for nickel, approximately $3.4\times10^{-5}M_0$. As the temperature is increased the $\alpha^{\frac{3}{2}}$ term will tend to dominate among the deviations in M_1, and grows linearly with T. The two anisotropic effects tend to cancel each other at about 7.5°K. At temperatures above this the M_1 effect dominates, i.e., the greater magnetization occurs in the easy directions. Below this temperature the greater magnetization occurs in the hard directions.

There is an interesting application of this work to the case of iron. Although the pseudodipolar coupling may be present, the problem of the anisotropy in iron has been treated more easily than that in nickel because of the possibility of using a pseudoquadrupolar interaction, which yields K_1 in first order.[10] This interaction must be measured by a parameter whose magnitude is $\sim D^2/J$, if it is to explain completely the anisotropy in iron. The anisotropy in magnetization resulting from pseudoquadrupolar coupling again has the two parts. The contribution to M_1 will be exactly the same as in the dipolar case. However, the contribution to M_2 must occur in a higher order of approximation than the contribution to K_1. That is, in second order, we may have a term in M_β of order of magnitude $(D/J)^4$ coming from the pseudoquadrupolar coupling. According to our discussion, such a term cannot be observed at any reasonable temperature. Thus, we can imagine the possibility of determining, experimentally, to what extent the anisotropy in iron is due to pseudodipolar coupling by observation of magnetization anisotropy.

[10] See reference 3; also C. Zener, Phys. Rev. **96**, 1335 (1954), and F. Keffer, Phys. Rev. **100**, 1692 (1955).

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Magnetic Scattering of Neutrons by Exchange-Coupled Lattices*

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The magnetic scattering of slow neutrons of arbitrary initial polarization by an extensive class of magnetically-coupled lattices is treated by a time-dependent operator approach for the case of complete orbital quenching of the magnetic ions. This magnetic scattering is carefully divided into purely magnetic and magnetovibrational scattering, the types thereof involving, respectively, only zero-phonon processes and solely nonzero-phonon processes, and general formulas for these two types of scattering are obtained. These formulas are applied in temperature regions which are sufficiently large (I) or sufficiently small (II) compared with the temperature above which paramagnetism obtains. In region I, for the purely magnetic scattering and under certain invariance requirements on the above magnetic coupling, we analyze the energy spectrum of outgoing initially unpolarized neutrons of sufficiently high incident energy by a moment method. We thus obtain general formulas for the energy-integrated effective differential cross section defined in this paper and for the moments of energy transfer defined therein. These formulas involve certain spin averages, explicit equations being given for a wide range of these averages for exchange-coupled lattices. These results are illustrated numerically and compared with experiment for the case of polycrystalline MnF_2. In region II, we discuss certain broad features of the purely magnetic one-magnon scattering of arbitrarily polarized neutrons by exchange-coupled lattices of the class alluded to previously and by more complex ones, studying this scattering in detail for ferromagnets and certain antiferromagnets. A new spin-wave effect is pointed out for polarized neutrons incident on ferromagnets. Brief treatments of the magnetovibrational scattering in regions I and II are also given.

I. INTRODUCTION

In this paper, we shall investigate the magnetic scattering of slow neutrons by a wide class of lattices having magnetic ions which are orbitally quenched and are magnetically coupled with one another. Our objective is twofold. First, we want to derive formulas which are general enough to encompass the case of neutrons of arbitrary initial polarization f incident on lattices of the above class at any temperature T. Second, we desire to employ these formulas in a detailed study of magnetic neutron scattering by certain lattices of this class, and particularly by exchange-coupled ones,[1] for temperatures T which are sufficiently high or low compared with the temperatures T_c above which they are paramagnetic, a restriction which permits us to base our results on reliable quantum-statistical methods.

In what follows, the neutron magnetic scattering processes of interest, in which the initial and final lattice states are the same or different with regard to their vibrational quantum numbers, shall be defined as corresponding to purely magnetic or to magnetovibrational scattering, respectively. In general, the purely magnetic scattering is of greater physical

* The main results of this paper were first reported in Bull. Am. Phys. Soc. **2**, 49 (1957); **3**, 203 (1958).

[1] 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.

interest than the magnetovibrational scattering, which could be of comparable or even larger magnitude, so that it is desirable, both experimentally and theoretically, to separate the first type of scattering from the second.

Halpern and Johnson[2] derived intensity and polarization formulas for the purely magnetic neutron scattering in a basic paper which is centered around the two extreme cases of a set of uncoupled paramagnetic ions and of ferromagnets with their electronic spins locked rigidly in an ordered arrangement. Van Hove[3,4] devised a very general and powerful time-dependent approach to both nuclear and magnetic scattering of slow neutrons which we shall exploit in the present study.

A number of theoretical and experimental investigations have dealt specifically with the effects of exchange coupling on the magnetic scattering of unpolarized neutrons in the paramagnetic and spin-wave regions.

In regard to the paramagnetic region, several investigations of the purely magnetic scattering by these lattices have been carried out, principally by means of moment methods. Van Vleck[5] first pointed out the relationship between the inelasticity of this type of paramagnetic scattering and the exchange constants of the pertinent lattices as early as 1939. Second and fourth moments of neutron energy transfer were computed for cases of nearest-neighbor exchange interactions, and a local cluster-model computation for such a case was also done. Interference effects due to short-range magnetic order were not considered by Van Vleck. Employing stationary-state methods, Slotnick[6] dealt with the purely magnetic scattering by paramagnetic exchange-coupled lattices with interactions between nearest and next-nearest neighbors, taking into account these interference effects. This author studied the dependence of the effective energy-integrated cross section $d\bar{\sigma}_0/d\Omega$ for a $1/v$-detector [see our Eqs. (3.6a)] on T and on the initial neutron energy $E_{\mathbf{k}}$, and presented calculations of first and second moments of energy transfer. However, a systematic procedure for computing the above cross section and moments was not given. After the substantial completion of our paper, we became aware that de Gennes[7] investigated the purely magnetic scattering of neutrons by Van Hove's approach.[4] De Gennes dealt mainly with such large values of T in the paramagnetic region that the T dependence of the quantities of interest could be disregarded. This author treated the energy transfer and the momentum transfer vector of a neutron as independent variables, in contrast with the viewpoint adopted, for instance, in reference 6 and in Sec. III of this paper. De Gennes derived some general properties of his moments of energy transfer, and formulas for the second and fourth of these moments for cases of next-nearest neighbor interactions. Various plausible formulas, giving explicitly the dependence of the scattered neutron intensity of the energy transfer and wave-number transfer vector of a neutron, have been obtained in references 5, 6, and in the third part of reference 7 for high enough T by fitting moments of energy transfer. Rigorously derived formulas of this kind are not available.

In regard to the spin-wave region, Van Hove[4] studied the purely magnetic one-magnon scattering of neutrons for ferromagnets by combining his method of time-dependent correlation functions with the spin-wave formalism of Holstein and Primakoff.[8] Elliot and Lowde[9] investigated this spin-wave scattering by both ferromagnets and antiferromagnets using the above formalism and that of Ziman[10] for antiferromagnetic substances. They also discussed the magnetovibrational scattering, and gave rules for separating it experimentally from the purely magnetic scattering. Maleev[11] performed an improved computation of the above type of ferromagnetic spin-wave scattering. Kaplan[12] treated this purely magnetic scattering by spin waves in normal spinels with nearest-neighbor A-B exchange interactions.[13] Earlier theoretical papers on the scattering of unpolarized neutrons by spin waves are listed in references 4, 9, and 11.

With respect to experimental studies of the magnetic scattering of neutrons in the paramagnetic region, Shull and his collaborators, Erickson, and Brockhouse and others[14] observed the effects of short-range magnetic order on the above effective integrated cross section in this region. Inelastic magnetic scattering was studied by indirect methods by Bendt[15] and by direct energy-analysis methods by Brockhouse and his collaborators.[16] In a broad manner, these studies confirm the theories of Van Vleck[5] and Slotnick.[6] More data on the paramagnetic scattering of neutrons by the simpler types of exchange-coupled lattices, under conditions for which the various theories for such scattering just mentioned[5–7] and the pertinent conclusions in Sec. III of this paper are valid, are required

[2] O. Halpern and M. H. Johnson, Phys. Rev. **55**, 848 (1939).
[3] L. Van Hove, Phys. Rev. **95**, 249 (1954).
[4] L. Van Hove, Phys. Rev. **95**, 1375 (1954).
[5] J. H. Van Vleck, Phys. Rev. **55**, 924 (1939).
[6] M. Slotnick, Phys. Rev. **83**, 1226 (1955).
[7] P. G. de Gennes, Service de Physique Mathématique, Centre d'Etudes Nucléaires à Saclay, Report No. 199, November 1956 (unpublished); Compt. rend. **244**, 752 (1957); J. Phys. Chem. Solids **4**, 223 (1958). The essential content of this work is in the first and third of these studies.
[8] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).
[9] R. J. Elliot and R. D. Lowde, Proc. Roy. Soc. (London) **A230**, 46 (1955).
[10] J. M. Ziman, Proc. Phys. Soc. (London) **A65**, 540 (1952).
[11] S. V. Maleev, J. Exptl. Theoret. Phys. U.S.S.R. **33**, 1010 (1957) [translation: Soviet Phys.-JETP **6** (**33**), 776 (1958)].
[12] T. A. Kaplan, Bull. Am. Phys. Soc. **4**, 178 (1959).
[13] Kaplan's treatment is based on his spin-wave analysis of these spinels in Phys. Rev. **109**, 782 (1958).
[14] C. G. Shull, W. A. Strausser, and E. O. Wollan, Phys. Rev. **83**, 333, (1951); R. A. Erickson, Phys. Rev. **90**, 779 (1953); B. N. Brockhouse, L. M. Corliss, and J. M. Hastings, Phys. Rev. **98**, 1721 (1955).
[15] P. J. Bendt, Phys. Rev. **89**, 562 (1953).
[16] B. N. Brockhouse, Phys. Rev. **99**, 601 (1955); P. K. Iyengar and B. N. Brockhouse, Bull. Am. Phys. Soc. **3**, 195 (1958).

before more definitive experimental tests of this theoretical work are possible.

Among the experimental investigations of spin-wave scattering, we mention that of Lowde for iron and that of Riste, Blinowski, and Janik for magnetite,[17] involving no energy analysis of the outgoing neutrons, and the studies of Brockhouse[18] on the energy spectrum of neutrons scattered by magnetite. These spin-wave experiments support the main conclusions of Elliot and Lowde[9] and Kaplan.[12,13,19]

We now summarize the contents of the present investigation.

In Sec. II, using the methods of reference 4, we derive general formulas for the differential cross section per unit energy range for the magnetic scattering of arbitrarily polarized neutrons into a given final spin state by the class of lattices alluded to previously, separating carefully the purely magnetic from the magnetovibrational scattering.

In Sec. III, we treat the scattering of neutrons by lattices of the above class in the paramagnetic domain, supposing that the magnetic interactions of the pertinent ions have certain invariance properties. This hypothesis is satisfied, in particular, in the case of lattices of this class for which these magnetic interactions are of the exchange type. We study the purely magnetic scattering of unpolarized neutrons for sufficiently large $E_{\mathbf{k}}$ and T by means of a systematic moment method, which yields equations for the previously mentioned energy-integrated cross section and suitably defined relative moments of general order in the form of power series in $1/E_{\mathbf{k}}$ and $1/T$, with coefficients involving certain averages. An extensive set of such averages is given explicitly for exchange-coupled lattices, which, when combined with the power-series formulas just alluded to, yields results which include previous ones[5–7] of this type as special cases, as well as new results. For the sake of completeness, we show that the moments defined by de Gennes, for example in the third part of reference 7, can be obtained from our absolute moments for the purely magnetic scattering by a simple limiting process. Our theory for this scattering is illustrated by numerical calculations for neutrons of wavelengths 1A and 2A incident on polycrystalline MnF_2 in the paramagnetic state, for two hypothetical cases of exchange-coupling. These numerical results are compared with experiment.[14,15] The rather unimportant case of polarized neutrons scattered purely magnetically in the paramagnetic domain is not treated. A rough equation is derived relating the magnetovibrational scattering to that portion of the inelastic incoherent nuclear scattering arising solely from the magnetic ions.

In Sec. IV, we begin by applying the usual spin-wave theories[8,10,20] to construct a framework for a systematic separation and calculation of the purely magnetic and magnetovibrational cross sections of various types for neutrons of arbitrary f incident on the lattices of the class specified in Sec. II for the case of exchange coupling and for $T \ll T_c$. Formulas for the one-magnon zero-phonon scattering are obtained for these lattices, and their straightforward extension to more complicated ones is indicated and compared with experiment.[18] The only restriction on the magnetic order in the above work is that the spins in each domain be aligned along a unique axis. This type of magnon scattering is studied in detail for ferromagnets and a certain type of antiferromagnets by means of these formulas and the customary spin-wave methods.[20] An interesting new effect for polarized neutrons incident on ferromagnets is discussed. Similar effects should occur for ferrimagnets, but we do not deal with this question here. In regard to the magnetovibrational scattering in the spin-wave region, we limit our attention to what is probably the most significant type thereof as far as a number of current experiments in this region are concerned, namely, the type involving only zero-magnon processes, confining ourselves to the case of certain ferromagnets and antiferromagnets for the sake of notational simplicity.[21]

In the Appendix, we prove two results concerning the spin averages of Sec. III.

II. GENERAL FORMULAS FOR MAGNETIC SCATTERING OF NEUTRONS BY LATTICES OF COUPLED MAGNETIC IONS

Consider a lattice with ν ions per primitive chemical unit cell. To avoid unessential complications, only one of these ions will be assumed to have a nonvanishing resultant electronic magnetic moment, which will be taken to arise from spin alone, it being supposed that the orbital angular momentum of the magnetic ions is completely quenched by the crystalline electric field due to the surrounding ions. We suppose that the portion of the electronic cloud of the magnetic ions which is effective in scattering neutrons of the wavelengths considered here moves rigidly with the corresponding nuclei. The interaction between the elec-

[17] R. D. Lowde, Proc. Roy. Soc. (London) **A235**, 305 (1956); T. Riste, K. Blinowski, and J. Janik, J. Phys. Chem. Solids **9**, 153 (1959).

[18] B. N. Brockhouse, Phys. Rev. **106**, 859 (1957); **111**, 1273 (1958).

[19] The experiments on Fe_3O_4 in references 17 and 18 were performed for temperatures such that the Fe^{+2} and Fe^{+3} ions were randomly distributed in equal numbers on the B sites, a case which, as is well known, cannot be dealt with, strictly speaking, by the usual spin-wave methods. In the above references, these experiments were compared with the pertinent spin-wave results, based on these methods, by making the usual approximation of ascribing identical effective electronic spin quantum numbers to all the B sites.

[20] A useful review of current spin-wave theories has been given by J. Van Kranendonk and J. H. Van Vleck, Revs. Modern Phys. **30**, 1 (1958).

[21] In view of the work of F. J. Dyson, Phys. Rev. **102**, 1217 (1956); and **102**, 1230 (1956), one expects that the spin-wave theories used in this paper should lead to correct results for the scattering of neutrons by one-magnon emission and absorption processes. No attempt is made to calculate higher order spin-wave effects on neutron scattering in the present study.

tronic and nuclear spins is neglected, and the latter are assumed to be oriented at random. The magnetic interactions of the ions of the lattices of interest with one another and with a uniform external magnetic field are specified by the Hamiltonian H. In this section, our only assumption concerning H is that it depends on no other operators outside of the resultant electronic spin vector operators $\mathbf{S}_0,\mathbf{S}_1,\mathbf{S}_2,\cdots,\mathbf{S}_{N-1}$ of the magnetic ions. We describe the relevant nonmagnetic interactions of the ions by the usual vibrational Hamiltonian, denoted by $\mathcal{H}$, which depends quadratically on the coordinate and momentum operators of the nuclei of the lattices of interest and involves no other operators. For mathematical convenience, we suppose in this section that the scattering crystals are in the form of rectangular parallelepipeds whose edges are parallel to the crystalline axes and we adopt periodic boundary conditions, in the sense that the magnetic and nonmagnetic ions near crystalline boundaries are coupled among themselves in the familiar cyclic manner.

The magnetic and nuclear interactions of a slow neutron with a magnetic lattice of the above type are described by the operators U and V, where U corresponds to the conventional interaction between a neutron and the orbitally quenched magnetic electrons of the pertinent ions of such a lattice according to the work of Halpern and Johnson,[2] and where V represents the Fermi[22] pseudopotential.

Let the incident neutron beam be characterized by a wave-number vector $\mathbf{k}$ and a polarization f along an arbitrary unit vector $\boldsymbol{\lambda}$. Let $\boldsymbol{\lambda}'$ be a second arbitrary unit vector and $\mathbf{s}$ the neutron spin vector operator, and denote the eigenvalues and eigenkets of $(\boldsymbol{\lambda}'\cdot\mathbf{s})$ by $s_\alpha'\equiv\frac{1}{2}\alpha$ and $|s_\alpha'\rangle$, respectively, where $\alpha\equiv\pm1$. Omitting the purely nuclear scattering, we are interested in calculating the differential cross section per unit-energy range for observing the magnetic scattering of a neutron of the above beam into a direction specified by a final wave-number vector $\mathbf{k}'$ and into a spin state $|s_\alpha'\rangle$, for given α, by a single crystal of the aforementioned kind in thermodynamic equilibrium at a given T. The relevant statistical-mechanical properties of the scatterers before collision are supposed to be described by a density operator which is the product of a density operator referring to the randomly oriented nuclear spins by $\exp[-\beta H]\exp[-\beta\mathcal{H}]$, where $\beta\equiv 1/k_BT$, k_B being Boltzmann's constant. We designate by $d^2\sigma(\alpha)/d\epsilon d\Omega$ this last cross section, which corresponds to a wave-number transfer vector

$$\mathbf{q}\equiv\mathbf{k}'-\mathbf{k}, \tag{2.1}$$

and to an energy transfer

$$\epsilon\equiv E_{\mathbf{k}'}-E_{\mathbf{k}}; \tag{2.2}$$

$$E_{\mathbf{k}}\equiv(\hbar^2/2m)k^2,\quad E_{\mathbf{k}'}\equiv(\hbar^2/2m)k'^2;$$

of a neutron, where m is the neutron mass, $k\equiv|\mathbf{k}|$, and $k'\equiv|\mathbf{k}'|$. We shall use the symbol $d^2\sigma/d\epsilon d\Omega$ to denote $\sum_{\alpha=\pm1}d^2\sigma(\alpha)/d\epsilon d\Omega$ and, in general, we shall write

$$\frac{d^2\sigma_{\ldots}}{d\epsilon d\Omega}\equiv\sum_{\alpha=\pm1}\frac{d^2\sigma_{\ldots}(\alpha)}{d\epsilon d\Omega} \tag{2.3}$$

for a partial magnetic cross section of any type of interest involving a summation over the final neutron spin states.

We now treat this magnetic scattering in the first Born approximation, employing the interaction potential $U+V$. In this approximation, it is well known that no extra term in this potential is necessary to take into account the effect of a uniform external magnetic field, so long as $q\neq0$. We shall avoid the trivial complications introduced by such an external field in what follows, by the understanding that the restriction $q\neq0$ is to be imposed whenever it is present. With this proviso, we can write in the first Born approximation:

$$\frac{d^2\sigma(\alpha)}{d\epsilon d\Omega}=\frac{d^2\sigma_u(\alpha)}{d\epsilon d\Omega}+\frac{d^2\sigma_{uv}(\alpha)}{d\epsilon d\Omega}. \tag{2.4}$$

The first term in the right-hand side of (2.4) is quadratic in matrix elements of U with respect to a complete set of initial and final states which are simultaneous eigenstates of the neutron momentum vector operator, H, $\mathcal{H}$, and some convenient complete set of operators involving the nuclear spins. The second term in this right-hand side involves products of such matrix elements of U with those of V. The scattering of purely nuclear origin arising from terms quadratic in matrix elements of V of the above type is well understood and shall not be considered here.

Letting $a=u$, uv, we divide the cross sections $d^2\sigma_a(\alpha)/d\epsilon d\Omega$ in (2.4) into two parts. These parts, denoted by $d^2\sigma_{a,0}(\alpha)/d\epsilon d\Omega$ and $d^2\sigma_{a,1}(\alpha)/d\epsilon d\Omega$, contain solely matrix elements of the class just specified whose initial and final phonon occupation numbers are either identical or different, respectively. From (2.4) and this definition, we obtain:

$$\frac{d^2\sigma(\alpha)}{d\epsilon d\Omega}=\sum_{r=0,1}\frac{d^2\sigma_r(\alpha)}{d\epsilon d\Omega};$$
$$\frac{d^2\sigma_r(\alpha)}{d\epsilon d\Omega}\equiv\frac{d^2\sigma_{u,r}(\alpha)}{d\epsilon d\Omega}+\frac{d^2\sigma_{uv,r}(\alpha)}{d\epsilon d\Omega},\quad r=0,1. \tag{2.5}$$

In the spirit of a schematic definition in the Introduction, we shall regard the indices $r=0$ and $r=1$ in (2.5) as referring exclusively to purely magnetic and to magnetovibrational scattering, respectively.

[22] E. Fermi, Ricerca Sci. 7, 13 (1936).

Before continuing, we shall need to define[23]

$$
\begin{aligned}
&A(t)\equiv\exp[iHt]A\exp[-iHt];\\
&\mathcal{A}(t)\equiv\exp[i\mathcal{H}t]\mathcal{A}\exp[-i\mathcal{H}t];\\
&\langle B\rangle_\beta\equiv\frac{\operatorname{trace}\{B\exp[-\beta H]\}}{\operatorname{trace}\{\exp[-\beta H]\}};\\
&\langle\mathcal{B}\rangle_\beta\equiv\frac{\operatorname{trace}\{\mathcal{B}\exp[-\beta\mathcal{H}]\}}{\operatorname{trace}\{\exp[-\beta\mathcal{H}]\}};
\end{aligned}
\tag{2.6}
$$

where A and B are any operators which depend solely on the resultant electronic spin operators; $\mathcal{A}$ and $\mathcal{B}$ are any operators dependent only on the coordinates and momenta of the nuclei of the lattice; and the traces in the definitions of $\langle B\rangle_\beta$ and $\langle\mathcal{B}\rangle_\beta$ are to be taken over a complete set of states pertaining to the resultant electronic spins and to the vibrational modes of the lattice, respectively.

We shall also require the definition[24]

$$
\begin{aligned}
&\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\,\exp[i\epsilon t]\langle\exp[i\mathbf{q}\cdot\mathbf{u}_{i,l}(0)]\exp[-\mathbf{q}\cdot\mathbf{u}_{j,m}(t)]\rangle_\beta\\
&\qquad\equiv\delta(\epsilon')\exp[-W_l(\mathbf{q})-W_m(\mathbf{q})]+\gamma_{ij}{}^{lm}(\epsilon',\mathbf{q}),
\end{aligned}
\tag{2.7}
$$

$$l,m=0,1,2,\cdots,\nu-1;$$

where ϵ' is independent of ϵ and of $\mathbf{q}$;

$$\left\{\begin{matrix}\mathbf{u}_{i,0}\\ \mathbf{u}_{i,l}\end{matrix}\right\}(l=1,2,\cdots,\nu-1)$$

denotes the displacement vector operator of the

$$\left\{\begin{matrix}\text{magnetic}\\ l\text{th nonmagnetic}\end{matrix}\right\}$$

ion in the ith unit cell from its equilibrium position

$$\left\{\begin{matrix}\mathbf{X}_{i,0}\\ \mathbf{X}_{i,l}\end{matrix}\right\};$$

and

$$
\begin{aligned}
&W_l(\mathbf{q})\equiv\langle|\mathbf{u}_{0,l}\cdot\mathbf{q}|^2\rangle_\beta,\\
&l=0,1,2,\cdots,\nu-1.
\end{aligned}
\tag{2.8}
$$

By procedures parallel to those used in a similar connection in reference 4, we find:

$$
\begin{aligned}
\frac{d^2\sigma_{u,0}(\alpha)}{d\epsilon d\Omega}&=\frac{\Gamma}{2N}F(q)\exp[-2W_0(\mathbf{q})]\frac{k'}{k}\\
&\quad\times\sum_{i,j=0}^{N-1}\exp[i\mathbf{q}\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,0})]G_{ij}(\epsilon,\mathbf{e};\alpha);\\
\frac{d^2\sigma_{u,1}(\alpha)}{d\epsilon d\Omega}&=\frac{\Gamma}{2N}F(q)\frac{k'}{k}\sum_{i,j=0}^{N-1}\exp[i\mathbf{q}\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,0})]\\
&\quad\times\int_{-\infty}^{\infty}d\epsilon' G_{ij}(\epsilon-\epsilon',\mathbf{e};\alpha)\gamma_{ij}{}^{00}(\epsilon',\mathbf{q});\\
G_{ij}(\epsilon',\mathbf{e};\alpha)&\equiv\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\,\exp[i\epsilon' t]\\
&\quad\times\langle([\mathbf{e}\times\mathbf{S}_i(0)]\cdot[\mathbf{e}\times\mathbf{S}_j(t)])\\
&\quad-if(\mathbf{e}\cdot\boldsymbol{\lambda})(\mathbf{e}\cdot[\mathbf{S}_i(0)\times\mathbf{S}_j(t)])\\
&\quad+\alpha f\{([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\mathbf{S}_i(0)])([\mathbf{e}\times\boldsymbol{\lambda}']\\
&\quad\cdot[\mathbf{e}\times\mathbf{S}_j(t)])+([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\mathbf{S}_i(0)])\\
&\quad\times([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\mathbf{S}_j(t)])-(\boldsymbol{\lambda}\cdot\boldsymbol{\lambda}')\\
&\quad\times([\mathbf{e}\times\mathbf{S}_i(0)]\cdot[\mathbf{e}\times\mathbf{S}_j(t)])+i(\mathbf{e}\cdot\boldsymbol{\lambda}')\\
&\qquad\times(\mathbf{e}\cdot[\mathbf{S}_i(0)\times\mathbf{S}_j(t)])\}\rangle_\beta;
\end{aligned}
\tag{2.9a}
$$

where $F^{\frac{1}{2}}$ denotes the magnetic form factor of a magnetic ion of the type of interest here, F being positive and depending on $\mathbf{k}$ and $\mathbf{k}'$ only through $q\equiv|\mathbf{q}|$; $\mathbf{e}\equiv\mathbf{q}/q$; and

$$\Gamma\equiv(\gamma r_0)^2,\tag{2.10}$$

γ being the magnetic moment of a neutron in units of nuclear Bohr magnetons and r_0 the classical electron radius.

By considerations analogous to those employed in obtaining (2.9a), we conclude:

$$
\begin{aligned}
\frac{d^2\sigma_{uv,0}(\alpha)}{d\epsilon d\Omega}&=\frac{\Gamma^{\frac{1}{2}}}{N}F^{\frac{1}{2}}(q_0)\exp[-2W_0(\mathbf{q}_0)]\\
&\quad\times\operatorname{Re}\{\sum_{l=0}^{\nu-1}a_l\sum_{i,j=0}^{N-1}\exp[i\mathbf{q}_0\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,l})]\\
&\qquad\times\mathcal{E}_i(\mathbf{e}_0;\alpha)\}\delta(\epsilon);\\
\frac{d^2\sigma_{uv,1}(\alpha)}{d\epsilon d\Omega}&=\frac{\Gamma^{\frac{1}{2}}}{N}F^{\frac{1}{2}}(q)\frac{k'}{k}\operatorname{Re}\{\sum_{l=0}^{\nu-1}a_l\sum_{i,j=0}^{N-1}\\
&\quad\times\exp[i\mathbf{q}\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,l})]\\
&\qquad\times\mathcal{E}_i(\mathbf{e};\alpha)\gamma_{ij}{}^{0l}(\epsilon,\mathbf{q})\};\\
\mathcal{E}_i(\mathbf{e};\alpha)&\equiv-([\mathbf{e}\times(f\boldsymbol{\lambda}+\alpha\{\boldsymbol{\lambda}'-i[\boldsymbol{\lambda}\times\boldsymbol{\lambda}']\})]\\
&\qquad\cdot\langle[\mathbf{e}\times\mathbf{S}_i]\rangle_\beta);
\end{aligned}
\tag{2.9b}
$$

---

[23] In order to simplify our formulas, we find it convenient to use a "time" t which has the dimension of inverse energy.

[24] Explicit equations for $\gamma_{ij}{}^{lm}(\epsilon',\mathbf{q})(l,m=0,1,2,\cdots,\nu-1)$ can be obtained by a number of known methods, for example, by that of P. O. Froman, Arkiv Fysik 4, 191 (1950). However, we shall not require such explicit results in this paper.

where $\mathbf{q}_0 \equiv \mathbf{q}|_{\epsilon=0}$; $q_0 \equiv |\mathbf{q}_0|$; $\mathbf{e}_0 \equiv \mathbf{e}|_{\epsilon=0}$; a_l is the coherent scattering length of the lth ionic species ($l=0,1,2,\cdots,\nu-1$); and Re denotes the real part of the pertinent expression inside curly brackets.

III. PARAMAGNETIC SCATTERING OF NEUTRONS BY LATTICES OF COUPLED MAGNETIC IONS

The results of the present section are perhaps of greatest interest when the magnetic couplings of the magnetic ions with one another are predominantly of the exchange type. In this case, we replace H in Sec. II by

$$H_0 \equiv -\sum_{i,j=0}^{N-1} J_{ij}(\mathbf{S}_i \cdot \mathbf{S}_j); \qquad J_{ii} \equiv 0; \tag{3.1}$$

where J_{ij} is the exchange-coupling constant of the ith and jth magnetic ions, and depends solely on the neighbor relation between these ions, its possible dependence on the lattice displacements being ignored here. We also assume that each magnetic ion is coupled by exchange interactions with a finite number of neighbors. The effects of external magnetic fields and of anisotropic magnetic couplings among the relevant ions on the paramagnetic scattering of neutrons will not be considered.

In view of the fact that, in principle, the moment methods of this section are not confined to the case of exchange coupling, our discussion of the above paramagnetic scattering is of a more general scope than would be required to treat only this important special case.

To every magnetic ion i, with equilibrium position $\mathbf{X}_{i,0}$, of the scattering parallelepiped of Sec. II, we associate the magnetic ions $i(\mathbf{d})$ and $i(-)$ of the parallelepiped, with equilibrium positions $\mathbf{X}_{i,0}+\mathbf{d}+\mathbf{R}_i(\mathbf{d})$ and $-\mathbf{X}_{i,0}+\mathbf{R}_i(-)$, respectively, where $\mathbf{d}$ is a vector independent of i connecting the equilibrium positions of any two magnetic ions of the scatterer, and $\mathbf{R}_i(\mathbf{d})$ and $\mathbf{R}_i(-)$ are vectors whose components are suitable integral multiples of the sides of the above parallelepiped. It is easy to see that $\mathbf{R}_i(\mathbf{d})$ and $\mathbf{R}_i(-)$, and, therefore, that the previously prescribed positions of $i(\mathbf{d})$ and $i(-)$ are uniquely specified by i and $\mathbf{d}$. The analogues of $i(\mathbf{d})$ and $i(-)$ in the case of a crystal unbounded in all directions are given by images of i under rigid crystallographic displacements and inversions, respectively.

We shall denote the invariance of H under the substitutions $\mathbf{S}_i \to \mathbf{S}_{i(\mathbf{d})}$ for all i and $\mathbf{d}$ as property A; the invariance of H under the substitutions $\mathbf{S}_i \to \mathbf{S}_{i(-)}$ for all i as property B; and the invariance of H under arbitrary rigid rotations of all the $\mathbf{S}_i$ as property C. Imposing the periodic boundary conditions of Sec. II, one can verify that properties A, B, and C hold for $H=H_0$.

We obtain from (2.6):

$$\langle S_{i,r}(0)S_{j,s}(t)\rangle_\beta = \langle S_{i(\mathbf{d}),r}(0)S_{j(\mathbf{d}),s}(t)\rangle_\beta, \tag{3.2a}$$

if H has property A; and

$$\langle S_{i,r}(0)S_{j,s}(t)\rangle_\beta = \langle S_{i(-),r}(0)S_{j(-),s}(t)\rangle_\beta, \tag{3.2b}$$

if H has property B. In (3.2a) and (3.2b), r and s refer to the components of the pertinent spin vector operators with respect to a set of Cartesian axes.

From the fact that $\mathcal{H}$, for the case of periodic boundary conditions, is invariant under substitutions of the coordinates and momenta therein for the various lattice sites analogous to the substitutions corresponding to property A, we find from (2.6) and (2.7), under these circumstances:

$$\gamma_{ij}{}^{lm}(\epsilon',\mathbf{q}) = \gamma_{i(\mathbf{d})j(\mathbf{d})}{}^{lm}(\epsilon',\mathbf{q}), \qquad l,m=0,1,2,\cdots,\nu-1. \tag{3.2c}$$

If H has property C, we obtain:

$$\langle S_{i,r}\rangle_\beta = 0; \qquad \langle S_{i,r}(0)S_{j,s}(t)\rangle_\beta = \tfrac{1}{3}\delta_{rs}\langle(\mathbf{S}_i(0)\cdot\mathbf{S}_j(t))\rangle_\beta. \tag{3.2d}$$

From (2.9b) and (3.2d), we conclude that the assumption that H has property C implies:

$$d^2\sigma_{uv,r}(\alpha)/d\epsilon d\Omega = 0, \quad r=0,1. \tag{3.3}$$

Define

$$L_i(\epsilon) \equiv \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, \exp[i\epsilon t]\langle(\mathbf{S}_i(0)\cdot\mathbf{S}_0(t))\rangle_\beta. \tag{3.4}$$

If H has the invariance properties A and C, we conclude from Eqs. (2.5), (2.6), (2.9a), (3.2a), (3.2c), (3.2d), (3.3), and (3.4) that $d^2\sigma_r(\alpha)/d\epsilon d\Omega$ ($r=0,1$) and the corresponding cross sections $d^2\sigma_r/d\epsilon d\Omega$ involving a summation over $\alpha=\pm 1$ are given by

$$\begin{aligned}
\frac{d^2\sigma_r(\alpha)}{d\epsilon d\Omega} &= \tfrac{1}{2}[1-\alpha f(\mathbf{e}\cdot\boldsymbol{\lambda})(\mathbf{e}\cdot\boldsymbol{\lambda}')]\frac{d^2\sigma_r}{d\epsilon d\Omega}, \\
r &= 0,1; \\
\frac{d^2\sigma_0}{d\epsilon d\Omega} &= \tfrac{2}{3}\Gamma F(q)\frac{k'}{k}\exp[-2W_0(\mathbf{q})] \\
&\quad \times \sum_{i=0}^{N-1} \exp[i\mathbf{q}\cdot\mathbf{X}_{i,0}]L_i(\epsilon); \\
\frac{d^2\sigma_1}{d\epsilon d\Omega} &= \tfrac{2}{3}\Gamma F(q)\frac{k'}{k}\sum_{i=0}^{N-1}\exp[i\mathbf{q}\cdot\mathbf{X}_{i,0}] \\
&\quad \times \int_{-\infty}^{\infty} d\epsilon'\, L_i(\epsilon-\epsilon')\gamma_{i0}{}^{00}(\epsilon',\mathbf{q}).
\end{aligned} \tag{3.5}$$

At this point, we mention for the sake of clarity and to avoid unnecessary repetitions, that all cross section

and moment formulas derived in this section hold, in particular, if H has properties A, B, and C.

The magnetic scattering of neutrons in the paramagnetic domain is not essentially different for polycrystals than for single crystals. The former have received much more attention experimentally and are more convenient from the standpoint of detailed theoretical analyses than the latter in this temperature region. Nevertheless, we shall deal with both of these types of scatterers in this section, for the sake of completeness.

Because of the remarks on the case $f\neq 0$ made later in this section, we shall only consider in detail the situation when $f=0$. Equations (3.5) imply that it is then sufficient to study $d^2\sigma_r/d\epsilon d\Omega$ $(r=0,1)$, a task to which we now address ourselves.

We define the effective differential cross sections

$$\frac{d\bar{\sigma}_r}{d\Omega}\equiv\int_{-E_{\mathbf{k}}}^{\infty}d\epsilon\,\rho\frac{d^2\sigma_r}{d\epsilon d\Omega},\tag{3.6a}$$

the relative moments of neutron energy transfer

$$E_r^{(m)}\equiv\int_{-E_{\mathbf{k}}}^{\infty}d\epsilon\,\epsilon^m\rho\frac{d^2\sigma_r}{d\epsilon d\Omega},\tag{3.6b}$$

and the corresponding absolute moments

$$e_r^{(m)}\equiv E_r^{(m)}\Big/\frac{d\bar{\sigma}_r}{d\Omega},\tag{3.6c}$$

where $m=1, 2, \cdots, r=0, 1$, and ρ is a positive quantity representing the detector efficiency. To avoid uninteresting complications, ρ is taken to depend on $\mathbf{k}$ and $\mathbf{k}'$ only through k and ϵ. In the case of polycrystals, (3.6a), (3.6b), and (3.6c) are to be understood as follows: the appropriate formulas for $d^2\sigma_r/d\epsilon d\Omega$, obtained by suitably averaging the pertinent Eqs. (3.5) over all crystal orientations, should be inserted into (3.6a) and (3.6b), and the moments in (3.6c) should be constructed from the foregoing results for $d\bar{\sigma}_r/d\Omega$ and $E_r^{(m)}$. In the integrations over ϵ in (3.6a) and (3.6b), $\mathbf{k}$, $\mathbf{k}'/k'$, and ϵ are to be regarded as independent variables, so that the integrated cross sections and moments in (3.6a), (3.6b), and (3.6c) depend on $\mathbf{k}$ and $\mathbf{k}'$ only through $\mathbf{k}$ and $\mathbf{k}'/k'$, a viewpoint adopted for the sake of experimental convenience.

We now consider the case of purely magnetic scattering in the paramagnetic region.

Because $W_l(\mathbf{q})$ in (2.8) reduces to $\langle|\mathbf{u}_{0,l}|^2\rangle_\beta q^2$ in the case of crystals of cubic symmetry, the formula for $d^2\sigma_0/d\epsilon d\Omega$ in (3.5) is valid both for single crystals and polycrystals in this case if the following replacement is carried out with respect to the explicitly appearing functions $\exp[i\mathbf{q}\cdot\mathbf{X}_{i,0}]$ therein:

$$\exp[i\mathbf{q}\cdot\mathbf{X}_{i,0}]\rightarrow\psi_i(\mathbf{q});$$

$$\psi_i(\mathbf{q})\equiv\begin{Bmatrix}\exp[i\mathbf{q}\cdot\mathbf{X}_{i,0}]\\ j_0(qX_{i,0})\end{Bmatrix}\quad\text{for}\quad\begin{Bmatrix}\text{single crystals}\\ \text{polycrystals}\end{Bmatrix};\tag{3.7}$$

$$X_{i,0}\equiv|\mathbf{X}_{i,0}|;$$

where the notation $j_n(\zeta)$ denotes the usual spherical Bessel function of the specified order and argument. In this section, barring a statement to the contrary, we shall suppose in the case of purely magnetic scattering that we are dealing with polycrystals whose component elementary single crystals have cubic symmetry.

It will be convenient to make the choice of origin

$$\mathbf{X}_{0,0}=\mathbf{0}.\tag{3.8}$$

In what follows, we suppose that

$$\lim_{k\rightarrow\infty}\rho=\rho_0(\text{independent of }\epsilon)>0,\tag{3.9}$$

where this limit is taken for any prescribed ϵ. Equation (3.9) holds, in particular, for $1/v$-detectors, where we take $\rho=k/k'$.

We now relate the moments for the purely magnetic scattering in (3.6c) to a somewhat more explicit form of the corresponding moments introduced by de Gennes in the third part of reference 7. Using a notation parallel to that employed therein, where $\hbar\omega\equiv-\epsilon$ and $\boldsymbol{\kappa}\equiv-\mathbf{q}$, we define $\langle\omega^m\rangle_{\mathbf{q}}$ $(m=1,2,\cdots)$ for single crystals and polycrystals, for arbitrary T in the paramagnetic region, by means of (1.8) in this last reference, where we replace $2n$ by m and where, in the case of polycrystals, we replace the functions $p_\kappa(\omega)$, given by (1.5) of this reference, by the appropriate average of these functions over all crystal orientations. Taking for granted that F is continuous in q for $q\geqslant 0$, we combine the foregoing definitions of $\langle\omega^m\rangle_{\mathbf{q}}$ with (3.4), (3.5) modified in the sense of (3.6a), (3.6b), (3.6c), (3.7), (3.9), the elementary identity

$$\mathbf{q}_0\equiv\lim_{k\rightarrow\infty}\mathbf{q},\tag{3.10}$$

valid for any fixed $\mathbf{q}_0$ and ϵ, and a formal interchange of the appropriate integrals and limits, obtaining for any given $\mathbf{q}_0$

$$\langle\omega^m\rangle_{\mathbf{q}=\mathbf{q}_0}=(-1)^m\hbar^{-m}\lim_{k\rightarrow\infty}e_0^{(m)}.\tag{3.11}$$

This is the relation alluded to in the Introduction.

In the remainder of this section it will be understood that the limit $k\rightarrow\infty$ is to be taken for any fixed $\mathbf{q}_0$. However, it is easy to see that the subsequent formulas of this section involving this symbol hold for polycrystals of the type specified above if only q_0 is kept constant, a remark which also applies to (3.11).

The identity

$$\int_{\infty}^{\infty} d\epsilon'\,\epsilon'^n L_i(\epsilon') = (-1)^n \frac{d^n}{(idt)^n}\langle(\mathbf{S}_i(0)\cdot\mathbf{S}_0(t))\rangle_\beta|_{t=0}$$
$$= (-1)^n\langle([\mathbf{S}_i,H]_n\cdot\mathbf{S}_0)\rangle_\beta$$
$$\equiv (-1)^n x\xi_{i,n}(\beta), \quad (3.12)$$

$n=0,1,2,\cdots;$

$$[a,b]_0\equiv a,$$
$$[a,b]_1\equiv ab-ba,$$
$$[a,b]_{m+1}\equiv[[a,b]_m,b], \quad m=1,2,\cdots;$$
$$x\equiv S(S+1),$$

where S is the resultant electronic spin quantum number of a magnetic ion, is of basic importance in our treatment of the purely magnetic scattering of neutrons in the paramagnetic region. It can be proved by means of (2.6) and (3.4).

From the definition of $\xi_{i,n}(\beta)$ in (3.12) we conclude for the case when H has property C[25]

$$\sum_{i=0}^{N-1}\xi_{i,n}(\beta)=0, \quad n=1,2,\cdots. \quad (3.13a)$$

At present, there is no rigorous method for evaluating exactly the spin averages $\xi_{i,n}(\beta)$ in (3.12). However, one can express these averages as power series in β, whose coefficients can be computed, at least in principle, by expanding $\exp[-\beta H]$ therein in such a series, and by employing (2.6) and (3.12), with the following results:

$$\xi_{i,n}(\beta)=\sum_{r=0}^{\infty}\beta^r\xi_{i,nr}, \quad n=0,1,2,\cdots;$$

where

$$\xi_{i,n0}(\beta)\equiv\xi_{i,n}(0)=b_{i,n0},$$
$$\xi_{i,nr}(\beta)\equiv\frac{1}{r!}\frac{\partial^r\xi_{i,n}(\beta)}{\partial\beta^r}\bigg|_{\beta=0}=b_{i,nr}-\sum_{s=0}^{r-1}c_{r-s}\xi_{i,ns},$$
$$n=0,1,2,\cdots, \quad r=1,2,\cdots; \quad (3.14)$$

and where

$$b_{i,nr}\equiv\frac{(-1)^r}{r!}\frac{\mathrm{trace}\{([\mathbf{S}_i,H]_n\cdot\mathbf{S}_0)H^r\}}{(2S+1)^N x},$$
$$c_s\equiv\frac{(-1)^s}{s!}\frac{\mathrm{trace}\{H^s\}}{(2S+1)^N}, \quad n,r,s=0,1,2,\cdots.$$

The averages $\xi_{i,nr}$ have several general properties required in the sequel. From (3.12) and (3.14) follow the elementary results

$$\xi_{i,00}=\delta_{i0};$$
$$\xi_{0,0r}=0, \quad r=1,2,\cdots; \quad (3.13b)$$

for all H in Sec. II; while we obtain

$$\sum_{i=0}^{N-1}\xi_{i,nr}=0, \quad n=1,2,\cdots, \quad r=0,1,2,\cdots; \quad (3.13a')$$

from (3.12), (3.13a), and (3.14) if H has property C. In the Appendix, we show that[26]

$$\xi_{i,2l+1\,0}=0, \quad l=0,1,2,\cdots; \quad (3.13c)$$
$$\xi_{i,2l+1\,1}=\tfrac{1}{2}\xi_{i,2l+2\,0}, \quad l=0,1,2,\cdots; \quad (3.13d)$$

in particular, when $H=H_0$, for large enough crystals, independently of the introduction of periodic boundary conditions; and when H has properties A and B.

In the case $H=H_0$, we find from (3.1), (3.12), and (3.14)

$$\xi_{i,nr}=O(J^{n+r}), \quad n,r=0,1,2,\cdots, \quad (3.15)$$

where J denotes a typical exchange constant.

In Eqs. (3.16) below we list some results for $\xi_{i,nr}$ for $i\neq0$ and $H=H_0$ which play a central role in the explicit calculations of $d\bar{\sigma}_0/d\Omega$ and $E_0^{(m)}$ for $0\leq m\leq4$ mentioned later in this section. These results, derived by means of straightforward but lengthy trace computations based on (3.14), are valid for crystals of any shape which are sufficiently large in all directions, in virtue of the assumed short-range nature of the coupling of interest between the magnetic ions.[27]

$$\xi_{i,10}=\frac{2}{3}xJ_{i0};$$
$$\xi_{i,02}=\frac{4}{9}x^2\{\sum_j J_{ij}J_{j0}-(3/4x)J_{i0}^2\};$$
$$\xi_{i,03}=\frac{8}{27}x^3\left\{\sum_{j,k}J_{ij}J_{jk}J_{k0}-\frac{3}{2x}\sum_j J_{ij}^2J_{j0}-2J_{i0}\right.$$
$$\left.\times\left[\sum_j J_{j0}^2+\frac{3}{4x}\sum_j J_{ij}J_{j0}\right]+\frac{1}{5}\left(2-\frac{3}{x}+\frac{3}{x^2}\right)J_{i0}^3\right\};$$
$$\xi_{i,12}=-2J_{i0}\xi_{i,02};$$
$$\xi_{i,13}=\frac{1}{2}\xi_{i,22}-\frac{1}{24}\xi_{i,40};$$
$$\xi_{i,20}=-\frac{8}{3}x\sum_j J_{i0}^2;$$
$$\xi_{i,21}=-4J_{i0}\xi_{i,20};$$

[25] A result equivalent to (3.13a) for $H=H_0$ is given in the third part of reference 7, Eq. (1.7).

[26] A result equivalent to (3.13c) for the case $H=H_0$ is stated without proof in the first part of reference 7, p. 3.

[27] It is, of course, unnecessary to introduce periodic boundary conditions in deriving (3.16). We have purposely refrained from applying (3.16) to various special crystal structures and schemes of exchange coupling, both because of lack of space and because these special results follow readily from our general ones.

$$\xi_{i,22}=\frac{8}{9}x^2\Big\{\sum_j J_{ij}J_{j0}{}^3$$
$$+J_{i0}\Big[-\frac{4}{3x}\sum_{j,k}J_{ij}J_{jk}J_{k0}-\sum_j J_{j0}{}^3+\sum_j J_{ij}J_{j0}{}^2\Big]$$
$$+J_{i0}{}^2\Big[\sum_j J_{ij}J_{j0}+\frac{8}{3}x\Big(1-\frac{3}{8x}\Big)\sum_j J_{j0}{}^2\Big]$$
$$-\frac{8}{15}xJ_{i0}{}^4\Big[1-\frac{3}{2x}+\frac{9}{16x^2}\Big]\Big\};\quad(3.16)$$
$$\xi_{i,40}=\frac{32}{3}x^2\Big\{\sum_j J_{ij}{}^2J_{j0}{}^2+\frac{4}{3}J_{i0}\sum_j J_{ij}J_{j0}{}^2-\frac{2}{3}J_{i0}{}^2$$
$$\times[\sum_j J_{ij}J_{j0}+5\sum_j J_{j0}{}^2]+\frac{4}{3}\Big(1+\frac{3}{8x}\Big)J_{i0}{}^4\Big\}.$$

The lattice sums in (3.16) range over all values of j and k for which the relevant coupling constants are not zero. From now on, lattice sums involving these constants explicitly are to be understood in a parallel manner.

Employing (3.5), (3.6a), (3.6b), and (3.7), we find:

$$\frac{d\bar{\sigma}_0}{d\Omega}=\frac{2}{3}\Gamma\sum_{i=0}^{N-1}\int_{-E_{\mathbf{k}}}^{\infty}d\epsilon\,\rho(\epsilon,k)\frac{k'}{k}F(q)$$
$$\times\exp[-2W_0(\mathbf{q})]\psi_i(\mathbf{q})L_i(\epsilon);\quad(3.17)$$
$$E_0{}^{(m)}=\frac{2}{3}\Gamma\sum_{i=0}^{N-1}\int_{-E_{\mathbf{k}}}^{\infty}d\epsilon\,\epsilon^m\rho(\epsilon,k)\frac{k'}{k}F(q)$$
$$\times\exp[-2W_0(\mathbf{q})]\psi_i(\mathbf{q})L_i(\epsilon),\quad m=1,2,\cdots.$$

Some formally exact limit formulas for $d\bar{\sigma}_0/d\Omega$ and $E_0{}^{(m)}(m=1,2,\cdots)$ will be given at this point.

From (3.7), (3.8), (3.9), (3.10), (3.12), (3.13a), and (3.17), we find by arguments parallel to those used in deriving (3.11)[28]:

$$\lim_{k\to\infty}\frac{d\bar{\sigma}_0}{d\Omega}=M\sum_{i=0}^{N-1}\xi_{i,0}(\beta)\psi_i(\mathbf{q}_0);$$
$$\lim_{k\to\infty}E_0{}^{(m)}=(-1)^{m+1}M\sum_{i=0}^{N-1}\xi_{i,m}(\beta)[1-\psi_i(\mathbf{q}_0)];\quad(3.18)$$
$$M\equiv\tfrac{2}{3}\Gamma x\rho_0F(q_0)\exp[-2W_0(\mathbf{q}_0)].$$

[28] It is interesting to compare the formally exact Eqs. (3.18), (3.19a), and (3.19b) with parallel results in references 6 and the third part of reference 7. The first of Eqs. (3.18), with $\xi_{i,0}$ replaced by the corresponding series in β in (3.14), is given rather implicitly for single crystals by Eq. (11) of reference 6. Equations (1.9) and (1.10) for even moments in the third part of reference 7 correspond, respectively, to evaluating $\lim_{k\to\infty}e_0{}^{(m)}$ by means of (3.18) in the limit $\beta\to0$ and to (3.19a) for m even in this last limit; and the conclusion immediately after (1.8) in this reference on the vanishing of odd moments for $\beta\to0$ is equivalent to (3.19b), if the moments in question are defined in the way prescribed earlier in this section.

With the aid of (3.6c), (3.13c), (3.14), and (3.18), we obtain:

$$\lim_{q_0\to0}\lim_{k\to\infty}e_0{}^{(m)}=0,\quad m=1,2,\cdots;\quad(3.19a)$$
$$\lim_{\beta\to0}\lim_{k\to\infty}e_0{}^{(2l+1)}=0,\quad l=0,1,2,\cdots.\quad(3.19b)$$

It is desirable to obtain formulas for $d\bar{\sigma}_0/d\Omega$ and $E_0{}^{(m)}(m=1,2,\cdots)$ which can be used for a larger range of $E_{\mathbf{k}}$ than (3.18). To obtain such formulas, we begin by assuming that, for sufficiently large k, $d^2\sigma_0/d\epsilon d\Omega$ is significantly different from zero only if $|\epsilon|\ll E_{\mathbf{k}}$.[29] A rough way of expressing this requirement is given by the inequality

$$[e_0{}^{(2)}]^{\frac{1}{2}}\ll E_{\mathbf{k}},\quad(3.20)$$

supposed to hold for k large enough. This inequality is easy to satisfy experimentally for a number of exchange-coupled compounds. Moreover, we suppose that, for any given $\mathbf{k}$ and $\mathbf{k}'/k'$, ρF is analytic in ϵ, say for $|\epsilon|<E_{\mathbf{k}}$, which can be readily seen to imply that the functions under the integral signs in (3.17) possess this analyticity property in this range of ϵ. It is easily verified that ρF has this last property for $|\epsilon|<E_{\mathbf{k}}$, in the case of $1/v$-detectors and for F analytic in q^2 for $q\geq0$. If the above two hypotheses concerning properties of $d^2\sigma_0/d\epsilon d\Omega$ and ρF as functions of ϵ are satisfied, one expects to obtain very good approximations for $d\bar{\sigma}_0/d\Omega$ and $E_0{}^{(m)}(m=1,2,\cdots)$ for sufficiently large k by treating (3.17) in the following manner: the lower limits $-E_{\mathbf{k}}$ in the integrals over ϵ in (3.17) are changed to $-\infty$; the functions under the integrands in (3.17) are replaced by the appropriate Taylor series in ϵ about $\epsilon=0$; and term-wise integrations over ϵ are then performed by means of (3.12). In order for the above integrated series for $d\bar{\sigma}_0/d\Omega$ or $E_0{}^{(m)}(m=1,2,\cdots)$ to be rapidly convergent for given $\mathbf{k}$ and $\mathbf{k}'/k'$, such that the hypothesis expressed crudely by (3.20) holds, it is essential that, in the nontrivial case $i\neq0$, $|\psi_i(\mathbf{q})-\psi_i(\mathbf{q}_0)|$ should be sufficiently small when ϵ varies over the range for which $L_i(\epsilon)$ or $\epsilon^mL_i(\epsilon)$ is appreciably different from zero. Because of (3.10), one does not expect this requirement to be satisfied unless k is sufficiently large. We shall not consider this condition on $\psi_i(\mathbf{q})$ any further.

With the aid of the treatment of (3.17) in the last paragraph, in conjunction with (3.13a′), (3.13b), (3.13c), (3.14), and (3.18), we obtain

$$\frac{d\bar{\sigma}_0}{d\Omega}=M\{1+\sum_{r=1}^{\infty}\sum_{i=1}^{N-1}\beta^r\xi_{i,0r}\psi_{i,0}$$
$$+\sum_{n=1}^{\infty}\sum_{r=1}^{\infty}\sum_{i=1}^{N-1}\beta^rE_{\mathbf{k}}{}^{-n}\xi_{i,nr}[\psi_{i,n}-\psi_{i,0}]\};$$

[29] A parallel hypothesis is used extensively in reference 6.

$$E_0^{(m)}=(-1)^m M\sum_{n=0}^{\infty}\sum_{r=0}^{\infty}\sum_{i=1}^{N-1}\beta^r E_{\mathbf{k}}^{-n}\xi_{i,\,n+m\,r}\qquad(3.21)$$
$$[\psi_{i,n}-\psi_{0,n}],\quad m=1,2,\cdots;$$
$$\psi_{i,0}\equiv\psi_i(\mathbf{q}_0),$$
$$\psi_{i,n}\equiv[(-1)^n/n!]\{\rho_0F(q_0)\exp[-2W_0(\mathbf{q}_0)]\}^{-1}\times(\partial^n/\partial y^n)\{\rho(\epsilon,k)y^{\frac{1}{2}}F(q)\times\exp[-2W_0(\mathbf{q})]\psi_i(\mathbf{q})\}|_{y=1},\quad n=1,2,\cdots;$$

where $y\equiv(k'/k)^2$; and where we carry out the indicated differentiations regarding $\mathbf{k}/k$, $\mathbf{k}'/k'$, and y as independent variables.[30]

From numerical calculations for typical exchange-coupled lattices, including the case of polycrystalline MnF_2 discussed in this section, one expects for such typical situations that the series in (3.21) for $d\bar{\sigma}_0/d\Omega$ and $E_0^{(m)}$ should converge well for $E_{\mathbf{k}}$ in the thermal and epithermal domains and for T sufficiently larger than T_c or, more precisely, than $|\Theta|$, where Θ is the high-temperature Curie-Weiss constant, given by $\frac{2}{3}k_B^{-1}x\sum_i J_{i0}$.[31]

For $H=H_0$, using (3.13c), (3.13d), and (3.16), one can write the following terms in the series in (3.21) as explicit lattice sums, involving the appropriate exchange constants and the functions $\psi_{i,n}$ in (3.21): (a) for $d\bar{\sigma}_0/d\Omega$, those with $0\leq r\leq 3$ if $n=0$, and those with $1\leq n+r\leq 4$ if $n>0$; (b) for $E_0^{(m)}$ those with $1\leq n+r+m\leq 4$.[32] Computations based on these results should permit one to analyze a wide range of paramagnetic scattering data for exchange-coupled compounds and thus to extend in a substantial way our present restricted knowledge of the coupling constants of these compounds.

For the important case of polycrystalline scatterers and $1/v$-detectors, the following rather explicit formulas, obtained by means of elementary differential identities, can be given for the functions $\psi_{i,n}$ with $n>0$:

$$\psi_{i,n}=\sum_{l=1}^{n}C_{nl}\phi_{i,l},\quad n=1,2,\cdots;$$

$$C_{nl}\equiv(-1)^n\sum_{m=0}^{\min\{n-l,\,l\}}[(n-l-m)!(l-m)!m!]^{-1}\times\frac{\partial^{n-l-m}}{\partial y^{n-l-m}}[(y^{\frac{1}{2}}+1)^{-(l+m)}]|_{y=1}q_0^{2(l-m)}k^{2m},$$
$$l=1,2,\cdots,n,\quad n=1,2,\cdots;\qquad(3.22)$$
$$\phi_{i,l}\equiv\{F(q_0)\exp[-2W_0(\mathbf{q}_0)]\}^{-1}\times\sum_{m=0}^{l}(-1)^m2^{-m}l![(l-m)!m!]^{-1}(X_{i,0}/q_0)^m\times j_m(q_0X_{i,0})\frac{d^{l-m}}{d(q_0^2)^{l-m}}\{F(q_0)\exp[-2W_0(\mathbf{q}_0)]\},\quad l=1,2,\cdots;$$

where, for real x and y,

$$\min\{x,y\}\equiv\begin{Bmatrix}x\\y\end{Bmatrix}\quad\text{for}\quad\begin{Bmatrix}x\leq y\\x>y\end{Bmatrix}.$$

We shall give a rough and provisional treatment of the magnetovibrational scattering for T large enough. Because of the occurrence of the functions $\gamma_{i0}^{00}(\epsilon',\mathbf{q})$ in (3.5), an accurate treatment of this scattering would entail considerable difficulties.

As a working hypothesis, we suppose that, for a given i, $\gamma_{i0}^{00}(\epsilon',\mathbf{q})$ is a much more slowly-varying function of ϵ' than is $L_i(\epsilon')$ near $\epsilon'=0$. An examination of the detailed structure of $\gamma_{i0}^{00}(\epsilon',\mathbf{q})$ and of the moments of $L_i(\epsilon')$ with regard to ϵ' for $H=H_0$ provides an indication, if not a proof, that these functions of ϵ' are negligible if ϵ' lies outside of the significant range of energy transfer of the scattered neutron spectrum corresponding, respectively, to the superposition of inelastic phonon processes of all orders and to the exchange-coupling between the pertinent ions for $T>T_c$. Our assumption is therefore reasonable in this special case only if the significant range of energy transfer of the first of these spectra is much larger than that of the second, which is plausible for typical exchange-coupled lattices.[33] In virtue of our hypothesis

[30] The terms of the series for $d\bar{\sigma}_0/d\Omega$ and $E_0^{(2l)}$ in (3.21) involving the lowest positive power of $1/k$ in the limit $q_0\to 0$ can be shown to be nonnegative in this limit if H has properties A, B, and C, if ρF is analytic in the sense mentioned previously, and if ρ satisfies certain requirements which are obeyed, in particular, for the case of $1/v$-detectors. In virtue of (3.19a), this nonnegative property constitutes a check on the correctness of the series for $E_0^{(2l)}$ in (3.21) for $q_0\to 0$.

[31] It is well known that, for $\langle\mathbf{S}_i\rangle_\beta=\mathbf{0}$, the portion $\lim_{k\to\infty}d\bar{\sigma}_0/d\Omega$ of $d\bar{\sigma}_0/d\Omega$ independent of k satisfies

$$\lim_{q_0\to 0}\lim_{k\to\infty}\left[\frac{d\bar{\sigma}_0}{d\Omega}\Big/M\right]=\beta^{-1}\chi(\beta)/\lim_{\beta\to 0}[\beta^{-1}\chi(\beta)],\qquad(\mathrm{I})$$

where $\chi(\beta)$ is the zero-field susceptibility [Compare, for example, with R. J. Elliot and W. Marshall, Revs. Modern Phys. **30**, 75 (1958), Eq. (3.36)]. Because of (I) and the fact that for exchange-coupled lattices the power series in β for $\chi(\beta)$ converges rapidly only if $T\gg|\Theta|$, we see that the series in β for $\lim_{k\to\infty}d\bar{\sigma}_0/d\Omega$ in (3.21) should only converge rapidly for $q_0\to 0$ if T satisfies this inequality. The numerical calculations alluded to above indicate that the rapidity of convergence of the last mentioned series with respect to β is not strongly dependent on q_0, so that one expects that $T\gg|\Theta|$ is also a necessary condition for its convergence for $q_0>0$ (Compare with the remarks on this condition in reference 6, p. 1229). Although no considerations of this type have been found in the case of the power series in (3.21) corresponding to the portion of $d\bar{\sigma}_0/d\Omega$ involving positive powers of $(1/k)$ and to $E_0^{(m)}$, they converge well in β in these numerical examples for $T\gg|\Theta|$ and for $E_{\mathbf{k}}$ in the thermal and epithermal ranges.

[32] The special explicit formulas for integrated cross sections and moments in reference 6 and for moments in the third part of reference 7, alluded to in the Introduction, can be readily obtained by employing a suitable proper subset of the terms of (3.21) listed in (a) and (b).

[33] From the work of G. Placzek and L. Van Hove, Phys. Rev. **93**, 1207 (1954) [see particularly pp. 1212–1213], one expects that the significant width of the energy spectrum of neutrons scattered incoherently by one-phonon processes in typical crystals is much larger than the width of the corresponding spectrum for the purely magnetic scattering of neutrons by typical exchange-coupled lattices in the paramagnetic region. Although it is

we can replace $\gamma_{i0}^{00}(\epsilon',\mathbf{q})$, approximately, by $\gamma_{i0}^{00}(\epsilon,\mathbf{q})$ in $d^2\sigma_1/d\epsilon d\Omega$ in (3.5). For T sufficiently high, we combine this replacement with the rough approximation $\langle(\mathbf{S}_i\cdot\mathbf{S}_0)\rangle_\beta \cong x\delta_{i0}$. It can then be seen that we obtain for single crystals and for polycrystals whose component elementary single crystals have cubic symmetry:

$$\frac{d^2\sigma_1}{d\epsilon d\Omega} \cong \frac{2}{3}\Gamma F(q)x\frac{k'}{k}\gamma_{00}^{00}(\epsilon,\mathbf{q}). \tag{3.23}$$

Since $\gamma_{00}^{00}(\epsilon,\mathbf{q})$ is proportional to the total inelastic phonon scattering of purely nuclear origin from the magnetic ions, (3.23) constitutes an approximate relation between this scattering and the magneto-vibrational scattering.

We now consider briefly the case $f\neq 0$. Superficially, it is tempting in this situation to define $d\bar{\sigma}_r(\alpha)/d\Omega$, $E_r^{(m)}(\alpha)$, and $e_r^{(m)}(\alpha)$ ($r=0,1$, $m=1,2,\cdots$) by means of (3.6a), (3.6b), and (3.6c), where one replaces $d^2\sigma_r/d\epsilon d\Omega$ by $d^2\sigma_r(\alpha)/d\epsilon d\Omega$. Formulas analogous to (3.18) for the quantities $d\bar{\sigma}_0/d\Omega$ and $E_0^{(m)}$ ($m=1,2\cdots$) can be easily derived. Series for these quantities analogous to (3.21) can also be readily deduced, but, unfortunately, terms of these series become unbounded for fixed k and $q_0\rightarrow 0$. This difficulty for $q_0\rightarrow 0$ arises from the fact that, for fixed $\mathbf{k}$ and for $q_0=0$, $\mathbf{e}$ is a nonanalytic function of ϵ. Scant new significant knowledge, not derivable from moment studies of the neutron intensity for $f=0$, would appear to result from investigations of the purely magnetic scattering for $f\neq 0$ of the type just mentioned. As far as the magneto-vibrational scattering is concerned, it is clear that an approximate relation for $d^2\sigma_1(\alpha)/d\epsilon d\Omega$ for arbitrary f, analogous to (3.23), can be obtained trivially from (3.5) and (3.23).

We conclude this section by presenting results of numerical calculations for $d\bar{\sigma}_0/d\Omega$ and $e_0^{(2)}$ in the case of MnF_2, a substance for which, for values of T and $E_\mathbf{k}$ which seem to be particularly convenient experimentally, the inequalities $T\gg|\Theta|$ and (3.20) are readily satisfied, and the relevant series in (3.21) appear to converge quite well for the choice of exchange-coupling constants used in our calculations. Since different exchange-coupling schemes have been proposed for MnF_2 [34] and since nothing certain is known on this matter, we have adopted the following simple model for the coupling between the Mn^{+2} ions: any one of these ions is connected by exchange constants $J<0$ and $\delta\times J$ with the ions of this type at the 8 nearest-neighbor sites and the 2 nearest ions of this kind along the c axis, respectively. The paramagnetic susceptibility data on MnF_2 of Foner[35] can be fitted by $\Theta=-80°K$. This fact and the low-temperature transverse susceptibility data of Griffel and Stout,[36] analyzed by means of Ziman's spin-wave results,[37] are not compatible with δ much smaller than -1. In order to study the above magnetic scattering for two widely different cases, we chose $\delta=0, -1$, for which one obtains $|J|/k_B=1.71°K$, $2.29°K$, respectively, using the above value of Θ. From Erickson's[14] coherent scattering data for MnF_2, the corresponding F was fitted by $\exp[-\alpha q^2]$, with $\alpha=0.145$ A^2. It was decided to choose $T=300°K$, $600°K$, and to select the initial neutron wavelengths $\lambda\rightarrow 0$, $\lambda=1A,2A$, where $\lambda\rightarrow 0$ corresponds to taking $\lim_{k\rightarrow\infty}$ keeping q_0 fixed. Since lattice vibrations are unimportant as far as the purely magnetic scattering by MnF_2 is concerned, we put $W_0=0$. For $W_0=0$, it can be seen that there are no restrictions of crystal symmetry on the applicability of (3.21) to polycrystals, so that they apply, in particular, to polycrystalline MnF_2, described according to our model.

Curves for

$$\frac{d\bar{\sigma}_0}{d\Omega}\bigg/\frac{2}{3}\Gamma x, \quad \text{and} \quad [e_0^{(2)}/e_{0,\infty}^{(2)}]^{\frac{1}{2}},$$

respectively, are given for this last polycrystalline substance, described in the above way, for the case of $1/v$-detectors, where

$$e_{0,\infty}^{(2)} \equiv \lim_{q_0\rightarrow\infty}\lim_{\beta\rightarrow 0}\lim_{k\rightarrow\infty} e_0^{(2)}, \tag{3.24}$$

i.e., $e_{0,\infty}^{(2)}$ corresponds to incoherent scattering at sufficiently high $E_\mathbf{k}$ and T. These curves were obtained with the aid of the IBM-704 at NBS, employing (3.6c), (3.22), and our explicit calculation of the terms of (3.21) listed in (a) and (b), closely following (3.21).[38] From (3.24), one obtains for our model of MnF_2 as a trivial by-product of these explicit results:

$$e_{0,\infty}^{(2)} = (560/3)[1+\tfrac{1}{4}\delta^2]J^2. \tag{3.25$$

plausible that the range of ϵ' for which $\gamma_{i0}^{00}(\epsilon',\mathbf{q})$ is appreciably different from zero is at least of comparable magnitude to that for the incoherent scattering discussed by Placzek and Van Hove, a careful study is needed to settle this point.

[34] Compare, for example, the exchange-coupling models proposed by J. S. Smart, Phys. Rev. **86**, 968 (1952) and by T. Nakamura and H. Taketa, Prog. Theoret. Phys. **13**, 129 (1955).

[35] S. Foner, J. phys. radium **20**, 336 (1959).

[36] M. Griffel and J. W. Stout, J. Chem. Phys. **18**, 1455 (1950).

[37] J. M. Ziman, Proc. Phys. Soc. (London) **A65**, 548 (1952), Eq. (16), and reference 10, Eq. (20).

[38] We shall make some nonrigorous remarks on the expected accuracy of our final results for $d\bar{\sigma}_0/d\Omega$ and $e_0^{(2)}$, on which we have based the curves in Figs. 1 and 2, with respect to the corresponding exact values. Our results for $d\bar{\sigma}_0/d\Omega$ for $\lambda\rightarrow 0$ and $\lambda=1A$, 2A, and of $e_0^{(2)}$ for $\lambda\rightarrow 0$ are expected to exhibit deviations of at most a few percent from these exact values. Roughly, this is also believed to be the accuracy of our values for $e_0^{(2)}$ for $\lambda=1A$, 2A and q_0 large enough, say, crudely, $q_0\gtrsim 0.5$. Reasonable though larger deviations are expected for our results on $e_0^{(2)}$ for $\lambda=1A$, 2A and $q_0\lesssim 0.5$. The deviations of our values for the changes in $[e_0^{(2)}]^{\frac{1}{2}}$ between $T=300°K$ and $T=600°K$ from the corresponding exact ones are believed to be about the same as those of the results in the last sentence. A rough criterion for the trustworthiness of approximate calculations of the type carried out here for $e_0^{(2)}$ in the case $\lambda\neq 0$ is that $|e_0^{(2)}-\lim_{k\rightarrow\infty} e_0^{(2)}|/e_{0,\infty}^{(2)}\ll 1$, an inequality suggested by the structure of (3.21) and by the shape of the appropriate curves in Fig. 2. The maximum value of the ratio on the left-hand side of this inequality is roughly $\frac{1}{4}$ in our numerical work.

Some common qualitative features of our numerical work are of interest. For given q_0, δ, and T, $d\bar{\sigma}_0/d\Omega$ and $e_0^{(2)}$ are monotonically increasing functions of λ for small enough q_0. This inelastic behavior is most pronounced in the neighborhood of $q_0=0$ and is noticeable for $q_0 \lesssim 1$. It is due mainly to the pertinent terms of $O(k^{-2})$ in (3.21).[39] For given q_0 and T, and for $\lambda=2$A, substantial differences exist between the results for $\delta=0$ and $\delta=-1$.

Some more specific features of our numerical calculations may be mentioned. The curves in Fig. 1 exhibit the so-called antiferromagnetic peaks of $d\bar{\sigma}_0/d\Omega$ for $T=300$°K. The maxima of all these peaks occur at about the same q_0 as found by Erickson[14] in the case of polycrystalline MnF_2 for $\lambda=1.21$A and $T=295$°K, thus showing that they are quite insensitive with respect to our choices of δ and λ, although this insensitivity does not always hold with respect to the shape of these peaks. For fixed q_0 and λ, the variation of $[e_0^{(2)}/e_{0,\infty}^{(2)}]^{\frac{1}{2}}$ between $T=300$°K and $T=600$°K, shown in Fig. 2, is much larger for $\delta=-1$ than for $\delta=0$. The oscillatory behavior of the curves for this variation is caused by interference effects due to short-range magnetic order. In his experiments on the scattering of neutrons by polycrystalline MnF_2 and MnO for $T>T_c$, Bendt[15] analyzed his data, which we shall not describe here for the sake of brevity, by assuming a Gaussian shape for the energy spectrum of the scattered neutrons. This analysis involved the use of data corresponding to values of λ somewhat larger than those employed in our numerical work. In the case of MnF_2, his values for the rms neutron energy transfer for $T=300$°K, 610°K and $q_0 \gtrsim 0.7$ are in rough agreement with those of $[e_0^{(2)}]^{\frac{1}{2}}$ obtained from Fig. 2 and (3.25), for essentially the same temperatures used by him and for $\delta=0, -1$, and they also agree, in the same sense, with parallel results deducible from the work in references 5 and 6. Bendt arrived at a value for the variation of this rms energy transfer with T between 300°K and 610°K much larger but of the same sign than the corresponding variations derived for $[e_0^{(2)}]^{\frac{1}{2}}$ from Fig. 2 and (3.25) for $\delta=0, -1$, and found no interference effects of the type mentioned in this paragraph. Aside from possible experimental errors, which are difficult to evaluate, and the contribution of magnetovibrational scattering to these experiments, which is probably unimportant, this disagreement is hardly surprising from a theoretical point of view, principally because the relevant series in (3.21) do not converge well for reasonable values of J and δ in the range of λ employed by Bendt, but also because of the arbitrariness of the parameters used in our calculations, whose purpose is to provide a broad illustration of the theory of the purely magnetic scattering of neutrons in this section.

[39] Where inelastic effects are particularly small, only the curves corresponding to $\lambda \to 0$ are shown in Fig. 1 and Fig. 2.

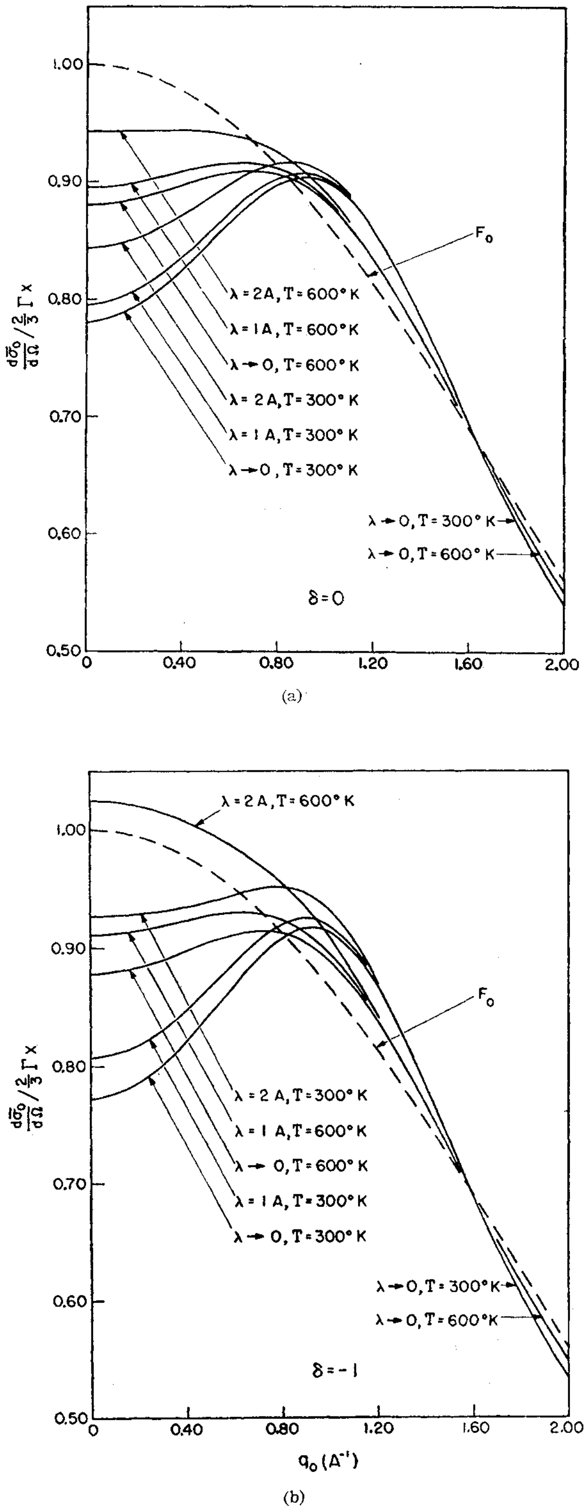

FIG. 1. Relative integrated differential cross sections $(d\bar{\sigma}_0/d\Omega)/\frac{2}{3}\Gamma x$ for neutrons incident on MnF_2, according to the coupling model in the text with $\Theta=-80$°K and for all combinations of the following values of δ, T, and λ: $\delta=0, -1$; $T=300$°K, 600°K; $\lambda \to 0$, and $\lambda=1$A, 2A. Curves (a) and (b) refer to $\delta=0$ and $\delta=-1$, respectively.

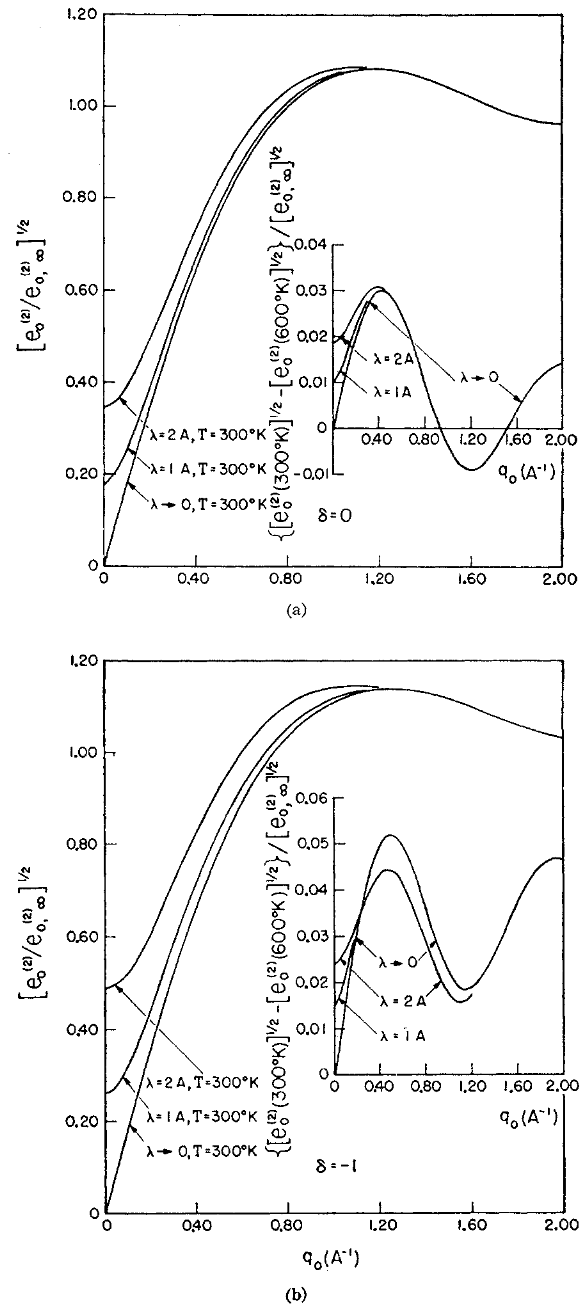

FIG. 2. Ratio of the relative rms neutron energy transfers $[e_0^{(2)}/e_{0,\infty}^{(2)}]^{\frac{1}{2}}$ for the coupling model of MnF_2 in the text with $\Theta=-80°K$ and for all combinations of the following values of δ, T, and λ: $\delta=0$, -1; $T=300°K$, $600°K$; $\lambda\rightarrow 0$, and $\lambda=1A$, $2A$. Curves (a) and (b) refer to $\delta=0$ and $\delta=-1$, respectively.

We conclude this section by remarking, in the spirit of a statement in the Introduction, that it would be of interest to obtain accurate experimental data for $d\bar{\sigma}_0/d\Omega$ and for the first few moments $e_0^{(m)}$ in the case of MnF_2 and of other exchange-coupled lattices of the class of interest here having exchange interactions of relatively simple types, for values of E_k and T within the domain of applicability of the pertinent theoretical work in this section.

IV. MAGNETIC SCATTERING OF NEUTRONS IN THE SPIN-WAVE REGION

This section is devoted to a study of the magnetic scattering of neutrons of arbitrary f by exchange-coupled lattices of the class specified in Sec. II at $T\gg T_c$, with special emphasis on ferromagnets and antiferromagnets. We shall deal with scatterers which are single magnetic domains, such that the vectors $\langle \mathbf{S}_i\rangle_\beta$ are almost parallel or antiparallel to a unique direction specified by the unit vector $\boldsymbol{\mu}$. Our results can be extended readily to situations involving any number of arbitrarily oriented domains of this type by suitable averaging operations.

Choosing a z axis parallel to $\boldsymbol{\mu}$, we adopt the following set of approximate operator equations for $T\gg T_c$, in agreement with the usual spin-wave theories[8,10,20]:

$$S_{i,x}\pm iS_{i,y}\cong(S/2)^{\frac{1}{2}}[(1\pm\sigma_i)a_i+(1\mp\sigma_i)a_i^+]\equiv S_{i,1}^{\pm};$$
$$S_{i,z}=\sigma_i[S-a_i^+a_i]; \tag{4.1}$$

where $\sigma_i=1(-1)$ if $\langle\mathbf{S}_i\rangle_\beta$ is almost parallel (antiparallel) to $\boldsymbol{\mu}$; and a_i^+ and a_i are the usual boson creation and annihilation operators, respectively. The restriction that the number operator $a_i^+a_i$ have eigenvalues ranging from 0 to $2S$ will be disregarded in this section.

A spin-wave theory of Holstein-Primakoff[8] type, applicable to a general class of exchange-coupled lattices with an arbitrary number of magnetic ions with arbitrary spin quantum numbers per primitive magnetic unit cell can be based on a slight extension of (4.1).[40]

To make the succeeding developments as clear as possible, we shall recall a general difficulty of the spin-wave theories of the above type when we put $H=H_0$ *ab initio*. In this case, $H=H_0$ can be written as a sum of a quadratic form in $p_i=(1/\sqrt{2}i)[a_i-a_i^+]$ and one in $q_i=(1/\sqrt{2})[a_i+a_i^+]$, whose corresponding matrices have at least one zero eigenvalue. This last fact implies that it is impossible to satisfy the double requirement that the transformation from the p_i and q_i to conjugate variables which diagonalize H be free from singularities for arbitrary exchange-coupled lattices and that the corresponding ground state energy of spin-wave excitation be positive, as demanded by stability considerations. A simple way of avoiding this difficulty is to introduce a new operator H_1 into H to represent schematically the effects of anisotropic couplings and of a uniform external magnetic field, as

[40] For a summary of this theory and of its application to the purely magnetic one-magnon scattering of neutrons of arbitrary f from such general lattices for the case of complete orbital quenching of the magnetic ions, see A. W. Sáenz, *Proceedings of the Fifth Symposium on Magnetism and Magnetic Materials*, J. Appl. Phys. **31** (Supplement), 108S (1960). We regret that a typographical error appears in the definition of $A_\pm(\epsilon)$ in Eqs. (5) of this paper, and that two other errors, of an unessential type, are present in this publication.

follows[41]:

$$H=H_0+H_1;$$
$$H_1\equiv -g\beta_B \sum_{i=0}^{N-1} (\mathrm{H}+\mathrm{H}_A\sigma_i)S_{i,z}; \tag{4.2}$$

where gS is the electronic magnetic moment of a magnetic ion in units of the electronic Bohr magneton β_B; and H and H_A are the respective magnitudes of an external magnetic field and of a hypothetical anisotropy field, whose directions are collinear with that of the z axis. If the matrices in the above quadratic forms of H_0 are positive semidefinite and if $\mathrm{H}_A-\mathrm{H}>0$, if all of the σ_i are positive, or $\mathrm{H}_A-\mathrm{H}>0$, if some of the σ_i are negative, then the previously stated double requirement is satisfied by $H=H_0+H_1$.

To within the approximations of the above spin-wave theories, H is invariant with respect to rigid rotations of all the spins about the z axis, in virtue of (3.1), (4.1), and (4.2). Combining this result with (2.6), where H is approximated in the sense of these theories, and with (4.1) and properties of the a_i^+ and a_i, we obtain for $T\ll T_c$, neglecting thermal averages of products of the a_i^+ and a_i of degree greater than two:

$$\begin{aligned}
&\langle([\mathbf{e}\times\mathbf{S}_i(0)]\cdot[\mathbf{e}\times\mathbf{S}_j(t)])\rangle_\beta \\
&\quad=[1-(\mathbf{e}\cdot\boldsymbol{\mu})^2](\langle\mathbf{S}_i\rangle_\beta\cdot\langle\mathbf{S}_j\rangle_\beta)+\tfrac{1}{4}[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2] \\
&\qquad\times\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t)+S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta; \\
&\langle([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\mathbf{S}_i(0)])([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\mathbf{S}_j(t)]) \\
&\qquad+([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\mathbf{S}_i(0)])([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\mathbf{S}_j(t)])\rangle_\beta \\
&\quad=2([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\mu}])([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\boldsymbol{\mu}]) \\
&\qquad\times(\langle\mathbf{S}_i\rangle_\beta\cdot\langle\mathbf{S}_j\rangle_\beta)+\tfrac{1}{2}\{([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\lambda}']) \\
&\qquad-([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\mu}])([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\boldsymbol{\mu}])\} \\
&\qquad\times\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t)+S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta; \\
&\langle(\mathbf{e}\cdot[\mathbf{S}_i(0)\times\mathbf{S}_j(t)])\rangle_\beta \\
&\quad=-(1/2i)(\mathbf{e}\cdot\boldsymbol{\mu})\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t) \\
&\qquad-S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta.
\end{aligned} \tag{4.3}$$

In order to proceed, it is essential to separate the elastic from the inelastic magnetic scattering contributions to G_{ij}. Using (2.9a) and (4.3), one obtains an expression for G_{ij} homogeneous and of the first degree in $(\langle\mathbf{S}_i\rangle_\beta\cdot\langle\mathbf{S}_j\rangle_\beta)$ and in $\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t)\pm S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta$. We denote the parts of G_{ij} containing only $(\langle\mathbf{S}_i\rangle_\beta\cdot\langle\mathbf{S}_j\rangle_\beta)$ and solely $\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t)\pm S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta$ by $G_{ij,0}$ and $G_{ij,1}$, respectively, so that

$$G_{ij}(\epsilon',\mathbf{q};\alpha)=\sum_{m=0,1} G_{ij,m}(\epsilon',\mathbf{q};\alpha); \tag{4.4}$$

where $m=0(1)$ corresponds to scattering processes in which exactly 0(1) magnons are emitted or absorbed. Employing (2.9a) and (4.3) in conjunction with these definitions of $G_{ij,0}$ and $G_{ij,1}$, and the notation

$$\begin{aligned}
&\phi_0(\mathbf{e};\alpha)\equiv[1-(\mathbf{e}.\boldsymbol{\mu})^2]+2\alpha f \\
&\qquad\times\{([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\mu}])([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\boldsymbol{\mu}]) \\
&\qquad\qquad-\tfrac{1}{2}(\boldsymbol{\lambda}\cdot\boldsymbol{\lambda}')[1-(\mathbf{e}\cdot\boldsymbol{\mu})^2]\}; \\
&\phi_1(\mathbf{e};\alpha)\equiv[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2]+2\alpha f\{([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\lambda}']) \\
&\qquad-([\mathbf{e}\times\boldsymbol{\lambda}]\cdot[\mathbf{e}\times\boldsymbol{\mu}])([\mathbf{e}\times\boldsymbol{\lambda}']\cdot[\mathbf{e}\times\boldsymbol{\mu}]) \\
&\qquad\qquad-\tfrac{1}{2}(\boldsymbol{\lambda}\cdot\boldsymbol{\lambda}')[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2]\}; \\
&\chi_0(\mathbf{e};\alpha)\equiv-([\mathbf{e}\times\boldsymbol{\mu}]\cdot[\mathbf{e}\times\{f\boldsymbol{\lambda}+\alpha\boldsymbol{\lambda}'\}]); \\
&\psi_1(\mathbf{e};\alpha)\equiv-2(\mathbf{e}\cdot\boldsymbol{\mu})(\mathbf{e}\cdot\{f\boldsymbol{\lambda}-\alpha\boldsymbol{\lambda}'\});
\end{aligned} \tag{4.5}$$

we obtain:

$$G_{ij,0}(\epsilon',\mathbf{e};\alpha)=(\langle\mathbf{S}_i\rangle_\beta\cdot\langle\mathbf{S}_j\rangle_\beta)\phi_0(\mathbf{e};\alpha)\delta(\epsilon');$$
$$G_{ij,1}(\epsilon',\mathbf{e};\alpha)=\tfrac{1}{2}[\phi_1(\mathbf{e};\alpha)M_{ij}(\epsilon')+\psi_1(\mathbf{e};\alpha)N_{ij}(\epsilon')];$$

$$\begin{aligned}
M_{ij}(\epsilon')&\equiv\frac{1}{4\pi}\int_{-\infty}^{\infty} dt\,\exp[i\epsilon' t] \\
&\qquad\times\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t)+S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta; \\
N_{ij}(\epsilon')&\equiv-\frac{1}{4\pi}\int_{-\infty}^{\infty} dt\,\exp[i\epsilon' t] \\
&\qquad\times\langle S_{i,1}{}^+(0)S_{j,1}{}^-(t)-S_{i,1}{}^-(0)S_{j,1}{}^+(t)\rangle_\beta.
\end{aligned} \tag{4.6}$$

We now carry out a formal separation of the magnetic scattering cross sections of interest into a set of partial cross sections involving simultaneous phonon and magnon processes as follows:

$$\frac{d^2\sigma_{rm}(\alpha)}{d\epsilon d\Omega}\equiv\frac{d^2\sigma_{u,rm}(\alpha)}{d\epsilon d\Omega}+\delta_{m0}\frac{d^2\sigma_{uv,r}(\alpha)}{d\epsilon d\Omega},\quad m,r=0,1; \tag{4.7}$$

where $d^2\sigma_{u,rm}(\alpha)/d\epsilon d\Omega$ is obtained from $d^2\sigma_{u,r}(\alpha)/d\epsilon d\Omega$ in (2.9a) by making the substitution

$$G_{ij}\rightarrow G_{ij,m} \tag{4.8}$$

therein. Our motivation for introducing δ_{m0} in (4.7) is the fact that $d^2\sigma_{uv,r}/d\epsilon d\Omega$ in (2.9b) involves no inelastic magnetic transitions. That for the substitution rule (4.8) is clear from the definition of $G_{ij,m}$ above.

From Eqs. (2.5), (2.9a), (2.9b), (4.4), (4.7), and (4.8), it can be verified that

$$\frac{d^2\sigma(\alpha)}{d\epsilon d\Omega}=\sum_{m=0,1}\sum_{r=0,1}\frac{d^2\sigma_{rm}(\alpha)}{d\epsilon d\Omega}, \tag{4.9}$$

In virtue of the physical significance of the indices r and m in (4.7) and (4.9), the pairs (r,m) of indices in these equations refer to scattering processes involving solely lattice transitions in which $m=0,1$ magnons and either 0 phonons, in the case $r=0$, or any nonzero number of phonons, in the case $r=1$, are emitted or absorbed. In terms of our earlier terminology, one sees that, to within the spin-wave approximation used here, the only processes contributing to the purely

[41] In principle, it would be straightforward to extend the results of this section to anisotropic interactions of a more realistic variety than those in (4.2).

magnetic and to the magnetovibrational scattering are those corresponding to $(0,m)$ and $(1,m)$ $(m=0,1)$, respectively.

Since the main features of the elastic magnetic scattering are well understood,[2] we shall not deal with this topic, except incidentally, in the discussions to follow, devoted to the inelastic magnetic scattering. Concerning the latter, we shall only treat the cases when (r,m) is equal to $(0,1)$ and $(1,0)$. In a number of experiments of current interest, carried out for $T\ll T_c$, these two cases can be expected to yield the largest contribution to the inelastic magnetic scattering.

We shall first study the case $(0,1)$.

The functions

$$\begin{aligned}\mathfrak{M}(\epsilon',\mathbf{q})&\equiv\frac{1}{N}\sum_{i,j=0}^{N-1}\exp[i\mathbf{q}\cdot(\mathrm{X}_{i,0}-\mathrm{X}_{j,0})]M_{ij}(\epsilon'),\\ \mathfrak{N}(\epsilon',\mathbf{q})&\equiv\frac{1}{N}\sum_{i,j=0}^{N-1}\exp[i\mathbf{q}\cdot(\mathrm{X}_{i,0}-\mathrm{X}_{j,0})]N_{ij}(\epsilon'),\end{aligned}\tag{4.10}$$

will play a central role in this investigation.

From (2.9a), (4.6), (4.7), (4.8), and (4.10), we get:

$$\frac{d^2\sigma_{01}(\alpha)}{d\epsilon d\Omega}=\tfrac{1}{4}\Gamma F(q)\exp[-2W_0(\mathbf{q})](k'/k)\times\{\phi_1(\mathbf{e};\alpha)\mathfrak{M}(\epsilon,\mathbf{q})+\psi_1(\mathbf{e};\alpha)\mathfrak{N}(\epsilon,\mathbf{q})\}.\tag{4.11}$$

Equations (4.11) imply, in virtue of (4.5), that the corresponding cross section obtained by summing over $\alpha=\pm1$ is given by

$$\frac{d^2\sigma_{01}}{d\epsilon d\Omega}=\tfrac{1}{2}\Gamma F(q)\exp[-2W_0(\mathbf{q})](k'/k)\times\{[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2]\mathfrak{M}(\epsilon,\mathbf{q})-2f(\mathbf{e}\cdot\boldsymbol{\lambda})(\mathbf{e}\cdot\boldsymbol{\mu})\mathfrak{N}(\epsilon,\mathbf{q})\}.\tag{4.12}$$

For complete orbital quenching of the magnetic ions and for $T\ll T_c$, one concludes from the spin-wave theory for the general class of exchange-coupled lattices alluded to earlier in this section,[40] by a straightforward extension of previous arguments, that (4.11) and, consequently, (4.12) hold in this general case, with $\mathfrak{M}(\epsilon,\mathbf{q})$ and $\mathfrak{N}(\epsilon,\mathbf{q})$ replaced by appropriate functions of ϵ and $\mathbf{q}$ which are independent of $\boldsymbol{\mu}$, $\boldsymbol{\lambda}$, and f. Earlier theoretical studies[9,11,12] for special exchange-coupled lattices of the above class have established for the case $f=0$ the fact that $d^2\sigma_{01}/d\epsilon d\Omega$ depends on $\boldsymbol{\mu}$ in the manner prescribed by (4.12). Brockhouse's experimental results in his second paper[18] on the spin-wave scattering of neutrons with $f=0$ by the (1,1,1) planes of Fe_3O_4 are in agreement with this $\boldsymbol{\mu}$ dependence. It would be desirable to extend this interesting experimental work, in the sense of testing thoroughly the dependence of the scattering of type (0,1) on $\boldsymbol{\mu}$, $\boldsymbol{\lambda}$, and f predicted by (4.11) and (4.12), modified as stated in this paragraph, for exchange-coupled lattices conforming closely to the conditions of applicability of the spin-wave analysis[40] used in deriving this dependence.[19] It should be kept in mind in an experimental investigation of this kind that the terms of $d^2\sigma_{01}/d\epsilon d\Omega$ involving f can vanish identically for certain exchange-coupled lattices, for example, for the class of antiferromagnets in this section, as can be seen from (4.14b). There are theoretical reasons[40] for believing that such vanishing does not occur for Fe_3O_4.

We proceed to evaluate $\mathfrak{M}(\epsilon,\mathbf{q})$ and $\mathfrak{N}(\epsilon,\mathbf{q})$ for ferromagnets and antiferromagnets restricted by the conditions in Sec. II. Moreover, we shall only deal with antiferromagnets which, outside of having one magnetic ion per primitive chemical unit cell, have two such ions per primitive magnetic unit cell, employing the word antiferromagnet only in this sense from now on. For this last class of substances, there exists a vector $\mathbf{w}$ with real components, such that[42]

$$\sigma_i\sigma_j=\exp[i\mathbf{w}\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,0})],\tag{4.13}$$

where $\mathbf{w}$ is independent of i and j. For simplicity, we restrict ourselves to the case when the anisotropy energy is negligible. *This restriction should be clearly understood in regard to the following considerations referring to processes of type* (0,1). In order to avoid singularities in the limit $H_1\to0$, we first calculate $\mathfrak{M}(\epsilon,\mathbf{q})$ and $\mathfrak{N}(\epsilon,\mathbf{q})$ by means of (2.6), (4.6), (4.10), (4.13), and familiar spin-wave methods,[20] with $\mathrm{H}_A+\mathrm{H}>0$ for ferromagnets and $\mathrm{H}_A-\mathrm{H}>0$ for antiferromagnets. After replacing the summations in reciprocal lattice space by integrals, we take the respective limits $\mathrm{H}_A+\mathrm{H}\to0+$ and $\mathrm{H}_A-\mathrm{H}\to0+$ in the final results for these two types of substances. We find for ferromagnets:

$$\begin{aligned}\mathfrak{M}(\epsilon,\mathbf{q})&=S\sum_{\tau}\int d\boldsymbol{\kappa}\,\{[\langle n_\kappa\rangle+1]\delta(\epsilon+\epsilon_\kappa)+\langle n_\kappa\rangle\delta(\epsilon-\epsilon_\kappa)\}\delta(\mathbf{q}-\boldsymbol{\kappa}-2\pi\boldsymbol{\tau});\\ \mathfrak{N}(\epsilon,\mathbf{q})&=-S\sum_{\tau}\int d\boldsymbol{\kappa}\,\{[\langle n_\kappa\rangle+1]\delta(\epsilon+\epsilon_\kappa)-\langle n_\kappa\rangle\delta(\epsilon-\epsilon_\kappa)\}\delta(\mathbf{q}-\boldsymbol{\kappa}-2\pi\boldsymbol{\tau});\\ \epsilon_\kappa&\equiv2S\sum_i J_{0i}[1-\cos(\boldsymbol{\kappa}\cdot\mathrm{X}_{i,0})];\\ \langle n_\kappa\rangle&\equiv\{\exp[\beta\epsilon_\kappa]-1\}^{-1};\end{aligned}\tag{4.14a}$$

where the integral over $\boldsymbol{\kappa}$ ranges over the fundamental zone of the reciprocal lattice of the lattice of magnetic ions; and $2\pi\boldsymbol{\tau}$ is an arbitrary vector of this reciprocal

[42] The representation (4.13) is used, for example, in references 9 and 10. More general representations of antiferromagnetic ordering have been derived by H. A. Gersch and W. C. Koehler, J. Phys. Chem. Solids 5, 180 (1958).

lattice. For antiferromagnets, we obtain:

$$\mathfrak{M}(\epsilon,\mathbf{q})=S\sum_{\tau}\int d\boldsymbol{\kappa}\,\{[\langle n_\kappa\rangle+1]\delta(\epsilon+\epsilon_\kappa)+\langle n_\kappa\rangle\delta(\epsilon-\epsilon_\kappa)\}\{[f_\kappa-g_\kappa]^2\delta(\mathbf{q}-\boldsymbol{\kappa}-2\pi\boldsymbol{\tau})+[f_\kappa+g_\kappa]^2\delta(\mathbf{q}-\boldsymbol{\kappa}-\mathbf{w}-2\pi\boldsymbol{\tau})\};$$

$$\mathfrak{N}(\epsilon,\mathbf{q})=0;$$

$$\begin{aligned}
\epsilon_\kappa&\equiv[A_\kappa^2-B_\kappa^2]^{\frac{1}{2}},\\
A_\kappa&\equiv 2S\sum_i J_{0i}\{\sigma_i-\tfrac{1}{2}(1+\sigma_i)\cos(\boldsymbol{\kappa}\cdot\mathbf{X}_{i,0})\},\\
B_\kappa&\equiv -S\sum_i J_{0i}(1-\sigma_i)\cos(\boldsymbol{\kappa}\cdot\mathbf{X}_{i,0}),\\
f_\kappa&\equiv 2^{-\frac{1}{2}}[A_\kappa/\epsilon_\kappa+1]^{\frac{1}{2}},\\
g_\kappa&\equiv 2^{-\frac{1}{2}}[A_\kappa/\epsilon_\kappa-1]^{\frac{1}{2}}\exp[i\gamma_\kappa],\\
\gamma_\kappa&\equiv\arg\{B_\kappa\},
\end{aligned}\tag{4.14b}$$

provided that $A_\kappa\geq|B_\kappa|$, a condition equivalent to the positive semidefiniteness of the matrices of the above quadratic forms in p_i and q_i pertaining to H_0; where the integral over $\boldsymbol{\kappa}$ extends over a fundamental zone of a sublattice with $\sigma_i=\pm1$; $2\pi\boldsymbol{\tau}$ is an arbitrary vector of the reciprocal lattice of the entire lattice of magnetic ions; and $\langle n_\kappa\rangle$ is defined by (4.14a), with ϵ_κ given by (4.14b).

From (4.11) and (4.14a), one obtains for ferromagnets:

$$\frac{d^2\sigma_{01}(\alpha)}{d\epsilon d\Omega}=\tfrac{1}{4}\Gamma F(q)\exp[-2W_0(\mathbf{q})](k'/k)\sum_{\eta=\pm1}[\phi_1(\mathbf{e};\alpha)-\eta\psi_1(\mathbf{e};\alpha)]U(-\eta\epsilon)\mathfrak{M}(\epsilon,\mathbf{q});\quad(4.15\text{a})$$

where $U(\zeta)\equiv1(0)$ for $\zeta>0(<0)$; and where it is clear that $\eta=1(-1)$ corresponds to one-magnon emission (absorption) processes.

Using (4.11) and (4.14b), one finds for antiferromagnets:

$$\frac{d^2\sigma_{01}(\alpha)}{d\epsilon d\Omega}=\tfrac{1}{4}\Gamma F(q)\exp[-2W_0(\mathbf{q})]\times(k'/k)\phi_1(\mathbf{e};\alpha)\mathfrak{M}(\epsilon,\mathbf{q}).\quad(4.15\text{b})$$

It can be seen from Eqs. (4.14a) to (4.15b) that the scattering of type (0,1) is most intense in the vicinity of the Bragg reflections specified by $\mathbf{q}_0=2\pi\boldsymbol{\tau}$ for ferromagnets, and $\mathbf{q}_0=2\pi\boldsymbol{\tau}$ and $\mathbf{q}_0=2\pi\boldsymbol{\tau}+\mathbf{w}$ for antiferromagnets, provided the average magnitude of the energy changes of the scattered neutrons is small compared with $E_{\mathbf{k}}$.[43]

[43] These conclusions are identical, as expected, with the corresponding ones for this type of scattering in the case $f=0$ in reference 9, Secs. 2.2 and 3.4.

It can be concluded, for example from (4.5), (4.15a), and (4.15b), that the inelastic magnetic cross sections of type (0,1) corresponding to a summation over $\alpha=\pm1$ are given by

$$\frac{d^2\sigma_{01}}{d\epsilon d\Omega}=\tfrac{1}{2}\Gamma F(q)\exp[-2W_0(\mathbf{q})](k'/k)\times\sum_{\eta=\pm1}[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2+2\eta f(\mathbf{e}\cdot\boldsymbol{\lambda})\times(\mathbf{e}\cdot\boldsymbol{\mu})]U(-\eta\epsilon)\mathfrak{M}(\epsilon,\mathbf{q});\quad(4.16\text{a})$$

$$\frac{d^2\sigma_{01}}{d\epsilon d\Omega}=\tfrac{1}{2}\Gamma F(q)\exp[-2W_0(\mathbf{q})](k'/k)\times[1+(\mathbf{e}\cdot\boldsymbol{\mu})^2]\mathfrak{M}(\epsilon,\mathbf{q});\quad(4.16\text{b})$$

for ferromagnets and antiferromagnets, respectively.

From (4.16a), one finds for ferromagnets that the relative contribution of magnon emission and absorption processes to $d^2\sigma_{01}/d\epsilon d\Omega$ in the case of strongly polarized neutrons can be altered markedly by varying the relative orientations of $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and $\mathbf{e}$. *This constitutes the spin-wave effect alluded to in the Introduction.* In sharp contrast to this situation, Eq. (4.16b) implies that $d^2\sigma_{01}/d\epsilon d\Omega$ is independent of f for antiferromagnets.

The above spin-wave phenomenon for ferromagnets can be exhibited experimentally very clearly in terms of the total cross section $\sigma_{01}{}^\tau$, obtained by integrating $d^2\sigma_{01}/d\epsilon d\Omega$ over the energies and the angular distribution of the neutrons scattered by processes of type (0,1) for a given $\mathbf{k}$ and $\boldsymbol{\tau}$. In our calculation of $\sigma_{01}{}^\tau$ we shall limit our attention to the case when the crystal is set sufficiently near to a Bragg position. Moreover, we shall deal solely with crystals of cubic symmetry and with the situation when only magnons of sufficiently long wavelengths are of importance in processes of type (0,1), so that we can replace ϵ_κ in (4.14a) by

$$\epsilon_\kappa\cong\frac{\hbar^2}{2m}a|\boldsymbol{\kappa}|^2,\tag{4.14a$'$}$$

where a is independent of $\boldsymbol{\kappa}$. Noting that one can replace $W_0(\mathbf{q})$ by $W_0(\mathbf{q}_0)$ in the vicinity of a Bragg reflection, and $\mathbf{e}$ by $\mathbf{e}_0$ in the neighborhood of such a reflection with $\boldsymbol{\tau}\neq\mathbf{0}$, we find in this last case by employing (4.16a), where we make these replacements, in conjunction with (4.14a), (4.14a)$'$, and elementary calculations[11]:

$$\sigma_{01}{}^\tau=\sum_{\eta=\pm1}\sigma_{01}{}^\tau(\eta);$$

$$\sigma_{01}{}^\tau(\eta)\equiv(\pi/2)S\Gamma F(q_0)\exp[-2W_0(\mathbf{q}_0)]\times[(\hbar^2/2m)a\beta kk_1]^{-1}[1+(\mathbf{e}_0\cdot\boldsymbol{\mu})^2+2\eta f(\mathbf{e}_0\cdot\boldsymbol{\lambda})(\mathbf{e}_0\cdot\boldsymbol{\mu})]\ln\{(\exp[\beta\epsilon_+(\eta)]-1)\times(\exp[\beta\epsilon_-(\eta)]-1)^{-1}\},$$

if

$$0\leqslant\frac{a+\eta}{a}\left[1-\frac{\eta k^2}{ak_1{}^2}\right]\leqslant1;\tag{4.17}$$

and $\sigma_{01}^{\tau}(\eta)\equiv 0$, otherwise;

$$\epsilon_{\pm}(\eta)\equiv\frac{\hbar^2}{2m}\left(k^2-\frac{a^2k_1^2}{(a+\eta)^2}\{1\mp\eta[1-a^{-1}(a+\eta)\times(1-\eta k^2/ak_1^2)]^{\frac{1}{2}}\}^2\right);$$

$$k_1\equiv|\mathbf{k}+2\pi\boldsymbol{\tau}|;$$

where $\eta=\pm1$ has the same physical meaning as in (4.15a). In the derivation of (4.17), the two conditions $a>1$ and $k^2/k_1^2<a$ were assumed to hold. Since $a\gg1$ for typical cases of experimental interest, the second condition is well satisfied in the vicinity of a Bragg reflection.

Define an angle of misset

$$d\theta\equiv\theta-\theta_B, \tag{4.18}$$

where θ is the glancing angle between $\mathbf{k}$ and the reflecting crystal planes specified by $\boldsymbol{\tau}$, and where θ_B is the corresponding Bragg angle. If $\boldsymbol{\tau}\neq\mathbf{0}$, if the first condition in the preceding paragraph holds, if $|d\theta|$ is small enough so that the second condition therein is satisfied and the above replacements in (4.17) are good approximations, and if $2a\sin2\theta_B\gg1$, which holds under typical experimental circumstances, one obtains from (4.17) and (4.18) the following simple rule for neutrons with arbitrary f incident on ferromagnets:

Let $|d\theta|>(2a)^{-1}\operatorname{cosec}2\theta_B$.[44] *Then the cross section* σ_{01}^{τ} *for ferromagnets is given by a factor independent of* $\boldsymbol{\mu}$, $\boldsymbol{\lambda}$, *and* f *times the factor* $[1+(\mathbf{e}_0\cdot\boldsymbol{\mu})^2\pm2f(\mathbf{e}_0\cdot\boldsymbol{\lambda})(\mathbf{e}_0\cdot\boldsymbol{\mu})]$, *where* $+(-)$ *corresponds to* $d\theta>0(<0)$.

The special case for which $|f(\mathbf{e}_0\cdot\boldsymbol{\lambda})(\mathbf{e}_0\cdot\boldsymbol{\mu})|=1$ appears to be particularly convenient experimentally, not only because of the obvious fact that the absolute value of the polarization-dependent portion of the factor in this rule is maximized, but also because the scattering of type $(r,0)(r=0,1)$ vanishes theoretically, either exactly $(r=0)$ or to a good approximation $(r=1)$, under the conditions of applicability of our rule given above. This last theoretical prediction follows from a result in the last paragraph of this section. It would be interesting to carry out such an experiment for iron, especially because of the controversial question of the applicability of the exchange Hamiltonian (2.1b) in this case.[45] A single crystal should be employed and, in the spirit of a previous suggestion in a similar connection,[46] it is desirable that this crystal should only contain iron isotopes whose coherent nuclear scattering lengths are small in comparison with their corresponding magnetic scattering lengths, so as to minimize coherent nuclear phonon scattering.

We shall now treat the magnetic scattering of type $(r,0)(r=0,1)$. Merely for the sake of obtaining simpler formulas, we shall only deal with ferromagnets which do not have any nonmagnetic ions $(\nu=1)$ and with antiferromagnets. No particular difficulties will be found in treating more complicated cases.

Employing (2.9a), (2.9b), (4.5), (4.6), (4.7), and (4.8) in conjunction with the equations

$$\langle\mathbf{S}_j\rangle_\beta=\boldsymbol{\mu}\left\{\begin{matrix}1\\ \exp[i\mathbf{w}\cdot\mathbf{X}_{j,0}]\end{matrix}\right\}\langle S_{0,z}\rangle_\beta,\quad\text{for}\ \left\{\begin{matrix}\text{ferromagnets}\\ \text{antiferromagnets}\end{matrix}\right\}; \tag{4.19}$$

which follow from (3.8), (4.13), and standard spin-wave results,[20] we obtain:

$$\frac{d^2\sigma_{00}(\alpha)}{d\epsilon d\Omega}=\frac{4\pi^3}{v_0}[b^2(q_0)\phi_0(\mathbf{e}_0;\alpha)+2a_0b(q_0)\chi_0(\mathbf{e}_0;\alpha)]\times\exp[-2W_0(\mathbf{q}_0)]\sum_\tau\delta(\mathbf{q}_0-2\pi\boldsymbol{\tau})\delta(\epsilon);$$
$$\frac{d^2\sigma_{10}(\alpha)}{d\epsilon d\Omega}=\frac{1}{2}\frac{k'}{k}[b^2(q)\phi_0(\mathbf{e};\alpha)+2a_0b(q)\chi_0(\mathbf{e};\alpha)]\times\frac{1}{N}\sum_{i,j=0}^{N-1}\exp[i\mathbf{q}\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,0})]\gamma_{ij}^{00}(\epsilon,\mathbf{q}); \tag{4.20a}$$

and

$$\frac{d^2\sigma_{00}(\alpha)}{d\epsilon d\Omega}=\frac{4\pi^3}{v_0}b^2(q_0)\phi_0(\mathbf{e}_0;\alpha)\exp[-2W_0(\mathbf{q}_0)]\times\sum_\tau\delta(\mathbf{q}_0-2\pi\boldsymbol{\tau}-\mathbf{w})\delta(\epsilon);$$
$$\frac{d^2\sigma_{10}(\alpha)}{d\epsilon d\Omega}=\frac{1}{2}\frac{k'}{k}b^2(q)\phi_0(\mathbf{e};\alpha)\frac{1}{N}\sum_{i,j=0}^{N-1}\times\exp[i(\mathbf{q}-\mathbf{w})\cdot(\mathbf{X}_{i,0}-\mathbf{X}_{j,0})]\gamma_{ij}^{00}(\epsilon,\mathbf{q}); \tag{4.20b}$$

for ferromagnets and antiferromagnets, respectively, where v_0 is the volume of a primitive chemical unit cell and

$$b(q)\equiv[\Gamma F(q)]^{\frac{1}{2}}\langle S_{0,z}\rangle_\beta. \tag{4.21}$$

If the average magnitude of the energy changes of the scattered neutrons in processes of type (1,0) is small compared with $E_{\mathbf{k}}$, it can be shown from Eqs. (4.20a) and (4.20b) that this scattering is most intense in the vicinity of the Bragg reflections specified by $\mathbf{q}_0=2\pi\boldsymbol{\tau}$ for ferromagnets and $\mathbf{q}_0=2\pi\boldsymbol{\tau}+\mathbf{w}$ for anti-

[44] As follows from (4.17) and (4.18), or from reference 9, p. 56, the restriction $|d\theta|>(2a)^{-1}\operatorname{cosec}2\theta_B$ excludes a mixture of magnon emission and absorption processes. This mixture would spoil the simplicity of the above rule. The presence of relatively weak dipole-dipole interactions among the magnetic ions makes the rule in question invalid for small enough $|d\theta|$. See reference 9, pp. 51–53, for details on the effect of dipole-dipole interactions on the purely magnetic scattering of neutrons with $f=0$ by ferromagnets at $T\ll T_c$.

[45] See, for example, reference 20, Sec. 11.

[46] Reference 17, p. 318.

ferromagnets.[47] For crystal settings which fulfill sufficiently closely one of these two Bragg conditions with $q_0 \neq 0$, we may replace $\phi_0(\mathbf{e};\ \alpha)$ and $\chi_0(\mathbf{e};\ \alpha)$ by $\phi_0(\mathbf{e}_0;\ \alpha)$ and $\chi_0(\mathbf{e}_0;\ \alpha)$, respectively. Carrying out this replacement in (4.20a) and (4.20b) for $r=1$, and using (4.5), it is seen that, for such settings and under the condition $|(\mathbf{e}_0\cdot\boldsymbol{\mu})|=1$, $d^2\sigma_{10}(\alpha)/d\epsilon d\Omega$ vanishes to a good approximation, while one easily verifies by a parallel argument the well-known fact that $d^2\sigma_{00}(\alpha)/d\epsilon d\Omega$ vanishes exactly under this condition. Under the same circumstances, these conclusions regarding the vanishing of the scattering of type $(r,0)$ can be shown to hold for the general class of magnetic lattices alluded to earlier in this section.[40] Exception made of the presence of $\mathbf{w}$ in (4.20b), one readily sees that the portions of $d^2\sigma_{r0}(\alpha)/d\epsilon d\Omega$ in (4.20a) and (4.20b) independent of $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and f are proportional to the differential cross sections per unit-energy range for the coherent, purely nuclear, scattering of neutrons corresponding to zero-phonon processes and to inelastic phonon processes of all orders, respectively, in the cases $r=0$ and $r=1$.

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APPENDIX. PROOF OF EQS. (3.13c) AND (3.13d)

We begin by proving (3.13c) under suitable restrictions on H.

According to (3.14), (3.13c) is equivalent to

$$\operatorname{trace}\{([\mathbf{S}_i,H]_{2l+1}\cdot\mathbf{S}_0)\}=0,\quad l=0,1,2,\cdots. \tag{A1}$$

Now,

$$\operatorname{trace}\{([\mathbf{S}_i,H]_n\cdot\mathbf{S}_0)\}=\operatorname{trace}\{([\mathbf{S}_0,H]_n\cdot\mathbf{S}_i)\},\quad n=0,1,2,\cdots; \tag{A2}$$

in particular, if H has properties A and B. This last result also holds for crystals sufficiently large in all directions without the introduction of periodic boundary conditions, for a wide class of Hamiltonians H, which includes $H=H_0$ as a special case, in virtue of the assumed short range of the corresponding magnetic interactions. This class, whose exact definition we shall omit, is characterized, roughly speaking, by the fact that the H therein involve couplings of a short enough range and are invariant under rigid crystallographic displacements and inversions of all the $\mathbf{S}_i$ for the case of lattices of infinite extent in all directions.

On the other hand, for any H,

$$\operatorname{trace}\{([\mathbf{S}_i,H]_n\cdot\mathbf{S}_0)\}=(-1)^n\operatorname{trace}\{([\mathbf{S}_0,H]_n\cdot\mathbf{S}_i)\},\quad n=0,1,2,\cdots. \tag{A3}$$

Equations (A1) follow immediately from (A2) and (A3).

If H has properties A and B, Eqs. (3.13c) can also be obtained with the aid of (2.6), (3.2a), and (3.2b), which imply that $\langle(\mathbf{S}_i(0)\cdot\mathbf{S}_0(t))\rangle_{\beta=0}$ is then even in t, and of (3.12) and (3.14).

We now prove (3.13d).

From (3.13c) and (3.14), we find:

$$\begin{aligned}\xi_{i,2r+1\ 1}&=-\operatorname{trace}\{([\mathbf{S}_i,H]_{2r+1}\cdot\mathbf{S}_0)H\}/(2S+1)^N x;\\ \xi_{i,2s\ 0}&=\operatorname{trace}\{([\mathbf{S}_i,H]_{2s}\cdot\mathbf{S}_0)\}/(2S+1)^N x;\\ r,s&=0,1,2,\cdots.\end{aligned} \tag{A4}$$

From (A4), (3.13d) is equivalent to

$$\operatorname{trace}\{([\mathbf{S}_i,H]_{2l+2}\cdot\mathbf{S}_0)\}=-2\operatorname{trace}\{([\mathbf{S}_i,H]_{2l+1}\cdot\mathbf{S}_0)H\},\quad l=0,1,2,\cdots. \tag{A5}$$

To prove (A5), we note that, for any H,

$$\begin{aligned}\operatorname{trace}\{([\mathbf{S}_i,H]_{2l+2}\cdot\mathbf{S}_0)\}&=-\operatorname{trace}\{([\mathbf{S}_i,H]_{2l+1}\cdot\mathbf{S}_0)H\}\\&\quad-\operatorname{trace}\{([\mathbf{S}_0,H]_{2l+1}\cdot\mathbf{S}_i)H\},\quad l=0,1,2,\cdots.\end{aligned} \tag{A6}$$

If H has properties A and B or belongs to the class of operators alluded to earlier in this Appendix, one can show that the two terms on the right-hand side of (A6) are equal and therefore that (A5) holds.

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[47] These results agree in essence, as expected, with the corresponding ones for this type of scattering in the case $f=0$ in reference 9, Sec. 4.