiv\sum_{\nu=\pm1}\frac{d^2\sigma(\nu)}{d\epsilon d\Omega}=\sum_\alpha\frac{d^2\sigma_\alpha}{d\epsilon d\Omega}, \quad (3.6)$$

$$\begin{aligned}\frac{d^2\sigma_\alpha}{d\epsilon d\Omega}&\equiv\sum_{\nu=\pm1}\frac{d^2\sigma_\alpha(\nu)}{d\epsilon d\Omega}\\ &=\sum_{\eta=\pm1}\{1+(\mathbf{e}\cdot\boldsymbol{\mu})^2+2\eta\omega_\alpha f(\mathbf{e}\cdot\boldsymbol{\lambda})(\mathbf{e}\cdot\boldsymbol{\mu})\}\times U(\eta\epsilon)\mathcal{P}_\alpha(\epsilon,\mathbf{q}).\end{aligned} \quad (3.7)$$

Let us consider variations of $\boldsymbol{\mu}$ for fixed values of the products $\sigma_\alpha\sigma_\beta$ $(\alpha\neq\beta)$, i.e., for a fixed relative orientation of the thermal expectation values of the $\mathbf{S}_{i,\alpha}$. For such variations, $d^2\sigma_\alpha(\nu)/d\epsilon d\Omega$ and $d^2\sigma_\alpha/d\epsilon d\Omega$ depend on $f\boldsymbol{\lambda}$, $\boldsymbol{\lambda}'$, and $\boldsymbol{\mu}$ only through the expressions inside of the curly brackets of (3.3) and (3.7), respectively, provided that the A_γ are independent of $\boldsymbol{\mu}$. This assertion can be verified with the aid of (2.6), (2.15), (2.16), (3.4), and (3.5), but we shall not prove it in detail here for the sake of brevity.

The dependence on the relative orientations of $\mathbf{e}$, $f\boldsymbol{\lambda}$, and $\boldsymbol{\mu}$ of the one-magnon zero-phonon scattering summed over the final neutron spins appears to be particularly accessible to experimental investigation. In principle, this dependence could be detected experimentally in the most direct manner by measuring the intensity of the neutrons scattered by magnons of a normal spin-wave mode whose energies do not coincide with those of any other such normal modes in the portion of $\boldsymbol{\kappa}$ space contributing to the pertinent one-magnon zero-phonon scattering. The following remarks concerning the situation when the energies of two or more such modes coincide for either some $\boldsymbol{\kappa}$ or all $\boldsymbol{\kappa}$ are experimentally relevant. The joint contribution, if any, of all these last-mentioned modes to $d^2\sigma/d\epsilon d\Omega$ is certainly dependent on f if all of the ω_α of the modes in question are identical. However, it should be kept in mind that this joint contribution may be independent of f if this last condition is violated. A realization of such a violation occurs for the simple antiferromagnets considered in Sec. IV of reference 9, for which it was shown therein that $d^2\sigma/d\epsilon d\Omega$ is independent of f in the absence of sig-

nificant external field and anisotropy effects, or more precisely, when $A_\beta \to 0+$.[25]

When $L_\kappa^{(0)}$ has the properties (a) and (b), (2.37) allows one to evaluate in a completely explicit manner the expressions inside of the curly brackets in (3.3) and (3.7) corresponding to the respective cross sections $d^2\sigma_1(\nu)/d\epsilon d\Omega$ and $d^2\sigma_1/d\epsilon d\Omega$ for the scattering by the spin-wave branch whose energies $\epsilon_{\kappa,1}$ tend to those of the unique acoustic branch when $A_\beta \to 0$. These explicit results are of particular relevance for ferromagnets and ferrimagnets.

Magnetite is a convenient solid for studying the dependence of the acoustic spin-wave scattering summed over the final neutron spins on the relative directions of $\mathbf{e}$, $f\boldsymbol{\lambda}$, and $\boldsymbol{\mu}$. It is known, for example from the experiments of reference 13, that (b) holds for magnetite.[26] In the experiments of reference 15, the scattering of initially polarized neutrons by acoustic magnons was measured in the vicinity of the (111) reflection of a magnetite crystal. In these last measurements, only the cases when $f\boldsymbol{\lambda}$ was parallel or antiparallel to $\boldsymbol{\mu}$ were considered, for experimental convenience, and no energy or polarization analyses of the scattered neutrons were carried out. The experimental results of reference 15 are in qualitative agreement with the dependence of $d^2\sigma_1/d\epsilon d\Omega$ on $\mathbf{e}$, $f\boldsymbol{\lambda}$, and $\boldsymbol{\mu}$ implied by the explicit bracketed expression in (3.7) alluded to in the preceding paragraph. We hope that quantitative tests of this dependence are forthcoming.

Outside of its intrinsic interest in testing the predictions of the spin-wave theory of Sec. II for various schemes of exchange coupling, the above dependence on the relative orientations of $\mathbf{e}$, $f\boldsymbol{\lambda}$, and $\boldsymbol{\mu}$ of the one-magnon zero-phonon scattering summed over the outgoing neutron spins may be of experimental importance in another connection, which we shall now mention. In the usual spin-wave scattering experiments, the only kinds of magnetic scattering which one would expect to interfere significantly with measurements of the one-magnon zero-phonon scattering are the magnetic elastic scattering and the type of magnetovibrational scattering denoted by the symbol (1,0) in Sec. IV of reference 9. The differential cross section per unit-energy range summed over the final neutron spins vanishes for the last two interfering scattering processes when $\mathbf{e}$, $\boldsymbol{\lambda}$, and $\boldsymbol{\mu}$ are collinear, while $d^2\sigma_\alpha/d\epsilon d\Omega$ can be doubled by an appropriate collinear arrangement of these unit vectors when $|f|=1$. In favorable cases, this fact could be employed to isolate the one-magnon zero-phonon scattering from the other two kinds of scattering processes mentioned above, by the use of highly polarized incident neutrons. This isolation method appears to be particularly promising in studies of the acoustic spin-wave scattering by suitable ferromagnetic and ferrimagnetic crystals. The method in question appears to be superior in certain respects to the standard procedure of magnetic separation of the one-magnon zero-phonon scattering of initially unpolarized neutrons, but a discussion of this matter lies beyond the bounds of the present paper.

From (3.5) and from (A1) of the Appendix, and taking it for granted that the f_β are continuous functions of $|\mathbf{q}|$ for all $\mathbf{q}$, one sees that (a) and (b) imply the following exact limit formula for the spin-wave scattering of interest by the branch with energies $\epsilon_{\kappa,1}^{(0)}$:

$$\lim_{\boldsymbol{\kappa}\to 0,\, A_\gamma \to 0} \mathcal{S}_{\kappa,1}{}^{\tau} = \left|\sum_\beta \sigma_\beta S_\beta f_\beta(2\pi|\boldsymbol{\tau}|) \times \exp[2\pi i \boldsymbol{\tau}\cdot\boldsymbol{\rho}_\beta]\right|^2 \Big/ \left|\sum_\gamma \sigma_\gamma S_\gamma\right| ; \quad (3.8)$$

where the limit is to be taken in precisely the same manner as in (A1). Furthermore under the conditions on $L_\kappa^{(0)}$ and the f_β just mentioned, $\mathcal{S}_{\kappa,1}{}^{\tau}$ is a continuous function of $\boldsymbol{\kappa}$ and of all A_α, provided that the absolute values of $\boldsymbol{\kappa}$ and of every one of the A_α are small enough, and that the A_α are such that L_κ is positive definite for all $\boldsymbol{\kappa}$. This assertion can be established by exploiting (3.5), (A4), and (A10), together with the continuity properties of $M_{\kappa,\alpha\beta}$ stated in the Appendix.

In the simple ferromagnetic case when $n=1$, one has

$$\mathcal{S}_{\kappa,1}{}^{\tau} = S_1[f_1(2\pi|\boldsymbol{\tau}|)]^2 \quad (3.9)$$

for any choices of $\boldsymbol{\kappa}$ and of the A_α, so that (3.8) is a trivial identity in this situation. To prove (3.9), we notice that, for $n=1$, the T_κ matrices contain but one element, which has an absolute value equal to unity in virtue of (2.16). Combining this observation with (3.5), the desired result (3.9) emerges.

Let us consider crystals with $n>1$ for which the hypotheses used in deriving (3.8) are valid, and such that the effects of magnetic anisotropy and of a possible external magnetic field are negligible with respect to the one-magnon zero-phonon scattering of neutrons. More precisely, let us suppose that these crystals are such that we can proceed to the limit $A_\alpha \to 0$, in the sense in which this limit is taken in (3.8), in the pertinent cross-section formulas without introducing significant errors. Furthermore, if the only acoustic magnons which contribute appreciably to this scattering are those with sufficiently small $|\boldsymbol{\kappa}|$, it would seem to be a reasonable approximation for the crystals in question to replace $\mathcal{S}_{\kappa,1}{}^{\tau}$ by the right-hand side of (3.8) in the expression for $\mathcal{P}_1(\epsilon,\mathbf{q})$ in (3.4), in virtue of the continuity property of $\mathcal{S}_{\kappa,1}{}^{\tau}$ in $\boldsymbol{\kappa}$ and in the A_β which was mentioned earlier. The approximate equation for $d^2\sigma_1(\nu)/d\epsilon d\Omega$ obtained in this manner has the same structure as the corresponding exact equation for $n=1$ which can be readily deduced with the aid of (3.9).

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[25] We are referring to the case in which $n=2$, $\sigma_1=-\sigma_2$, and $\epsilon_{\kappa,1}^{(0)}=\epsilon_{\kappa,2}^{(0)}$ for all $\boldsymbol{\kappa}$. Clearly, $\omega_1=-\omega_2$ for these simple antiferromagnets.

[26] For completeness, we list the values of the ω_α for the exchange-coupled model of magnetite in reference 4, choosing $\boldsymbol{\mu}$ in such a way that $\sum_\beta \sigma_\beta S_\beta > 0$. In this model, $J_{i\alpha,j\beta}=-J<0$ ($J_{i\alpha,j\beta}=0$) for any two sites which are (are not) nearest-neighbors $A-B$ sites, and $S_\alpha=S_A(S_B)$ for $A(B)$ sites. The two identical branches with energies $12JS_A$ for all $\boldsymbol{\kappa}$ when $A_\beta=0$ have equal ω_α, namely 1. For the remaining branches, whose energies are 0, $12J(2S_B-S_A)$, $12JS_A$, and $24JS_B$ for $\boldsymbol{\kappa}=0$ and $A_\beta=0$, one finds that the respective ω_α are 1, -1, 1, -1.

In his work on spin-wave scattering alluded to in Sec. I, Kaplan[11] derived an equation essentially equivalent to (3.8) for the special normal spinels considered by him in reference 4. He employed this result to construct an approximate cross-section formula for initially unpolarized neutrons which follows as a particular case from our approximate cross-section equation mentioned in the preceding paragraph.

APPENDIX

In this Appendix, we shall prove that, if $L_\kappa^{(0)}$ possesses the properties (a) and (b), then one has the following limit formula for the elements $T_{\kappa,\alpha 1}$ of any matrices T_κ which satisfy (2.15) and (2.16):

$$\lim_{\boldsymbol{\kappa}\to 0, A_\gamma\to 0} \{T_{\kappa,\alpha 1}T_{\kappa,\beta 1}^*\} = (S_\alpha S_\beta)^{\frac{1}{2}}/|\sum_\gamma \sigma_\gamma S_\gamma|\,; \quad \text{(A1)}$$

where the limit in (A1) is to be taken with respect to $\boldsymbol{\kappa}$ and all the A_γ in such a way that $A_\gamma \to 0$ only through values of the A_γ for which L_κ is positive definite for all $\boldsymbol{\kappa}$, for example, only through positive A_γ; and where the order of the limiting operations on $\boldsymbol{\kappa}$ and the A_γ is immaterial. Our motivation for letting the $A_\gamma \to 0$ in (A1) in the manner just specified is the fact, whose proof was outlined in **B** of Sec. II, that solutions T_κ of (2.15) and (2.16) exist when L_κ is positive definite for every $\boldsymbol{\kappa}$.

Define

$$M_\kappa \equiv \sigma L_\kappa - \mu_{\kappa,1} I, \quad \text{(A2)}$$

where I is the $n\times n$ unit matrix.

The following properties of M_κ will play an essential role in our proof of (A1):

$$\text{cof}[M_{\kappa,\alpha\beta}] \neq 0, \quad \text{(A3)}$$

if (b) holds, and provided that $|\boldsymbol{\kappa}| < \rho$ and $|A_\gamma| < \mathfrak{A}$. The symbols ρ and $\mathfrak{A}$ denote two positive numbers which are the same for all α and β, and which are independent of $\boldsymbol{\kappa}$ and of the A_γ. Moreover, if (a) and (b) hold, and if these last restrictions on $\boldsymbol{\kappa}$ and on the A_γ are satisfied, then

$$\text{sgn}\{\sum_\gamma \sigma_\gamma |\text{cof}[M_{\kappa,\alpha\gamma}]|^2\} = \omega_1. \quad \text{(A4)}$$

We proceed to establish these properties of M_κ. When (b) obtains, (A3) holds for $\boldsymbol{\kappa}=\mathbf{0}$ and $A_\gamma=0$. In fact, if (b) is fulfilled, then (A2), the vanishing of $\mu_{0,1}^{(0)}$, (2.33), and the inequality in (2.36) lead to the result:

$$\text{cof}[M_{0,\alpha\beta}] = \Gamma\sigma_\alpha (S_\alpha S_\beta)^{\frac{1}{2}} / \prod_\gamma \sigma_\gamma S_\gamma \neq 0, \quad \text{(A5)}$$

when $A_\gamma = 0$. Because of (A5) and (2.37), (a) and (b) imply that (A4) is true for $\boldsymbol{\kappa}=\mathbf{0}$ and $A_\gamma=0$.

The existence of the above positive numbers ρ and $\mathfrak{A}$, such that (A3) and (A4) obtain for $|\boldsymbol{\kappa}| < \rho$ and $|A_\gamma| < \mathfrak{A}$ under the stated requirements on $L_\kappa^{(0)}$, now follows directly from the conclusions in the preceding paragraph, and from the joint continuity of the $M_{\kappa,\alpha\beta}$ in $\boldsymbol{\kappa}$ and in the A_γ.

Let R be the nonempty set of all points $(A_1, A_2, \cdots, A_n)$ whose A_α are such that L_κ is positive definite for all $\boldsymbol{\kappa}$ and are such that $|A_\alpha| < \mathfrak{A}$ [R is nonempty because the points $(A_1, A_2, \cdots, A_n)$ which obey the inequalities $0 < A_\alpha < \mathfrak{A}$ are in R in virtue of the fact, stated in B of Sec. II, that L_κ is positive definite for all $\boldsymbol{\kappa}$ when $A_\alpha > 0$]. It is obvious that (A3) and (A4) hold when $|\boldsymbol{\kappa}| < \rho$ and when $(A_1, A_2, \cdots, A_n) \in R$, provided, of course, that the conditions on $L_\kappa^{(0)}$ stated in connection with (A3) and (A4) obtain. The conclusions in B of Sec. II imply trivially that solutions T_κ of (2.15) and (2.16) certainly exist when the conditions on $\boldsymbol{\kappa}$ and on the A_α in the last sentence are fulfilled.

According to (2.15), (2.16), and (A2), the $T_{\kappa,\alpha 1}$ obey the equations:

$$\sum_\beta M_{\kappa,\alpha\beta} T_{\kappa,\beta 1} = 0, \quad \text{(A6)}$$

$$\sum_\beta \sigma_\beta |T_{\kappa,\beta 1}|^2 = \omega_1. \quad \text{(A7)}$$

If (b) obtains and if $|\boldsymbol{\kappa}| < \rho$ and $(A_1, A_2, \cdots, A_n) \in R$, then one can employ (A3) to show that M_κ has rank $n-1$. One therefore sees that, under these conditions, (A3) implies that the most general solution of the linear equations (A6) is

$$T_{\kappa,\alpha 1} = c_{\kappa,\delta}\, \text{cof}[M_{\kappa,\delta\alpha}]; \quad \text{(A8)}$$

where $\alpha = 1, 2, \cdots, n$; δ is any of these n numbers and is the *same* for all α; and $c_{\kappa,\delta}$ is a complex number independent of α.

Let (a) and (b) hold, and let $|\boldsymbol{\kappa}| < \rho$ and $(A_1, A_2, \cdots, A_n) \in R$. We then conclude from (A4) that the $T_{\kappa,\alpha 1}$ in (A8) satisfy (A7) if and only if we choose the $c_{\kappa,\delta}$ in such a way that

$$|c_{\kappa,\delta}|^2 = 1/|\sum_\gamma \sigma_\gamma |\text{cof}[M_{\kappa,\delta\gamma}]|^2|, \quad \text{(A9)}$$

which is always possible under these circumstances, because then (A4) guarantees that the denominator of the right-hand side of (A9) is nonvanishing.

If the conditions on $L_\kappa^{(0)}$, $\boldsymbol{\kappa}$, and the A_β stated in the preceding paragraph obtain, then (A8) and (A9) imply that any pair of elements $T_{\kappa,\alpha 1}$ and $T_{\kappa,\beta 1}$ of an arbitrary solution T_κ of (2.15) and (2.16) satisfy the equation:

$$T_{\kappa,\alpha 1} T_{\kappa,\beta 1}^* = \text{cof}[M_{\kappa,\delta\alpha}]\, \text{cof}[M_{\kappa,\delta\beta}^*] / |\sum_\gamma \sigma_\gamma |\text{cof}[M_{\kappa,\delta\gamma}]|^2|. \quad \text{(A10)}$$

Under the requirements specified in the first paragraph of this Appendix, the limit formula (A1) is a direct consequence of (A5), (A10), and the continuity properties of the $M_{\kappa,\alpha\beta}$ mentioned above.