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Abstract. For a ^{4}He liquid surface, we derive an analytical form of the ripplon dispersion relation considering surface diffusiveness. The diffusiveness reduces the ripplon frequency for a ^{4}He liquid with an abrupt surface. Comparison of our dispersion curve with the recently measured spectrum gives the width of the diffusiveness and surface tension. Using the dispersion relation and applying the Atkins theory, we can explain well the measured surface tension in the temperature range from 0 K to $T_\lambda = 2.17$ K.

Surface vibrations of a ^{4}He liquid have been investigated in recent years. Among recent research, an important contribution has been made by Lauter *et al* [1]. They have measured the energy loss spectrum of inelastically scattered neutrons from ^{4}He thin liquid films on a graphite substrate. They have directly observed, for the first time, the ripplon dispersion relation at 0.65 K. The results show a lower dispersion curve than a simple dispersion curve $\omega_k = \sqrt{\sigma/\rho}k^{3/2}$ where σ is surface tension and ρ is the mass density. Edwards *et al* [2] have derived a softened dispersion relation including an effect of the curvature dependence of the surface tension. For a certain choice of two parameters, their dispersion curve, however, exhibits an anomalous curve resulting in a divergent group velocity of the ripplon. On the basis of a density functional theory, Dalfovo *et al* [3] have derived the ripplon dispersion curve which deviates from data observed by Lauter *et al* [1].

Concerning the temperature dependence of the surface tension $\sigma(T) = \sigma(0) + \delta\sigma(T)$, Atkins [4] has proposed for the first time that the ripplon is responsible for it. Assuming that the ripplon dispersion relation is $\omega_k = \sqrt{\sigma/\rho}k^{3/2}$, Atkins [4] has derived the $T^{7/3}$ dependence of $\delta\sigma(T)$ at sufficiently low temperatures. In the course of calculations, the integral over k has been carried out from 0 to infinity. At finite temperatures, however, this procedure is unrealistic. Because an infinite k means an infinitesimal wavelength, an excess number of ripplon modes is counted. Moreover, an expansion factor $T^{4/3}$—as a leading term of $T^{7/3}$ and following terms of $T^{11/3}$, $T^{15/3}$, . . .—results in slow convergence. Zinov'eva and Boldaev [5], Eckardt *et al* [6] and Iino *et al* [7] have measured $\delta\sigma(T)$. Results by Eckardt *et al* [6] and Iino *et al* [7] are consistent. Though several kinds of theory have been proposed to explain $\delta\sigma(T)$, none of them have obtained a quantitative $\delta\sigma(T)$ in a wide range of temperature less than $T_\lambda = 2.17$ K. Some researchers [8–10] have considered the effect of Bose–Einstein condensation that induces superfluidity. On the basis of a density functional method, Pricaupenko and Treiner [11] have derived the ripplon dispersion relation and $\delta\sigma(T)$. Using a variational method, Gernoth and Clark [12] also have derived these quantities. Those $\delta\sigma(T)$, however, begin to deviate from the measured data as T increases.

In addition to this problem, the value of the surface tension at $T = 0$ K is in controversy even now. Among measured data, 354.4 μJ m^{-2} measured by Iino *et al* [7] had been thought to be the most accurate. Recently, Roche *et al* [13] have measured the value as 375 μJ m^{-2} that is close to the value 378 μJ m^{-2} measured by Eckardt *et al* [6]. Here we clarify this uncertainty by calculating $\delta\sigma(T)$.

To investigate the validity of a proposed theory for analysing the ripplon, it is necessary to check three quantities: the dispersion relation, the temperature dependence of surface tension and the surface tension at $T = 0$ K. The purpose of the present report is, by introducing surface diffusiveness, to derive these quantities.

Since the ripplon is localized near the surface, the effect of surface diffusiveness on the ripplon frequency is important. Here we set the density distribution of a liquid film to be a type of Fermi–Dirac distribution:

$$\rho(\boldsymbol{r}_\parallel, z) = \frac{\rho_d}{1 + \exp\{[z - \eta(\boldsymbol{r}_\parallel)]/d\}} \tag{1}$$

where d is related to the thickness of surface diffusiveness, $\boldsymbol{r}_\parallel$ is a position vector parallel to the surface, z is a vertical position taken from the bottom plane and $\eta(\boldsymbol{r}_\parallel)$ represents a distance from the bottom plane to a point on the diffusive surface. We expand the displacement of $\eta(\boldsymbol{r}_\parallel)$ from an equilibrium height h as

$$\eta(\boldsymbol{r}_\parallel) = h + \sum_{\boldsymbol{k}} \alpha_{\boldsymbol{k}}^* \exp(i\boldsymbol{k} \cdot \boldsymbol{r}_\parallel) = h + \delta\eta \tag{2}$$

where coefficients $\alpha_{\boldsymbol{k}}^*$ represent normal coordinates. By expanding equation (1) around $d = 0$, we find

$$\rho(\boldsymbol{r}_\parallel, z) = \rho_d[\theta(\eta(\boldsymbol{r}_\parallel) - z) - (\pi^2/6)d^2\delta'(z - \eta(\boldsymbol{r}_\parallel)) + \cdots] \tag{3}$$

where $\theta(x)$ represents the step function, and a prime above a delta-function represents the derivative with respect to z. The first term denotes the constant density with an abrupt surface. To lowest order in $\delta\eta$ and first order in d^2, we find an expression for the density as

$$\rho(\boldsymbol{r}_\parallel, z) = \rho_d[\theta(h - z) - \varepsilon(\pi^2/6)d^2\delta'(z - h) + \cdots] \tag{4}$$

where ε means an expansion parameter. Hereafter, a dot above a variable represents the derivative with respect to time.

To define the diffusiveness d, we introduce the following density with $\delta\eta = 0$:

$$\rho_d(z) = \frac{\rho_d}{1 + \exp[(z - h)/d]}. \tag{5}$$

By setting the thickness of the diffusiveness D to be a distance between two sites at which $\rho_d = 0.9\rho_d$ and $0.1\rho_d$, respectively, we find $d = D/4.4$. The $\rho(\boldsymbol{r}_\parallel, z)$ of equation (4) should not be taken literally as the real density. It behaves as a weighting function with which a physical quantity is integrated over a certain region. Such an integrated quantity is non-singular. A typical example of this kind is the internal energy of free electrons at finite temperatures. It has an additional term proportional to $(k_B T)^2$ [14].

At first we assume that the liquid is incompressible. Then the equation of continuity becomes $\nabla \cdot \boldsymbol{v} = 0$. We assume further that the liquid is irrotational. The flow velocity, therefore, is written as the gradient of a velocity potential χ: $\boldsymbol{v} = \nabla\chi$. Since the velocity potential χ satisfies the Laplace equation $\nabla^2\chi = 0$, the solution can be written as

$$\chi(\boldsymbol{r}, t) = \sum_{\boldsymbol{k}} c_{\boldsymbol{k}}^*(t) \exp(i\boldsymbol{k} \cdot \boldsymbol{r}_\parallel)[\exp(kz) + \exp(-kz)]\theta(z)\theta(h - z). \tag{6}$$

Because the vertical velocities $\partial \chi / \partial z$ and $\dot{\eta}(r_{\parallel})$ should be continuous at the surface, we find $c_k = [2k \sinh(kh)]^{-1} \dot{\alpha}_k$.

We define the kinetic energy for the ripplon as

$$K = \frac{1}{2} \int \rho(r_{\parallel}, z) |\nabla \chi(r, t)|^2 \, dr \equiv K_0 + \varepsilon K_1 + O(\varepsilon^2). \tag{7}$$

By using $\rho(r_{\parallel}, z)$ of equation (4) and $\chi(r, t)$ of equation (6), we obtain

$$K_0 = \rho_d A_0 \sum_k k \sinh(2kh) |c_k|^2 \tag{8}$$

where A_0 represents the surface area of the liquid film in the equilibrium state. In the same way, we find K_1 as follows.

$$K_1 = (2\pi^2/3) d^2 \rho_d A_0 \sum_k k^3 \sinh(2kh) |c_k|^2. \tag{9}$$

These calculated results lead us to a final expression for the kinetic energy:

$$K = K_0 + \varepsilon K_1 = \frac{1}{2} \rho_d A_0 \sum_k \frac{[1 + \varepsilon(2\pi^2/3) d^2 k^2]}{k \tanh(kh)} |\dot{\alpha}_k|^2 \tag{10}$$

and the canonical momentum π_k is defined as

$$\pi_k = \frac{\partial K}{\partial \dot{\alpha}_k} = \rho_d A_0 \frac{[1 + \varepsilon(2\pi^2/3) d^2 k^2]}{k \tanh(kh)} \dot{\alpha}_k^*. \tag{11}$$

The coefficient $\rho_d A_0 [1 + \varepsilon(2\pi^2/3) d^2 k^2]/[k \tanh(kh)]$ in the right-hand side represents an effective mass associated with the liquid flow. The mass becomes large with the increase in the diffusiveness.

Neglecting the effect of gravity and considering the enlarged surface area during surface vibrations, we find the potential energy as

$$V = \frac{1}{2} \sigma(T) \int dr_{\parallel} |\nabla \eta|^2 = \frac{1}{2} \sigma(T) A_0 \sum_k k^2 |\alpha_k|^2 \tag{12}$$

where $\sigma(T)$ represents the surface tension at a temperature T.

From the kinetic energy of equation (10) and the potential energy of equation (12) we arrive at a Hamiltonian for the ripplon as follows.

$$H = \frac{1}{2} \sum_k \left\{ \frac{k \tanh(kh)}{\rho_d A_0 [1 + (2\pi^2/3) d^2 k^2]} |\pi_k|^2 + \sigma(T) A_0 k^2 |\alpha_k|^2 \right\} \tag{13}$$

where the expansion parameter ε is put equal to 1. Thus we obtain an analytical expression for the frequency as

$$_s\omega_{kd}^2 = \frac{\sigma(T)}{\rho_d} \frac{k^3}{[1 + (2\pi^2/3) d^2 k^2]} \tanh(kh). \tag{14}$$

If $d = 0$ and $kh \gg 1$, we reproduce $\omega_k = \sqrt{\sigma/\rho} k^{3/2}$. Hereafter, we label $_s\omega_{kd}$ as ω.

In the experiments carried out by Lauter *et al* [1], the average distance between neighbouring layers is about 3.3 Å, and h of the liquid films are larger than 5.0 Å. Thus, in the range $k > 0.4$ Å^{-1}, a term $\tanh(kh)$ is set to be 1 in equation (14). In this k-range the velocity field v is localized near the surface, and we can regard the ripplon as that of a semi-infinite liquid having the mass density ρ_d.

To determine the diffusiveness D, we transform equation (14) into

$$\frac{k}{\omega^2} = \frac{\rho_d}{\sigma(T)} \left[\frac{1}{k^2} + \frac{2\pi^2}{3} \left(\frac{D}{4.4} \right)^2 \right] \tag{15}$$
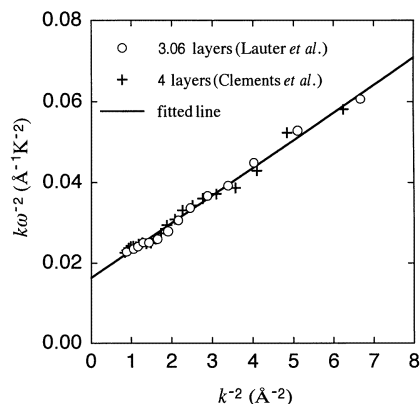
Figure 1. Scaled data $k\omega^{-2}$ as a function of k^{-2}. A thick line represents the fitted line for measured data of both 3.06 layer and 4 layer films.

where $\tanh(kh) = 1$. Figure 1 shows this dispersion relation in which the measured data with $k > 1.06$ Å^{-1} are neglected. The reason for this is given below concerning the maximum wave number of ripplons. Open circles refer to the data points for 3.06 layers taken from Lauter *et al* [1], and crosses refer to the middle data points for 4 layers measured by Clements *et al* [15]. In contrast to other films exhibiting large fluctuations, both data of 3.06 and 4 layers clearly show a linear dependence on the abscissa k^{-2}. This linearity justifies our dispersion relation of equation (14). Since two values 3.06 and 4 are very close to integers, those films are thought to be almost saturated. A thick line shows a fitted line—by the least squares method—to the measured data of both 3.06 layer and 4 layer films: $k\omega^{-2} = 0.006\,82(k^{-2} + 2.39)$. From the value 2.39, we have $D = 2.65$ Å. Clements *et al* [16] have calculated the density profiles of liquid films and obtained $D = 2.7$ Å that agrees well with our D. From the coefficient 0.006 82, we have $\sigma(0.65\text{ K}) = 365\ \mu\text{J m}^{-2}$ by taking $\rho_d = \rho_B$ for simplicity. It should be emphasized that these values can be derived only from the measured data through the analytical expression of equation (15). Figure 2 shows the dispersion relation of equation (14) with $\tanh(kh) = 1$. Our dispersion curve traces well the data of 3.06 layer and 4 layer films. In this way, the dispersion relation of equation (14) applies well to a liquid film having an almost saturated top layer.

According to the Atkins theory, the surface free energy is written as

$$F = A_0[\sigma(0) + \delta\sigma(T)]. \tag{16}$$

The first term shows surface tension at 0 K. The second term is defined as

$$\delta\sigma(T) = \frac{k_{\rm B}T}{2\pi} \int_0^{k_{\rm m}} \ln[1 - \exp(-\hbar\omega/k_{\rm B}T)]k\,\mathrm{d}k. \tag{17}$$

Though the integration of equation (17) has been often carried out from 0 to ∞, it gives no quantitative $\delta\sigma(T)$ except at sufficiently low temperatures. Here we set the $k_{\rm m}$ to be the maximum wavenumber associated with the minimum wavelength defined by $k_{\rm m} = \pi/a$ where a is an interatomic distance. The $k_{\rm m}$ corresponds to the zone boundary of a system having a symmetry of translation a. Here we put $a = 2.96$ Å derived for an interatomic potential by Aziz *et al* [17]. Karlos *et al* [18] has shown, by a Monte Carlo method, that the pair correlation function $g(r)$ of a superfluid ^{4}He is equal to 1 at $r = a$.

Since the frequency ω includes surface tension $\sigma(0) + \delta\sigma(T)$, we need to calculate, with $D = 2.65$ Å, $\delta\sigma(T)$ self-consistently. Figure 3 shows $\delta\sigma(T)$ as a function of $T^{7/3}$.
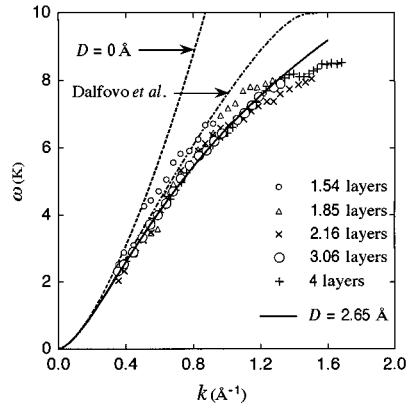
Figure 2. Dispersion relation of the ripplon. Symbols show the data measured by Lauter *et al* [1] and Clements *et al* [15].
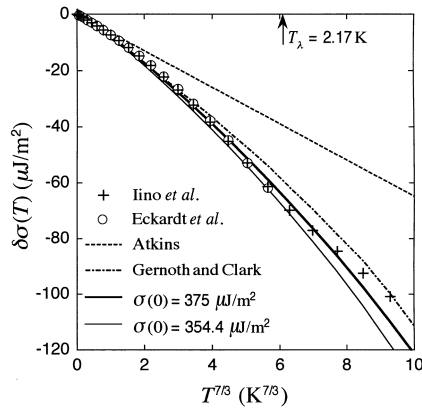

Figure 3. Temperature dependence of surface tension. Solid curves show the self-consistent solutions for equation (17).

In the range 0 K < T < 1.9 K our $\delta\sigma(T)$ with $\sigma(0) = 375$ μJ m^{-2} traces well the data measured by Eckardt *et al* [6] and Iino *et al* [7]. Our result with $\sigma(0) = 354.4$ μJ m^{-2} slightly deviates from the measured data in the range less than 1.9 K. Thus we support $\sigma(0) = 375$ μJ m^{-2} and the $\delta\sigma(T)$ measured by Eckardt *et al* [6] and Iino *et al* [7]. A dashed line shows $\delta\sigma(T)$ calculated by Atkins [4] with $\omega_k = \sqrt{\sigma/\rho}k^{3/2}$. The difference from the measured data is great except at sufficiently low temperatures. A dash–dotted curve shows the result calculated by Gernoth and Clark [12] and it underestimates $-\delta\sigma(T)$ at high temperatures.

Lauter *et al* [1] have measured the ripplon dispersion relation at 0.65 K. From our calculated result of $\delta\sigma(T)$, we obtain $\delta\sigma(0.65$ K$) = 372$ μJ m^{-2}. The data measured by Iino *et al* [7] read $\sigma(0.65$ K$) = 352$ μJ m^{-2}. As is shown in the paragraph below equation (15), the surface tension estimated from the measured data is $\sigma(0.65$ K$) = 365$ μJ m^{-2}. The value 372 μJ m^{-2} is closer to this estimated value.

In the above treatment, a ^{4}He liquid is assumed to be incompressible. Extension to the case of a compressible liquid is straightforward. In that case we take account of the coupling between ripplons and phonons located near the surface. To derive a Hamiltonian

of the ripplon, we consider that the restoring force is composed of two factors: surface tension and compressibility of the liquid. For a semi-infinite liquid with an abrupt surface, we have the frequency $_c\omega_{k0}$ satisfying $_c\omega_{k0}^2 = (\sigma/\rho_d)k^2\nu_0$, where $\nu_0 = \sqrt{k^2 - (_c\omega_{k0}^2/u^2)}$ and u is the speed of sound. This expression is the same as that derived by García-Moliner and Flores [19]. The $_c\omega_{k0}^2$ can be expanded as

$$_c\omega_{k0}^2 = \frac{\sigma(T)}{\rho_d}k^3 - \frac{1}{2u^2}\left[\frac{\sigma(T)}{\rho_d}\right]^2 k^4 + \text{O}(k^5) \tag{18}$$

and $_c\omega_{k0}^2$ is softened because of the coupling with phonons. In the case of a diffusive surface, we find the frequency $_c\omega_{kd}$ defined by

$$_c\omega_{kd}^2 = \frac{u^2(k^2 - \nu_d^2)^2}{(k^2 + \nu_d^2)[1 + (2\pi^2/3)d^2\nu_d^2]} + \frac{2\sigma(T)k^2\nu_d^3}{\rho_d(k^2 + \nu_d^2)[1 + (2\pi^2/3)d^2\nu_d^2]} \tag{19}$$

where $\nu_d = \sqrt{k^2 - (_c\omega_{kd}^2/u^2)}$. The $_c\omega_{kd}^2$ can be expanded as

$$_c\omega_{kd}^2 = {}_s\omega_{kd}^2 \left\{1 - \frac{_s\omega_{kd}^2}{2u^2k^2[1 + (2\pi^2/3)d^2k^2]} + \cdots\right\}. \tag{20}$$

Comparing with $_c\omega_{k0}$ of an abrupt surface, the softening induced by the coupling with phonons is suppressed mainly by a term $[1 + (2\pi^2/3)d^2k^2]$. Decrease in the frequency from $_s\omega_{kd}$ amounts to a few per cent of $_s\omega_{kd}$, which does not yield an appreciable change in $\delta\sigma(T)$ obtained with $_s\omega_{kd}$. In summary, the coupling with phonons has little contribution to the ripplon frequency. We can extend the present treatment to a compressible liquid film. Details of deriving these quantities will be reported elsewhere.

Our treatment is based on classical hydrodynamics and our results show that the temperature dependence of surface tension is independent of the superfluidity of ^{4}He; all ^{4}He atoms, irrespective of whether they are normal liquid or superfluid, contribute to surface tension. Thus, the quantum effect is thought to be included implicitly in the surface diffusiveness.

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