Critical Casimir force in \(^{4}\)He films: confirmation of finite-size scaling

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We present new capacitance measurements of critical Casimir force-induced thinning of \(^{4}\)He films near the superfluid/normal transition, focused on the region below \(T_{\lambda}\) where the effect is the greatest. \(^{4}\)He films of 238, 285, and 340 Å thickness are adsorbed on N-doped silicon substrates with roughness \(\approx 8\) Å. The Casimir force scaling function \(\vartheta\), deduced from the thinning of these three films, collapses onto a single universal curve, attaining a minimum \(\vartheta = -1.30 \pm 0.03\) at \(x = t d^{1/\nu} = -9.7 \pm 0.8\) Å\(^{1/\nu}\). The collapse confirms the finite-size scaling origin of the dip in the film thickness. Separately, we also confirm the presence down to \(2.13K\) of the Goldstone/surface fluctuation force, which makes the superfluid film \(\sim 2\) Å thinner than the normal film.

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An important focus in condensed matter physics is understanding how the properties of a thermodynamic system evolve as its size is shrunk to ever smaller dimensions. Near a continuous phase transition or critical point, the theory of finite-size scaling offers a testable prediction. According to finite-size scaling, the correction to the free energy per unit area of a planar film of thickness \(d\) due to confinement of critical fluctuations has a simple, universal form \(\Theta_1\).

\[
\delta F_{12} = \frac{k_B T_c}{d^2} \Theta_{12}(d/\xi).
\]

where \(T_c\) is the transition temperature and the correlation length \(\xi = \xi_0|t|^{-\nu}\) measures the spatial extent of fluctuations in the bulk. \(t = T/T_c - 1\) is the reduced temperature. The scaling function \(\Theta_{12}\) is predicted to be a dimensionless, universal function of the ratio \(d/\xi\) and the boundary conditions that the order parameter satisfies at the confining interfaces.

While finite-size scaling is applicable to all critical systems, the most rigorous experimental tests to date have focused on the scaling behavior of the specific heat anomaly of \(^{4}\)He near the superfluid transition \(\nu \approx 3\). This is due to the nearly-ideal, impurity-free nature of liquid \(^{4}\)He and the low-sensitivity of this system to gravitational rounding errors. For the superfluid transition \(T_c = T_{\lambda} = 2.1768K\) and \(\nu = 0.67016 \pm 0.00008\). For a 57 μm thick film, the magnitude and temperature dependence of the specific heat is found to be in reasonable agreement with finite-size scaling predictions \(\Theta_2\). However, for films 500 – 7000 Å thick, the situation is not as clear-cut. The temperature-dependence of the specific heat is as expected from the universal \(d/\xi\) dependence in Eq. \(\Theta_1\). The maximum specific heat occurs at a common value \(x = (d\xi_0/\xi)^{1/\nu} = t d^{1/\nu} = -9 \pm 1\) Å\(^{1/\nu}\) for all films, where the negative \(x\) refers to the maximum occurring below \(T_{\lambda}\). However, the magnitude of the specific heat shows an unexpected, systematic non-collapse \(\Theta_3\).

The critical Casimir force is another fundamental manifestation of finite-size scaling that is open to experimental testing. Just as the Casimir force between two conducting plates arises due to the confinement of zero-point electromagnetic fluctuations between the plates \(\Theta_4\), a completely analogous thermodynamic Casimir force is expected between the substrate and vapor interfaces of adsorbed liquid films, due to the confinement of critical fluctuations within the thickness of the film \(\Theta_5\). The theoretically-predicted critical Casimir force per unit area

\[
f = -\frac{\partial \delta F_{12}}{\partial d} = \frac{k_B T_c}{d^3} \vartheta_{12}(d/\xi)
\]

where Casimir scaling function \(\vartheta(z) = 2\Theta(z) - z\partial\Theta/\partial z\). Because in \(^{4}\)He films the superfluid order parameter vanishes at both film interfaces, the critical Casimir force is attractive \(\vartheta < 0\) \(\Theta_6\), producing a dip in the equilibrium film thickness near \(T_{\lambda}\). The existence of this dip, first observed by \(\Theta_7\), has been confirmed in a quantitative experiment using as substrates five pairs of capacitor plates made of polished Cu set at different heights above bulk liquid helium \(\Theta_8\). The interpretation of this experiment is complicated by the roughness of the Cu surface, which changes the effective areas of the Cu surface and makes it impossible to accurately determine the film thickness. AFM scans over 2500μm\(^2\) areas of the surfaces show they are not ideal, with 10-130 Å rms roughness and occasional micron-deep scratches and dust particles. \(\vartheta\)
is calculated using DLP theory \([14]\) and by assuming that the Cu surfaces at different heights are flat. The result of the experimental analysis \([8]\) is that the scaling function \(\vartheta\) exhibits a behavior suggestively similar to the specific heat. The minimum in the dip occurs at a common value \(x = -9.2 \pm 0.2 \, \text{Å}^{1/\nu}\) for all the films 257-423 Å thick. The temperature-dependence is exactly that expected from \(d/\xi\) dependence in Eq. (2), but the magnitude of \(\vartheta\) shows an unexpected non-collapse, the minimum of \(\vartheta\) increasing systematically from \(-1.85\) to \(-1.4\) as \(d\) increases from 257 Å to 423 Å \([8]\). To address whether this systematic trend in the magnitude of \(\vartheta\) is an artifact due to the non-ideal surface or is truly related to the non-collapse observed for the specific heat, we have undertaken improved capacitance measurements of the critical Casimir force similar to \([8]\) but using flat N-doped silicon surfaces with roughness \(\approx 8\) Å.

A sketch of the experimental cell machined from oxygen-free high conductivity Cu is presented in Fig. 1. Two silicon (100) wafers highly-doped with phosphorous \((1-5 \, \text{mΩ/□})\) are configured as parallel plates forming a capacitor with a gap \(G = 235 \, \mu\text{m}\). According to a tapping-mode AFM, the rms roughness of the wafers is 8 Å. To minimize surface roughness, dust particles, and scratches, the experimental cell is washed, dried, assembled and sealed in the Penn State Nanofabrication facility, a class 10 clean room. Virgin wafers, completely intact, are rubber-cemented into the Cu guard rings used to position the electrodes. The top electrode is a 1 inch wafer, and the bottom is 2 inch. To minimize error from the fringe field, the top and bottom guard rings are grounded and the 1 inch wafer is placed at virtual ground in the AC bridge circuit used to measure the capacitance \(C\) \([8, 11]\). To determine \(T_\lambda\), a fixed point device anchored to the cell bottom is used, following the procedure described by \([12]\).

The temperature control scheme of our experiment is similar to that of the original experiment of \([8]\). A needle valve is used to close the helium fill line just above the cell. The data are taken with the cell slowly drifting through the lambda point, at 10-40μK/h near \(T_\lambda\) where equilibration takes longer and at 70-300μK/h below \(T_\lambda\). We use two thermal control stages. The first outer stage is maintained at constant temperature with less than 50μK noise. To achieve a uniform temperature drift rate, we apply heat to a second stage, just above the cell. After dosing helium into the cell, we typically observe signs of capillary condensation, where liquid droplets condense in the gap between the silicon electrodes. To get rid of these droplets, we very slowly \((100 - 300\, \text{μK/h})\) thermally cycle the cell through \(T_\lambda\), each time looking for a distinctive drop in capacitance that signals the flowing of liquid from the gap. This procedure is repeated until a reproducible \(C(T)\) dependence is obtained.

To calculate the film thickness \(d\) from the measured \(C(T)\), we model \(C(T)\) as the equivalent capacitance due to three dielectric layers added in series: adsorbed film, vapor phase, and adsorbed film, obtaining

\[
d = \frac{G}{2} \left( \frac{1}{\varepsilon_{\text{vapor}}} - \frac{1}{\varepsilon(T)} \right) / \left( \frac{1}{\varepsilon_{\text{vapor}}} - \frac{1}{\varepsilon_{\text{film}}} \right) \tag{3}
\]

where, if \(C_0(T)\) is the temperature-dependent empty capacitance, the effective dielectric constant \(\varepsilon(T) = C(T)/C_0(T)\). As in \([8]\), the dielectric constant of the film \(\varepsilon_{\text{film}} = 1.05760 \pm 0.00005\) and the dielectric constant of the vapor \(\varepsilon_{\text{vapor}}\) is calculated using the Clausius-Mossotti equation, taking the molar polarizability of helium to be 0.123296 ± 0.000030 cm\(^3\)/mol. The vapor density is calculated from the pressure \(P(T)\), using the second virial coefficient \(B(T)\) from \([13]\).

The temperature-dependence of \(C_0\) is due to a small linear increase in \(G\) caused by a combination of liquid surface tension acting on the Cu spacers and differential thermal contractions among the various materials that make up the capacitor, including between the silicon wafer and the rubber cement.
than 5% or 10˚C of silicon. Nevertheless, the error is estimated to be less than 0.1%. The effect of the small 20˚C temperature dependence of the equilibrium film thickness is negligible.

In our analysis, we assume that $C_0(T) = C_0(T\lambda)(1 - 3.5 \times 10^{-5}(T - T\lambda))$. This results in a temperature-independent $d$ for all films for $T$ sufficiently above or below $T\lambda$. Each time we dose liquid into the cell to make a new film, we characterize observe an additional small shift in $G$ (and $C_0$) on the order of 50ppm. To correct for this, we adjust $C_0(T\lambda)$ in order to obtain the theoretically-predicted thickness in the regime above $T\lambda$ where the critical Casimir force is negligible and the equilibrium thickness $d$ on the silicon wafer is expected to be determined solely by a competition between temperature-independent van der Waals and gravitational forces.

In this regime, the film thickness is given by

$$mgh = \frac{\gamma_0}{d^3} \left( 1 + \frac{d}{d_{1/2}} \right)^{-1} \left( 1 + \frac{d}{d_{1/2}} \right)^{-1}$$

where, on the left side $mgh$ is the chemical potential due to gravity, fixed by the height $h$ above the bulk liquid, $g$ is the gravitational acceleration and $m$ the atomic weight of helium. On the right side, $d_{1/2}$ is a simplified expression for the chemical potential due to van der Waals forces, where $\gamma_0 \approx 1950 K \AA^3$ and $d_{1/2} \approx 230 \AA$ are substrate-specific interpolation parameters that characterize the net attraction of the helium to the silicon, including retardation effects. The parameters $\gamma_0$ and $d_{1/2}$ are approximate, ignoring the effect of the small 20 Å natural oxide layer on the silicon. Nevertheless, the error is estimated to be less than 5% or 10 Å and the same for all the films studied.

In Fig. 2, we show the measured change in the film thickness in response to the temperature-dependent Casimir force near and below $T\lambda$, for different values of the height $h = 15.00$, 8.01, and 4.22 ±0.05mm. The films are labeled by their thicknesses above $T\lambda$ calculated from Eq. (6), namely 238, 285, and 340 ±10Å. As seen previously, due to the Casimir force, thicker films exhibit larger dips which occur closer to $T\lambda$. Including the additional contribution to the chemical potential from the critical Casimir force, the equilibrium film thickness is expected to be given by

$$mgh = \frac{\gamma_0}{d^3} \left( 1 + \frac{d}{d_{1/2}} \right)^{-1} + k_B T\lambda V \frac{\partial}{d^3} \vartheta(d/\xi)$$

where $V = 45.81 \AA^3/atom$ is the specific volume of liquid $^4$He, and $\vartheta$ is the dimensionless scaling function for the Casimir force. The observed dip in $d$ is due to $\vartheta < 0$, i.e. an attractive Casimir force between the substrate and vapor interfaces, as expected due to the superfluid order parameter satisfying Dirichlet boundary conditions at the two film interfaces.

In Fig. 3, we show the Casimir force scaling function calculated using Eq. (6) and the data of Fig. 2. Because it was necessary to disrupt data collection every 2.5 days to transfer cryogen, each curve, which takes about 2 weeks to complete, actually consists of 4-5 overlapping data sets that are spliced together; this results in additional noise and a small discrepancy close to $T\lambda$, not present in the earlier work. Nevertheless, within the scatter, all three data sets collapse onto a single curve, with a minimum value of
\( \theta = -1.30 \pm 0.03 \) at \( x = -9.7 \pm 0.8 \Delta^{1/\nu} \). The collapse of the data verifies that the dip in the film thickness near \( T_\lambda \) is due to fluctuation-induced forces. It is noteworthy that the measured \( \theta \) shows quantitative agreement with the \( \theta \) obtained previously \(^5\) for 423 A thick \( ^4 \)He films on Cu, but disagrees with the results obtained for thinner films that, presumably, would be more sensitive to surface non-idealities. These results suggest the non-collapse is the result of inadequate corrections for the effects of surface roughness and not due to \( \theta \) depending on the additional off-coexistence variable \( h \Delta^{1/\nu} / \nu \), where \( \Delta / \nu = 2.47 \). This is expected to have important implications for the analysis of specific heat and wetting experiments \(^2\ 3\ 12\ 13\ 17\).

The new measurements, which focus on obtaining data near the minimum of the dip and over a wide range below \( T_\lambda \), confirm an additional important aspect of earlier experiments: for all the films studied, we find the superfluid film is \( \sim 2 \) A thinner than the normal film down to 2.13K. Experiments indicate that the onset of superfluidity in the films occurs somewhere between \( x = -7 \) and \( -12 \Delta^{1/\nu} \). Thus it has been suggested that the thinner superfluid film is caused by Casimir forces due to fluctuations involving superfluidity in the film, such as Goldstone modes, second and third sound \(^16\). As seen from Fig. 3, the thinning in the superfluid film is consistent with an asymptotic, low-temperature value of the Casimir force \( \approx -(0.30 \pm 0.10) k_B T / d^3 \). This force is marginally larger than the \( -0.15 k_B T / d^3 \) force predicted by \(^16\).

In summary, the current experiment confirms the validity of finite-size scaling formula for the critical Casimir force in adsorbed \( ^4 \)He films between 230 and 340 A thick. Measurements down to \( \sim 2.13K \) also show the presence of an additional, non-critical, attractive fluctuation-induced force in the superfluid film. Our study underscores the importance of smooth surfaces for these types of measurements. For future work, it would be desirable to test the scaling of Casimir forces in a much wider thickness range that overlaps the range covered by specific heat measurements. We would like to thank A. Maciokle, R. Zandi, J. Rudnick, M. Krech, D. Dantchev, S. Dietrich, M. Kardar, S. Balibar, G. Williams, F. M. Gasparini, and M. W. Cole for useful discussions and comments. This research was supported by NASA grant NAG3-2891.

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