RECENT NEUTRON STUDIES OF ELEMENTARY EXCITATIONS IN LIQUID 3He AND 4He

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Résumé.- La diffusion inélastique des neutrons est un outil particulièrement puissant pour étudier les excitations de l'hélium-4 superfluide. Au cours des dernières années, ces études ont été étendues à l'isotope ^{3}He très absorbant. Nous présentons ici des travaux récents sur ^{4}He et ^{3}He, ainsi que sur les mélanges des deux isotopes

Abstract.- Neutron inelastic scattering has been shown to be a particularly powerful tool for the study of the excitations of superfluid ^{4}He. In the last few years, these studies have been extended to the highly absorbing isotope ^{3}He. In this paper, recent neutron work on both ^{4}He and ^{3}He, and on mixtures of the two isotopes, is discussed.

1. INTRODUCTION.- After more than twenty years of intensive study by neutron inelastic scattering (N. I. S.) methods, probably more is known about the behaviour of the elementary excitations of superfluid ^{4}He than for any other material. Why there should be this continuing interest is not difficult to explain. Firstly, there is the innate attraction of this unique substance. The hope that neutron scattering would help provide a microscopic understanding of the phenomenon of superfluidity, however, has gone largely unfulfilled. But there is also the interest in the study of the elementary excitations themselves and in their interactions. ^{4}He may be considered as a "model" substance for the study of excitations in solids as there is only one well-defined mode to consider, and, of course, helium is isotropic. The excitations have energies of a few meV, (1meV$\simeq$11.6 K) well-matched to the energies of thermal neutrons, while the inter-atomic distances are very similar to the wavelengths of thermal neutrons. Although neutron scattering yields information complementary to that obtained by other techniques, for instance ultrasonic attenuation measurements, it is the only method by which detailed dynamic information may be obtained at values of wavevector Q greater than about 0.1 Å^{-1}.

The very first N.I.S. results on ^{4}He were reported in 1957 by Palevsky and his colleagues /1/, and demonstrated directly the existence of the "roton" excitations predicted some years earlier by Landau /2/. The work of several groups, in particular that at A.E.C.L., Chalk River /3/, has since contributed to our knowledge of the "phonon-roton" dispersion curve of superfluid ^{4}He. Current work is attempting to supply the missing detail, particularly in the field of the interactions between these excitations. The most recent advances in these studies have been concerned with the excitations in ^{4}He films and with the excitations of liquid ^{3}He and of mixtures of ^{3}He and ^{4}He, until lately considered too difficult to observe with neutrons.

As an introduction to a discussion of some of the recent experiments, we shall outline briefly the techniques employed in N.I.S. studies of liquid helium.

The usual aim of a neutron scattering experiment is to obtain knowledge about the Van Hove scattering function $S(Q,\omega)$ over some range of wavevector Q and frequency ω. $S(Q,\omega)$ describes the fluctuations in the density of the system and is directly related to the dynamical susceptibility $\chi(Q,\omega)$ which describes the response of the system to an external perturbation. The quasi-momentum transfer $Q = |\vec{Q}|$ is given by the difference between the wavevectors of the incident neutron beam, $\vec{k}_o$, and the scattered beam $\vec{k}_F$. Thus

$$\vec{Q} = \vec{k}_o - \vec{k}_F$$

and the energy transferred to the sample is

$$\hbar\omega = (\hbar^2/2M_n)\ (k_o^2 - k_F^2)$$

where M_n is the neutron mass. Thus by defining both $\vec{k}_o$ and $\vec{k}_F$ we may obtain information on the scattering function by measuring the intensity of the scattered neutron beam. The two principal instruments employed to study the inelastic scattering from liquid helium are the triple-axis crystal spectrometer and the time-of-flight (t.o.f.) spectrometer.

In the first of these, a monoenergetic neutron beam is obtained by Bragg reflection from a suitable single crystal. The energy of the neutrons scattered by the sample is then measured using a second single crystal, suitably oriented. Figure 1 shows a diagram of the IN3 triple-axis spectrometer at I.L.L., used for some of the measurements to be discussed in section 2.

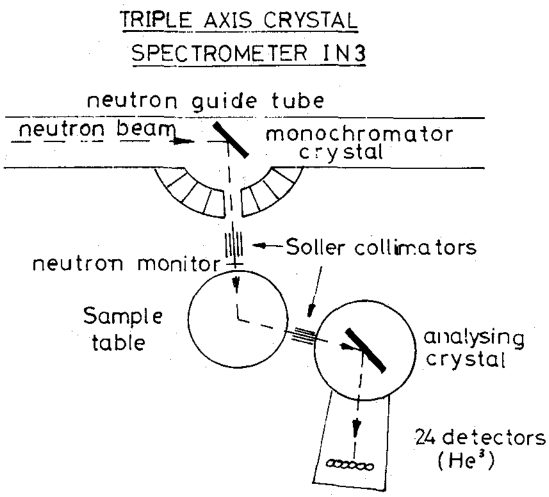

Fig. 1 : Schematic diagram of the IN3 triple-axis crystal spectrometer (I.L.L.).

The particular advantage of this technique is that the energy spectrum of the neutrons scattered by a sample can be determined at preselected constant wavevectors, Q. But normally only one angle of scatter at the sample can be examined at a time. This mathod is especially useful where the sample has a well-defined ("sharp") dispersion relation $\hbar\omega(Q)$; much of the detailed measurement of the phonons and rotons of superfluid ^{4}He has been made in this way (see for example reference /3/).

Time-of-flight spectrometers normally use either a single crystal or a mechanical rotor system to define the energy of the incident neutron beam while the energy of the scattered beam is determined by measuring the time-of-flight of the neutrons from the sample to the detector bank. In figure 2, we present a diagram of the IN5 time-of-flight spectrometer at the I.L.L., used for several of the experiments discussed in sections 2 and 3. Although many different scattering angles can be examined simultaneously, both $\vec{Q}$ and ω vary over the measured spectrum. Where $S(Q,\omega)$ does not exhibit sharp delta-function features, the t.o.f. technique may be many times more efficient than the triple-axis method. All the measurements to date on liquid ^{3}He have been made using time-of-flight spectrometers.

Since the aim of this paper is to discuss some of the more recent work on helium, and not to provide an exhaustive review, we refer the reader who desires a more detailed treatment to several recent reviews.

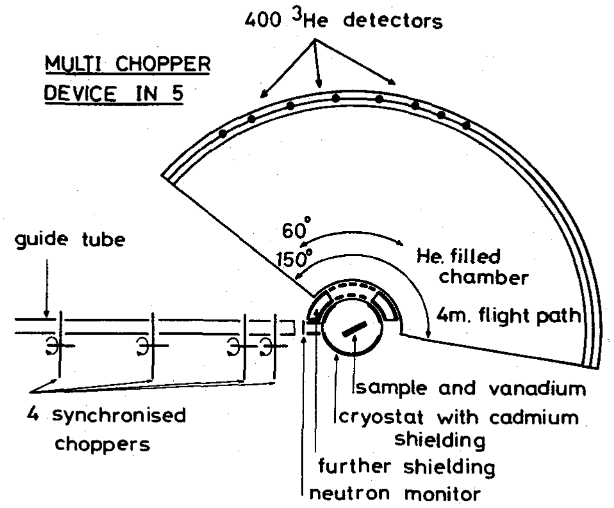

Fig. 2 : Schematic diagram of the IN5 multichopper time of flight spectrometer (I.L.L.)

Woods and Cowley, 1973 /4/, review work on the excitations in liquid ^{4}He, while ^{3}He is included in the articles of Price, 1976 /5/, Lovesey and Copley, 1977 /6/ and Cowley, 1976 /7/ and 1977 /8/.

2. BULK ^{4}He AND ^{4}He FILMS.-One of the few really new topics in neutron research on helium is the study of thin films. In this section we shall describe the observation of roton excitations in films only a few atomic layers thick. Returning to the bulk liquid we describe a few experiments which have attempted to increase our detailed knowledge of the excitation spectrum of ^{4}He and of the interactions between these excitations.

Figure 3 represents the well-known phonon-roton dispersion curve of superfluid ^{4}He. There have been recent neutron studies of the excitations in the phonon region A, the roton region B, and in region C where the one-phonon excitations merge into the band of multiphonon excitations.

It has been known for many years that the behaviour of liquid ^{4}He is different in confined geometry to that in the bulk.

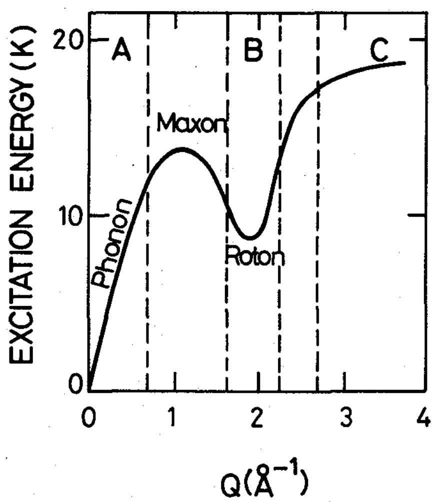

Fig. 3 : The phonon-roton excitation curve of superfluid ^{4}He. The dashed lines represent an arbitrary separation into "phonon", "roton", and "large wavevector" regimes.

For instance the temperature of the superfluid transition is lower for helium in narrow channels than for bulk helium /9/. It has been suggested that the roton energy of constricted helium should decrease and that new excitations - "ripplons" - might occur at the free surface of the film /10/. To date, two neutron experiments have been reported which investigate these effects. Both experiments studied superfluid films on exfoliated graphite.

In the experiments of Carneiro et al. /11/, sharp excitations at Q = 2.0 Å^{-1} could be observed for coverages down to less than 6 atomic layers. Within experimental uncertainty, the energies of these roton excitations were found to be identical to those of bulk ^{4}He. No additional scattering was observed which could be attributed to ripplons. Lambert et al. /12/ came to very similar conclusions. Roton peaks were observed with a decrease in energy of only about 0.24 K compared with the bulk roton energy of 8.62 K. In both experiments the lineshapes were asymmetric and this was interpreted /11/ as arising from out-of-plane contributions to the scattering. There was also agreement in the value of the coverage, 2 1/2 layers, at which the integrated roton scattering falls to zero. So it seems that the liquid film lies on a layer of solid ^{4}He, 2 1/2 atomic layers thick. Thus, even for films of only 3 1/2 atomic layers, bulk behaviour predominates. It is certain that in the near future there will be further neutron experiments on ^{4}He films.

Even in bulk helium there are still outstanding problems. There is considerable interest in the "anomalous" or "positive" dispersion of the phonon dispersion curve for wavevectors below 1 Å^{-1}. If there is upwards dispersion, then the decay of one phonon into two others is possible, and so the shape of the dispersion curve controls the decay processes which can occur. This unusual behaviour has been extensively studied using a variety of techniques - see the review by Maris /13/. However, although it is now generally accepted that the phase velocity ω/Q does exceed the sound velocity C_o at some finite wavevector Q, the evidence was somewhat indirect. In principle, neutron scattering allows the direct determination of $\omega(Q)$. But neutron measurements on phonons in ^{4}He are fairly difficult since normally rather small scattering angles must be employed and so there are problems with the experimental geometry. Further, for wavevectors $\sim$ 0.5 Å^{-1}, the one phonon intensity is only about 10 % of its maximum value at the roton minimum /3/. The experimental data of Cowley and Woods /3/ at s.v.p. and of Svensson et al. /14/ at pressures up to 24 atm. are consistent with the existence of anomalous dispersion at pressures of up to 17 atm. but, taken alone, are not sufficiently accurate to demonstrate this phenomenon. For this reason, the phonon region was reexamined /15/ using a t.o.f. spectrometer with low energy neutrons to provide many points (ω,Q) on the dispersion curve, with improved resolution. In this way, about 40 phonon frequencies were determined between 0.1 Å^{-1}and 0.9 Å^{-1}. The measured phase velocities are presented in figure 4. It is clear that the phase velocity does increase from its long wavelength limit, with a maximum deviation of about 4 % at a wavevector of about 0.42 Å^{-1}. These measurements confirm the existence of anomalous dispersion in superfluid ^{4}He. The predicted functional forms of Maris /13/ and Aldrich et al. /16/ are shown as the lines of figure 4. Both the parametrized expression of Maris, fitted to the data of Cowley and Woods /3/, and the polarization potential calculation of Aldrich et al. /16/ agree well with these experimental results.

We turn now to recent results at larger wavevectors. For several years there has been speculation as to the existence of a two-roton bound state /17/. High precision Raman scattering /18/

which measures the scattering from pairs of rotons with approximately equal and opposite momenta, has shown the existence of a peak at 16.97 ± .03 K. This has to be compared with twice the roton energy Δ as measured by neutron scattering. Cowley and Woods /3/ measured $\Delta/k = 8.67 \pm .04$ K in 1971. So the two-roton Raman peak occurs at an energy slightly lower than 2Δ.

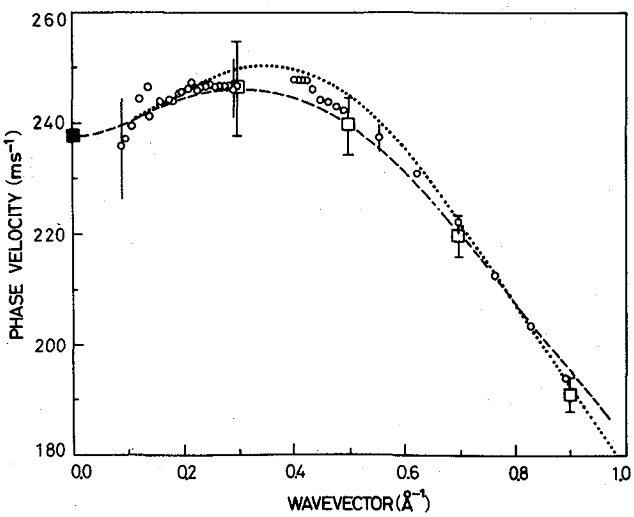

Fig. 4 : Measured phase velocities in superfluid ^{4}He at 1.2 K /15/. The solid square is the experimental sound velocity and the circles are the measurements of reference /15/. The open squares are representative data of Cowley and Woods /3/. The dashed curve is calculated using the parameters of Maris /13/ while the dotted curve is the calculation of Aldrich, Pethick and Pines /16/.

This difference, the binding energy E_B of the two rotons, was not consistent with the value of E_B determined independently from a lineshape analysis of the Raman results /18/. In an attempt to improve upon the accuracy of the neutron value of Δ, and hence give a more reliable value of E_B, an accurate determination of the roton energy was made by Woods et al. /19/. The measured dispersion curve for the roton region is presented in figure 5 where the data points are compared with the "best-fit" parabola of the form

$$\hbar\omega = \Delta + \frac{\hbar^2}{2m^*}\left[Q - Q_R\right]^2$$

where m^* is the roton effective mass. From this fit a value of Δ/k of 8.618 ± 0.009 K was obtained. So $2\Delta/k$ is 17.24 K which, when compared with the Raman value of 16.97, yields a binding energy E_B of 0.27 ± 0.04 K. This value compares favourably with that obtained from their lineshape analysis by Murray et al. /18/ and provides solid support for the belief in the existence of a two-roton bound state.

While it seems certain that pairs of rotons with almost zero total momentum interact attractively, it is not clear that the interaction has the same sign at larger values of the total pair momentum. Graf et al. /20/ have studied the neutron scattering at the "maxon" position - $Q = 1.1$ Å^{-1} - as a funciton of pressure.

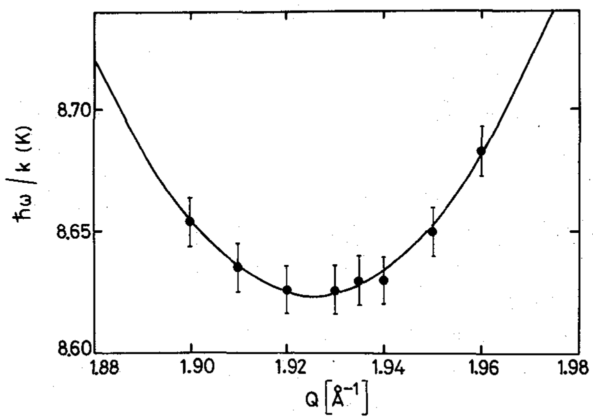

Fig. 5 : The measured dispersion curve near the roton minimum /19/. The curve is a parabola, fitted to the data points.

The maxon frequency increases with pressure and exceeds 2Δ at above about 20 atm., while Δ itself decreases with increasing pressure. A more recent experiment /21/ has been concerned with the region about 3 Å^{-1} where the one-phonon excitations merge into the "upper branch" of multiphonon excitations. Once again at the higher pressures, the one-phonon peaks were found to have frequencies above 2Δ, for Q values above about 2.6 Å^{-1}. Figure 6 shows the observed dispersion relation at a pressure of 24.3 atm. These results were interpreted using the theory of Zawadowski et al. /22/ in which the basic approximation is that the observed spectrum is dominated by the decay of the excitations into pairs of rotons. There are then two principal parameters of the theory, g_3 which describes the interaction between 2 rotons and a single excitation, and g_4 describing the direct roton-roton interaction. These parameters were obtained by fitting the theoretical expressions to the observed intensity distributions.

As expected from the result that the one-phonon peaks have frequencies above 2Δ, the roton-roton interaction was found to be repulsive. A similar analysis of the results of Graf et al. /20/ for $Q = 1.1$ Å^{-1} leads to the same conclusion.

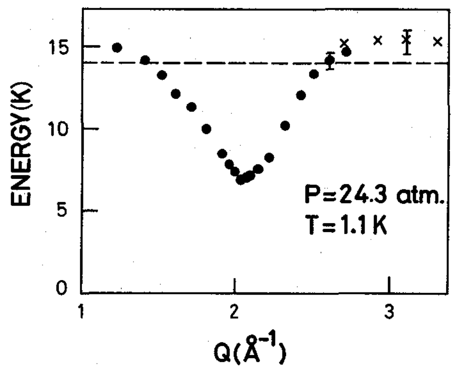

Fig. 6 : The dispersion relation of superfluid ^{4}He at 24.3 atm. as measured by Smith et al. /21/.

However, there remains some controversy ower this result. Tüttö and Zawadowski /23/ argue that the relevant quantity is not g_4 but rather and effective g_4 which combines both g_4 and g_3. It is this effective roton-roton coupling which should be used to describe the neutron lineshapes. These authors argue that even if the experiments of Smith et al. /21/ yield positive values for g_4, and hence a repulsive interaction, the effective coupling may be negative leading to an attractive roton-roton interaction.

Further measurements at wavevectors near the "end" of the one-phonon spectrum may help to clarify this point. The principal problem lies in the difficulty of handling with sufficient accuracy the multi-phonon distribution which is very broad and which persists to high energies. To make a separate estimate of the parameters g_3 and g_4 requires a clear separation between the one-phonon and multiphonon scattering, a task which the present author considers almost impossible.

3. NORMAL LIQUID ^{3}He.-At first sight, the excitation spectrum of liquid ^{3}He might be expected to be very similar to that of ^{4}He. This not so as ^{3}He is a system of interacting Fermions, whereas ^{4}He is a Bose system and so the quantum statistics are very different. In a Fermi liquid there is the possibility of observing both "sharp" collective excitations and a broad continuum of quasi-particle - quasi-hole excitations which arise from the excitation of a ^{3}He atom from an energy close to the Fermi surface to a higher energy state.

Whereas the scattering from ^{4}He is wholly coherent, ^{3}He has a spin-1/2 nucleus and so spin-dependent incoherent scattering must also be considered. This scattering is by no means negligible as the incoherent cross-section is about 25 % of the coherent cross-section /24, 25/. The reason that neutron studies of ^{3}He are not as developed as those of ^{4}He is the extremely large thermal neutron absorption of the lighter isotope-about 10000 b a wavelength of 4 Å. Nevertheless, two groups have now made extensive measurements on liquid ^{3}He.

The first neutron results were reported by Scherm et al. in 1974 /26/. Using a conventional "multichopper" time of flight spectrometer, figure 2, they observed broad distributions of scattering at wavevectors between 1.3 Å^{-1} and 2.2 Å^{-1} for a sample temperature of 1.3 K. These measurements were later improved upon in a series of experiments at 0.63 K for wavevectors between 1.0 and 2.5 Å^{-1} /27/.

The observed scattering was assumed to arise from excitations within the particle-hole continuum but no sharp collective excitations were observed. The maxima of the continuum peaks exhibited a roton like dip near 1.6 Å^{-1}. These results were compared with lineshapes calculated using variants of the random phase approximation. The theory gives reasonable agreement with experiment but with rather extensive modifications.

In parallel with this work, measurements on ^{3}He at 0.015 K were made at Argonne National Laboratory by Sköld et al. /25,28/ using the statistical or correlation time-of-flight technique /29/. In these measurements, peaks were observed at energies of about 12.8 K for wavevectors in the range 0.8 Å^{-1} to 1.3 Å^{-1}. These peaks have been identified with scattering from the zero sound mode, observed in the sound attenuation measurements of Abel et al. /30/. As well as the zero sound peak, scattering from the particle-hole continuum was present and a low frequency peak, identified with the spin fluctuation excitations. Specimens of the data of this experiment and of that of reference /27/ are shown in figure 7. For wavevectors above 1.4 Å^{-1}, the two sets of data are in good agreement, suggesting that the continuum scattering is largely temperature independent. Indeed, the polarization potential calculations of Aldrich et al. /31/ used the results of Scherm et al. /26/ to fit some parameters of the theory and obtained satisfactory agreement between the theoretical and experimental /25/ dispersion relation for zero sound. Figure 8 taken from the paper of Aldrich et al. /31/, presents a comparison between this theory and the experiments of references /25/ and /27/.

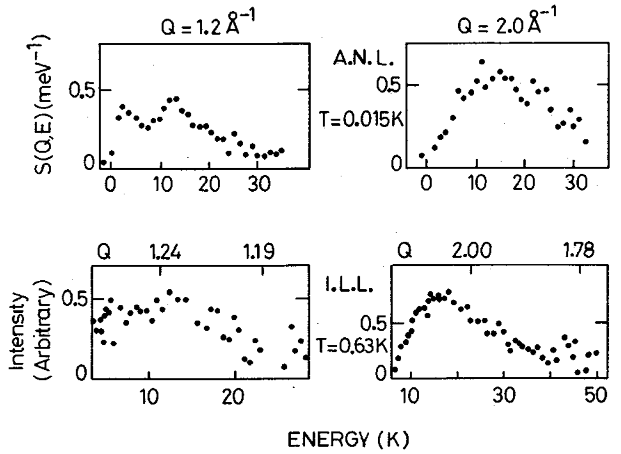

Fig. 7 : Comparison of ^{3}He data of references /25/ and /27/. The A.N.L. data is interpolated to give the energy distribution at constant wavevector while the I.L.L. data is presented in the constant scattering angle representation and so both energy and wavevector vary. At the smaller wavevector (Q $\sim$ 1.2 Å^{-1}) the upper figure shows clearly the existence of a low energy spin fluctuation peak and a higher energy zero sound peak while this structure is lost in the statistical scatter of the points of the lower figure. For Q $\sim$ 2 Å^{-1}, the two sets of data are in good agreement.

A more recent set of experiments at A.N.L. /28/ indicates that the zero sound mode bends over towards the sound velocity line, at small wavevectors.

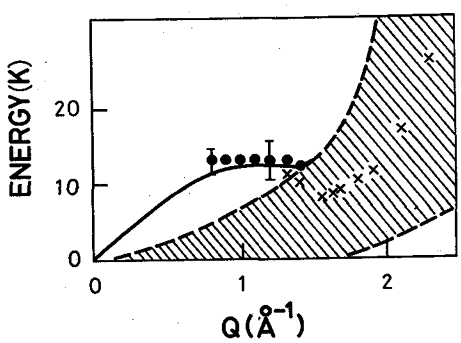

Fig. 8 : The ^{3}He zero sound peak positions of Sköld et al. /25/ at 0.015 K, closed circles, and the maxima of the spectra observed by Stirling et al. /27/ at 0.63 K, crosses. The full-line represents the zero sound calculation of Aldrich et al. /31/ and the hatched area is the band of particle-hole excitations.

Since there is some overlap in wavevector between the I.L.L. and A.N.L. measurements, it has been suggested that the zero sound mode is damped with increasing temperature and so might not be separable from the continuum scattering at the much higher temperature of the I.L.L. measurements. Using the theory of Akhiezer et al. /32/ which is based on Landau theory and so is strictly valid only for small Q and ω, Lovesey and Copley /6/ show that there is a "dramatic change" in S(Q,ω) on changing the temperature from 0.016 K to 0.65 K. The temperature dependence of the zero sound mode has also been considered by Glyde and Khanna /33/, within the framework of the random phase approximation. These authors also find a loss of structure of S(Q,ω) at higher temperatures. On the other hand, Aldrich and Pines /34/ consider the way in which their low temperature s.v.p. calculations will be changed by increasing the temperature and pressure. They conclude that neither the frequency nor the width of the zero sound mode at about Å^{-1} are much altered by increasing the temperature to as much as 1.2 K. Sköld and Pelizzari have now made measurements at both 0.04 K and 1.2 K /35/ and find that the zero sound mode is still observable at the higher temperature for wavevectors between about 0.36 Å^{-1} and 1.24 Å^{-1}. It appears that for these extremely difficult neutron measurements on ^{3}He, the correlation technique provides significantly improved sensitivity as compared with the conventional time-of-flight method. For a detailed comparison of the experimental techniques used in these measurements on liquid ^{3}He, the reader is referred to the original papers and to the review of Lovesey and Copley /6/.

There remains much experimental work to be done. In particular, the pressure dependence of the excitations remains to be studied in detail. Hilton et al. /36/ have measured the inelastic scattering at the saturated vapour pressure and at 10 and 20 bars, for a temperature of 0.7 K. Figure 9 gives the peak and half height positions of the energy distributions observed by Hilton et al. /36/. No well-defined zero sound peak was observed, even though measurements were made in the region of (Q,ω) space in which such a peak was expected. However, there are marked differences in the spectra at s.v.p. and at elevated pressure. As the pressure is increased, the maxima in the energy distributions shift to smaller energies. This effect is particularly marked at the "roton-minimum" wavevector, Q $\sim$ 1.9 Å^{-1}, where the maximum decreases from about 10 K at s.v.p. to less than 4 K at 20 bars. This is a much greater change than in liquid

^{4}He as the same pressure increase decreases the roton energy from 8.6 K to 7.3 K. For further details of these results, we refer the reader to the paper of Hilton et al. in these proceedings.

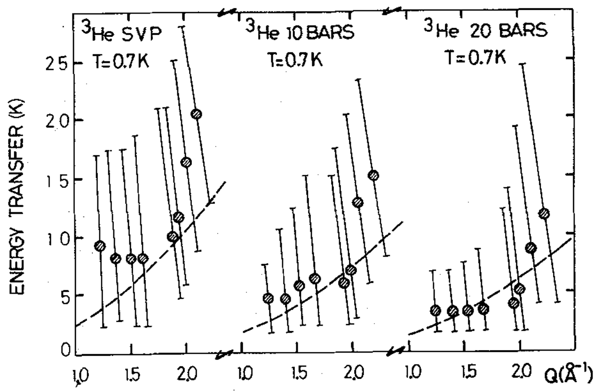

Fig. 9 : The peak positions and half-height positions of the energy distributions observed by Hilton et al. /36/ for ^{3}He at 0.7 K and at s.v.p., 10 bars and 20 bars pressure. The dotted lines show the free-particle dispersion relation $\hbar\omega = \hbar^2Q^2/2M^*$ for effective masses M^* of 3.08 m_3 at s.v.p., 4.4 m_3 at 10 bars and 5.2 m_3 at 20 bars.

It is certain that further experiments on liquid ^{3}He will be performed in the near future. The A.N.L. group intends studying the pressure dependence of the excitations while the I.L.L. group will re-examine the excitations at s.v.p. at much lower temperatures. These experiments have stimulated a great deal of theoretical effort which will doubtless continue as more experimental information becomes available.

4. MIXTURES OF LIQUID ^{3}He and ^{4}He.- A simple model for the neutron scattering from a dilute ^{3}He - ^{4}He mixture might be considered as being merely the weighted superposition of scattering from ^{3}He and ^{4}He. We would then expect to see the phonon-roton excitations of ^{4}He, perhaps modified slightly by the addition of the ^{3}He "impurity", as well as scattering from the ^{3}He continuum of particle-hole excitations.

Rowe et al. /37/ reported the first neutron measurements on a mixture of 5 % of ^{3}He in ^{4}He. Although more difficult to observe due to the ^{3}He absorption, the ^{4}He excitations were only slightly different from those of pure ^{4}He. Small energy shifts of up to 0.5 K, and increases in width of about 1 K were detected, but there were no large changes in energy as had previously been suggested [e.g. /38/]. Similar effects on the phonon-roton excitations were observed in the measurements of Hilton et al. /39/ on a 6 % micture, but in addition peaks at lower energy were seen, which were identified with the particle-hole continuum. The excitation spectrum of this mixture is shown in figure 10. It is clear that the continuum excitations deviate from a simple quadratic dispersion relation, but whether or not there is a roton-like dip as has been suggested /40/ is impossible to say.

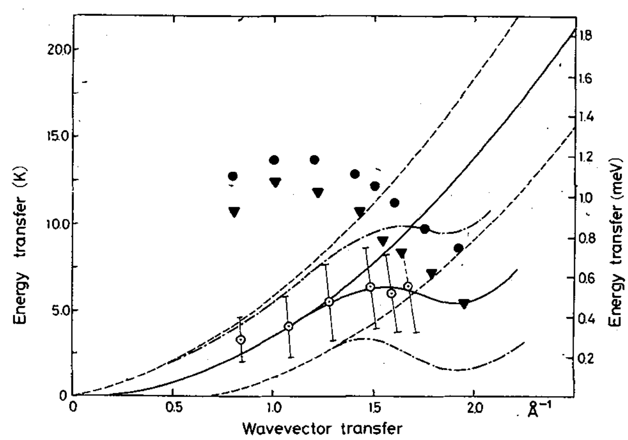

Fig. 10 : The excitation spectrum of a mixture of 6 % ^{3}He at 0.6 K /39/. The open circles are the positions of the particle-hole continuum peaks while the "error bars" represent the full-widths at half maximum of these peaks. The solid circles show the phonon-roton positions and the triangles give the lower limit of effects due to these peaks. Two possible forms for the continuum are represented by the dashed (quadratic) and dot-dash ("quadratic plus roton-like") lines

As one might expect, increasing the ^{3}He concentration increases the roton line-width, but for concentrations of up to 25 %, the roton energy shifts are found to be negligible. At wavevectors smaller than that of the roton, both shifts and line widths increase with increasing temperature. Without doubt, more detailed measurements of the excitations in mixtures of ^{3}He in ^{4}He will soon be performed.

5. CONCLUSION.- Of necessity, much has been omitted from this short "sketch" of current neutron scattering research on liquid helium. Considerations of space have dictated that no mention was made of work on normal ^{4}He or of the continuing effort to prove -or disprove- the existence of a Bose condensate. Although over twenty years of continuous work lie behind us there remains much that needs to be done.

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