INELASTIC SCATTERING OF COLD NEUTRONS BY CONDENSED ARGON

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In recent years the question of the existence of collective modes of heat motion in liquids has been put forward by different authors. In order to investigate this problem we have done neutron inelastic scattering measurements on liquid argon (88°K). As a comparison, solid polycrystalline argon (80°K) was also studied. The experiment was carried out with the time-of-flight spectrometer at the Studsvik R2 reactor. The incident wavelength was 4.1 Å and the energy spectra of the scattered neutrons were analyzed at 9 angles between 60° and 100°.

Theoretically, it is well known[1,2] that the coherent single-phonon scattering by a polycrystal will show discontinuities at angles and energies determined by the dispersion relations and the crystal structure. These breaks will occur at energies given by equations of the following form:

$$\kappa(\Theta,\omega)_{\max} - q(\omega) = 2\pi\tau \cdots \quad (1a)$$

or

$$\kappa(\Theta,\omega)_{\min} + q(\omega) = 2\pi\tau \cdots, \quad (1b)$$

where κ is the momentum change in the scattering process, q is the phonon wave number, and τ is a vector of the reciprocal lattice. By measuring the position of the breaks we could, with the help of Eqs. (1a) or (1b), determine an average dispersion relation.

Figure 1(a) shows a typical example of the spectra scattered by the polycrystalline sample. Figure 1(b) shows the dispersion relation determined from such measurements.

In the liquid phase there are no longer distinct values of τ. Instead of a set of δ-functions in the angular distribution of the elastically scattered neutrons, there is a continuous distribution $[1+\gamma(\vec{\kappa}_0)]$. Equations (1a) and (1b) also apply to the scattering from a liquid[3] if the continuous pair correlation function $[1+\gamma(\vec{\kappa}_0)]$ is substituted for the set of distinct τ values. Assuming that the concept of phonons has some meaning in a liquid, it can be shown that the first peak in $[1+\gamma(\kappa)]$ is broadened at $\omega \neq 0$. For a certain energy the broadening is related to the average phonon wave vector at this energy. According to this the dispersion relation in a liquid can be defined in the form

$$\omega = \omega(\Delta\kappa), \quad (2)$$

where we might use as an approximation

$$\Delta\kappa = [(\Delta\kappa_\omega)^2 - (\Delta\kappa_0)^2]^{1/2}; \quad (3)$$

$\Delta\kappa_\omega$ = width of the first diffraction peak at energy ω.

After subtraction of the incoherent contribution we evaluated $\Delta\kappa_\omega$ from the experimental data around the first diffraction peak for a set of energies. Examples for three energies of observed intensities divided by κ^2 as a function of κ are shown in Fig. 2(a).

The dispersion relation obtained by the method described above is plotted in Fig. 2(b). From

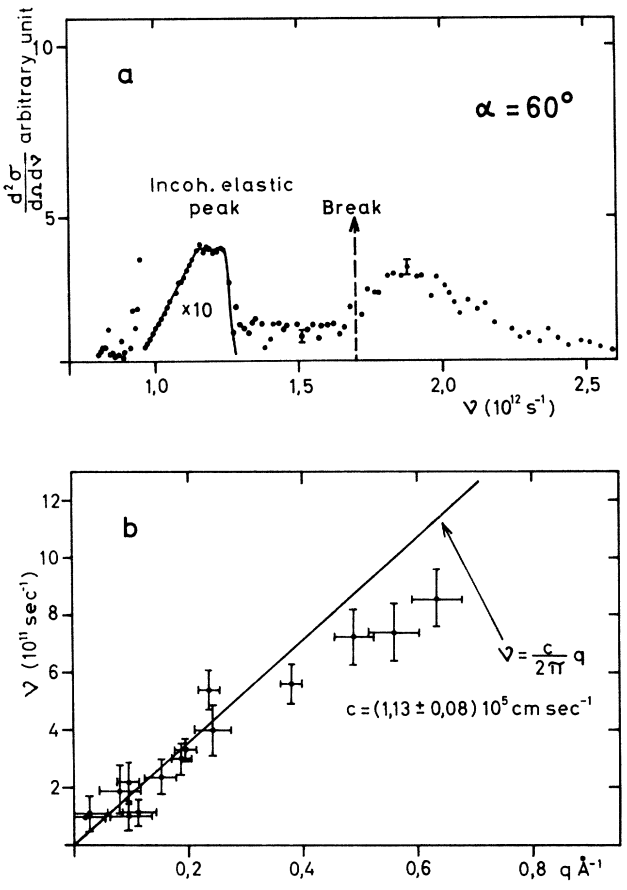

FIG. 1. (a) Energy distribution of neutrons scattered at 60° by solid argon (T = 80°K). (b) Average dispersion curve in solid argon (T = 80°K).

the initial slope of our measured dispersion curves we determined the sound velocities. They were found to be

$$c_{\text{solid exp.}} = (11.3 \pm 0.8) \times 10^4 \text{ cm sec}^{-1}$$

$$c_{\text{liquid exp.}} = (9.1 \pm 0.5) \times 10^4 \text{ cm sec}^{-1}.$$

The sound velocities in polycrystalline argon at 80°K were found by ultrasonic methods[4] and are

$$c_{\text{long.}} = 13.2 \times 10^4 \text{ cm sec}^{-1},$$

$$c_{\text{transv.}} = 7.5 \times 10^4 \text{ cm sec}^{-1}.$$

The sound velocity in liquid argon at 90.3°K was measured by acoustical methods[5] and found to be

$$c_{\text{liquid}} = 8.2 \times 10^4 \text{ cm sec}^{-1}.$$

As argon shows both coherent and incoherent scattering, the coherent inelastic scattering effects are not clearly visible in one single observation for the liquid state as is the case for the solid state as exemplified in Fig. 1(a). The implication of our result for the liquid as shown in Fig. 2(a) is, however, that the same coherent scattering rules are operative in the liquid state as in the solid. The size of the region over which cooperative or coherent scattering occurs may not at present be estimated with certainty but is large enough to allow the development of a distinct phase relation between wavelets scattered from various argon nuclei.

A study of the time behavior of the atomic motions is best performed by an analysis of the incoherently scattered quasi-elastic neutron intensity. As neutron scattering experiments on other incoherently scattering liquids have indicated a minimum residence time of an atom in a quasi-equilibrium position of about 10^{-12} seconds,[6] we are performing studies on liquid argon to find if a similar situation exists in this case. Such measurements are performed at small scattering angles where the coherent effects are unimportant. It seems already quite clear that cross-section formulas based on the elementary diffusion model[7] are incapable of explaining the data. We are therefore trying more elaborate models for the combined diffusive and vibratory motions like the stochastic model.[8]

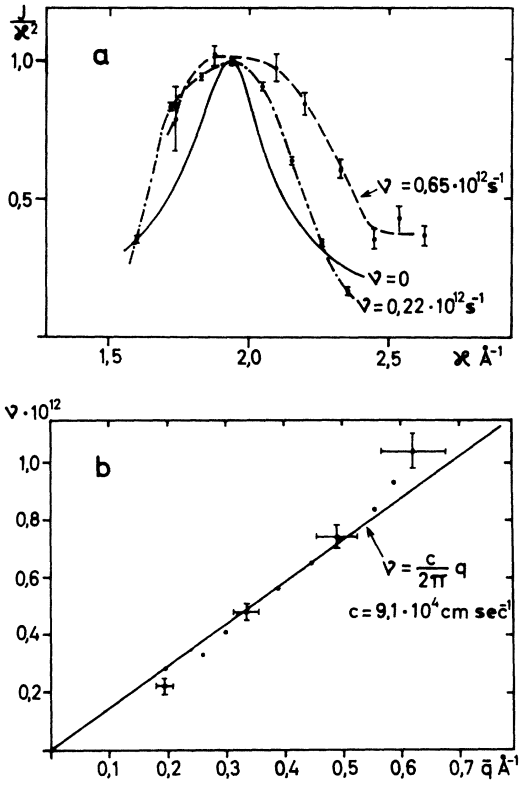

FIG. 2. (a) Examples of observed intensity at constant ω divided by κ^2 as a function of κ around the first diffraction peak in liquid argon ($T = 88$°K). (b) Average dispersion curve in liquid argon ($T = 88$°K).

A detailed report on this work will be published.

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