THE SCATTERING OF SLOW NEUTRONS IN FERRITES AND ANTIFERROMAGNETICS

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Submitted to JETP editor July 2, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) 39, 1430-1436 (November, 1960)

We use the spin-wave method to evaluate cross sections for the elastic scattering of a neutron in ferrites and antiferromagnetics and the cross sections for the scattering of a neutron accompanied by the emission or absorption of one spin wave.

RECENTLY the scattering of slow neutrons has gained ever greater importance as a method for studying solids and liquids. This refers in particular to the study of the energy spectra of different substances. One can, for instance, point to the investigations[1] to determine the spectrum of the elementary excitation in He II, to the steadily increasing number of investigations to study the phonon dispersion law in different crystals, and also to the first attempt to determine the spin wave dispersion law in magnetite.[2]

All these papers are based upon the following fact, first noted by Placzek and Van Hove,[3] that when a neutron is scattered coherently while at the same time one elementary excitation (a phonon, spin wave, and so on) is absorbed or emitted the laws of conservation of energy and momentum are of the form

$$\hbar^2 p^2 / 2m \mp \varepsilon(\mathbf{k}) = \hbar^2 p'^2 / 2m, \qquad \mathbf{p} \mp \mathbf{k} = \mathbf{p}' - \boldsymbol{\tau}, \qquad (1)$$

where $\hbar\mathbf{p}$ and $\hbar\mathbf{p}'$ are the neutron momenta before and after the scattering, $\hbar\mathbf{k}$ the quasi-momentum of the elementary excitation, $\epsilon(\mathbf{k})$ its energy, and τ a reciprocal lattice vector multiplied by 2π. Here and henceforth the upper sign refers to the scattering of a neutron accompanied by the emission of a quantum and the lower one to the scattering accompanied by the absorption of a quantum.

For given values of $\mathbf{p}$, $\mathbf{p}'$, and τ, the set of equations (1) determines the values of $\mathbf{k}$ and $\epsilon(\mathbf{k})$ uniquely. If the values of $\mathbf{p}$ and τ are given and also the direction of the scattering (the direction of $\mathbf{p}'$), then, for a given dispersion law $\epsilon(\mathbf{k})$, the magnitude of $\mathbf{p}'$ can only take on a finite number of values. At the same time one verifies easily that if more than one elementary excitation quantum takes part in the scattering, the magnitude of $\mathbf{p}'$ can take on a continuous range of values for given values of $\mathbf{p}$, τ, and the direction of the scattering. The energy distribution of neutrons scattered in a given direction has thus steep maxima corresponding to single-quantum scattering events. By determining the positions of these maxima for different scattering angles one can establish the dispersion law for the elementary excitations, using Eqs. (1).

In the present paper we use the phenomenological spin-wave theory to obtain an expression for the cross section for elastic magnetic scattering in antiferromagnetics and ferrites. These yield, in particular, the temperature dependence of the Bragg maxima and also expressions for the cross sections for scattering accompanied by the absorption or emission of one spin wave. These expressions determine also the intensities of the above-mentioned maxima of the single-quantum scattering in the cases considered.

One may also think that processes involving a large number of spin waves will play a small part in substances with a sufficiently high Curie temperature. This statement is proved in the case where the spin-wave energy[4] is $\epsilon(\mathbf{k}) \approx J(ak)^2$, where J is the exchange integral and a the lattice constant.*

It is well known[6] that the matrix element of the magnetic scattering of a neutron by a system of spin waves is of the form

$$V_{A\mathbf{p}}^{A'\mathbf{p}'} = -\frac{4\pi\hbar^2}{m} r_0 \gamma_0 (A' \Big| \sum_l e^{i\mathbf{q}\mathbf{R}_l} F_l(q) S_l^\alpha \Big| A) \{s_n^\beta - e^\alpha (s_n \mathbf{e})\}, \qquad (2)$$

where m is the neutron mass, r_0 the classical electron radius, γ_0 the neutron magnetic moment in nuclear magnetons, A the state of the system

*We use this opportunity to note that although Maleev's statement[4] that multi-magnon scattering processes play a small role is correct, the classification of these processes given by him[4,5] is not altogether correct.

before the scattering, A' the state after the scattering, $\mathbf{q} = \mathbf{p} - \mathbf{p}'$ the momentum transfer, $\mathbf{e} = (\mathbf{q}\cdot\mathbf{q})^{-1}$, $\mathbf{s}_n$ the neutron spin, and $\mathbf{S}_l$, $F_l(q)$, and $\mathbf{R}_l$ the spin, the magnetic form factor, and the coordinate of the l-th atom; the summation over l is over all the magnetic atoms of the system.

It is convenient to split the sum over l into a sum over the elementary cells of the magnetic lattice and one over the atoms inside the elementary cell.* We then get

$$\sum_l e^{i\mathbf{q}\mathbf{R}_l}\mathbf{S}_l = \sum_j e^{i\mathbf{q}\mathbf{R}_j} \sum_\nu e^{i\mathbf{q}\mathbf{r}_\nu} F_\nu(q)\mathbf{S}_{j\nu}, \qquad (3)$$

where j is the index of the elementary cell, and ν the index of the atom inside the elementary cell.

As at the present there exists no consistent microscopic spin-wave theory for antiferromagnetics or ferrites, we shall in the following use the results of a recently developed phenomenological spin-wave theory.[7,8] In this theory the basic role is played by the densities of the magnetic moments $\mathbf{M}_\nu(\mathbf{r})$ of the magnetic sublattices; these can be connected with the atomic spins through the following relation

$$\mathbf{S}_{j\nu} \to (v_0/\mu n)\,\mathbf{M}_\nu(\mathbf{R}_j), \qquad (4)$$

where n is the number of atoms in the elementary cell, v_0 the volume of the elementary cell, and μ the electronic magnetic moment. Using Eqs. (3) and (4) we can write the matrix element for the magnetic scattering of a neutron in the form

$$V_{A\mathbf{p}}^{A'\mathbf{p}'} = -\frac{4\pi\hbar^2}{m}\frac{v_0}{\mu n} r_0\gamma_0 (A'|\sum_j e^{i\mathbf{q}\mathbf{R}_j}\sum_\nu e^{i\mathbf{q}\mathbf{r}_\nu} F_\nu(q) M_\nu^\alpha(\mathbf{R}_j)\,|A)$$
$$\times\ \{s_n^\alpha - e^\alpha(\mathbf{s}_n\mathbf{e})\}. \qquad (5)$$

We first of all shall consider elastic scattering. The vectors $\mathbf{M}_\nu(\mathbf{R}_j)$ have non-vanishing diagonal matrix elements only for the components along the corresponding axes of the spontaneous magnetization. Taking the translational symmetry of the sublattice into account, we get thus

$$(A|\mathbf{M}_\nu(\mathbf{R}_j)|A) = \boldsymbol{\varepsilon}_\nu M_{0\nu}\{1 - (\mu n/M_{0\nu} v_0)\, n_\nu(A)\}, \qquad (6)$$

where $\boldsymbol{\epsilon}_\nu$ is a unit vector in the direction of the magnetization of the sublattice, $M_{0\nu}$ is the maximum density of the magnetic moment, and $(\mu n/M_{0\nu}v_0)\,n(A)$ is the relative average deviation from the maximum magnetization in the state A.

Using Eqs. (5) and (6) we can obtain the following expression for the cross section for the elastic scattering of a neutron

$$\frac{d\sigma_0}{d\Omega} = r_0^2\gamma_0^2\frac{(2\pi)^3}{v_0}\sum_\tau \delta(\mathbf{q}+\boldsymbol{\tau})\frac{1}{n}\sum_{\nu\nu'}[\boldsymbol{\varepsilon}_\nu\boldsymbol{\varepsilon}_{\nu'} - (\mathbf{e}\boldsymbol{\varepsilon}_\nu)(\mathbf{e}\boldsymbol{\varepsilon}_{\nu'})]\,S_\nu S_{\nu'}$$
$$\times F_\nu(q)F_{\nu'}^*(q)\exp\{i\mathbf{q}(\mathbf{r}_\nu - \mathbf{r}_{\nu'})\}\exp\{-W_{\nu q}$$
$$- W_{\nu' q}\}[1 - G_\nu(T) - G_{\nu'}(T)], \qquad (7)$$

where $S_\nu = v_0 M_{0\nu}/n\mu$ is the effective spin of the atoms in the sublattice, $\exp(-W_{\nu q})$ the thermal Debye-Waller factor for the atoms in the sublattice, which is introduced in the usual way, and

$$G_\nu(T) = \langle\frac{\mu n}{M_{\nu 0}v_0}n(A)\rangle_T = \frac{M_{\nu 0} - M_\nu(T)}{M_{\nu 0}} \ll 1 \qquad (8)$$

the relative deviation from the spontaneous magnetization of the sublattice from its maximum value ($T \ll \Theta_C$). When deriving Eq. (8) we have used the following well-known relation[9]

$$\frac{n}{N}\sum_{jj'}\exp\{i\mathbf{q}(\mathbf{R}_j - \mathbf{R}_{j'})\} = \frac{(2\pi)^3}{v_0}\sum_\tau \delta(\mathbf{q}+\boldsymbol{\tau}), \qquad (9)$$

where N is the number of atoms in the scatterer.

We note that since the magnetic elementary cell is larger than the nuclear elementary cell the Bragg maxima of the magnetic scattering will include some that do not coincide with maxima of nuclear scattering. This fact has already often been used to study the magnetic structure of antiferromagnetics (see, for instance, reference 10). We must also note that the temperature dependence of the Bragg maxima is determined, together with the Debye-Waller factor, by the quantities $G_\nu(T)$. The experimental determination of the temperature dependence of these quantities enables us to find the change in the magnetic moment of the sublattices with temperature. The comparison of this dependence with the theoretically evaluated one is an additional check on the theory.

For a scatterer consisting of two sublattices magnetized in the opposite direction, Eq. (7) can be simplified considerably. We have in that case for antiferromagnetics

$$\frac{d\sigma_0}{d\Omega} = r_0^2\gamma_0^2 s^2\frac{(2\pi)^3}{v_0}|F(q)|^2[1 - 2G(T)]e^{-2W_q}\sum_\tau \delta(\mathbf{q}$$
$$+\boldsymbol{\tau})(1 - e_z^2)\ 1 + \cos \mathbf{r}_{12}\boldsymbol{\tau}). \qquad (10)$$

When deriving Eq. (10) we took into account the equivalence of the magnetic lattices in an antiferromagnetic.

For ferrites we have

$$\frac{d\sigma_0}{d\Omega} = \frac{1}{2}r_0^2\gamma_0^2\frac{(2\pi)^3}{v_0}\{S_1^2|F_1(q)|^2[1 - 2G_1(T)]e^{-2W_{1q}}$$
$$+ S_2^2|F_2(q)|^2[1 - 2G_2(T)]e^{-2W_{2q}} - 2S_1S_2[1 - G_1(T)$$
$$- G_2(T)]e^{-W_{1q}-W_{2q}}\mathrm{Re}\,F_1(q)F_1^*(q)e^{i(\mathbf{q},\,\mathbf{r}_1-\mathbf{r}_2)}\}\sum_\tau \delta(\mathbf{q}$$
$$+\boldsymbol{\tau})(1 - e_z^2), \qquad (11)$$

*In the following we shall talk simply about "an elementary cell" instead of about "a magnetic elementary cell" wherever this cannot lead to any misunderstanding.

where e_z is the component of $\mathbf{e}$ in the direction of the magnetization.

According to the spin-wave theory, the function $G_\nu(T)$ has for an antiferromagnetic the form

$$G(T) = \begin{cases} \dfrac{\gamma\mu^2}{12a^3\Theta_c}\left(\dfrac{T}{\Theta_c}\right)^2, & \dfrac{\mu M_0\sqrt{2\beta\gamma}}{T} \ll 1. \\ \dfrac{\gamma\mu^2}{\pi^2 a^3\Theta_c}\left(\dfrac{T}{\Theta_c}\right)^2\left(\dfrac{2\mu M_0\sqrt{2\beta\gamma}}{T}\right)^{1/2}\exp\left\{-\dfrac{\mu M_0\sqrt{2\beta\gamma}}{T}\right\}, & \dfrac{\mu M_0\sqrt{2\beta\gamma}}{T} \gg 1. \end{cases}$$

For ferrites we have

$$G_{1,2}(T) = \frac{\mu M_{0(1,2)}\,\Gamma(3/2)\zeta(3/2)}{4\pi^2 a^3\,(M_{10}-M_{20})^2}\left(\frac{T}{\Theta_c}\right)^{3/2}.$$

Here β is the magnetic anisotropy constant, and $\gamma \sim \Theta_c/\mu M_0$ (the notation is the same as in reference 11).

We now consider the inelastic scattering of neutrons. We restrict ourselves for the sake of simplicity to the case where the scatterer consists of two sublattices magnetized in opposite directions. To evaluate the matrix elements corresponding to the scattering of a neutron accompanied by the emission or absorption of spin waves we follow Holstein and Primakoff[12] and go over from the operators of the moments $\mathbf{M}_\nu$ to the spin-wave creation operators c_k^+ and annihilation operators c_k. If we are interested in the scattering of a neutron with the emission or absorption of one spin wave, we can restrict ourselves to the first terms in the expansion of the operators of the moments in terms of the spin-wave creation and annihilation operators; this expansion is of the form[11]

$$M_{1k}^{\pm} = M_{1k}^x \pm iM_{1k}^y = \sqrt{\mu M_{10}}\begin{cases} -u_k c_{1k} + v_k c_{2,-k}^+ \\ -u_k c_{1,-k}^+ + v_k c_{2k} \end{cases}, \tag{12}$$

$$M_{2k}^{\pm} = M_{2k}^x \pm iM_{2k}^y = \sqrt{\mu M_{20}}\begin{cases} -u_k c_{2,-k}^+ + v_k c_{1,k} \\ -u_k c_{2k} + v_k c_{1,-k}^+ \end{cases}, \tag{13}$$

where c_{1k}^+ and c_{1k} are the creation and annihilation operators of spin waves with energy ϵ_{1k}, while c_{2k}^+ and c_{2k} are those for spin waves with energy ϵ_{2k}.

In the case of a ferrite, when $M_{10} \sim M_{20}$, the quantities u_k, v_k, and ϵ_k are determined by the equations[11] [for the case where $(ka)^2 \ll 1$]

$$u_k \approx \sqrt{\frac{M_{10}}{M_{10}-M_{20}}} = \sqrt{\frac{s_1}{s_1-s_2}},$$
$$v_k \approx \sqrt{\frac{M_{20}}{M_{10}-M_{20}}} = \sqrt{\frac{s_2}{s_1-s_2}}; \tag{14}$$

$$\varepsilon_{1k} \approx \mu(M_{10}-M_{20})^{-1}(\alpha_1 k^2 M_{10}^2 + \alpha_2 k^2 M_{20}^2 - 2\alpha_{12}k^2 M_{10}M_{20}) + \mu H,$$
$$\varepsilon_{2k} \approx \mu(M_{10}-M_{20})^{-1}M_{10}M_{20}(\alpha_1+\alpha_2-2\alpha_{12})k^2 + \mu\,[\gamma(M_{10}-M_{20}) - H]. \tag{15}$$

In the case of an antiferromagnetic[13]

$$u_k \approx v_k \approx \sqrt{\gamma\mu M_0/2\varepsilon_k} \gg 1; \tag{16}$$

$$\varepsilon_{1,2k} = \mu M_0\sqrt{2\gamma[\beta + (\alpha-\alpha_{12})k^2]} \mp \mu H,$$
$$\alpha_1 \sim \alpha_2 \sim \alpha_{12} \sim \Theta_c a^2(\mu M_0)^{-1}. \tag{17}$$

Using Eqs. (5), (12), and (13) one obtains easily the cross section for the scattering of a neutron accompanied by the emission or absorption of one spin wave

$$\frac{d\sigma_{\pm1,2}}{d\Omega\, dE'} = \frac{r_0^2\gamma_0^2}{4}\sqrt{S_1S_2}\int d\mathbf{k}\,\frac{p'}{p}\sum_\tau \delta(\mathbf{q}\mp\mathbf{k}+\boldsymbol{\tau})(n_{1,2}+\tfrac{1}{2}\pm\tfrac{1}{2})$$
$$\times\{u_k^2|F_1(q)|^2e^{-2W_{1q}} + v_k^2|F_2(q)|^2e^{-2W_{2q}}$$
$$-2u_kv_k e^{-W_{1q}-W_{2q}}\,\mathrm{Re}\,F_1(q)F_2^*(q)e^{i\mathbf{q}(\mathbf{r}_1-\mathbf{r}_2)}\}(1$$
$$+e_z^2)\,\delta(E-E'\mp\varepsilon_{1,2}(\mathbf{k})),$$
$$n_{1,2}(k) = [\exp\{\varepsilon_{1,2}(k)/T\}-1]^{-1}. \tag{18}$$

The integration with respect to k is over the volume of the elementary cell that contains the origin. Consequently, the only terms that do not vanish, in the sum over τ, which occurs in Eq. (18), are those for which the vector $\mathbf{q}+\tau$ lies in the elementary cell of the reciprocal lattice centered about the origin. Then, if $a|\mathbf{q}+\tau| \ll 1$, it is convenient to write Eq. (18) in the form

$$\frac{d\sigma^\tau_{\pm1,2}}{d\Omega\, dE'} \approx \frac{1}{4}Sr_0^2\gamma_0^2\frac{p'}{p}\frac{\gamma\mu M_0}{\varepsilon(\mathbf{q}+\boldsymbol{\tau})}\Big[n_{1,2}(\mathbf{q}+\boldsymbol{\tau})$$
$$+\tfrac{1}{2}\pm\tfrac{1}{2}\Big]|F(\boldsymbol{\tau})|^2 e^{-2W_\tau}$$
$$\times(1-\cos\mathbf{r}_{12}\mathbf{q})(1+\tau_z^2)\,\delta[E-E'\mp\varepsilon_{1,2}(\mathbf{q}+\boldsymbol{\tau})] \tag{19}$$

in the antiferromagnetic case* and

$$\frac{d\sigma^\tau_{\pm1,2}}{d\Omega dE'} \approx r_0^2\gamma_0^2\frac{\sqrt{S_1S_2}}{4(S_1-S_2)}\frac{p'}{p}\Big[n_{1,2}(\mathbf{q}+\boldsymbol{\tau})$$
$$+\tfrac{1}{2}\pm\tfrac{1}{2}\Big]\{S_1|F_1(\tau)|^2e^{-2W_{1\tau}} + S_2|F_2(\tau)|^2e^{-2W_{2\tau}}$$
$$-2\sqrt{S_1S_2}\,\mathrm{Re}\,F_1(\tau)F_2^*(\tau)e^{i(\mathbf{r}_1-\mathbf{r}_2,\,\tau)}e^{-W_{1\tau}-W_{2\tau}}\}$$
$$\times(1+\tau_z^2)\,\delta[E-E'\mp\varepsilon_{1,2}(\mathbf{q}+\boldsymbol{\tau})] \tag{20}$$

in the ferrite case.

*Elliott and Lowde were the first[14] to obtain this equation from a microscopic spin-wave theory for antiferromagnetics.

One can see from these formulae that the cross section for the scattering of a neutron accompanied by the absorption or emission of a spin wave is proportional to the first power of the temperature in the temperature range $\Theta_C \gg T \gg \epsilon(\mathbf{q}+\tau)$, and the scattering in ferrites occurs mainly only when spin waves with energy ϵ_1 take part, since the number of spin waves with energy ϵ_2 is exponentially small $[\sim \exp(-\Theta_C/T)]$. If the temperature is such that $T \ll \epsilon(\mathbf{q}+\tau)$ the cross section for the scattering accompanied by the absorption of one spin wave is proportional to $\exp\{-\epsilon(\mathbf{q}+\tau)/T\}$ and the cross section for the scattering accompanied by the emission of one spin wave tends to a constant limit which is independent of the temperature.

Let us consider in more detail the scattering cross section in antiferromagnetics. From the expression for the spin wave energy in antiferromagnetics when there is no magnetic field it follows that when

$$|\mathbf{q}+\boldsymbol{\tau}|^2 \ll \beta/(\alpha-\alpha_{12}) \sim \beta\mu M_0/\Theta_c a^2 \ll a^{-2} \qquad (21)$$

the energy of the scattered neutrons is independent of the direction and is given by the expression

$$E' = E \mp \mu M_0 \sqrt{2\beta\gamma}. \qquad (22)$$

The quantity $\mu M_0\sqrt{2\beta\gamma}$ is of the order of several degrees for several antiferromagnetics (MnF_2 and others), and for neutrons with energies of the order of several hundreds of degrees the change in energy is thus several percent. One verifies easily that this lack of dependence of the energy on the scattering angle occurs near those angles where the Bragg condition $|\mathbf{p}+\tau| = p$ is satisfied and for which elastic scattering is possible.

It is well known that if the antiferromagnetic is placed in a constant external magnetic field, the structure of the ground state changes,[8,15] in fields $H = \sqrt{2\beta\gamma}(1+\beta/4\gamma)$. If the magnetic moments are oriented along a chosen axis of the antiferromagnetic, in the absence of a field, then a field $H \gtrsim \sqrt{2\beta\gamma}(1+\beta/4\gamma)$ causes the magnetic moments of the sublattices to be oriented almost perpendicular to the chosen axis. Together with the change in the ground state, the character of the spin-wave dispersion law is also changed.[8,16] Magnetic and thermal measurements[17] on $CuCl_2 \cdot 2H_2O$ indicate that such a transition indeed takes place. It would be of interest to observe by neutron diffraction the change in the structure of the ground state and, in particular, the change in the spin-wave dispersion law.

We must note that it is experimentally simplest to observe inelastic magnetic scattering in substances with a high Curie temperature (several hundred degrees). The reason is, first, that when inelastic scattering takes place in such substances one can more easily discover a change in the neutron energy (which, roughly speaking, is proportional to Θ_C), and second that the scatterer can be at a relatively high temperature.

Borovik-Romanov[18] has recently studied in detail the magnetic properties of antiferromagnetics with weak ferromagnetism ($MnCO_3$, $CoCO_3$, and others) comparing the experimental data with the results obtained from spin-wave theory. He noted a considerable discrepancy between the experimental and the theoretical results. In view of this it would be very important to establish the spin-wave dispersion law in those substances by studying experimentally neutron scattering accompanied by the absorption or emission of one spin wave. Neutron-scattering experiments with these substances are, however, extremely complicated, as they have a very low Curie temperature (tens of degrees).

The authors express their deep gratitude to A. I. Akhiezer for suggesting this topic and for his interest in this paper.

Note added in proof (October 12, 1960). One should note that near the Bragg maximum for which $\cos(\mathbf{r}_{12}\cdot\boldsymbol{\tau}) = 1$ the cross section for the inelastic scattering in antiferromagnetics [which is given by Eq. (19)] becomes very small. It is then no longer possible to assume that $u_k = v_k$ and we must use Eq. (18) to evaluate the cross section, substituting into it the exact expressions for v_k and u_k given in reference 11.

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Translated by D. ter Haar
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