Overdamped Phonons in Fluid Helium at 4 K

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From inelastic neutron scattering we determine the dynamic structure factor $S(q, \omega)$ of fluid helium at 4 K and 1 bar as a function of frequency ω for wave numbers $2.5 < q < 30 \text{ nm}^{-1}$. $S(q, \omega)$ is completely given by only two phonon contributions located at the sound propagation frequencies $\pm \omega_s(q)$. For $18 < q < 21 \text{ nm}^{-1}$ they are overdamped, i.e., $\omega_s(q) = 0$.

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The behavior of elementary microscopic excitations in dense monatomic fluids in equilibrium has been extensively studied by means of inelastic neutron scattering experiments. In such experiments one determines the dynamic structure factor $S(q, \omega)$ as a function of wave number q and frequency ω from which one derives the detailed properties of the microscopic fluid excitations [1]. Obviously there are two very different extreme cases: superfluid helium and classical liquids. In this Letter we discuss how the behavior of superfluid helium could be connected to that of classical liquids by considering a quantum fluid which is not superfluid, i.e., ^4He at 4 K.

For temperatures T below the λ -transition temperature T_λ ($= 2.17 \text{ K}$ at STP), $S(q, \omega)$ of superfluid helium consists of *two* sharp contributions located at the frequencies $\pm \omega_s(q)$ [2–4]. Here $\omega_s(q)$ is the well-known dispersion curve of phonons in superfluid helium. For $q < 3 \text{ nm}^{-1}$, $\omega_s(q) = cq$ with c the adiabatic speed of sound (due to hydrodynamic sound excitations), for $q = q_M \sim 10 \text{ nm}^{-1}$, $\omega_s(q)$ has a maximum (maxon excitations), and for $q = q_R \sim 20 \text{ nm}^{-1}$ $\omega_s(q)$ shows a minimum (roton excitations) with a minimum frequency $\omega_s(q_R) > 0$ that is strictly positive for all superfluid states.

On the other hand, there are classical fluids like Rb [5], Kr [6], Ar [7,8], Ne [9], and He [10] at $T \gtrsim 30 \text{ K}$. Then $S(q, \omega)$ consists of *three* contributions. There is one strong central line in $S(q, \omega)$ that is due to the microscopic cage diffusion process [11]. For $q \rightarrow 0$, this central line tends continuously to the macroscopic Rayleigh line of hydrodynamics that is caused by heat diffusion in the fluid [1]. In addition, there are two weak side lines in $S(q, \omega)$, located at $\pm \omega_s(q)$, that are due to the extended sound modes of the fluid. For $q \rightarrow 0$ these contributions tend continuously to the Brillouin lines of hydrodynamics, caused by the propagation of sound, located at $\pm \omega_s(q) = cq$. Usually for classical liquids [7,9] one finds a finite q region near $q \approx 20 \text{ nm}^{-1}$, where $\omega_s(q) = 0$ (so-called “overdamped phonons”).

In this Letter we present the results of an inelastic neutron scattering experiment on liquid helium at tempera-

ture $T = 4.0 \text{ K}$ and pressure $p = 1.0 \text{ bar}$ (number density $n = 19.51 \text{ nm}^{-3}$). We obtain the dynamic structure factor $S(q, \omega)$ for $2.5 \leq q \leq 30 \text{ nm}^{-1}$. For all q , $S(q, \omega)$ is completely given by only two phonon contributions located at the sound propagation frequencies $\pm \omega_s(q)$. For $18 \leq q \leq 21 \text{ nm}^{-1}$ the phonons are overdamped, i.e., $\omega_s(q) = 0$. Thus, helium at 4 K and 1 bar is different from both superfluid helium (where there are no overdamped phonons) as well as classical liquids (where there is a central contribution from “cage diffusion”). In this peculiar way helium at 4 K is intermediate between a superfluid and a classical fluid.

Our inelastic neutron scattering experiment on helium at 4 K and 1 bar was performed on the time-of-flight spectrometer RKS at the Interfaculty Reactor Institute in Delft. The wavelength of the neutrons in the incident beam (cross section $2.5 \times 10 \text{ cm}^2$) was 0.240 nm . Time-of-flight spectra were collected in 75 detectors at scattering angles $4.4^\circ \leq \phi \leq 86^\circ$. The sample holder was a single aluminum cylinder with height 10 cm , diameter 2.8 cm , and wall thickness 0.6 mm . The spectra were corrected for background, detector efficiency, and interpolated from constant ϕ to constant q . The ω resolution of the spectrometer is $\Delta\omega = 1.3 \text{ ps}^{-1}$ (FWHM).

The results for $S(q, \omega)$ are shown in Fig. 1 as a function of ω , for $q = 7, 10.5, 14, 21$, and 28 nm^{-1} . The spectra obey the detailed balance condition $S(q, -\omega) = \exp(-\beta\hbar\omega)S(q, \omega)$ with $\beta = 1/k_B T$ and $\hbar = h/2\pi$ and, therefore, are not symmetric around $\omega = 0$ (cf. Fig. 1). We analyze $S(q, \omega)$ in terms of the symmetrized dynamic structure factor $\chi(q, \omega)$ defined by [12,13]

$$\chi(q, \omega) \equiv \frac{1 - \exp(-\beta\hbar\omega)}{\beta\hbar\omega} S(q, \omega), \quad (1)$$

so that $\chi(q, -\omega) = \chi(q, \omega)$. In Fig. 2 we show $\chi(q, \omega)$, corresponding to the $S(q, \omega)$ in Fig. 1. From Eq. (1) follows that $\chi(q, 0) = S(q, 0)$ but $\chi(q, \omega)$ and $S(q, \omega)$ become very different as $|\omega|$ increases (cf. Fig. 1 and Fig. 2), which is a typical quantum effect [for $\beta\hbar \rightarrow 0$, $\chi(q, \omega) = S(q, \omega)$].

FIG. 1. Dynamic structure factor $S(q, \omega)$ of He (4 K, 1 bar) as a function of ω for $q = 7, 10.5, 14, 21$, and 28 nm^{-1} . Experiment (error bars) and best model fit (solid line). The model is convoluted with the experimental resolution function.

FIG. 2. Symmetrized dynamic structure factor $\chi(q, \omega)$ of He (4 K, 1 bar) as a function of ω for $q = 7, 10.5, 14, 21$, and 28 nm^{-1} . Experiment (error bars) and best model fit (solid line). The model is not convoluted with the experimental resolution function.

In Fig. 3 we show the static structure factor $S(q) = \int_{-\infty}^{\infty} d\omega S(q, \omega)$. The overall shape of $S(q)$ is much like that in dense classical fluids including the typical maximum in $S(q)$ at $q = 20 \text{ nm}^{-1}$. We note that for the smallest observed q values, $S(q)$ is still far from its asymptotic value $S(0) = 0.43$, obtained from the compressibility theorem, $S(0) = k_B T (\partial n / \partial p)_T$.

In Fig. 3 we also show the response function $\chi(q) = \int_{-\infty}^{\infty} d\omega \chi(q, \omega)$. We remark that for small q , $\chi(q) \approx S(q)$ (cf. Fig. 3) and that $\chi(0) = S(0)$. This is due to the fact that for decreasing q , $S(q, \omega)$ becomes increasingly narrow in ω so that $\chi(q, \omega) = S(q, \omega)$ for $q \rightarrow 0$. The large difference between $\chi(q)$ and $S(q)$ seen in Fig. 3 for finite q is typical for a quantum fluid [for classical fluids $\chi(q) = S(q)$ for all q].

We express the ω dependence of $\chi(q, \omega)$ in terms of the complex, symmetric 2×2 coupling matrix [13],

$$H(q) = \begin{pmatrix} 0 & i f_{un}(q) \\ i f_{un}(q) & z_u(q) \end{pmatrix}, \quad (2)$$

where $f_{un}(q) = [\beta m \chi(q)]^{-1/2} q$ describes the coupling between microscopic density (n) and longitudinal momen-

tum (u) fluctuations (with m the mass of a helium particle), and where $z_u(q)$ is the decay rate of momentum fluctuations, for which explicit expressions are given in the literature [13]. Then one has

$$\chi(q, \omega) = \frac{\chi(q)}{\pi} \text{Re} \left\{ \frac{1}{i\omega + H(q)} \right\}_{11}. \quad (3)$$

We find that our experimental data for $\chi(q, \omega)$ and $S(q, \omega)$ [via Eq. (1)] are for all q very well represented by the 2×2 coupling model of Eqs. (2) and (3).

The best fits are shown in Figs. 1 and 2 as full curves. In the comparison of Fig. 1 we have convoluted the model of Eq. (3) with the experimental resolution function, yielding a perfect fit. The results for $f_{un}(q)$ and $z_u(q)/2$ are shown in Fig. 4(a) as functions of q . We note that the 2×2 matrix $H(q)$ of Eq. (2) describes for all q the time evolution of a damped harmonic oscillator, i.e., $(\dot{x}(t), \dot{y}(t)) = -H(x(t), y(t))$ with $x(t)$ the deviation at time t , $y(t) = i\dot{x}(t)/f_{un}$ the reduced velocity, f_{un} the reduced spring constant, and z_u the reduced damping rate. Hence oscillatory solutions in time exist when $f_{un} > z_u/2$, while overdamping occurs when $f_{un} < z_u/2$.

FIG. 3. $S(q)$ (closed circles) and $\chi(q)$ (crosses) of He (4 K, 1 bar) as functions of q . The arrow points to $S(0) = \chi(0) = 0.43$.

The representation of $\chi(q, \omega)$ given by Eq. (3) is in fact a description in terms of two separate phonon contributions, as can be seen as follows. The coupling matrix $H(q)$ has two eigenvalues $\lambda_j(q)$ ($j = \pm$) given by

$$\lambda_{\pm}(q) = \frac{z_u(q)}{2} \pm \sqrt{\frac{z_u(q)^2}{4} - f_{un}(q)^2}, \quad (4)$$

so that the phonon (sound) propagation frequency $\omega_s(q) = |\text{Im}\lambda_j(q)|$ and the phonon (sound) damping $\Gamma_{s,j}(q) = \text{Re}\lambda_j(q)$ with $j = \pm$.

When $f_{un}(q) > z_u(q)/2$ [cf. Fig. 4(a)], the eigenvalues $\lambda_j(q)$ are complex and each others complex conjugates. Then Eq. (3) implies a description of $\chi(q, \omega)$ in terms of two propagating contributions located at $-\omega_s(q)$ and $+\omega_s(q)$, with equal half-width $\Gamma_s(q) = \Gamma_{s,+}(q) = \Gamma_{s,-}(q) = z_u(q)/2$, i.e.,

$$\begin{aligned} \chi(q, \omega) &= \frac{\chi(q)}{\pi} \frac{2\Gamma_s(q)[\omega_s(q)^2 + \Gamma_s(q)^2]}{[\omega^2 - \omega_s(q)^2 - \Gamma_s(q)^2]^2 + 4\omega^2\Gamma_s(q)^2} \\ &= \frac{\chi(q)}{2\pi} \left(\frac{2\Gamma_s(q) + \omega\Gamma_s(q)/\omega_s(q)}{\Gamma_s(q)^2 + [\omega + \omega_s(q)]^2} \right. \\ &\quad \left. + \frac{2\Gamma_s(q) - \omega\Gamma_s(q)/\omega_s(q)}{\Gamma_s(q)^2 + [\omega - \omega_s(q)]^2} \right). \end{aligned} \quad (5)$$

One observes these two separate contributions clearly in Fig. 2 for $q = 7, 10.5$, and 28 nm^{-1} . We remark that Eq. (5) for $\chi(q, \omega)$ is exactly the damped harmonic oscillator model which is extensively used to describe the dynamics of superfluid helium [cf. Eqs. (6) and (7) of Ref. [2], and Eqs. (1) and (2) of Ref. [4]].

When $f_{un}(q) \leq z_u(q)/2$ [cf. Fig. 4(a)], the eigenvalues $\lambda_j(q)$ are real. Then $\chi(q, \omega)$ is given by two central Lorentz lines with different widths $\Gamma_{s,j}(q) = \lambda_j(q)$ ($j =$

FIG. 4. Frequencies as functions of q : $z_u(q)/2$ [error bars in (a)], $f_{un}(q)$ [crosses in (a)], $\omega_s(q)$ [error bars in (b)], and $\Gamma_{s,j}(q)$ [error bars in (c)]. The solid straight line in (b) is $\omega_s(q) = cq$. For $18 \leq q \leq 21 \text{ nm}^{-1}$, $f_{un} < z_u/2$, $\omega_s(q) = 0$, and $\Gamma_{s,j}(q)$ ($j = \pm$) is double valued. The solid line in (c) is the half-width of an equivalent hard sphere fluid.

$\pm$), i.e.,

$$\chi(q, \omega) = \frac{\chi(q)}{\pi} \sum_{j=\pm} \frac{A_j(q)\Gamma_{s,j}(q)}{\Gamma_{s,j}(q)^2 + \omega^2} \quad (6)$$

and amplitudes $A_{\pm} = \{1 \mp [1 - (2f_{un}/z_u)^2]^{-1/2}\}/2$. This central behavior of $\chi(q, \omega)$ is clearly seen in Fig. 2 for $q = 21 \text{ nm}^{-1}$.

The eigenvalues $\lambda_j(q)$ of $H(q)$ are plotted in Fig. 4 as a function of q . The imaginary part $\omega_s(q)$ is for $q \leq 4 \text{ nm}^{-1}$ given by $\omega_s(q) = cq$, with $c = 184.3 \text{ ms}^{-1}$ the adiabatic speed of sound; for $4 < q < 11 \text{ nm}^{-1}$, $\omega_s(q) > cq$ (so-called “anomalous dispersion” [14,15] while for $18 \leq q \leq 21 \text{ nm}^{-1}$, $\omega_s(q) = 0$ (overdamped phonons) [cf. Fig. 4(b)]. The real parts $\Gamma_{s,j}(q)$ are equal for $q \leq 18 \text{ nm}^{-1}$ and $q \geq 21 \text{ nm}^{-1}$ and given by $\Gamma_s(q) = z_u(q)/2$. In the overdamped region $18 \leq q \leq 21 \text{ nm}^{-1}$ of $\omega_s(q)$, the two eigenvalues $\lambda_j(q)$ of $H(q)$ are real and different [cf. Fig. 4(c)].

We end with a number of remarks.

(1) The most salient difference between He at 4 K and classical fluids is the complete absence of a central line in $S(q, \omega)$ for $2.5 < q < 30 \text{ nm}^{-1}$. From hydrodynamics [1] follows that eventually, for $q \rightarrow 0$, $S(q, \omega)$ of He at 4 K and 1 bar must be given by the Rayleigh-Brillouin triplet. Then $S(q, \omega)$ consists of one central Rayleigh line with area $S(0)(\gamma - 1)/\gamma$ and two Brillouin lines located at $\pm\omega_s(q) = \pm cq$ each with area $S(0)/2\gamma$. Here $\gamma = 3_p/c_v = 1.76$ is the ratio of specific heats at constant pressure and volume. Hence the central Rayleigh line, with 43% of the total area, must be prominently present in $S(q, \omega)$ for $q \rightarrow 0$ but disappears from the spectrum at a wave number q_c with $q_c < 2.5 \text{ nm}^{-1}$.

(2) Woods, Svensson, and Martel [14] have measured $S(q, \omega)$ of He at 4.2 K and saturated vapor pressure for $1 \leq q \leq 22 \text{ nm}^{-1}$. They observe the Rayleigh-Brillouin triplet in $S(q, \omega)$ directly for $q = 1 \text{ nm}^{-1}$ but not for $q > 2 \text{ nm}^{-1}$, consistent with the present results.

(3) A striking difference between superfluid helium and helium at 4 K is the behavior of $\omega_s(q)$ for q near the roton wave number $q_R = 20 \text{ nm}^{-1}$. For superfluid helium $\omega_s(q_R)$ is strictly positive, while at 4 K, $\omega_s(q_R) = 0$. Dietrich *et al.* [16] have analyzed $S(q, \omega)$ for $q = q_R$ and temperatures T near T_λ , albeit in a different manner. They find a strong decrease of $\omega_s(q_R)$ when T approaches T_λ from below, hinting at a relationship between the vanishing of $\omega_s(q_R)$ and that of superfluidity.

(4) The half-widths $\Gamma_{s,j}(q)$ show a minimum $\Gamma_{s,-}(q) = 1.7 \text{ ps}^{-1}$ at $q = q_R = 20 \text{ nm}^{-1}$ [cf. Fig. 4(c)] equal to that of a classical fluid of hard spheres with diameter $\sigma = 0.28 \text{ nm}$ [11]. For classical fluids the minimum in the half-width is due to the "De Gennes-narrowing" caused by the cage effect. Thus in the overdamped phonon region near $q_R = 20 \text{ nm}^{-1}$, He at 4 K shows a behavior similar to classical particles locked up in cages of size $2\pi/q_R \approx \sigma = 0.28 \text{ nm}$.

(5) For $2.5 \leq q \leq 4 \text{ nm}^{-1}$ we find that $\omega_s(q) = cq$ [cf. Fig. 4(b)]. It is not clear why this hydrodynamic relation holds in a q region where hydrodynamics is not valid [i.e., $S(q) \neq S(0)$, cf. Fig. 3]. Possibly, the hydrodynamic relation $c = [\gamma/\beta m S(0)]^{1/2}$ can be generalized to finite q , i.e., $c = [\gamma(q)/\beta m S(q)]^{1/2}$ with constant $\gamma(q)/S(q)$. This implies that a decrease of $S(q)$ from $S(0) = 0.43$ causes a decrease of $\gamma(q)$ from $\gamma(0) = \gamma = 1.76$. Indeed, the contribution of the central Rayleigh line $S(q)[\gamma(q) - 1]/\gamma(q)$

then vanishes at $q_c < 2.5 \text{ nm}^{-1}$, where $\gamma(q_c) = 1$. Further experiments for $q < 2.5 \text{ nm}^{-1}$ are needed to verify this assumption.

(6) For $4 < q < 11 \text{ nm}^{-1}$, $\omega_s(q) > cq$ so-called anomalous dispersion. This effect is observed in both superfluid helium [14,15] and ^3He [17], as well as in classical liquids [7,9] and is, therefore, a quite common phenomenon.

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