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SURFACE TENSION OF ^{4}He NEAR THE CRITICAL POINT

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We have measured the surface tension σ of ^{4}He near its liquid–gas critical point. The effective critical exponent for σ in the temperature interval $0.030 \lesssim (T_c - T)/T_c \lesssim 0.30$ is $\mu = 1.10$, distinctly smaller than the value 1.26 found for other simple liquids.

The behaviour of simple liquids in the vicinity of the critical point is generally well understood. The relevant bulk quantities like compressibility, correlation length and density difference between the coexisting liquid and vapour phase are described by power laws, whose critical exponents are in close agreement with renormalization group calculations; relationships between these critical exponents are given by the scaling laws [1,2].

The critical behaviour of the bulk materials is also reflected in the properties of the interface between the coexisting phases, in particular the interfacial tension σ. At the critical temperature T_c, σ has to vanish because the liquid and vapor phase become identical. One therefore expects a behaviour of the form

$$\sigma = \sigma_0 t^\mu , \tag{1}$$

where σ_0 is a constant, $t = (T_c - T)/T_c$ is the reduced temperature, and the critical exponent μ has a positive value. According to the theory of Widom [2] μ is related to the bulk properties via

$$\mu = \gamma - \nu + 2\beta , \tag{2a}$$

which is equivalent, using scaling relationships, to

$$\mu = (d - 1)\nu . \tag{2b}$$

Here β, γ, and ν are the critical exponents of the density difference, compressibility and correlation length, respectively, and the dimensionality of the system is $d = 3$. With the usual set of exponents for a scalar order parameter one obtains for μ a value of 1.26.

Although the theoretical basis for (2) is not quite as sound as for the critical exponents of the bulk quantities, this relation is found to be well fulfilled in the systems studied so far. For simple liquids like Ne, Xe and N_2 the experiment yields values for μ between 1.21 and 1.29, and also for more complex binary systems such as cyclohexane–methanol similar results are found (see table 1).

The system we have studied, ^{4}He, also belongs to the class of simple liquids. Our investigation was motivated by earlier measurements well below the critical temperature [3,4] which suggest a temperature dependence for σ distinctly weaker than predicted by (2). As our measurements show, the effective critical exponent μ is indeed significantly smaller than the expected value even relatively close to T_c; thus ^{4}He at its liquid–gas critical point appears to deviate in its behaviour from all the other systems investigated until now, including ^{3}He.

Table 1

System	μ	Ref.
^{3}He	1.24	5
Ne	1.21	6
Ar	1.281	2
Xe	1.287	2
N_2	1.247	2
cyclohexane – methanol	1.23	2

Our method for determining σ is based on the excitation of surface waves. We have used a technique where the interface is slightly charged with electrons in the presence of a vertical electric field E. Resonances of the surface waves in the cylindrical sample cell can then easily be excited by a small modulation of the holding field [7]. The dispersion relation of these waves is given by [8]

$$\omega^2 = \frac{(\rho_\ell - \rho_v)gk + \sigma k^3 - E^2k^2/2\pi}{\rho_\ell + \rho_v}, \tag{3}$$

where ω and k are the angular frequency and the wave vector, g is the acceleration due to gravity, and ρ_ℓ and ρ_v are the densities of the liquid and the vapor phase. In the present case the field E was chosen weak enough so that its contribution to eq. (3) was negligible. (This was checked by changing the field by a factor of two, which left the wavelength unaffected within our accuracy.) Likewise the electrons, having spacings larger than 10^4 Å, should be of negligible influence on σ. Taking into account the finite depths h_ℓ and h_v of the liquid and the vapor phase one then obtains [5]

$$\omega^2 = \frac{(\rho_\ell - \rho_v)gk + \sigma k^3}{\rho_\ell \coth(kh_\ell) + \rho_v \coth(kh_v)}. \tag{4}$$

A schematic drawing of the set up is shown in fig. 1a. Since we had optical access to the sample cell from above, the wave vector of the standing surface could be determined by an imaging technique which allowed to visualize surface deformations by stroboscopic illumination of the cell. An example of a resulting pattern is given in fig. 1b.

Values for the surface tension at varous temperatures were determined in the following way:

From a set of resonance frequencies and corresponding wave vectors the dispersion relation of surface waves was obtained for each temperature. A small correction to eq. (4) had to be taken into account because the observed surface waves are eigenmodes of the cylindrical cell, which gives rise to Bessel functions for the spatial dependence. Values for ρ_ℓ and ρ_v, which also appear in (4), were taken from the literature [9].

Fit parameters for the dispersion relation were the surface tension as the quantity of interest, and the depth h_ℓ of the liquid. The latter had to be included because our set up did not allow to measure h_ℓ precisely. (Near the critical point h_ℓ may vary considerably if the overall density deviates from the critical value.) It should be noted that in eq. (4) the ratio of the contribution due to surface tension (second term on the right-hand side) to the gravity contribution (first term) scales as $t^{3\beta-\mu}$ near T_c and is thus nearly independent

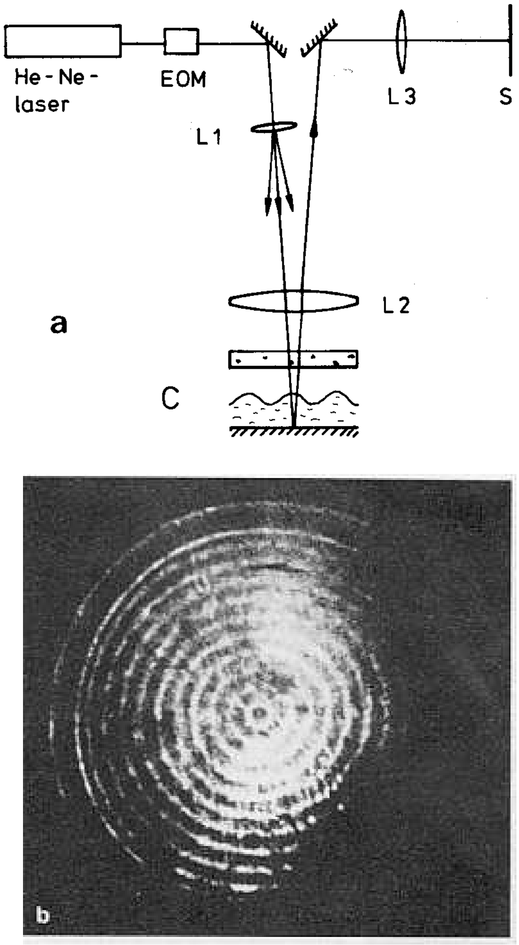

Fig. 1. (a) Sketch of set up for measuring the surface tension by the excitation of surface waves. The light beam of a He–Ne laser is chopped by an electrooptic modulator (EOM) and expanded by the lenses L_1 and L_2 to the diameter of the sample cell C. Lens L_3 images the cell onto the screen S. Curved portions of the helium surface modify the beam path, giving rise to a redistribution of the light intensity on S.
(b) Example of a standing wave pattern on the surface of liquid ^{4}He, obtained by illuminating the sample cell stroboscopically at the frequency of the surface waves. The wavelength in this case is 2 mm, the amplitude several μm.

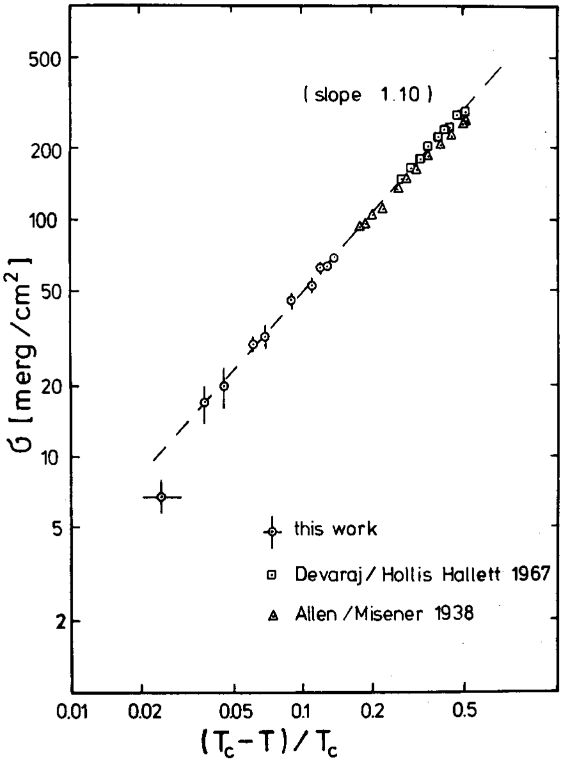

Fig. 2. Surface tension σ of ^{4}He versus the reduced temperature $t = (T_c - T)/T_c$. For the critical temperature we have taken the value $T_c = 5.198$ K [11]. ○, this work: △, Allen and Misener [3]; □, Deveraj and Hollis-Hallett [4].

of temperature. (This is the case, if one uses always the same frequency range, as was done in our experiment.)

Fig. 2 shows a plot of our data on logarithmic scales, together with results from previous measurements [3,4]. Apart from the point closest to T_c (at $t = 0.024$) the data are well represented by an expression

$$\sigma = \sigma_0 t^{1.10 \pm 0.05} ,$$

where $\sigma_0 = 0.62$ erg/cm^2. The effective critical exponent $\mu = 1.10$ for the range $0.038 \leqslant t \leqslant 0.3$ clearly deviates from the value 1.26 expected for a simple liquid. This discrepancy is somewhat reduced when the datum point at $t = 0.024$ (which has relatively large error bars, however) is included in the analysis. The damping of the surface waves in the investigated range was so weak that a noticeable influence on the value of σ is not to be expected. Besides, even if a weak divergence of the viscosity near T_c would be incorporated into the analysis, this would *increase* σ for the data nearest T_c rather than decreasing it [10], thus leading to an even smaller value of μ.

Two interpretations for the observed behaviour come to mind:

(i) Possibly – as suggested by the point at $t = 0.024$ – the asymptotic critical region where the surface tension displays its true critical behaviour is not reached in our experiment. In fact, for most other critical properties the effective exponents in the temperature range considered here differ from their asymptotic value closer to T_c.

(ii) A much more spectacular interpretation would be that the critical exponent μ is indeed distinctly different from the usual value 1.26, which would be at variance with universality.

Although at first glance argument (i) appears as the more obvious one, it would imply that the asymptotic critical region of σ is at least an order of magnitude smaller for ^{4}He than for any other simple liquid studied to date. Typically, for all the systems listed in table 1 an effective exponent $\mu \approx 1.26$ is found for temperatures down to about $0.7T_c$, equivalent to a critical region for σ extending as far as $t \approx 0.3$. In particular, such a wide critical region is found also for ^{3}He [5] and for ^{3}He–^{4}He mixtures near the tricritical point [12]. This rules out quantum effects [13] as a possible source for the anomalous behaviour of ^{4}He, since these effects should be even larger in ^{3}He.

In conclusion, we have observed an effective critical exponent μ for the surface tension of ^{4}He which is unusually small. Clearly experiments closer to T_c are required to distinguish between the various interpretations. Whatever the result – a small asymptotic critical region, or an asymptotic exponent μ which differs from 1.26 – the surface of ^{4}He appears to deviate in its behaviour from that of most other simple liquids. An explanation for such a discrepancy is presently not at hand.

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Note added. In a recent publication by Iino et al. [14] results for σ of ^{4}He near T_c are presented which are consistent with the data in fig. 2.

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