Small-Angle X-Ray Scattering from Liquid Helium I and Liquid Helium II†*

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X-rays from a rotating Cu anode x-ray tube have been scattered from liquid helium at angles up to 5 degrees in the temperature range from 1.5°K to 4.2°K. The helium was contained in a thin-walled aluminum tube enclosed in a metal cryostat with beryllium windows. The liquid structure factor obtained experimentally, agreed well with the theoretical zero-angle liquid structure factor between 3°K and 4.2°K. Below about 2°K, the observed value of the zero-angle structure factor is larger than the theoretical value by an amount believed to be outside the experimental error. Angular distributions at 4.2°K are typical of liquid scattering patterns near the critical point. No scattering anomaly was observed near the λ point.

I. INTRODUCTION

THE x-ray scattering by liquid helium has previously been measured[1-3] at several temperatures above and below the λ point and at angles from 15 degrees to 155 degrees. There are no appreciable differences between the characteristic liquid diffraction patterns obtained at different temperatures. The data seem to be consistent with some sort of very open liquid structure, with a range of order of perhaps 8A or so.

In the present paper, we report x-ray scattering measurements at temperatures from 1.5°K to 4.2°K and at angles less than 5 degrees. In this angular range the scattering is determined, principally, by the liquid density fluctuations rather than by the details of the short-range order.

The data are discussed in terms of the classical expression for the zero-angle liquid structure factor and certain possible discrepancies between experiment and the classical theory are considered in the light of the recent theoretical work.[4-6]

II. EXPERIMENTAL

The x-ray source[7] used in this experiment was a continuously evacuated beryllium window x-ray tube with a rotating copper water-cooled anode. It was run at 80 ma and 30 kv from a current and voltage regulated power supply. The electron stream was focused to a line 1 mm wide and 1 cm high. As viewed by the collimating system, the focal line was about 0.1 mm wide and 1 cm high. The flux density of x-rays incident on the sample was of the order of 10^9 Cu $K\alpha$ photons/ cm² sec. These constituted about ½ the total x-ray flux.

The x-rays were collimated[8] by a pair of tantalum slits in a fixed collimating tube and then were incident on the helium sample holder. The scattered radiation traversed a second pair of slits in a tube mounted on an arm which could be rotated about a line passing through the center of the sample holder. The collimating tubes were continuously evacuated and the windows in their end plates covered with thin mica. The slits were separated by 50 cm and were 0.16 cm wide and 1 cm high. The resolution was approximately 0.35 degrees. The smallest angle at which data could be taken was about 0.015 radian. Monochromatization of the x-ray beam to the Cu $K\alpha$ line was accomplished by the use of Ross balanced filters. The x-rays were detected with a commercial end-window Geiger counter mounted behind the filters on the movable arm.

The sample holder was of pure aluminum (Alcoa 2S) 6 in. long, i.d. 0.940 in., machined in two sections to minimize the rather considerable difficulty of construction. The wall of the lower section was 0.040 in. thick, except for a distance of ¾ in. near the bottom where it was turned down to 0.002 in., thus creating a window for the x-ray beam. The upper section was joined to a thin-walled stainless steel connecting tube with a graded seal, using copper for the intermediate metal. The two aluminum sections of the sample holder were joined with Araldite Type 1 cement, a temperature-polymerizing casting resin, which was applied to etched surfaces. This seal withstood repeated cycling between room temperature and liquid helium temperature without developing leaks.

The sample holder was attached to the cryostat with a gasket seal between two polished steel flanges. The gaskets were of 0.025-in. nominally pure gold wire, self-welded with an oxyhydrogen torch and annealed.

The cryostat, which was adapted from a modification by Dr. F. A. Rodgers of a cryostat described by Harvey and Dolecek[9] is shown in Fig. 1. It consisted of an outside vacuum shell A, whose upper section was a

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* Based on a thesis presented in partial fulfillment of the requirements for the Ph.D. degree at the University of Wisconsin.
‡ Now at the General Electric Research Laboratories, Schenectady, New York.
[1] W. H. Keesom and K. W. Taconis, Physica **5**, 270 (1938).
[2] J. Reekie, Proc. Cambridge Phil. Soc. **36**, 236 (1940); **43**, 262 (1946).
[3] Reekie, Hutchison, and Beaumont, Proc. Phys. Soc. (London) A**66**, 409 (1953).
[4] L. Goldstein, Phys. Rev. **83**, 289 (1951).
[5] L. I. Schiff, Phys. Rev. **57**, 844 (1940).
[6] P. J. Price, Phys. Rev. **86**, 495 (1952).
[7] B. R. Leonard, Jr., thesis, University of Wisconsin, 1951 (unpublished).

[8] Ritland, Kaesberg, and Beeman, J. Appl. Phys. **21**, 838 (1950).
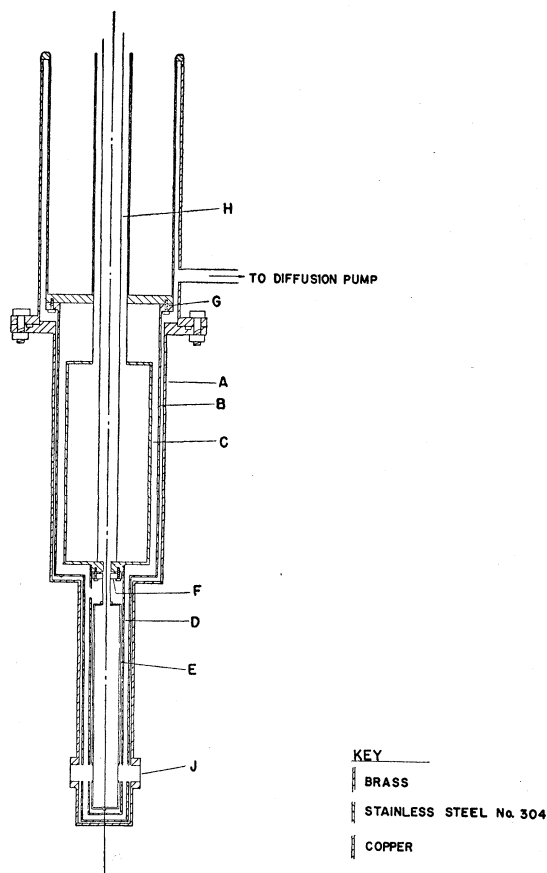[9] W. E. Harvey and R. L. Dolecek, Rev. Sci. Instr. **21**, 496 (1950).

FIG. 1. Vertical section of cryostat and sample holder used in the scattering experiment.

double-walled chamber open at the top. The inside of the chamber was stainless steel and held liquid air for cooling the radiation shield B. Inside the liquid air radiation shield was a liquid helium reservoir C, whose sole purpose was to provide thermal isolation for the sample holder E, by means of the copper radiation shield D. The flange connection for attaching the sample holder to the bottom of the liquid helium reservoir can be seen at F.

Slots were cut in both radiations shields at the level of the x-ray window and covered with 0.0003-in. aluminum foil. The outside vacuum windows J at room temperature were made of beryllium sheet cemented in place with Araldite Type 1.

The stainless steel tube K which attached the helium reservoir to the bottom of the liquid air reservoir was turned to a 0.016-in. wall thickness. The thermal path from liquid helium to liquid air temperatures was thus quite long; the loss rate of the helium reservoir was about 30 cm³/hr. A thin-walled stainless steel tube H passed through the helium reservoir and terminated at the throat of the sample holder, providing a means of filling and pumping on the sample holder. A ¼-in. bellows valve on the vacuum lead to the diffusion

pump made the cryostat portable. All joints were silver-soldered except those on the sample holder.

The end of the sample holder pumping tube H entered a box into which also came connections to the oil and mercury manometers as well as the coarse and fine vacuum leads for pumping off helium vapor at any desired rate. In addition, the box contained a rack and pinion gear system, the rack controllable through a rotary vacuum seal in the side of the box, which provided up and down motion for a combination sample holder depth gauge and stirring rod. Wollaston wire depth gauges[10] were used exclusively and proved to be moderately satisfactory.

The cryostat, manometers, pumping box, diffusion pump, and ion gauge were mounted on a conventional cart made of angle iron. The cart itself was mounted on a heavy base which allowed it to be rotated 90 degrees and translated 6 in. or so with considerable precision, permitting rapid and accurate positioning of the cryostat in the collimating system.

III. PROCEDURE

The cryogenic apparatus was designed so that the cryostat could be filled with liquid helium directly from the liquefier. Prior to filling, the sample holder was flushed and filled with warm helium gas. The stopcock was closed with the insulating vacuum space at a pressure of $\sim 3 \times 10^{-6}$ mm Hg, and the cryostat was then precooled with liquid air. When precooling was complete, the cryostat was carried to the Collins Helium Liquefier and the sample holder and liquid helium reservoir were filled, taking great care to prevent contamination of the sample by solid air. The cryostat was then set in position in the x-ray collimating system.

The method of taking data varied according to the plan for a given run. Since the evaporation rate of the sample was relatively large (approximately 5 cm³/hr out of a total capacity of 65 cm³), and since a minimum of one-third of the liquid was lost in pumping down to 1.5°K, it was not possible to cover the entire range of temperature in detail in any one run. However, the general procedure in taking data was to make a reading of the intensity at a given angle and temperature with the two Ross filters. Then either the angle or the temperature was varied and the process repeated. It was customary to wait 15 minutes at the beginning of a run and several minutes between readings where the temperature was varied for the sample to come to thermal equilibrium.

At the end of the run, the sample holder was evacuated and a background taken. The cryostat was then allowed approximately 12 hours to warm to room temperature. It was not moved during this time. The sample holder was then filled with sulfur hexafluoride at room temperature and pressure and the scattering measured as a function of angle in the same way that

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[10] A. Wexler and W. S. Corak, Rev. Sci. Instr. 22, 941 (1951).

it had been for liquid helium. The use of the SF_6 permitted calibration of the liquid helium scattering in a way which will be described in detail below. The SF_6 scattering was checked with the scattering from freon 12 (dichlorodifluoromethane) from time to time, and the ratio of the scattering by the two gases was found to agree satisfactorily with theory. The anomalous dispersion of Cu $K\alpha$ x-rays in these gases is negligible. The incident x-ray beam intensity was monitored from time to time during a run with an iron foil fluorescer. Fluctuations in intensity from one half of a run to the next were not found to be excessive.

A total of ten runs was made, of which one was thrown out because of difficulties with the x-ray voltage control. A number of angular distributions were measured at 4.2°K, and one each at 2.2°K, and 2.0°K, for angles between 0.015 radian and 0.090 radian. The scattering was measured as a function of temperature between 1.5°K and 4.2°K for angles from 0.020 radian to 0.040 radian.

The counting rates for the liquid helium scattering were about 17 counts/sec Cu $K\alpha$ at 4.2°K and 0.015 radian, including a background of 10 counts/sec. The background, which was principally due to the beryllium outside windows on the cryostat, dropped off rapidly with angle to about 1.7 counts/sec at 0.030 radian and essentially to zero at 0.090 radian. The SF_6 counting rates were about 30 counts/sec.

It was customary to take 6400 counts with the nickel filter and 3200 counts with the cobalt filter for the heavy gas measurements, and between 640 and 6400 counts for the liquid helium, depending on the angle and temperature.

IV. ANALYSIS OF THE DATA

The liquid structure factor, the quantity calculated in all theoretical treatments of the liquid scattering, is defined to be the ratio of the intensity actually scattered by the liquid sample to that which it would scatter if all the molecules were to scatter independently. By definition

$$\mathcal{L} = I_{He}/N_{He}I_m, \tag{1}$$

where $\mathcal{L}$ is the liquid structure factor, I_{He} is the actual liquid scattering, N_{He} is the number of helium atoms irradiated, and I_m is the intensity scattered by an isolated helium atom illuminated with the incident intensity I_0. The absolute determination of N_{He} and I_0 would be difficult and subject to large systematic errors. However, they may be eliminated from the expression for $\mathcal{L}$ by the use of the comparison gas SF_6.

At standard temperature and pressure, intermolecular interference is negligible in SF_6, and hence

$$I_g = N_g I_e F_g^2, \tag{2}$$

where I_g is the scattering by SF_6 at STP, N_g is the number of gas molecules irradiated, I_e is the intensity scattered by a single electron, and F_g is the SF_6 mo-

lecular structure factor. F_g was calculated by standard methods using the known constants of the SF_6 molecule.[11] At angles less than 5° F_g does not deviate greatly from the number of electrons in the SF_6 molecule. Equation (1) may be rewritten

$$I_{He}/\mathcal{L} = N_{He}I_e F_{He}^2, \tag{3}$$

where F_{He} is the atomic structure factor for helium. Dividing (2) by (3), we get

$$\mathcal{L} = \frac{F_g^2}{F_{He}^2} \cdot \frac{n_g}{n_{He}} \cdot \frac{I_{He}}{I_g}, \tag{4}$$

where n_g and n_{He} are the number densities of SF_6 molecules and helium atoms, respectively. Explicit use has been made of the fact that the scattering geometry was identical for the SF_6 and the liquid helium. Also, it has been assumed that the incident intensity was the same during both halves of a run. The iron fluorescer monitoring verifies this.

The scattered intensities to be inserted in Eq. (4) must be gotten from the raw data by the application of a series of corrections. The data must be corrected for the appreciable absorption of the Cu $K\alpha$ x-rays by the heavy gas and the liquid helium, and the background scattering from the slit system and evacuated cryostat must be subtracted. In the case of the SF_6 scattering, the resulting expression must then be corrected to STP. For the SF_6 data, therefore,

$$I_g = [S_g/X - B]\frac{T}{273} \cdot \frac{76}{P},$$

where S_g is the raw heavy gas scattered intensity, X is the transmission of the gas at T, the atmospheric temperature in degrees Kelvin, and P is the barometric pressure in cm Hg. B is the background. Similarly, for the liquid helium data,

$$I_{He} = [S_{He}/X(T) - B],$$

where S_{He} is the raw scattered intensity and $X(T)$ is the liquid transmission, which is a function of temperature since the absorption coefficient is a linear function of the liquid density.

The rest of the quantities on the right-hand side of Eq. (4) are known; hence, $\mathcal{L}$ may be obtained immediately.

The conventional theories[12,13] of the liquid structure factor predict that in the limit of x-ray scattering at vanishing angles, the liquid structure factor reduces to $\mathcal{L}_0 = nkT\chi_T$ where n is the number density of particles, k is Boltzmann's constant, T is the temperature in degrees Kelvin, and χ_T is the isothermal compressibility. Unfortunately, the calculation of this quantity

[11] L. O. Brockway and L. C. Pauling, Proc. Natl. Acad. Sci. U. S. 19, 68 (1933).
[12] F. Zernicke and J. Prins, Z. Physik 41, 184 (1927).
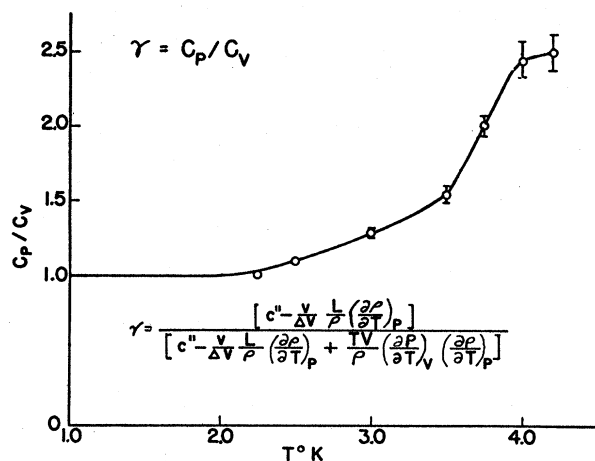[13] L. Brillouin, Ann. phys. 17, 88 (1922).

$$\gamma = C_P/C_V$$

$$\gamma = \frac{\left[c'' - \frac{V}{\Delta V} \frac{L}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P\right]}{\left[c'' - \frac{V}{\Delta V} \frac{L}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P + \frac{TV}{\rho}\left(\frac{\partial P}{\partial T}\right)_V\left(\frac{\partial \rho}{\partial T}\right)_P\right]}$$

FIG. 2. The ratio of the specific heats as a function of temperature for liquid helium. The error bars result from estimated uncertainties in the experimental values of the partial derivatives and the latent heat.

for comparison with experiment cannot be done in a straightforward manner for liquid helium, since X_T has not been measured directly for the liquid under its saturated vapor pressure. However, X_S, the adiabatic compressibility, can be obtained from measurements of the velocity of first sound[14] and $X_T = \gamma X_S$, where γ is the ratio of the specific heats. Using a method first employed by Groenewold[15] it is possible to calculate γ. From a relation between c'' and c_P,[16] it can be shown that

$$\gamma = \frac{\left[c'' - \frac{V}{\Delta V} \frac{L}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P\right]}{\left[c'' - \frac{V}{\Delta V} \frac{L}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P + \frac{TV}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P\left(\frac{\partial P}{\partial T}\right)_V\right]},$$

where c'' is the specific heat at saturated vapor pressure, V is the volume of a unit mass of the liquid, ΔV is the change in volume on evaporation, L is the latent heat of evaporation, and ρ is the liquid density. A plot of γ vs T is given in Fig. 2. The error bars refer to estimated uncertainties in the partial derivatives and the latent heat. All the other quantities are assumed to be exact.[17]

V. RESULTS AND ERROR DISCUSSION

The results of the scattering as a function of angle at 4.2°K, 2.2°K, and 2.0°K are shown in Figs. 3 and 4. The error bars refer to uncertainties due to counting statistics alone. In each case, the point at zero angle

[14] K. R. Atkins and C. E. Chase, Proc. Phys. Soc. (London) A64, 826 (1951).
[15] H. J. Groenewold, Nature (London) 142, 956 (1938).
[16] E. A. Guggenheim, Thermodynamics, An Advanced Treatise for Physicists and Chemists (Interscience Publishers, New York, 1949), p. 124.
[17] The numerical values of the quantities in this formula, as well as other numerical data on liquid helium used in this experiment, are taken from W. H. Keesom, Helium (Elsevier, Amsterdam, 1942).

is the calculated value of the theoretical zero-angle liquid structure factor. The experimental results for $\mathcal{L}$ as a function of temperature for various angles are shown in Fig. 5. Each of the points indicated in Fig. 5 is actually an average of two individual datum points in most cases, with the error bars reduced accordingly. The solid line is the calculated value of $\mathcal{L}_0 = nkTX_T$.

It is easily seen from the length of the error bars that most of the spread in the data can be attributed to counting statistics, which ran between 4 percent and 13 percent. Examination of the other errors shows that the rest of the spread in the data can be largely accounted for in terms of other known random errors. An error was assumed to be random, if it could change sign unpredictably from one measurement to the next. The significant known random errors and their estimated sizes are as follows:

(a) Errors in measurement of the temperature of the liquid helium scattering sample from measurements of the vapor pressure—less than 1 percent.

(b) Errors in measurement of the pressure and temperature of the SF_6 comparison gas in the sample holder—1 percent.

(c) Fluctuation in x-ray beam intensity during a count—2 percent.

(d) Fluctuation in x-ray beam intensity for one-half of a run to the next as estimated with the fluorescent monitor—3 percent.

The square root of the sum of the squares of these errors is 4 percent.

In addition to the random errors, there were possible systematic errors, due to instrument design and procedure, whose effect on the measured value of $\mathcal{L}$ can be estimated. They are as follows:

(a) Incorrect value for the transmission of the liquid helium—2 percent. This was due to uncertainty in the value of the mass absorption coefficient for helium, which was taken to be 0.69 for Cu $K\alpha$, by graphical interpolation in a table in Compton and Allison.[18]
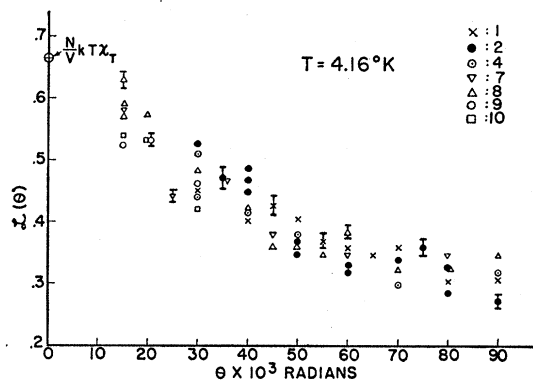

FIG. 3. Experimental values of the liquid structure factor of liquid helium as a function of angle at 4.16°K.

[18] A. H. Compton and S. K. Allison, X-Rays in Theory and Experiment (D. Van Nostrand Company, Inc., New York, 1935), p. 802.

(b) Effect of non-uniformity of the sample holder window thickness—2 percent. Since the sample holder shifted position in the x-ray beam from one-half of a run to the next due to thermal expansion, the thickness of the window must be uniform. The estimate given here is believed to be conservative.

(c) Impurities in the SF_6—1 percent. The SF_6 used in this experiment was at least 98 percent pure by volume, the principal impurity being air.[19]

The sum of these systematic errors is 5 percent.

Besides the relatively large errors described above, there were a number of smaller sources of error, largely systematic, which could add another 1.5 percent to the total error. They included such things as a temperature gradient in the He I, thermal expansion of the sample holder, possible error in the accepted value of the density of liquid helium, nonideality of the SF_6,[20] imbalance in the Ross filters, and the finite resolution of the slit system.

Finally, there were two sources of error which it was impossible to estimate quantitatively.

There is a possibility that the liquid helium sample bubbled, since its temperature was lowered by pumping directly on its surface. This would make the measured value of the liquid structure factor too low. It is believed that the sample holder evaporation rate of about 5 cm³/hr was small enough so that bubbling was not important, particularly in view of the experimental procedure described above. The fact that there was no sudden change in the scattered intensity as the temperature dropped below the λ point is the best indication that bubbling of the He I was not a serious problem.

Contamination of the liquid helium scattering sample could have been a very troublesome source of error in this experiment. The impurity content of the Amarillo well helium used was so small ($\sim 10^{-3}$ molar percent)[21] that it was not an important source of contamination.
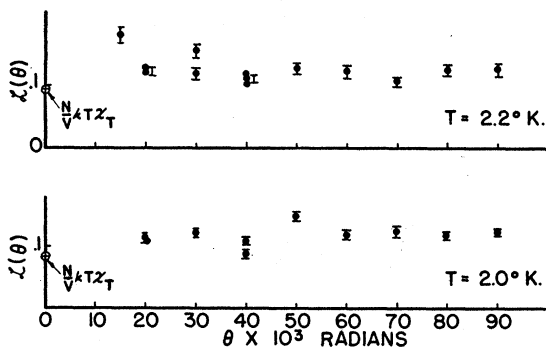

Fig. 4. Experimental values of the liquid structure factor of liquid helium as a function of angle at 2.2° and 2.0°K.

[19] M. O. Ericson, General Chemical Company, 205 South 16th Street, Milwaukee, Wisconsin (private communication).
[20] K. E. MacCormack and W. G. Schneider, J. Chem. Phys. 19, 845 (1951).
[21] G. E. Erlandson, Chief, Production Branch, Helium Division, Region VI, P. O. Box 911, Amarillo, Texas (private communication).
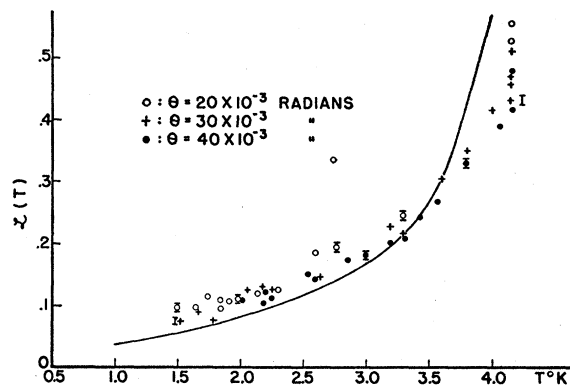
Fig. 5. Experimental values of the liquid structure factor of liquid helium at various angles and temperatures. The solid line is the value of the zero-angle liquid structure factor, as calculated from the ratio of the specific heats (see Fig. 2) and the velocity of first sound.

Great precautions were taken during filling of the cryostat to prevent contamination through the condensation of solid air and water vapor into the scattering sample. It can be shown, with the aid of Stokes' law, that particles of solid air of radius $>10^{-4}$ cm were not a source of contamination, since they will not remain suspended in the liquid sample more than 10 minutes. The surest indication that solid air did not seriously contaminate the sample is that there is no unaccounted for variation in the data for different runs. Since the procedure for filling the cryostat was not identical for all runs, it is extremely unlikely that the same amount of solid air and water with the same particle size distribution would have gotten into the sample holder each time. Consequently, it is believed that the contamination problem was effectively overcome.

VI. DISCUSSION OF RESULTS

In Fig. 3, it appears that the experimental curve of liquid-structure factor vs angle at 4.16°K, when extrapolated to zero angle, matches the theoretical value of $\mathcal{L}_0$ quite well. Moreover, the shape of this angular distribution is typical of the scattering by liquids not too far from the critical point. The critical temperature and pressure of helium are small enough, 5.2°K and 2.3 atmospheres, respectively, so that at 4.2°K and 76-cm Hg helium is still at a point on its diagram of state where the angular dependence of its scattering exhibits a maximum at zero angle and an appreciable decrease with increasing angle. Eisenstein and Gingrich,[22] in an extensive series of measurements of the x-ray scattering from liquid argon under its saturated vapor pressure from the triple point to the critical point, have demonstrated how the peak at zero angle, which is much more pronounced near the critical point, flattens out more and more with decreasing pressure and temperature until the curve of $\mathcal{L}$ vs angle

[22] A. Eisenstein and N. S. Gingrich, Phys. Rev. 62, 261 (1942).

eventually has a positive curvature at zero angle near the triple point. Similar results have been obtained by Wild[23] on nitrogen. The data in this experiment, while incomplete, seem to indicate a similar behavior on the part of liquid helium. The angular distributions near 2°K shown in Fig. 4 are considerably flatter than that of Fig. 3. It is almost possible within the experimental error, to draw a line of zero slope through the data points on both plots. However, such a line does not extrapolate to the theoretical value of the zero-angle liquid structure factor.

In view of the probable shapes of the angular distributions for temperatures somewhat below 4°K, it is believed that there is substantial agreement between the experimental and theoretical curves of $\mathcal{L}$ vs T shown in Fig. 5 from 3°K to the normal boiling point. The relative positions in Fig. 5 of the points taken at different angles (see legend) and roughly the same temperature, support this conclusion.

There is no evidence in Fig. 5 of a peak in the liquid structure factor near 2°K as predicted by Goldstein[4] for a perfect Bose-Einstein gas asymptotic model of liquid helium. This indicates that there is no anomalous ordering of the configuration of liquid helium at the λ point. Thus, statistical correlations in the liquid are considerably less important than interatomic forces in governing the structure. A similar conclusion can be reached by considering the results of De Boer and Lunbeck's[24] investigation of the quantum corrections to the equation of corresponding states for He^3 and He^4

It is believed that the discrepancy between theory and experiment in Fig. 5 is outside the experimental error below about 2°K and that it probably cannot be explained as an angular effect. Consequently, it may be necessary to find some other explanation of this apparent difference. It is interesting to note that, if one inserts the experimentally observed temperature dependence of the fraction of atoms in the condensed state into Goldstein's final formula for the liquid structure factor[25] in place of the $\frac{3}{2}$ power expression obtained in the Bose-Einstein condensation theory, the predicted peak in the liquid structure factor occurs at 1.5°K rather than at 2°K for a scattering angle of 0.03 radian. Such an *ad hoc* substitution is rather dubious, since it essentially takes account of the interactions at the end of the calculations instead of at the beginning. However, careful calculation of the liquid structure factor based on a more realistic model of liquid helium than the perfect Bose-Einstein gas might very well reveal

peculiarities in its behavior below the λ point. Recent work[26] seems to indicate that such a model need not be very complicated in order to be a good approximation to the actual liquid. It would appear that an adaptation of the methods of Fournet[27] might be made so as to take into account the effects of both statistics and atomic interaction in the calculations.

Price[6] has pointed out the possibility of anomalous scattering in liquid helium caused by the existence of zero-point fluctuations in density.

A final point is concerned with the problem of the proper way to obtain χ_T. It may be calculated directly from equation of state data, or it may be determined from the velocity of first sound and the ratio of the specific heats. The latter method was used since it does not depend as sensitively on the equation of state data, which are difficult to obtain accurately. This method is particularly desirable below about 2.5°K, where γ is very close to unity, and it is necessary only to know the first sound velocity in order to get the isothermal compressibility. However, first sound is strongly attenuated near the λ point and the velocity is complex. Consequently, the simple expression for the velocity will not hold, and it is possible that it is not correct to compare the x-ray scattering data with the theoretical expression in the way it has been done here.§

ACKNOWLEDGMENTS

[23] R. L. Wild, J. Chem. Phys. 18, 627 (1950).
[24] J. De Boer and R. J. Lunbeck, Physica 14, 139, 510 (1948).
[25] Reference 4, Eq. 20.

[26] M. H. Friedman and S. T. Butler, Phys. Rev. 91, 465 (1953).
[27] G. Fournet, thesis, University of Paris, 1950 (unpublished).
§ *Note added in proof.*—The author wishes to thank Dr. L. Goldstein of the Los Alamos Scientific Laboratory for stimulating correspondence on various ways of obtaining $L_0 = nkT\chi_T$ from the equation of state data. In connection with this correspondence, we have rechecked our computations for Fig. 2 and discovered a numerical error whose correction reduces the calculated values of γ at 4.16°K and at 4.0°K to 2.3 and 2.25, respectively. Other calculated values of γ are unchanged, and the error bars in Fig. 2 remain the same length. This change lowers the value of $L_0 = nkT\chi_T$ at 4.16°K shown in Fig. 3 to 0.16.