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Collective excitations of normal liquid ^{4}He, at 3.1 K, studied by neutron inelastic scattering

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Neutron inelastic scattering from normal liquid ^{4}He at $T=3.1$ K and wave vectors in the region $0.2 \leqslant \kappa \leqslant 2.2$ Å^{-1} reveals the existence of propagating collective excitations, as measured by peaks at finite energies in the dynamical structure factor $S(\kappa, \omega)$. The spectra are in some respects similar to spectra of the superfluid liquid. The relation to other normal liquids, in particular to Rb and Ar, is discussed.

Since the original proposal by Feynman[1] that the characteristics of superfluid ^{4}He could be explored by neutron scattering, the unique properties of ^{4}He II have been studied extensively by this technique.[2,3] However only relatively little interest has been devoted to ^{4}He I, the normal liquid above the λ temperature $T_\lambda = 2.17$ K. Here we present experimental neutron results which, when compared to earlier investigations, show that ^{4}He I is of interest in its own right. Further our results elucidate the general problem of when liquids sustain propagating short-wavelength collective excitations.

Two neutron experiments on He I prior to our study should be mentioned. Woods *et al.*[4] have studied zero sound at $T=2.3$ K, showing that also in the normal phase is there evidence of propagating modes, but their wave vector transfer did not exceed 0.8 Å^{-1}. At $T=4.2$ K Woods *et al.*[5] have reported that at wave vectors $\kappa \geqslant 0.1$ Å^{-1} the dynamical structure factor $S(\kappa, \omega)$ is quasielastic, which we find is reminiscent of overdamped, nonpropagating collective motion. We here present $S(\kappa, \omega)$ of ^{4}He I at 3.1 K as determined by neutron inelastic scattering for wave vectors ranging from $0.2 \leqslant \kappa \leqslant 2.2$ Å^{-1}. The measurements were made at the triple-axis spectrometer TAS I at the cold source of the DR 3 reactor at the Risø National Laboratory. The spectrometer was run in the constant wave vector mode, with a fixed incident neutron energy of 5 meV. For comparison measurements on superfluid liquid ^{4}He II at 1.9 K were also made. The results obtained for $\kappa = 0.2$ and 2.0 Å^{-1} are shown in Fig. 1 together with the scattering results for the empty sample chamber, an aluminium cylinder with an inner radius of 0.9 cm.

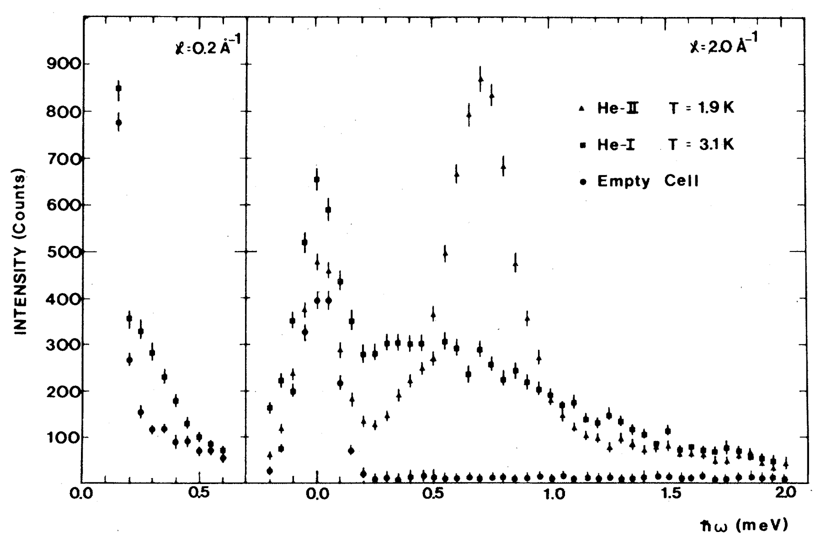

FIG. 1. Measured intensity data from the empty cell (●), the cell filled with liquid ^{4}He at 1.9 K (▲), and the cell filled with liquid ^{4}He at 3.1 K (■).

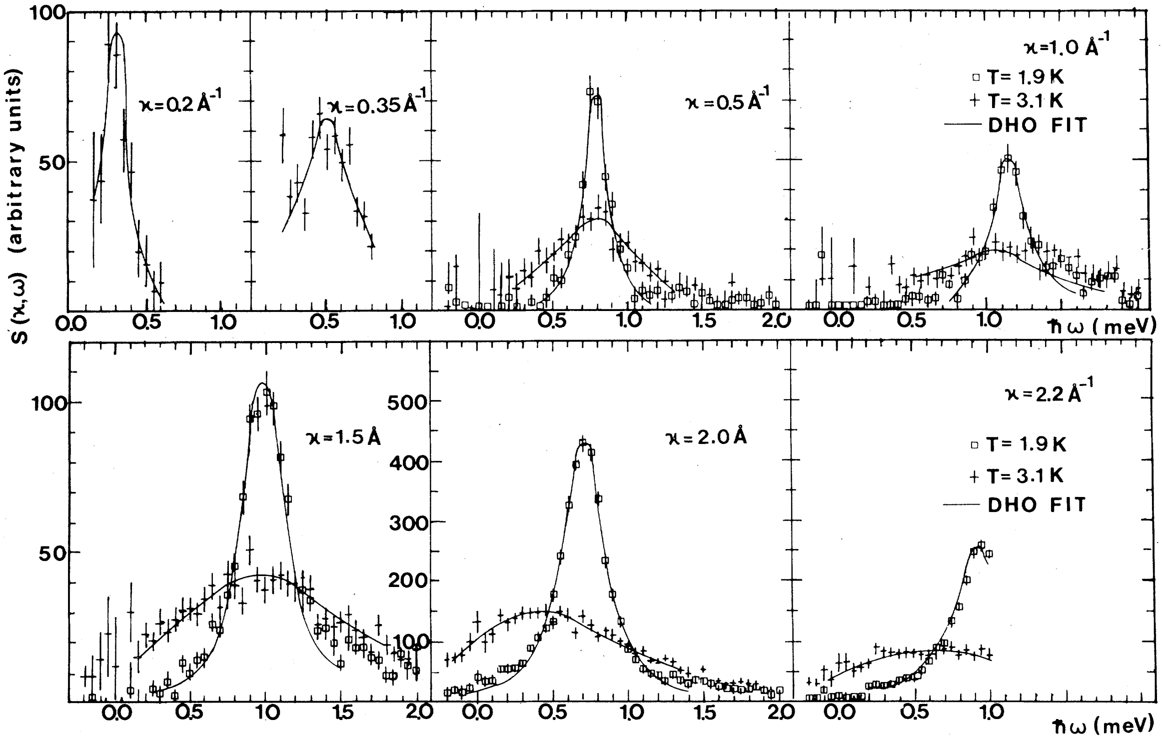

FIG. 2. Dynamical structure factor $S(\kappa,\omega)$ for liquid ^{4}He at 1.9 K (□) and at 3.1 K (+) broadened by instrumental resolution. Full lines show fits to the data in terms of a damped harmonic oscillator (DHO).

Figure 2 shows the results for normal and superfluid ^{4}He after subtraction of the intensity from the empty sample chamber and corrected for instrumental sensitivity.[6] Our spectra at $T=1.9$ K agree with the results of both Cowley and Woods[2] as well as with the recent results of Refs. 7 and 8, giving the well-defined dispersion relation with the roton minimum at $\kappa \simeq 2$ Å^{-1} and $\hbar\omega=0.7$ meV. Compared to these spectra, the spectra at $T=3.1$ K show the following features: (i) At small wave vectors, $\kappa \leq 1$ Å^{-1} a broad but well-defined excitation exists, which seems to be symmetrically broadened with respect to the sharp low-temperature excitation. (ii) At larger wave vectors, the line shapes are asymmetrical, and, although a maximum at finite energy can be located, it is now questionable whether this maximum stems only from the asymmetry owing to detailed balance, which states

$$S(\kappa,\omega)=e^{\hbar\omega\beta}S(\kappa,-\omega) \tag{1}$$

or whether it originates from propagating collective excitations. In Eq. (1) $\beta=(k_BT)^{-1}$. Further it is notable from Fig. 2, that at high frequencies, the spectra appear to be the same at the two temperatures. In an attempt to resolve the nature of the excitations in particular at $\kappa \geq 1$ Å^{-1} we analyze $S(\kappa,\omega)$ by assuming a damped harmonic oscillator

$$S(\kappa,\omega) \propto (1-e^{-\hbar\omega\beta})^{-1}\frac{4\Gamma(\kappa)\omega}{[\omega^2-\Omega(\kappa)^2]^2+\Gamma(\kappa)^2\omega^2}\ , \tag{2}$$

where Ω and Γ are the wave-vector-dependent frequency of oscillation and damping, respectively. The results of these fits are shown in Fig. 3.

Our results for ^{4}He I at $T=3.1$ K differ from the earlier results at 4.2 K in that we observe peaks in $S(\kappa,\omega)$ at appreciably higher κ's, indicating a rather solidlike behavior as regards microscopic excitations of normal ^{4}He at the lower temperatures, a feature which has traditionally been attributed solely to the superfluid phase. In order to demonstrate that our result is independent of the specific form (2), fits to a Lorentzian line shape

$$S(\kappa,\omega) \propto (1-e^{-\hbar\omega\beta})^{-1}\left(\frac{\omega\Gamma(\kappa)}{[\omega-\Omega(\kappa)]^2+\Gamma^2(\kappa)} + \frac{\omega\Gamma(\kappa)}{[\omega+\Omega(\kappa)]^2+\Gamma^2(\kappa)}\right) \tag{3}$$

are also shown in Fig. 3. Although Ω does depend upon the model,[9] in both cases dispersion curves

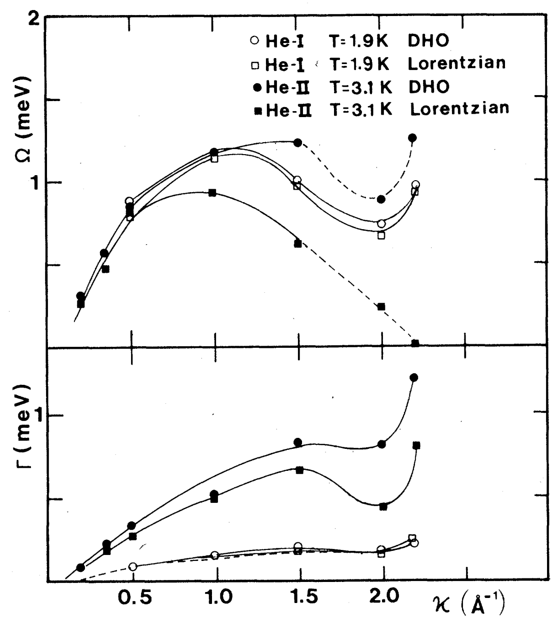

FIG. 3. Frequency $\Omega(\kappa)$ and damping $\Gamma(\kappa)$ for liquid ^{4}He at 1.9 and 3.1 K obtained by fitting the data for the dynamical structure factor to an expression for a damped harmonic oscillator (DHO) and to a Lorentzian form. The dashed line indicates experimental resolution; and the solid lines are guides to the eye.

$\Omega(\kappa)$ are well defined up to $\kappa \simeq 1.5$ Å^{-1}, i.e., well beyond the maximum in $\Omega(\kappa)$.

Few theoretical attempts have been made to calculate $S(\kappa,\omega)$ for ^{4}He I. However, the recent work of Harris and Evans[10] gives results which are consistent with our conclusions. Further using the molecular dynamics computer simulation, Rahman *et al.*[11] found the scattering law for a Lennard-Jones amorphous solid at very low temperatures similar to the one we have found for ^{4}He I.

In view of our results for ^{4}He I, it is interesting to illuminate the two very recent interpretations of $S(\kappa,\omega)$ in ^{4}He II. Tarvin and Passell[6] concluded that Eq. (2) gave a somewhat better fit than Eq. (3), which led to an $\Omega(\kappa)$ rather independent of T below T_λ. This can apparently be extended above T_λ leading to the conclusion that Ω (or the restoring force $\propto \Omega^2$) is independent of phase as opposed to $\Gamma(\kappa)$. An alternative interpretation presented by Woods and Svensson[7] assumes that the normal (n) and the superfluid (s) fractions of ^{4}He-II scatter distinctly differently so that $S(\kappa,\omega) = S_s(\kappa,\omega) + S_n(\kappa,\omega)$, where the integrated intensity of $S_s(\kappa,\omega)$ follows the superfluid fraction ρ_s. Their analysis is based on the assumption that $S_n(\kappa,\omega)$ only varies with temperature via thermal population of identical excitations. It seems therefore worth pointing out that our results at $T = 3.1$ K, when compared to the earlier reported results at 4.2 K, indicate a nontrivial temperature dependence of $S(\kappa,\omega)$ for ^{4}He I. As a consequence we find it likely that the appreciable high-frequency scattering in $S_s(\kappa,\omega)$ as deduced in Ref. 8 might in part stem from insufficient subtraction of the normal component, $S_n(\kappa,\omega)$.

Finally we would like to discuss $S(\kappa,\omega)$ for ^{4}He I in the general context of liquid dynamics, in order to elucidate the basic problem of what requirements are to be satisfied in the liquid state, if short-wavelength propagating modes are to be sustained. Our starting point is here to note that our data for ^{4}He I look very similar to $S(\kappa,\omega)$ for liquid Rb, where also a dispersion curve $\Omega(\kappa)$ could be identified somewhat beyond the maximum of the dispersion curve, as opposed to liquid Ar, where no wave propagation takes place for $\kappa > 0.2$ Å^{-1}. If we follow the theory of phonons in crystals, the starting point is the pair potential $U(r)$, which may be expanded around the minimum value $-\epsilon_0$ at the distance r_0:

$$U(r) = \epsilon_0\left[-1 + \tfrac{1}{2}\alpha_2\left(\frac{r}{r_0}-1\right)^2 - \tfrac{1}{6}\alpha_3\left(\frac{r}{r_0}-1\right)^3 + \cdots\right], \tag{4}$$

where $\alpha_n = (-1)^n \epsilon_0^{-1} r_0^n [\partial^n U(r_0)/\partial r^n]$. Hence α_2 and α_3 characterize the harmonic and anharmonic part of the potential. It is also well known that phonon damping depends on thermal population and hence we define the reduced temperature $t = k_B T/\epsilon_0$, where T is the temperature where the scattering experiments have been made. ϵ_0, r_0, and t are given in Table I for the liquids Ar (Ref. 12) and Rb (Ref. 13) together with the ratio α_3/α_2 for liquid Ar and liquid Rb. Also given is the answer to the question: "Are well-defined collective excitations observed; i.e., is $\Omega(\kappa) > 0$ well above $\kappa = 0.2$ Å^{-1}?" It is obvious from Table I that the Rb potential is substantially less

TABLE I. Some characteristics of the liquids ^{4}He, Ar, and Rb of importance for the short-wavelength collective dynamics in the liquid state. The parameters are defined in the text.

	^{4}He	Ar	Rb
ϵ_0/k_B(K)	10	120	403
r_0(Å)	2.90	3.83	4.94
$\frac{1}{6}\alpha_3/\alpha_2$	$\cdots$	3.50	1.43
γ_G	3.1	2.6	1.9
t	0.31	0.72	0.87
Well-defined excitations?	yes	no	yes

anharmonic than the Ar potential consistent with the fact that collective excitations are observed in Rb but not in Ar. Although no reliable pair potential exists for ^{4}He we show the generally accepted Lennard-Jones parameters.[12] Also given in Table I are the experimental Grüneisen parameters γ_G for the solids ^{4}He (Ref. 14), Rb (Ref. 15), and Ar (Ref. 16). According to Lewis and Lovesey[17] $\gamma_G = \frac{1}{6}\alpha_3/\alpha_2$ must be less than 2 for well-defined collective excitations to exist. This is in agreement with what is observed for Rb and Ar, whereas ^{4}He does not follow this rule. On the other hand, t is much lower for ^{4}He than for Rb and Ar.

The conclusion from Table I is the following. In classical liquids (e.g., Rb and Ar) t is large. Here the anharmonicity determines whether propagating modes exist in a liquid, whereas at low t such as in ^{4}He modes are sustained in spite of an appreciable anharmonicity. Our analysis shows that despite the lack of stringent foundation of the lattice dynamical theory of phonons in the liquid state, this theory provides a useful guide into the dynamics of liquids.

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