Two-Dimensional Interface Excitations: Measurements in ^3He-^4He Mixtures

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The experimental data which we have gathered on the superfluid mass content in films of ^3He-^4He mixtures yield a characteristic length which varies markedly both with temperature and ^3He concentration. The data are analyzed to present their implications on the energy gap and on other parameters characterizing the two-dimensional helium-boundary-interface excitation spectrum.

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Several years ago it was demonstrated[1,2] that the existence of two-dimensional helium interface excitations is clearly evidenced by data on the superfluid mass in adsorbed thin films. We deduced the quantitative features of the rotonlike dispersion curve characterizing these excitations from the results then available. We were gratified to find, in the elegant neutron-scattering[3] experiments recently reported by Thomlinson, Tarvin, and Passell, substantial confirmation of our conclusions. The single and important difference between these works lies in the value of the energy gap. An energy gap of 6.3°K (0.55 meV) is deduced from the neutron-scattering data; our data yielded 4.5°K The neutron experiments, having been carried out at only one temperature, did not confirm our finding that the dispersion-curve parameters are temperature independent.

The essential conclusion—a two-dimensional (2-D) excitation—rests, in both of these methods, upon a similar fundamental observation. Size-dependent features which, in thin-film experiments, persist at constant strength regardless of film material thickness must be attributable to a surface rather than to a volume effect. In our case, the superfluid deficit in a thickening film persists as a constant independent of film thickness; in neutron experiments, a particular inelastic process exhibits a persistent saturated intensity independent of film extent at large thicknesses. We have found that thin-film ^3He-^4He mixtures exhibit the same essential feature signaling an interface excitation; a superfluid deficit persists in ever thickening films (cf. Fig. 1).

The relevant experimental results are shown by

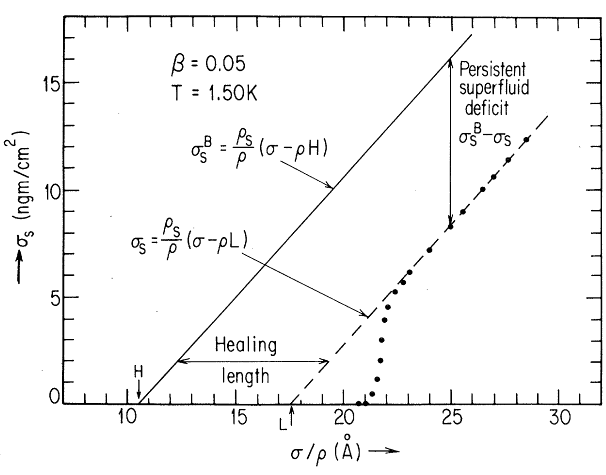

FIG. 1. The superfluid mass in a ^3He-^4He mixture film as a function of the total amount of film mass present for a 5% ^3He concentration at temperature 1.50°K. The points are experimental data. The superfluid deficit persists independent of film material increase as shown.

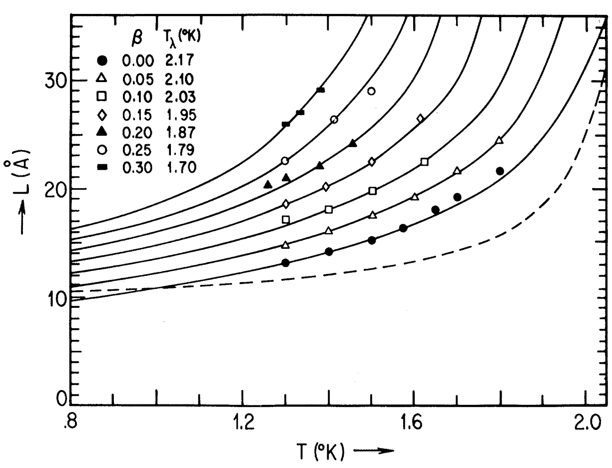

FIG. 2. The characteristic intercept length as a function of temperature. The points are the experimental data. The full curves represent the formula $L = H + \hbar\, k^3(m^*/8\pi KT)^{1/2}\exp(-\Gamma/KT)$, where the parameters employed are those listed in the $\Gamma = 4.5$°K column of Table I. The dashed curve represents the same formula plotted for a 5% ^3He concentration but with the single change that 4.5°K is replaced by 6.3°K in the exponent.

the data points of Figs. 1 and 2. The mass per unit area of superfluid in the film is σ_s and σ is the total areal mass density of the film. The quantity $\rho=\rho(\beta,T)$ is the bulk volume mass density of a liquid ^{3}He-^{4}He mixture at temperature T with ^{3}He concentration β. It is employed to form the film-thickness estimator σ/ρ, used as abscissa in Fig. 1. The dashed straight line drawn through the experimental points is always found to have the expected slope of $\rho_s(\beta,T)$—the bulk volume superfluid mass density corresponding to a mixture at concentration β and temperature T. The intercept of this line with the horizontal axis yields a characteristic length L. The function $L=L(\beta,T)$ constitutes a central finding of our experimental data. An inspection of Fig. 1 makes it evident that ρL is just that mass (per unit area) of helium which never contributes to the superfluid in the system no matter how thick the sample becomes. In Fig. 2 the points show the experimental values found for L. The data were gathered via quartz-crystal adsorptometer measurements. Detailed experimental expositions can be found in previous and forthcoming works.[4-8]

The essential clue to the data interpretation lies in Fig. 1. In this figure the unbroken straight line shows the asymptotic behavior expected[2,5,9] of the film as its thickness grows indefinitely large. This line follows the equation

$$\sigma_s^B=(\rho_s/\rho)(\sigma-\rho H). \tag{1}$$

Equation (1) merely states the evident notion that for thick films the bulk idealized superfluid mass (areal density σ_s^B) is directly proportional to the total mass present less some mechanically excluded mass. We denote the "excluded mass" by ρH. This represents the physical amount of solid plus high-density compressed mass present in the film. Because of its compressed state, this portion of the film cannot support any superfluidity. Having little to do with superfluidity, H does not vary appreciably with temperature.[5,9] By contrast, the measured intercept, L, varies quite appreciably with temperature (cf. Fig. 2). The "superfluidless" excluded mass determined experimentally is ρL; the excluded mass expected is ρH.

Insofar as L differs from H, there is a persistent superfluid deficit to be accounted for. As shown in Fig. 1, the superfluid mass is less than that expected ideally. And this difference is seen to be constant, independent of film thickness, as the film thickens indefinitely. This feature signals a surface effect; it excludes a volume one. The surface area remains constant as the film thickens; so does the superfluid mass deficit.

Less superfluid signifies more normal fluid. Hence we interpret the superfluid deficit as revealing the normal-fluid excitations associated with a surface of the helium. Denoting the surface excitation normal-fluid contribution by $\delta\sigma_n$, the concept is summarized by

$$\sigma_s^B-\sigma_s=\rho_s(L-H)=\delta\sigma_n. \tag{2}$$

This equation refers only to sufficiently thick films in which the superfluid deficit $\sigma_s^B-\sigma_s$ is independent of thickness. Thin films merely provide experimental access to the phenomenon. But the effect is a surface one on bulk helium; it is not characteristic of very thin films for which Eq. (2) is quite untrue.

How to calculate the normal-fluid mass for a 3-D bath of excitations was first demonstrated many years ago by Landau.[10] It is a simple matter to carry his ideas over to a gas of excitations characterized by only two independent spatial degrees of freedom.[2,11] The contribution from the 2-D high-momentum rotonlike excitations is

$$\delta\sigma_n=\hbar k^3(m^*/8\pi KT)^{1/2}\exp(-\Gamma/KT), \tag{3}$$

where k is the momentum at which the "roton dip" occurs and Γ is the energy at the dispersion curve minimum—the 2-D roton energy gap. The reciprocal of m^* represents the curvature of the dispersion curve at the roton minimum. Of course, h and K are Planck's and Boltzmann's constants, respectively.

The combination of Eqs. (2) and (3) yields an unequivocal prediction for the expected behavior of L upon T for each β. The form is clearly defined via $\rho_s=\rho_s(\beta,T)$, a bulk parameter tabulated from experimental data,[12] and the exponential and square-root temperature dependences. We demand of the data that they yield the values of Γ, the product $k^3\sqrt{m^*}$, and H while demonstrating a firm fit to the form of the equation at all T for every β. On physical grounds we certainly expect H to vary with β.[5,9] Our analysis yielded β-independent values for the other two parameters. They are, of course, forbidden to vary with temperature as is H; i.e., the temperature dependence of L at each β is firmly fixed by the form of (2) and (3).

The results of our analysis are summarized by the curves drawn in Fig. 2 and the contents of Table I. It is clear from this figure that the data are fitted quite well at all β by the single energy gap $\Gamma=4.5\,^\circ$K. The values of $H=H(\beta)$ and $k^3(m^*/$

TABLE I. Representative values of the parameters which produce a match of the equation $L=H+\hbar\, k^3(m^*/8\pi KT)^{1/2}\exp(-\Gamma/KT)$ to the experimental points for the two choices of special interest for Γ.

β	H (Å) $\Gamma=4.5\,°\mathrm{K}$ $k^3(m^*/m)^{1/2}=4.2$ Å^{-3}	$\Gamma=6.3\,°\mathrm{K}$ $k^3(m^*/m)^{1/2}=10.5$ Å^{-3}
0.00	9.2	10.6
0.05	10.5	12.1
0.10	11.6	13.5
0.15	12.6	15.0
0.20	13.5	16.3
0.25	14.3	17.4
0.30	15.2	19.3

$m)^{1/2}$, characterizing the curves exhibited, are in Table I. For $k=2$ Å^{-1}, as suggested[3] by the neutron scattering researchers, we find $m^*/m=0.27$, a result close to that for 3-D bulk ^{4}He.

Motivated by the neutron-scattering results we carefully explored other values for the three parameters Γ, $k^3(m^*/m)^{1/2}$, and H for fits to our data. Our conclusions are the following:

(1) Within a range of Γ extending down to about 2 °K and up to about 8 °K, the data can always be fitted. But once a value of Γ is chosen within this range, the other two parameters are unequivocally determined by the data.

(2) Setting Γ equal to the bulk energy gap of 8.8 °K (neutron scattering 0.76 meV) reveals a systematic deviation from experiment. Our data clearly forbid this and higher values of Γ. The form of Eqs. (2) and (3) cannot be fit with $\Gamma=8.8$ °K regardless of the choice for the other parameters.

(3) The value, $\Gamma=6.3$ °K, derived from the neutron-scattering experiments, can be fitted to our data. The curves derived look much like those shown in Fig. 2. However, the values of $k^3(m^*/m)^{1/2}$ and $H=H(\beta)$ must be readjusted so as to secure this fit. Their values, for $\Gamma=6.3$ °K, are given in Table I. Note that for the expected $k\simeq 2$ Å^{-1} the implication at $\Gamma=6.3$ °K, is that $m^*/m=1.7$, a value over eight times greater than that for bulk.

(4) The dashed curve of Fig. 2 illustrates the sensitivity of the fit to our data for visual comparison. It is a plot for a 5% ^{3}He concentration with $\Gamma=6.3$ °K but employing $k^3(m^*/m)^{1/2}=4.2$ Å^{-3}. It is clear that this cannot fit the data no matter what H is chosen. In general, for a particular Γ, the other parameters are determined to within about 15%.

(5) The values deduced for $H(\beta)$, for both energy-gap cases presented, are entirely commensurate with expectations. They imply a mean density for the compressed region of about twice that for bulk ^{4}He taking this region to be between one and two atom diameters thick, i.e., about 5.4 Å. That H rises with β is also commensurate with other measurements on the compressed mass region.[5,7,9]

The energy-gap value, Γ, may reflect the nature of the substrate via the van der Waals force which produces the compression at the substrate. The neutron scattering substrate, Graphon, is characterized by a van der Waals strength about three and a half times greater than that for our substrate.

In the literature on superfluid critical distances an intercept length is rarely measured. Usually the length reported is the onset one—where the precipitous onset of superfluidity is observed. The data points in Fig. 1 show a precipitous step at a point, L_0, on the abscissa where, of course, $L_0>L$. The onset length is L_0. However, by virtue of the recently verified finding that there is a zero point of areal mass density of superfluid,[4,13,14] the two lengths L_0 and L are related. The simple geometry of Fig. 1 implies that

$$L=L_0-\sigma_s{}^0/\rho_s(\beta,T). \tag{4}$$

The quantity $\sigma_s{}^0$ is the zero-point areal mass density mentioned. It has a temperature variation —linear in T—and no ^{3}He concentration dependence.[4] Hence the two lengths differ considerably in temperature dependence. The surface excitations are seen directly through L but not through L_0 except via Eq. (4).

The ^{3}He acts as a spatial probe within the film. The concentration of ^{3}He is a strong function of position. Almost pure ^{4}He characterizes the substrate side of the film while the ^{3}He concentrates on the free surface side.[15-17] We find the excitation gap energy to be relatively independent of the ^{3}He in the film. The implication is that the substrate, ^{4}He rich, side of the film is the one whose excitations are seen. This supports our earlier finding[2] and that of Thomlinson, Tarvin, and Passell.[3]

In view of Fig. 1 it is quite natural to take $L-H$ to be the "healing length," l. The difference $L-H$ measures the extent to which, in helium able to support it, superfluidity disappears before the helium itself does. So defined, the healing length has a particular physical basis; l directly measures the contribution from the bounding (solid) surfaces of the helium to the spectrum of normal-fluid excitations.

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Vortices and Superfluid Decay in ^{4}He Films

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All previous theoretical predictions of the decay of superflow in thin ^{4}He films are in conflict with the recent data obtained by Ekholm and Hallock. For the first time an analysis is presented of two effects: vortex creation and annihilation at the edges of a film of finite width, and the effect of vortex density relaxation. The latter result is used to discuss the above-mentioned discrepancy between theory and experiment.

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Recent experiments[1] on the decay of superflow in very thin ^{4}He films have indicated significant deviations, in my opinion, from all the current predictions. If one denotes by d the film thickness in units of layers, then for $d > 8$, observation shows a slow, logarithmic decay similar to that predicted by the older Langer-Reppy *Ansatz*,[2] and which is at best only qualitatively approximated by a more careful calculation based upon vortex-pair dissociation theory for an infinite film.[3] For $d < 8$, a much stronger initial decay rate is observed, which is approximated by a much slower, roughly t^{-1} decay at long times. The current theory based upon an unbounded film[3] can fit the latter thin-film results (films with $d < 8$) only by assuming unrealistically small values for the dimensionless vortex coupling parameter λ.

In order to state carefully the discrepancy referred to above, I begin with the "Langer-Reppy *Ansatz*" as corrected by Ambegaokar *et al.*,[3] and I consider a semi-infinite strip where the superfluid flows with speed U_s along the negative x axis, and is bounded by parallel film edges at $y = 0$ and $y = L$. Then the decay of superflow is described by

$$dU_s/dt = -|\kappa| j_y, \tag{1}$$

where j_y is the average current density for vortices of both sign of circulation ($\kappa = \pm h/m$) created near the film edge at $y = 0$, or in the bulk, and which annihilate at the film boundary at $y = L$. In the limit of an infinitely wide film,[3,4] $j_y = 2\pi D\lambda n U_s$, where D is the vortex diffusion coefficient, $\lambda = \sigma\kappa^2(2\pi kT)^{-1}$ is the dimensionless vortex coupling parameter [σ is the two-dimensional (2D) super-