AN APPROXIMATE DETERMINATION OF FREQUENCY SPECTRUM OF PHONONS IN CRYSTALS BY NEUTRON SCATTERING

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Recently Kagan[1,2] and Oskot·skiĭ[3] have suggested methods for determining the frequency distribution function for crystals from neutron scattering experiments. The idea behind these methods is to find conditions in which coherent scattering is suppressed. Though, in theory, such conditions have been shown to exist, it is pointed out here that they would be very difficult to attain experimentally. It is suggested that a fair idea of the frequency distribution function of a solid can be obtained easily by studying the energy distribution of neutrons inelastically scattered from a polycrystalline sample.

IN some recent papers[1-3] different methods have been suggested for determining the phonon frequency distribution function for crystals from neutron scattering experiments. The main idea behind these methods is to suppress coherent neutron scattering and observe only the incoherently scattered neutrons. Kagan[2], for example, suggests the study of the energy spectrum of those inelastically scattering neutrons (through one-phonon exchange) for which the change in the neutron wave vector $\mathbf{K}$ ($\mathbf{K} = \mathbf{k}_1 - \mathbf{k}_2$, $\mathbf{k}_1$ being the wave vector of incident neutrons and $\mathbf{k}_2$ that of the scattered neutrons) is equal to 2π times some reciprocal lattice vector τ of the crystal. As has been pointed out by Kagan himself, this method is not very practicable from the experimental point of view.

Besides the above method, Kagan[2] has suggested two other alternatives, (1) study of inelastically scattered neutrons at very small angles or (2) the measurement of the differential scattering cross section $\partial^2\sigma/\partial E_2\partial\Omega$ for a few fixed directions of $\mathbf{k}_2$. According to Kagan the last method is the simplest but it is the least accurate.

Oskot·skiĭ[3] has criticized Kagan's approach and has suggested another method. According to him one should study inelastically scattered neutrons (through one-phonon exchange) in a direction (or close to the direction) of Bragg reflection. Oskot·skiĭ[3] shows that if the velocity of incident neutrons is larger than the maximum velocity of sound in the crystal c_{max}, then no coherent scattering with energy gain by neutrons is possible. It is not difficult to see that Oskot·skiĭ's suggestion is also not practicable. For his suggestion to work, the incident neutrons must have an energy $E_1 > \frac{1}{2} m_0 c_{max}^2$, where m_0 is the neutron mass. This energy will range from 400 k to 10,000 k (k is the Boltzmann constant) depending upon the value of c_{max} ($c_{max} \approx (2.5 - 13) \times 10^5$ cm/sec). A crystal with low c_{max} will also have a low Debye temperature Θ, so that the incident-neutron energy required for such an experiment would be many times $k\Theta$ (see the table). This implies that even for crystals at room temperature the contribution from two- and higher-order phonon processes will be large and the cross section for scattering with energy gain will be negligible[4]. Decreasing the crystal temperature would also not improve matters.

Metal	$c_{max} \times 10^{-5}$ cm/sec	$\epsilon = m_0 c_{max}^2/2k$	Θ	ϵ/Θ
Be	12.89	10290	1000	10
Al	6.40	2483	398	6
Mg	4.60	1280	305	4
Fe	5.93	2128	420	5
Pb	2.40	348.5	88	4

If one were to consider the neutrons scattered with energy loss, then the minimum incident energy required to avoid all coherent scattering would be even higher[3].

Another point which one has to bear in mind is that for all metals, except vanadium, the bound incoherent scattering cross section per atom s is very much smaller than S, the bound coherent scattering cross section per atom. This implies that only a small fraction of neutrons scattered by a crystal are incoherently scattered.

In spite of these handicaps, one of the above methods will have to be used to study the exact

nature of the frequency distribution function close to its singularities and extremum points. It would appear, however, that, to within a reasonable degree of accuracy (in present-day neutron-scattering experiments it is difficult to attain better than a few per cent accuracy), the best method for determining the general shape of the frequency distribution function of a solid would be to study the energy distribution $d\sigma/dE_2$ of neutrons inelastically scattered by a polycrystalline sample. The incident-neutron energy should be nearly 500 k or higher, which (except in the case of lead) is of the order of $k\Theta$ ($\frac{1}{2}k\Theta$ in case of beryllium). It has been shown in case of beryllium[4,5] (but this will be true generally) that at these energies the number of contributing reciprocal-lattice vectors is so large that the energy distribution of the coherently scattered neutrons is almost the same as that of incoherently scattered ones. Thus by studying the neutrons inelastically scattered from a polycrystalline sample, it should be possible to deduce its frequency distribution function.

Marshall and Stuart[6] have given the following expression for one-phonon coherent differential scattering cross section for neutron scattering from a polycrystalline sample

$$\frac{\partial^2\sigma^{\mathrm{coh}}}{\partial E_2\partial\Omega}=\frac{NS}{4\pi}\sqrt{\frac{E_2}{E_1}}\frac{\hbar K^2}{2M}e^{-2W}\sum_{n=0}^{\infty}G(n)\frac{\sin nK}{nK}$$

$$\times\int_{-\infty}^{\infty}\frac{d\omega}{\omega}\frac{g(\omega)}{e^{\hbar\omega/kT}-1}\frac{\sin nf}{nf}\delta(E_2-E_1-\hbar\omega);$$

where N is the total number of atoms in the crystal, S is the bound coherent scattering cross section per atom, M is the mass of the scatterer atom, $\mathbf{K}=\mathbf{k}_1-\mathbf{k}_2$ is the difference of the incident neutron wave vector $\mathbf{k}_1$ and the final neutron wave vector $\mathbf{k}_2$, $\mathbf{f}$ is the phonon wave vector, and ω is the corresponding angular frequency. The frequency distribution function is represented by $g(\omega)$ whereas G(n) represents the number of atoms with position vector of magnitude n. The Debye-Waller factor is e^{-2W} and the crystal temperature is T. (Similar expression had been obtained earlier by Kothari and Singwi[7], but they omitted the factor $\sin n\mathbf{K}/n\mathbf{K}$.)

The major contribution to the above expression comes from the term with n = 0, which is identical with the expression for the incoherent scattering cross section provided one replaces S by s (the bound incoherent scattering cross section per atom). It is also seen that the correction term (i.e., the sum of all terms with $n \geq 1$) is not strongly angle-dependent. Marshall and Stuart[6] have shown by numerically evaluating the correction term that the ratio of the total correction term to the total scattering cross section in the incoherent approximation (i.e., taking the expression for incoherent scattering and replacing s by (S + s)) seldom exceeds 2% or so, particularly in the thermal energy region. This small correction leads one to expect a reasonably accurate determination of the frequency distribution function by the method discussed above.

The use of "isotopic alloys" in cases where one of the isotopes of the element under consideration has a negative scattering length has also been suggested[8]. For example, nickel containing 45.9% of Ni^{62} would exhibit no coherent scattering, nor would an alloy of 26.3% of Li^6 and 73.7% of Li^7.

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