Liquid-Solid ^{4}He Interfacial Tension: Temperature Variation near the Superfluid Transition

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Accurate measurements of the interfacial tension between solid and superfluid ^{4}He show for the first time that it decreases between 1.2 and 1.76 K. The magnitude of this effect is compared with various recent theoretical predictions. A significant contribution is found to come from the entropy of Rayleigh waves. We also discuss the effect of the progressive disappearance of the condensate along the melting curve, near the λ transition.

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The interfacial tension $\tilde{\alpha}$ between solid ^{4}He and superfluid ^{4}He has recently been measured by means of various techniques.[1–3] We present here more accurate measurements near the λ transition on the melting curve, where the liquid in contact with the solid becomes normal. These measurements show, for the first time, a definite temperature variation of $\tilde{\alpha}$. After having described our experimental method, we compare the observed variation with various recent predictions. Indeed, Andreev and Parshin[4] predicted that the existence of melting-freezing waves should lead to a $T^{7/3}$ variation of the interfacial free energy. Uwaha and Baym[5] reexamined this prediction, including elasticity effects and the existence of Rayleigh waves. Melting-freezing waves are probably highly damped in the temperature range of the experiment, but Rayleigh waves are found to give a significant contribution. The effect of bulk excitations[6,7] is found to be negligible. Another contribution comes from the distortion of the superfluid order parameter near the interface. According to various authors,[8] the energy associated with this distortion can be calculated. Recently, Campbell[8] extracted a very reasonable value for the condensate fraction n_0 at saturated vapor pressure from surface tension measurements.[6,7] These latest developments stimulated this work since a lot of interest is still devoted to the theoretical[9] and experimental[10] determination of n_0, in particular at high pressure. The surface energy associated with the condensate has to vary near the λ transition, but the observed variation of $\tilde{\alpha}$ is 3 times greater than what is expected from Campbell's estimate. A final suggestion is made about the dependence of $\tilde{\alpha}$ on the difference in density $\Delta\rho$ between the liquid and the solid.

In order to measure the liquid-solid interfacial tension near the λ transition, none of the very accurate methods used for the liquid-vapor interface[6,7] can be used. Indeed, the angle of contact of our interface[1,2] with solid walls is neither 0° nor 180°, and thus the capillary rise or depression method measures a different quantity.[1] The capillary waves (or melting-freezing waves in this case) are weakly damped only at low temperature[3] and the measurement of their dispersion relation cannot be used above 1 K as a precise measurement of $\tilde{\alpha}$. Thus, in an experiment similar to those described by Balibar, Edwards, and Laroche[1] or Wolf, Balibar, and Gallet,[11] we measured the average curvature of a meniscus bent under the effect of a known pressure difference. A small box is suspended in the pressure cell of our cryostat and observed through windows. It has transparent walls and a 1.00-mm hole in its bottom plate. By growing a crystal from the outside a bent meniscus can be anchored on the hole. Within a few seconds, it usually reaches an equilibrium shape which depends on $\tilde{\alpha}$ and the height difference H between the interfaces inside and outside of the box. With the aid of a cathetometer, we measured the height h of the meniscus above the hole as a function of H at various temperatures.

As a first approximation, the hole radius R being smaller than the capillary length, one can neglect the effect of gravity on the shape of the meniscus. The interfacial tension $\tilde{\alpha}$ is then given by

$$\tilde{\alpha}/\Delta\rho = (h^2+R^2)gH/4h. \tag{1}$$

In this simple formula, possible anisotropy effects have been neglected as well. For bcc crystals, this was justified since all the meniscii looked quasicircular and since, from one crystal to another, no significant variation of $\tilde{\alpha}$ was found. However, the results presented here have to be considered as average values. For hcp crystals, where $\tilde{\alpha}$ is known to be anisotropic at low temperature,[3] we consider our results as only typical of a given orientation. This orientation was such that no facets appeared in the profile of the meniscus which looked quasicircular as for bcc crystals.

In fact, the effect of gravity is not fully negligible.

After a numerical calculation of the exact shape of the meniscus, more exact values of $\tilde{\alpha}$ were obtained, which are 5% to 10% lower than those we obtained when using Eq. (1). We got final values by a weighted average of our measurements at each temperature. With account taken of a random error of ± 0.02 mm in the height measurements, the final accuracy for $\tilde{\alpha}$ is found to be about 4%.

Our results are presented in Fig. 1. They are consistent with previous measurements which were less accurate and could not show the temperature variation found here. The data are limited to the range 1.2 to 1.76 K. Indeed, 1.2 K is close to the temperature of the first roughening transition, where c facets appear and change the meniscus shape.[11] Above 1.76 K, the liquid is normal[12] and our technique appeared impossible to use: First, the growth of crystals is dendritic under usual conditions; second, the temperature in the cell is no longer homogeneous. The results show a discontinuity of $\tilde{\alpha}$ at the hcp-bcc transition. In the two small adjacent domains of temperature, we observe a decrease which we describe by an average slope since the exact law of variation cannot be extracted. These slopes are respectively $(-27 \pm 13) \times 10^{-3}$ and $(-72 \pm 15) \times 10^{-3}$ erg cm^{-2} K^{-1} in the hcp (1.2–1.46 K) and bcc (1.46–1.76 K) phases.

Let us first examine Campbell's ideas. According to Ref. 8, the distortion of the condensate fraction n_0 near an interface where it vanishes gives a contribution

$$\alpha_c(T) = (\sqrt{2}\hbar^2/3m^2\xi)\rho_l n_0(T)$$

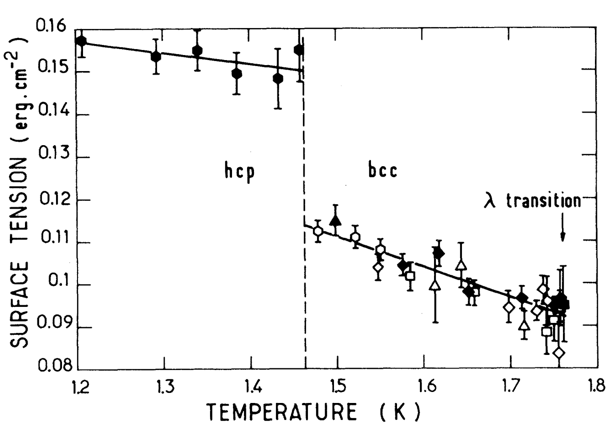

FIG. 1. Temperature variation of the interfacial tension. Different symbols correspond to different crystals. The discontinuity at 1.46 K is due to the hcp-bcc transition. The two solid lines correspond to least-squares fits. Their respective slopes are $-(27 \pm 13) \times 10^{-3}$ erg cm^{-2} K^{-1} (hcp phase) and $-(72 \pm 15) \times 10^{-3}$ erg cm^{-2} K^{-1} (bcc phase).

to its energy (ξ is the coherence length, m the ^{4}He atomic mass, and ρ_l the liquid density). After a careful analysis of experimental data concerning the liquid-gas interface,[6] Campbell found a value of 2.3×10^{-2} erg cm^{-2} for $\alpha_c(0)$ and 0.13 for $n_0(0)$, a result in agreement with other calculations[9] or measurements[10] of the condensate fraction at saturated vapor pressure. Along the melting curve, we estimate an upper bound of this effect by supposing first that the relative variation of n_0 with the reduced temperature T/T_λ is universal, and second that when the pressure rises up to ~ 26 bars, n_0 is approximately divided by two[9] and ξ does not increase significantly.[13] These various hypotheses lead to a maximal variation of 3.7×10^{-3} erg cm^{-2} between 1.2 and 1.46 K and 8.1×10^{-3} erg cm^{-2} between 1.4 and 1.76 K. This is only about one-third of the observed variation. It means that the effect calculated by Campbell might explain part of our experimental result, but that another phenomenon has to be taken into account. An even more precise measurement of $\tilde{\alpha}$, below *and* above T_λ, showing for example a change in slope at T_λ would have been very helpful to test Campbell's ideas. Unfortunately, this seems difficult in the present stage of our experimental technique.

Atkins and Narahara[14] showed that the ripplons play a major role in the free superfluid surface energy. According to Andreev and Parshin,[4] one could think that melting-freezing waves have a similar importance here. Let us first remark that the dispersion relation for melting-freezing waves contains a factor $(\rho_l/\Delta\rho)^2$ which makes them much more energetic than the usual ripplons of the free superfluid surface. However, as calculated by Uwaha and Baym,[5] elasticity effects limit their velocity to a fraction of the transverse sound velocity in the solid. This might give a significant contribution to the interface entropy at low temperature but we do not think that it is the case here. Indeed, if one extrapolates the results of Keshishev, Parshin, and Babkin,[3] one finds that these melting-freezing waves are highly damped above 1 K. This is due to the very small value of the growth coefficient at high temperature which was actually measured by Bodensohn, Leiderer, and Savignac.[15] Whatever the variation of their lifetime with frequency,[16] we do not think that these melting-freezing waves really exist between 1.2 and 1.76 K.

Let us now consider the Rayleigh waves. Uwaha and Baym[5] showed that when the growth coefficient goes to zero, the spectrum of surface excitations reduces to only one branch called "Rayleigh waves without melting." The associated free energy is

$$F_R = -[\zeta(3)/2\pi](k_B^3/\hbar^2 c_R^2)T^3,$$

where c_R is the velocity of this mode, which depends on the sound velocity in the solid and in the liquid. c_R is smaller than the transverse sound velocity c_t which is a rather anisotropic quantity in the bcc phase. Using Greywall's results[17] on the bcc crystal, we estimate c_R as 75 ms^{-1} in the $[1\bar{1}0]$ direction of the (110) plane and 235 ms^{-1} in the $[1\bar{1}0]$ direction of the (001) plane. In other directions, c_R lies between these two values. For example, it is 185 ms^{-1} in [001]-type directions. Then, we estimate F_R in various planes by averaging c_R^{-2}. For a (001) plane, we find $\langle c_R^{-2}\rangle = 0.214\times10^{-4}$ s^2 m^{-2}, and 0.56×10^{-4} $\text{s}^2\ \text{m}^{-2}$ for a (110) plane. Finally, we calculate the corresponding entropies at 1.6 K. Numerical values are, respectively, 7×10^{-3} and 19×10^{-3} erg cm^{-2} K^{-1} for (100) and (110) planes. The measured entropy corresponds to a further average over nearly all possible orientations of interfaces and the resulting anisotropy should be smaller than our error bars. In the case of the hcp crystal, where the transverse sound velocity is nearly isotropic, we find $c_R \simeq 190$ m s^{-1} and the entropy at 1.33 K is 6.7×10^{-3} cgs. Therefore, as a conclusion about Rayleigh waves, we think that their entropy amounts to about one-fourth of the experimental result. It is slightly smaller than the above estimate concerning the condensate.

Let us consider the influence of bulk phonons and rotons, which has been analyzed by Brouwer and Pathria[8] and Edwards and co-workers[6] in the case of the free superfluid surface. Bulk excitations were found to give a contribution to the surface free energy, whose sign depends on the boundary condition at the interface. For the liquid-gas interface which is free to move, this free energy is positive. For the liquid-solid interface which has a small mobility in our temperature range, this free energy should be negative. We neglect longitudinal phonons whose transmission coefficient is nearly 1.[18] Supposing that transverse phonons (with velocity c_ϕ) are not transmitted through the interface, we find that their maximum contribution is

$$F_\phi = -[\zeta(3)k_B^3/8\pi\hbar^2]\langle c_\phi^{-2}\rangle T^3.$$

Taking into account the two transverse polarizations, we have $+4.2\times10^{-3}$ erg cm^{-2} K^{-1} for the corresponding entropy at 1.6 K. This is significantly smaller than the Rayleigh-wave term, and can be neglected in a first approximation. The roton term was calculated by Brouwer and Pathria[8]; it is even smaller in our temperature range. In other words, the problem of bulk excitations is not found important for the interpretation of our experiment.

A last phenomenon might have to be considered. Namely, the difference in density $\Delta\rho$ between liquid and solid He varies along the melting curve. It decreases by $\sim 4\%$ from 1.2 to 1.46 K and by $\sim 30\%$ from 1.46 to 1.76 K. This decrease does not change significantly the sound velocities and the calculations above, but we expect the interfacial energy to depend on $\Delta\rho$. The magnitude of this effect might be obtained by applying density functional theories to the liquid-solid interface as was done for the free superfluid surface.[19] Such a calculation would be very helpful in order to decide which part of the experimental variation of $\tilde{\alpha}$ has to be attributed to the disappearance of the condensate.

To sum it up, we have presented here the first measurements of a temperature dependence of the liquid-solid ^{4}He interfacial tension. A significant part of this dependence is attributed to the entropy of Rayleigh waves. A larger part might arise from the condensate which disappears at the λ transition (1.76 K). Melting-freezing waves or bulk excitations are found unimportant in the temperature range of the experiment (1.2 to 1.76 K). In order to obtain a more complete understanding of the interfacial free energy, a calculation of its dependence on the density difference Δ_ρ between the liquid and the solid phase would be very useful.

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