

Study on Surface Tension of Fluid Helium Three

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Abstract As a standard property of helium-3, the surface tension is not only an important aspect to study the normal characteristics of this special cryogenic fluid, but also helpful to understand the Fermi quantum effects at low temperatures. After completion of studies on the thermodynamic and two typical transport properties (thermal conductivity and viscosity) of helium-3, all of the published experimental data of the surface tension of ^3He have been collected. Different measurement techniques are compared and analyzed. The peculiar behavior of ^3He surface tension, perhaps dominated by Fermi-Dirac quantum statistics, is analyzed and discussed at temperatures starting from zero to its critical point (3.3157 K). Based on a regular theoretical model for surface tension, a semi-empirical correlation is proposed for ^3He covering the whole temperature range. The surface tension extrapolated to zero temperature by this equation is $1.5579 \times 10^{-4} \text{ N} \cdot \text{m}^{-1}$. In the vicinity of the critical point, the equation could be smoothly switched to the known scaling law, which takes the critical index 1.289. Comparison for the surface-tension behavior is performed between ^3He and its isotope ^4He , which obeys Bose-Einstein statistics.

Keywords Helium-3 · Surface tension · Equation

1 Introduction

^3He , the second stable natural isotope of helium, has been dramatically applied in many fields, such as cryogenics and fundamental physics, because of its particular thermophysical properties. Since being first liquefied in 1948, many experimental and

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theoretical studies have been carried out internationally on the properties of this special fluid, especially at temperatures lower than 1 K. However, no compiled reference data for ^3He has been announced in a wide range of temperature and pressure, until the recent work by Chen and Huang [1–5] on raw data mining and equation-of-state development. Recently, the transport properties of ^3He are being studied, and encouraging progress has been achieved for thermal conductivity, viscosity, and surface tension. The surface tension properties will be introduced by this article, which is one of the important components of our overall research on ^3He properties.

2 Reference Data Sources and Analysis

Our reference data are all from a comprehensive literature survey. Experimentally, Esel'son and Bereznyak [6], Lovejoy [7], Zinov'eva [8,9], and Iino et al. [10,11] measured the surface tension of ^3He at temperatures from 0.3 K to the vicinity of the critical point, although their data differ from each other by up to 25 %. All of the studies, except for Iino et al., used a conventional capillary rise method. The main source of the experimental difficulties was the large heat resistance in a narrow channel with a width of 10 μm . Hence, a relatively large uncertainty was introduced into the results of the measurements.

Esel'son and Bereznyak [6] were the first to measure the surface tension of ^3He , in the temperature range from 1.32 K to 2.13 K with an uncertainty of 2.5 %. Perhaps because the article was written in Russian, it was seldom cited by later researchers. After that, Lovejoy [7] did a similar study and obtained data ranging from 1.08 K to 2.32 K, except in the range of (1.5–2.1) K where liquid ^3He had a strong tendency to bubble up in the capillary. Zinov'eva [8,9] initially measured the surface tension of ^3He from 0.93 K to 3.34 K by using three 2 cm long capillary tubes with diameters of 0.360 mm, 0.224 mm, and 0.188 mm. Then she extended the temperature from 1 K down to 0.35 K and obtained 13 points. After the rapid and large development of measurement technologies over 30 years, Iino et al. [10] achieved much more accurate measurements of ^3He in a wider range by using a completely new method. The surface tension was determined by measuring the surface wave velocity in a resonant system. The bubble-up problem no longer existed, and hence the accuracy was improved by 2–3 orders of magnitude. This is the most important criterion for us to give high weight to Iino et al.'s data during fitting the semi-empirical equation for the surface tension of ^3He . Compared to the early authors' results, measurements by Iino et al. roughly agree with them within their own reported uncertainties above 1.5 K. However, there are apparent differences below 1 K, especially approaching 0.32 K. Iino et al. [11] also measured the surface tension in the region very close to the critical point ($3 \times 10^{-4} < \tau = (T_c - T)/T_c < 2 \times 10^{-1}$), which is of great concern for determining the behavior of the equation near the critical temperature. The data point numbers, temperature range, uncertainties, etc. for all of the above references are list in Table 1.

3 Surface-Tension Equation for ^3He

Since the data from different authors introduced in Table 1 exhibit scatter and cover different temperature ranges but do not completely overlap, they are not enough

Table 1 Experimental data for ^3He surface tension

Year	Author	References	No. of points	T (K)	Uncertainty
1954	Esel'son and Bereznyak	[6]	18	1.324 to 2.128	2.5 %
1955	Lovejoy	[7]	27	1.08 to 2.32	3.0 %
1955	Zinov'eva	[8]	22	0.93 to 3.34	Max. 7 % (Ave. 5 %)
1956	Zinov'eva	[9]	13	0.35 to 1.0	5 %
1985	Iino et al.	[10]	62	0.32 to 3.02 $T < 1$ K	$< 1 \times 10^{-6} \text{ N} \cdot \text{m}^{-1}$ $1 \times 10^{-7} \text{ N} \cdot \text{m}^{-1}$
1986	Iino et al.	[11]	61	$3 \times 10^{-4} < (T_c - T)/T_c$ $< 2 \times 10^{-1}$ $T < 1$ K	$< 1 \times 10^{-6} \text{ N} \cdot \text{m}^{-1}$ $1 \times 10^{-7} \text{ N} \cdot \text{m}^{-1}$

to accurately describe the surface tension behavior of ^3He . It will be convenient to have an accurate mathematical equation with some physical significance to not only represent all the scattered measurements, but also extend to a wider temperature range.

As we know, in order to escape from a liquid into the saturated vapor, a molecule must have sufficient energy to overcome the barrier set up by the surface tension. Consequently the number of molecules per unit volume in the vapor must be a function of the amplitude of the barrier. In terms of measurable quantities, it amounts to say that the vapor pressure must be some function of the surface tension. Based on this idea, Pollara [12] proposed the following empirical relationship between the surface tension and the vapor pressure:

$$T \log_{10} p = -a\sigma \left(\frac{M}{\rho_l - \rho_g} \right)^{2/3} + b \quad (1)$$

where a and b are constants, p is the vapor pressure, σ is the surface tension, T is the temperature, M is the molar mass, and ρ_l and ρ_g are the densities of the saturated liquid and vapor, respectively. Equation 1 should work for most of the normal fluids in their dominant liquid–vapor coexistence region. If we plot graphs of $T \log_{10} p$ versus $\sigma[M/(\rho_l - \rho_g)]^{2/3}$, it will be found that the plots are remarkably straight, even near the critical points, thus, bearing out the fact that a and b are constants. Since $\sigma = 0$ at the critical point, the constant b is found to be $T_c \log_{10} p_c$, where $T_c = 3.3157$ K and $p_c = 114.6039$ Pa for ^3He .

However, ^3He is not such a “normal fluid”. It becomes superfluid at temperatures going down below 2.6 mK in place of the triple point of a normal fluid. On the other hand, the strong quantum effect of ^3He appearing at low temperatures dominantly forced the departure of its thermophysical properties from regular laws. For these two reasons, Eq. 1 could not be applied to ^3He directly or without modification. In fact, the ^4He isotope has similar problems as ^3He . Is it then possible to apply the relatively mature theoretical model of surface tension for ^4He to ^3He ? According to

the theory of Atkins [13], the zero-point energy of the atoms plays an important role in determining the surface energy of the liquid at very low temperatures. The variation in surface tension of ^4He at these temperatures has been thought to be produced predominantly by riplons, and, in addition, the behavior at higher temperatures may come from the reflection at the surface of internal excitations, which would first be phonons. The energy of the surface waves could be determined by following a procedure similar to that adopted in the Debye theory of solids. This treatment leads to the expression [13],

$$\sigma_0 - \sigma = bT^{7/3} \quad (2)$$

where σ_0 and σ are the surface tensions at 0 K and temperature T , respectively; b is a constant. Since the value of σ_0 for the fluid is fixed, the relationship could be simplified as $\sigma \propto T^{7/3}$.

However, the big difference between ^3He and ^4He quantum statistics should not be neglected. ^4He obeys Bose-Einstein (BE) statistics and transitions to superfluid below 2.172 K through BE condensation. But ^3He obeys Fermi-Dirac statistics and becomes superfluid only below 2.6 mK through a condensation similar to the BCS mechanism of a superconductor. The temperature dependence of the surface tension should be quite different between ^3He and ^4He , since the riplons on the latter might be damped, at least at very low temperatures, because of the substance's large bulk viscosity. The internal excitations in ^3He should also be different from ^4He . An experiment by Guo et al. [14] on ^3He floating on bulk liquid ^4He has shown a quadratic temperature dependence of the surface tension. They inferred that the surface tension of bulk liquid ^3He has T^2 dependence like the ^3He layer on ^4He , as expected for a Fermi system at low temperatures. On the other hand, an explanation of the temperature dependence of the surface tension of ^4He was attempted by Singh [15] who adopted the ideal-gas model and made use of an enumeration of the eigenfunctions over the surface area. If this treatment is applied to ^3He , the surface tension also becomes proportional to T^2 .

According to the plot containing all the experimental data listed in Table 1, the tendency of the scatter from Zinov'eva [9] is almost flat below 0.7 K. It may not correctly reflect the behavior of the surface tension of ^3He in this region, because the measurement uncertainty here is very large. In contrast, Iino et al. [10] have more reliable and comprehensive data below 0.9 K, which follows the T^2 proportion relationship. The distribution of the surface tension data versus temperature begins to depart from the T^2 line further and further above 1 K with an increase of temperature, until it joins a linear line at around 1.5 K. The possible reason is that liquid ^3He begins to lose Fermi degeneracy above 0.9 K until it totally disappears. It also can be found from the experimental data that the decrease in the density difference between liquid and gaseous ^3He from 0 K to 2 K is about 10 %, whereas the surface tension decreases by about 45 %. It is unlikely that this drastic change of the surface tension could be brought about only by the change of density. The kinetic energy of the surface waves is thought to be the other important cause of the observed decrease in the surface tension, except perhaps near the critical temperature.

Starting from Eq. 1 with the above considerations, a correlation is proposed for the surface tension of ^3He ;

$$\sigma = (c_1 - c_2 T \log_{10} p) \left(\frac{\rho_l - \rho_g}{M} \right)^{2/3} + c_3 T^2 + c_4 T^3 + c_5 T^4 \tag{3}$$

where the molar mass $M = 3.01603$, the vapor pressure p in Pa is taken from Ref. [2], the densities for the liquid and vapor (ρ_l and ρ_g) are from Ref. [4] (in $\text{kg} \cdot \text{m}^{-3}$), and the temperature (in K) is denoted by T . Based on the data analysis and filter of the measurements in Table 1, high weight was given to the high accuracy data from Iino et al. [10, 11]. A nonlinear least-squares fit to the model using all the reference data gives the coefficients: $c_1 = 1.694 \times 10^{-3}$, $c_2 = 2.4869 \times 10^{-5}$, $c_3 = -4.75792 \times 10^{-6}$, $c_4 = -8.42267 \times 10^{-6}$, and $c_5 = 2.5 \times 10^{-6}$. The maximum residual between the calculations by Eq. 3 and the experimental data from Iino et al. is $1.08 \times 10^{-6} \text{ N} \cdot \text{m}^{-1}$. The maximum relative residual is 1.06 % and the average is 0.34 %, which means the uncertainty of the equation is completely within that of all the measurements.

Like the vapor pressure and saturated density, the surface tension should also follow the scaling law when approaching the critical point. Iino et al. [11] measured the surface tension of liquid ^3He near its gas–liquid critical point and found the critical exponent to be 1.289 ± 0.015 , which is close to the value of normal fluids (=1.3). Thus, the scaling law applied to the surface tension of ^3He could be expressed as

$$\sigma = 3.07 \times 10^{-4} \left(\frac{T_c - T}{T_c} \right)^{1.289} \text{ N} \cdot \text{m}^{-1} \tag{4}$$

when T is equal to T_c , the surface tension becomes zero in Eq. 4. Equation 3 could be switched to Eq. 4 at 3.065 K with no discontinuity in the function and an acceptable discontinuity in the slope to give a more accurate description of the properties in this

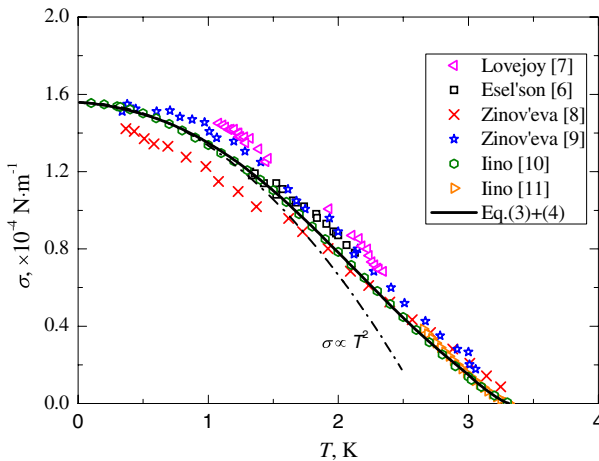


Fig. 1 Measurements and equation for the surface tension of ^3He

region. The switching point is just within the working temperature range determined by Iino et al. for Eq. 4.

The surface tension versus temperature curve generated by Eq. 3 combined with Eq. 4 is plotted in Fig. 1, in which the reference experimental data are also included as a comparison. It could be found that the surface tension equations represent the measurements within their uncertainties. At zero temperature, the surface tension of ^3He extrapolates to a maximum, without consideration of superfluidity below 2.6 mK. A dash line is plotted for the theoretical relation $\sigma \propto T^2$ which is almost overlapped by the curve of Eq. 3 below 1 K. Above 1.3 K, they depart from each other. The surface tension decreases by more than 50% when the temperature is increased from 1.08 K to 2.32 K.

4 Discussion and Conclusion

Zinov'eva [8] concluded that the surface-tension curves of ^3He and ^4He coincide on the σ/σ_0 versus T/T_c dimensionless diagram; thus, the different statistics of the isotopes leads to no noticeable difference in the temperature dependence of surface tension. In other words, on a σ versus T diagram, the curve for ^3He should appear to the left of that of ^4He by an offset of the distance between their critical points, also with a smaller slope. In order to verify this statement, the curve of the reduced surface tension for ^3He from this study and the curve for ^4He cited from the authoritative data by Donnelly and Barenghl [16] are plotted together in Fig. 2. Obviously these two curves do not coincide well. It should be pointed out that the ^3He surface tension versus temperature relation could be simplified as $\sigma_0 - \sigma = 2.23 \times 10^{-5} T^2 \text{ N} \cdot \text{m}^{-1}$ in the temperature range from 0.9 K down to 0 K, which is completely different from ^4He .

Another key point is about the surface tension value at absolute zero temperature. Lovejoy [7] deduced the surface tension of ^3He from the rules of ^4He within the lim-

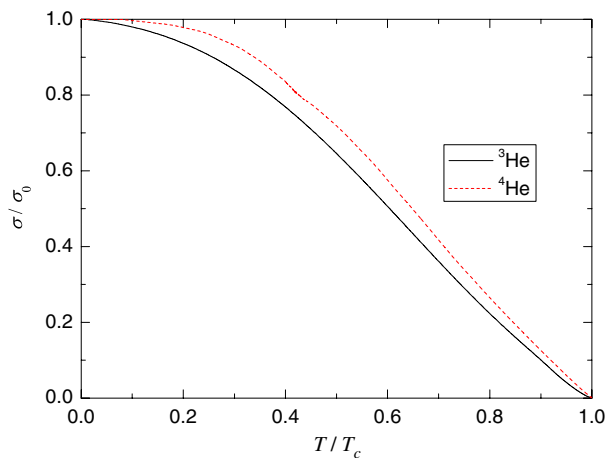


Fig. 2 Reduced surface tension of ^3He and ^4He

itation of then current theories and got a value $\sigma_0 = 1.54 \times 10^{-4} \text{N} \cdot \text{m}^{-1}$. Iino et al. [10] extrapolated their measurements down to 0K and got $\sigma_0 = (1.557 \pm 0.005) \times 10^{-4} \text{N} \cdot \text{m}^{-1}$. The value calculated by Eq. 3 is $1.5579 \times 10^{-4} \text{N} \cdot \text{m}^{-1}$, which is very close to Iino et al.'s value.

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