Ripplons in ^{4}He Films Observed by Neutron Scattering

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We report the first observation of ripplon excitations at large wave vectors. The experiments were performed on liquid-^{4}He films of atomic thickness adsorbed on a graphite substrate. Neutron-scattering techniques were used to determine the inelastic structure factor of surface (ripplon) and bulklike (phonon-roton) elementary excitations. The lowest lying mode at low wave vectors is experimentally shown to be a surface mode; its dispersion relation agrees well with the quantum-hydrodynamic theory of Edwards and Saam. The predictions of recent microscopic models are compared to the data at high energies and momenta.

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The study of interfaces and surfaces has motivated substantial theoretical and experimental research in recent years. Whereas elementary excitations at solid surfaces are rather well understood, relatively little is known about the elementary excitations at liquid surfaces, which are quantized capillary waves, also called ripplons. In a normal fluid these excitations are overdamped at large wave vectors due to the effect of the viscosity. In a superfluid, however, the damping is expected to be much lower and hence the observation of well-defined excitations with wavelengths down to interatomic distances should be possible. An obvious system for this purpose is ^{4}He below the lambda temperature, and in fact the existence of large-wave-vector ripplons both on bulk ^{4}He and in films has been predicted by theory, and has indirectly been confirmed by experiment [1-3]. As long as the liquid can be treated as a continuum the dispersion relation of ripplons follows from the hydrodynamics of an incompressible fluid as $\omega^2=(\alpha/\rho)k^3$, where α is the surface tension, ρ the liquid density, and k the wave vector. A correction factor $\tanh(kd)$ is applied when the depth of the liquid is smaller than the ripplon wavelength. A contribution due to gravity becomes important only at extremely small k. The validity of this hydrodynamic formula is restricted to wavelengths sufficiently large compared to an atomic scale.

The ripplon dispersion relation can be used to derive the temperature dependence of the surface tension at very low temperatures. However, detailed measurements [3] of $\alpha(T)$ revealed a much larger temperature dependence than expected from the classical dispersion relation mentioned above. To explain this discrepancy, several authors proposed modified dispersion curves for wave vectors above 0.5 Å^{-1}. The generalized quantum hydrodynamic model [1,3], in particular, extends the validity of the hydrodynamic approach to the quasimicroscopic regime.

This subject has been reinvestigated recently [4,5] by theorists interested in the properties of inhomogeneous quantum many-body systems. Detailed calculations for the free surface of bulk ^{4}He, and for films of atomic thickness adsorbed on a solid substrate, at selected coverages, have yielded the dispersion relation and the dynamic structure factor of the elementary excitations in the microscopic regime, i.e., for large wave vectors. Experimentally, however, little direct evidence has been available up to now [6] on that part of the ripplon dispersion curve. A study in this regime requires a microscopic probe like inelastic neutron scattering (INS). Because of the low neutron cross section of ^{4}He, the measurement has to be performed on samples with a large surface-to-volume ratio; also, a very high neutron flux, a low and clean background, and long counting times are needed in order to observe surface excitations.

We have measured the inelastic structure factor of ^{4}He adsorbed on the basal plane of graphite as a function of coverage. The INS measurements were performed at the time-of-flight spectrometer IN6 at the Institut Laue-Langevin's reactor using a neutron wavelength of 5.12 Å. The detectors are located in the angular range 11.9°-113°; this corresponds, for elastically scattered neutrons, to momentum transfer (Q) between 0.254 and 2.046 Å^{-1}. Spectra were taken for 37 angles corresponding to equally spaced elastic Q values. The measured energy resolution at zero energy transfer is slightly Q-dependent due to sample size effects, increasing with scattering angle from 0.080 to 0.11 meV. The sample consisted of 31.70 g of Papyex [7] sheets oriented with their c axis normal to the scattering plane, separated by gadolinium foils to reduce multiple scattering. The sample was located in an aluminum container and thermally connected by a copper rod to the ^{3}He stage of a cryostat. During the measurements the temperature was kept at 0.65 K, far below the lambda temperature $T_\lambda=2.17$ K, but an annealing of the adsorbed films above 2 K was performed after each change in coverage. The total surface area determined by adsorption isotherms and neutron diffraction was 730 m$^2\pm2\%$; the ^{4}He monolayer coverage (0.112 atom/Å^2) was 304 cm^3 STP. Data obtained before any ^{4}He was adsorbed were used as back-

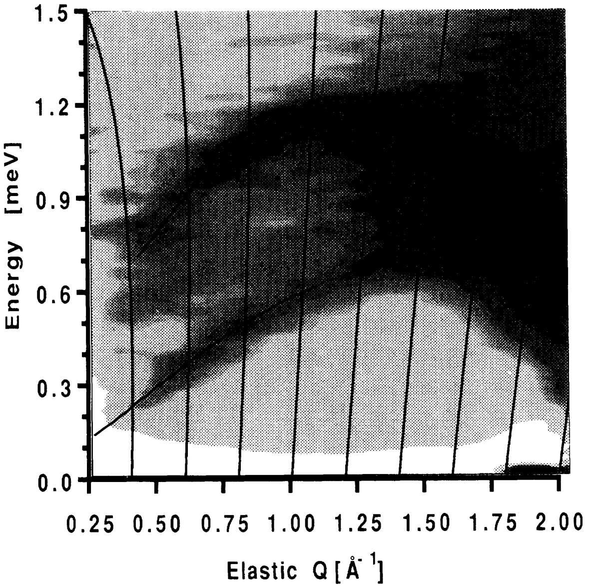

FIG. 1. Contour plot of the inelastic structure factor $S(Q,\omega)$ for a coverage of 0.448 atom/Å^2 (5.06 layers). Elastic Q values, given on the abscissa, mark the origin of constant-Q lines. The magnitude of $S(Q,\omega)$, indicated in arbitrary units by different shades of grey, increases from white 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 ^{4}He phonon-roton dispersion relation. The lower line is the ripplon dispersion curve calculated by Edwards and Saam [1], using $a=+1.0$ Å^2, $\delta=-0.336$ Å.

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 atom/Å^2. Finally, a scan with the cell filled with bulk superfluid ^{4}He 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 atom/Å^2 for the first layer and 0.094 atom/Å^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 ^{4}He, obtaining somewhat arbitrarily 0.078 atom/Å^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 ~5 Å [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, 4.46, 4.76, and 5.06 layers.

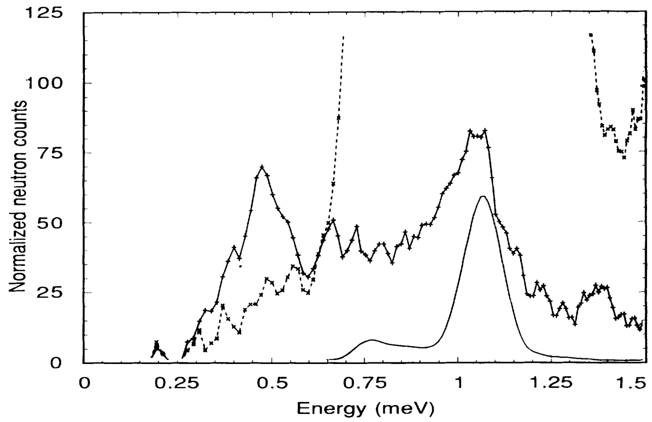

FIG. 2. Inelastic spectrum (arbitrary units) for $Q=0.8$ Å^{-1} as a function of energy. The solid line with +'s corresponds to a coverage of 5.06 atomic layers. The ripplon peak is found at 0.47 meV and the bulk phonon at 1.05 meV. Dashed line with ×'s: spectrum with the cell filled with bulk liquid; 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).

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-roton 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 Papyex sample. This enabled us to determine the lower-branch dispersion relation unambiguously up to wave vectors ~1 Å^{-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 ^{4}He, 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 $\delta=d(\ln\alpha_0)/dK$, 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 ($a=+1.5$ Å^2, $\delta=0$ [3] and $a=+1.0$ Å^2, $\delta=-0.336$ Å [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* Å^{-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 ripplons 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-roton 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 ^{4}He. 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 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 ripplons 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-roton 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 ($Q \geq 1.5$ Å^{-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 $Q \geq 1$ Å^{-1}, and that for $Q \sim 2$ Å^{-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

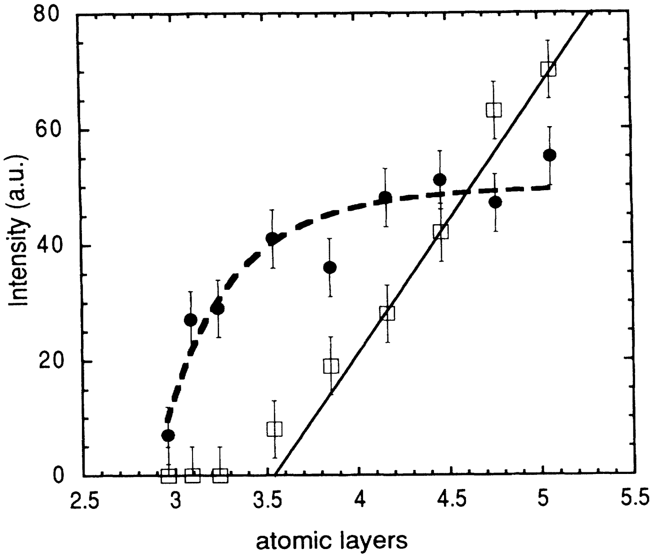

FIG. 3. Intensity (height of the observed peak, arbitrary units) of the phonon (□) and the ripplon (●) measured at $Q = 0.8$ Å^{-1} as a function of coverage at $T = 0.7$ K. The slope of the slid line corresponds to that measured for the phonon in bulk liquid ^{4}He.

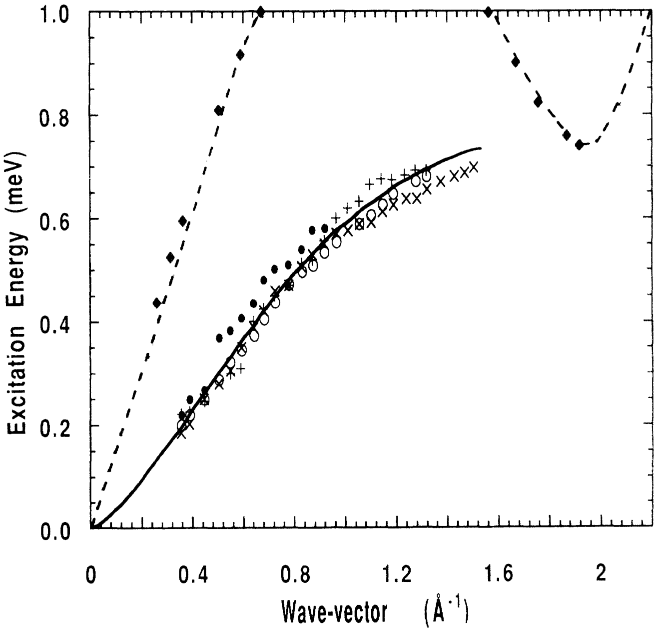

FIG. 4. The ripplon dispersion for various coverages: 5.06 (o), 4.16 (×), 3.85 (+), and 3.54 (●) layers. Solid line: quantum hydrodynamic theory [1]. The dispersion relation of bulk ^{4}He at zero pressure is shown by a dashed line, and the results with the cell filled with ^{4}He by ◆.

dispersion curves (Fig. 4) may be fortuitous above 1 Å^{-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.

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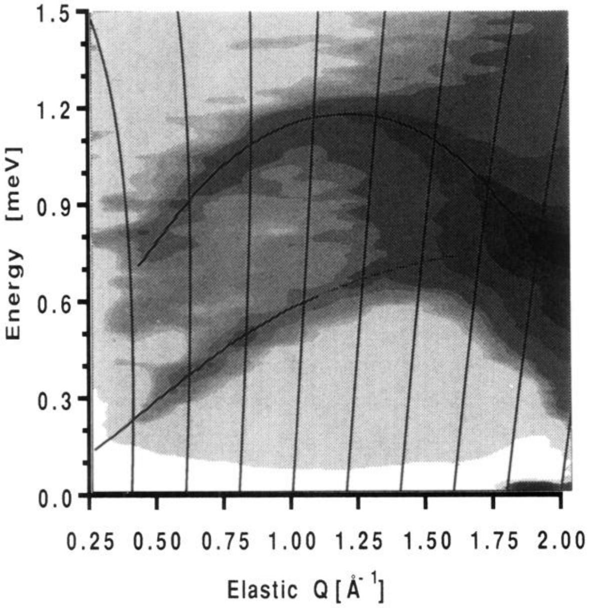

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