SUM RULES, STRUCTURE FACTORS, AND PHONON DISPERSION IN LIQUID He^4 AT LONG WAVELENGTHS AND LOW TEMPERATURES*

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Combining simple sum-rule arguments and a calculation of departures from the Feynman excitation spectrum, we demonstrate the existence of an inflection point in the liquid structure factor S_q, and relate S_q to the phonon dispersion coeffieient γ and the strength and average frequency of the multiparticle excitation spectrum in liquid He^4. We find in particular $\gamma \cong 0$, a result recently obtained by Woods and Cowley, to be in accord with other parameters determined by x-ray and neutron-scattering measurements.

Hallock[1] has recently reported an x-ray scattering measurement of the liquid structure factor, S_q, at very long wavelengths and low temperatures. His data, of greater accuracy and extending to smaller wave vectors than previous measurements, provide experimental confirmation for the hitherto theoretically conjectured inflection point in S_q.[2-4] On the other hand, Woods and Cowley,[5] using neutron-scattering techniques, measured the single-phonon part of S_q, the dispersion of the phonon spectrum, as well as the average multiparticle excitation energy $\overline{\omega}$. We have present simple sum-rule arguments for the existence of an inflection point, and show how these may be combined with a calculation[6] of departures from the Feynman excitation spectrum and experimental data to yield an estimate of phonon dispersion and of the strength of the multiparticle excitation spectrum in liquid He^4. We find

$$\gamma \cong 0, \tag{1}$$

a result reported by Woods and Cowley,[5] consistent with the experimental results on S_q.

The relevant sum rules for the dynamic structure factor $S(\vec{q}, \omega)$ are as follows[7]:

$$\int_0^\infty d\omega S(\vec{q}, \omega) = NS_q, \tag{2}$$

$$\int_0^\infty d\omega \omega S(\vec{q}, \omega) = \frac{Nq^2}{2m}, \tag{3}$$

$$\int_0^\infty \frac{d\omega}{\omega} S(\vec{q}, \omega) = \frac{-\chi_q}{2}, \tag{4}$$

where χ_q is the static wave-vector–dependent density-density response function. In working with the sum rules it is useful to divide the contributions to the dynamic structure factor into those coming from single-particle excitation from the condensate and multiparticle excitations, as was first done by Miller, Pines, and Nozières.[2] In the long-wavelength limit, we may write

$$S(\vec{q}, \omega) = Z_q \delta(\omega - \omega_q) + \xi (q/ms)^4 f(\omega), \tag{5}$$

where Z_q and ω_q are the strength and energy of the single-particle excitations (presumed to possess series expansions in integral powers of q), and ξ measures the strength of the multiparticle excitations from the condensate, which contributes terms of order q^4 to all three sum rules in this limit. We further argue that χ_q possess a series expansion in powers of q^2 which takes the form

$$\chi_q = \frac{-N}{ms^2}\left[1 - \chi_2\left(\frac{q}{ms}\right)^2 + \cdots\right] \tag{6}$$

(where χ_2 is positive), a result which follows from the assumption that only short-range correlations are present in the density-density correlation function. On making use of the sum rules, Eqs. (2)-(4), and Eq. (6), we obtain directly the following long-wavelength expansions:

$$Z_q = \frac{Nq}{2ms}\left[1 + z_2\left(\frac{q}{ms}\right)^2 + \cdots\right], \tag{7}$$

$$\omega_q = sq\left[1 + \omega_2\left(\frac{q}{ms}\right)^2 + \cdots\right], \tag{8}$$

$$\chi_q = \frac{-N}{ms^2}\left[1 + (z_2 - \omega_2)\left(\frac{q}{ms}\right)^2 + \cdots\right], \tag{9}$$

$$S_q = \frac{q}{2ms}\left[1 + z_2\left(\frac{q}{ms}\right)^2 + \xi\left(\frac{q}{ms}\right)^3 + \cdots\right]. \tag{10}$$

Note that since $\omega_2 = -\gamma(ms)^2$ is from all experimental indications zero or negative, if χ_2 is positive, Z_2 must be negative. And since ξ must be positive, one will always expect a point of inflection in the static structure factor S_q.

If further we write

$$\omega_q = \frac{q^2}{2mS_q}\left[1 + C_2\left(\frac{q}{ms}\right)^2 + C_3\left(\frac{q}{mc}\right)^3 + \cdots\right], \quad (11)$$

we find by comparison

$$C_2 = \omega_2 + z_2 = -\overline{\omega}\xi/ms^2, \quad (12)$$

where $\overline{\omega}$ is the average multiparticle excitation energy in the long-wavelength limit, defined by

$$\overline{\omega} = \int_0^\infty \omega f(\omega) d\omega / \int_0^\infty f(\omega) d\omega. \quad (13)$$

In Ref. 6, C_2 was calculated to a considerable degree of accuracy to yield a series whose leading terms are −1.04 and −0.30. Assuming a geometric progression, we find $C_2 = -1.46$. From Ref. 5, we find

$$\omega_2 = 0, \quad (14)$$

$$\overline{\omega} = 20°\mathrm{K}. \quad (15)$$

Hence,

$$z_2 = -1.46, \quad (16)$$

and

$$\xi = -C_2 m s^2/\overline{\omega} = 2.0. \quad (17)$$

Substituting these results into Eqs. (7) and (10), we find Z_q and S_q in good agreement with the data of Woods and Cowley[5] and Hallock[1] at long wavelengths (≲0.4 Å^{-1}). Beyond 0.4 Å^{-1}, higher orders in q than those considered here become important and must be taken into account. It should be noted that the previously accepted value of γ (3×10^{37} g^{-2} cm^{-2} sec^2) corresponds to $\omega_2 = -0.77$, and leads to large discrepancies between the calculated and experimental results for Z_q and S_q.

Further information is provided by the ω^3 sum rule[8]:

$$\int_0^\infty \omega^3 S(\vec{q}, \omega) d\omega = \frac{Nq^2}{2m}\left[\left(\frac{q^2}{2m}\right)^2 + 4\left(\frac{q^2}{2m}\right)\frac{\overline{E}_{\mathrm{kin}}}{N} + \frac{\rho}{m}\int (1-\cos\vec{q}\cdot\vec{r}) g(r) \frac{(\vec{q}\cdot\vec{\nabla})^2}{q^2} v(r) d\vec{r}\right]. \quad (18)$$

For the Lennard-Jones 6-12 potential and long wavelengths it reduces[9] to

$$\frac{s^2}{2m} + \overline{\omega}^3 \frac{\xi}{2(ms)^4} = \left(\frac{1}{2m}\right)^2\left[\frac{272}{15}\frac{\overline{E}_{\mathrm{kin}}}{N} - \frac{144}{5}\frac{E}{N}\right]. \quad (19)$$

Using the experimental value of −7.14°K for E/N, and a recent theoretical estimate[10] of 14.3°K for $\overline{E}_{\mathrm{kin}}/N$, we find

$$(\overline{\omega^3})^{1/3} = 42.3°\mathrm{K}. \quad (20)$$

Equations (15) and (20) indicate that the multiparticle contribution to the dynamic structure factor, or $f(\omega)$, peaks toward higher frequencies.

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