Excitations in Few-Atomic-Layer Adsorbed Helium Films: The Two-Dimensional Roton

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Inelastic neutron scattering from helium adsorbed on Graphon indicates the presence of two types of excitations: one bulklike, which appears to be associated with the normal density part of the film; and a second, at lower energy, which is thought to originate in the high-density liquid layer immediately adjacent to the solid layers of the film. The latter is believed to be responsible for the reduced roton gap inferred from bulk studies of few-atomic-layer superfluid films.

Recent studies of few-atomic-layer superfluid ^{4}He films show them to differ in a number of interesting respects from their bulk counterparts. One of the more striking of these differences —particularly evident in third[1-3] and fourth[4] sound measurements—is that the superfluid fraction is relatively smaller in films. This has led to suggestions that the roton gap is smaller in films than in bulk liquid, a view also supported by heat capacity[5] measurements.

Thus far, attempts to observe directly the renormalization of roton energies in surface superfluid films have not met with notable success. Early neutron studies of helium adsorbed on Grafoil, an exfoliated graphite foil, yielded excitation spectra in the region of the roton minimum not significantly different from those of the bulk liquid even when the films contained, on average, as few as six layers of atoms.[6] Later, evidence of broadening of the roton peaks and structure on their low-energy side was reported.[7] In other respects, however, the excitations were not observed to be noticably altered.

Some time ago, we began to explore the possibility of using Graphon rather than Grafoil as a substrate for neutron studies. Graphon[8] is a graphitized carbon powder of random orientation with three to four times the specific surface area of Grafoil. Its surfaces, like those of Grafoil, consist primarily of basal planes but they are neither as homogeneous nor as uniform. Ultimately, our Graphon experiments proved to be more sensitive than the earlier studies. As we will show, they reveal that the scattering from helium films on graphite is actually composed of two parts: one coming from bulklike excitations, which we suspect has its origin in the normal-density part of the film; and a second, not present in scattering from bulk liquid, which we tentatively identify with excitations of the high-density liquid layer adjacent to the solid layers of the film. Our reasons for suggesting these identifications will become clear in due course.

All of the measurements to be described were made on a triple-axis neutron spectrometer operating in the constant-Q mode with a fixed incident neutron energy of 5.0 meV. As is usual, the scattering from the surface film was taken as the difference between intensities with and without helium in the cell. The sample consisted of 34.6 g of Spheron-6 powder. Nitrogen and helium adsorption isotherm measurements were used to determine monolayer coverage. The specific area of the substrate, as obtained from these measurements, was 79 m^2/g, almost four times that of typical Grafoil samples.

Before considering the experimental data, it will be helpful to review briefly what is currently known about how helium overlayers are deposited on graphite surfaces. Heat-capacity[9] and neutron-diffraction measurements[6] show that at low temperatures the first monolayer forms an incommensurable, two-dimensional solid with a triangular lattice structure. It is highly compressed, having a nearest-neighbor spacing of 3.17 Å. The second layer is deposited as a liquid and remains so until solidified by the compression of a partial third layer. When solidified, the second layer forms a triangular lattice with a nearest-neighbor spacing[10] of 3.54 Å; it should be noted that its atomic density is only slightly greater than that in the basal plane of hcp solid helium at the solid-liquid boundary. Layers subsequent to the second are always liquid unless the system is under pressure. In converting our data to total layers we assumed the first-layer density to be 0.115 atom/Å^2, the second to be 0.092 atom/Å^2, the third 0.086 atom/Å^2, and subsequent layers 0.077 atom/Å^2.

Several sequences of measurements were made, all at temperatures less than 1.3 K. The first was a study of the Q dependence (scattering vector dependence) of the inelastic scattering from helium films of 7.3 total layers nominal thickness. What was observed over the entire Q range from 0.45 to 2.3 Å^{-1} were peaks characteristic

of the scattering from the bulk liquid at the saturated vapor pressure, superimposed on a background of multiple scattering due to inelastic processes in the film in combination with elastic (Bragg) processes in the substrate. The multiple-scattering part of the spectral distributions, we found, could be satisfactorily reproduced by taking account of two-scattering interactions alone. Accordingly, we computed the two-scattering distributions at each Q and used these to correct the 7.3-layer spectra for multiple-scattering background. When this was done we found that not only did the single-scattering peaks have the same energies as those from bulk helium but their intensities exhibited exactly the Q dependence observed in the bulk liquid as well.

Next we investigated the coverage dependence of the scattering at $Q = 2.0$ Å^{-1}, a value chosen to be as close to the roton minimum as the intense background of scattering from the (002) Bragg peak of the substrate would allow. Representative difference spectra from this scan sequence, corrected for multiple scattering (the corrections never amounted to more than 8%), are shown in the upper part of Fig. 1. At higher coverages the intense bulk-roton peak at 0.76 meV is the dominant feature. With decreasing coverage the bulk signal broadens and decreases in intensity but there is no significant downward shift in energy. Finally, at the lowest coverage, 4.0 total layers, the bulk signal becomes so broad as to be virtually unrecognizable.

To parameterize the bulklike part of the data we folded a cross section of Lorentzian form with the resolution function of our spectrometer and fitted the resulting peak shapes to the experimental spectra. The fitted shapes appear as the solid lines and their dashed extensions in Fig. 1. In Fig. 2 we show the coverage dependence of the bulklike excitation energies (open triangles) and their inverse linewidths (solid circles); the solid circles in Fig. 3 indicate the coverage dependence of the corresponding integrated intensities. Note that the excitation energies appear to be independent of coverage and that there is a linear relationship between intensity and coverage over a range extending from 5 to 25 total layers. Only at the lowest coverage is there any evidence of a departure from linearity, possibly as a result of a systematic error in the background determination but more likely a manifestation of the fact that in the thinnest films excitations with wave vectors parallel to the film plane are more stable than those with wave vectors of other orientations.[6] Both the inverse linewidth and the integrated intensity extrapolate to zero in the neighborhood of three total layers of which it should be remembered that two are solid and one liquid. Thus the bulklike part of the signal disappears at the point where only a single atomic layer (or at most two layers) of liquid remain in the system.

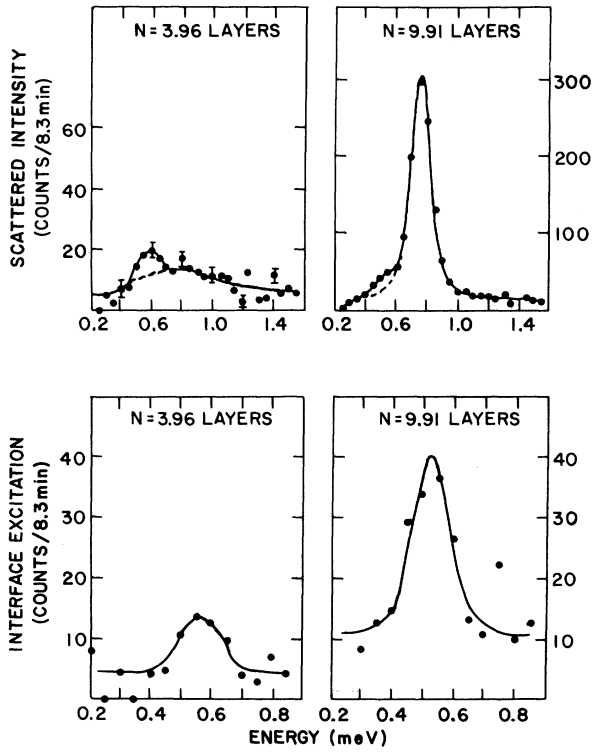

FIG. 1. Upper: Inelastic scattering at $Q = 2.0$ Å^{-1} from ^{4}He films adsorbed on Graphon (multiple-scattering and unfilled-cell backgrounds have been subtracted); $T \leq 1.32$ K. The solid lines are Lorentzians fitted to the bulklike roton peak at 70 meV and to the peak at 0.54 meV which we identify with scattering from the liquid-solid interface. Lower: Inelastic scattering in the neighborhood of 0.54 meV with background of scattering from bulklike rotons subtracted. The solid lines are fitted Lorentzians.

Aside from the bulklike peaks so prominent at higher coverages, there is significant excess scattering (also noted in Ref. 7) in the region between 0.5 and 0.7 meV. At the lowest coverage, this scattering is the only obviously recognizable feature in the data. In the lower part of Fig. 1 we show it as it appears when the bulklike part of the signal (estimated by extrapolating the fitted line shapes) is removed by subtraction. Although there is more statistical scatter than

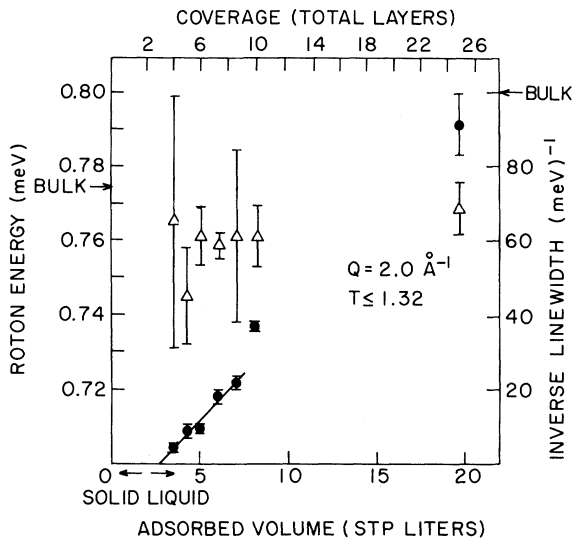

FIG. 2. Coverage dependence of the excitation energies (open triangles) and the inverse linewidths (solid circles) of the bulklike rotons of the film. Note the suppressed zero on the excitation energy scale.

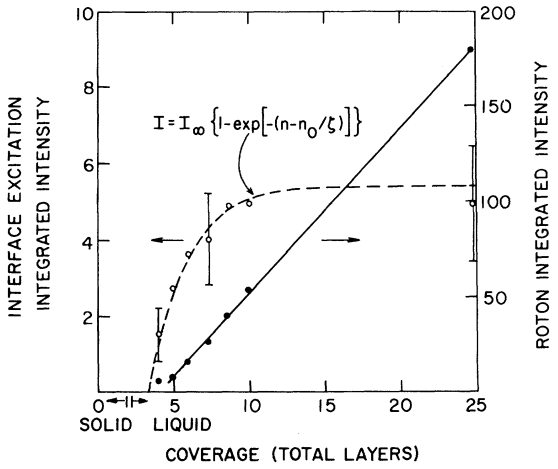

FIG. 3. Coverage dependence of the integrated intensity of the bulklike-roton scattering (solid circles) and the scattering we associate with the liquid boundary layer (open circles). ζ and n_0 were assigned values of 2.3 and 3.3 layers in plotting the dashed line.

might be wished, it nevertheless seems clear that another excitation of lower energy is present in the system. Fitting Lorentzians to these peaks yielded an excitation energy of 0.54± 0.03 meV; linewidths could not be determined because the observed peak widths were resolution limited. The open circles in Fig. 3 show how the integrated intensity of this part of the scattering varies with coverage. There is a region of increase up to about eight total layers; then the intensity saturates and does not change further. Extrapolation to zero intensity indicates that this scattering disappears at about three total layers. It is essential to add that at a coverage of 7.4 total layers and with $Q = 2.08$ Å^{-1} we saw a similar peak at an energy of 0.60± 0.03 meV but there was no evidence of any extra low-energy scattering (or other anomalies) outside the region of the roton minimum. At $Q = 2.08$ Å^{-1}, the bulklike component had a 50% greater width than at $Q = 2.0$ Å^{-1}; however, the width of the low-energy peak was, as before, resolution limited. A further point to note is that the low-energy excitations fall within the span of roton gap energies deduced from macroscopic studies of few-layer helium films.[1-5]

One plausible approach to understanding the bulklike linewidths is to assume, as the few-atomic-layer geometry suggests, that the bulk excitation lifetimes are limited by boundary scattering. If this is so, then it should follow that the line widths are governed by the simple relation $\Gamma = \hbar v/\lambda$, where λ is the mean free path between boundaries and v the excitation velocity. Near the roton minimum, Q_0, the velocity is $v = \hbar(Q - Q_0)/\mu$, with μ the effective mass. The linewidth relation therefore takes the form $\Gamma = \hbar^2(Q - Q_0)/\mu\lambda$. When the widths of Fig. 2 are substituted into this expression together with values of Q_0 and μ appropriate to bulk helium at the saturated vapor pressure, values for λ increasing linearly from 4.2 to 86 Å are obtained, depending on the coverage. These seem physically reasonable. Furthermore, the increased width of the bulklike excitation at $Q = 2.08$ Å^{-1} is seen as a natural consequence of the fact that its velocity is larger. We come thus to the view that rotons in helium overlayers are identical to those in bulk helium; their lifetimes are, however, limited by the thickness of the film.

To reach an equivalent understanding of the low-energy part of the neutron scattering is less straightforward. The film geometry suggests that the observed excitations are associated with either the free surface of the liquid or the boundary between liquid and solid layers. Since fourth-sound studies of helium in full pores indicate a reduced roton gap even with no free liquid surfaces,[4] the first alternative appears unlikely. The second implies that the low-energy mode is an excitation of the dense liquid layer immediately adjacent to the solid-helium boundary. If this is

correct, it is easy to understand why the mode is so long-lived: The mean free path is not limited (as for bulk rotons) by the thickness of the film but rather by the mean distance between discontinuities along the boundary, which can be presumed to be much greater. Moreover, the assumption of a boundary mode leads to an expected variation of scattered-neutron intensity with film thickness of the form $I = I_\infty\{1 - \exp[-(n-n_0)/\zeta]\}$, where ζ is a parameter characterizing the depth to which the excitation penetrates into the film and n_0 is the minimum thickness film in which the excitation is viable. This expression is plotted in Fig. 3 with $\zeta = 2.3$ and $n_0 = 3.3$ layers. It is clearly an excellent representation of the data. Finally, we should note that in bulk liquid of the same density as the boundary layer, the roton gap is 0.60 meV,[11] not very different from the value we observe. Taken together, the data suggest an excitation confined to the high-density liquid boundary layer: in effect, a two-dimensional roton.[12]

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Two-Dimensional ^{3}He in ^{4}He Films

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Measurements and analysis of heat capacity for ^{3}He in ^{4}He films at temperatures below 350 mK are reported. The data show that ^{3}He is less bound to the free surface of the ^{4}He film than to the bulk ^{4}He surface, and that it has a larger effective mass. Both of these parameters depend on ^{3}He surface coverage. The ^{3}He dissolved within the film behaves as a two-dimensional system for sufficiently thin ^{4}He films.

In liquid mixtures of ^{3}He in ^{4}He with a free surface the ^{3}He resides preferentially at this surface in a state which is about 2.2 K below the binding energy to bulk ^{4}He.[1] This state becomes appreciably occupied at low temperatures thus yielding a most ideal realization of a two-dimensional (2D) Fermi system. The ^{4}He surface is completely homogeneous and, at sufficiently low temperatures, there are very few excitations; thus one may think of the ^{4}He as a substrate at very nearly its quantum-mechanical ground state. With suitable choice of experimental parameters, surface-to-volume ratio, amount of ^{3}He, and temperature, one can readily achieve and study surface coverages from as low a density as one can detect, up to a full atomic layer. Beyond about a full layer, at least for the surface of bulk ^{4}He, there will be an appreciable concentration of ^{3}He within the ^{4}He. This concentration can be increased up to the phase-separation limit. The measurements which have been made to probe properties of the surface ^{3}He are surface ten-

