Then the crystal is rotated until each face is eclipsed. The dial angle (ψ_A) and the scale reading (D_A) where the face cuts the x, y line are noted; the crystal is then rotated through 180° and the measurements repeated (ψ_B, D_B). To obtain θ, the stage is rotated until each face, in its eclipsed position, is parallel to one of the crosswires. A note is also made of whether the face interesects the $+z$ or $-z$ axis. If the relationship between ψ and the $x-y$ direction is as in Fig. 2 then $\varphi=90-(\psi-\psi_0)$, where ψ is the dial reading when the plane is on the 'positive' side of the z axis (*i.e.* the side to which $+y$ projects when the x axis is vertical). The distance D, from the z axis is given by half the difference between D_A and D_B, while the z axis cuts the $x-y$ plane at a scale value D_A+D. The plane equation is then:

$$x \sin(\psi-\psi_0)+y \cos(\psi-\psi_0)\pm z \tan\theta=\tfrac{1}{2}(D_B-D_A)\,.$$

For example:

$\psi_0=256°$. 1 drum unit=0·00004283 cm. Positive side of z axis is to the lower scale value.

$\psi_A=64°$, $D_A=376$ drum units.

$\psi_B=244°$ $D_B=596$ drum units.

$\theta=20°$ intersecting $+z$ (away from the goniometer head).

This gives $D=110$, the z axis is at 486, $\psi=\psi_A-\psi_0=168°$, and the plane is:

$$x \sin 168+y \cos 168+z \tan 20=110*0{\cdot}00004283$$

or

$$0{\cdot}2079\,x-0{\cdot}9871\,y+0{\cdot}3640\,z=0{\cdot}004711\,.$$

For planes which do not cut the $x-y$ line, two courses are possible. If, when eclipsed, they cut the crosswire parallel to the z axis (at 500 units on the screw-micrometer eyepiece) the eyepiece can be rotated through 90° and the distance of this intersection from the $x-y$ line measured. Then, using θ, the intersection of the eclipsed plane with the z axis can be calculated (D_z) and then the distance $D=D_z \cos\theta$. For a basal plane, of course, the equation is simply:

$$z=D_z\,.$$

Alternatively, the distance (parallel to the z axis) to some prominent point can be measured (by rotating the eyepiece), the crystal translated until this point lies on the $x-y$ line, and a further series of measurements made with a new origin, and referred to the first origin.

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Artificial Splitting of One-Phonon Neutron Groups due to Relaxed Vertical Collimation

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It is shown that in certain circumstances the relaxed vertical collimation of a triple-axis spectrometer can lead to the measured one-phonon neutron groups having two peaks, and experimental measurements which clearly show the splitting and indicate its origin are presented. In the form of dispersion curve considered, two branches of the curves are degenerate along a symmetry direction but split for any adjacent wave vector with the splitting directly proportional to the distance from the symmetry direction. An additional requirement is that the resolution function of the spectrometer should be squashed along the symmetry direction and expanded perpendicular to it. Calculations which also show the splitting of the neutron groups are described.

Introduction

In inelastic neutron scattering experiments performed with a triple-axis spectrometer, it is customary for the vertical divergence of the spectrometer to be greater than the horizontal divergence by a factor of two or three. Whenever possible specimens are mounted with a mirror plane horizontal. This normally results in the frequencies of the observed phonons or magnons not changing, to first order, for wave vectors slightly out of the scattering plane. The relaxed vertical collimation then leads to a useful increase in intensity with negligible broadening of the neutron groups. The presence of a mirror plane is not, however, sufficient to ensure that the constant frequency surfaces cut the scattering plane normally. If two branches of the dispersion curve are degenerate along a line in the scattering plane, for wave-vectors with a small off-symmetry-

direction component the two branches can split, with a splitting directly proportional to the off-symmetry-direction component of the wave vector. Examples of this occur for transverse modes propagating along the [111] direction in common cubic structures (Gilat, 1969), and for Λ_3 modes propagating along the [111] direction in calcite. It is clear that in this case any relaxing of the vertical divergence will at best give rise to a broadening of the neutron groups. We present here experimental measurements made on calcite which show more serious deformation of the neutron groups caused by this type of constant frequency surface. Such measurements can easily be misinterpreted, especially if the shape of the dispersion curves is not more or less known in advance. It therefore seems worth while to point out the possibility of this splitting arising.

Recently Raunio, Almqvist & Stedman (1969) and Raunio & Almqvist (1969) have reported measurements on sodium chloride and potassium chloride in which similar difficulties were encountered, which they ascribe to the same cause as in the present work.

Experimental results

The experiments were carried out on the McMaster University triple-axis spectrometer on the NRU reactor at Chalk River (Brockhouse, deWit, Hallman & Rowe, 1968). In normal operation the vertical divergences of the incident and scattered beams are determined principally by the sizes of the specimen and the monochromating and analysing crystals, and the distances between them. This results in a vertical divergence of approximately 1 in 60 for the incident beam, and 1 in 20 for the scattered beam. These values are normally found to be acceptable in that peak centres do not change when finer collimation is used.

Fig. 1(*a*) shows a neutron group obtained from a natural single crystal of calcite for a wave vector corresponding to the zone boundary in the [111] direction, that is, the point Z in the Brillouin zone (Cowley, 1969). For all the measurements shown here the incident neutrons had an energy of 40 meV. This neutron group was originally interpreted by us as consisting of two partially resolved peaks caused by phonons with frequencies of approximately 2·4 and $3{\cdot}0 \times 10^{12}$ cps. Measurements made for neighbouring wave vectors were consistent with this interpretation and we were able to trace out what appeared to be two branches of the dispersion curves over most of the [111] direction. In addition measurements made in a second region of reciprocal space gave very similar results.

Fig. 1(*b*) shows the neutron group measured under the same conditions except that the vertical divergence of both incident and scattered beams was reduced to 1 in 80, equal to the horizontal divergences. The overall width of this group is only slightly greater than typical values of one-phonon groups, and it appears that the double peak in Fig. 1(*a*) arises in some way from the imperfect resolution of the spectrometer and not from the existence of two branches of the dispersion curves. The neutron groups shown in Fig. 1(*c*) and 1(*d*) were measured under exactly the same conditions as for 1(*b*),

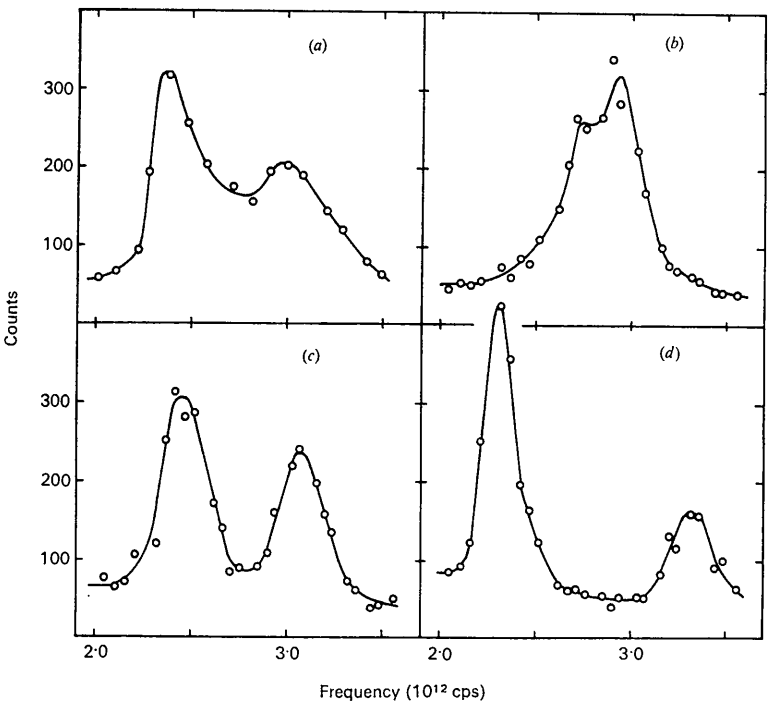

Fig. 1. Measured neutron groups at the [111] zone boundary: (*a*) relaxed vertical collimation, (*b*) fine vertical collimation, (*c*) and (*d*) as for (*b*) with crystal tilted by $\frac{3}{4}°$ and $1\frac{1}{2}°$. The group shown in (*a*) has been scaled down by a factor of four.

that is, with fine collimation, except that the crystal was tilted along the scattering vector, **Q**, by $\frac{3}{4}°$ and $1\frac{1}{2}°$ respectively, deliberately introducing an out-of-plane component of the wave vector. These measurements show clearly the splitting of the neutron groups varying linearly with the amount of tilt, and suggest that the splitting observed in Fig. 1(*a*) is caused by contributions from phonons with wave-vectors slightly off the [111] direction.

Interpretation

We interpret the splitting of the neutron group shown in Fig. 1(*a*) as arising from the peculiar form of the constant frequency surfaces around the [111] direction. Gilat (1969) has shown that in common cubic structures the frequencies of the two transverse acoustic branches in this region of reciprocal space are given by

$$\nu_T = \nu_T(q_A) \pm A\eta$$

where q_A is the component of the wave vector along the [111] direction and η is the component of the wave vector perpendicular to [111]. The same expression applies to the transverse acoustic modes along the [111] direction in calcite.

The splitting is then essentially a density of states effect. Due to the finite resolution of the spectrometer each neutron group contains contributions from a range of wave-vectors around the central value. If the dispersion curves have the form indicated above, for any wave-vector slightly away from the symmetry direction there are two frequencies, one slightly below the central value and one slightly above it. Depending on the form of the resolution function there can thus be more modes contributing to the neutron group with frequencies slightly above or slightly below the central value than there are at that value.

An easily visualised limiting case would occur if the resolution function was a thin disc with its axis along the [111] direction. The number of modes sampled with wave-vectors between η and $\eta + d\eta$ from the [111] direction is then proportional to $2\pi\eta d\eta$, so that if no other factors entered the cross section the neutron group would show a V-shaped dip at $\nu_T(q_A)$, with the intensity rising linearly on either side. Any thickness of the resolution function along the [111] direction smears out the dip, the amount of smearing depending on the change of $\nu_T(q_A)$ across the thickness of the resolution function.

To demonstrate that the splitting can still remain visible with a fairly realistic resolution function we have carried out a calculation using the elastic constants of calcite to generate the constant frequency surfaces and eigenvectors in the neighbourhood of the [111] direction and assuming that the contribution made by any mode to the neutron group was proportional to a factor

$$\exp -\{(\delta q_x/R_x)^2 + (\delta q_y/R_y)^2 + (\delta q_z/R_z)^2\}$$

where $(\delta q_x, \delta q_y, \delta q_z)$ is the distance in reciprocal space between the wave-vector of the mode and the wave-vector for which the neutron group is measured. The resolution function of a triple-axis spectrometer has been considered in detail by Cooper & Nathans (1967). Compared with their results the above formula neglects

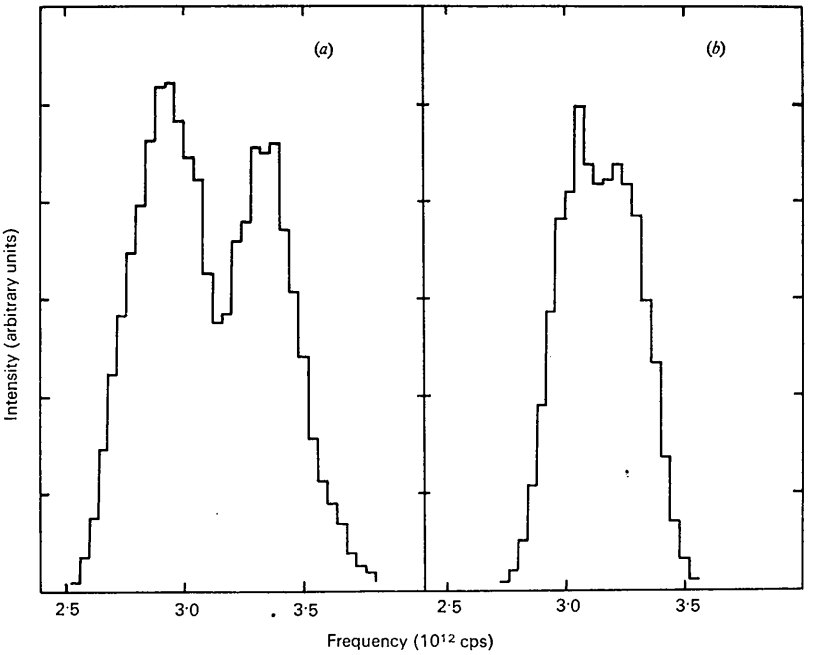

Fig. 2. Calculated peak shapes corresponding to (*a*) and (*b*) of Fig. 1.

all correlation between momentum transfer and energy transfer, but is otherwise a good approximation for the geometry of our experiment, in that by coincidence the principal axes of the resolution ellipsoids coincided with the crystal axes. Focusing effects were omitted to simplify the calculation, but their omission is to some extent justified by the fact that we have observed split peaks under several focusing conditions. Their inclusion would therefore not add anything to the calculation and would obscure the essential density of states argument.

The use of the elastic constants to generate the dispersion curves was also made for speed of calculation. We have also used a microscopic force constant model to calculate dispersion curves, and for the modes of interest the continuum calculation gives frequencies and eigenvectors in error by only a few per cent. Also we have observed split peaks for the transverse acoustic branches over almost the whole of the [111] direction, so that the splitting is not due to any property of the dispersion curves special to the zone boundary.

For a given arrangement of the spectrometer the constants R_x, R_y, and R_z can be estimated, and the two peaks shown in Fig. 2 were calculated using values for these constants corresponding to the neutron groups shown in Figs. 1(*a*) and (*b*). The centres of the calculated peaks come at too high a value because no dispersion was allowed for in the calculation of the frequencies, but the overall widths are in good agreement with the measured values and the splitting effect is clearly shown.

An essential feature of the resolution function for this splitting to occur is that the elliposidal surfaces of constant resolution are squashed along the [111] direction and expanded perpendicular to it, so that in an extreme case they would become discs perpendicular to the [111] direction. A calculation using equal resolution in all directions, with R_x, R_y, and R_z equal to the geometric mean of the values used for Fig. 2(*a*) gave rise to a single, flat-topped peak.

Discussion

We have shown that a particular form of dispersion curve, where two branches of the dispersion curves are degenerate along a symmetry direction but are split for any adjacent wave vector, can lead to neutron groups with two peaks, which can easily be interpreted as due to two distinct phonons in the symmetry direction. This distortion of the neutron groups is most marked if the constant resolution surfaces are squashed along the symmetry direction. This is, however, often the case in measurements on transverse phonons.

The magnitude of the effect observed in calcite is much larger than has been seen in other materials. The size of the effect depends both on the size of the resolution function used and on the rate of divergence of the two transverse branches away from the symmetry direction. The relevant parameter here is the ratio

$$\mu = \left| \frac{\partial v_T}{\partial \eta} \Big/ \frac{\partial v_T}{\partial q_A} \right|.$$

Gilat (1969) has given an expression for μ appropriate to the elastic constant limit in cubic crystals and gives values of 0·091 for aluminum and 0·597 for copper. An exactly similar calculation for rhombohedral crystals leads to the expression

$$\mu = |2c_{14}/c_{44}|$$

using the crystal orientation commonly used in elastic constant measurements (Peselnick & Robie, 1963). For calcite this has the value 1·256. This large value is probably the main cause of the pronounced splitting we observe, and the effect is possibly also enhanced by the fact that the only points in reciprocal space with favourable structure factors for the phonon examined here occur at larger values of the scattering vector **Q** than would normally be used. This necessitated the use of larger incident and scattered wave-vectors resulting in poorer resolution.

For simple materials the spurious nature of the extra neutron groups would soon be discovered, but for a crystal with a complicated structure there is no simple way of knowing in advance how many branches of the dispersion curves to expect in any frequency range and results of the type shown here can be misleading. The simple remedy is to use finer collimation when such an effect is possible. In the present example, however, the improvement in resolution was accompanied by a loss of a factor of five in intensity so that measurements must usually be made initially with somewhat relaxed collimation. Awareness that this type of effect can occur should then enable spurious peaks to be detected.

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