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^{4}He FILMS ON GRAPHITE STUDIED BY NEUTRON SCATTERING

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INTRODUCTION

A ^{4}He film shows particular properties. For example the superfluid onset temperature is shifted to lower temperatures with decreasing film thickness and a reduced superfluid fraction is detected [1-3]. This may be caused by a different excitation spectrum with respect to the bulk [e.g. 3]. In addition a ^{4}He film has two interfaces the gas-liquid boundary and the liquid-solid boundary. At the gas-liquid boundary quantized capillary waves (ripplons) [4-7] can be excited. Concerning the liquid-solid boundary it was measured that if the film is deposited on a graphite substrate, that the two layers adjacent to the graphite are solid [8]. The interesting interface is thus the liquid-solid ^{4}He interface. Here the freezing-melting wave could be excited [9]. In addition this interface could play a role in the anomalous small Kapitza-resistance. In this report we will give some new information about excitations in a ^{4}He film and at its boundaries.

EXPERIMENT

In contrast to previous neutron scattering experiments of ^{4}He films [10,11], which where carried out using graphite powder as substrate, we used this time Papyex [12] as substrate. Papyex has besides a powder contribution oriented graphite crystallites which show a 30º (FWHM) mosaic distribution with respect to the c-axis. A densest first layer (which under consideration of the pressure of the second layer has a density of 0.115 atoms/Å^2 [8]) consisted of 312 cc (STP) ^{4}He. The subsequent layers are taken to have densities of 0.95 atoms/Å^2 [8] , 0.078 atoms/Å^2, 0.078.atoms/Å^2...

The inelastic neutron scattering experiments were performed on the time-of-flight spectrometer IN6 [13] at the ILL with a chosen wavelength of 5.12Å. The detectors are located in an angular range of 11.9º - 113º ; this corresponds, for elastically scattered neutrons, to momentum transfers (Q) between 0.254 - 2.046Å^{-1}. The energy resolution is only slightly dependent on Q and is about 0.055 μeV for elastically scattered neutrons. The sample was mounted with the c-axis perpendicular to the scattering plane. The data obtained from the sample cell before any ^{4}He was adsorbed were used as background and subtracted from subsequent measurements with adsorbed ^{4}He.

EXCITATIONS AT THE LIQUID-SOLID BOUNDARY

In previous experiments it was shown that excitations can be measured at the liquid-solid ^{4}He interface, which have a constant dispersion ("flat modes") indicating a localized

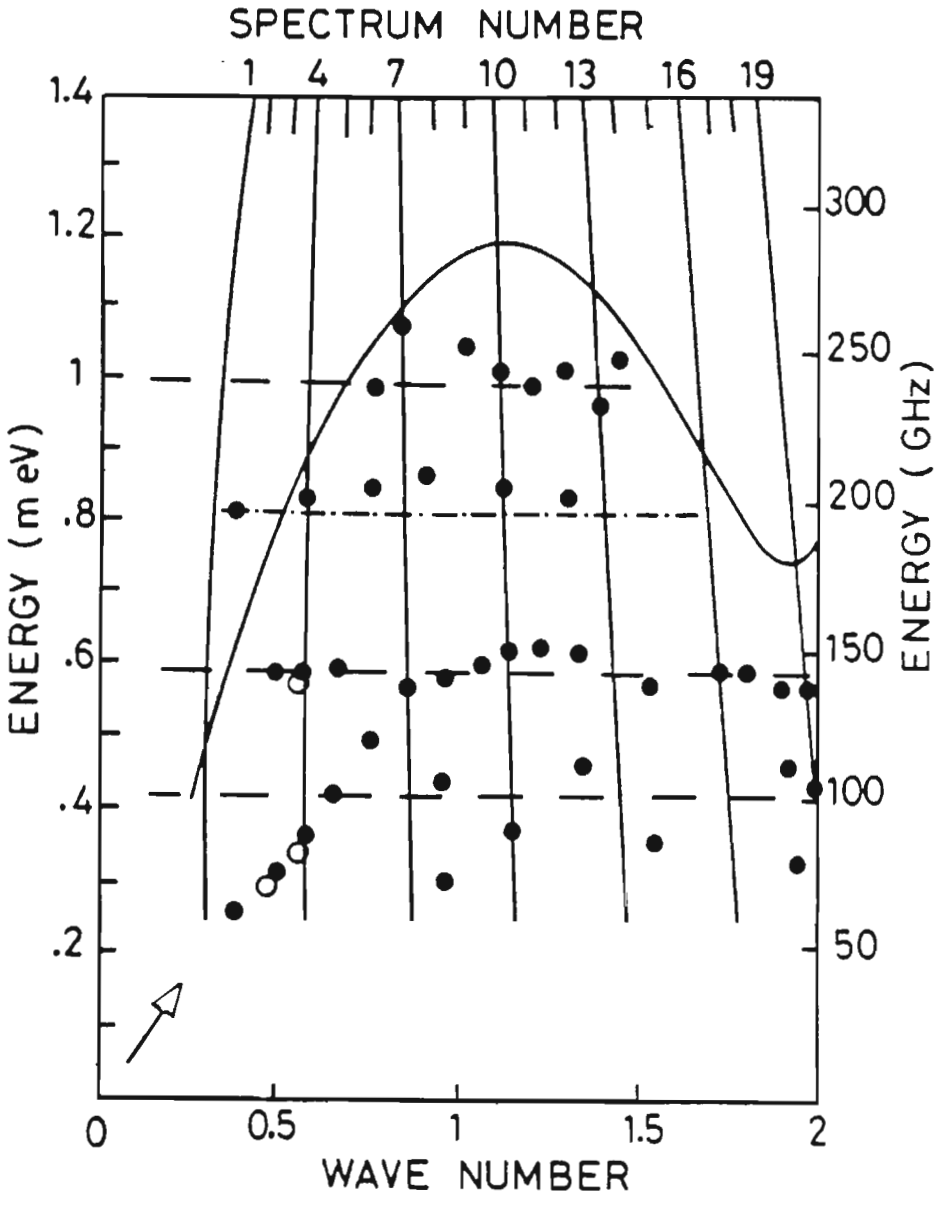

Fig.1: Position of the signals in the energy-wavenumber plane from 6.4 ^{4}He layers on top of the Ne coated graphite powder at T=0.5 K [10]. The spectra have been measured on a time-of-flight spectrometer, therefore the spectrum number indicates the path along which the spectra are taken with constant scattering angle. The solid line represents the bulk ^{4}He signals. The dashed-dotted line shows the peak position of the multiple scattering (diffraction of neutrons by the (002) graphite reflection and creation of a roton). Accidently, this signal is superimposed on a localized mode which are otherwise marked by dashed lines. The arrow points to the first indication of the ripplon, which has been measured in 4.8 ^{4}He layers on Papyex (o) [10].This mode does not depend on the amount of adsorbed ^{4}He beyond a coverage of 4 layers [11]. Now we changed from the powder substrate to Papyex and the flat mode lost about 30% in intensity with respect to the roton intensity. Thus a more homogeneous substrate and/or the preferential orientation may contribute to the decrease of the signal.

excitation [10,11]. This is shown in figure 1, where for a 6.4 ^{4}He layer film on Ne coated graphite powder in addition to the bulk excitations these flat modes show up. Also in the case of Ne coated graphite a solid ^{4}He layer is next to the Ne layers. The proof that these flat modes are localized at the liquid-solid ^{4}He boundary was given by the fact that this mode still persists in a completely with ^{4}He filled sample cell and that the intensity of

this mode does not depend on the amount of adsorbed ^{4}He beyond a total coverage of 4 layers [11]. Now we changed from the powder substrate to Papyex and the flat modes lost about 30% in intensity with respect to the roton intensity. Thus a more homogeneous substrate and/or the preferential orientation may contribute to the decrease of this signal.

The main origin of the flat modes was suggested to be an exchange of atoms between the second solid ^{4}He layer (pure graphite substrate) and the liquid ^{4}He. In order to test this idea we preplated the graphite with two layers of H_2. The ^{4}He isotherm taken on the preplated graphite [14] shows that the density of the first ^{4}He layer of 0.072 atoms/Å^2 is not high enough to be a solid at least at the temperature of T=1.96 K. From Ref.15 it can be deduced that about a density of 0.08 atoms/Å^2 is needed to solidify an incommensurate monolayer of ^{4}He.

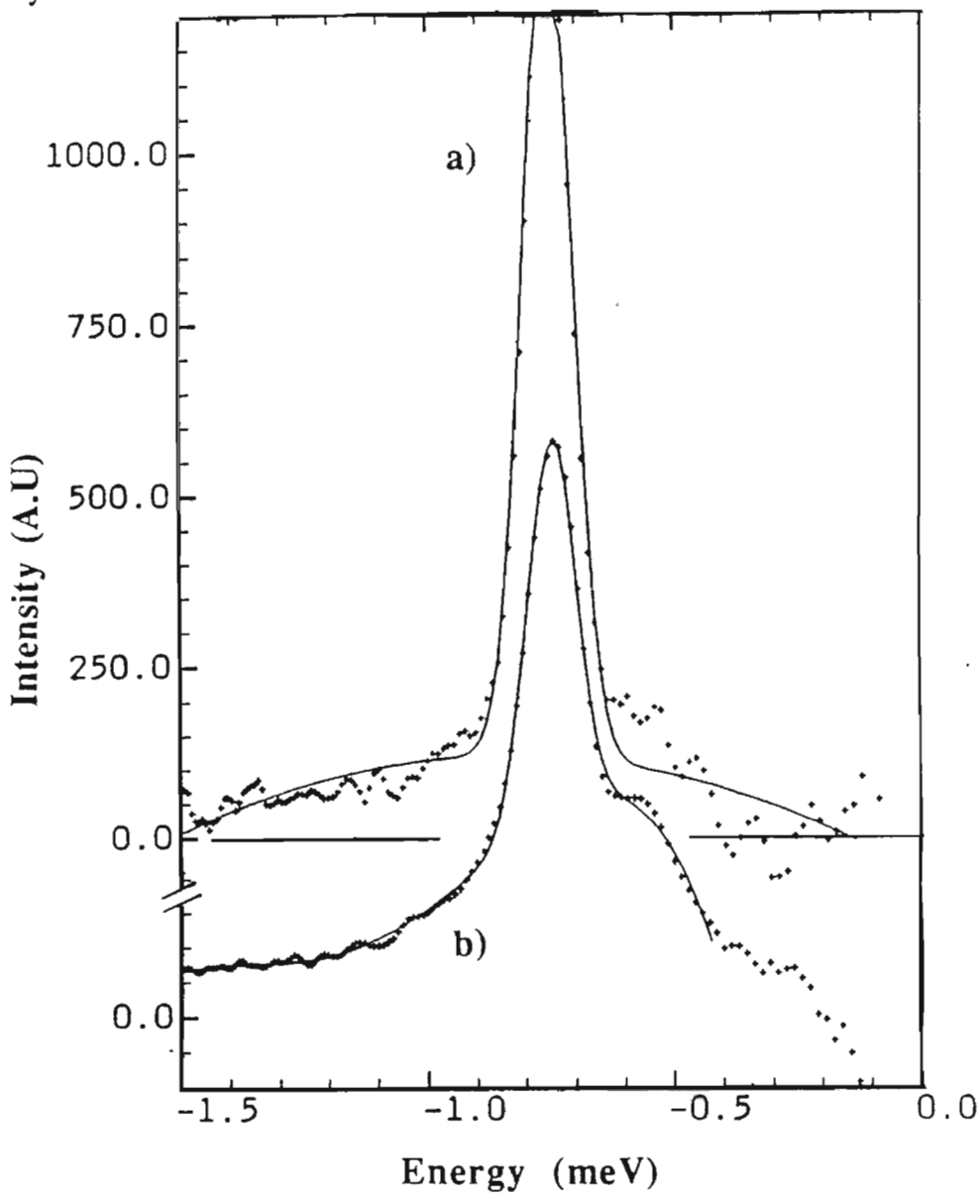

Fig.2: Inelastic neutron scattering scans along the path of spectrum number 18 in fig.1 with an elastic Q = 2.00 Å^{-1}. In a) (sample 1) the graphite was preplated by two layers of H_2 and subsequently 4.6 layers of ^{4}He are adsorbed. In b) (sample 2) a pure graphite substrate was used with 5.06 adsorbed layers of ^{4}He in total. In both cases the roton shows up at an energy transfer of 0.755 meV. The flat mode intensity is at about 0.6 meV. The fit is a very rough one and focuses mainly on the roton. The temperature is 0.7 K. The ordinate is displaced by 250 units between 2a) and 2b). The energy scale is negative due to a convention on the IN6 spectrometer for the energy transfer. The absolute value is the energy transfered to the excitation.

The comparison of a spectrum taken with and without preplating is shown in fig.2. The spectra have been taken at a constant angle, which corresponds to a Q of 2.00 $Å^{-1}$ on the elastic line. From figure 1 it is seen that this corresponds roughly to spectrum number 18 and this scan crosses the bulk dispersion at a Q of 1.92 $Å^{-1}$ and an energy of 0.755 meV. In both cases the roton excitation can be clearly seen. At a lower energy of 0.6 meV the intensity of a flat mode shows up. It is evident that this intensity is decreased by a factor of two in the case of the preplated sample with respect to the intensity of the not preplated sample. This already shows that the suppression of the solid ^{4}He layers decreases the intensity of the flat modes by an appreciable amount, so that the conclusion that the solid layers are involved in the local modes is correct. Remember that the intensity of the flat modes does not depend on the ^{4}He layer thickness in the range of thickness displayed in fig.2 [11]. These solid layers are present in most examples of Kapitza-resistance measurements and should be taken into consideration, which means that the excitation of a localized mode can render possible a transmission of a phonon from a solid through an interface to liquid ^{4}He.

Another comparison has to compare the intensity of the bulk roton signal of the film on the different substrates. The intensity increases in the case of the preplated sample (called sample 1 with a filling of 4.6 layers) by a factor of 2.2 (same linewidth) with respect to the not preplated sample (sample 2 with a filling of 5.06 layers including the solid ones). This is mainly due to the replacement of the solid ^{4}He layers by liquid layers. A more careful consideration gives the following result. The filling was 5.06 layers of ^{4}He for sample 2. It turns out (see paragraph: Phonons and Rotons in a ^{4}He-film) that 3.5 layers do not contribute to the intensity of phonons and rotons in the case of sample 2, thus a layerthickness of 1.56 does only contribute to the signal intensity. An intensity gain of 2.2 was measured in switching to sample 1 (see fig.2, this factor is also confirmed at other Q's). This gives 3.43 layers which do contribute to the signal intensity for sample 1. The filling of sample 1 was 4.6 layers. Thus 1.2 layers do not contribute to the intensity of phonon and roton excitations. This is about the same number as on the pure graphite substrate, where 3.5 layers are inert, but two of them are solid so 1.5 inert liquid layers are left. The information which can be taken from the phonon and roton signal intensity for the two different substrates is twofold. It firstly confirms that no solid ^{4}He layers are present in the case of the preplated sample and secondly it demonstrates a liquid ^{4}He layer (1.2 to 1.5 layers) which does not contribute to the intensity of the phonons and rotons.in a ^{4}He film on different substrates.

PHONONS AND ROTONS IN A ^{4}He-FILM

A long outstanding question is how the dispersion curve of bulk liquid ^{4}He is changed if the ^{4}He is confined in a film. A very simple approach was given in Ref.3, where a change in the static structure factor was calculated in going from 3-dimensional ^{4}He to 2-dimensional ^{4}He. This shift results in a lowering of the roton minimum and a shift towards lower energies. Our scans hardly approached the roton minimum in Q and on the other hand this theory was not made for the phonons and maxons. Thus an other theory was considered [5,16]. Here a clear increase of the energy in the maxon region was predicted due to the higher density regions of the liquid layers near the solid. However our fits to the experimental data, exhibited in fig.3, show that a lowering of the dispersion curve takes place in the maxon region if the film thickness is reduced. The lowering of the dispersion curve is continued towards the roton minimum. However the roton minimum was not covered by the scans. The phonon branch is not influenced. But phonons were only visible for our thickest film. Our data from the completely with ^{4}He filled cell agree with the bulk Dispersion curve [17]. But we have no explanation for the softening of the maxon and the roton branch, where the rotons have with negative group velocity .

The intensity of the phonons and rotons is plotted in fig.4. A calibration to the bulk data was done for all film thicknesses at a Q of 1.92 Å-1. The data taken with the

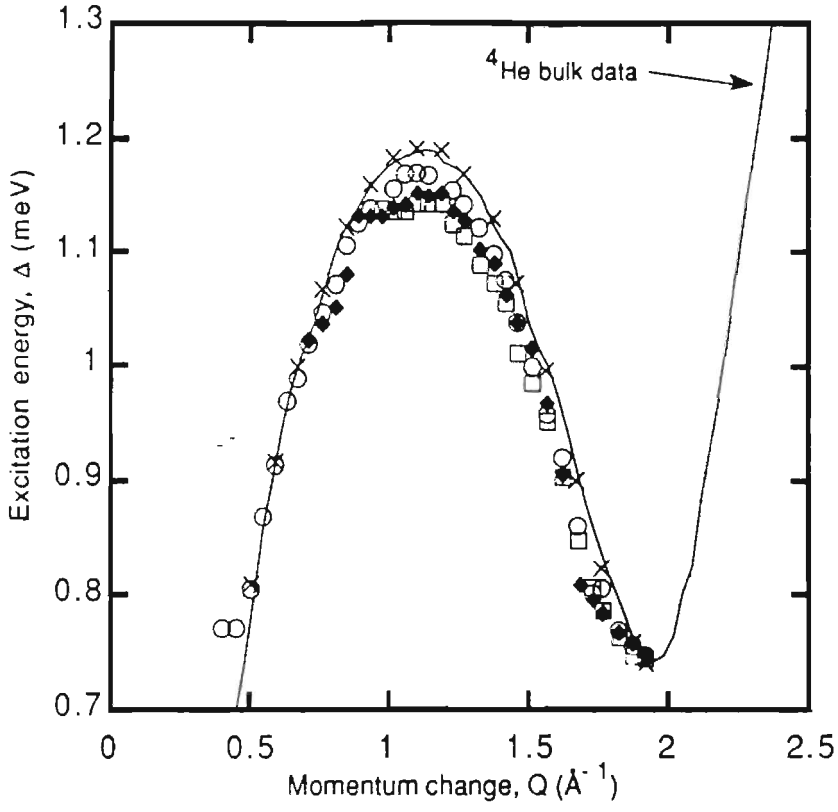

Fig.3: Variation of the dispersion relation of the phonons and rotons as a function of film thickness at a temperature of 0.7 K. The total ^{4}He layer thickness is 5.06 layers (○), 4.16 layers (◆) and 3.85 layers (❑). (×) marks data from the completely with he filled sample cell.

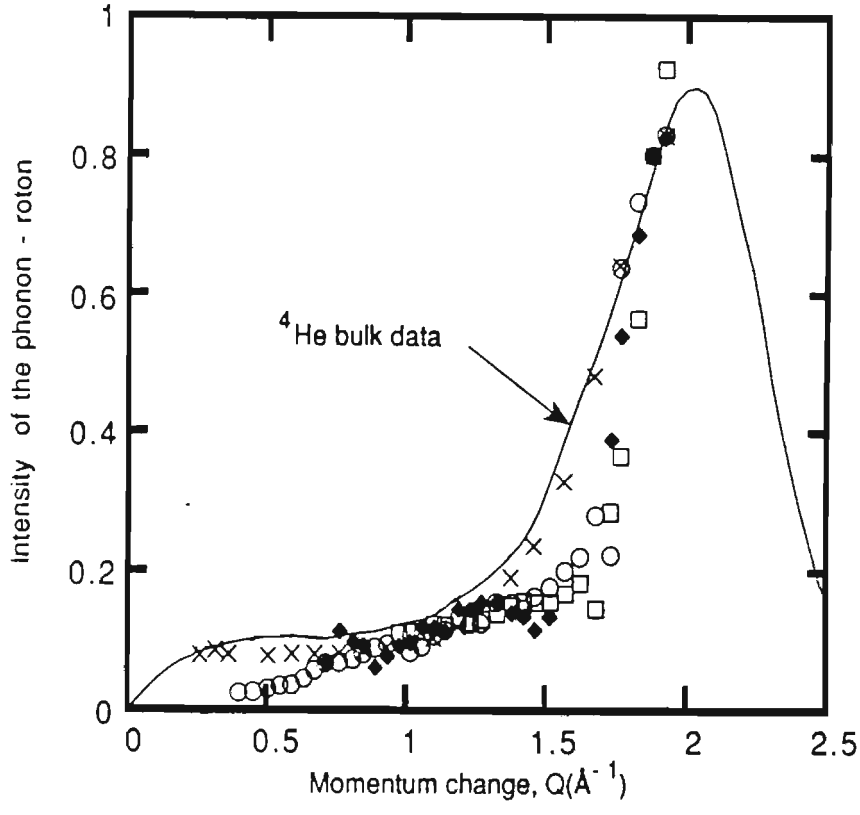

Fig.4: Intensity of the phonons and rotons as a function of Q at a temperature of 0.7K.The total ^{4}He layer thickness is 5.06 layers (○), 4.16 layers (◆) and 3.85 layers (❑). (×) marks data from the completely with he filled sample cell. The intensities of the different coverages are calibrated to 0.8 , the value of the bulk data at 1.92 Å^{-1}

completely ^{4}He filled cell show towards lower Q a small deviation to lower intensities with respect to the bulk data. We do not know yet whether this is significant. Important deviations appear however for the ^{4}He films. The thick film (5.06 layers) looses much intensity below a Q of 0.7 Å^{-1}. This Q corresponds just to the liquid filmthickness. Thus the loss of the third dimension reduces the intensity of the phonon. The thinner films show at low Q no more intensity. Thus a loss in intensity is detected if the filmthickness correlates with the momentum transfer of the excitation and the intensity disappears even at still lower Q.

Another remarkable feature in figure 4 is the loss of intensity with respect to the bulk data around 1.7 Å^{-1}. This is again the region, where rotons have negative group velocity and where a shift in the dispersion relation was measured. We can only suggest that this effect may again result from the reduced dimensionality.

The behavior of the intensity of the phonon is plotted in fig.5 against the filmthickness for a Q of 1.2 Å^{-1}. The intensity extrapolates to zero intensity at a total coverage of 3.5 layers. This coincides with measurements in Ref.18, where a loss of superfluidity is detected at about 3 adsorbed layers.

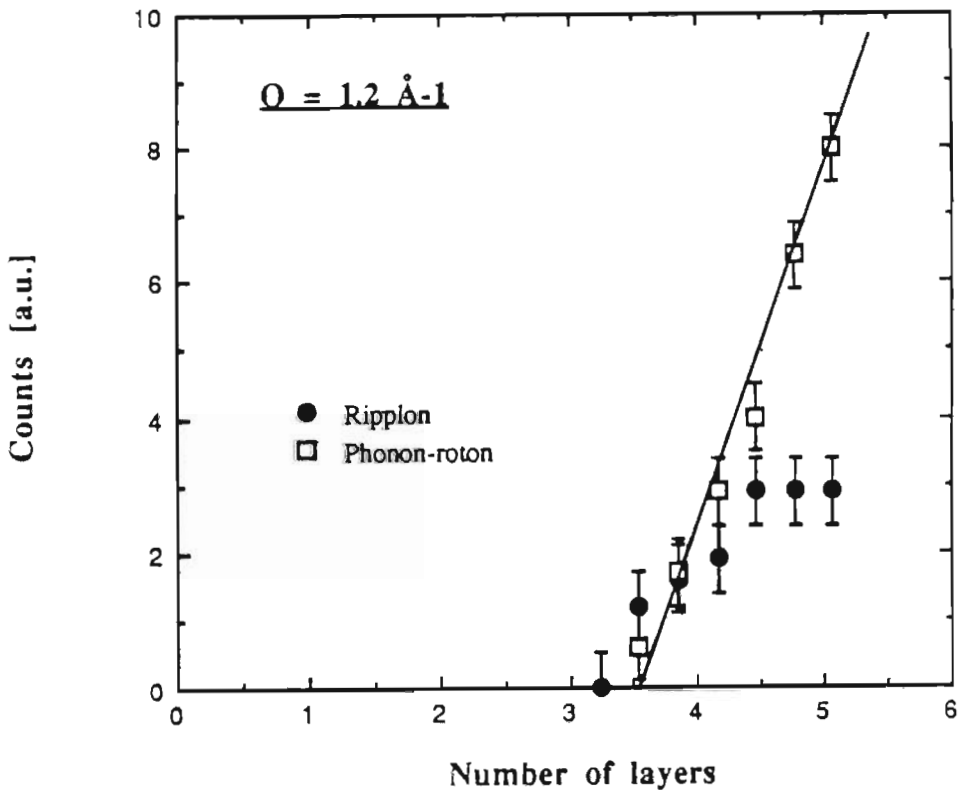

Fig.5: Intensities of the phonon (□) and ripplon (○) as a function of coverage at T=0.7K.

RIPPLON

Quantized capillary waves (ripplons) are the elementary excitations of a free liquid surface. Their existence at the bulk ^{4}He surface and in films has been predicted by theory and indirectly confirmed by experiment [4,19]. At long wavelengths the ripplon dispersion relation is easily evaluated using hydrodynamic relations for an incompressible fluid:

$$\omega^2 = (\alpha_0/\rho_0)\, k^3 \qquad \{1\}$$

where α_0 is the zero temperature surface tension, ρ_0 the ^{4}He density at zero pressure and k the wavevector. The temperature dependence of the surface tension ($\alpha(T)$) at very low temperatures can be deduced from the ripplon dispersion relation. Detailed measurements

[20] of $\alpha(T)$ revealed a much larger temperature dependence than expected from formula {1}. Several modified dispersion curves have been proposed which differ mainly for wavevectors above 0.5Å^{-1}. The idea of a 'surface roton', with a minimum at ~2K, was introduced by Reut and Fisher [21] improving the agreement with the available thermodynamic data. Edwards et. al. [4,20], taking into account the curvature dependence of α, were able to fit the experimental data on the excess surface entropy. Their model involves two parameters: a length $\delta = d(\ln \alpha_0)/dK$ where $K = (r_1^{-1} + r_2^{-1})$ is the curvature of the surface, and an area $a = d\delta/dK$. Within the precision of the entropy data, several sets of parameters have been used ($a = +1.5$Å^2, $\delta = 0$ [20] and $a = +1.0$Å^2, $\delta = -0.336$Å [4]), the latter giving a better agreement. Such a large variation in the parameters corresponds to very different ripplon dispersion curves at wavevectors ~1Å^{-1}, with a common trend indicating the presence of a downward curvature. Little direct experimental evidence was available [10], however, on the ripplon dispersion curve at these wavevectors. Such a study requires a microscopic probe like inelastic neutron scattering (INS), but due to the low neutron cross section of ^{4}He the measurement has to be performed on samples with a large surface to volume ratio. The success of a neutron total reflection experiment is however not yet excluded [22].

A first outline of the experiment of which some results are depicted in fig.6 was already given in Ref.23. In figure 6 the different colors indicate the behavior of the intensity as a function of energy and momentum transfer. It is clearly seen in fig.6a that besides the intensity on the phonon-roton curve there is intensity on an energetically lower lying branch. Evidence of the existence of this branch has been given previously on measurements [10,11]. This branch coincides with the calculated dispersion of the ripplon using the parameter set in Ref.4. The agreement is very good, it seems to be even up to 1.5A-1. At still higher Q the roton intensity combined with the one of the flat modes becomes to high to distinguish the ripplon signal. This good agreement allows to say that the temperature dependence of the surface tension is really based on an experimentally verified dispersion relation. It still remains to prove the modified parameter set [4] by a theory.

In fig.6b the result of the completely filled sample cell is shown. In the region where the ripplon should show up, the colors are the same as in fig.6a. Only near the phonon-roton intensity and the flat bar of the multiple scattering [10,11] the attribution of the colors to intensity has been modified. Thus this figure shows that no signal of the ripplon intensity is visible in fig.6b the filled cell, although below 0.7 Å^{-1} it would have been distinguishable from the overwhelming quasi bulk phonon roton intensity. This disappearance proves that the ripplon signal is really bound to the gas-liquid interface.

A more detailed description of the ripplon dispersion is seen in fig.7. The measured dispersion agrees indeed very well with the calculation in Ref.4 up to $Q = 1$Å^{-1}. But between 0.6 to 1 Å^{-1} a small but detectable increasing slope of the dispersion curve with decreasing film thickness is detectable. The behavior at higher Q lets assume a minimum in the dispersion curve. However this comes about due to crossing the flat mode at 0.6 meV and each negative slope is due to the fit-routine, which clamps with increasing Q to the higher intensity of the flat mode. An interaction between the ripplon and the flat mode in a thin film is not excluded [11] but needs further consideration. We can confirm a positive slope of the dispersion curve in agreement with Ref.4 up to 1.3 Å^{-1} for certain coverages. At higher Q's the ripplon intensity gets lost.

The intensity of the ripplon is plotted in fig.8 as a function of Q for the different coverages. The ripplon is most intense at low Q loosing its intensity rapidly up to 0.6 Å^{-1}. At higher Q's again the interference with the flat modes makes a correct data analysis impossible in particular to the half automatic fit-routine. Perhaps the experiments on a H_2 preplated graphite surface will render the analysis more effective.

From preferential Q's, which are free from the flat mode intensity, ripplon intensities can be taken by careful "by hand" assisted fitting as a function of coverage. For a Q of 1.2 Å^{-1} this has been done and is shown in fig.5. The ripplon intensity seems to saturate

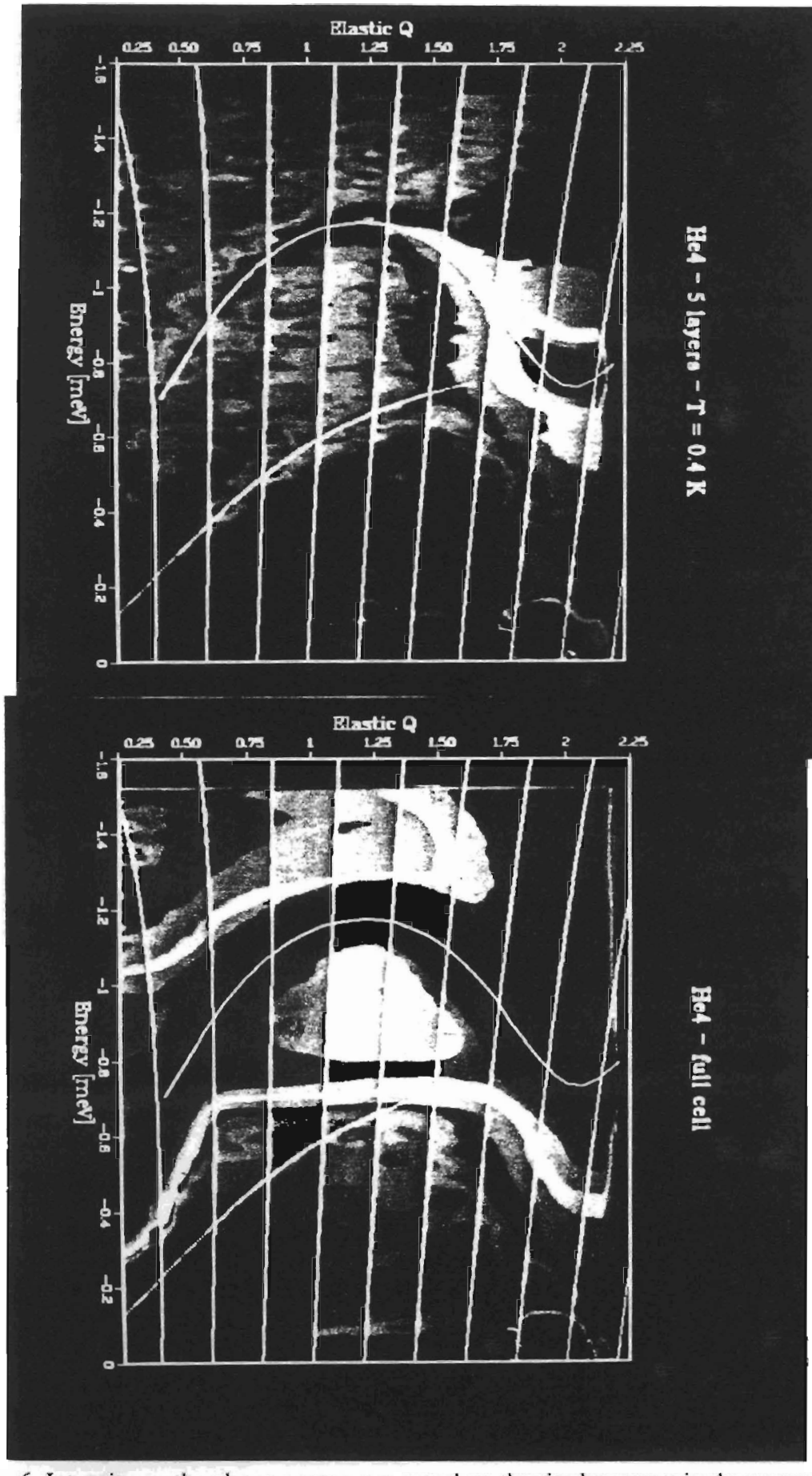

Fig.6: Intensity on the phonon-roton curve and on the ripplon curve in the energy-Q plane. Fig.6a shows the signal from 5.06 adsorbed layers on graphite and fig.6b the signal from the completely filled sample cell (including the graphite).

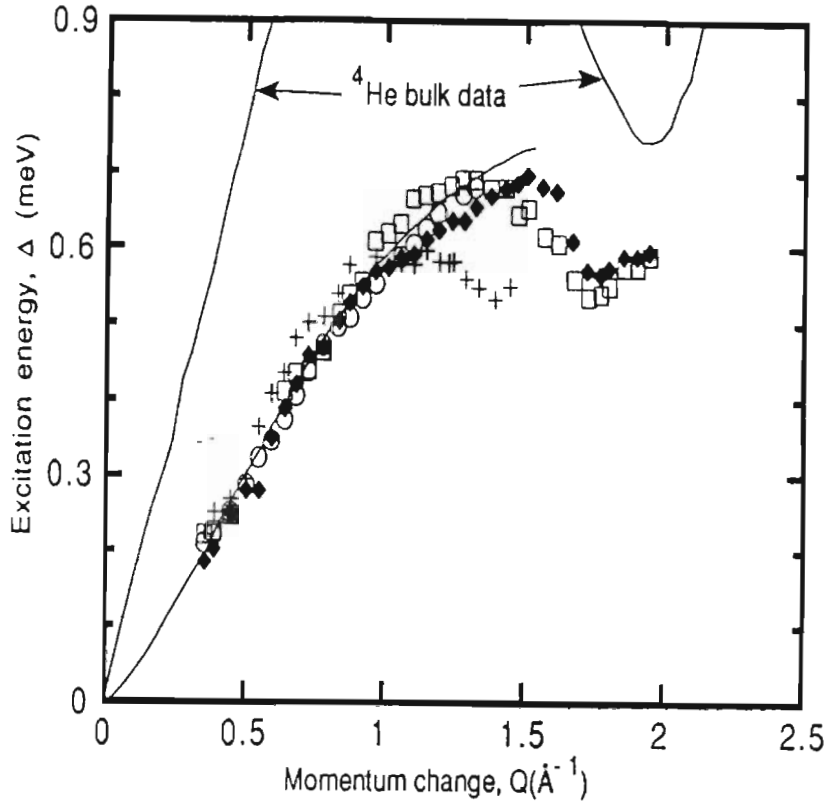

Fig.7: The ripplon dispersion for various coverages.The total He layer thickness is 5.06 layers (○), 4.16 layers (◆), 3.85 layers (❑)and 3.54 layers (+).

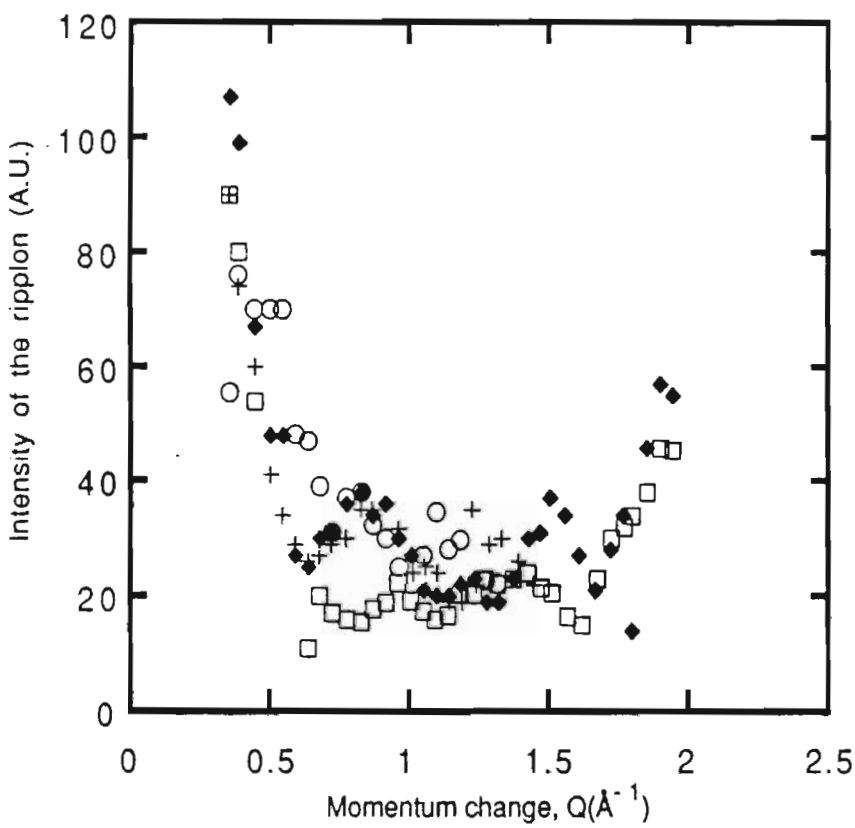

Fig.8: The intensity of the ripplon as a function of Q for various coverages. The total ^{4}He layer thickness is 5.06 layers (○), 4.16 layers (◆), 3.85 layers (❑)and 3.54 layers (+).

beyond a coverage of 5 layers. This is in agreement with the picture of a surface wave, whose intensity does not depend on the amount of bulk liquid below the surface. On the other hand the ripplon intensity disappears near 3 layers. This is in agreement with a penetration depth of about an atomic layer.[4,6] However this intensity disappears definitely at a lower coverage than the phonon-ripplon intensity. This phenomena is not yet understood.

Other modes are predicted for a thin film between the ripplon and the continuum (the phonon-roton dispersion curve) [5,6,16] but up to now no other modes could be detected.

CONCLUSION

In summary it turns out that a liquid helium film exhibits a lot of interesting features in the excitation spectrum. The excitation spectrum of the bulk phonon-roton curve is modified in a helium film as well as the dependence of the intensity on the momentum transfer. There are in addition excitations which have no dispersion (localized modes) at the solid helium-liquid-helium interface. Finally the dispersion curve of the ripplon at the gas-liquid boundary could be measured for the first time to relatively high momentum transfers.

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