

neiro, to be published.

<sup>12</sup>P. W. Anderson and H. Suhl, *Phys. Rev.* **116**, 898 (1959).

<sup>13</sup>N. R. Werthamer, in *Superconductivity*, edited by

R. D. Parks (Dekker, New York, 1969), p. 321.

<sup>14</sup>W. F. Brinkman and P. W. Anderson, to be published.

<sup>15</sup>R. E. Webb, T. J. Greytak, R. T. Johnson, and J. C. Wheatley, *Phys. Rev. Lett.* **30**, 210 (1973).

## Two-Roton Raman Scattering in He<sup>3</sup>-He<sup>4</sup> Solutions

C. M. Surko and R. E. Slusher

*Bell Laboratories, Murray Hill, New Jersey 07974*

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The two-roton Raman spectra of superfluid solutions of He<sup>3</sup> and He<sup>4</sup> are measured at a temperature of 1.3 K for molar concentrations of He<sup>3</sup> up to 31%. At 31% He<sup>3</sup> concentration, the shift in the roton energy is inferred from these spectra to be  $+0.3 \pm 0.5$  K, in striking contrast to the  $-3.5$ -K shift inferred from previous measurements of normal-fluid density. The width of the two-roton peak is resolved and is a measure of the He<sup>3</sup>-roton interaction.

In spite of extensive study of the roton excitation in superfluid helium, many facets of its behavior remain to be explained. In particular, the effect of He<sup>3</sup> impurities on the phonon-roton dispersion relation of superfluid helium has been studied both experimentally and theoretically.<sup>1-5</sup> It has recently been predicted<sup>4,5</sup> that the addition of a few percent He<sup>3</sup> would drastically modify the phonon-roton curve by essentially hybridizing the phonon-roton branch and the He<sup>3</sup> quasiparticle branch. Experiments involving fourth-sound propagation,<sup>1</sup> ion mobilities,<sup>2</sup> and an "oscillating disk-stack" measurement of normal-component density<sup>3</sup> have been used to infer a significant decrease in the roton energy  $\Delta$  in superfluid solutions of He<sup>3</sup> and He<sup>4</sup>. Specifically,  $\Delta$  was found to decrease from 8.5 to 5 K when the molar concentrations of He<sup>3</sup> was increased from 0 to 30%. These experiments, however, do not investigate the roton directly, but measure quantities such as normal-fluid density. It is, therefore, particularly desirable to study the roton in these solutions with a more direct probe. Since neutron scattering is difficult in strong solutions of He<sup>3</sup> because of the large nuclear absorption cross section, inelastic light scattering is a unique choice for such a study. Since the light has a small wave vector compared to that of the roton, light couples only to pairs of rotons with nearly equal and opposite wave vector. The high density of states near the roton minimum produces a sharp peak in the spectrum of the scattered light at a frequency shift corresponding to the energy of the roton pair.<sup>6</sup> We report here measurements of two-roton Raman spectrum in He<sup>3</sup>-He<sup>4</sup> solu-

tions containing up to 31% molar concentration of He<sup>3</sup>. In striking contrast to the work referred to above, we infer an increase of  $0.6 \pm 1$  K in the energy necessary to create a pair of rotons. An increase in the two-roton linewidth due to the He<sup>3</sup>-roton interaction was also measured and was found to be approximately linear with He<sup>3</sup> concentration.

The experimental apparatus has been described previously,<sup>7</sup> except that for these experiments the sample cell was made from beryllium-copper. An argon-ion laser beam at 5145 Å with power of 100 to 200 mW is focused through indium-sealed 0°-sapphire windows into the sample cell. Depolarized scattering at 90° to the incident beam is collected with  $f/3$  optics, analyzed with a 0.75-m double-grating spectrometer, and detected with a cooled Channeltron photomultiplier. The noise in the spectra is predominantly statistical arising from the low level of scattered light ( $\sim 10$  counts per second per resolution interval). The possibility that laser heating might cause "heat flush" effects and thereby decrease the concentration of He<sup>3</sup> in the laser beam was checked by observing that the spectrum of scattered light is independent of laser power when the sample cell is maintained at a fixed temperature.

In Fig. 1 is shown the Stokes-shifted Raman spectrum of superfluid helium for several concentrations of He<sup>3</sup>. These spectra were taken at a temperature of 1.30 K with an instrumental resolution [half width at half-maximum (HWHM)] of  $0.75 \text{ cm}^{-1}$ . The peak at a frequency shift of approximately 17.5 K is due to scattering from

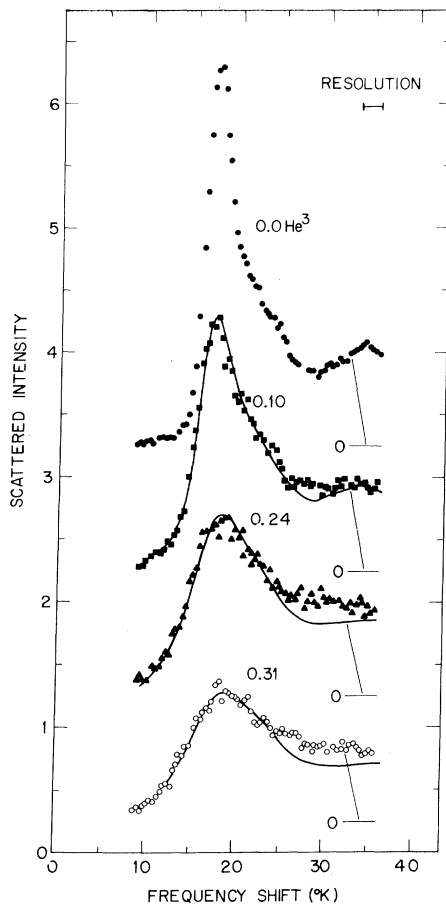


FIG. 1. Two-rotor Raman scattered intensity of superfluid  $\text{He}^4$  containing the indicated molar concentrations of  $\text{He}^3$ , shown as a function of frequency shift of the scattered light. The ordinate is in arbitrary units with the zero shifted for each concentration. See text for details.

two rotors.<sup>6</sup> Qualitatively, as the concentration of  $\text{He}^3$  is increased this peak increases in width and remains at nearly the same position, while the broad spectrum on the high-frequency side of the roton is unaffected in spectral shape and intensity. To analyze the data quantitatively we Lorentz broaden each point of the pure  $\text{He}^4$  spectrum and fit the finite-concentration  $\text{He}^3$  data using the Lorentz width, the peak position, and the overall intensity as adjustable parameters.<sup>8</sup> This least-squares fit is carried out for frequency shifts between 8 and 25 K, where the two-rotor spectral weight is the dominant contribution to the spectrum. The resulting curves are shown by the solid lines in Fig. 1. Figure 2 shows as a function of  $\text{He}^3$  concentration the roton peak position  $\Delta$  and the Lorentzian width  $\Gamma$  which re-

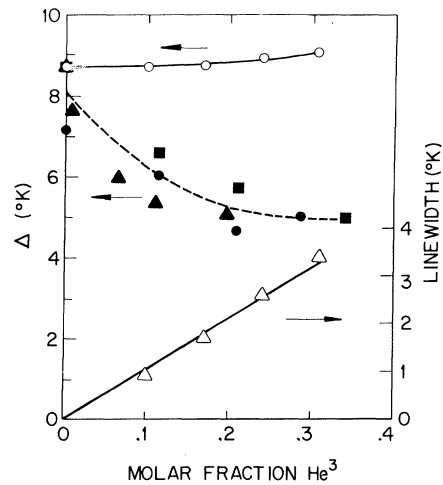


FIG. 2. Open circles, roton energy  $\Delta$  as a function of  $\text{He}^3$  concentration inferred from two-rotor Raman scattering; open triangles,  $\text{He}^3$  contribution to the two-rotor linewidths (HWHM). Solid circles, squares, and triangles: values of  $\Delta$  inferred from fourth-sound, oscillating-disk, and ion-mobility measurements, respectively. The solid and dashed lines are guides to the eye and have no special significance.

sult from this analysis. Here  $\Delta$  is taken as one-half of the position of the two-rotor peak of the pure  $\text{He}^4$  spectrum at 1.30 K after broadening and translation along the energy axis to best fit the finite  $\text{He}^3$  concentration data. The linewidth shown is HWHM for the two-rotor peak. The error in  $\Delta$  is estimated to be  $\pm 0.2$  K at 10%  $\text{He}^3$  and increases to  $\pm 0.5$  K at 31%. The error in linewidth is  $\pm 10\%$  of the value shown.

Shown for comparison in Fig. 2 is the value of  $\Delta$  inferred from oscillating-disk,<sup>3</sup> fourth-sound,<sup>1</sup> and ion-mobility<sup>2</sup> data. The two-rotor Raman-scattering inference of  $\Delta$  as a function of  $\text{He}^3$  concentration is clearly in qualitative disagreement with these measurements. The origin of this discrepancy is not immediately obvious. There are several points at which one might question the analyses which lead to this discrepancy. There is no precise theory of ion mobilities (particularly in superfluid He containing such strong concentrations of  $\text{He}^3$ ), so that it is difficult to derive a value of  $\Delta$  from these measurements. The oscillating-disk and fourth-sound measurements involve inference of  $\Delta$  from the normal-fluid density, and at these high  $\text{He}^3$  concentrations, the assumed separation of the normal-fluid density into a  $\text{He}^3$  component and a roton component cannot be rigorously justified. In spite of these reservations, the three separate

experiments do give a consistent shift in  $\Delta$  as a function of  $\text{He}^3$  concentration. With regard to the Raman-scattering measurements of  $\Delta$ , the coupling of light to the liquid is rigorously proportional to a four-density correlation function. The common assumption that this can be approximated as coupling to pairs of elementary excitations has no firm theoretical justification and may be inappropriate. Even within the framework of this assumption, it should be mentioned that light only couples to pairs of rotons and only to those in an  $l=2$  angular-momentum state,<sup>6</sup> and it is conceivable that this selective coupling might also lead to the discrepancy. Finally, there is evidence that the light creates the roton pair in close proximity in a bound state,<sup>6</sup> which may also affect the concentration dependence of  $\Delta$ .

The linewidth  $\Gamma$  shown in Fig. 2 is a measure of the  $\text{He}^3$ -roton interaction and is approximately linearly proportional to concentration. It is difficult to relate  $\Gamma$  simply to a roton- $\text{He}^3$  cross section  $\sigma_{3r}$  for several reasons. First of all, at these strong concentrations the  $\text{He}^3$ -roton mean free path is comparable to the distance between  $\text{He}^3$  atoms. Secondly, if the rotons are formed in a bound state this effect must be taken into account. Finally, any dependence of  $\sigma_{3r}$  on the roton or the  $\text{He}^3$  momentum, as has been suggested recently,<sup>9,10</sup> must be accounted for. If, however,  $\sigma_{3r}$  is estimated as  $\sigma_{3r} = 2\pi\Gamma/n_3\bar{v}_3$ , where  $n_3$  is the number density of  $\text{He}^3$  and  $\bar{v}_3$  is the average thermal velocity of a  $\text{He}^3$  quasiparticle with an effective mass of 2.0  $\text{He}^4$  atoms, we find  $\sigma_{3r} \approx 0.7 \times 10^{-14} \text{ cm}^2$ . This is a factor of 2 less than that deduced from transport measurements.<sup>9</sup> Inclusion of a nonzero roton velocity would increase the  $\text{He}^3$  velocity relative to that of the roton thereby decreasing  $\sigma_{3r}$ .

The integrated intensity of the two-roton contribution to the scattered spectrum decreases by about 30% as the  $\text{He}^3$  concentration is increased to 30%. The shapes of the spectra shown in Fig. 1 exhibit very little change as a function of  $\text{He}^3$  concentration at energies greater than that of the two-roton peak. In particular, just as in the pure fluid, we see no increase in scattering from the maximum in the phonon-roton dispersion relation which would be expected to occur near 28 K. Also, we do not observe any contribution to the spectrum at energies less than  $2\Delta$  which could be interpreted as arising from excitations with energy values comparable to those inferred from the normal-fluid data.<sup>1-3</sup>

The spectra show no evidence of the predicted

hybridization<sup>4,5</sup> of the phonon-roton excitations and  $\text{He}^3$  quasiparticle excitations. Such an effect could be expected to lead to two peaks in the spectrum and/or a sizable shift in  $\Delta$ . Reference 4 makes specific predictions for 1%  $\text{He}^3$  concentration at 1.25 K.<sup>11</sup> The Raman spectrum of a 1% solution was measured at 1.30 K and found to exhibit no measurable shift or broadening of the two-roton peak, consistent with the solid curves in Fig. 2 and in apparent disagreement with these predictions.

In summary, we have measured the two-roton spectrum in superfluid helium in the presence of molar concentrations of  $\text{He}^3$  up to 31%. The position of the two-roton peak is very nearly constant, and a measurable broadening is observed. These data for peak position are in apparent contradiction with the simplest interpretation of previous measurements of  $\Delta$  as a function of  $\text{He}^3$  concentration. The data presented above for the linewidth  $\Gamma$  are a new measure of the  $\text{He}^3$ -roton interaction and should prove useful as a guide to its theoretical description.<sup>12</sup>

We wish to acknowledge helpful conversations with T. J. Greytak, P. Hohenberg, J. Ruvalds, N. R. Werthamer, and V. K. Wong; and the cooperation of T. J. Greytak in coordinating the publication of a similar experimental study<sup>13</sup> of the roton in  $\text{He}^3$ - $\text{He}^4$  solutions.

<sup>1</sup>N. E. Dyumin, B. N. Esel'son, E. Ya. Rudavskii, and I. A. Serbin, *Zh. Eksp. Teor. Fiz.* **56**, 747 (1969) [*Sov. Phys. JETP* **29**, 406 (1969)].

<sup>2</sup>B. N. Esel'son, Yu. Z. Kovdrya, and V. B. Shikin, *Zh. Eksp. Teor. Fiz.* **59**, 64 (1970) [*Sov. Phys. JETP* **32**, 37 (1971)].

<sup>3</sup>V. I. Sobolev and B. N. Esel'son, *Zh. Eksp. Teor. Fiz.* **60**, 240 (1971) [*Sov. Phys. JETP* **33**, 132 (1971)].

<sup>4</sup>D. L. Bartley, J. E. Robinson, and V. K. Wong, to be published.

<sup>5</sup>T. Soda, to be published.

<sup>6</sup>T. J. Greytak and J. Yan, *Phys. Rev. Lett.* **22**, 987 (1969); T. J. Greytak, J. Woerner, J. Yan, and R. Benjamin, *Phys. Rev. Lett.* **25**, 1547 (1970).

<sup>7</sup>R. E. Slusher and C. M. Surko, *Phys. Rev. Lett.* **27**, 1699 (1971).

<sup>8</sup>Two comments should be made concerning this analysis. Firstly, the actual spectrum should first be Lorentz broadened and then convoluted with the spectrometer resolution function. Model calculations indicate that our procedure of Lorentz broadening the instrument-broadened spectrum introduces errors of less than 1% and hence is a good approximation. Secondly, at the temperature of 1.3 K the change in the spectrum produced by inclusion of the Planck distribution of thermal excitations [M. J. Stephen, *Phys. Rev.* **187**, 279

(1969)] is in all cases smaller than our random error (e.g., less than 0.2% for frequency shifts greater than 12 K), and we have therefore neglected this factor.

<sup>9</sup>G. A. Herzlinger and J. G. King, *Phys. Lett.*, **40A**, 65 (1972).

<sup>10</sup>H. T. Tan and C.-W. Woo, *Phys. Rev. Lett.*, **30**, 365 (1973).

<sup>11</sup>The theories of Refs. 4 and 5 do not predict Raman spectra, but only the dynamic structure factor  $S(k, \omega)$ . We have assumed that the changes in  $S$  will lead to mod-

ification of the Raman spectra. The authors of Ref. 1 point out that their assumption of the validity of quantum hydrodynamics is untested. The failure of this assumption could be the origin of the discrepancy between theory and experiment.

<sup>12</sup>The Raman data for both  $\Delta$  and  $\Gamma$  are in reasonable agreement with very recent theoretical estimates by A. Bagchi and J. Ruvalds, to be published.

<sup>13</sup>R. L. Woerner, D. A. Rockwell, and T. J. Greytak, following Letter [*Phys. Rev. Lett.*, **30**, 1114 (1973)].

## Measurement of Roton Energies and Linewidths in He<sup>3</sup>-He<sup>4</sup> Solutions\*

R. L. Woerner, D. A. Rockwell, and T. J. Greytak†

*Department of Physics, Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*

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Using Raman scattering from rotons in liquid He<sup>3</sup>-He<sup>4</sup> solutions for He<sup>3</sup> concentrations  $0 \leq X \leq 0.11$  at  $T = 0.60^\circ\text{K}$ , we have measured the two-roton energy and linewidth. Our measurements indicate no change in the single-roton energy as a function of concentration, whereas the single-roton linewidth increases nonlinearly with  $X$ .

Although neutron scattering has provided detailed information on the elementary excitations in pure liquid He<sup>4</sup>,<sup>1</sup> this technique has not been applied to solutions of He<sup>3</sup> in He<sup>4</sup> because of the very strong absorption of neutrons by the He<sup>3</sup> atoms. Therefore, only indirect information has been available about the influence of the He<sup>3</sup> atoms on one of the most interesting excitations in the pure He<sup>4</sup>, the roton. Measurements of fourth-sound velocity,<sup>2</sup> ion mobility,<sup>3</sup> and normal-fluid fraction<sup>4</sup> in the solutions all infer that the roton energy  $\Delta(T, X)$  first decreases rapidly with increasing He<sup>3</sup> molar concentration  $X$  ( $\Delta^{-1} d\Delta/dX \sim -3$  near  $X=0$ ) and then levels out at concentrations  $X \sim 0.2$ . From values of the He<sup>3</sup>-roton scattering cross section  $\sigma$  obtained from measurements of the diffusion of He<sup>3</sup> atoms in the solutions,<sup>5</sup> estimates of the concentration dependence of the roton linewidth  $\Gamma(T, X)$  can be made. We report here direct measurements by Raman scattering of the energy and linewidths of zero-momentum pairs of rotons at a temperature of  $0.60^\circ\text{K}$  and concentrations  $0 \leq X \leq 0.11$ . In contrast with previous results, we find that  $\Delta$  is essentially independent of concentration in this region. The linewidth we measure is consistent with simple order-of-magnitude estimates based on  $\sigma$ . In addition, our measurements indicate a nonlinear dependence of  $\Gamma$  on  $X$ .

The experimental apparatus is the same as

that used in earlier experiments<sup>6,7</sup> except that the liquid-helium sample is now cooled by a continuous He<sup>3</sup> refrigerator. The spectrometer is a pressure-swept flat Fabry-Perot type with a free spectral range of  $48.6^\circ\text{K}$  and an instrumental full width of  $0.70^\circ\text{K}$  ( $1^\circ\text{K} = 0.6952 \text{ cm}^{-1}$ ). The mixtures were prepared in gaseous form at room temperature and then condensed into the scattering cell at a temperature below  $0.30^\circ\text{K}$ . The cell was sealed for an experimental run during which the solution was at saturated vapor pressure. As a check for heat flush, the  $5145\text{-\AA}$  incident laser power was varied between 300 and 750 mW. No changes in the measured spectrum were observed.

The most prominent feature in the spectrum of Raman scattering from pure liquid He<sup>4</sup> is a peak corresponding to the creation of a pair of rotons with zero total momentum.<sup>6</sup> The location and width of this peak are related to the energy and linewidth of the pair state. Figure 1 shows typical experimental spectra in the vicinity of the two-roton peak at  $0.6^\circ\text{K}$  for three different concentrations. Note that the position of the two-roton peak is essentially unchanged by the addition of He<sup>3</sup>, whereas the broadening of this peak is quite evident.

Quantitative values for the changes in the energy and linewidth of the two-roton state were obtained as follows.<sup>7</sup> An experimental trace tak-