as a result of the complicated nonstationary velocity field generated by the presence of quantized vortices, and that as one passes through onset, the density of these vortices increases rapidly. However, in the thin films the entrance of vortices seems to be energetically unfavored, and it may be that in this case the uncertainty in V_s is related to a macroscopic zero-point motion.

While some success has perhaps been achieved by grafting various (macroscopic) quantum ideas onto the Landau two-fluid equations, the following basic question is forced upon us: What is the single macroscopic Schrödinger theory which contains the macroscopic quantum ideas such as (1) and (2), and in some limit reduces to the Landau two-fluid theory (macroscopic correspondence principle)?

It is a pleasure to thank T. Wang for making available prior to publication his experimental results on the absorption of third sound.

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[1] L. Onsager, cited in F. London, *Superfluids* (Wiley, New York, 1954), Vol. 2, p. 151, note; L. Onsager, Nuovo Cimento Suppl. 6, 249 (1949). For experimental verifications of the quantization condition, see W. F. Vinen, Proc. Roy. Soc., Ser. A, 260, 218 (1961); S. C. Whitmore and W. Zimmermann, Jr., Phys. Rev. 166, 181 (1968); G. W. Rayfield and F. Reif, Phys. Rev. Lett. 11, 305 (1963).

[2] For He II the "classical" dynamics are the Landau two-fluid equations: L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon, London, 1959), Chap. 16.

[3] In Eq. (5) it is appropriate to exclude the nonsuperfluid part (the so-called solid layer) of the film adjacent to the substrate which is commonly taken to be one atomic layer thick.

[4] I. Rudnick and J. C. Fraser, J. Low Temp. Phys. 3, 225 (1970).

[5] A careful consideration of the attenuation and propagation of third sound according to the Landau two-fluid theory is given by D. Bergman, Phys. Rev. A 3, 2058 (1971).

[6] See K. R. Atkins and I. Rudnick, in *Progress in Low Temperature Physics VI*, edited by C. J. Gorter (North-Holland, Amsterdam, 1970), Chap. 2, for early experimental values of the attenuation coefficient of third sound.

[7] T. Wang, Ph.D. thesis, University of California, Los Angeles, 1971 (unpublished).

[8] Still another way of obtaining this condition is to equate the van der Waals potential experienced by a single particle with the quantum fluctuation energy given to this particle by localizing it in the shallow superfluid layer. It is important, however, to notice the following difference between this condition and a corresponding condition that could be written, for instance, for the propagation of long gravity waves in an ordinary fluid. In the latter case we expect that the waves will still propagate even when the quantum fluctuation energy of the single particles is greater than the potential energy. This is so because for an ordinary fluid the individual particles move incoherently, whereas for He II the superfluid particles move coherently in a macroscopically occupied quantum state.

Raman Scattering from Condensed Phases of He^3 and He^4

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Inelastic light scattering with frequency shifts from 10 to 150 cm^{-1} has been measured in liquid and solid He^3 and He^4 under pressures up to 150 atm. In the hcp solids a transverse optic phonon was observed and resolved. In addition a broad peak was observed which is remarkably similar in both liquids and solids. In the solid this peak is interpreted as scattering from two or more damped, coupled phonons. Scattering intensities and spectral forms are compared with recent theoretical calculations.

Light scattering is a useful and often unique probe of density fluctuations in quantum crystals and fluids. It is complementary to neutron scattering but is capable of greater resolution. Light scattering is particularly important for He^3 since neutron scattering is made impractical because of the large nuclear absorption. Since the light interacts with the density fluctuations only via the weak polarizability of the bound electrons, the scattered signal-to-noise ratios in helium are small. In spite of experimental difficulties, a number of interesting results have been observed, such as light scattering from first and second sound and from two rotons.[1-4] We report here the scattering of laser light from liquid He^3 and liquid He^4 under pressure, from the bcc and hcp phases of solid He^3, and from the hcp phase of solid He^4. In the hcp phase of the solids, first-order Raman scattering from the transverse optic (TO) phonon was observed. In both liquids and solids, scattering by multiple excitation of vibrational modes produces a broad peak in the spectrum extending out to nearly 150 cm^{-1}.

Raman scattering in liquid and solid helium has been studied theoretically by Stephen[5] and by Werthamer *et al.*[6,7] They assume that the light couples to density fluctuations via the dipole coupling of point dipoles induced by the electric field of the incident laser light. In the solid conservation of wave vector requires single-phonon scattering from phonons near the zone center or multiphonon scattering from two or more phonons whose wave vector sum is nearly zero modulo a reciprocal-lattice vector. For solid hcp helium this process yields

$$\mathcal{R}_1 \approx \left(\frac{u}{a}\right)^2 \frac{\alpha_0^4}{a^6} \epsilon_{1\perp}^2 \epsilon_{2\perp}^2 \approx 10^{-12}\ \mathrm{cm}^{-1}\ \mathrm{sr}^{-1}, \qquad (1)$$

$$\mathcal{R}_2 \approx (u/a)^4 (\alpha_0^4/a^6) \int_0^\infty \sum_{ij} C_{ij} \rho_{ij}\, d\omega \approx 10^{-11}\ \mathrm{cm}^{-1}\ \mathrm{sr}^{-1}, \qquad (2)$$

where $\mathcal{R}_1$ and $\mathcal{R}_2$ are the Raman efficiencies for one- and two-phonon scattering, u is the amplitude of motion about the equilibrium atomic positions at spacing a, α_0 is the atomic polarizability, ρ_{ij} is the joint density of states of the ith and jth phonon branch, C_{ij} is a factor relating the symmetries of the phonons to the light directions and polarizations, and $\epsilon_{1\perp}$ and $\epsilon_{2\perp}$ are the components of the input and scattered light polarizations perpendicular to the c axis of the hcp phase. Similar estimates can be obtained using the simple classical model of a diatomic vibrator where each atom is polarized by the external field of the light and the pair interacts via dipole coupling of the induced dipoles. The single-phonon efficiency $\mathcal{R}_1$ is smaller for helium than for other solids because for helium the larger u^2/a^8 factor is dominated by the smaller α_0^4 factor. However, the two-phonon efficiency $\mathcal{R}_2$ is of the same order of magnitude for He, H_2, Ne, and Ar since the u^4/a^{10} factor compensates for the change in α_0.

The small Raman efficiencies for helium require careful experimental techniques (e.g., shielding against leaked elastic scattering), but all of the equipment which we used is readily available. Approximately 200 mW of linearly polarized Ar laser radiation at 5145 Å was used to excite the helium in a focal volume 20 μm in. diameter and 2 mm long. The stainless-steel helium cell containing a volume of 8 cm^3 was cooled from the bottom by copper contact to a pumped liquid-helium-II pot. Optical access was achieved through three 0° sapphire windows sealed to the cell with indium alloy *O*-rings. In order to eliminate laser elastic scattering and heating due to dust and impurities, we used a crushed stainless filter in the helium input capillary. The cell temperature was monitored by Ge thermometers and calibrated by the passage of the solid-liquid phase boundary through the focal volume. Scattered light of all polarizations at 90° to the incident beam was collected with $f/4$ optics. The image of the scattered light was rotated 90° with three reflections from plane mirrors and focused into a double grating spectrometer. Because of its low dark current (≈ 0.17 counts/sec) and high efficiency, a cooled BX 754 Channeltron photomultiplier was used to detect the weak scattered light (≈ 10 photons/sec). The signal was digitally averaged using a small computer to obtain the data in Fig. 1. These spectra were swept 50 to 100 times, sampling in each sweep at 50 to 100 points and integrating for 1 sec per point. In Fig. 1 adjacent points are connected with straight lines and the noise is predominately statistical. We subtracted constant background of 0.3–0.5 counts/sec caused by Channeltron dark current and leaked elastic scattering. For the sharp one-phonon peaks in hcp He^3 and He^4 both the background and the two-phonon scattering has been subtracted. Only Stokes scattering is shown in Fig. 1 since at 2°K anti-Stokes scattering from thermally excited phonons of these energies is much weaker.

The spectral feature most readily analyzed is the single-phonon peak observed near 10 cm^{-1} in both hcp He^3 and He^4. It is due to scattering from the Raman-active, degenerate, TO phonons near the zone center. Single-phonon scattering from the higher-frequency longitudinal optic mode is forbidden by symmetry. At a molar volume of 19.2 ± 0.1 cm^3 the measured frequency for the TO mode is 9.4 ± 0.2 cm^{-1} for He^4 and 11.6 ± 0.2 cm^{-1} for He^3 compared with the theoretical calculations[7,8] of 11.2 and 12.15 cm^{-1}, respectively. The agreement with theory is considered good since second-order perturbations due to phonon interactions are not included and are expected to shift the theoretical predictions toward lower frequencies. The width of the TO mode was measured to be 1.3 ± 0.5 cm^{-1} (full width at half-maximum). Calculations are now in progress which include damping and second-order frequency

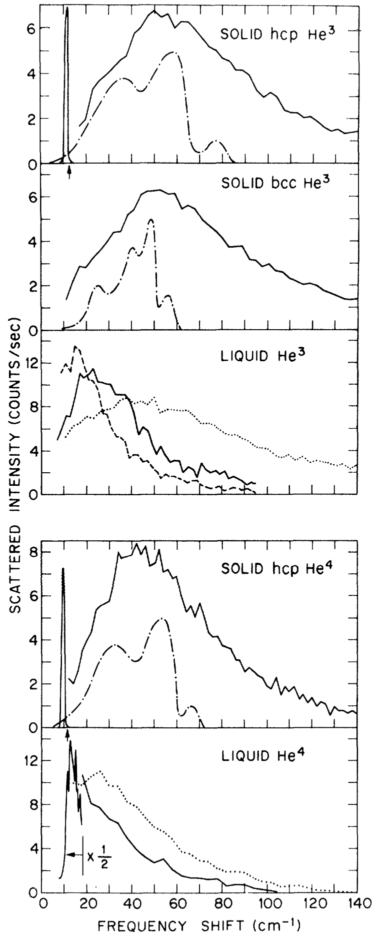

FIG. 1. The Raman spectra of liquid and solid He^3 and He^4. Dash-dotted curves and arrows are theory (Ref. 7). For liquid He^3: broken line, 0.8 atm; solid line, 14.8 atm; dotted line, 94 atm. For liquid He^4: solid line, 10 Torr; dotted line, 21 atm. For the solids: hcp, 19.2 ± 0.1 cm^3/mole; bcc, 20 ± 0.2 cm^3/mole. Resolution is 4 cm^{-1} except for the TO modes (1 cm^{-1}) and liquid data (2 cm^{-1}). The temperature is 2.3°K except liquid He^4 (1.7°K).

shifts.[9] The integrated intensity of the TO mode in the solid He^4 with $\epsilon_{1\perp}$ maximized was about 10% of the integrated two-roton intensity in liquid He^4 at 1.7°K (Fig. 1). Comparison with the intensity of the two-roton scattering quoted by Greytak and Yan[4] yields a TO-mode Raman efficiency of 1×10^{-12} cm^{-1} sr^{-1} in fair agreement with the estimate of Eq. (1) ($\mathcal{R}_1 = 3\times10^{-12}$ cm^{-1} sr^{-1}).[6] (Similar agreement with the dipole scattering theory has been obtained for the TO mode in hcp solid hydrogen.[10]) The pressure dependence of the TO frequency ω_T in solid He^4 yields a preliminary value of the Grüneisen constant $[-d(\ln\omega_T)/d(\ln V)]$ of 3 ± 0.5. The c-axis orientation and the perfection of the crystal in the scattering volume can, in principle, be determined by the dependence of the scattered intensity on the polarization of the input and scattered beams.[7,10] We found a pronounced dependence of the intensity of polarization but quantitative data were not possible because of depolarization of the light by the cell windows. Judging from the experience of others,[11] it seems quite likely there was only one single crystal in the small focal volume which we used.

A broad peak was observed in the spectra of both liquids and solids between 10 and 150 cm^{-1}. For the solids, a spectrum in this range caused by two-phonon scattering has been calculated[6,7] and the predictions corresponding to the experimental parameters are sketched in Fig. 1 (dash-dotted curves). The calculations neglect phonon damping and interactions. These theoretical curves are equally weighted averages of the spectra calculated for various orientations and are not normalized to the data. The theoretical structure is caused by maxima in the two-phonon joint density of states at various critical points. The measured integrated intensity of the broad peak in the solids is about a factor of 10 larger than that of the single TO phonon in agreement with the theoretical predictions.[6,7] However, the measured spectra exhibit none of the expected structure and extend to considerably higher frequencies than predicted by theory.[6,7] It is clear that in the framework of a phonon theory for solid helium, phonon interactions and light scattering from more than two phonons must be important.

It is interesting that the broad peak changes very little in intensity or spectral form when the crystal is melted (e.g., in Fig. 1, compare liquid He^3 at 94 atm with bcc solid He^3 at 113 atm). This is in contrast to Ar and Kr where the liquid spectrum has no peak and is nearly exponential.[12-14] In addition, the scattered intensity in liquid Kr and Ar decreases by a factor of from 10 to 10^3 upon solidification.[14] Only the exponential high-frequency tails for the liquids in Fig. 1 resemble the spectra observed for the other rare-gas liquids. The peak which we observe in liquid He^3 shifts to lower frequencies as the pressure is decreased but persists down to 0.8 atm where liquid and vapor are in equilibrium at 2.3°K. At constant pressure the spectral form and intensity are nearly independent of temperature in the 2 to 4°K range. In superfluid He^4 at 1.7°K similar behavior is observed down to about 10 atm where two-roton scattering begins to appear. As the pressure is increased from 0.01 to 10 atm the two-roton scattering shifts to lower frequencies and decreases in intensity.

The similarity of the broad peak in the liquids to that in the solids probably indicates the existence of both short-range correlated motions in the liquids (similar to zone-boundary phonons in the solid) and considerable disorder in the solids near the melting point. The existence of a peak in liquid He and the absence of a peak in the classical liquids such as Ar and Kr appears to indicate that short-range correlated motions have much better definition in helium. However, the high-frequency tails in the scattered spectra are similar and probably can be regarded as collision induced scattering from hard binary collisions. It is interesting to note that the classical liquids are at temperatures near the Debye temperature in the solid (for Ar, $T \approx 90^\circ\mathrm{K}$, $\theta_\mathrm{D} \approx 80^\circ\mathrm{K}$) while the temperature of liquid helium is much less than the Debye temperature of solid He ($T \approx 4^\circ\mathrm{K}$, $\theta_\mathrm{D} \approx 20^\circ\mathrm{K}$) (melting in helium is associated with quantum motion). The higher reduced temperature (T/θ_D) in the classical liquids may cause a relative decrease of short-range correlation and increase in damping. The large increase in the intensity of scattering upon melting solid Ar and Kr is still not understood.

In summary, density fluctuations at high frequencies have been observed by light scattering in liquid and solid helium. The results in the solid indicate a well-defined single-phonon peak and a broad, featureless multiphonon peak. The observed frequencies of the single-phonon peaks for both He^3 and He^4 are 5 to 10% lower than those predicted by the self-consistent phonon calculations. The light-scattering intensities agree in order of magnitude with a simple dipole-interaction model. In the liquids the spectra are similar in both spectral form and intensity to the multiphonon spectra in the solids. Interesting extensions of this work include scattering from He^3:He^4 mixtures and scattering from liquid He^3 at lower temperatures where degeneracy effects and zero sound may be important.

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