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LIGHT SCATTERING FROM ROTONS IN LIQUID HELIUM*

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We have measured the spectrum, intensity, and polarization of light Raman scattered from rotons and higher energy quasiparticle excitations in liquid He^4 for temperatures between 1.16 and 2.14°K. The effect on the spectrum of the temperature-dependent energy and lifetime of the rotons is clearly visible. At a scattering angle of 90° the light is highly depolarized and about three orders of magnitude weaker than the Brillouin scattering from the phonons.

Using a 1-W argon laser, an Ebert-grating monochromator, and photoelectron counting detection we have measured the spectrum, intensity, and polarization of light Raman scattered from liquid helium. In the scattering process two elementary excitations are created in the liquid with nearly equal and opposite momenta $\vec{k}$ and $\vec{k}'$. This is indicated schematically on the dispersion curve for liquid helium shown in Fig. 1. By the conservation of momentum the sum $\vec{k}+\vec{k}'$ is equal to the change in the momentum of the scattered photon. However, the momentum of an optical photon is small compared with that of the elementary excitations involved in this process; for example, it is less than 10^{-3} times the momentum of a roton (an excitation near the local minimum of the dispersion curve). Therefore the magnitudes k and k' of the momenta of the elementary excitations will be nearly equal. Since the liquid is isotropic, the energy of an excitation depends only on the magnitude of its wave vector, and, as shown in Fig. 1, the energies E_k and $E_{k'}$ of the elementary excitations created in the scattering process will be virtually identical. This result, together with the conservation of energy, shows that the scattered photon will suffer an energy loss corresponding to twice the energy of an elementary excitation in the liquid.

As Halley has recently proposed,[1,2] this type of scattering can complement neutron inelastic scattering as a tool for studying the elementary excitations in liquid helium. Halley shows[2] that the intensity of the Raman spectrum at an energy shift $2E$ will be the product of two terms, the density of states for the elementary excitations

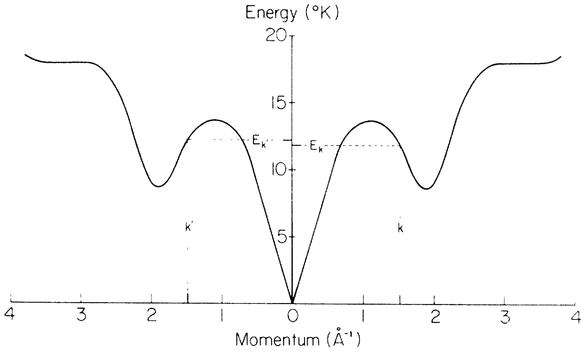

FIG. 1. The dispersion curve for liquid He^4 taken from neutron inelastic-scattering data. E_k and $E_{k'}$ are the energies of a possible pair of excitations, of nearly equal and opposite momenta k and k', created in the liquid during the scattering of a photon.

evaluated at an energy E and a term indicating the strength of the interaction between these excitations and the electric field of the light beam. Thus a peak should occur in the Raman spectrum corresponding to each region in which the dispersion curve for the elementary excitations has zero slope and, therefore, a high density of states. Figure 1 shows that there are at least three such regions in the dispersion curve[3] for liquid helium.

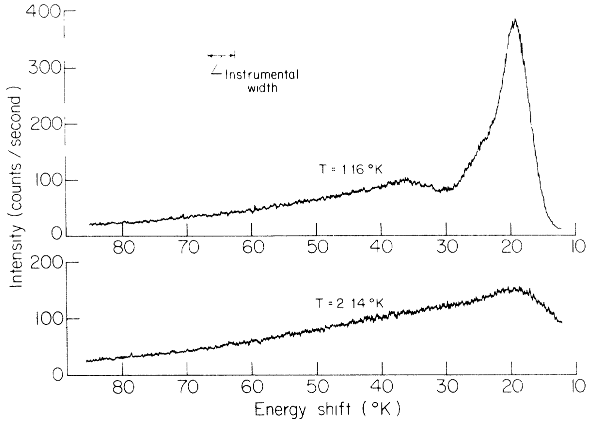

FIG. 2. The Raman spectrum of liquid He^4 at a scattering angle of 90°, in a direction parallel to the electric field vector of the incident light. The instrumental width is 4.2°K.

In our experiment approximately 1 W of linearly polarized 5145-Å radiation from a Coherent Radiation Laboratories Model No. 52 argon laser was focused to a narrow beam inside the sample cell. The cell was filled by means of a superfluid leak to avoid the problem of Tyndall scattering from frozen air and other particulate matter present in the main helium reservoir. The Brillouin scattering from the thermal phonons was strong enough to see by eye; therefore, it was used to align the optical system and also served as a reference against which to measure the relative intensity of the Raman scattering. The scattered light was collected within a solid angle of about 0.08 sr centered on a scattering angle of 90°. The image of the beam in the liquid was rotated by a Dove prism and focused on the input slit of the spectrometer. The spectrometer was a modified Perkin-Elmer Model No. E-1 grating monochromator used in first order and double pass. The light passing through the exit slit was detected by a cooled ITT Model No. FW130 photomultiplier tube. Puise-height discrimination was used at the output of the tube and reduced the dark count to below 1 photoelectron/sec.

Figure 2 shows experimental traces of the Raman spectrum in liquid helium at two different temperatures below the lambda point. In the literature on liquid helium it is standard practice to give energies of excitations on the dispersion curve in °K. We will follow this convention here, and note that an energy shift of 1°K corresponds to a frequency shift of 0.695 cm^{-1}, or 20.9 GHz. The principal features of the spectrum at a temperature of 1.16°K are a sharp asymmetric peak with a maximum corresponding to an energy shift of 18.5 ± 0.5°K, a broad peak at an energy shift of 35.3 ± 1.0°K, and a gradual decrease in the intensity at higher energy shifts. When the temperature is raised to 2.14°K the sharp peak spreads markedly and the broad peak can no longer be distinguished, but the spectrum for energy shifs beyond about 40°K does not change. The energy of a single quantum excitation at the roton minimum of the dispersion curve approaches a value[4] $E_{\text{min}} = 8.65$°K at low temperatures. The maximum of the sharp peak in our low-temperature spectra occurs at an energy shift which differs slightly from $2E_{\text{min}}$. This difference results from a distortion of the true spectrum produced by its convolution with the instrumental profile of the spectrometer. We have investigated this effect by convolving the density of states corresponding to a parabolic fit to the roton region of the dispersion curve with several analytic approximations to our instrumental profile. We find that this convolution shifts the maximum of the true spectrum toward higher energies by about one third of the full width at half-height of the instrument. In our case this correction amounts to 1.4°K and shifts the predicted maximum to 18.7°K. The maximum of the experimental trace occurs at 18.5 ± 0.5°K. The shape of the experimentally measured spectrum near the sharp maximum is in qualitative agreement with the spectrum resulting from our convolution. Figure 3 shows a study made of the temperature dependence of the roton peak using an instrumental width of 2.1°K, half that used in Fig. 2. Smooth curves were drawn through four experimental traces so that they could be superimposed conveniently on the same plot. As the temperature increases toward T_λ the peak is seen to move toward smaller energy shifts and increase in width. The magnitudes of these effects are consistent with the temperature dependence of the roton energies and roton lifetimes measured by neutron inelastic scattering.[4] The intensity of the scattering depends on the energy shift through the density of states, the dependence of

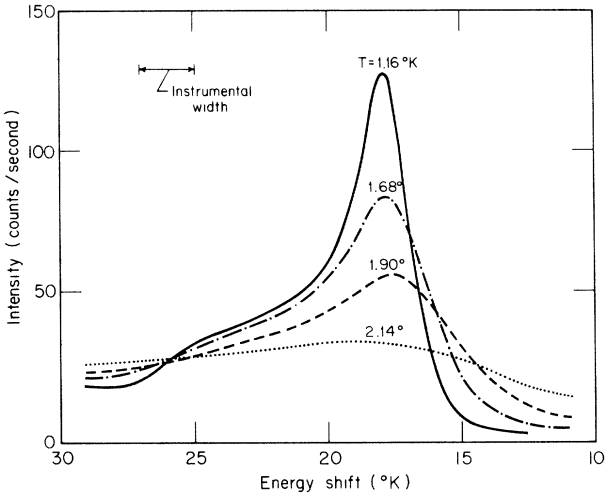

FIG. 3. The Raman spectrum of liquid helium about the roton peak at four different temperatures, at a scattering angle of 90°, in a direction parallel to the electric field vector of the incident light. The instrumental width is 2.1°K.

the coupling constant between the light and the excitations on the energy of the excitation, and the lifetimes of the excitations. At the present time the relative contributions of these three factors to the spectrum are not sufficiently understood theoretically to allow us to obtain quantitative values for the roton lifetimes from our data. However, we will be able to obtain precise values for the roton energies and lifetimes as a function of temperature from spectra similar to those in Fig. 3 when the dependence of the theoretical spectra on these parameters is known in more detail.

The density of states near the local maximum of the dispersion curve, $E_{\mathrm{max}} = 13.7°\mathrm{K}$,[4] has a magnitude similar to that for the rotons; therefore, this region could also contribute a peak to the Raman spectrum. In fact, the calculation of Halley[2] predicts a peak in the scattering due to these excitations which is roughly as strong as that due to the rotons. Our spectra show no evidence of such a peak. At an energy shift of $2E_{\mathrm{max}}$ our measured intensity is about a factor of 7 below that at the roton peak. The absence of a peak in the spectrum at $2E_{\mathrm{max}}$ shows that the coupling between the light and the elementary excitations varies markedly along the dispersion curve.

The dispersion curve for liquid helium contains a plateau at an energy of 17.9°K.[3] This will certainly give rise to a high density of states and could, therefore, cause a peak in the spectrum at an energy shift of twice this value, or 35.8°K. Since the maximum of the broad peak we see at low temperatures occurs at an energy shift consistent with this figure, we attribute the broad peak to scattering from excitations in the vicinity of the plateau in the dispersion curve.

Stephen informed us that his calculations[5] predict that the Raman-scattered light should be highly depolarized; we have verified this experimentally. We made measurements at a temperature of 1.16°K on light scattered at an angle of 90° in the region of the spectrum corresponding to the maximum of the roton peak. Let $\vec{l}$ and $\vec{\mathcal{E}}$ indicate, respectively, the propagation direction and the electric field polarization of the incident light. With $\vec{\mathcal{E}}$ oriented perpendicular to the direction of observation, the ratio of scattered intensity polarized parallel to $\vec{l}$ to that polarized perpendicular to $\vec{l}$ was 0.9 ± 0.2. With $\vec{\mathcal{E}}$ oriented parallel to the direction of observation, the ratio of scattered intensity polarized parallel to $\vec{l}$ to that polarized perpendicular to $\vec{l}$ was 1.15 ± 0.20. The total intensity scattered at 90° regardless of polarization, when $\vec{\mathcal{E}}$ was oriented parallel to the direction of observation, was 0.84 ± 0.08 times smaller than that scattered when $\vec{\mathcal{E}}$ was perpendicular to the direction of observation. The above ratios have been computed taking into account the depolarizing effect of the Dove prism and the polarization dependence of the spectrometer transmission. At least half the indicated uncertainty in these ratios is caused by the difficulty of taking these effects into account quantitatively. We noticed no dependence of the spectrum of the Raman-scattered light on the direction of scattering or the polarization.

We have measured, at a temperature of 1.16°K and in a direction perpendicular to $\vec{l}$ and $\vec{\mathcal{E}}$, the intensity of the Raman scattering relative to the Brillouin scattering. The Brillouin shift is small compared with the instrumental widths used in this experiment, so all of the Brillouin intensity appears to be concentrated in a very narrow line at zero energy shift. The stray elastically scattered light present in the cell due to the passage of the laser beam through the Dewar and cell windows was measured to be less than 1% of this Brillouin intensity. The stray light which would appear in the Raman spectra due to scattering of the Brillouin light inside the spectrograph was measured to be 3×10^{-6} times smaller than the Brillouin peak. The total Raman intensity of both polarizations with energy shifts between 14 and 92°K (approximately the region covered in Fig. 2) is $1.6 \pm 0.3 \times 10^{-3}$ times smaller than the Brillouin intensity. The intensity of Brillouin scattering in liquid helium below the lambda point has been shown[6,7] to be given accurately by the theory of thermodynamic fluctuations[8]. When combined with this theoretical estimate of the Brillouin scattering intensity, our measurements indicate a differential extinction coefficient[8] for the Raman scattering into the direction and range of energy shifts mentioned above of $6 \pm 2 \times 10^{-12}$ cm^{-1} per unit solid angle. We can also give an estimate of the intensity of the scattering due to the roton portion of the dispersion curve alone. With an instrumental width of 3.6°K the ratio of the intensity at the maximum of the roton peak to that at the Brillouin line is $3.2 \pm 0.3 \times 10^{-4}$. The energy interval of 3.6°K in the spectrum at the roton peak corresponds to scattering from an interval in momentum of width 0.44 Å^{-1} centered at the roton minimum of the dispersion curve. The differential extinction coefficient for the scattering from these excitations in the geometry mentioned above would then be approximately $1.1 \pm 0.2 \times 10^{-12}$ cm^{-1} per unit solid angle. These figures for the extinction coefficient for Raman scattering in liquid helium are considerably larger than the initial estimates made by Halley,[2] but are in rough agreement with more recent calculations by Stephen.[5]

We have shown that light scattering is a useful tool for investigating the elementary excitations in liquid helium. The very high resolution which is available through conventional optical techniques will make possible measurements of certain features of the excitations, such as roton energies and lifetimes, which are more precise than those presently obtainable through neutron inelastic scattering. Also, isotopic mixtures of He^3 and He^4, as well as pure He^3, can be studied with this technique, whereas the neutron scattering studies of mixtures are limited by the strong absorption of the neutrons by the He^3.

We would like to thank Professor J. W. Halley for suggesting Raman scattering as a means of studying liquid helium, Professor M. Stephen for helpful discussions of the theory and a calculation of the extinction coefficient which indicated that the experiment was feasible, and Professor G. Benedek for many stimulating discussions. We also wish to thank Professor R. A. Smith, former director of the Center for Materials Science and Engineering, Massachusetts Institute of Technology, for making available the monochromator used in this experiment.

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