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DIRECT OBSERVATION OF RIPPLONS IN ^{4}He FILMS BY NEUTRON SCATTERING

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Quantized capillary waves (ripplons) are the elementary excitations of a free liquid surface. Their existence in bulk ^{4}He and in films has been predicted by theory and indirectly confirmed by experiment[1,2]. 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[3] 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[4] improving the agreement with the available thermodynamic data. Edwards et al.[1,3], 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, $\delta = 0$ [3] and $a = +1.0$Å^2, $\delta = -0.336$Å [1]), the latter giving a somewhat better agreement. Such a large variation in the parameters corresponds to very different ripplon dispersion curves at wavevectors ~1Å^{-1}. Little direct experimental evidence is available[5], 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.

We have measured the inelastic structure factor of ^{4}He adsorbed on the basal plane of graphite. The INS measurements were performed at the time of fligth spectrometer IN6 at the Institut Laue-Langevin's reactor using a wavelength of 5.12Å. The elastic energy resolution depends sligthly on the momentum transfer Q due to sample size effects, increasing from 80 to 110μeV with scattering angle.

The sample consisted of 31.70g of Papyex[6] sheets oriented with their c-axis normal to the scattering plane. The temperature of the sample was kept at 0.65K for all the measurements. An adequate annealing of the adsorbed films was performed after each change in coverage. The total surface area was determined by adsorption isotherms and neutron diffraction to be 730m^2 ± 2%. The ^{4}He monolayer coverage (0.112 atoms/Å^2) was 304cc STP. The data obtained before any ^{4}He was adsorbed were used as background and substracted from subsequent measurements. The result of two measurements will be reported here: a) at a total coverage of 0.448atoms/Å^2, equivalent roughly to 5 atomic layers and b) a scan with the cell filled with bulk superfluid ^{4}He.

The first and second layers are solid at these coverages and their density is well known[7,8] (1st layer=0.115at/Å^2, 2nd layer=0.094at/Å^2). Thus, 0.209at/Å^2 correspond to the solid and the remaining amount of ^{4}He to the liquid layers. We use a mean density of 0.078at/Å^2 for a liquid layer to evaluate the thickness of the film. This value has to be taken with care, since microscopic calculations showed that the density of the bulk liquid-vacuum interface decreases slowly within a distance of ~5Å[9].

The result of the measurement for 5 layers is depicted in figure 1 as a contour plot of $S(Q,\omega)$ in order to give a general overview of all channels and detectors. One can easily recognize two excitation branches. The higher energy one agrees well with the bulk phonon-roton dispersion relation (solid line in fig.1). The lower branch, located at about half the energy of the previous one, is the main object of this paper. Evidence of the existence of this branch has been found previously on measurements[5] done on a different substrate (Vulcan III graphite powder), together with the observation of dispersionless modes. In the present experiment no dispersionless modes were observed for $Q < 1.5\text{Å}^{-1}$; this may be due to the larger coherence length or to the preferential orientation of our Papyex sample. The absence of these flat modes enables us to determine the lower branch dispersion relation unambiguously up to $Q \sim 1.5\text{Å}^{-1}$.

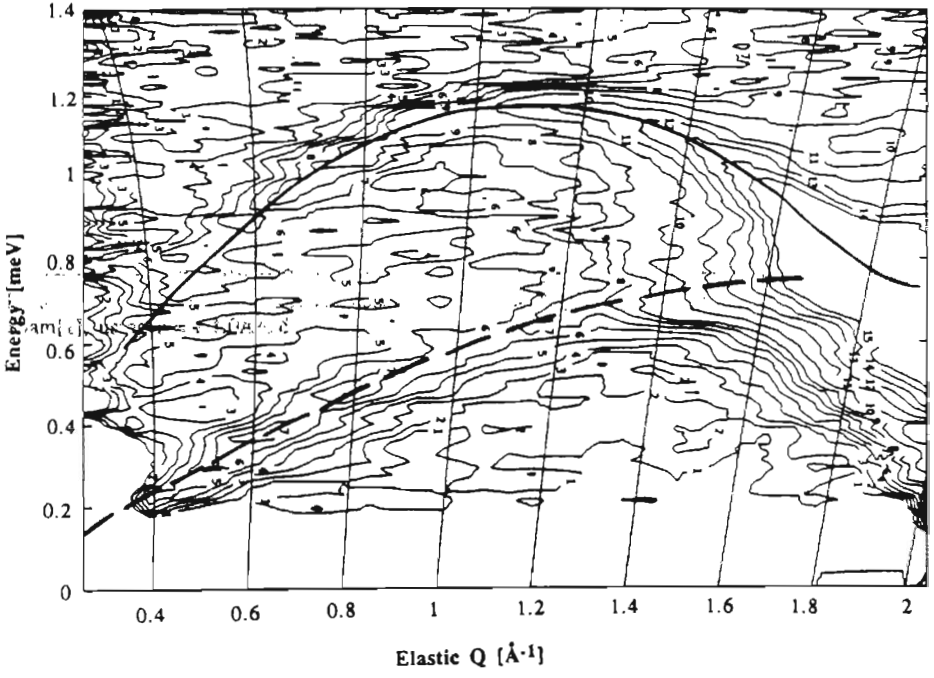

Figure 1: Contour plot of $S(Q,\omega)$ (arbitrary units) for a coverage of 0.448at/Å2. The contour lines 1 to 9 are separated by 10 units, lines 10 to 15 by 30 units. The thick solid line is the bulk ^{4}He phonon-roton dispersion relation. The elastic Q values are given in abscissa, constant Q lines are indicated by thin solid lines in the graph. The dashed line corresponds to the ripplon dispersion curve determined by Edwards and Saam[1], using $a = +\ 1.0\text{Å}^2$, $\delta = -\ 0.336\text{Å}$.

To determine the origin of the lower excitation branch we performed a measurement with the cell filled with bulk liquid. This procedure suppresses the liquid-vapour interface, but does not affect the adsorbed solid-liquid interface. As seen in figure 2, the phonon-roton part is strongly enhanced while the peak at 0.47meV is suppressed. This is observed for all the measured spectra. Therefore, we conclude that the low energy excitation branch belongs to the liquid free surface and is identified as a ripplon.

The experimental ripplon dispersion curve displays a strong downward curvature and seems to merge with the bulk roton minimum for Q~2Å^{-1}. Due to the high intensity of the roton signal it is difficult to follow the ripplon peak at these wavevectors.

Below Q=1.5Å^{-1} our result agrees well with the calculation of Edwards and Saam[1] for a=+1.0Å^2, δ=-0.336Å and does not agree with the dispersion curves given in references 3 and 9.

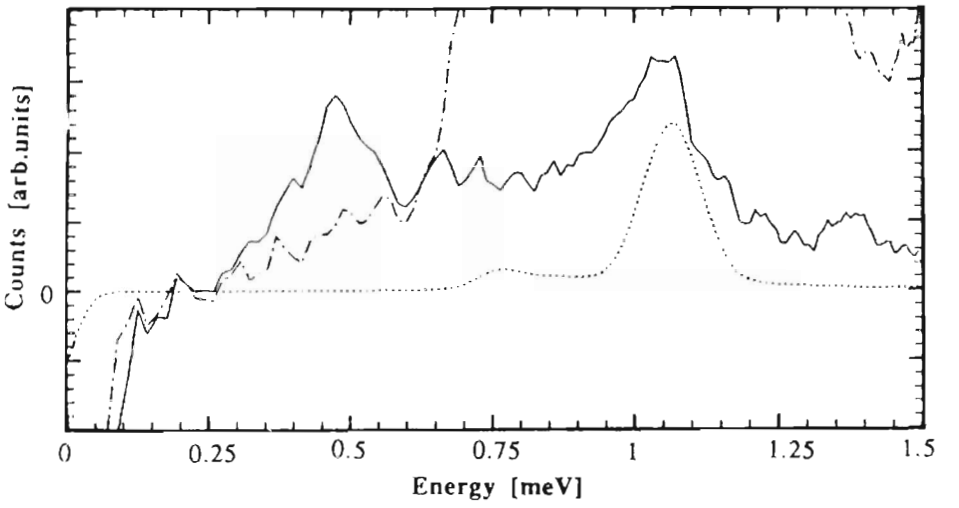

Figure 2: Inelastic spectrum (arbitrary units) for Q=0.8Å^{-1} as a function of energy. The solid line corresponds to the measurement with coverage 0.448at/Å^2 (5 atomic layers). The peak at ~0.47meV corresponds to the ripplon and the one at ~1.05meV to the bulk phonon. Dashed line: same spectrum with the cell filled with bulk liquid; note that the ripplon peak has now disappeared. Dotted line: bulk spectrum divided by 100; the peak at 0.75meV is due to multiple scattering.

In conclusion, we have directly determined the ripplon spectrum at large wavevectors for the first time. With the cell filled with ^{4}He, the surface mode is suppressed, showing clearly that this excitation corresponds to the liquid-gas interface. Measurements of the temperature and coverage dependence and at higher Q are in progress.

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