THEORY OF SCATTERING OF NEUTRONS IN DISORDERED CRYSTALS

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Inelastic scattering of neutrons accompanied by emission of a localized phonon or of a spin wave is investigated. It is shown that the differential cross sections for these processes are proportional to the spectral density $\nu(\omega, c)$ in the impurity band (ω is the frequency of the emitted quantum). This conclusion does not depend on special features of the classification of localized states.

1. INTRODUCTION

IN systems without translational symmetry the concept of quasimomentum loses its meaning. Therefore, both the spectrum and the classification of elementary excitations in such systems are considerably more complicated than in ideal crystals. Investigation of the structure of the energy spectrum of quasiparticles in disordered crystals is a very important problem of the theory of the condensed state which has not been solved to date. Recently many papers have been published on this subject (cf., for example,[1-7]). Of particular interest are the recent results of Lifshitz[1,2] who has investigated the properties of the spectrum and the classification of states near singular points of the spectral density in disordered solid solutions for low concentrations of the impurity.

The addition of disordered impurities leads, in particular, to the possibility of appearance of new impurity bands. In the limit of small concentrations $c \rightarrow 0$ a singular point of the spectral density $\nu(\epsilon, c)$ in the impurity band is the point $\epsilon = \epsilon_0$, where ϵ_0 is the "initial" impurity level, i.e., the level which splits off from the continuous spectrum as a result of adding a single impurity atom.

The thermodynamic properties of a material depend only on the integrated characteristics of the energy spectrum and practically do not vary at low impurity concentrations. At the same time the kinetic phenomena in disordered crystals are in many cases altered essentially as the concentration is increased. Investigation of such phenomena, can in principle, yield detailed information on the properties of the energy spectrum in the presence of impurities.

In this paper we shall discuss inelastic one-phonon (or one-magnon) scattering of neutrons in disordered solid solutions at low impurity concentrations, which is accompanied by the excitation in the crystal of vibrations characterized by one of the impurity frequencies.[1] The differential cross-sections for these processes at frequencies close to the "initial" local frequency ω_0 turn out to be proportional to $\nu(\omega, c)$. This conclusion does not depend on the characteristic features of the classification of localized states.[2]

Thus, an investigation of the phenomena indicated above can serve as a method for determining the spectral density $\nu(\omega, c)$ in the impurity band in the neighborhood of $\omega = \omega_0$, i.e., just in the very region where $\nu(\omega, c)$ is of the greatest interest. Practically the whole integrated intensity of the function $\nu(\omega, c)$ is concentrated in this region.

2. VIBRATIONS OF DISORDERED CRYSTALLINE LATTICES

In this section we shall discuss the properties of local vibrations which are essential for the theory of neutron scattering.

Usually the theory of vibrations of disordered lattices is constructed on the assumption that the force constants are unchanged when foreign atoms are dissolved in it[3,7]. Also the deformation of the lattice in the neighborhood of the impurity atom is not taken into account, i.e., it is assumed that the difference between the original and the dissolved atoms consists only of a difference in their masses.

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[1]In the paper by Kagan and Iosilevskiĭ [3] a discussion was given of the possibility of observing discrete frequencies of lattice vibrations in experiments on neutron scattering. In this discussion it was assumed that the distinction between the components of the solid solution consists only of a difference of masses, and the concentration broadening of levels was not taken into account.

[2]However, we should remember that the spectral density itself $\nu(\omega, c)$ depends in an essential manner on the special features of the classification [1,2] which, therefore, indirectly are also manifested in these processes.

We shall not be interested in the position of the "initial" impurity level and in the other characteristics of the impurity band. For our problem only the fact itself of the presence of localized states is significant, and, therefore, we can take all the factors enumerated above into account by regarding them as being responsible for the appearance and for the characteristic features of the impurity band.

In their most general form the equations for the normal vibrations $\psi_n^{(\lambda)}$ have the form

$$M_n \omega_\lambda^2 \psi_{n\alpha}^{(\lambda)} = \sum_{n', \alpha'} A_{\alpha\alpha'}(n, n') \psi_{n'\alpha'}^{(\lambda)}, \tag{1}$$

λ is the ordinal number of a normal vibration, and ω_λ is its frequency, M_n is the mass of the atom, $A_{\alpha\alpha'}(n, n') = A_{\alpha'\alpha}(n', n)$ are the force constants; the subscript n serves to number the atoms.

It is convenient to normalize the functions ψ in the following manner:

$$\sum_n M_n \psi_n^{(\lambda)} \psi_n^{(\lambda')*} = \delta_{\lambda\lambda'}. \tag{2}$$

Then the relations which express the displacement operator u_n in terms of the operators for the creation and annihilation of phonons b_λ^+, b_λ, have the form ($\hbar = 1$)

$$u_n = \sum_\lambda (2\omega_\lambda)^{-1/2} (\psi_n^{(\lambda)} b_\lambda + \psi_n^{(\lambda)*} b_\lambda^+). \tag{3}$$

We shall assume that one impurity atom in an infinite crystal gives rise to the appearance of a nondegenerate [3] local level ω_0 with the corresponding normal vibration ("wave function") $\psi_n^{(0)}$. "Interaction" between impurity atoms leads to concentration broadening of this level.

In the presence of several impurity centers the "wave functions" in zero order approximation with respect to the "interaction" of the impurities are equal to $\psi_{n-n_j}^{(0)}$ (n_j is the position of the j-th impurity atom). In the same approximation the characteristic frequencies are equal to ω_0, i.e., the degree of degeneracy of the level ω_0 is equal to the number of impurity atoms. The correct "wave function" in zero order approximation has in the general case the form

$$\psi_n^{(\lambda)} = \sum_j c_j^{(\lambda)} \psi_{n-n_j}^{(0)}, \qquad \sum_j |c_j^{(\lambda)}|^2 = 1, \tag{4}$$

i.e., the states turn out to be collective states with respect to several impurity centers. The coefficients $c_j^{(\lambda)}$, the level shifts and the spectral density $\nu(\omega, c)$ can be determined by solving the corresponding secular equation. This problem has been considered in two limiting cases by I. Lifshitz[1,2]. In the "classical" limiting case each of the states remains localized near one of the impurity centers. In the "quantum" case [4] it is possible to have collective states between two impurity centers: $\psi_n = 2^{-1/2}(\psi_{n-n_1}^{(0)} \pm \psi_{n-n_2}^{(0)})$. The appearance of collective states between three, or a greater number of, impurity centers turns to have a very low probability.

Assuming that the spectral density $\nu(\omega, c)$ is known we shall not restrict ourselves to a discussion of the above limiting cases, but shall investigate the more general picture of states in an impurity band, i.e., we shall assume that the coefficients $c_j^{(\lambda)}$ in formula (4) are arbitrary.

3. INELASTIC ONE-PHONON SCATTERING OF NEUTRONS

We shall take the energy of interaction of a neutron with atomic nuclei of a crystal to be equal to

$$V(r) = \sum_n \alpha_n \delta(r - r_n), \tag{5}$$

where r and r_n are the coordinates of the neutron and the nucleus, α_n determines the amplitude for the scattering by a free nucleus. The coefficients α_n are treated as random quantities which take on two values with probabilities equal to the concentrations of the corresponding components of the solid solution. For the sake of simplicity we consider each of the components to be isotopically homogeneous and we do not take nuclear spins into account. The final results are also valid without these restrictions.

The differential effective cross-section for nuclear one-phonon scattering per unit solid angle and per unit energy interval has the form

$$\frac{d^2\sigma}{d\Omega_{p'} dE_{p'}} = \frac{m^2}{(2\pi)^2} \frac{p'}{p} \sum_\lambda \left| \left\langle \sum_\lambda \alpha_n e^{-iqr_n} b_\lambda^+ \right\rangle \right|^2 \delta(E_p - E_{p'} - \omega_\lambda). \tag{6}$$

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[3] Generally speaking, it is possible for the "initial" level to be degenerate. Thus, for example, in a cubic crystal the "initial" level is three-fold degenerate, and the corresponding normal vibrations are polarized along the principal crystallographic axes. This case is somewhat more complicated, but the final results remain unaltered.

[4] This case is realized if the essential factor is only a change of mass. It is evident that in the "quantum" case the vibrations of different impurity atoms are not independent even in the zero order approximation. The "pairing" of impurity atoms must be taken into account in the theory of neutron scattering, and this was not done in the paper of Kagan and Iosilevskiĭ [3].

In this formula m is the neutron mass, p and p' are the momenta before and after scattering, E_p and $E_{p'}$ are the corresponding energies, $q = p - p'$ is the transferred momentum. As usual[3,8,9], we have restricted ourselves to the first approximation of the Born scattering theory. In future we shall discuss the case of zero temperature (the results, of course, will also hold for temperatures small in comparison to the Debye temperature); therefore, the averaging in formula (6) is performed with respect to the ground state of the crystal.

We decompose the radius-vector r_n into an equilibrium part x_n, and the displacement u_n: $r_n = x_n + u_n$ and evaluate the average $\langle \exp(-iq \cdot u_n) b_\lambda^+ \rangle$. By expanding the exponential $\exp(-iq \cdot u_n)$ in series in powers of the displacement and commuting the terms of this series with the operator b_λ^+ it can be shown that

$$\langle e^{-iqu_n} b_\lambda^+ \rangle = -i(2\omega_\lambda)^{-1/2} q\varphi_n^{(\lambda)} \langle e^{-iqu_n} \rangle. \qquad (7)$$

As a result of some simple transformations formula (6) assumes the form

$$\frac{d^2\sigma}{d\Omega_{p'} dE_{p'}} = \frac{m^2}{2(2\pi)^2} \frac{p'}{p} \sum_\lambda \frac{1}{\omega_\lambda} \left| \sum_n \alpha_n e^{-iqx_n} \right.$$
$$\left. \times q\varphi_n^{(\lambda)} \langle e^{-iqu_n} \rangle \right|^2 \delta(E_p - E_{p'} - \omega_\lambda). \qquad (8)$$

In an ideal crystal the average $\langle \exp(-iq \cdot u_n) \rangle$ does not depend on λ and is expressed in terms of the Debye-Waller factor. In our case this is not so. We make use of the fact that near an impurity atom this quantity, as well as the lattice parameters, is practically unaffected by the presence of other impurity centers. We shall be interested in frequencies ω_λ close to the "initial" frequency ω_0. For such frequencies the "wave functions" are linear combinations of the functions $\psi_{n-n_j}^{(0)}$ [cf., formula (4)]. Consequently we have

$$\sum_n \alpha_n \exp(-iqx_n) q\varphi_n^{(\lambda)} \langle \exp(-iqu_n) \rangle$$
$$= \sum_j c_j^{(\lambda)} \exp(-iqx_{n_j}) \sum_{n} \alpha_n \exp(-iq(x_n - x_{n_j}))$$
$$\times q\psi_{n-n_j}^0 \langle \exp(-iqu_n) \rangle. \qquad (9)$$

The function $\psi_{n-n_j}^{(0)}$ falls off rapidly with increasing distance from the impurity center n_j. Therefore the summation over n on the right hand side of relation (9) does not depend on the position of the impurity centers and can be evaluated for the case when there exists only a single impurity atom (this will be denoted by the subscript "zero"):

$$\sum_n \alpha_n \exp(-iq(x_n - x_{n_j})) q\psi_{n-n_j}^{(0)} \langle \exp(-iqu_n) \rangle$$

$$\approx \left\{ \sum_n \alpha_n \exp(-iqx_n) q\varphi_n^{(0)} \langle \exp(-iqu_n) \rangle \right\}_0. \qquad (10)$$

We further have

$$\left| \sum_j c_j^{(\lambda)} \exp(-iqx_{n_j}) \right|^2 = 1 + \sum_{j \neq j'} c_j^{(\lambda)} c_{j'}^{(\lambda)*} \exp(-iqx_{jj'}),$$
$$x_{jj'} = x_{n_j} - x_{n_{j'}}. \qquad (11)$$

The second term on the right hand side of formula (11), generally speaking, is not small compared to unity and depends on the position of those impurity atoms to which the given state "belongs." However, averaging the quantities $\exp(-iq \cdot x_{jj'})$ over all the orientations of the vector $x_{jj'}$, we obtain[5]

$$\{\exp(-iqx_{jj'})\}_{av} = \sin qx_{jj'}/qx_{jj'} \ll 1, \qquad (12)$$

since the values of q of interest to us are of the order of the period of the reciprocal lattice (this follows from the condition $E_p - E_{p'} \approx \omega_0$; $E_p \sim \omega_0$). Consequently, the cross terms in formula (11) can be left out in calculating the scattering cross-section. Then it turns out that

$$\frac{d^2\sigma}{d\Omega_{p'} dE_{p'}} = \frac{m^2}{2(2\pi)^2} \frac{p'}{p} \left| \sum_n \alpha_n e^{-iqx_n} q\varphi_n^{(0)} \langle e^{-iqu_n} \rangle \right|_0^2$$
$$\times \sum_\lambda \frac{1}{\omega_\lambda} \delta(E_p - E_{p'} - \omega_\lambda). \qquad (13)$$

(We have made use of the fact that the summation within the absolute value signs in this formula is evaluated for a crystal with a single impurity atom and is, therefore, independent of λ.)

Taking into account the fact that

$$\sum_\lambda \delta(\omega - \omega_\lambda) = N\nu(\omega, c), \qquad (14)$$

where N is the number of atoms in the crystal, and replacing ω_λ approximately by ω_0, we obtain the final expression for the neutron scattering cross-section:

$$\frac{d^2\sigma}{d\Omega_{p'} dE_{p'}} = \frac{Nm^2}{2(2\pi)^2 \omega_0} \frac{p'}{p}$$
$$\times \left| \sum_n \alpha_n e^{-iqx_n} q\varphi_n^{(0)} \langle e^{-iqu_n} \rangle \right|_0^2 \nu(E_p - E_{p'}, c). \qquad (15)$$

Thus, by measuring the differential cross section for one-phonon scattering we can determine the spectral density $\nu(\omega, c)$ for the impurity band.[6]

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[5] Such an averaging is automatically carried out in summing over λ, because the differential scattering cross-section is a "self-averaging" quantity i.e., one valid for a macroscopic crystal. In averaging one can leave out of account the discrete structure, since the distances between impurity atoms are large.

[6] For the normalization of $\nu(\omega, c)$ one should make use of the relation $\int \nu(\omega, c)d\omega = c$ (cf., for example, [1,2]).

4. SCATTERING ACCOMPANIED BY THE EMISSION OF A SPIN WAVE [7]

In the mathematical sense the theory of one-quantum magnetic scattering is almost completely equivalent to the theory presented in the preceding section. Therefore, we shall obtain the general expression for the differential cross-section for one-quantum scattering, and we shall then formulate the final result without detailed derivations.

The matrix element of the magnetic interaction between a neutron and a scatterer taken between neutron states with momenta p and p', is equal to [9,11,12]

$$V_{pp'} = -\frac{4\pi}{m} r_0 \gamma \sum_n F_n(q) e^{-iqr_n} (S_n, S - (eS)e). \quad (16)$$

In this formula $r_0 = e^2/m_0 c^2$ is the electromagnetic electron radius, $\gamma = -1.93$ is the magnetic moment of the neutron in nuclear Bohr magnetons, $F_n(q)$ is the magnetic form factor of the atom, S_n and S are the spins of the atom and of the neutron, $e = q/|q|$, parentheses denote a scalar product.

We consider the scattering of unpolarized neutrons. After averaging over the spin orientations in the neutron beam the differential effective cross-section for scattering accompanied by the emission of a spin wave has the form

$$\frac{d^2\sigma}{d\Omega_{p'} dE_{p'}} = (r_0\gamma)^2 \frac{p'}{p} \sum_\lambda \{A_\lambda A_\lambda^* - |(eA_\lambda)|^2\}$$

$$\times \delta(E_p - E_{p'} - \omega_\lambda), \quad (17)$$

$$A_\lambda = \sum_n F_n(q) e^{-iqx_n} \langle e^{-iqu_n} \rangle \langle S_n b_\lambda^+ \rangle, \quad (18)$$

b_λ^+ is the operator for the creation of a spin wave of frequency ω_λ. We remind the reader that we are discussing the case of zero temperature.

Further, we shall utilize the approximate relations which express the spin operators in terms of the operators for the creation and annihilation of spin waves;

$$S_n^{(x)} + iS_n^{(y)} \approx (2S_n)^{1/2} \sum_\lambda \psi_n^{(\lambda)} b_\lambda,$$

$$S_n^{(x)} - iS_n^{(y)} \approx (2S_n)^{1/2} \sum_\lambda \psi_n^{(\lambda)*} b_\lambda^+. \quad (19)$$

In these formulas $\psi_n^{(\lambda)}$ are the normalized wave functions for the spin waves

$$\sum_n \psi_n^{(\lambda)} \psi_n^{(\lambda')*} = \delta_{\lambda\lambda'}, \quad \sum_\lambda \psi_n^{(\lambda)} \psi_{n'}^{(\lambda)*} = \delta_{nn'}. \quad (20)$$

They are analogous to the functions $\psi_n^{(\lambda)}$ introduced in section 2 and have the same properties.

With the aid of relations (19) we can evaluate the vectors A_λ. These calculations will be omitted and we shall give only the final result:

$$\frac{d^2\sigma}{d\Omega_{p'} dE_{p'}} = \frac{N}{2} (r_0\gamma)^2 \frac{p'}{p} (1 + \cos^2\theta_q)$$

$$\times \left| \sum_n S_n^{1/2} F_n(q) e^{-iqx_n} \psi_n^{(0)} \langle e^{-iqu_n} \rangle \right|_0^2 \nu(E_p - E_{p'}, c). \quad (21)$$

Here θ_q is the angle between the vector q and the direction of the axis of easiest magnetization.

It can be seen that investigations of magnetic scattering of neutrons can be utilized to determine the spectral density in the impurity band of spin waves.

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