Experimental Evidence for a Two-Roton Bound State in Superfluid Helium

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(Received 29 October 1970)

We have used Raman scattering to study roton pairs in liquid helium at 1.2°K. We find that the energy required to create two rotons is less than twice the energy of a single roton. This result can be explained by the existence of a two-roton bound state. Comparison of our spectra with theoretical calculations gives a binding energy of (0.37 ± 0.10)°K and shows that the pair is in a D state ($L=2$) of angular momentum.

We have been using Raman scattering as a probe of the elementary excitations in superfluid helium.[1,2] In the scattering process two elementary excitations of equal and opposite wave vector are created in the helium.[3,4] The energy difference between the incident and scattered photons is equal to the energy required to create the pair. Therefore, the spectrum of the scattered light is determined by the density of pair states of the excitations as a function of energy, and by the coupling between the pairs and the light. The most prominent feature in our initial spectra was a peak due to the creation of two rotons. Equally noticeable was the absence of a peak corresponding to the creation of two excitations near the maximum of the dispersion curve. Ruvalds and Zawadowski[5] and Iwamoto[6] have shown that this absence is due to a depletion of the density of pair states in this region caused by an interaction between the elementary excitations. They predict that the same interaction, if it is present between rotons, would be attractive and could result in a two-roton bound state. We report here the results of higher-resolution studies of the roton portion of the spectrum. We find that at 1.2°K the two-roton peak occurs at an energy shift which is less than twice the energy of a single roton at the same temperature. This result, which cannot be explained on the basis of noninteracting rotons, provides direct evidence for the existence of a two-roton bound state. Our measurement of the depolarization ratio of the scattered light, when compared with the theory of the coupling between the light and the excitations, indicates that the bound state which we observe is a D state (angular momentum $L=2$). The exact position and shape of the peak in the Raman spectrum depends on the entire density of D states, unbound as well as bound, for the interacting roton pair. We here analyze our data in terms of a density of states based upon the simplest possible form for the roton-roton interaction which gives rise to a bound D state, and we arrive at a binding energy of (0.37 ± 0.10)°K for the roton pair.

The experimental arrangement is the same as that employed in our initial work except in place of the grating monochromator we now use a Fabry-Perot spectrometer whose free spectral range, 48.6°K, is about three times the shift of the two-roton Raman-scattered light. Figure 1 shows a typical experimental trace taken at a scattering angle of 90° with the electric field of the incident 4880-Å laser light pointing toward the spectrometer. The sharp spikes on the trace are interferometrically generated marker pulses used to obtain a precise measure of the energy shift. The peak on the right is the convolution of the instrumental profile with the two lines corresponding to Brillouin scattering from the phonons. The polarization of the laser light was purposely oriented to suppress this strong peak, and that fraction which still occurs in the trace is due to the finite acceptance angle, $f/2$, of the collection optics. The separation and width of the Brillouin lines are small and do not contribute appreciably to the width of this component of the spectrum. Therefore, this peak serves to determine both the zero of energy shift and the instrumental profile of the system. The instrumental width measured from this peak is 0.68°K and corresponds to an effective finesse of 71 for the interferometer. The much weaker peak shown on an expanded vertical scale at the left is caused by the two-roton Raman scattering. The sweep rate is increased in the interval between the Brillouin and Raman peaks, and this is indicated by the decreased spacing between the marker pulses. In the Raman region the detected photons are counted and displayed for 10-sec intervals, giving rise to the steplike nature of the experimental trace. The dot-dashed line is the background signal due in part to the incomplete extinction of the Brillouin components by the spec-

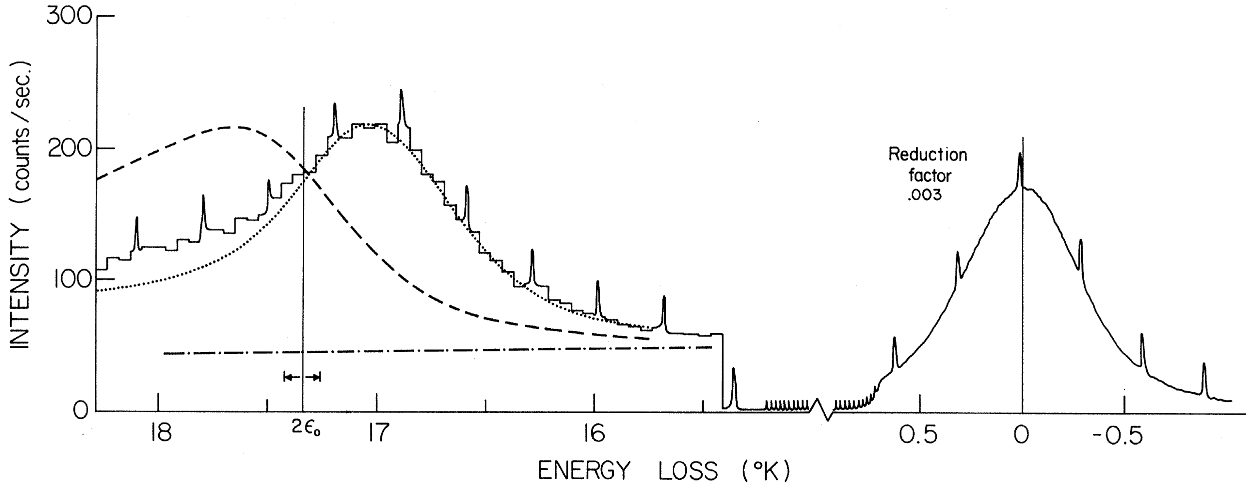

FIG. 1. The Raman spectrum of liquid He^4 at 1.2°K. The strong peak at zero energy shift is caused by Brillouin scattering and indicates the instrumental profile. The dotted curve is a theoretical fit to our data based on a two-roton bound state. The dashed curve would correspond to noninteracting rotons. The dot-dashed line is the background level.

trometer.

The energy shift corresponding to the maximum in the Raman spectrum, determined from the nine traces similar to the one shown, is (17.022 ± 0.027)°K. As a check of our determination of the energy shift we ran five traces with the 5145-Å argon laser line. We obtained the same result with a somewhat larger statistical uncertainty. The minimum energy ϵ_0 that a single roton can possess has been measured by neutron scattering to be (8.67 ± 0.04)°K.[7] In Fig. 1, a vertical line on our experiment trace indicates an energy shift of $2\epsilon_0 = (17.34 \pm 0.08)$°K. The difference, 0.32°K, between $2\epsilon_0$ and the energy shift at the maximum of our trace cannot be interpreted directly as the binding energy of a roton pair because the position of the maximum is also influenced by the lifetime of the excitations and the instrumental profile of the spectrometer. We can, however, compare our experimental spectrum with that which would arise in two simple cases. We first show that our result is qualitatively inconsistent with that expected for noninteracting but lifetime-broadened rotons. We then consider rotons interacting through a simple potential known to produce a bound D state and show that the energy difference mentioned above is, indeed, quite close to the binding energy of the pair.

The Raman spectrum of noninteracting rotons, each having an intrinsic energy width $\delta\epsilon$, has been considered by Stephen.[4] He finds that the spectrum at an energy shift E is proportional to the density of two-roton states, $\rho_2(\vec{K}=0, E)$, available to a pair whose total momentum $\vec{K}$ is zero. At zero temperature ρ_2 is given by

$$\rho_2(\vec{K}=0, E) = A(E-2\epsilon_0)^{-1/2}\Theta(E-2\epsilon_0), \qquad (1)$$

where $\Theta(x)$ is a unit-step function which is 1 for $x > 0$ and 0 for $x < 0$, and A depends on the mean wave vector k_0 and effective mass μ of the rotons: $A = (k_0/2\pi)^2\mu^{1/2}\hbar^{-1}$. ρ_2 at finite temperatures is given by the convolution of the zero-temperature result with a Lorentzian line of unit area and width $2\delta\epsilon$. By width we shall always mean full width at half-height. The maximum of the resulting curve occurs at an energy $(2/\sqrt{3})\delta\epsilon$ above $2\epsilon_0$. The convolution of this spectrum with the instrumental profile will also act to move the maximum toward increased energy shifts by an additional amount which is of the order of half of the instrumental width. For example, the dashed curve in Fig. 1 shows the spectrum which we would measure in the absence of roton-roton interactions for $\delta\epsilon = 0.15$°K (a value found to apply in the interacting case). From these considerations it is clear that a movement of the maximum to smaller energy shifts, as observed in our experiment, cannot be explained on the basis of noninteracting but lifetime-broadened rotons.

Ruvalds and Zawadowski[5] have developed a theory of the effects of roton-roton interactions on the single and pair excitation spectra of liquid helium. Their calculations are based on a model in which the interaction in k space between two rotons is of the separable form $U(\vec{k}, \vec{k}') = g_4$, where $\hbar(\vec{k}-\vec{k}')$ is the momentum transfer and g_4

is a coupling constant. This interaction gives rise to a single bound S state. In Stephen's theory[4] of the Raman scattering from liquid helium the light can couple only to the D-wave portion of the wave function for the roton pair,[8] and the scattered light should have the angular dependence and depolarization characteristic of D partial-wave scattering. The depolarization ratios which we measured for 90° scattering in our initial work[1] are indeed those given by this theory. Therefore we conclude that the bound state which we observe is a D state. Since the interaction U given above does not have a bound D state, Stephen has suggested[9] the use of an alternative separable interaction, $U(\vec{k},\vec{k}') = 5gP_2(\cos\theta_{kk'})$, where P_2 is a Legendre polynomial and $\theta_{kk'}$ is the angle between $\vec{k}$ and $\vec{k}'$. This is one of the simplest potentials which can give rise to a bound D state, and it has the additional advantage that the expressions for ρ_2 found by Ruvalds and Zawadowski will still apply with g_4 replaced by g. Therefore, we will use this interaction model to analyze our data. While this may not be a complete representation of the actual roton-roton interaction,[10] it does give a simple expression for the D-wave part of ρ_2 which we measure, and allows us to relate the position of the maximum in our spectra to the energy of the bound state.

Using the expressions of Ref. 5, we find that at zero temperature and for $g<0$, which corresponds to an attraction between the rotons, ρ_2 reduces to

$$\rho_2(K=0,E) = 2\pi A E_B^{1/2}\delta(E-2\epsilon_0+E_B) + A(E-2\epsilon_0)^{1/2}(E-2\epsilon_0+E_B)^{-1}\Theta(E-2\epsilon_0). \tag{2}$$

The δ function in Eq. (2) represents a single bound state with a binding energy $E_B = A^2g^2$ and which, for our form of the interaction, has $L=2$ symmetry. The formation of a bound state in this system is the analog of the Cooper problem for interacting electrons.[11] The remaining states are unbound and it can be seen from Eq. (2) that for $E-2\epsilon_0 \gg E_B$ their density approaches that found for noninteracting rotons. When $g>0$ the bound state disappears, but the density of unbound states is the same as that given by Eq. (2). The theory of Ruvalds and Zawadowski includes lifetime effects only through the individual roton linewidths $\delta\epsilon$ and it can be shown that, as in the case of noninteracting rotons, their expression for ρ_2 at finite temperatures can be obtained by convolving Eq. (2) with a Lorentzian of unit area and width $2\delta\epsilon$. It should be noted that at sufficiently low temperatures the lifetime of the bound state would not be expected to be determined solely by the free lifetimes of its constituent excitations, and this method of dealing with lifetime effects would no longer be applicable.

We have compared the spectra computed from this model with our experimental traces by varying the binding energy and the single roton linewidth and we find that the best fit is obtained when $E_B = 0.37$°K and $\delta\epsilon = 0.15$°K. The resulting theoretical spectrum is represented by the dotted curve in Fig. 1. This value of E_B corresponds to $g = -1.2\times10^{-39}$ cm^3 ergs, which is in good agreement with the value $g_4 = -1.0\times10^{-39}$ cm^3 ergs estimated by Ruvalds and Zawadowski[5] from the suppression of the scattering at the maximum of the dispersion curve. By studying the temperature dependence of the width of the two-roton peak at higher temperatures we have measured[2] roton linewidths above 1.3°K where they are determined by roton-roton collisions. An extrapolation of these results to our temperature of 1.2°K gives a contribution to $\delta\epsilon$ from this mechanism alone of 0.07°K, in fair agreement with our estimate here of the actual width. The main source of error in our determination of both E_B and $\delta\epsilon$ is the uncertainty in the neutron measurements of $2\epsilon_0$. In arriving at the above fit we used $2\epsilon_0 = 17.34$°K. However, when we use a smaller value of $2\epsilon_0$ within the neutron uncertainty we find that the fit to our spectra is improved in the region above 17.5°K, and the corresponding optimum values of E_B and $\delta\epsilon$ decreases in magnitude. This leads us to believe that with higher resolution experiments we may be able to determine both E_B and ϵ_0 self-consistently. At present these factors are the major source of our uncertainty of ±0.10°K in both E_B and $\delta\epsilon$.

We have shown that pairs of rotons from at least one bound state, and that Raman scattering measurements at low temperatures, can be used to determine the symmetry, binding energy, and lifetime of this state. In addition, roton-roton collision times can be determined from the temperature dependence of the Raman line shape at higher temperatures.[2] These experiments can play a central role in establishing the detailed nature of the roton-roton interaction.

We would like to thank Professor M. Stephen and Professor J. Ruvalds for valuable discussions of the theory.

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*Work supported by the Advanced Research Projects Agency under Contract No. DAHC 15-67-C-0222.

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Temperature Dependence of Normal Modes in a Nematic Liquid Crystal

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(Received 16 October 1970)

We have measured the temperature dependence of the intensity and spectrum of light scattered by fluctuations in the nematic phase of *p*-methoxy-benzylidene *p*-*n*-butylaniline. Our measurements are interpreted using two phenomenological theories of nematic liquid crystals.

In this Letter we report the results of an experimental study of the temperature dependence of the intensity and spectrum of light scattered by fluctuations in the ordered phase of the nematic liquid crystal *p*-methoxybenzylidene *p*-*n*-butylaniline (MBBA). From these measurements we determine the mean squared amplitude and time dependence of the normal modes of the ordered phase as a function of temperature. We first analyze our result using the phenomenological theory of the Orsay Liquid Crystal Group[1] to determine the temperature dependence of the elastic constants and viscosity coefficients of the model. Recently an attractively simple macroscopic model of liquid crystals has been proposed by Martin, Pershan, and Swift[2]; we analyze our data to indicate how the normal-mode displacements of this theory couple to the dielectric constant of the liquid crystal.

The order in a nematic liquid crystal of uniaxial symmetry is specified by the direction of the alignment of the liquid-crystal molecules (the optic axis) and their degree of alignment along this direction. The Cartesian dielectric-constant tensor for a uniaxial nematic may, therefore, be written

$$\delta\epsilon_{\alpha\beta}=\bar{\epsilon}\delta_{\alpha\beta}+Q(\Delta\epsilon/3)(3n_\alpha n_\beta-\delta_{\alpha\beta}), \qquad (1)$$

where n_α and n_β are the Cartesian components of a unit vector (called the director) parallel to the local optic axis. An order parameter specifying the degree of alignment is[3] $Q=\frac{3}{2}\langle\cos^2\theta-\frac{1}{3}\rangle$, where θ is the angle between the long axis of a molecule and the local optic axis. The other quantities are $\bar{\epsilon}=\frac{1}{3}(\epsilon_p+2\epsilon_t)$ and $\Delta\epsilon=\epsilon_p-\epsilon_t$, where ϵ_p and ϵ_t are the dielectric constants parallel and transverse, respectively, to the optic axis of a completely ordered sample ($Q=1$). The value of Q is temperature dependent and is given approximately by the mean-field model of Maier and Saupe.[3] A uniform vector field $\vec{n}$ describes the lowest energy state of the ordered liquid.

The value of $\Delta\epsilon$ in (1) is of order unity; the order in a liquid crystal therefore has a pronounced effect upon the index of refraction, and optical methods are ideally suited to study these materials. Fluctuations in Q will scatter light and have been studied in the isotropic phase of MBBA by Stinson and Litster.[4] In the ordered liquid the strongest light scattering results from fluctuations in $\vec{n}$; these are normal modes analogous to spin waves in a ferromagnet. From the intensity and spectrum of light scattered by these fluctuations one may determine the mean squared amplitude and time dependence of the normal modes in the liquid crystal. The intensity of light scattered by these modes in *p*-azoxyanisole (PAA) has been studied by Chatelain[5]