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Light Scattering in Liquid Helium

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Classical formulas appear to describe adequately the data reported here on the relative scattering of light by liquid and gaseous helium. Anomalous scattering at the λ point, if present, is less than the relatively large experimental error of about 20 percent.

THE light scattering of liquid helium has been discussed theoretically by Goldstein,¹ Schiff,² Galanin,³ and Ginsburg.⁴ The lowest predicted value for the scattering is given by Rayleigh's classical theory of density fluctuations.⁵

According to this approach, the fraction of light scattered per unit volume, per unit solid angle, is given by

$$\frac{i}{I} = \frac{\pi^2}{18\mu_0^4\lambda_0^4}(\mu_0^2 - 1)(\mu_0^2 + 2)^2 kT\chi, \quad (1)$$

where μ_0 is the index of refraction corresponding to the wavelength λ_0 of the scattered light, k is the Boltzmann constant, T the absolute temperature, and χ is the isothermal compressibility. Despite the fact that the temperature of liquid helium is considerably less than its Debye temperature, Brillouin⁶ has demonstrated that Eq. (1) is still valid in this temperature range for optical wavelengths. If, however, the λ -point phenomena are related to the condensation of an *ideal* Bose-Einstein gas, the scattering below the λ point should be by orders of magnitude higher and critical opalescence should be expected at the λ point itself.

Two experimental observations of the light scattering in liquid helium are recorded in the literature, one by McLennan, Smith, and Wilhelm,⁷ and the other by Jakovlev.⁸

McLennan, Smith, and Wilhelm found that the intensity of the light scattered by liquid helium could not be distinguished from the background in their experiment, which indicates that the scattering is of the order of magnitude given by Eq. (1). Jakovlev, comparing visually the scattering of air with that of liquid helium, came to the same conclusion.

We have measured the light scattering of liquid helium in a more quantitative manner. The apparatus is shown schematically in Fig. 1. The scattering chamber consisted of the lower end of a thin-walled stainless steel tube of 1-in. o.d., with a quartz window on top and another at the side. These windows were mounted on the metal with Araldyte plastic.⁹ A concave surface mirror on the bottom of the scattering chamber reflected the light beam back out of the cryostat. A high-pressure H-4 mercury arc lamp mounted outside the cryostat at the top of the stainless steel tube served as light source. A metal stopcock was used as light switch. No effort was made to use monochromatic light, as the available intensity at all wavelengths was needed to achieve adequate sensitivity. The scattered light was measured with a photomultiplier tube No. P-28 mounted immediately in front of the side window of the scattering chamber. It was separated by a quartz

¹ L. Goldstein, Phys. Rev. **57**, 241 (1940).

² L. I. Schiff, Phys. Rev. **57**, 844 (1940).

³ A. Galanin, J. Exptl. Theoret. Phys. **10**, 1267 (1940).

⁴ V. L. Ginsburg, J. Phys. (U.S.S.R.) **7**, 305 (1943).

⁵ See, for instance, R. H. Fowler, *Statistical Mechanics* (Macmillan Company, New York, 1936), second edition, p. 768.

⁶ Brillouin's theory [L. Brillouin, Ann. Phys. **17**, 88 (1922)] does not include zero-point energy as it is based on classical quantum theory. Introduction of the zero-point correction into his theory affects the values computed by Eq. (1) by less than 5 percent.

⁷ McLennan, Smith, and Wilhelm, Phil. Mag. **14**, 161 (1932).

⁸ I. A. Jakovlev, J. Phys. (U.S.S.R.) **7**, 307 (1948).

⁹ Araldyte is the trade name of Ciba Company for ethoxyline resins.

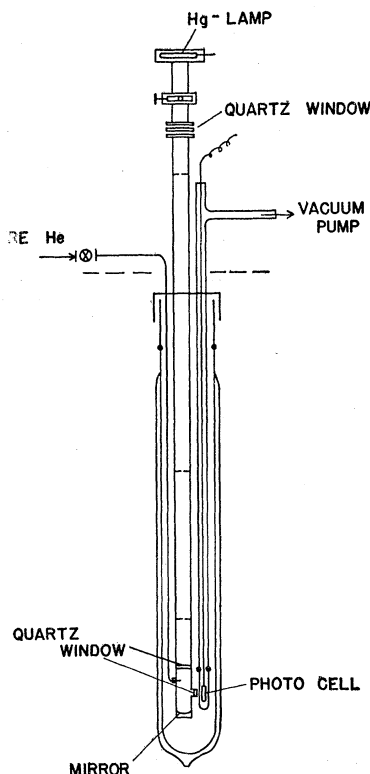


FIG. 1. Apparatus for measuring the scattering of light by liquid helium.

bulb and by high vacuum from the helium bath.¹⁰ The photocurrent was measured with a galvanometer arrangement with a sensitivity of 3×10^{-11} amp/mm deflection. The scattering chamber could be filled with helium via a stainless-steel capillary. The purification of the helium used for the scattering experiments required extreme care. The helium was first purified under 2000 lb/sq in. over charcoal at 80°K, and then once more led over well-degassed charcoal in a liquid nitrogen bath before entering the vacuum system connected with the scattering chamber. During filling, the gas flow rate had to be kept below 1 liter per minute. The liquid used for the scattering experiments was condensed from this highly purified gas.

¹⁰ Originally the photocell was placed directly in the liquid helium surrounding the scattering chamber. Under these conditions the sensitivity of the cell became extremely low, and the results were erratic. This behavior is probably due to the fact that the Cs-Sb surfaces of these cells are semiconductors with an extremely small gap between the valence and conduction bands, and so become practically insulators at helium temperatures. Surrounding the photomultiplier tube with high vacuum was sufficient to keep it at a temperature around 50°K in spite of the fact that the quartz vacuum chamber was covered for more than 10 inches with He II. The latter temperature seemed to provide nearly optimum working conditions for this type of phototube, because then the sensitivity was unimpaired and the noise level low.

The standard equipment for transferring liquid helium into the cryostat, for pumping on the helium bath, etc., are not shown in Fig. 1, nor is the Dewar with liquid nitrogen surrounding the helium Dewar.

As the measurement of the absolute ratio of intensity of the primary beam to that of the light scattered under a 90° angle seemed rather complicated, we preferred to calibrate the system by determining the light scattered by helium gas under 1 atmosphere at its boiling point. By taking the ratio of the light scattered by the liquid at different temperatures, $S_L(T)$, to the light scattered by the gas at the boiling point under the same conditions, $S_g(\text{bp})$, we eliminate the necessity of knowing the intensity of the primary beam, the geometric factors, etc. Equation (1) then reduces to¹¹

$$\frac{S_L(T)}{S_g(\text{bp})} = \frac{(\epsilon_L - 1)^2}{(\epsilon_g - 1)^2} \frac{T}{4.2} \chi_L P_g. \quad (2)$$

The results of a typical run are given in Table I.

TABLE I. Comparison of observed and calculated values of $S_L(T)/S_g(\text{bp})$.

1	2	3	4	5	6	7	8
$T^\circ\text{K}$	P atmos	Phase	Galv. deflect. in cm	Back- ground deflection in cm	4 minus 5	$S_L(T)/$ $S_g(\text{bp})$ obs	$S_L(T)/$ $S_g(\text{bp})$ calc. Eq. (2)
4.2	1	gas	3.0 ₀	2.8 ₀	0.2 ₀	1.0	
4.2	1	liq.	3.6 ₀	2.8 ₀	0.8 ₀	4.0	3.8
2.1	1	liq.	3.0 ₀	2.8 ₀	0.2 ₀	1.0	1.0
1.5	1	liq.	2.9 ₅	2.8 ₀	0.1 ₅	0.7 ₅	0.7

Within our accuracy of about 20 percent, the light scattering of liquid helium is in experimental agreement with Eq. (2) if one uses the experimentally determined values for μ_0^2 or ϵ and χ .¹² No excess scattering larger than our experimental error is observed at the λ point. This conclusion was confirmed by several runs in which the temperature of the bath was allowed to warm up slowly from 2.1 to 2.2°K during an interval of from 10 to 20 minutes. The galvanometer readings remained constant within the experimental accuracy and did not

¹¹ Equation (2) is derived by using the ideal gas law. Actually, the deviation from the ideal gas law at the helium bp is about 25 percent. However, using the second virial coefficient B as measured by Keesom and Walstra [Physica 7, 985 (1940)], not only χ , but also the value of ϵ changes, since it has to be calculated from the values given for -191° and 0°C by means of the gas density at 4.2°K. Both corrections cancel each other within 10 percent, which does not exceed the accuracy of our measurements and the accuracy to which the compressibility of the liquid is known.

¹² See W. H. Keesom, *Helium* (Elsevier, Amsterdam, 1942), pp. 136, 319, 134, 236.

deviate perceptibly from the normal value as the λ point was passed.¹³

It should be emphasized that the principal source of error in this experiment is impurities. Although it might be presumed that it would be relatively simple to purify helium, our experience indicates that extra-

¹³ The liquid in the scattering chamber was kept under 1-atmosphere pressure to avoid bubble formation during warming or cooling of the bath, so the λ point of the liquid helium in the chamber was a few thousandths of a degree lower than that in the bath. In some runs the temperature of the bath was kept as near as possible ($\pm 0.0003^\circ$) to the λ point of the liquid helium in the scattering chamber. Even then there was not the slightest indication of an increase in scattering.

ordinary precautions must be taken to avoid the introduction of miniscule particles of dust. Any deviation from our strict routine in handling the helium gas inevitably led to a considerably enhanced background scattering.

The results reported here are consistent with those of Taconis¹⁴ and Reekie,¹⁵ who were unable to detect any change in the x-ray diffraction pattern of liquid helium at the λ point.

¹⁴ W. H. Keesom and K. W. Taconis, *Physica* **5**, 270 (1938).

¹⁵ Reekie, Hutchison, and Beaumont, *Proc. Phys. Soc. (London)* **A66**, 409 (1953).