SOVIET PHYSICS JETP VOLUME 24, NUMBER 5 MAY, 1967

INCOHERENT SCATTERING OF NEUTRONS AND THE PROBLEM OF RECONSTRUCTING THE MAGNON SPECTRUM

Yu. A. IZYUMOV and M. V. MEDVEDEV

Institute of Metal Physics

Submitted to JETP editor April 22, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) **51**, 1423—1429 (November, 1966)

A method is developed for determining the state density in the magnon spectrum of a ferromagnetic crystal on basis of data pertaining to inelastic scattering of neutrons. It is shown that the scattered neutron energy distribution should be measured not for a perfect ferromagnetic crystal but for a crystal containing a small concentration of nonmagnetic substitution atoms. It is found that in this case the cross section defined by the incoherent single-magnon scattering impurities can be expressed in terms of the density state in the magnon spectrum of a perfect crystal.

IT is well known that a study of inelastic nuclear scattering of neutrons in crystals makes it possible to determine the phonon dispersion law and the frequency distribution function (the density of states). The possibility of obtaining such information concerning the crystal is based on the properties of the cross section for single-phonon scattering: its coherent part contains peaks in the energy distribution of the scattered neutrons, the positions of which determine the energy of the phonon participating in the scattering, while the incoherent part is proportional to the density of the states in the phonon spectrum. Incoherent nuclear scattering in the crystal is due either to the presence of spin in the nuclei, or to the isotopic composition of the elements, therefore even a crystal which is ideal from the crystal-chemical point of view leads to incoherent scattering of neutrons, thus affording a possibility of determining the density of states in the phonon spectrum.

In analogy with phonon scattering, coherent single-magnon scattering of neutrons in ferromagnets makes it possible to determine the magnon dispersion law, but an ideal ferromagnetic crystal does not produce incoherent magnetic scattering that can be related to the density of states in the magnon spectrum. To obtain such a possibility, the magnetic crystal must be made irregular, for example by introducing in it a certain concentration of nonmagnetic impurity substitutional atoms. Then, however, the structure of the excitation spin-wave spectrum may change, but in the case of low impurity concentrations the problem may be solved relatively easily.

The introduction of non-magnetic atoms is convenient because it does not introduce the impurity—host exchange parameter J′, which is difficult to determine, so that the cross section of the incoherent single-magnon scattering can be most easily connected with the density of states for an ideal ferromagnetic crystal.

The doubly-differential cross section of the magnetic scattering of neutrons in a crystal can be expressed in terms of space-time correlations of the spin projections of the atoms by means of the well known formula[1]

$$\frac{d^2\sigma}{d\Omega\, dE'} = (r_0\eta)^2 \frac{p'}{p} \sum_{nm} F_n(\mathbf{q}) F_m(\mathbf{q}) e^{-i\mathbf{q}(\mathbf{n}-\mathbf{m})} \sum_{\alpha\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta)$$

$$\times \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\Delta E t} \langle S_n{}^\alpha(0) S_m{}^\beta(t) \rangle\, dt, \quad (1)$$

where E, p and E′, p′ are the energy and momentum of the incident and scattered neutrons, q = p − p′ is the scattering vector, $\Delta E = |E - E'|$ is the transition energy, e is the unit scattering vector, S_n^α is the α projection of the spin vector of the atom at sight n, F(q) is the magnetic form factor of the atom, r_0 is the classical electromagnetic radius of the electron, and $\eta = -1.913$ is the magnetic moment of the neutron.

The part of the cross section (1) which is responsible for the single-magnon scattering can be readily obtained for a ferromagnetic crystal by expanding the spin operators in powers of the Bose operators a_n and a_n^+ of creation and annihilation of spin deviations by means of the relations[2]:

$$S_n^+ = (2S_n)^{1/2}a_n, \quad S_n^- = (2S_n)^{1/2}a_n^+,$$
$$S_n^z = S_n - a_n^+a_n \tag{2}$$

(the spontaneous magnetic moment of the crystal is directed along the z axis).

If we introduce the advanced Green's function[3]

$$G_{nm}(t-t') = i\theta(t'-t)\langle[a_n(t), a_m^+(t')]\rangle, \tag{3}$$

then we can readily show that the cross section of the single-magnon scattering is expressed in terms of its Fourier component $G_{nm}(E)$ with respect to the variable $t - t'$ by means of the relation

$$\frac{d^2\sigma}{d\Omega\, dE'} = \frac{1}{2}(r_0\eta)^2\frac{p'}{p}[1+(\mathbf{em})^2]\left\{\begin{matrix} n(\Delta E) \\ n(\Delta E)+1\end{matrix}\right\}\sum_{nm}F_n(\mathbf{q})$$
$$\times F_m(\mathbf{q})\,(S_nS_m)^{1/2}\exp(-i\mathbf{q})(\mathbf{n}-\mathbf{m}))\frac{1}{\pi}\operatorname{Im}G_{nm}(\Delta E). \tag{4}$$

The upper relation is taken here for scattering with absorption of a magnon, and the lower for emission; $n(\Delta E)$ is the Bose distribution function, and **m** is a unit vector in the direction of the spontaneous moment.

For an ideal crystal consisting of one species of atoms, this term determines the cross section of the wholly coherent scattering of the neutrons. In the presence of randomly distributed atoms of the difference species, incoherent single-magnon scattering is also produced. For small concentrations of the impurity magnetic atoms, the corresponding parts of the cross sections were calculated by one of the authors in[4]. However, from the results of that paper it is impossible to obtain the cross section for the case when the impurity atom is nonmagnetic, by simply taking the limit to $S' = 0$ and $J' = 0$ in the final results, without taking account of the fact that in this case the number of states of the single-particle excitations in the system decreases compared with the case of magnetic impurity atoms. This fact makes it necessary, in addition, to assume from the very outset the identical vanishing of $G_{00} = G_{0n} = G_{n0} \equiv 0$ (for all the sites 0 occupied by the nonmagnetic impurities).

As seen from (4), calculation of the cross section in the presence of impurities in the crystal calls for calculation of the Green's function G_{nm} and subsequent averaging of expression (4) over different impurity configurations. This procedure was developed in detail by a diagram technique by one of the authors[5] who has shown that for small impurity concentrations the results can be obtained by considering expression (4) for a crystal containing a single impurity atom, making a rather simple subsequent generalization.

We have thus reduced our problem to the calculation of the Green's function of a crystal containing one non-magnetic impurity atom. Let us locate it at the site $n = 0$. Since $G_{00} = G_{0n} = G_{n0} = 0$, only $(N-1)(N-1)$ matrix elements G_{nm} will differ from zero, but is most convenient to consider the Green's function in the space of N sites of the crystal, including also the site $n = 0$. The corresponding matrix will be denoted by G, so that

$$\bar{G} = \begin{pmatrix} 0 & 0 \\ 0 & G \end{pmatrix}, \tag{5}$$

where G is a matrix of dimension $(N-1)(N-1)$ for all the magnetic sites of the excited crystal.

Using the definition of the Green's function (3), we can readily setup an equation of motion for G using the scheme previously employed by us in[6]. We thus find that G satisfies the equation

$$\bar{G} = G^0\bar{1} + G^0V\bar{G}, \tag{6}$$

where $\bar{1}$—is a matrix of dimension $N \cdot N$ in the form

$$\bar{1} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix},$$

and G^0 is the Green's function of an ideal crystal with elements

$$G_{nm}{}^0 = \frac{1}{N}\sum_{\mathbf{k}}\frac{\exp[i\mathbf{k}(\mathbf{R}_n - \mathbf{R}_m)]}{E - \varepsilon_{\mathbf{k}} - i0^+}, \tag{7}$$

where ϵ_k is the energy of the spin wave in the ideal crystal. We assume throughout that there is one magnetic atom per unit cell of the crystal, and therefore only one branch of spin waves. V is the matrix of the perturbation introduced by the impurity atom. We confine ourselves in this paper to the investigation of a cubic crystal, for which

$$V = 2JS\begin{pmatrix} -z & 1 & 1\ldots & 1 \\ 1 & -1 & 0\ldots & 0 \\ 1 & 0 & -1\ldots & 0 \\ \cdots & \cdots & \cdots & \cdots \\ 1 & 0 & 0\ldots & -1 \end{pmatrix}, \tag{8}$$

z is the number of nearest neighbors.

Solving (6) by using the cubic symmetry of the crystal, we can show that G_{nm} breaks up into a sum of contributions corresponding to irreducible representations of the point group of the crystal. For example, for a simple cubic lattice ($z = 6$) we have

$$G_{nm} = G_{nm}{}^0 + \Delta_{nm}(s) + \Delta_{nm}(p) + \Delta_{nm}(d), \tag{9}$$

where s, p, and d denote the irreducible representations of the cubic group O_h: Γ_1, Γ_{15}, and Γ_{12} (in the nomenclature of[7]). The contribution $\Delta_{nm}(s)$ turns out to be

$$\Delta_{nm}(s) = E(1 - EG_0(E))^{-1} G_{n0}{}^0(E) G_{0m}{}^0(E), \quad (10)$$

where we put for brevity

$$G_0(E) \equiv G_{00}{}^0(E) = \frac{1}{N} \sum_{\mathbf{k}} \frac{1}{E - \varepsilon_{\mathbf{k}} - i0^+}. \quad (11)$$

Expression (10) is valid for all three types of cubic lattices. As regards the contributions $\Delta_{\mathrm{nm}}(\mathrm{p})$ and $\Delta_{\mathrm{nm}}(\mathrm{d})$, we can use for a simple cubic lattice the results obtained in[4] (formulas (2.12) and (2.13), in which we must put $\rho = -1$). We shall not write out the corresponding formulas.

We now calculate the sum over n and m in (4). Since summation over n and m is only over the magnetic atoms for which the spin and form factors are the same and equal respectively to S and F(q), in the presence of non-magnetic impurity atoms the problem reduces to the calculation of the sum

$$\frac{1}{N} \sum_{n,m} e^{-i\mathbf{q}(\mathbf{n}-\mathbf{m})} \left\langle \frac{1}{\pi} \operatorname{Im} G_{nm} \right\rangle, \quad (12)$$

where the symbol $\langle \ldots \rangle$ denotes averaging over the impurity configurations.

Let us examine this expression when there is only one impurity atom in the crystal. Using (9) and (10) we can write an exact expression

$$\frac{1}{N} \sum_{n,m\neq 0} e^{-i\mathbf{q}(\mathbf{n}-\mathbf{m})} G_{nm}(E)$$

$$= \left\{ G_0(\mathbf{q}) + \frac{1}{N} G_0^2(\mathbf{q}) W(\mathbf{q}) \right\} + \frac{1}{N} \frac{G_0(E)}{1 - EG_0(E)}, \quad (13)$$

where

$$G_0(\mathbf{q}) = (E - \varepsilon_{\mathbf{q}})^{-1}, \quad (14a)$$

$$W(\mathbf{q}) = \frac{E - 2G_0^{-1}(\mathbf{q})}{1 - EG_0(E)}$$

$$+ \sum_{nm} e^{-i\mathbf{q}(\mathbf{n}-\mathbf{m})} [\Delta_{nm}(p) + \Delta_{nm}(d)] G_0^{-2}(\mathbf{q}). \quad (14b)$$

The quantity 1/N in (13) plays the role of the impurity concentration c. Accurate to terms of order c^2, the expression in the curly brackets in (13) can be written in the form

$$G_0(\mathbf{q}) + cG^2(\mathbf{q}) W(\mathbf{q}) \approx (G_0^{-1}(\mathbf{q}) - cW(\mathbf{q}))^{-1} \equiv \langle G(\mathbf{q}) \rangle, \quad (15)$$

It is clear therefore that $\langle G(\mathbf{q}) \rangle$ is the Green's function of the spin-wave excitations of the irregular crystal, averaged over the impurity configurations, while W(q) has the meaning of the self-energy part. In[5] there is a rigorous proof, with the aid of a diagram technique, of an expression for the Green's function of type (14), averaged over the impurity configurations, in the asymptotic limit of low concentrations. At the same time, a connection was indicated there between the rigorous results and the terms in the single-impurity problem. We have used this connection here, in order not to repeat all the complicated arguments of[5], to which we refer the reader for details.

Calculation of the sums over n and m in (14b) leads to the following result for the self-energy part:

$$W(\mathbf{q}) = \frac{E - 2G_0^{-1}(\mathbf{q})}{1 - EG_0(E)} + \frac{Z_p(\mathbf{q})}{D_p(E)} + \frac{Z_d(\mathbf{q})}{D_d(E)}, \quad (16)$$

where

$$Z_p(\mathbf{q}) = -2JS\{1 - {}^1\!/_3[\cos\mathbf{q}(1-2) + \cos\mathbf{q}(3-4) + \cos\mathbf{q}(5-6)]\}, \quad (17)$$

$$\begin{aligned} Z_d(\mathbf{q}) = & -2JS\{1 + {}^1\!/_3[\cos\mathbf{q}(1-2) \\ & + \cos\mathbf{q}(3-4) + \cos\mathbf{q}(5-6)] \\ & - {}^1\!/_6[\cos\mathbf{q}(1-3) + \cos\mathbf{q}(1-4) \\ & + \cos\mathbf{q}(1-5) + \cos\mathbf{q}(1-6) \\ & + \cos\mathbf{q}(2-3) + \cos\mathbf{q}(2-4) \\ & + \cos\mathbf{q}(2-5) + \cos\mathbf{q}(2-6) \\ & + \cos\mathbf{q}(5-3) + \cos\mathbf{q}(5-4) \\ & + \cos\mathbf{q}(6-3) + \cos\mathbf{q}(6-4)]\}; \end{aligned} \quad (18)$$

$$\begin{aligned} D_p(E) &= 1 + 2JS[G_0(E) - G_{12}{}^0(E)], \\ D_d(E) &= 1 + 2JS[G_0(E) + G_{12}{}^0(E) - 2G_{13}{}^0(E)]. \end{aligned} \quad (19)$$

Here the indices, 1, 2, ..., 6 number the sites from the first coordination sphere. The sites connected by inversion are numbered by the pairs 1 and 2, 3 and 4, and 5 and 6. The three terms in W(q) describe the contributions of the s, p, and d states.

Summing now the results of (13), (14), and (15), we can write for the quantity (12), pertaining to crystals containing a certain impurity concentration c, the expression

$$\frac{1}{N} \sum_{nm} e^{-i\mathbf{q}(\mathbf{n}-\mathbf{m})} \frac{1}{\pi} \langle \operatorname{Im} G_{nm}(E) \rangle = \frac{1}{\pi} \frac{\gamma_{\mathbf{q}}}{(E - E_{\mathbf{q}})^2 + \gamma_{\mathbf{q}}^2}$$

$$+ c \frac{g_0(E)}{[1 - E \operatorname{Re} G_0(E)]^2 + [\pi E g_0(E)]^2}, \quad (20)$$

$$g_0(E) = \pi^{-1} \operatorname{Im} G_0(E) \quad (21)$$

is the density of states in the ideal crystal, and E_{q} and γ_{q} are the energy and damping of the spin wave in an irregular crystal containing impurities; in first order in c we have

$$E_{\mathbf{q}} = \varepsilon_{\mathbf{q}} + c\,\mathrm{Re}\,W(\mathbf{q}, \varepsilon_{\mathbf{q}}), \qquad \gamma_{\mathbf{q}} = c\,\mathrm{Im}\,W(\mathbf{q}, \varepsilon_{\mathbf{q}}). \tag{22}$$

At low values of the quasimomentum q, expressions (22) yield, with account of the relations (15)–(19).

$$E_{\mathbf{q}} = \left\{1 - c\frac{1+\Lambda}{1-\Lambda}\right\}\varepsilon_{\mathbf{q}}, \tag{23}$$

$$\gamma_{\mathbf{q}} = c\pi\left\{1 + \frac{4}{3}\frac{1}{(1-\Lambda)^2}\right\}\varepsilon_{\mathbf{q}}^2 g_0(\varepsilon_{\mathbf{q}}), \tag{24}$$

where Λ is a numerical parameter, equal to in the case of a simple cubic lattice

$$\Lambda = 2JS[\mathrm{Re}\,G_{12}^0(0) - \mathrm{Re}\,G_0(0)] \approx 0.21. \tag{25}$$

We now return to formula (4) for the cross section, in which the sum over n and m is given by (20). It is clear that the first term in (20) leads to coherent single-magnon scattering, whereas the second leads to incoherent scattering. Thus, for incoherent single-magnon scattering we obtain the following result:

$$\frac{d^2\sigma_{\mathrm{incoh}}}{d\Omega\, dE'} = Nc\frac{1}{2}(r_0\eta)^2 SF^2(\mathbf{q})\frac{p'}{p}[1 + (\mathbf{em})^2]\left\{\begin{matrix} n(\Delta E) \\ n(\Delta E)+1 \end{matrix}\right\}$$
$$\times g_0(\Delta E)\left\{\left[1 - \Delta E\int_0^{E_m}\frac{g_0(E')}{\Delta E - E'}dE'\right]^2 + [\pi\Delta E g_0(\Delta E)]^2\right\}^{-1}. \tag{26}$$

We have used here a relation between the real and imaginary parts of the Green's function $G_0(E)$:

$$\mathrm{Re}\,G_0(E) = \int_0^{E_m}\frac{g_0(E')\,dE'}{E - E'}. \tag{27}$$

Expression (26) was obtained here as a result of concrete calculations for a simple cubic lattice. It can be shown that it remains valid for all three types of cubic lattices with a single magnetic atom in the primative cell. Relation (26) allows us to reconstruct from the experimentally measured incoherent magnetic scattering the density of states of the magnon spectrum $g_0(E)$, obviously, by solving the corresponding integral equation, the structure of relation (26) as a function of the transition energy ΔE being such that an approximate method of solving the integral equation can be easily developed.

At low transition energies, when $\Delta E \ll E_m$, the denominator of expression (26) can be approximated by unity, and the cross section turns out to be proportional to $g_0(\Delta E)$, in analogy with the situation that takes place for incoherent phonon scattering. We note that although the cross section of the incoherent magnetic scattering is proportional to the concentration of the impurity, which is assumed to be small, it should be readily separable from the cross section for nuclear incoherent scattering because of the factor $1 + (\mathbf{e}\cdot\mathbf{m})^2$, which depends on the orientation of the spontaneous moment of the crystal, which usually is varied with the aid of an external magnetic field. The incoherent scattering should be measured in the region of small scattering angles, where the cross section of the magnetic scattering is maximal, and that of the phonon incoherent scattering is small. It would be of interest to carry out the corresponding measurements on known cubic ferromagnetic crystals such as iron, nickel, and europium oxides.

It must be borne in mind that these measurements require neutron beams of appreciable intensity, for by virtue of the low concentration of the impurities the incoherent magnetic scattering produced by them will be suppressed by the incoherent nuclear scattering of the matrix. To this end it is necessary to choose ferromagnetic crystals with the smallest nuclear incoherent cross section. Among the known ferromagnets, the most convenient object is apparently iron. Its cross section for incoherent nuclear scattering is 0.3 b, whereas the cross section of the magnetic scattering per atom $(r_0\eta)^2 \approx 0.3$ b. When referred to a single atom in the crystal, this cross section amounts to $c(r_0\eta)^2$, where the concentration c should be taken quite small (in order to use formula (26)), not exceeding 5–7%. The additive could be aluminum, which is not magnetic, is well dissolved in iron within the limits of the indicated concentrations, and produces no nuclear incoherent scattering.

With respect to formula (26) it is useful to make the following remark. In spite of the resonant character of the denominator, the expression written out has no sharp maximum at the "resonance" point (when the first term in the denominator vanishes), for at this point the second term of the denominator is not small. For example, in a simple cubic lattice the first term vanishes when ΔE is equal to 0.6 of the width of the spin-wave band, but at this place the density of states is maximal, so that the second term in the denominator turns out to be equal to approximately 3. The absence of sharp maxima in the "resonant" factor of the function (26) should facilitate the reconstruction of the function $g_0(E)$ from data on the incoherent cross section.

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Translated by J. G. Adashko
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