

Critical Casimir Force in ^4He Films: Confirmation of Finite-Size Scaling

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

$$\delta F_{12} = \frac{k_B T_c}{d^2} \Theta_{12}(d/\xi), \quad (1)$$

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 and ν is the correlation length exponent. The scaling function Θ_{12} is predicted to be a dimensionless, universal function of the ratio d/ξ 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 in ^4He films near the superfluid transition [2,3]. This is due to the nearly ideal, impurity-free nature of liquid ^4He and the low sensitivity of this system to gravitational rounding errors. For the superfluid transition, $T_c = T_\lambda = 1.1768 \text{ K}$ and $\nu = 0.67016 \pm 0.00008$ [4]. For a $57 \mu\text{m}$ thick film, the magnitude and temperature dependence of the specific heat is found to be in reasonable agreement with finite-size scaling predictions [2]. However, for films 500–7000 Å thick [3], the situation is not as clear-cut. The temperature dependence of the specific heat is as expected from the universal d/ξ dependence in Eq. (1). The maximum specific heat occurs at a common value $x = (d\xi_0/\xi)^{1/\nu} = td^{1/\nu} = -9 \pm 1 \text{ \AA}^{1/\nu}$ for all films, where the negative x refers to the maximum occurring below T_λ . However, the *magnitude* of the specific heat shows an unexpected, systematic noncollapse [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 [5], 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 [1,6–9]. The theoretically predicted critical Casimir force per unit area

$$f = -\frac{\partial \delta F_{12}}{\partial d} = \frac{k_B T_c}{d^3} \vartheta_{12}(d/\xi), \quad (2)$$

where the Casimir scaling function $\vartheta(z) = 2\Theta(z) - z\partial\Theta/\partial z$. Because in ^4He films the superfluid order parameter vanishes at both film interfaces, the critical Casimir force is attractive ($\vartheta < 0$) [6], producing a small dip near T_λ . The existence of this dip, first observed in Ref. [10], 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 [7]. The interpretation of this experiment is complicated by the roughness of the Cu surface, which changes the effective areas of the Cu plates and makes it impossible to accurately determine the film thickness. Atomic force microscope (AFM) scans of the surfaces show they are not ideal, with 10–130 Å rms roughness and occasional micron-deep scratches and dust particles. ϑ is calculated using Dzyaloshinskii-Lifshitz-Pitaevskii theory [11] and by assuming that the Cu surfaces at different heights are flat. The result of the experiment [7] is that the thinning above T_λ ($x > 0$) agrees with the theoretically calculated ϑ for Dirichlet boundary conditions, which exists only for $x > 0$ [6]. The minimum in the scaling function occurs at a common value $x = -9.2 \pm 0.2 \text{ \AA}^{1/\nu}$ for all the films 257–423 Å thick. ϑ exhibits a behavior suggestively similar to the specific heat. The temperature dependence is exactly that expected from the d/ξ dependence in

Eq. (2), but the magnitude of ϑ shows an unexpected noncollapse, the minimum of ϑ increasing systematically from -1.85 to -1.4 as d increases from 257 to 423 Å [7]. To address whether the systematic trend in the magnitude of ϑ is an artifact due to the nonideal surface or is truly related to the noncollapse observed for the specific heat, we have undertaken improved capacitance measurements of the critical Casimir force similar to Ref. [7] but using atomically smooth, flat N -doped silicon surfaces.

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 ($1\text{--}5$ m Ω/\square) are configured as parallel plates forming a capacitor with a gap $G = 235$ μm . Tapping-mode AFM scans over 0.01 μm^2 areas of these surfaces yields an rms roughness of 1 Å, but scans over larger 1 μm^2 regions give 8 Å due to occasional scratches. 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 inches. 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 [7,12]. To determine T_λ , a fixed point device anchored to the cell bottom is used, following the procedure described by Ref. [13].

The temperature control scheme of our experiment is similar to that of the original experiment of Ref. [7]. A needle valve is used to close the helium fill line just above

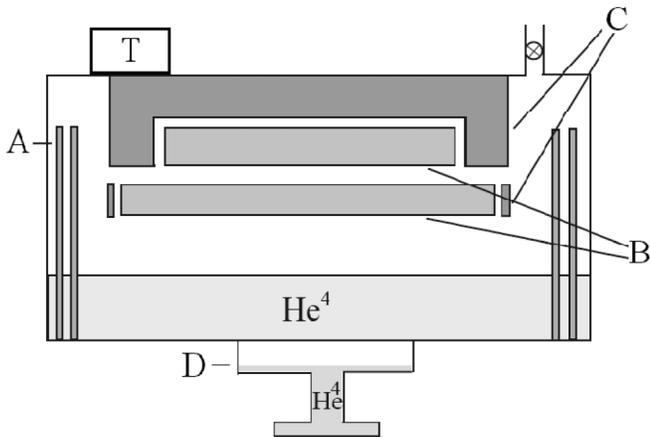


FIG. 1. The sketch of the experimental cell. The level of the bulk ^4He at the bottom of the cell is measured using an annular capacitor (A). The ^4He film is adsorbed on the two silicon plates (B) attached to electrically grounded copper guard rings (C) held 0.2 mm apart by Cu shim spacers. The temperature of the cell was measured using a germanium thermometer T attached to the top of the cell, which is calibrated vs the ^3He vapor curve and the lambda fixed point device (D) at the bottom.

the cell. The data are taken with the cell slowly drifting through the lambda point, at $10\text{--}40$ $\mu\text{K}/\text{h}$ near T_λ , where equilibration takes longer, and at $70\text{--}300$ $\mu\text{K}/\text{h}$ below T_λ . We use two thermal control stages. The first outer stage is maintained at constant temperature with less than 50 μK 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 ($100\text{--}300$ $\mu\text{K}/\text{h}$) thermally cycle the cell through T_λ , 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

$$d = \frac{G}{2} \left(\frac{1}{\epsilon_{\text{vapor}}} - \frac{1}{\epsilon(T)} \right) / \left(\frac{1}{\epsilon_{\text{vapor}}} - \frac{1}{\epsilon_{\text{film}}} \right), \quad (3)$$

where, if $C_0(T)$ is the temperature-dependent empty capacitance, the effective dielectric constant $\epsilon(T) = C(T)/C_0(T)$. As in Ref. [7], the dielectric constant of the film $\epsilon_{\text{film}} = 1.05760 \pm 0.00005$, and the dielectric constant of the vapor ϵ_{vapor} is calculated using the Clausius-Mossotti equation, taking the molar polarizability of helium to be 0.123296 ± 0.000030 cm^3/mol [14]. The vapor density is calculated from the pressure $P(T)$, using the second virial coefficient $B(T)$ from Ref. [15].

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 underneath. In our data 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 far above as well as below T_λ . Each time we dose liquid into the cell to make a new film, we characteristically observe an additional small shift in G (and C_0) on the order of 50 ppm. To correct for this, we adjust $C_0(T_\lambda)$ in order to obtain the theoretically predicted thickness sufficiently far above T_λ 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 [11]

$$mgh = \frac{\gamma_0}{d^3} \left(1 + \frac{d}{d_{1/2}} \right)^{-1}, \quad (4)$$

where, on the left side, mgh is the chemical potential due to gravity, fixed by the height h above the bulk liquid, where g

is the gravitational acceleration and m the atomic weight of helium. On the right side is a simplified expression for the chemical potential due to van der Waals forces, where $\gamma_0 \approx 1950 \text{ K \AA}^3$ and $d_{1/2} \approx 230 \text{ \AA}$ are substrate-specific interpolation parameters that characterize the net attraction of the helium to the silicon, including retardation effects [11]. The parameters γ_0 and $d_{1/2}$ are approximate, ignoring the effect of the small 20 \AA natural oxide layer on the silicon. Nevertheless, the error is estimated to be less than 5% or 10 \AA 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_λ , for three different values of the height $h = 15.00, 8.01,$ and $4.22 \pm 0.05 \text{ mm}$. The films are labeled by their thicknesses above T_λ calculated from Eq. (4), namely, 238, 285, and $340 \pm 10 \text{ \AA}$. As seen previously [7], due to the Casimir force, thicker films exhibit larger dips which occur closer to T_λ . Including the additional contribution to the chemical potential from the critical Casimir force [6], the equilibrium film thickness is expected to be given by

$$mgh = \frac{\gamma_0}{d^3} \left(1 + \frac{d}{d_{1/2}}\right)^{-1} + \frac{k_B T_\lambda V}{d^3} \vartheta(d/\xi), \quad (5)$$

where $V = 45.81 \text{ \AA}^3/\text{atom}$ is the specific volume of liquid ^4He , and ϑ 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 [6].

In Fig. 3, we show the Casimir force scaling function calculated using Eq. (5) and the data in Fig. 2. Because it

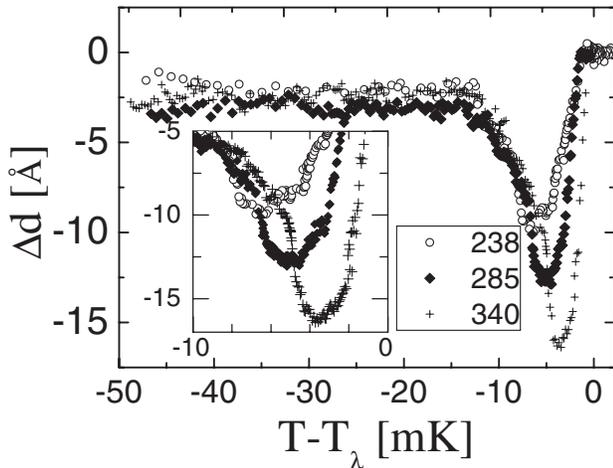


FIG. 2. The thinning of the film plotted vs temperature T : the data shown over a wide range of temperature and (inset) a blowup near the minimum. The films are labeled by the thickness in \AA in the region above T_λ where the Casimir force is negligible.

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 very close to T_λ not present in the earlier work [7]. Nevertheless, within the scatter, all three data sets collapse onto a single curve, with a minimum value of $\vartheta = -1.30 \pm 0.03$ at $x = -9.7 \pm 0.8 \text{ \AA}^{1/\nu}$. The collapse of the data verifies that the dip in the film thickness near T_λ is due to fluctuation-induced forces [6]. It is noteworthy that the measured ϑ shows quantitative agreement with the ϑ obtained previously [7] for 423 \AA thick ^4He films on Cu but disagrees with the results obtained for thinner films that, presumably, would be more sensitive to surface nonidealities. These results suggest that the non-collapse is the result of inadequate corrections for the effects of surface roughness and not due to ϑ depending on the additional off-coexistence variable $hd^{\Delta/\nu}$, where $\Delta/\nu = 2.47$ [6]. This is expected to have important implications for the analysis of specific heat and wetting experiments [2,3,16].

The new measurements, which focus on obtaining data near the minimum of the dip and over a wide range below T_λ , confirm an additional important aspect of earlier experiments: For all the films studied, we find the superfluid film is $\sim 2 \text{ \AA}$ thinner than the normal film down to 2.13 K . Experiments indicate that the onset of superfluidity in the films occurs somewhere between $x = -7$ and $-12 \text{ \AA}^{1/\nu}$ [3,17]. 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 and second and third sound [18]. 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 larger than the $-0.15k_B T/d^3$ force predicted by Ref. [18].

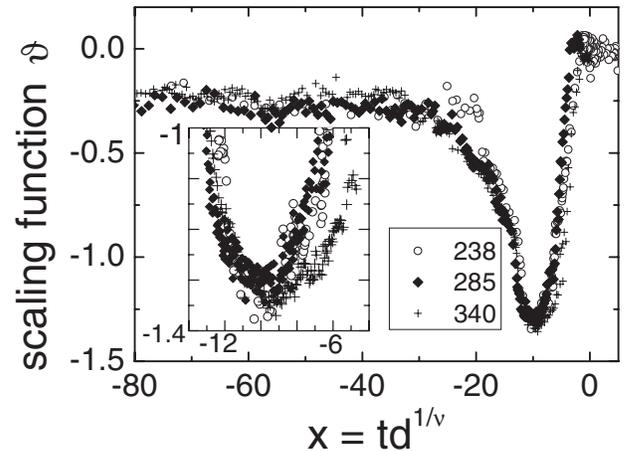


FIG. 3. The scaling function ϑ vs the scaling variable x . The minimum occurs at $x = -9.7 \pm 0.8 \text{ \AA}^{1/\nu}$, for all three films.

In summary, the current experiment confirms the validity of finite-size scaling formula for the critical Casimir force in adsorbed ^4He films between 230 and 340 Å thick. Measurements down to ~ 2.13 K also show the presence of an additional, noncritical, 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.

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