

High-Energy Neutron Scattering from Liquid He⁴

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(Received 8 July 1966)

An experiment is proposed using high-energy neutrons to probe the momentum distribution of helium atoms in liquid helium, and detect the presence of a zero-momentum condensate below T_λ . It is suggested that for momentum transfers to the neutron much larger than a roton momentum, the energy transfer should be equal to the recoil energy of a single helium atom, Doppler-shifted by its initial motion in the helium bath. Thus, if a finite fraction of atoms are initially in the zero-momentum state, they will contribute a peak to the spectrum of scattered neutrons. Corrections due to final-state interactions are discussed briefly and estimated.

INTRODUCTION

CONSIDER the scattering of neutrons from He⁴. If the scattering is weak (i.e., the intensity of the scattered beam is much less than the intensity of the incident beam), then the differential cross section $d\sigma/d\omega d\Omega$ for scattering with momentum transfer k and energy transfer $\hbar\omega$ is proportional to the structure factor $S(k, \omega)$ of the liquid (see Fig. 1).¹ The neutron scattering acts as a microscopic probe whose resolution has a characteristic length $\lambda_{\text{res}} \sim \hbar/k$. This length may be compared to the typical interparticle distances over which the wave function of the helium will vary, $\lambda_{\text{int}} \approx \lambda_{\text{roton}} \approx 1 \text{ \AA}$. In the long-wavelength limit ($\lambda_{\text{res}} \gg \lambda_{\text{int}}$), the scattering takes place from a large number of He⁴ atoms, and the dependence of $S(k, \omega)$ on k and ω gives information about the collective (or quasi-particle) excitations in the liquid.¹ These are the well-known phonons and rotons. In the short-wavelength limit ($\lambda_{\text{res}} \ll \lambda_{\text{int}}$) the scattering takes place from individual He⁴ atoms (bare particles). If in addition, the scattering takes place in a time which is short compared to interparticle collision times, i.e., by the uncertainty principle, if the recoil energy $k^2/2m_{\text{He}}$ is much greater than a characteristic helium atom energy ($\hbar v_{\text{thermal}}/\lambda_{\text{int}}$ or ϵ_{roton}) then the $S(k, \omega)$ can give information about the single-particle momentum distribution of helium atoms. In effect the high-energy neutron scatters from an individual helium atom, catching it between collisions, and experiences a Doppler shift due to the

initial motion of the atom in the helium bath. We propose to use neutrons in the range of 1 eV to measure the momentum distribution of helium atoms and in particular the occupancy of the zero-momentum state in He II.

A similar situation occurs in the scattering of x rays from an electron gas. This case was studied in Ref. 2. In terms of the well-known formula

$$\frac{d\sigma}{d\omega d\Omega} \propto \sum_{\mathbf{p}, \mathbf{p}'} \int dt e^{-i\omega t} \langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}+\mathbf{k}}(t) a_{\mathbf{p}'+\mathbf{k}}(0) a_{\mathbf{p}'}(0) \rangle, \quad (1)$$

it was shown in Ref. 2 that the qualitative discussion given above corresponds to the quantitative statement that the operator $a_{\mathbf{p}+\mathbf{k}}(t)$ in (1) behaves like a free-particle operator, i.e.,

$$a_{\mathbf{p}+\mathbf{k}}(t) \sim e^{-i\epsilon_{\mathbf{p}+\mathbf{k}}t/\hbar} a_{\mathbf{p}+\mathbf{k}}, \quad \epsilon_{\mathbf{p}+\mathbf{k}} = (\mathbf{p}+\mathbf{k})^2/2m_{\text{He}}. \quad (2)$$

Since the bracket in (1) denotes an average over the equilibrium ensemble for the helium at some temperature, the momenta \mathbf{p} and \mathbf{p}' will be typical helium-particle momenta, $p \sim \hbar/\lambda_{\text{int}} \sim p_{\text{roton}}$. Since $k \gg p$ or p' , the momentum $\mathbf{p}+\mathbf{k}$ corresponds to the high-energy recoiling helium atom, and assumption (2) implies the neglect of the interaction of this high-energy particle with the remainder of the helium bath. If we make this assumption and in addition neglect² energies of order $P\epsilon_{\text{roton}}$,³

$$\frac{d\sigma}{d\omega d\Omega} \propto \int \frac{d^3p}{(2\pi\hbar)^3} \delta\left(\omega - \frac{k^2}{2m_{\text{He}}} - \frac{\mathbf{p} \cdot \mathbf{k}}{m_{\text{He}}}\right) n_{\mathbf{p}}. \quad (3)$$

Here

$$n_{\mathbf{p}} = \langle a_{\mathbf{p}}^\dagger a_{\mathbf{p}} \rangle$$

is the momentum distribution of helium atoms in the interacting finite-temperature ensemble. Equation (3) states that for fixed momentum transfer \mathbf{k} ($k \gg p_{\text{roton}}$) the scattered neutrons are shifted in energy by the recoil energy of the helium atom $k^2/2m_{\text{He}}$, plus the Doppler shift $\mathbf{p} \cdot \mathbf{k}/m_{\text{He}}$, weighted by the initial momentum distribution $n_{\mathbf{p}}$ in the helium bath. Before discussing the

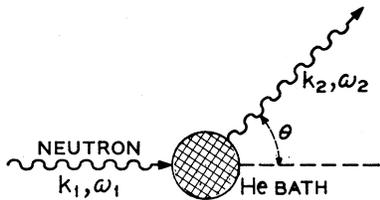


FIG. 1. Diagrammatic description of the inelastic scattering of neutrons from He.

$$\text{TAKE } k = |\mathbf{k}_1 - \mathbf{k}_2| \gg k_{\text{rot}}$$

$$\omega = \omega_1 - \omega_2 \gg \omega_{\text{rot}}$$

¹ M. Cohen and R. P. Feynman, Phys. Rev. **107**, 13 (1957).

² P. Platzman and N. Tzoar, Phys. Rev. **139**, A410 (1965).

³ G. F. Chew, Phys. Rev. **80**, 196 (1950).

applicability of formula (3) to realistic experimental situations we shall examine certain limiting cases.

THE IDEAL BOSE GAS

In this case the assumption made in (2) is exact, as is Eq. (3). The spectrum of scattered neutrons is shown in Fig. 2 at various temperatures. The zero-momentum state shows up as a delta function whose intensity is reduced at finite temperatures by depletion effects. Above the Bose condensation temperature, the delta function disappears.

NONIDEAL BOSE GAS, BOSE LIQUID FOR $k \rightarrow \infty$

In order to discuss the effect of interparticle interactions on the high-energy recoiling helium atom we shall assume that their effect is to introduce an imaginary part into the energy in (2) (lifetime effect). Since the recoiling particle has a high energy and since its scattering from the medium involves high momentum transfers we may estimate these final-state interactions by assuming that the recoiling helium atom collides independently with the other atoms in the bath. Thus

$$\text{Im} \epsilon_k \sim (k/m_{\text{He}}) \hbar n \sigma_{\text{total}}(k), \quad (4)$$

where $\sigma_{\text{total}}(k)$ is the He-He scattering cross section and n is the He atom number density.

In a Bose liquid, (He II) in the $k \rightarrow \infty$ limit, $\sigma_{\text{total}}(k)$ should decrease to a very small number (for example the cross section for He⁴ nuclear scattering $\sim 10^{-26}$ cm²) and the correction in (4) becomes negligible at high momentum transfers.⁴ In this limit Eq. (3) once again becomes applicable.³ In Fig. 3 we show schematically the spectrum of scattered neutrons in this case. There is a finite delta-function piece which is depleted even at $T=0$, and whose weight is reduced and finally disappears as the temperature is raised to the transition.

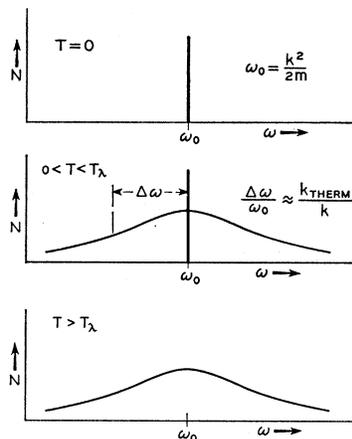
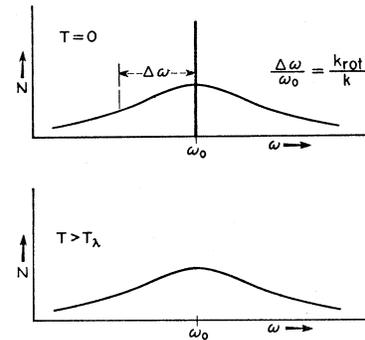


FIG. 2. The differential scattering cross section of neutrons from an ideal Bose gas for a fixed momentum transfer k . The ordinate N is proportional to the number of counts per unit solid angle.

⁴ A. Miller, D. Pines, and P. Nozières, Phys. Rev. **127**, 1452 (1962), see p. 1463.

FIG. 3. The differential scattering cross section of neutrons from a Bose liquid for a fixed momentum transfer $k \rightarrow \infty$. The ordinate N is proportional to the number of counts per unit solid angle.



The remaining particles are spread out in momentum space in a way which is not known in detail.

LIQUID HELIUM AT FINITE k

In practice it is impossible to obtain enough neutrons in the domain of k for which $\sigma_{\text{total}}(k)$ is reduced from its $k=0$ value.⁵ Expression (4) may still be taken as our estimate of the smearing of the delta function due to final-state interactions $\Delta\omega_0 \sim (k/m_{\text{He}}) n \sigma_{\text{tot}}(0)$. The particles not in the condensate will be spread out owing to the Doppler shift by an amount of the order of $\Delta\omega \sim k p_{\text{rot}}/m_{\text{He}}$. Using the values⁶ $\sigma = 2 \times 10^{-15}$ cm², $p_{\text{rot}}/\hbar = 2 \times 10^8$ cm⁻¹, $n = 2 \times 10^{22}$ cm⁻³, we find a "resolution"

$$R \equiv \frac{\Delta\omega_0}{\Delta\omega} = \frac{\hbar n \sigma}{p_{\text{rot}}} = \frac{1}{5}. \quad (5)$$

Needless to say, (5) is only a crude estimate of the effect of final-state interactions. Since it turns out that $R < 1$, we might expect the spectrum of scattered neutrons to be schematically as shown in Fig. 4. The important point is that the "delta-function part" corresponding to a helium atom initially in the condensate, will have a characteristic temperature dependence which will distinguish it from the contribution of the Doppler-shifted noncondensed particles. At incident neutron energies of 1 eV the energy resolution required to see the bump coming from the condensate is about

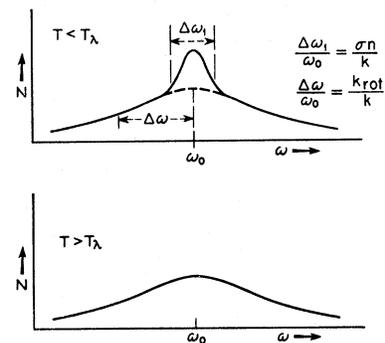


FIG. 4. The differential scattering cross section of neutrons from a Bose liquid for a fixed but finite momentum transfer k . The ordinate N is proportional to the number of counts per unit solid angle.

⁵ This would require neutron energies in the 100-eV range before a significant decrease set in.

⁶ I. Amdur and A. L. Harkness, J. Chem. Phys. **22**, 664 (1964).

1%, which seems to be attainable in this range of energies.

Although the relationship between superfluidity and macroscopic occupation of one mode is generally accepted, its experimental demonstration has been rather indirect. The proposed experiment, if even qualitatively successful, would yield information on the wave function of He II which would confirm the basic postulate of Fritz London on the relationship

between Bose-Einstein condensation and the superfluidity of He⁴.

ACKNOWLEDGMENTS

One of us (P.H.) wishes to thank Professor P. Nozières for an early discussion on this subject. We have also benefited from a discussion with Professor Michael Cohen.

Density, Coefficient of Thermal Expansion, and Entropy of Compression of Liquid He⁴ under Pressure Below 1.4°K†

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(Received 11 July 1966)

The density of liquid He⁴ at pressures up to 24.5 atm and at temperatures between 0.5 and 1.4°K has been measured by the dielectric-constant method. From the density data we derive the coefficient of thermal expansion, the entropy of compression, and the compressibility as a function of temperature and pressure. The results are discussed in terms of the Landau theory for liquid He⁴, and the parameters Δ , μ , and p_0 of the roton excitation spectrum are determined at several pressures and compared with previous determinations.

I. INTRODUCTION

IN this paper we describe measurements of the density ρ , the isobaric coefficient of thermal expansion α_p and the deduction of related thermodynamic properties for liquid He⁴ under pressure between 0.5 and 1.4°K. The principle of the method is the determination of the dielectric constant ϵ and the use of the Clausius-Mosotti relation

$$\rho = \frac{3M}{4\pi A} \frac{\epsilon - 1}{\epsilon + 2} \quad (1)$$

to derive the density ρ for a given pressure and temperature. Here M is the molecular weight and A is the polarizability. Until last year, only the early results of the *PVT* studies of Keesom and Keesom¹ were available down to 1.2°K, as well as measurements on the density and coefficient of thermal expansion for the liquid at saturated vapor pressure^{2,3} extending down to 0.9°K. Recently, Mills and Sydoriak⁴ presented measurements of α_p by the method of adiabatic expansion between

† Research supported by a grant of the National Science Foundation and of the U. S. Army Research Office (Durham). Data of this research were recently presented with tabulations in the Ph.D. thesis by C. Boghosian, Duke University, August 1965. A preliminary report was given by C. Boghosian and H. Meyer, *Bull. Am. Phys. Soc.* **10**, 258 (1965).

¹ W. H. Keesom and A. P. Keesom, *Commun. Kamerlingh Onnes Univ. Leiden Lab.*, 224e (1933). W. H. Keesom, *Helium* (Elsevier Publishing Company, Amsterdam, 1942).

² K. R. Atkins and M. H. Edwards, *Phys. Rev.* **97**, 1429 (1955).

³ E. C. Kerr and R. D. Taylor, *Ann. Phys. (N.Y.)* **26**, 292 (1964).

⁴ R. L. Mills and S. G. Sydoriak, *Ann. Phys. (N.Y.)* **34**, 277 (1965).

0.5 and 1.5°K. They also calculated the entropy of compression and the change in compressibility as a function of temperature. Mills⁵ then interpreted the results in terms of the Landau model of liquid helium.⁶⁻⁸

After a short description of the experiment in Sec. 2, the results obtained by the dielectric constant method will be presented and tabulated. Relevant thermodynamic properties such as the entropy of compression are tabulated also and compared with previous results. In Sec. IV, we will derive the Landau parameters Δ , μ and p_0 as a function of density and compare these results with those obtained by previous authors.

II. APPARATUS AND EXPERIMENTAL PROCEDURE

The apparatus and the principle of measurement are the same as for the work on liquid He³, described in another paper.⁹ The discussion⁹ on the validity of the Clausius-Mosotti relation is also relevant here. The measurements had to be limited to temperatures above 0.5°K because of the large heat influx into the cryostat resulting from the superfluidity of He II. At temperatures below 0.65°K, the value of the coefficient of thermal expansion became rather small. Its absolute value

⁵ R. L. Mills, *Ann. Phys. (N.Y.)* **35**, 411 (1965).

⁶ L. D. Landau, *J. Phys. (USSR)* **5**, 71 (1941).

⁷ L. D. Landau, *J. Phys. (USSR)* **11**, 91 (1947).

⁸ See also K. R. Atkins, *Liquid Helium* (Cambridge University Press, New York, 1959).

⁹ C. Boghosian, H. Meyer, and J. E. Rives, *Phys. Rev.* **146**, 110 (1966).