SMALL ENERGY TRANSFER SCATTERING OF COLD NEUTRONS FROM LIQUID ARGON

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(Received 18 August 1967)

The quasielastic scattering of 4.1-Å neutrons from liquid argon at 94 ± 2°K has been studied for wave-vector transfers in the region 1.3 $Å^{-1} < Q < 2.6$ $Å^{-1}$ which covers the main peak in the liquid structure factor. The peak intensity and peak width exhibit the same type of oscillatory behavior as observed earlier in other coherently scattering liquids. A simple modification of Vineyard's convolution approximation is described by which these oscillations are quantitatively explained.

An extensive study of the scattering of 4.1-Å neutrons from liquid argon at 94 ± 2°K has been reported in an earlier paper[1] together with a thorough discussion of the inelastic energy transfer region of the neutron spectra. In the present paper the small energy transfer or quasielastic scattering is considered and a model is proposed for the coherent contribution to this scattering.

Spectra observed at 17 angles of scattering from liquid argon at 94 ± 2°K are shown in Fig. 5 of the article referred to above. The experimental details and a description of the data treatment are also given there. For the present purpose, it is assumed that the quasielastic scattering, which is associated with diffusional motion, can be specified by the height and the width of the peak at each angle, and that the influence of the vibrational motion is negligible for frequency transfers smaller than the width of the peak.

It was shown by de Gennes[2] that the second and fourth energy moments of the coherent scattering law vary with Q in such a way that the curve is expected to be high and narrow at values of Q for which the structure factor $S(Q)$ is large, and vice versa. These predictions are verified by observations of the scattering from liquid lead[3] and liquid argon[4,5] and also by the present experiment. The first few moments thus describe the broad features of the scattering law correctly, but they are not sufficient to predict the exact value of the width and the height unless the shape of the curve is specified.

Vineyard[6] proposed that the continuous diffusion model be used to describe the incoherent scattering and he derived a simple expression for the coherent scattering law $S_c(Q, \omega)$ in terms of the incoherent scattering law $S_i(Q, \omega)$ and the structure factor $S(Q)$:

$$S_c(Q, \omega) = S_i(Q, \omega) S(Q). \quad (1)$$

A serious objection often raised against this approximation is that it violates the moment relationships and that it predicts an energy width of $S_c(Q, \omega)$ which is the same as that of $S_i(Q, \omega)$ for all values of Q. It is thus unable to explain the observed fluctuations in the width of $S_c(Q, \omega)$. A simple ad hoc modification of Eq. (1) will now be outlined by which these difficulties can be overcome, at least in part.

The first moment and, in the classical limit, also the second moment of the scattering laws differ only in that M in the incoherent moment is replaced by $MS(Q)$ in the coherent moment. An earlier proposal[7] suggested that the convolution approximation of Vineyard's Eq. (1) could be improved by substituting $MS(Q)$ for M in the expression for $S_i(Q, \omega)$. Qualitatively, this substitution may be understood in the following way: The coherently scattered intensity is composed of the scattering from several atoms, $S(Q)$ being a measure of the effective number of atoms which contribute to the intensity at wave vector Q. The recoil energy transferred to the system is therefore $\hbar^2Q^2/2MS(Q)$–i.e., the same as if the recoiling mass were $MS(Q)$.

We may equally well consider this as a scattering process in which the wave-vector transfer is $Q[S(Q)]^{-1/2}$. If, therefore, the incoherent scattering law is used to describe the dynamical part of $S_c(Q, \omega)$, it seems reasonable to assume that it should be evaluated at $Q[S(Q)]^{-1/2}$, and we arrive at the following modified form of Eq. (1):

$$S_c(Q, \omega) = S_i(Q[S(Q)]^{-1/2}, \omega) S(Q). \quad (2)$$

The structure-dependent scaling of the Q values in $S_i(Q, \omega)$ allows this formula to fulfill the first and, at least in the classical limit, also the second moment relationship.

In order to compare Eq. (2) with the observed quasielastic scattering from liquid argon, we

must specify $S_i(Q,\omega)$. Since we are interested in small energy transfers only, we may use the simple diffusion formula.[6] This is in accordance with the observations of Dasannacharya and Rao[8] and leads to the following expression for the double differential scattering cross section including both coherent and incoherent scattering:

$$\frac{d^2\sigma}{d\Omega d\omega}=\left(\frac{\omega+\omega_0}{\omega_0}\right)^{1/2}\times\left[a_i^{\,2}\frac{DQ^2}{(DQ^2)^2+\omega^2}+a_c^{\,2}\frac{DQ^2}{[DQ^2/S(Q)]^2+\omega^2}\right]. \quad (3)$$

The originally proposed modification of Eq. (1)[7] yields the same result in this case since substituting $MS(Q)$ for M is equivalent to substituting $D/S(Q)$ for D if simple diffusion is considered. This may be seen from the relation between D and M given by the Langevin diffusion equation. A formula similar to Eq. (3) was recently used by Venkataraman, Dasannacharya, and Rao[9] to describe the scattering from liquid CD_4. The width of the incident spectrum is comparable with the width of the cross section which must therefore be folded over the incident spectrum before it is compared with the observed curves. The width at half-maximum and the height of the peak obtained when the cross section is folded over the incident spectrum is shown in Figs. 1(a)-

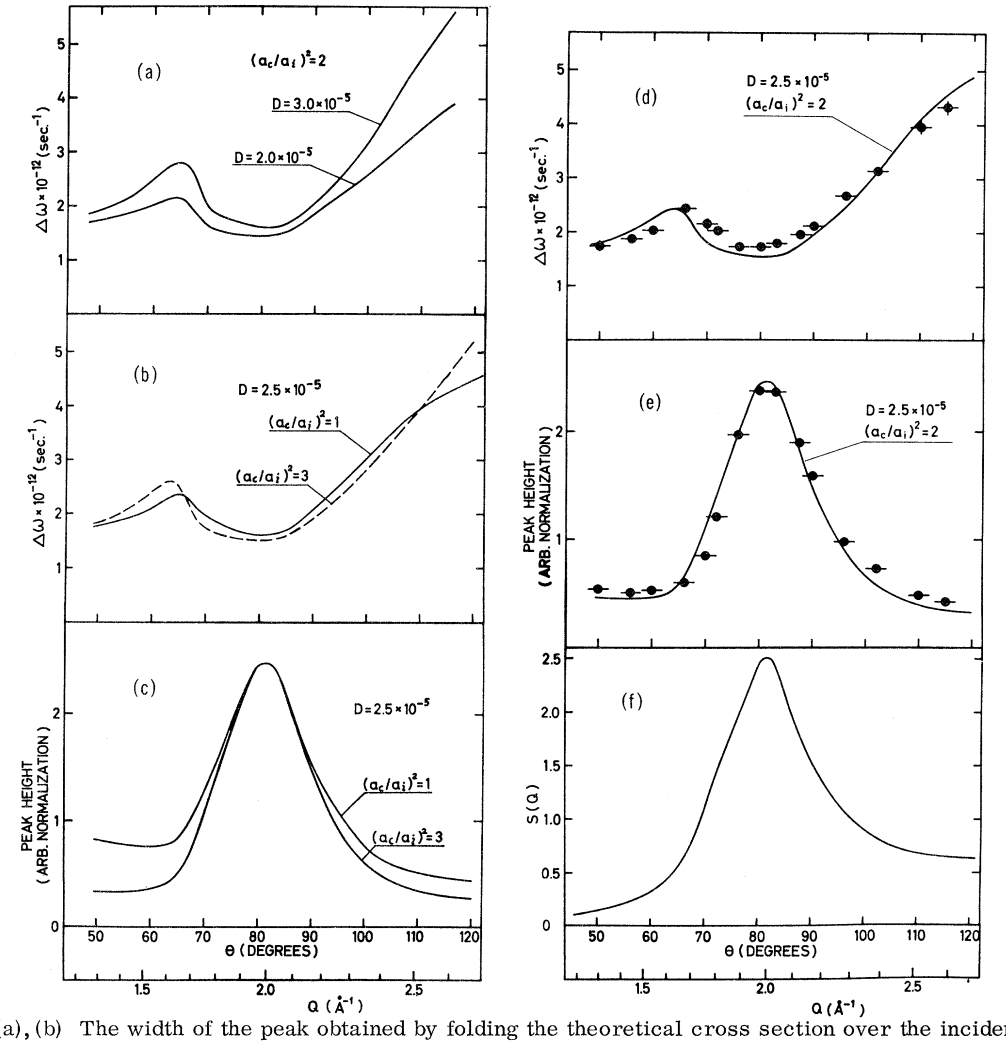

FIG. 1. (a), (b) The width of the peak obtained by folding the theoretical cross section over the incident spectrum is shown as a function of Q for various values of the parameters. (c) The height of the peak obtained by folding the theoretical cross section over the incident spectrum is shown as a function of Q for various values of the parameters. (d) The experimentally determined width of the quasielastic peak from liquid argon at 94.4°K is shown as a function of Q. The solid line is the theoretical curve. (e) The experimentally determined height of the quasielastic peak from liquid argon at 94.4°K is shown as a function of Q. The solid line is the theoretical curve. (f) $S(Q)$ for liquid argon at 84°K. This curve is due to Henshaw (Ref. 10).

1(c) for various values of D and $(a_c/a_i)^2$. In all these calculations the structure factor obtained by Henshaw[10] at 84°K was used. The effect of variations in D on the shape of the peak height curve is small and is therefore not shown in Fig. 1.

The full width at half-peak height and the maximum intensity of the observed spectra are shown in Figs. 1(d) and 1(e), respectively, as functions of scattering angle and wave-vector transfer. Q is calculated assuming that both the incident and the scattered neutron wavelength is 4.1 Å. Vertical bars in Fig. 1 indicate errors in the counting statistics and horizontal bars represent the angular resolution of the spectrometer. The solid lines in Figs. 1(d) and 1(e) are the curves obtained from Eq. (3) for the values of the parameters shown.

The value used for the ratio of the scattering lengths is the one reported by Henshaw,[10] and the value used for the diffusion constant should be compared with the values $D=2.43\times10^{-5}$ cm^2/sec and $D=3.5\times10^{-5}$ cm^2/sec, obtained by Naghizadeh and Rice[11] at $T=90$ and $T=100$°K, respectively.

The agreement between the predictions of Eq. (3) and the experimental results must be considered satisfactory. The "theoretical" value for the width depends strongly on the exact shape of $S(Q)$ which was not available at the appropriate temperature. The peaks in $S(Q)$ are expected to broaden when the temperature increases and this will decrease the fluctuations in the calculated width and also in the peak height. Use of a structure factor obtained at 94°K could therefore be expected to improve the agreement.

The author is indebted to Professor Waller for valuable discussions on the subject matter of this paper.

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PROPAGATION OF SOUND IN MONATOMIC GASES

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(Received 12 September 1967)

Since the days of Kirchhoff, the velocity of propagation and the absorption of sound in gases are usually derived from the Navier-Stokes hydrodynamical equations. For monatomic gases the results are especially simple. One finds for the velocity U and the absorption coefficient in amplitude α, as function of the frequency ω, expansions of the form

$$U_0/U = 1-a_1\xi^2+a_2\xi^4-\cdots,$$

$$\alpha U_0/\omega = b_1\xi-b_2\xi^3+\cdots, \qquad (1)$$

where

$$\xi = \mu\omega/\rho U_0^2, \qquad (2)$$

with ρ and μ the mass density and viscosity coefficient of the gas, respectively, and $U_0 = (5kT/3m)^{1/2}$. The coefficients a_i and b_i are constants if one assumes that the heat conductivity κ is related to μ by

$$\kappa = \frac{15}{4}\frac{k}{m}\mu,$$

which is closely fulfilled for all monatomic gases. One obtains

$$b_1 = 7/6; \quad a_1 = 141/72; \quad b_2 = 1559/432; \qquad (3)$$