

FIG. 1. Contour plot of the inelastic structure factor $S(Q, \omega)$ for a coverage of $0.448 \text{ atom}/\text{\AA}^2$ (5.06 layers). Elastic Q values, given on the abscissa, mark the origin of constant- Q lines. The magnitude of $S(Q, \omega)$, indicated in arbitrary units by different shades of grey, increases from white to black; white: ≤ -80 (negative values originate from the subtraction of an intense background signal around the elastic line); greys: -80 to 25 , and then to $30, 35, 40, 50, 60, 120, 250, 400, 600$; black: ≥ 600 . The upper solid line is the bulk ^4He phonon-rotor dispersion relation. The lower line is the ripplon dispersion curve calculated by Edwards and Saam [1], using $a = +1.0 \text{ \AA}^2$, $\delta = -0.336 \text{ \AA}$.

ground and subtracted from subsequent measurements. Ten coverages between 1 and 5 layers were investigated: $0.118, 0.283, 0.294, 0.306, 0.329, 0.354, 0.377, 0.401, 0.424$, and $0.448 \text{ atom}/\text{\AA}^2$. Finally, a scan with the cell filled with bulk superfluid ^4He was performed.

Before presenting results for the inelastic spectra at the various coverages, we briefly discuss the structure of the adsorbed films as it follows from diffraction measurements [8,9]. On graphite substrates as they were used here the first two layers are solid, with densities which are slightly coverage dependent; except for our lowest coverage, their densities can be considered as constant: $0.115 \text{ atom}/\text{\AA}^2$ for the first layer and $0.094 \text{ atom}/\text{\AA}^2$ for the second layer. The remaining amount of helium is liquid. To estimate the total thickness of the film we use for the liquid layers the density of bulk liquid ^4He , obtaining somewhat arbitrarily $0.078 \text{ atom}/\text{\AA}^2$ for the mean density of a liquid layer. This value has to be taken with care, since microscopic calculations show that the density at the bulk-liquid-vacuum interface decreases slowly with a spatial extension of $\sim 5 \text{ \AA}$ [4,5]. Besides, a small increase in the density next to the two solid layers is expected due to the van der Waals interaction with the substrate. Using the values given above, the coverages investigated here are $1.05, 2.96, 3.09, 3.24, 3.54, 3.85, 4.16,$

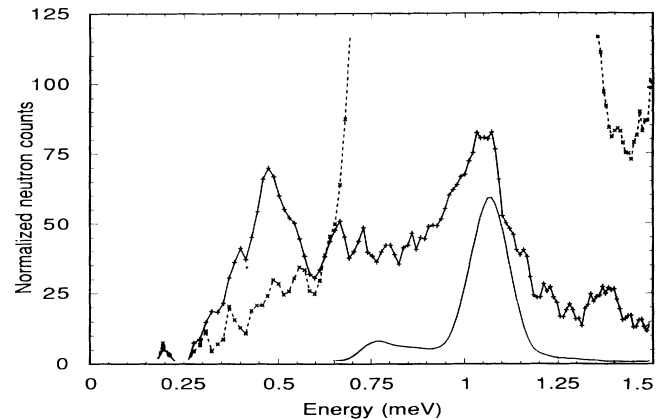


FIG. 2. Inelastic spectrum (arbitrary units) for $Q = 0.8 \text{ \AA}^{-1}$ as a function of energy. The solid line with +'s corresponds to a coverage of 5.06 atomic layers. The ripplon peak is found at 0.47 meV and the bulk phonon at 1.05 meV . Dashed line with x's: spectrum with the cell filled with bulk liquid; note that the ripplon peak has disappeared. The full line shows the same spectrum divided by 100 (the feature at 0.75 meV is due to multiple scattering).

$4.46, 4.76,$ and 5.06 layers.

The results for 5.06 layers, depicted in Fig. 1, give an overview of all channels and detectors. Two excitation branches are clearly seen. The one at higher energies agrees well with the bulk phonon-rotor dispersion relation. 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 in measurements [6] done on a different substrate (Vulcan III graphite powder). However, the simultaneous observation of dispersionless modes made an assignment of the modes difficult. In the present experiment the intensity of the dispersionless modes was very small (except in the high- Q region to be discussed below); this may be due to the larger coherence length or to the preferential orientation of the Papyx sample. This enabled us to determine the lower-branch dispersion relation unambiguously up to wave vectors $\sim 1 \text{ \AA}^{-1}$.

Our interpretation of the new branch to be a mode of the liquid surface is substantiated by the following experiment: Filling the cell completely with superfluid ^4He , thus suppressing the free surface, yields an inelastic spectrum with an intense bulk excitation branch, but with the lower branch having totally disappeared (Fig. 2). Therefore, the latter is clearly associated with the free surface of the film. Furthermore, its dispersion relation at low wave vectors agrees well with thermodynamic data, and in particular with the calculation by Edwards and Saam [1]. [Their model involves two parameters: a length $\delta = d(\ln a_0)/dK$, where $K = r_1^{-1} + r_2^{-1}$ is the curvature of the surface, and an area $a = d\delta/dK$. Several sets of parameters have been used, determined by fitting the thermodynamic data ($a = +1.5 \text{ \AA}^2$, $\delta = 0$ [3] and $a = +1.0 \text{ \AA}^2$, $\delta = -0.336 \text{ \AA}$ [1]), the latter giving a somewhat

better agreement.]

The coverage dependence of the intensity of these excitation branches is also of interest (Fig. 3). Let us consider first the case of *wave vectors smaller than 1.5 \AA^{-1}* , where the branches are well separated.

For thin films (3 to 3.5 layers), only ripplon excitations are observed. Their intensity is very small for 2.96 layers, then increases rapidly between 3.09 and 3.54 layers, and saturates above 4 layers. That means that ripplon excitations are observed in films as thin as one liquid atomic layer (remember that the two layers adjacent to the substrate are solid). Also, only two liquid layers are necessary to observe the full intensity of ripples in the wave-vector range explored here. These effects are due to the small ripplon penetration depth expected at such high wave vectors [4,5]. The ripplon dispersion curve displays only a weak dependence on coverage. As seen in Fig. 4, some deviation from the "thick film" curve towards higher energy is found for the film of 3.54 layers; larger deviations are seen at 3.24 layers (i.e., 1.24 liquid layers), but a precise fit of the peak positions is rendered more difficult due to the reduced intensity. Qualitatively, this trend seems to agree with theory [4].

As the coverage is increased above 3.5 layers, the intensity on the phonon-rotor branch (Fig. 3) starts to develop slowly, reaching a linear dependence as a function of coverage—with the slope expected for bulk liquid—at about 4 layers. The spectrum of this branch practically coincides with the roton-phonon excitations of bulk superfluid ^4He . For very thin films, however, a small shift to lower energies is observed, mainly around the maxon region; this reflects the lower average density of

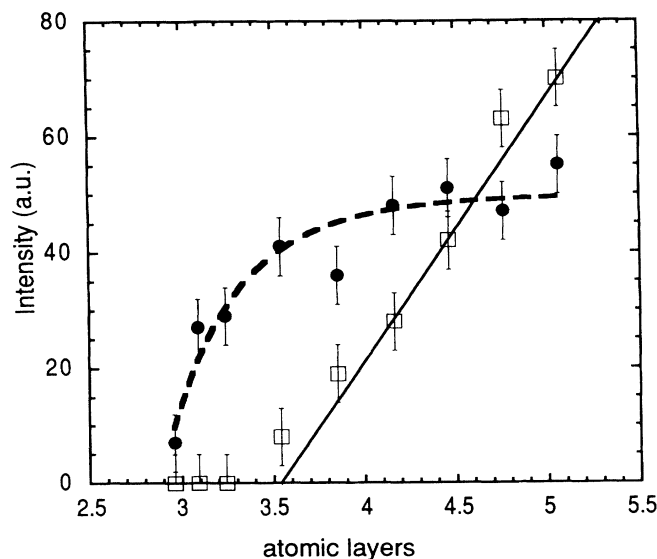


FIG. 3. Intensity (height of the observed peak, arbitrary units) of the phonon (\square) and the ripplon (\bullet) measured at $Q=0.8 \text{ \AA}^{-1}$ as a function of coverage at $T=0.7 \text{ K}$. The slope of the solid line corresponds to that measured for the phonon in bulk liquid ^4He .

the film compared to bulk. The characteristic differences in the growth of the ripplon and phonon intensities as a function of coverage, depicted in Fig. 3, obviously support the interpretation of the signals as surface and bulk excitations, respectively.

Present microscopic theories provide only a qualitative picture of the dispersion relations of ripples and phonons and their coverage dependence. At low Q , the ripplon is found to be the lowest-lying excitation, in agreement with our experimental data. Moreover, the existence of several "ripplonlike" branches lying above the ripplon dispersion curve, but still below the bulk phonon-rotor excitations, has been predicted [4,5]. In this region some intensity is observed in our spectra between the ripplon and phonon branches. This effect is more clearly seen at our highest coverages (see Fig. 1). However, the theoretical coverage dependence of those branches, and even their number, do not seem to agree with our results.

The *high- Q region* ($Q \geq 1.5 \text{ \AA}^{-1}$) is certainly the most difficult to analyze, despite the relatively large intensity of the neutron spectra. Present theories indicate that a level crossing of the ripplon branch with the phonon (bulklike) branch is expected for $Q \geq 1 \text{ \AA}^{-1}$, and that for $Q \sim 2 \text{ \AA}^{-1}$, the lowest-lying mode is a volume excitation [5]. The data clearly show that the ripplon branch points towards the roton minimum. This provides support to the hybridization with the volume mode proposed theoretically [4,10]. Note that this effect is not included in the theory of Ref. [1] and therefore the agreement between the theoretical and the experimental

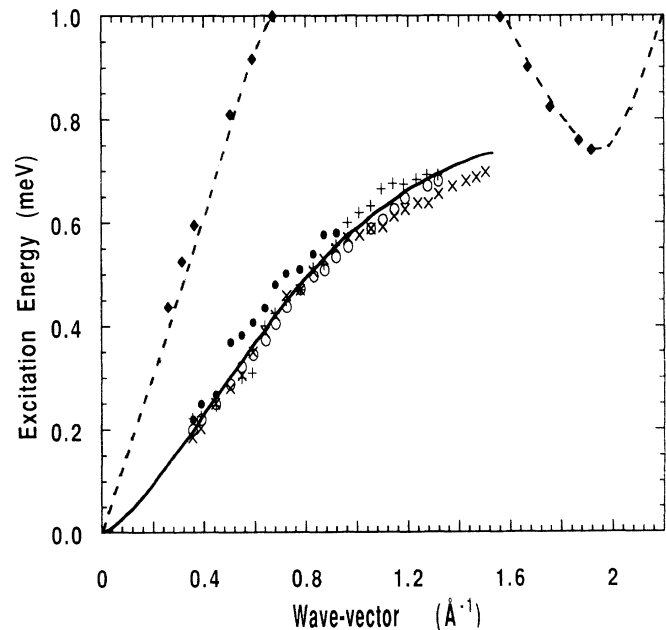


FIG. 4. The ripplon dispersion for various coverages: 5.06 (\circ), 4.16 (\times), 3.85 ($+$), and 3.54 (\bullet) layers. Solid line: quantum hydrodynamic theory [1]. The dispersion relation of bulk ^4He at zero pressure is shown by a dashed line, and the results with the cell filled with ^4He by \blacklozenge .

dispersion curves (Fig. 4) may be fortuitous above 1 \AA^{-1} .

The region of the spectrum below the roton minimum has been studied previously [11]. An excitation was observed and "tentatively identified as an excitation of the high-density liquid layer adjacent to the solid layers of the film," a "surface roton." Our former [6,12] and present results on different graphite samples show that this mode, at an energy slightly below 0.6 meV, persists even when the cell is filled with bulk helium, indicating that it is not related to the free surface of the liquid (this corroborates the assumption made in Ref. [11] based on indirect experimental evidence). Clearly, the signal observed in Fig. 1 below the roton minimum should not be interpreted as belonging to the ripplon dispersion curve.

In conclusion, the ripplon spectrum up to very large wave vectors, corresponding to wavelengths of atomic scale, has directly been observed for the first time. The data at intermediate wave vectors support quantitatively the semiempirical theoretical prediction of Edwards and Saam for the lowest-lying ripplon state. Our results also display qualitative features which agree with recent microscopic theories of inhomogeneous quantum fluids: In films the spectrum is not simply characterized by a bulk-like phonon-roton branch and a single ripplon branch with a rotonlike minimum, as demonstrated by the additional intensity observed between those branches, and also by the complexity of the spectra at high wave vectors. The excitations observed around the wave vector of the roton minimum, but at lower energy, do not seem to be connected to the ripplon spectrum.

The experimental and theoretical understanding of the high-wave-vector region is still incomplete; the expected development of quantitative microscopic models should allow a better analysis of our data in this region.

This work has been partially supported by the German

Federal Ministry of Research and Technology (BMFT).

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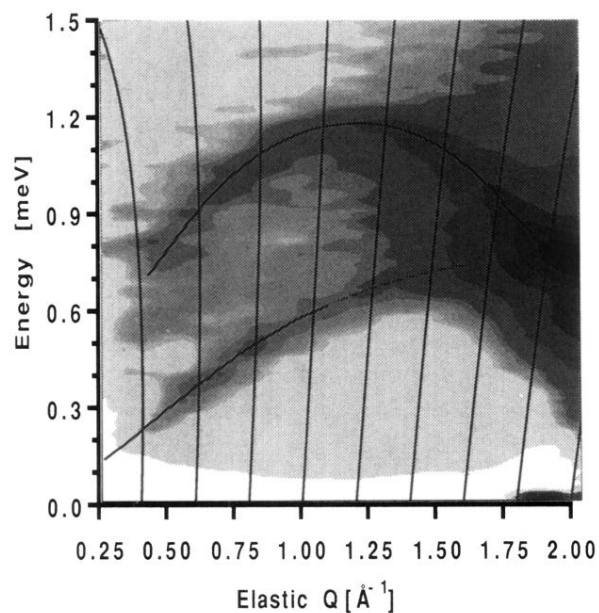


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