Surface normal component of a superfluid liquid

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The surface density of the normal component of a superfluid liquid, which characterizes the superfluid flow along the surface of a solid, is governed by the statistical properties of the surface roughness features. The effect of the roughness features of the substrate surface on the velocity of third sound in helium films is found. Experimental data are discussed.

The surface normal component of a superfluid liquid has been studied previously[1] in connection with the properties of a free surface of a liquid. In that case the surface normal component arises because of elementary surface excitations (ripplons), and its density is expressed in terms of the energy spectrum of the excitations by a formula which is a 2D analog of the Landau formula. In the present letter a surface normal component is introduced as a macroscopic characteristic of a liquid–solid interface. The density of the surface normal component is determined by statistical properties of the random static roughness features of the solid surface. These roughness features give rise to irregularities in the uniform flow of the superfluid component near the surface. As a result, the average mass flux is reduced. Correspondingly, in a macroscopic description, an additional density of the normal component, at rest with respect to the walls, arises. A characteristic property of this surface normal component is that it is finite at absolute zero; this circumstance plays an important role at low temperatures.

The surface normal component is manifested in the propagation of third-sound waves in superfluid films. The magnitude of the surface normal component determines the effect of the substrate roughness features on the behavior of the third-sound velocity as a function of the film thickness, as we will see below. It is interesting to note that the contribution of the surface roughness features to the density of the normal component is determined by the same correlation function of the roughness features which determines (if, of course, the solid is a metal) the reflection coefficient of this surface for electrons incident from the side of the metal (Ref. 2, for example). The superfluid properties of a film are directly related to the electronic properties of the metal substrate.

1. We write as $z=\zeta(\boldsymbol{\rho})$, $\boldsymbol{\rho}=(x,y)$ the equation of the surface which separates the solid $(z<\zeta)$ from the superfluid liquid $(z>\zeta)$. The statistical properties of random roughness features of the surface can be characterized by specifying a correlation function

$$\langle\zeta(\boldsymbol{\rho})\zeta(\boldsymbol{\rho}')\rangle=\xi^2 w(\boldsymbol{\rho}-\boldsymbol{\rho}'), \tag{1}$$

where $\xi^2\equiv\langle\zeta^2\rangle$ is the mean square amplitude of the roughness, and $w(0)=1$. We assume that the surface is planar on the average, so we have $\langle\zeta(\boldsymbol{\rho}\ )\rangle\ =0$.

Introducing Fourier harmonics of the function $\zeta(\boldsymbol{\rho}\ )$ by means of

$$\zeta(\boldsymbol{\rho})=\int\frac{d^2k}{(2\pi)^2}\zeta_{\mathbf{k}}\,e^{i\mathbf{k}\boldsymbol{\rho}},$$

and following the corresponding procedure for $w(\boldsymbol{\rho})$, we find

$$\langle\zeta_{\mathbf{k}}\zeta_{\mathbf{k}'}\rangle=(2\pi)^2\delta(\mathbf{k}+\mathbf{k}')\xi^2 w_{\mathbf{k}}, \tag{2}$$

where

$$\int\frac{d^2k}{(2\pi)^2}w_{\mathbf{k}}=1. \tag{3}$$

The quantities w_{k} are positive, since by virtue of (2) we have $\langle|\zeta_{\mathbf{k}}|^2\rangle = (2\pi\xi)^2\delta(0)w_{\mathbf{k}} = S\xi^2 w_{\mathrm{k}}$, where S is the total area of the surface. These quantities represent the distribution of roughness features with respect to values of the 2D wave vector **k**. Equation (3) serves as a normalization condition on the probabilities.

We assume that the typical values of the roughness wave vector satisfy the condition $k\xi\ll 1$, and we consider a perturbation introduced by the roughness in a uniform flow of a superfluid liquid at a velocity $\mathbf{v}_s^{(0)}$. This velocity is directed parallel to the average surface of the solid, $z=0$. At low velocities, the superfluid component can be treated as incompressible, and the potential of the superfluid velocity satisfies the Laplace equation

$$\Delta\varphi=0,\quad v_s=\nabla\varphi. \tag{4}$$

The amplitude of the roughness, $\zeta(\boldsymbol{\rho})$, is also small, as we assumed above. We therefore expand all quantities in power series in ζ, using the notation $\varphi^{(1)}$, $\varphi^{(2)}$, ... and $\mathbf{v}_s^{(1)}$, $\mathbf{v}_s^{(2)}$, ... for the terms in the expansion of the potential and the velocity which are proportional to ζ, ζ^2, ..., respectively.

A general solution of Eq. (4), which satisfies the condition that $\varphi^{(1)}$ be finite at infinity, $z=\infty$, is

$$\varphi^{(1)}(\boldsymbol{\rho},z)=\int\frac{d^2k}{(2k)^2}\,a_{\mathrm{k}}\,e^{i\mathbf{k}\boldsymbol{\rho}-kz}. \tag{5}$$

The quantities a_{k} are found from the relation

$$v_{sz}^{(1)}=-\mathbf{v}_s^{(0)}\,\frac{\partial\zeta}{\partial\boldsymbol{\rho}}$$

at $z=0$. This equation follows from the condition that the velocity component normal to the solid surface must vanish at this surface. We thus have

$$a_{\mathbf{k}}=-i\frac{\mathbf{k}\cdot v_s^{(0)}}{k}\zeta_{\mathbf{k}}. \tag{6}$$

Let us consider the variation p_α ($\alpha = x, y$) caused in the total mass flux of the liquid by the surface roughness:

$$p_\alpha = \int_{\zeta(\rho)}^{\infty} dz (j_\alpha - \rho_s v_{s\alpha}^{(0)}), \tag{7}$$

where $\mathbf{j} = \rho_s \mathbf{v}_s$ is the vector mass flux density, and ρ_s is the mass density of the superfluid component. We wish to calculate the mean value $\langle p_\alpha \rangle$. Since the mean value of any quantity which is linear in ζ is zero, we find from (7)

$$\langle p_\alpha \rangle = -p_s \left\langle \zeta(\rho) \int \frac{d^2k}{(2\pi)^2} a_{\mathbf{k}} e^{i\mathbf{k}\rho} i k_\alpha \right\rangle + \rho_s \int_0^{\infty} dz \langle v_{s\alpha}^{(2)} \rangle. \tag{8}$$

The mean value $\langle v_{sz}^{(2)} \rangle$ is independent of $\boldsymbol{\rho}$ simply because the average here must be understood as an average over $\boldsymbol{\rho}$. By virtue of the condition curl $\mathbf{v}_s^{(2)} = 0$, the mean values $\langle v_{s\alpha}^{(2)} \rangle$ in (8) are independent of z. They are thus simply constants, which must be incorporated in a renormalization of $v_{s\alpha}^{(0)}$. The second term in (8) should therefore be discarded, and we should understand $v_{v\alpha}^{(0)}$ as the value of the velocity $v_{s\alpha}$ in the limit $z \to \infty$. A direct calculation of $v_{s\alpha}^{(2)}$ through a power-series expansion in ζ leads to divergent expressions.

The first term in (8) can be evaluated easily by substituting in a Fourier expansion for $\zeta(\boldsymbol{\rho})$ and by taking the average in accordance with (2). As a result, we find

$$\langle p_\alpha \rangle = -\nu_{\alpha\beta}^{(n)} v_{s\beta}^{(0)}, \tag{9}$$

where the quantities

$$\nu_{\alpha\beta}^{(n)} = \rho_s \xi^2 \left\langle \frac{k_\alpha k_\beta}{k} \right\rangle \tag{10}$$

have the meaning of a 2D tensor of the unknown density of the surface normal component. The averaging symbols, when applied to the functions f($\mathbf{k}$) of the 2D wave vector in (10) and below, mean an average over distribution (3):

$$\langle f(\mathbf{k}) \rangle = \int \frac{d^2k}{(2\pi)^2} f(\mathbf{k}) w_{\mathbf{k}}. \tag{11}$$

In order of magnitude we have $\nu^{(n)} \sim \rho_s \xi^2 k \sim \rho_s \xi^2 / l$, where $l \sim 1/k$ is a characteristic "wavelength" of the roughness. The typical thickness of the layer, in which the effective density of the superfluid component is substantially lower than that at infinity, $z \to \infty$, is ξ^2/l in order of magnitude. By virtue of the assumption $\xi \ll l$, made above, this thickness is much smaller than ξ in the range of applicability of these equations, but it may be much larger than the interatomic distance.

2. The equations derived here can be used to clarify the effect of substrate roughness on the propagation velocity of third-sound waves in superfluid films. The square of the third-sound velocity is given by an expression of the following type (Ref. 4, for example):

$$u^2 = \nu_{\alpha\beta}^{(s)} n_\alpha n_\beta F(T, d), \tag{12}$$

where $F(T, d)$ is a function of the film thickness d, the temperature T, and also the type of substrate; the quantity $\nu^{(s)}_{\alpha\beta}$ is the total "mass" of the superfluid component per unit area of the film; and the unit vector $\mathbf{n}$ runs along the propagation direction of the third-sound wave. Using the results of the preceding section of this letter, we can calculate $\nu^{(s)}_{\alpha\beta}$ for the case in which the film thickness is the largest length parameter of the problem: $d \gg l \gg \xi$. In this case we find the following relation by virtue of (7) and (9):

$$\nu^{(s)}_{\alpha\beta} = \rho_s d\delta_{\alpha\beta} - \nu^{(n)}_{\alpha\beta}. \tag{13}$$

Relation (13) is analogous to the empirical formula of Sholtz, McLean, and Rudnick,[3]

$$\nu^{(s)} = \rho_s (d - D), \tag{14}$$

which describes[4] the region of small film thicknesses, $d < 5$–6 atomic layers. We will discuss the relationship between (13) and the experimental data found by Smith and Hallock[4] for large values of d later on. Here we would simply point out that the constant D, which is always on the order of 2 or 3 atomic layers, is independent of the nature of the substrate, differing from $\nu^{(n)}$ in this regard. In particular, Smith and Hallock[4] demonstrated that D has identical values for helium films on smooth and etched silicon surfaces.

Let us consider two films, each of thickness d, under identical conditions. One of the films (r) is on a rough substrate, while the other (i) is on a flat substrate of the same material. According to (12) and (13), the ratio of the square third-sound velocities for these two films is

$$\frac{u_r^2}{u_i^2} = 1 - \frac{\nu^{(n)}}{\rho_s d}, \tag{15}$$

where $\nu^{(n)} = \nu^{(n)}_{\alpha\beta} n_\alpha n_\beta$. The second term in (15), which is a correction term under the condition $d \gg l$, is therefore inversely proportionally to d.

3. We now consider the opposite limit, in which the condition $d \ll l \sim 1/k$ holds. In this case the film thickness is a small parameter, and all quantities can be expanded in powers of the coordinate z, reckoned from the middle of the film. In a zeroth approximation, the velocity of the superfluid component is constant over the film thickness and is parallel to the surface of the film:

$$v_{s\alpha} = v^{(0)}_{s\alpha}, \quad v_{sz} = v^{(0)}_{s\alpha} \frac{\partial \zeta}{\partial \rho_\alpha}, \tag{16}$$

where $v^{(0)}_{s\alpha}$ are constants.

From the condition under which the problem is of a potential nature, $\partial v_{s\alpha}/\partial z = \partial v_{sz}/\partial \rho_\alpha$, we find

$$\frac{\partial v_{s\alpha}}{\partial z} = \frac{\partial}{\partial \rho_\alpha}\left(v^{(0)}_{s\beta} \frac{\partial \zeta}{\partial \rho_\beta} \right) = v^{(0)}_{s\beta} \frac{\partial^2 \zeta}{\partial \rho_\alpha \partial \rho_\beta}$$

and thus

$$v_{s\alpha}(z) = v_{s\alpha}^{(0)} + z\left(\mathbf{v}_s^{(0)}\frac{\partial}{\partial\boldsymbol{\rho}}\right)\frac{\partial\zeta}{\partial\rho_\alpha}. \quad (17)$$

We can evidently assume that values $z = z(\boldsymbol{\rho}) = \pm(d/2) + \zeta(\boldsymbol{\rho})$ correspond to the two boundaries of a film on a rough substrate with $l \gg d$. We can calculate the mean value of the total mass flux:

$$\langle J_\alpha\rangle = \rho_s\left\langle \int_{-\frac{d}{2}+\zeta(\rho)}^{\frac{d}{2}+\zeta(\rho)} dz v_{s\alpha}(z)\right\rangle$$

$$= \rho_s d v_{s\alpha}^{(0)} + \left\langle\left(\mathbf{v}_s^{(0)}\frac{\partial}{\partial\boldsymbol{\rho}}\right)\frac{\partial\zeta}{\partial\rho_\alpha}\int_{-\frac{d}{2}+\zeta(\rho)}^{\frac{d}{2}+\zeta(\rho)} z dz\right\rangle$$

$$= \rho_s d v_{s\alpha}^{(0)} + \rho_s d\left(\mathbf{v}_s^{(0)}\frac{\partial}{\partial\boldsymbol{\rho}}\right)\left\langle\frac{\partial\zeta}{\partial\rho_\alpha}\zeta\right\rangle.$$

Substituting in the Fourier expansion for $\zeta(\boldsymbol{\rho})$, and taking an average in accordance with (2), we find

$$\langle J_\alpha\rangle = \nu_{\alpha\beta}^{(s)} v_{s\beta}^{(0)}, \quad (18)$$

where $\nu_{\alpha\beta}^{(s)}$ is given by (13), as above, but in place of (10) we now have

$$\nu_{\alpha\beta}^{(n)} = \rho_s d\xi^2\langle k_\alpha k_\beta\rangle. \quad (19)$$

According to (19), the effective 2D density of the normal component in a film of thickness $d \ll l$ is, in order of magnitude, $\rho_s d(\xi^2/l^2)$.

The ratio of the squares of the third-sound velocities for a film on a rough substrate and for a film on a plane surface of a substrate of the same material, under the condition $d \ll l$, is

$$\frac{u_r^2}{u_i^2} = 1 - \xi^2\langle(\mathbf{kn})^2\rangle, \quad (20)$$

i.e., independent of the film thickness d.

4. Finally, we consider the more general case in which the surface has two types of roughness simultaneously, whose characteristic "wavelengths" are much larger than the film thickness in one case and much smaller in the other. The large-scale and small-scale roughness features in this case evidently contribute independently to $\nu_{\alpha\beta}^{(n)}$. As a result, we find

$$\frac{u_r^2}{u_i^2} = 1 - \xi_1^2\langle(\mathbf{k}\cdot\mathbf{n})^2\rangle_1 - \frac{\xi_2^2}{d}\left\langle\frac{(\mathbf{k}\cdot\mathbf{n})^2}{k}\right\rangle_2, \quad (21)$$

where ξ_1^2 and ξ_2^2 are the mean square amplitudes of the large-scale and small-scale roughness features, respectively, and the indices 1 and 2 mean an average over the roughness spectrum of the corresponding type. The behavior of the ratio of the

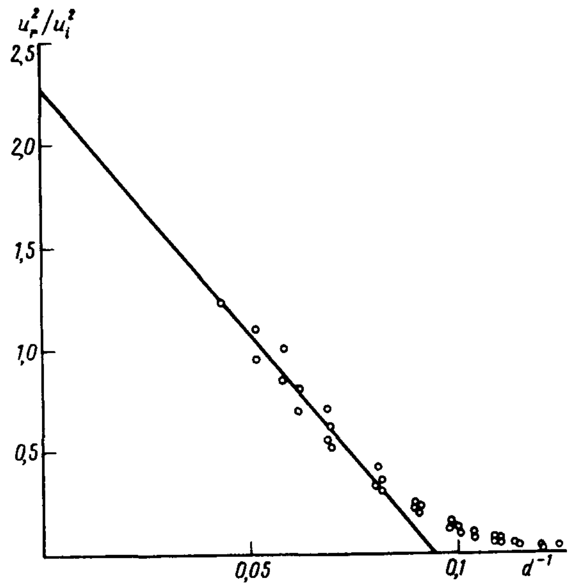

FIG. 1. Ratio of the squares of the third-sound velocities, u_r^2/u_i^2, for films of liquid ^{4}He on the surface of etched silicon (r) and on a smooth surface (i) at T = 1.42 K. The independent variable is the reciprocal of the film thickness, expressed in atomic layers (1 atomic layer = 3.6 Å; see Ref. 3). The straight line is a plot of the equation $u_r^2/u_i^2=2.28–24.2/d$.

squared third-sound velocities as a function of the film thickness is therefore governed by an expression of the type $A-B/d$. The constant B is determined by the small-scale irregularities. The large-scale irregularities cause A to differ from one.

Smith and Hallock[4] have simultaneously measured the third-sound velocities for films of liquid ^{4}He on rough and smooth surfaces of silicon. The behavior of u_r^2/u_i^2 as a function of the film thickness d was found to be complicated and nonmonotonic at small thicknesses. Figure 1 shows some data from Ref. 4, for thicknesses greater than 8 atomic layers. A linear dependence on the reciprocal thickness corresponding to Eq. (21) is observed at thicknesses greater than 12. This result agrees completely with the theory developed above, since that theory is macroscopic and applies at sufficiently large thicknesses. There is, on the other hand, a surprising result: The ratio of the squares of the velocities in the limit $d\to\infty$ is about 2.3, i.e., greater than one. Along the approach of this letter, the only way to explain this result would be to suggest that the i surface is actually less smooth than the etched surface r in terms of large-scale roughness features. We might add that the constant $B=24.2$ [in contrast with D in Eq. (14)] is considerably greater than the interatomic distance. This result agrees completely with the theory derived above, as we have already mentioned.

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