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Small-angle scattering of neutrons on normal and superfluid liquid helium

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Experiments on small-angle scattering of neutrons on liquid helium at temperatures of 1.0–5 K at the ISIS pulsed neutron source (England) are reported. Detailed measurements of the angular distribution of the scattered neutrons reveal a significant change in the temperature dependence of the second moment of the pairwise correlation function (the first derivative of the angular distribution for small scattering angles). At high temperatures the angular distribution of the scattered neutrons follows the classical description for small-angle scattering, but at temperatures below the λ-point, quantum mechanical behavior is observed (scattering of neutrons on quantum fluctuations). It is confirmed experimentally that over the entire temperature range the neutron scattering cross section at zero angle is determined by classical thermodynamic fluctuations in the density. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4821758]

Introduction

Neutrons are well known to be ideally suited for studies of excitations in condensed matter and their structure since the wavelengths of thermal neutrons, which range from 0.5 to 10 Å, are comparable to the interatomic distances and their energies are of the same order of magnitude as the elementary excitations. For example, experiments on the inelastic scattering of neutrons in liquid helium have helped establish many properties of this quantum liquid, such as the excitation spectrum advanced theoretically by Landau, the character of the pulsed distribution of helium atoms in the normal and superfluid phases, and the relative fraction of helium atoms in a Bose condensate.[1–3] On the other hand, measurements of elastic scattering of neutrons in liquid helium over a wide range of angles by Svensson *et al.*,[4] have made it possible to measure the structure factor over a wide temperature interval. In particular, it has been shown that the position of the main peak in the structure factor $S(Q,T)$ (here Q is the scattering momentum, which is related to the scattering angle θ by $|Q| = (4\pi/\lambda)\sin(\theta/2))$ is essentially invariant, and its absolute magnitude only changes slightly (by 6%) on passing through the λ-point.

These measurements[4] were made in the $Q > 1$ Å^{-1} range; i.e., the small-angle neutron scattering (SANS) was not measured. For small angles, the authors of Ref. 4 used x-ray scattering data[5] obtained at several temperatures, 4.6, 4.0, 3.3, 1.45, and 0.38 K. The only SANS experiment was done quite some time ago, in 1953, by Egelstaff and London.[6] There[6] the measurements were made only for a few temperatures and with a quite large statistical error, so only qualitative conclusions could be drawn regarding the nature of SANS before and after the superfluid transition. Of course, the quality of the data can be greatly improved using modern nuclear reactors and pulsed accelerator neutron sources (spallation sources) along with the associated equipment.

The absence of experimental data on SANS in liquid helium has led us to make some detailed measurements at a modern level. A detailed study of SANS in liquid helium was undertaken by one of us (Yu.M.Ts.) at ILL (France).[7] However, these results were criticized from an experimental standpoint, since the experiments were done with ordinary helium and it was entirely possible that solid hydrogen and neon, which could distort the data significantly, were present. In addition, there was evidently some microscopic contamination on the walls of the cryostat, and this could also increase the background for the small Q being observed in the experiments.

It should be noted that small-angle scattering offers the possibility of getting information on the character of thermal fluctuations in a liquid. Small-angle elastic scattering of neutrons is, in fact, scattering on "prepared" quasiparticles. In helium at finite temperatures, phonons and rotons are excited thermally; in helium at temperatures from 1 to 2.2 K rotons are predominantly excited and above the superfluid transition, phonons. The "size" of the rotons, as opposed to that of the phonons, does not depend on temperature (all have the same quasimomentum and energy), so that it is possible to hope that this will show up in some way in SANS. Burkova and Pitaevskii[8] made an estimate of the roton size based on the Feynman wave function[9] that gave a fully satisfactory description of the spectrum of the excitations.

Experiment

These experiments were done on an SANS2D system with the ISIS pulsed neutron source. Spectroscopically pure helium was condensed in a thin-walled aluminum working vessel with a volume of 20 cm^3 using a standard system for filling helium cryostats from the firm Various Oxford Instruments. The temperature was stabilized to within ± 10 mK. Measurements were made over temperatures from 1 to 5 K. The wavelengths of the incident neutrons ranged from 2–12 Å.

We paid special attention to the reliability of the data. To account for background (neutron scattering on the cryostat walls), measurements were made with the working volume empty at various temperatures. These measurements showed that the background had no observable temperature dependence. In addition, in order to confirm that scattering on bubbles formed during boiling of the helium is negligible, SANS measurements were made at 4.4 K with the equilibrium vapor pressure, as well as at an elevated pressure (where boiling is absent). No difference in the radial distributions of the scattered neutrons was detected.

In discussing small-angle scattering it is always assumed that neutron scattering is elastic, but for scattering in liquids this is not strictly so. The Placzek correction, which takes the inelastic nature of the scattering into account, must be applied to the data, but it has not been calculated for superfluid helium. Thus, the characteristics of the angular distributions obtained at a fixed temperature but with different wavelengths of the incident neutrons (from 2 to 12 Å) were compared. With an accuracy of order 5%, no difference (i.e., no contribution from inelastic processes) was detected in these distributions.

Theoretical description of small-angle scattering

One of the basic characteristics for describing a liquid is the static structure factor $S(Q)$, which is a measure of the correlation between the positions of the atoms. This quantity is directly related to the macroscopic neutron scattering cross section $d\Sigma/d\Omega$, as

$$\frac{d\Sigma}{d\Omega}(Q) = \frac{\sigma_b}{4\pi}\rho_{\rm at}S(Q), \tag{1}$$

where σ_b is the cross section for scattering of neutrons on a bound helium atom (=1.172 b) and $\rho_{\rm at}$ is the density of helium.

Goldstein has shown[10] that for finite temperatures and low Q, the structure factor can be written in the form of the series

$$S(Q) = n_0 kT\chi_T + \sum_{n=1}^{\infty}(-1)^n Q^{2n} r_g^{2n}[(2n+1)!]^{-1}, \tag{2}$$

where n_0 is the concentration of helium atoms, χ_T is the isothermal compressibility, and r_g^{2n} is the even moment of the pairwise correlation function $g(r)$,

$$r_g^{2n} = \int r^{2n} g(r) d^3 r. \tag{3}$$

Small-angle scattering is scattering with small amounts of momentum transfer Q. In that case, Eq. (2) takes the form

$$S(Q) \simeq n_0 kT\chi_T - r_g^2\frac{Q^2}{6}. \tag{4}$$

The first term in this formula, $S(0)$, describes the scattering cross section for angle of 0° and is determined by purely thermodynamic hydrodynamic fluctuations in the liquid, i.e., such that the liquid can be treated as a continuous medium. Here we are interested in the second term in this expression.

The resulting expression for the static structural factor is valid for small Q and is analogous to the well known approximation of Guinier[11] for the angular distribution of the intensity of neutrons scattered on an ensemble of spatially randomly distributed particles,

$$I(Q) = I(0)\exp(-Q^2R_g^2/3) \simeq I(0)(1 - Q^2R_g^2/3). \tag{5}$$

If all the particles are spheres with the same radius R_0, then the parameter R_g in Eq. (5) (referred to as the radius of inertia) is equal to $3R_0^2/5$. It is, therefore, clear that the mean square radius of the pairwise correlation function of the liquid is $r_g^2 = R_g^2/2$.

Experimental data and analysis

According to Eqs. (4) and (5), the scattered neutron intensity I (like the static structure factor) depends quadratically on Q; this, of course, most be verified. Figure 1 is a plot of the experimentally determined neutron scattering cross section (the radial intensity distribution) as a function of the square of the momentum transfer for several temperatures (1, 2.1, 4.2, and 4.6 K). The data represent the difference between the intensities of scattering on the cryostat filled with liquid helium and the empty cryostat. It should be noted that the neutron scattering cross section in helium is rather small and, at low temperatures the scattering on helium becomes comparable to the scattering on the cryostat walls; this leads to large errors in the determination of the neutron scattering cross section for helium.

A quadratic dependence is actually found, but, as can be seen in the figure, this occurs within a very small range of Q. The derivative dS/dQ for small Q is $-(r_g^2/3)Q$, and is easily calculated from the linear segment of the $I(Q^2)$ curve.

It is natural, first of all, to see how $S(0)$ behaves as the temperature is varied. Equation (4) implies that there should be a linear dependence, with $S(0) = 0$ at zero temperature. In fact, as Fig. 2 shows, the experiment confirms this dependence quite well.

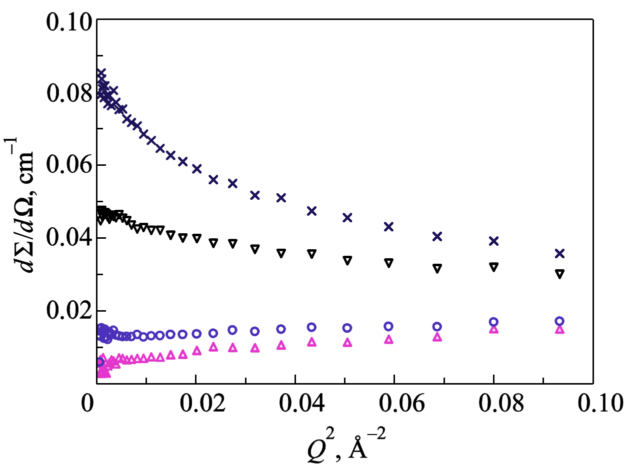

FIG. 1. The radially averaged neutron scattering cross section (cm^{-1}) as a function of the square of the momentum transfer at different temperatures T (K): 1 (Δ), 2.1 (○), 4.2(∇), 4.6 (×).

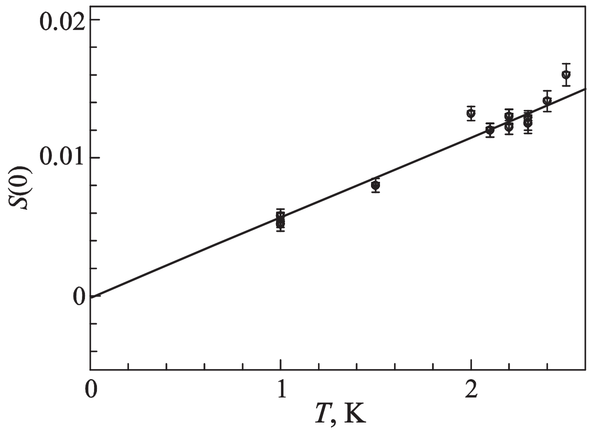

FIG. 2. The static structure factor at zero angle as a function of temperature. A linear least squares fit is drawn through the experimental points.

This result is of fundamental importance, since the validity of a classical description of thermodynamic fluctuations in a quantum liquid has only been demonstrated theoretically by Price.[12]

It can be seen from Fig. 1 that the slope of the curves changes sign at temperatures below the λ-point. Figure 3 shows the results of a linear approximation for $I(Q^2)$ and, in fact, the derivative $dI/dQ^2(T)$ is negative for $T < 3$ K.

That figure shows the temperature dependence of the mean square radius of the pairwise correlation function for liquid helium (the derivative of the scattered intensity with respect to the square of the momentum transfer). A second order least squares polynomial fit to the experimental points is shown there for clarity.

This indicates that a description of neutron scattering in terms of scattering on thermal fluctuations at low temperatures is still not adequate. As Feynman showed,[9] at zero temperature the structure factor should depend linearly on the momentum, i.e., be given by

$$S(Q) = \hbar Q/2ms\,, \qquad (6)$$

where s is the first sound speed and m is the mass of a helium atom. This form of the structure factor $S(k)$ for the ground state changes markedly as the temperature is raised. Then the probability of finding an oscillator represented by a phonon with wave number k in its n-th state is proportional to $\exp(-E/kT)$. This implies[13] that

$$S(Q) \simeq kT/ms^2 + \hbar^2 Q^2/12mkT + \ldots. \qquad (7)$$

As $Q \rightarrow 0$ this expression transforms to

$$S(Q) = \rho kT\chi_T\,, \qquad (8)$$

i.e., it is the same as the result from the classical theory of fluctuations.[14] It should be noted that the expansion for this function given here, which is valid at elevated temperatures, does not contain a term that is linear in Q.

Therefore, the linear variation in $S(Q)$ is caused by zero-point oscillations and, as the temperature is raised, thermal fluctuations come to play an ever larger role. Our experiment

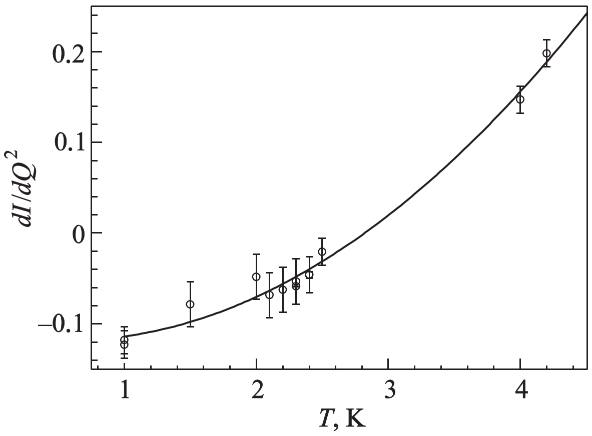

FIG. 3. The temperature variation of the mean square radius of the pairwise correlation function for liquid helium (the derivative of the scattering intensity with respect to the square of the momentum transfer). For clarity an approximation curve is drawn through the experimental points.

shows that the contribution of quantum mechanical zero-point oscillations is significant up to temperatures of $\sim$3 K.

Conclusion

It has been shown that the scattering of neutrons in liquid helium at high temperatures is well described by the classical theory as scattering on thermal oscillations and that scattering at an angle of 0° is linear over the entire range of temperatures and goes to zero at zero temperature. The experiments also indicate that as the temperature is lowered, scattering on quantum mechanical oscillations makes an ever larger contribution.

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