

Van der Waals effects at the superfluid transition of confined ^4He

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We explore the role which the van der Waals field plays in the behavior of the superfluid density of confined helium near T_λ . This effect can be calculated in the context of Ψ -theory. The influence of a solid wall is incorporated into the equation for the order parameter via a spatially-dependent transition temperature. The equation is solved numerically. We first calculate the scaling function without wall interactions for the case of 2D, 1D, and 0D dimensionality crossover. We find that upon introducing the van der Waals field, its influence is too small, and in the wrong direction to explain experimental observations.

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1. INTRODUCTION

The critical behavior of a confined system is a fundamental problem in statistical mechanics. Unlike the situation achieved in the thermodynamic limit, the confined system's response is dependent on geometry and boundary conditions. Nevertheless, near a second order phase transition, the growth of the correlation length allows one to scale finite systems so that equivalent geometries, such as films, can be expected to collapse on a universal locus. This collapse was first predicted by Fisher and Barber^{1,2}. Recently, experimental results for the specific heat of ^4He confined in a planar geometry, where one crosses over from three dimensions(3D) to 2D, have confirmed these predictions³⁻⁶. The predicted scaling is obtained for data in a planar confinement up to a temperature near the specific heat maximum. Beyond this, scaling is observed to fail^{4,5}. In the case of the superfluid density, for planar confinement, it is observed that the data do not scale at all⁷.

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For helium at a rigid wall, one may take as a boundary condition one where the order parameter vanishes. However, a wall will have additionally a van der Waals interaction with the helium, which will produce non-scaling effects on the critical behavior. Because this field is localized, one expects this effect to become negligible as the confinement becomes large. It is of interest to estimate this effect, especially for the superfluid density which is observed not to scale. One can do this in the context of Ψ -theory^{8,9}. This is a phenomenological theory which has been applied to a number of experimental situations by Ginzburg and Sobyenin¹⁰⁻¹³. It is, to be sure, an approximation of the critical behavior of ^4He in the sense that the exponents in the theory are not correct; the specific heat exponent α is taken as 0 instead of -0.013, and that of the superfluid density ν as 2/3 rather than 0.671. Limitations of Ψ -theory are discussed by Ginzburg and Sobyenin¹⁰.

2. Ψ -THEORY FORMALISM

In Ψ -theory one starts with an expansion of the Gibbs free energy in the order parameter ψ . This is related to the superfluid density ρ_s via

$$\rho_s = m|\psi^2|. \quad (1)$$

When the free energy is minimized one obtains,

$$\nabla_*^2 \left(\frac{\psi}{\psi_0} \right) = \frac{3}{3+M} \left(-\epsilon^{4/3} \left(\frac{\psi}{\psi_0} \right) + (1+M)\epsilon^{2/3} \left(\frac{\psi}{\psi_0} \right)^3 + M \left(\frac{\psi}{\psi_0} \right)^5 \right) \quad (2)$$

where $\epsilon = |T_\lambda - T|$, $\psi_0^2 = (k\rho_\lambda/mT_\lambda^{2/3}) = 1.43(\rho_\lambda/m)K^{-2/3}$, $k = 2.395$ is the dimensionless prefactor in the power-law behavior of ρ_s/ρ , and M is a dimensionless constant which determines the strength of the term $|\psi|^6$ in the free energy. The spatial variable has also been scaled, $r_* = r/\xi_{0M}$, and $\xi_{0M}^2 = \hbar k T_\lambda^{2/3} / (2m^2 \Delta C_\mu)$. One can further reduce Eq. 2, by introducing the variables $\eta = \psi/(\psi_0 \epsilon^{1/3})$ and $u = r_* \epsilon^{2/3}$ to obtain

$$\nabla^2 \eta = \frac{3}{3+M} \left(-\eta + (1-M)\eta^3 + M\eta^5 \right). \quad (3)$$

Solution of this equation over the confinement region will yield a function $\eta^2(r) = \rho_s/\rho_{sb}$, where ρ_{sb} is the bulk superfluid density. This can then be averaged over the spatial confinement to connect with experimental results. From the form of Eq. 3, η must depend on the ratio r/l , where l is a temperature dependent length diverging as $\epsilon^{-2/3}$. Upon averaging over a spatial distance L , the ratio $\bar{\rho}_s(\epsilon, L)/\rho_{sb}(\epsilon)$ will depend on the variable

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$\epsilon L^{3/2}$. This is the expected correlation-length scaling variable. The specific dependence on $\epsilon L^{3/2}$ is determined by the geometry.

With a van der Waals potential $U(r)$, assuming the helium is incompressible, one can write the chemical potential μ as¹¹,

$$\mu = \mu_0 + U(r). \quad (4)$$

The variation in μ as the surface is approached results in a local shift of T_λ so that one has

$$\epsilon = \epsilon_0 + \frac{dT_\lambda}{d\mu} U(r) \quad (5)$$

where ϵ_0 is $T_\lambda - T$ in the absence of $U(r)$, and

$$\frac{dT_\lambda}{d\mu} = -s_\lambda + v_\lambda \frac{dP_\lambda}{dT} = -311 \text{J/mol K}. \quad (6)$$

We consider first helium in a planar geometry between two surfaces at separation L and infinite lateral extent. One has, at a point z away from one surface,

$$U(z) = \frac{-\alpha_0}{z^3} \left(1 + 1.64 \left(\frac{z}{d_{1/2}} \right)^{1.4} \right)^{-1/1.4} - \frac{\alpha_0}{(L-z)^3} \left(1 + 1.64 \left(\frac{L-z}{d_{1/2}} \right)^{1.4} \right)^{-1/1.4}. \quad (7)$$

Note z^{-3} is the dependence close to the surface and z^{-4} reflects the crossover into a retardation regime¹⁴. For the case of a silicon surface, one has $\alpha_0 = 1950 \text{ K}\text{\AA}^3$ and $d_{1/2} = 230\text{\AA}$ ¹⁴.

To incorporate $U(z)$ into the equation for the order parameter, one introduces a new variable $u = z\epsilon^{2/3}/\xi_{0M}$ and the constant

$$u_0 = \left| \frac{dT_\lambda}{d\mu} \frac{\alpha_0 \epsilon_0}{\xi_{0M}} \right|^{1/3}. \quad (8)$$

For simplicity, consider the case with no retardation first. The above constant is a spatial cutoff which is temperature dependent. Beyond this cutoff ρ_s will be zero. This reflects the finite slope of the λ -line as a function of μ , or pressure. The equation for η becomes

$$\begin{aligned} \frac{d^2\eta}{du^2} = \frac{3}{3+M} & \left\{ -\eta \left[1 - u_0 \left(\frac{1}{u^3} + \frac{1}{\left(\frac{L\epsilon_0^{2/3}}{\xi_{0M}} - u \right)^3} \right) \right]^{4/3} \right. \\ & \left. + (1+M)\eta^3 \left[1 - u_0 \left(\frac{1}{u^3} + \frac{1}{\left(\frac{L\epsilon_0^{2/3}}{\xi_{0M}} - u \right)^3} \right) \right]^{2/3} + M\eta^5 \right\}. \end{aligned} \quad (9)$$

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For a numerical calculation one chooses an L , and for any ϵ_0 , this fixes the quantity $L\epsilon_0^{2/3}/\xi_{0M}$, and the cutoff for the numerical integration. In practice one searches for non oscillatory solutions with values η at $z = L/2$ which yield $\eta = 0$ at the cutoff points. One then averages $\eta^2(u)$ over the confinement space.

3. RESULTS

We consider first $U(r) = 0$, and confinements to films, cylinders and spheres. In these cases, Eq. 3 can be solved as one-variable equation, but with different expressions for ∇^2 . The results with $M = 0$ are shown in Fig. 1. Here we plot the ratio of confined to bulk superfluid density as function of the scaling variable $tL_{eff}^{3/2}$, where $t = \epsilon_0/T_\lambda$ and L_{eff} in μm is the *thickness* in the planar case, or the *radius* for the cylindrical and spherical cases. These curves are the scaling functions in Ψ -theory for these confinements.

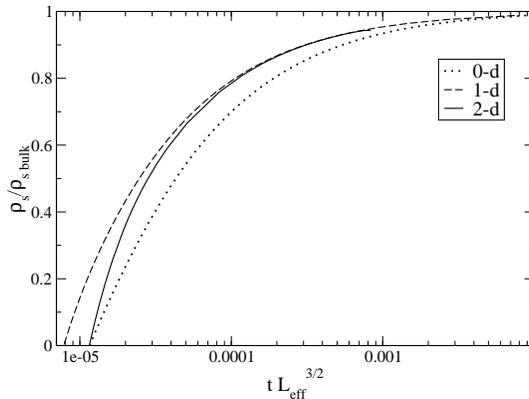


Fig. 1. Scaling functions for different confinements, and $U(r) = 0$.

Results for cylindrical and planar confinements have been obtained previously by Bot and Zimmermann¹⁵ and Wang *et al.*¹⁶ respectively. We are in good agreement with these calculations. Also the point at which the superfluid density vanishes has been obtained for these geometries by Ginzburg and Sobyenin¹⁰.

The calculation of the superfluid density ratio for planar geometry and with $U(r)$ for several L 's (these are L 's for which data exist) is done using

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Eq. 9. Results are shown in Fig. 2. As expected, $U(r)$ lowers ρ_s and the curves for different L 's no longer collapse. The effect is largest for the smallest confinement. The inset in this figure shows these results as a percent difference between the scaling function and each individual L . These are "correction" curves which could be used to eliminate from the experimental data the effect of $U(z)$. We have also investigated using the full form for $U(z)$, Eq. 7, in Eq. 9. This has negligible effects on the results.

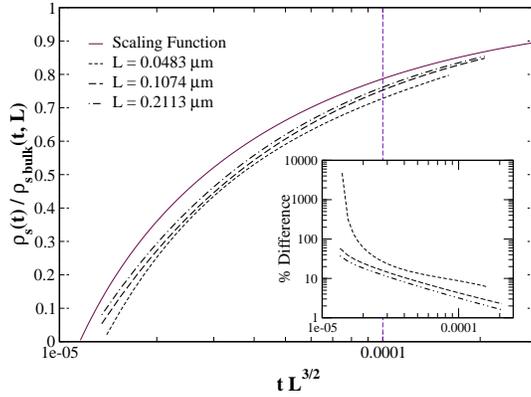


Fig. 2. Effects of $U(r)$ for planar confinement

For cylindrical confinement the function $U(r)$ is given by¹⁷,

$$U(r) = -(3\pi\alpha_0/2R^2)F(3/2, 5/2, 1, r/R), \quad (10)$$

where F is a hypergeometric function. We have used this to calculate the values of ρ_s/ρ_{sb} for the radii equal to the film thickness in Fig. 2 (no data exist at present for these confinements). One finds that the effect of $U(r)$ is less for cylindrical confinement. This result is somewhat surprising because the function given in Eq. 10 represents a stronger attraction for the helium than the function given by Eq. 7. What appears to be the case is that the $\frac{1}{r}\frac{\partial}{\partial r}$ operator gives an overall smaller curvature to the order parameter, thus it does not vanish quite as rapidly as in the planar case. Finally, we note that if one were to correct experimental data^{7,18} for the effect of $U(r)$, one would have to *increase* the measured value of ρ_s/ρ_{sb} . This would make the disagreement with scaling even worse.

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