

# Fluctuation induced thinning of superfluid films: Confirmation of finite size scaling

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- Introduction
  - Analogy between quantum and critical Casimir effects
  - Superfluid and tri-critical point phase transitions in liquid helium
- Critical Casimir effect near the superfluid transition, confirmation of finite size effect
- Casimir effect near the tricritical point

# Casimir Force - due to fluctuations

Specifically confinement of quantum fluctuations of the electromagnetic field



Not allowed because  
 $E=0$  inside metal



H. G. B. Casimir, Proc. K. Ned.  
Acad. Wet. 51, 793 (1948).

From QM, all fields have  
fluctuations even at  $T = 0$ ,

$$\mathcal{E} = \sum \frac{1}{2} \hbar \omega_n \rightarrow \infty$$

Since  $E=0$  at boundary,

$$\omega_n = \omega_n(d) = 2\pi c / \lambda_n$$

where  $\lambda_n = 2d/n$

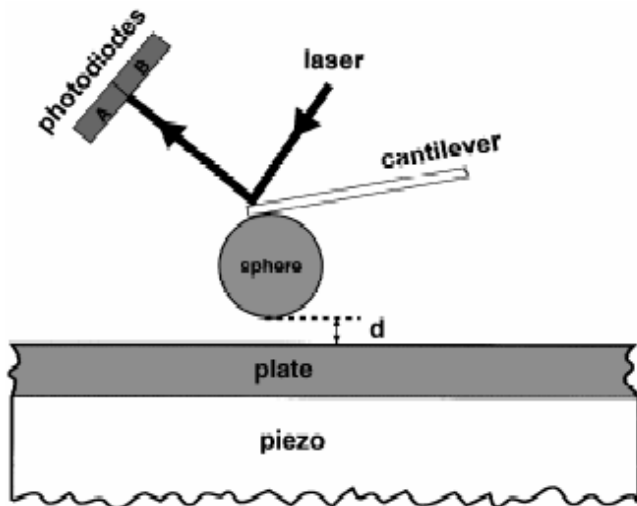
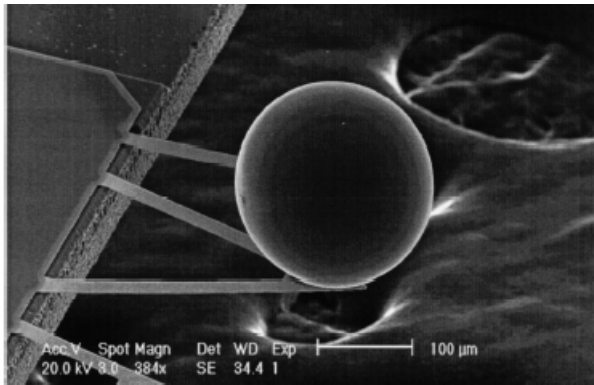
Thus, force =  $-\partial \mathcal{E}(d) / \partial d \neq 0$

→ force =  $-A\pi\hbar c / 480d^4$   
attractive!

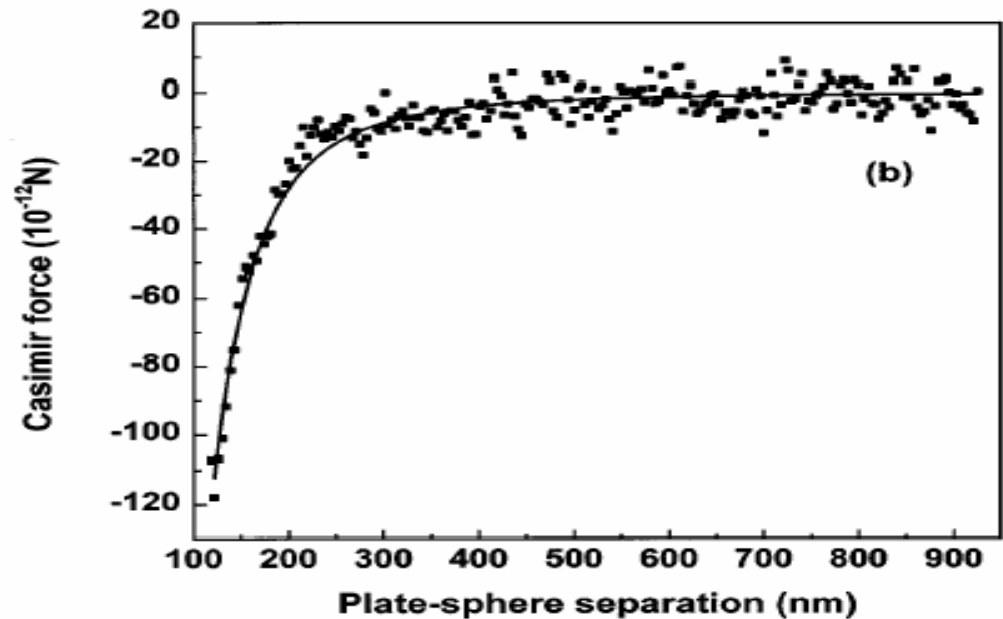
# Experimentally confirmed recently

S. Lamoreaux, Phys. Rev. Lett. 78, 5 (1997); U. Mohideen and A. Roy., PRL 81, 4549 (1998)

H. B. Chen et al., Science 291, 1941 (2001); Bressi et al., PRL, 88, 041804 (2002)



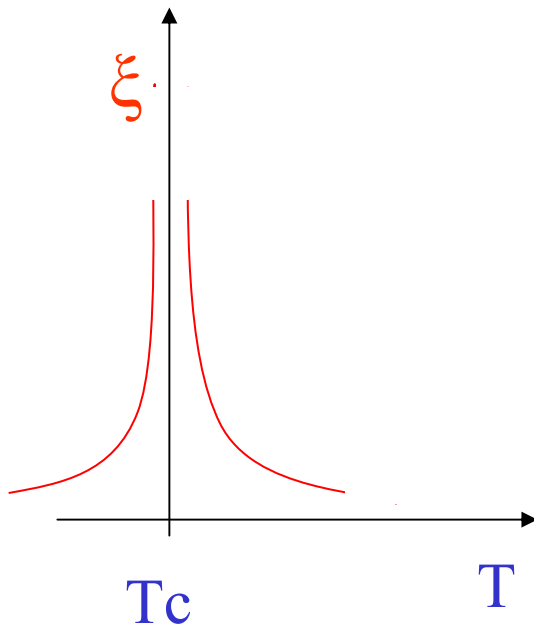
Between sphere and plate:  
Force =  $-\frac{2\pi R}{3} \frac{\pi \hbar c}{480d^4}$



Near a critical point, the thermodynamic behavior of the system is dominated by the fluctuations of the "order parameter".

The spatial extent of this "critical fluctuation" is known as the correlation length,  $\xi$

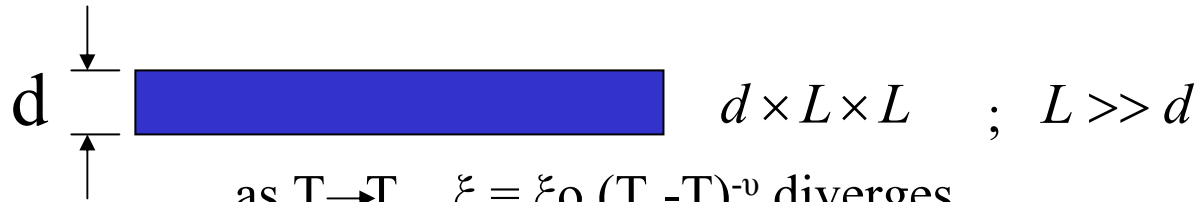
$$\xi = \xi_0 (T_c - T)^{-\nu}$$



As  $\xi$  becomes much larger than the atomic spacing, "chemistry" is no longer important; only the **dimension** of the system and the **symmetry** of the order parameter matters

Are there consequences if we limit the spatial extent of the fluctuation?

# Origin of Critical Casimir Effect



as  $T \rightarrow T_c$ ,  $\xi = \xi_0 (T_c - T)^{-\nu}$  diverges

and when  $\xi \geq d$ , critical fluctuation of length scale larger than  $d$  is cutoff in one direction

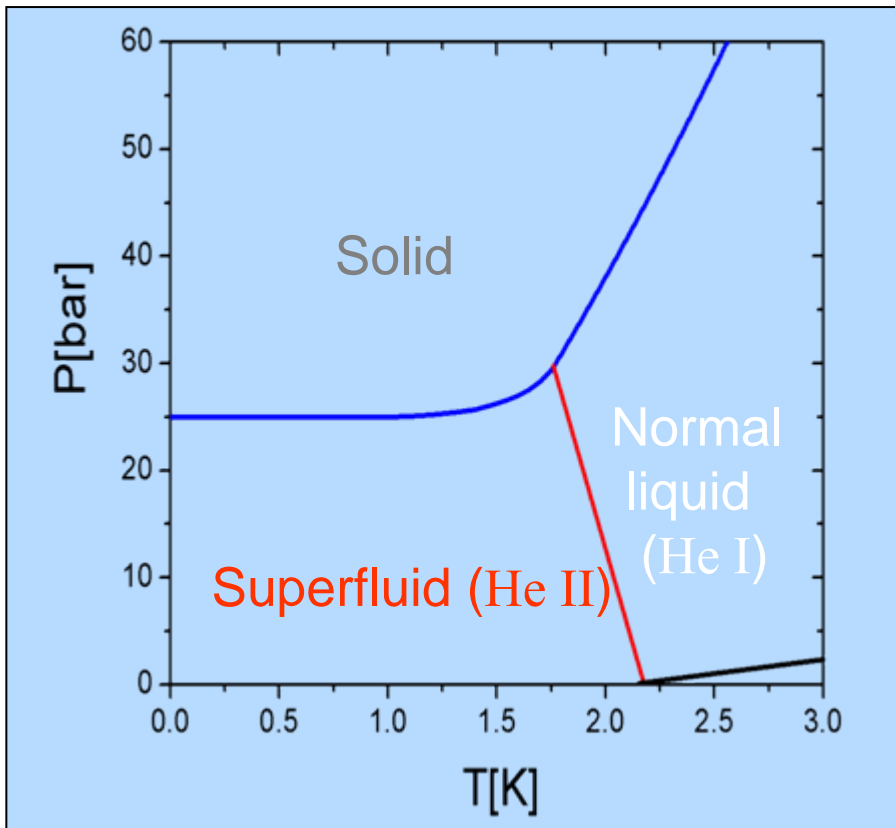
**Question: Is there a Critical Casimir effect?**

shown by Fisher & de Gennes (1978) that the change in free energy due to this cut-off is:

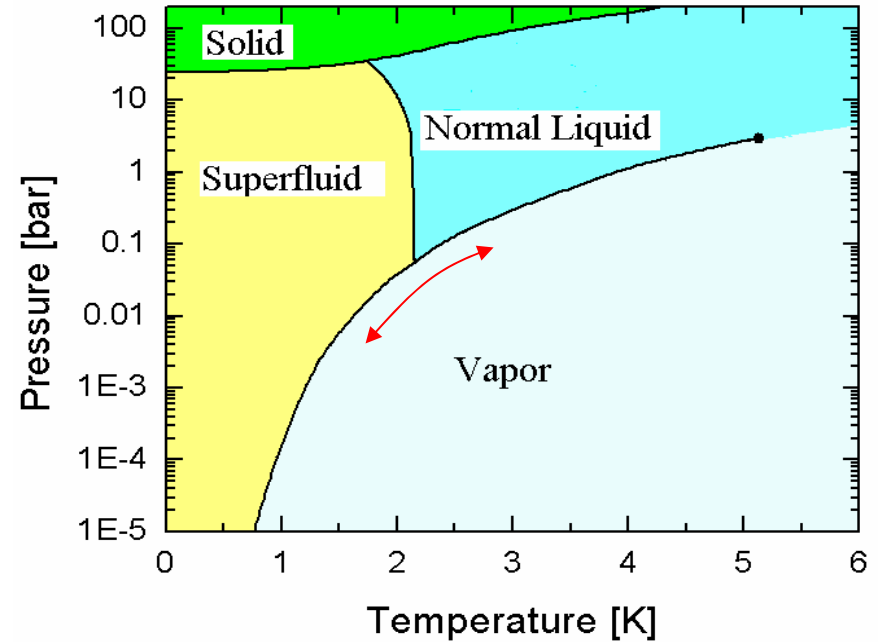
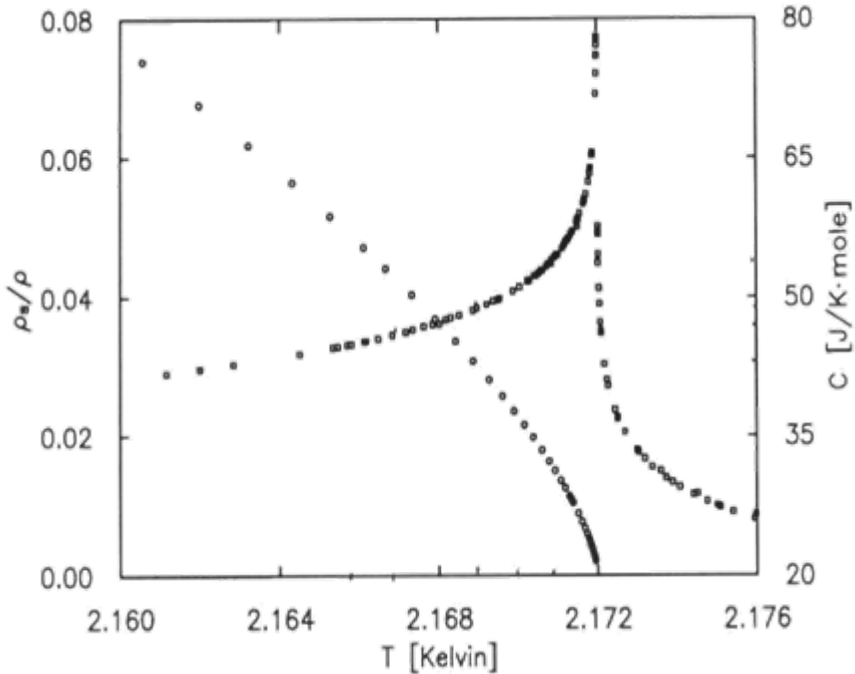
$$f(d, \xi) = \frac{\text{Fluctuation Energy}}{\text{Unit Area}} = \frac{k_b T_c}{d^2} \Theta\left(\frac{d}{\xi}\right)$$

$\Theta\left(\frac{d}{\xi}\right)$  is dependent on the specific universality class and on the boundary condition of the order parameter at the confining interfaces

# Phase diagram of $^4\text{He}$



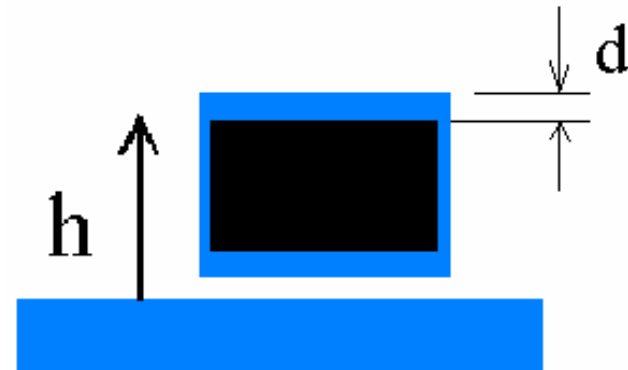
# Superfluid Transition in $^4\text{He}$



$$\Psi^* \Psi = \rho_s(t) = \rho_{s0} t^v ;$$

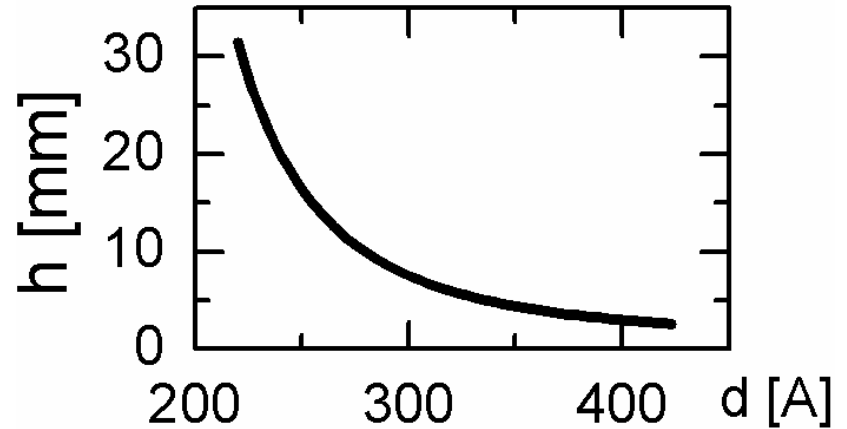
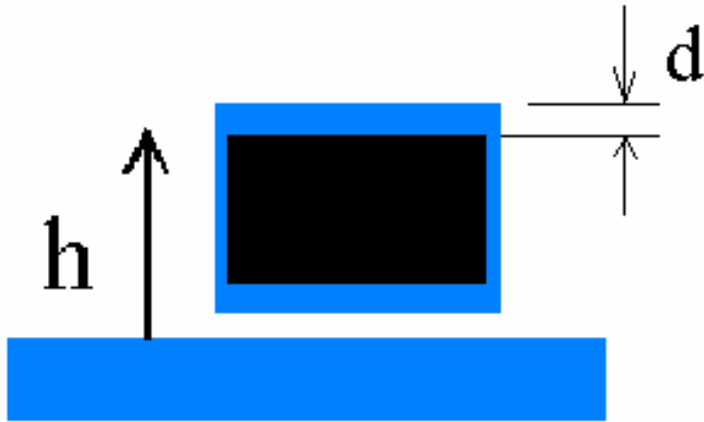
$$\xi = \xi_0 t^{-\nu} = (m^2 k_B T) / (\hbar^2 \rho_s(t))$$

$$t = (T_\lambda - T) / T_\lambda \quad ; \quad \nu = 0.67$$





# Physical adsorption of helium on a solid substrate



$$\frac{\gamma}{d^3(1+d/\lambda)} = mgh$$

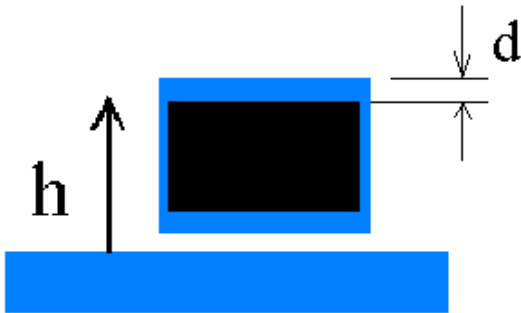
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Van der Waal's
Gravity

I.E. Dzyaloshinskii et al., Adv. Phys. 10, 165 (1961);

E. Cheng and M. Cole, Phys. Rev. B 38, 987 (1998)

# Contribution due to Casimir effect




$$f(d, \xi) = \frac{\text{Fluctuation Energy}}{\text{Unit Area}} = \frac{k_b T_c}{d^2} \Theta\left(\frac{d}{\xi}\right)$$

$$\mu = \frac{\partial Af}{\partial N} = \frac{\bar{V}}{A} \frac{\partial Af}{\partial d} = \frac{\bar{V} k_B T_c}{d^3} \mathcal{G}\left(\frac{d}{\xi}\right)$$

$N$  = No. of helium atoms =  $Ad/V$

$\bar{V}$  = atomic volume of liquid  ${}^4\text{He}$  =  $46(\text{\AA})^3/\text{atom}$

Equilibrium film thickness at height  $h$  above bulk is determined by competition between van der Waals, gravity, and critical Casimir forces

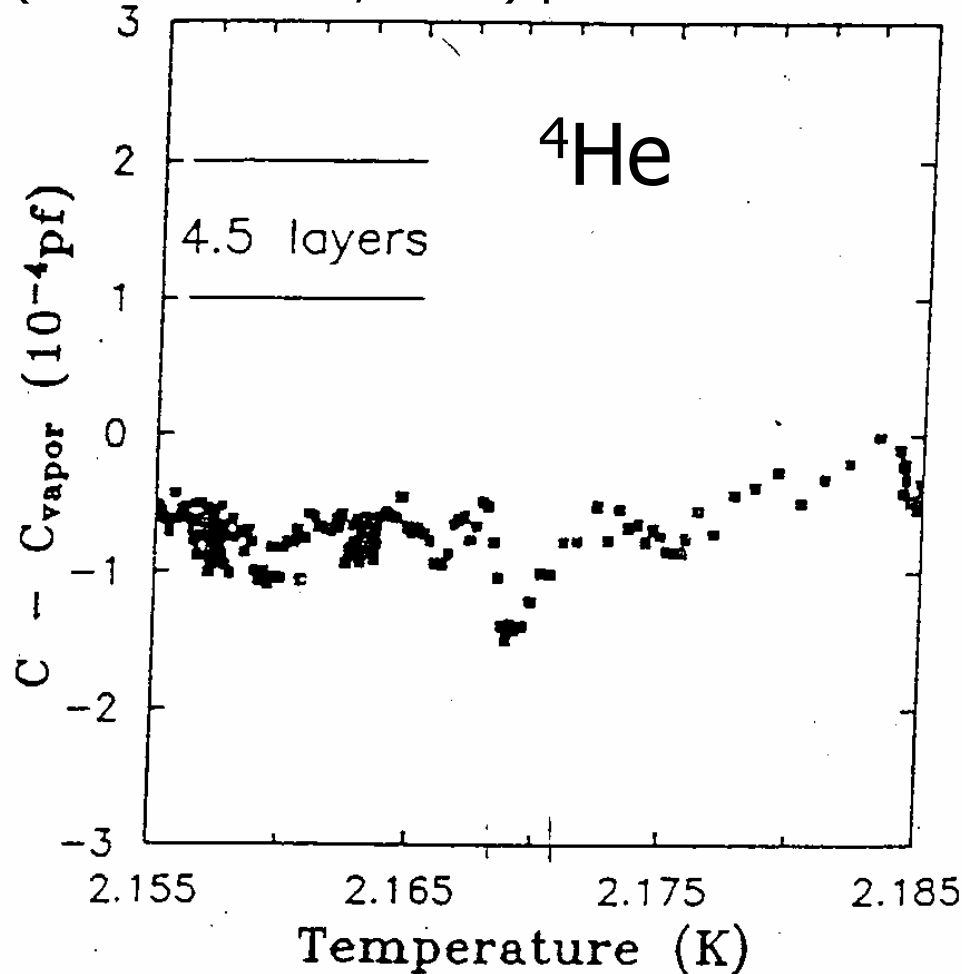


$$\frac{\gamma}{d^3(1+d/\lambda)} + \frac{k_B T_C \bar{V} \mathfrak{G}(d/\xi)}{d^3} = mgh$$

Van der Waals
Critical Casimir
gravity

# Dip in film thickness previously observed

R.J. Dionne and R.B. Hallock, AIP Conf. Proc. No 194  
(AIP New York, 1989) p.199.



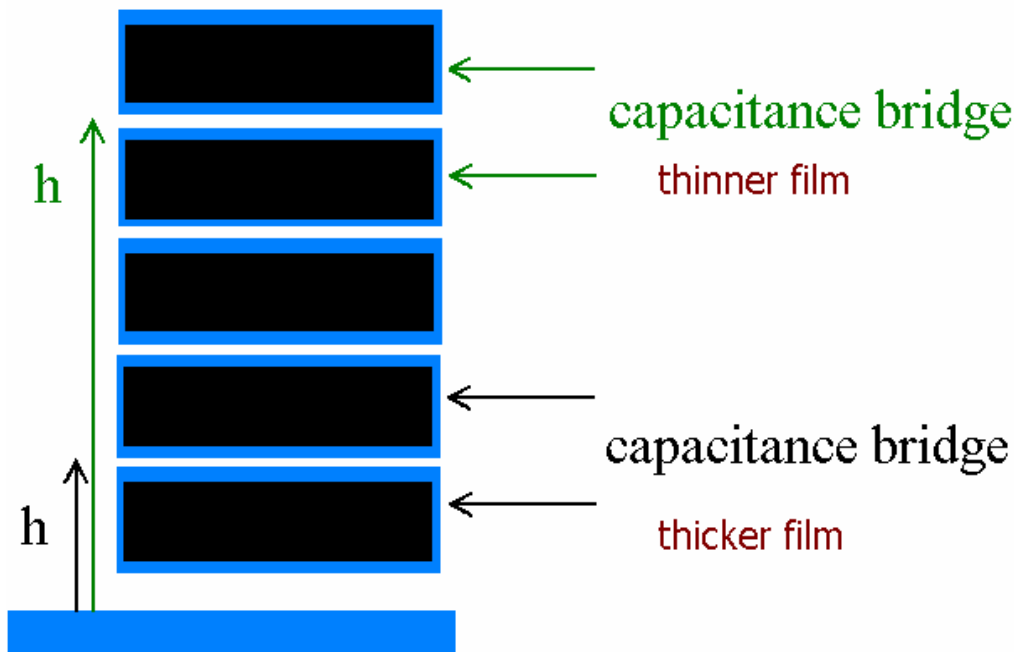
- It was not entirely clear that the dip in the film thickness was due to Casimir force



# Crucial Test:

## Does thinning depend on $d/\xi$ ?

$$\frac{\gamma}{d^3(1+d/\lambda)} + \frac{k_B T_C \bar{V} \vartheta(d/\xi)}{d^3} = mgh$$



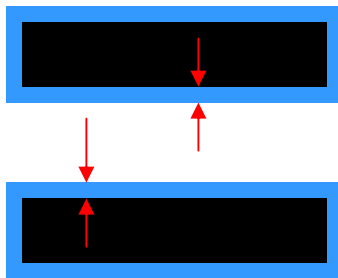
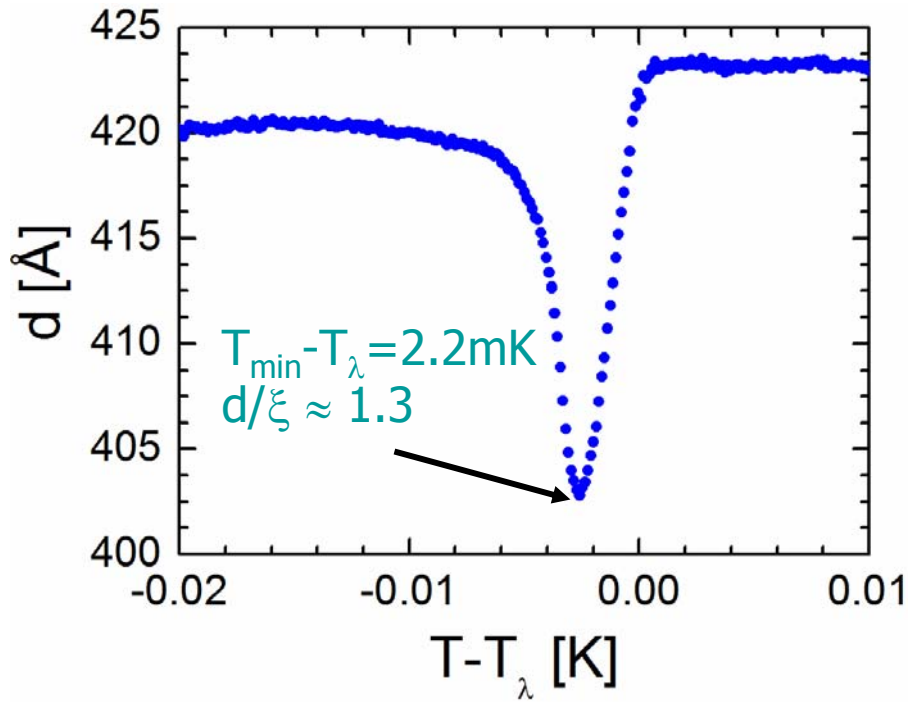
Capacitances in Series

$$\frac{1}{C_{\text{meas}}} = \frac{1}{C_{\text{film}}} + \frac{1}{C_{\text{vapor}}}$$

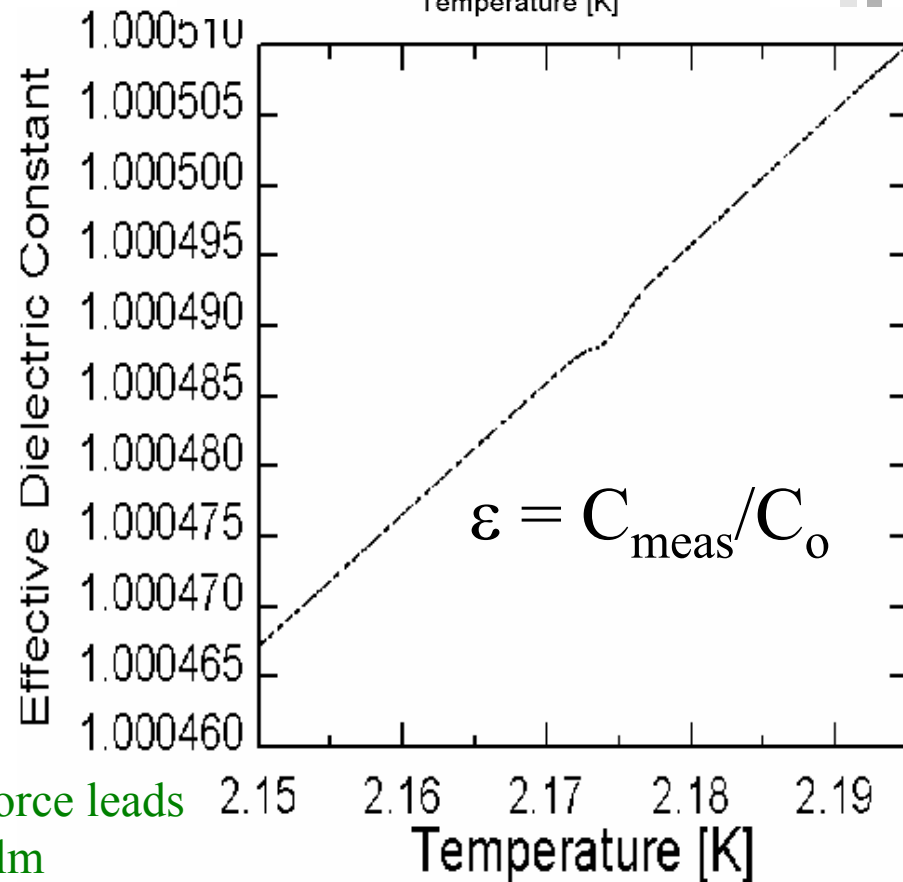
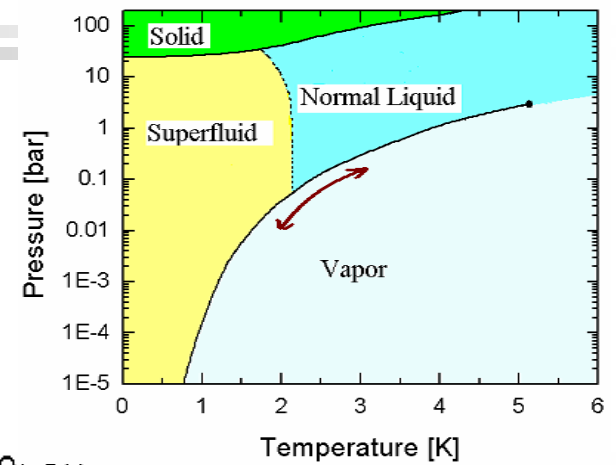
Six copper plates each 0.27cm thick and 1.7cm diameter; form 5 sets of capacitors with gaps of 0.2mm. The height  $h$  of the capacitor (gap) above the bulk liquid are 0.228, 0.516, 0.806, 1.091 and 1.382 cm.

- Change in capacitance  $C \Rightarrow$  change in Film Thickness  $d$ .

# Film Thickness



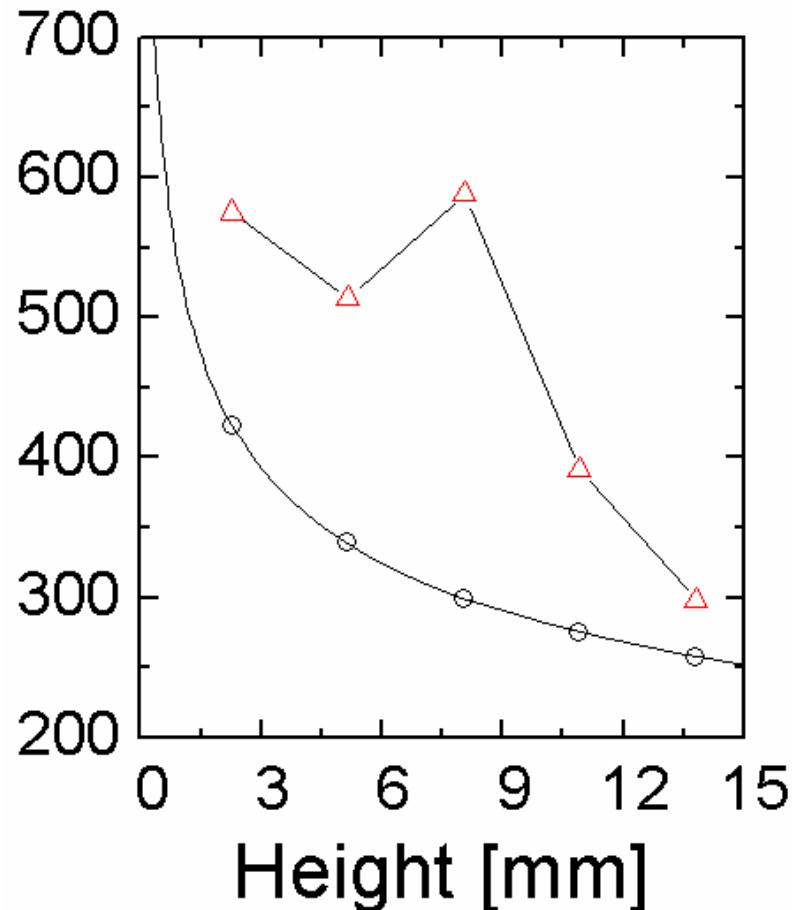
attractive Casimir force leads to thinning of the film

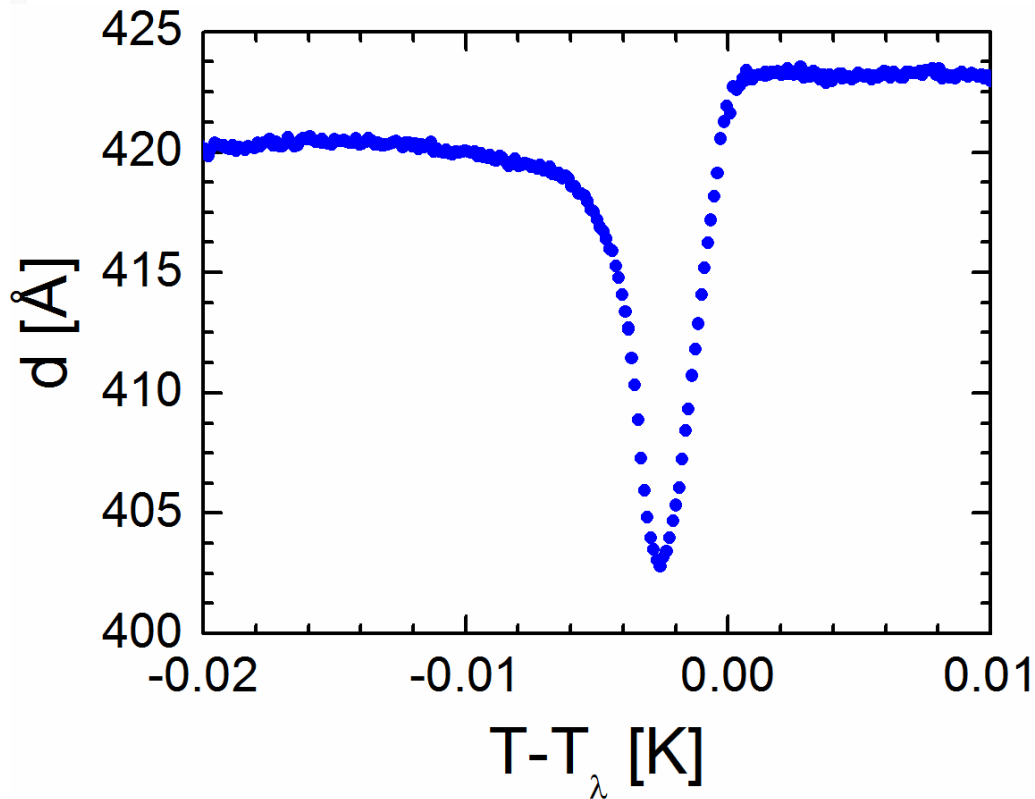


# Capacitance technique susceptible to inaccuracies in the background $d$

- Nevertheless, change in  $d$  measured very accurately.

Theoretically Expected  $d$  vs.  
 $d$  Naively Calculated from  $\epsilon$





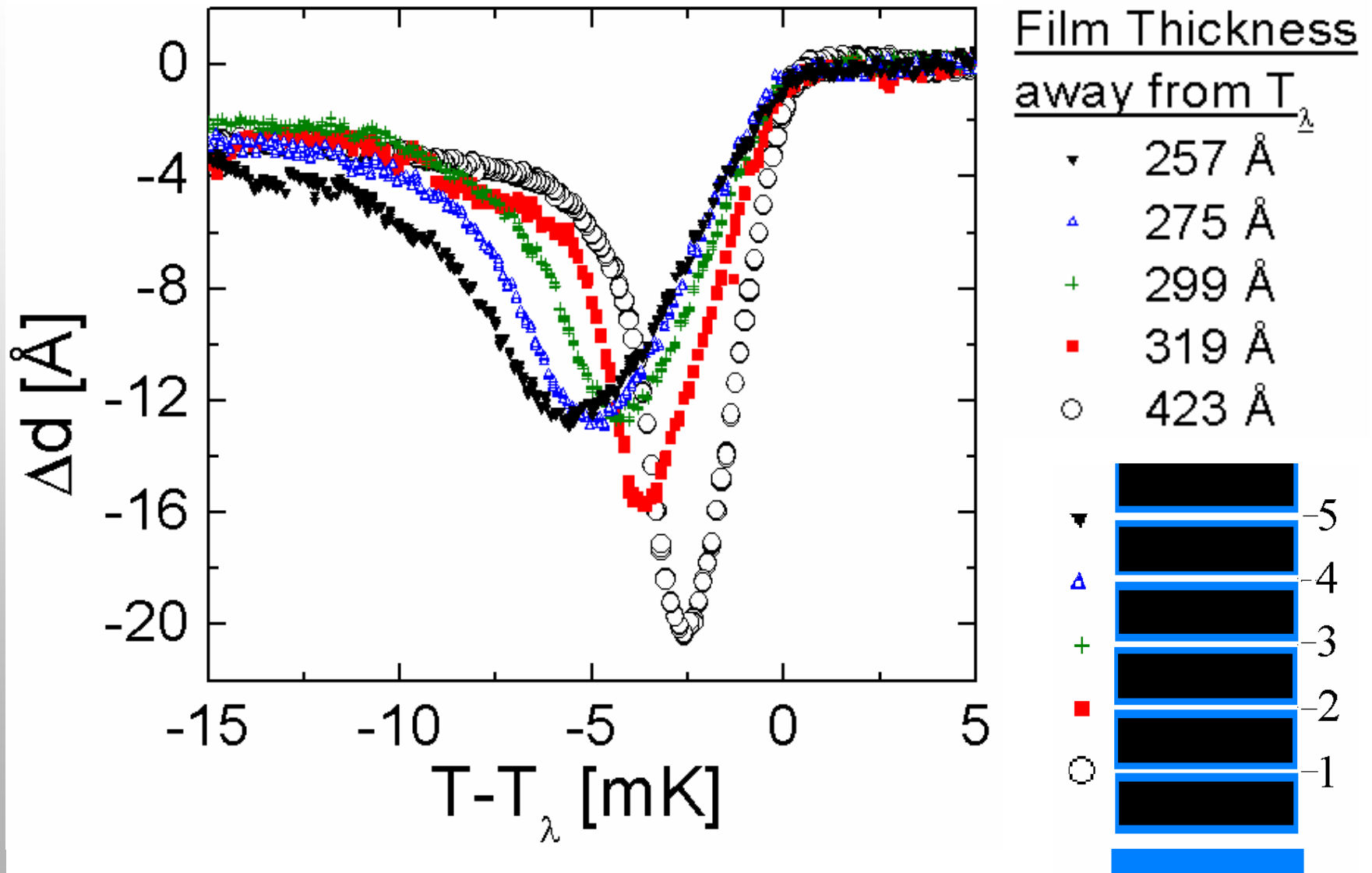
The film thickness in the superfluid phase is found to be thinner than that in the normal phase. This difference has been attributed to the Casimir force resulting from Goldstone modes and surface fluctuations of superfluid

M. Kardar and R. Golestanian, Rev. Mod. Phys. 71, 1233 (1999)

R. Zandi, J. Rudnick, and M. Kardar, PRL 93, 155302 (2005)

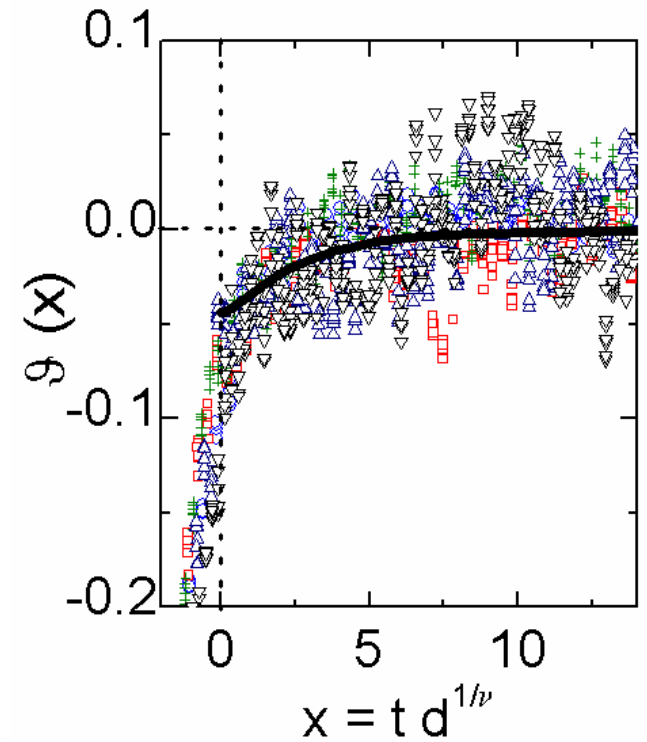
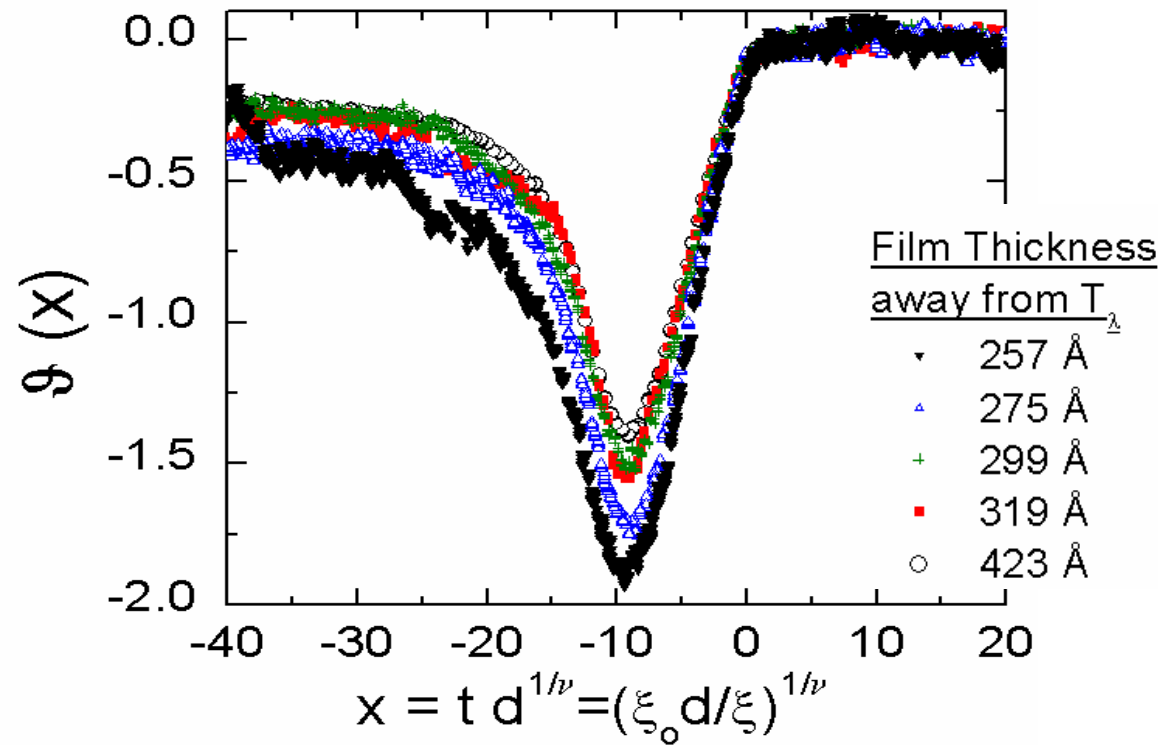


# Film Thickness $d(T)$ calc. from $\epsilon(T)$



# Scaling Function $\vartheta(x)$

$$\frac{\gamma}{d^3(1+d/\lambda)} + \frac{k_B T_C \bar{V} \vartheta(d/\xi)}{d^3} = mgh$$



The minima of the scaling functions appear at the same  $x$  (good!) ; but the scaling functions do not collapse. Is this real or an artifact ?

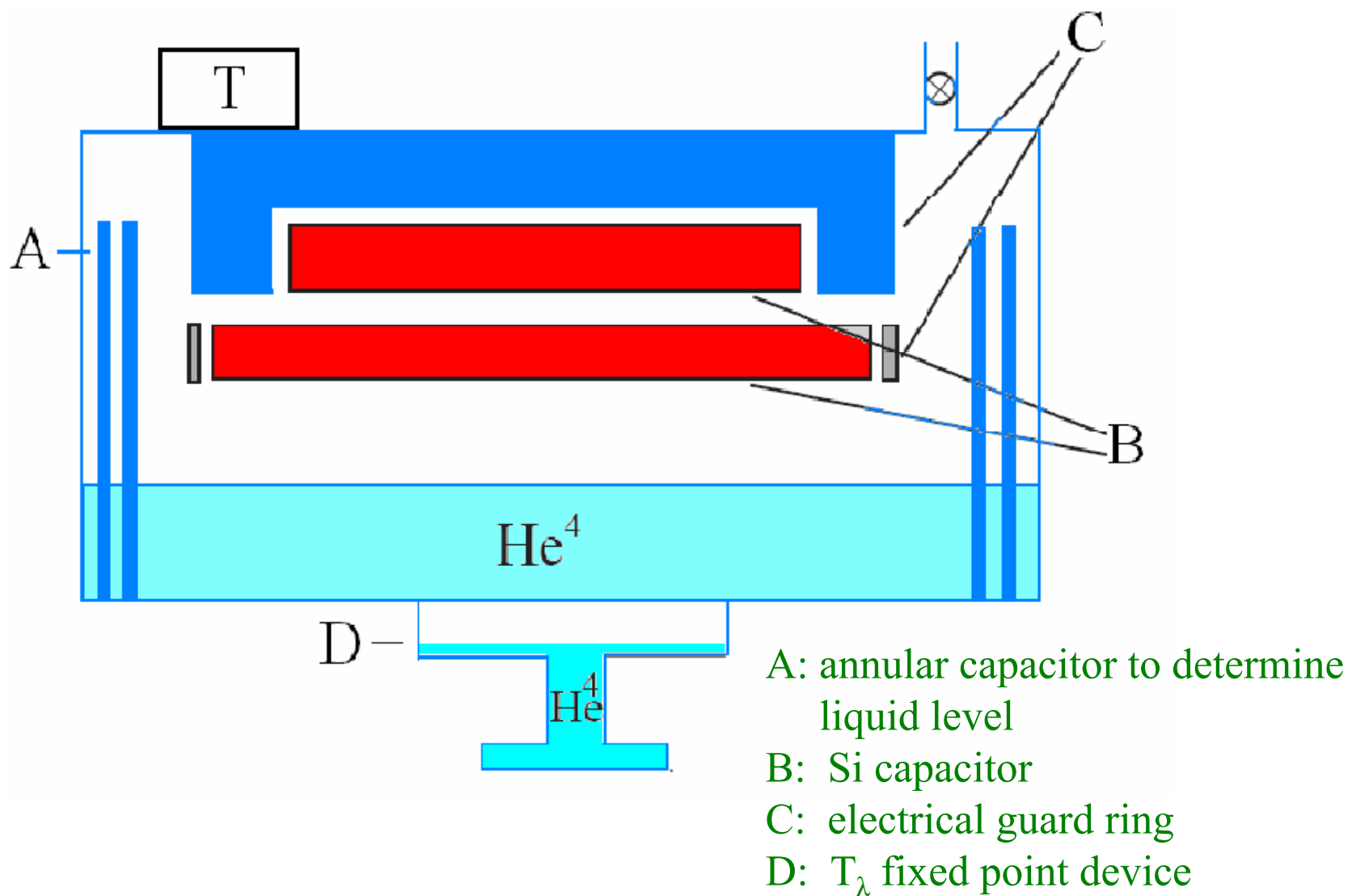
Garcia and Chan, PRL 83, 1187 (1999)

Comparison with theoretical (Krech and Dietrich) scaling function for  $T \geq T_\lambda$

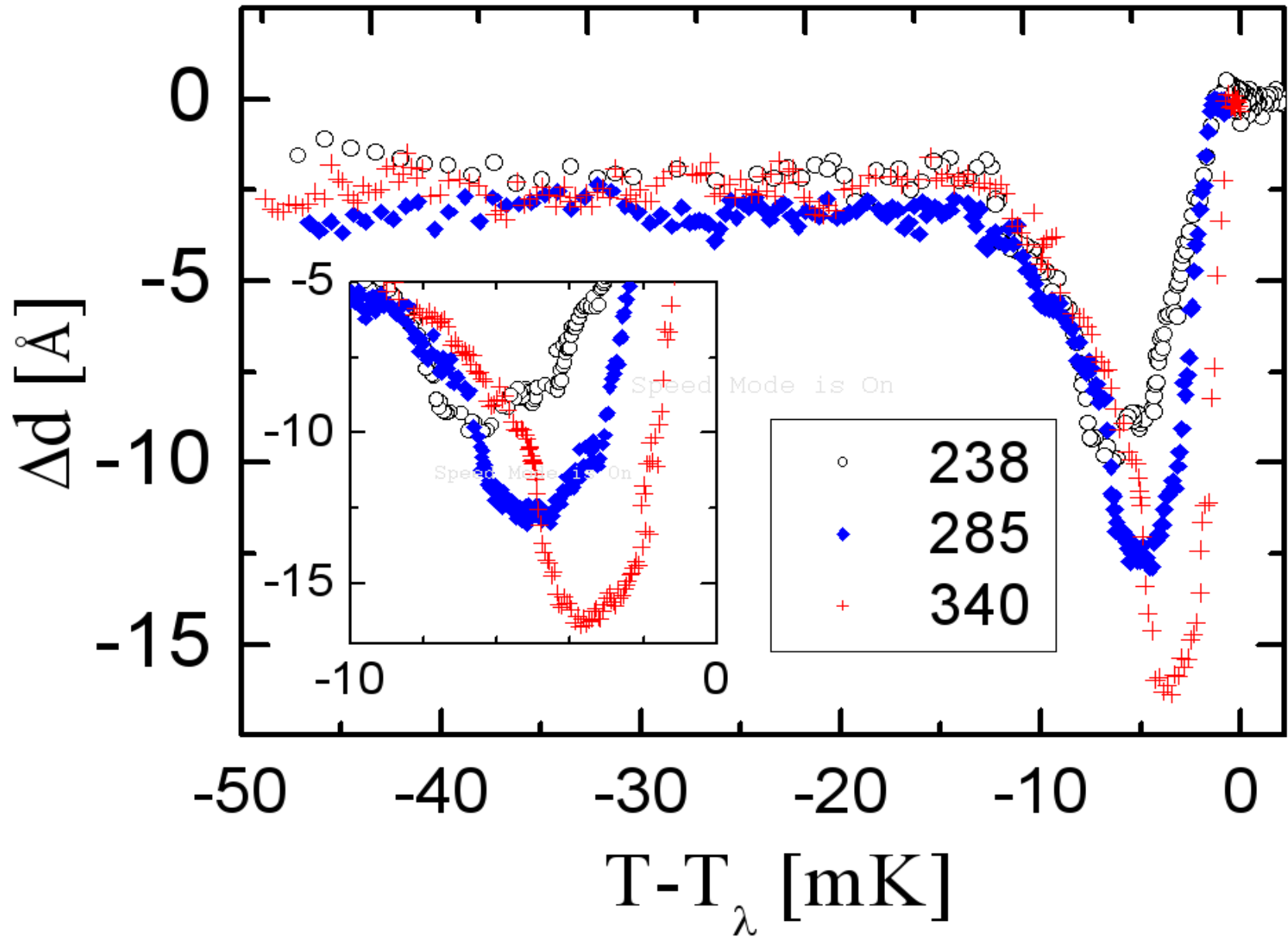
The surface of the copper electrodes is less than ideal ( rms roughness is 10 nm) and there are scratches and foreign particles.

New experiment employs doped silicon electrodes assembled in clean room. Surface roughness is  $\sim 0.8\text{nm}$ .

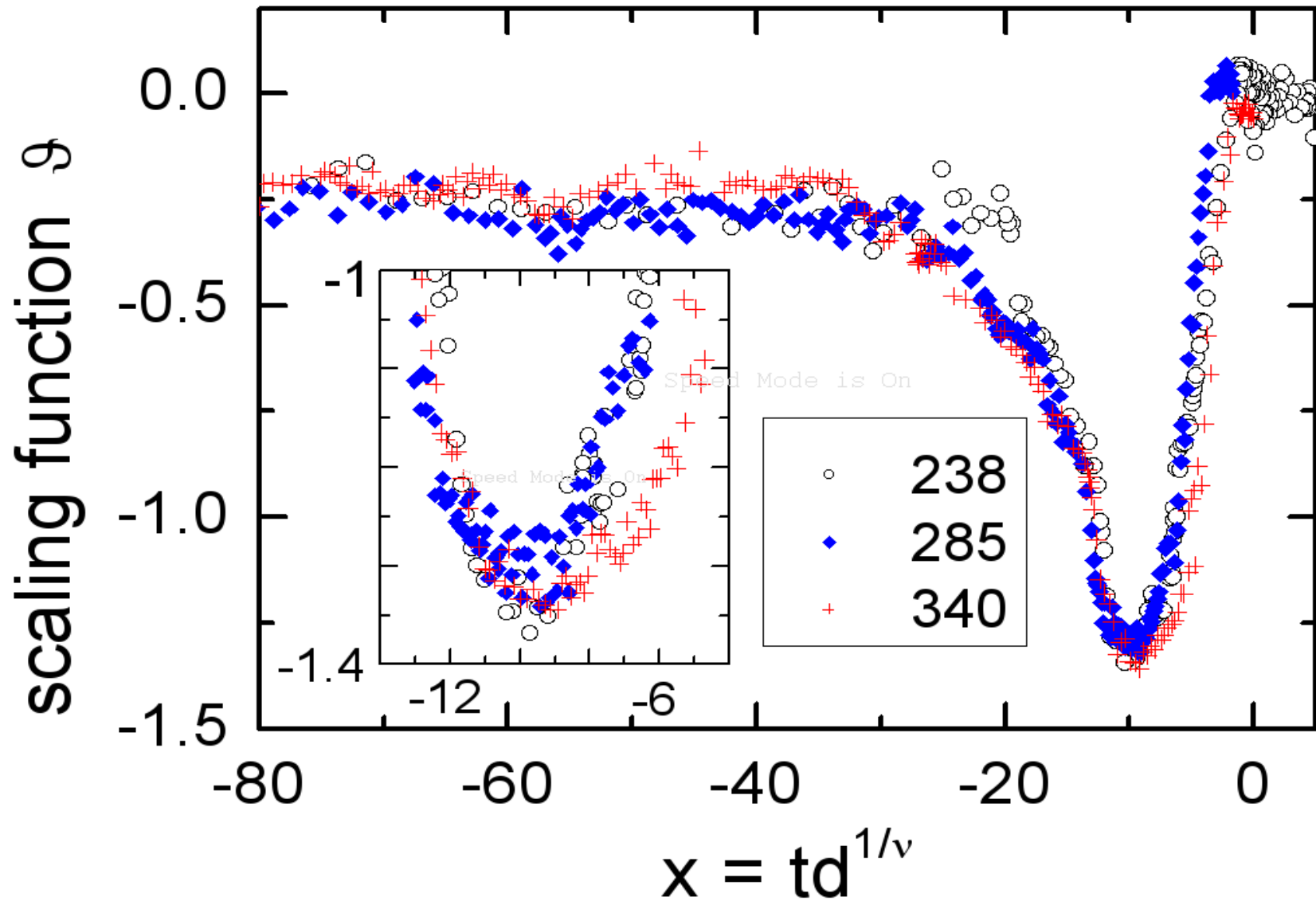
# New experiment with atomically flat silicon substrate



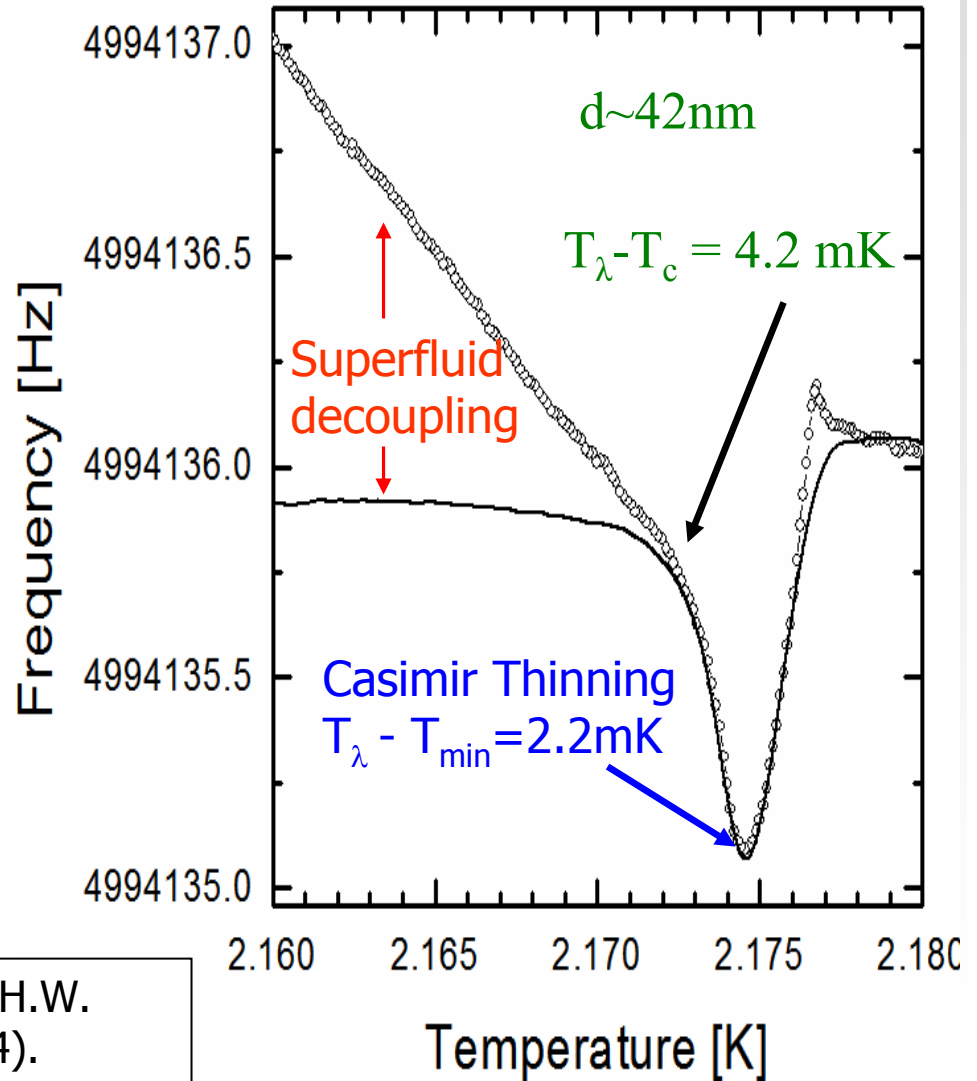
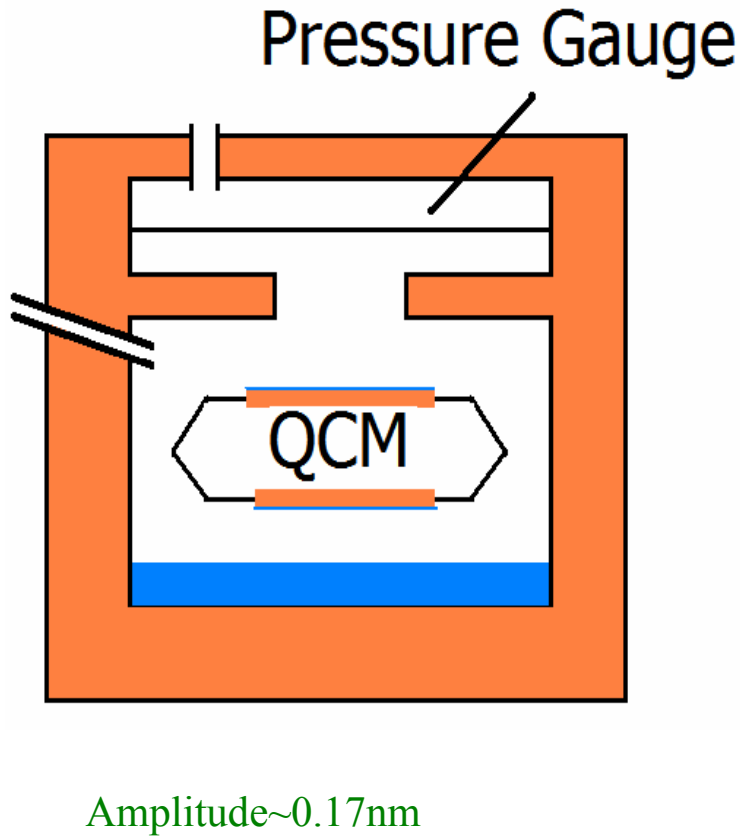
# Film Thickness $d(T)$ calc. from $\epsilon(T)$



$$\frac{\gamma}{d^3(1+d/\lambda)} + \frac{k_B T_C \bar{V} \vartheta(d/\xi)}{d^3} = mgh$$



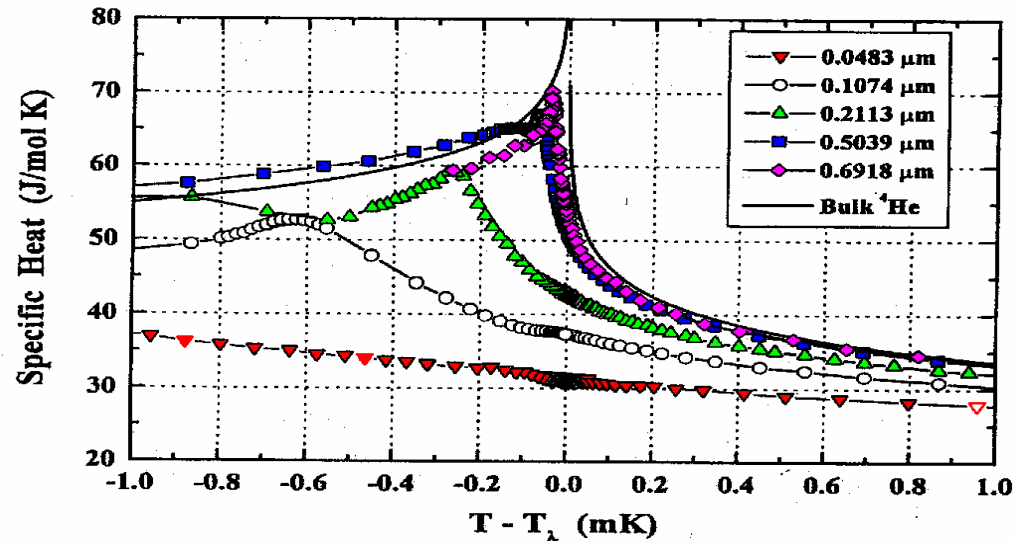
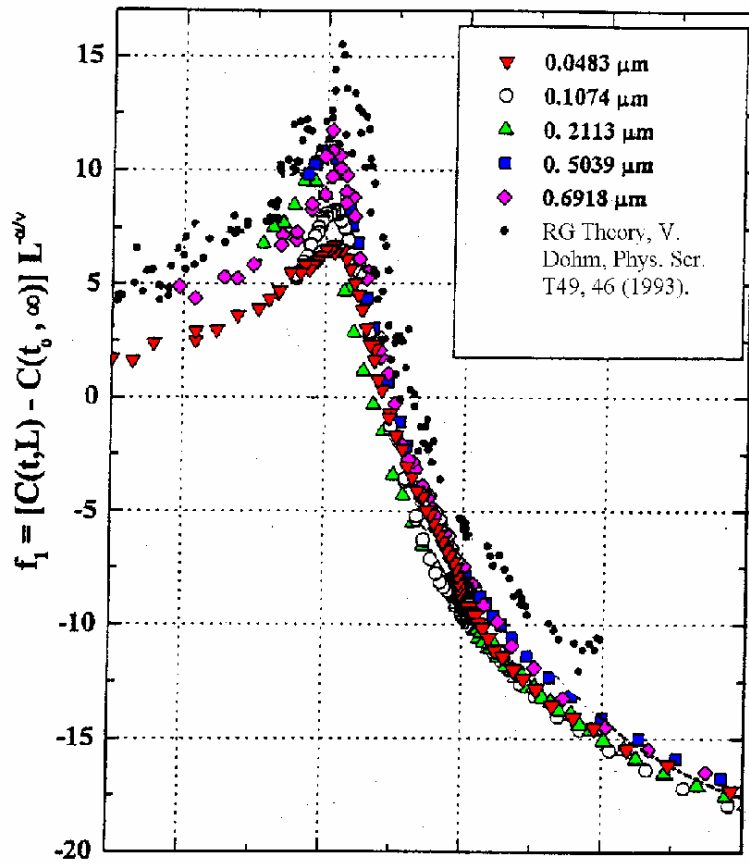
# Where is the superfluid transition temperature?



R. Garcia, S. Jordan, J. Lazzaretti, and M.H.W. Chan, J. Low Temp. Phys. **134**, 527(2004).

# Finite size scaling from specific heat of helium films

$$C_V = T^{-1} (\partial^2(F/V) / \partial T^2)_V$$



Gaspirini and students.

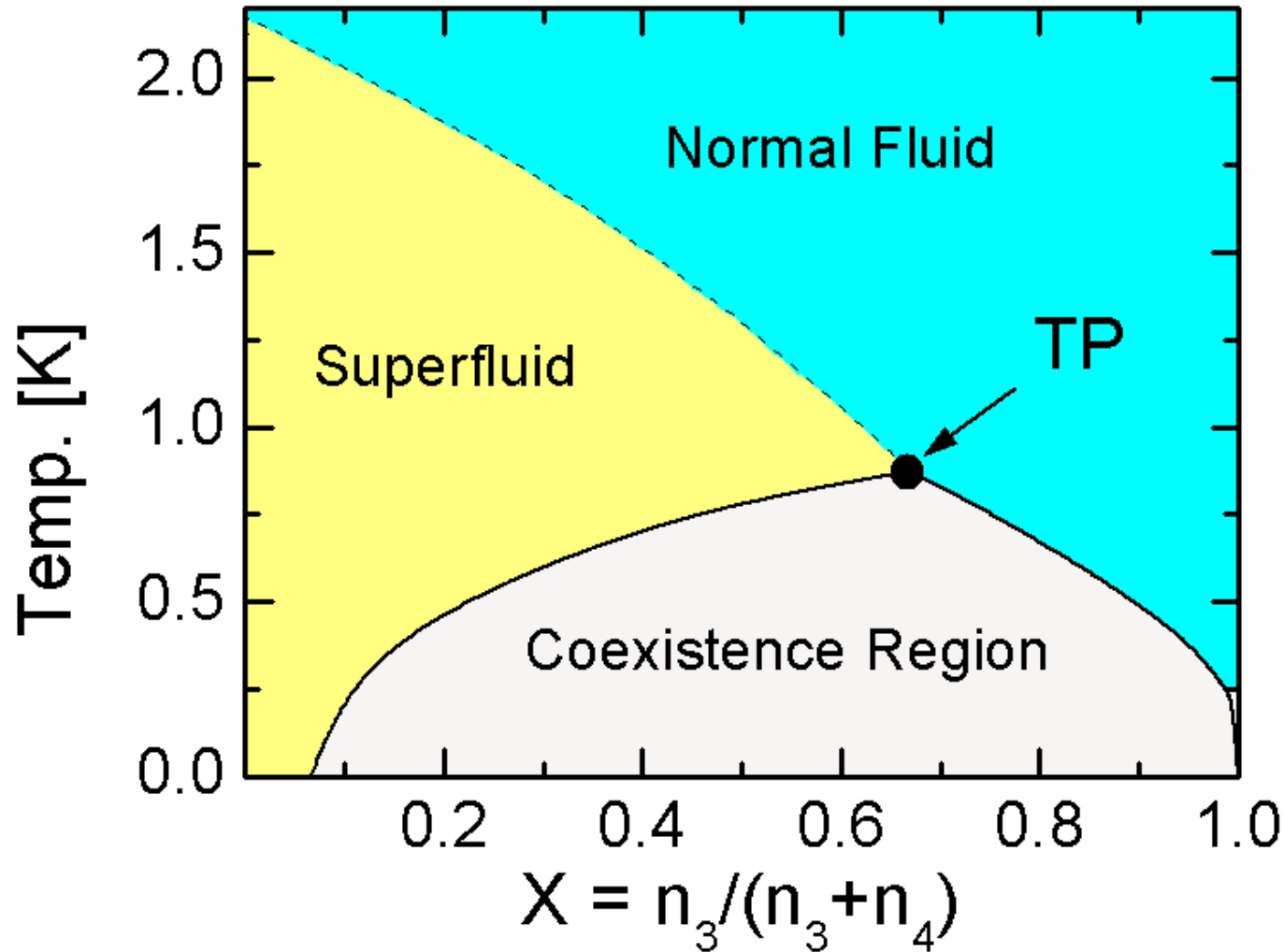
$$x = td^{1/\nu} = (\xi_0 d / \xi)^{1/\nu}$$



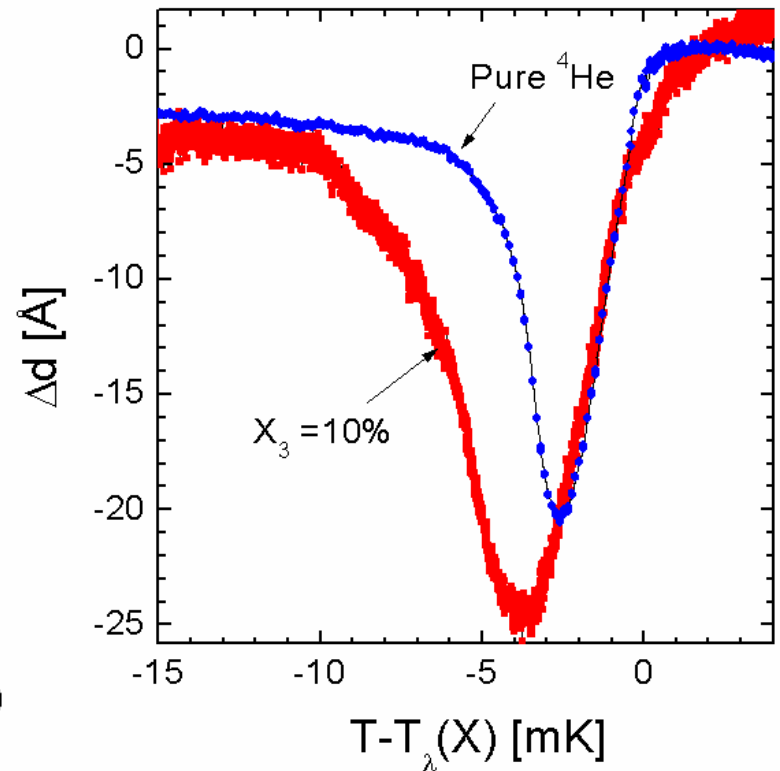
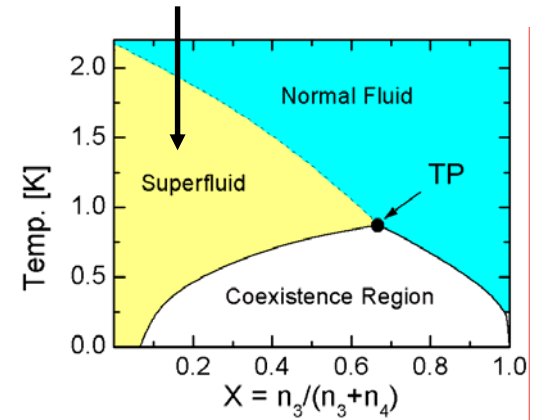
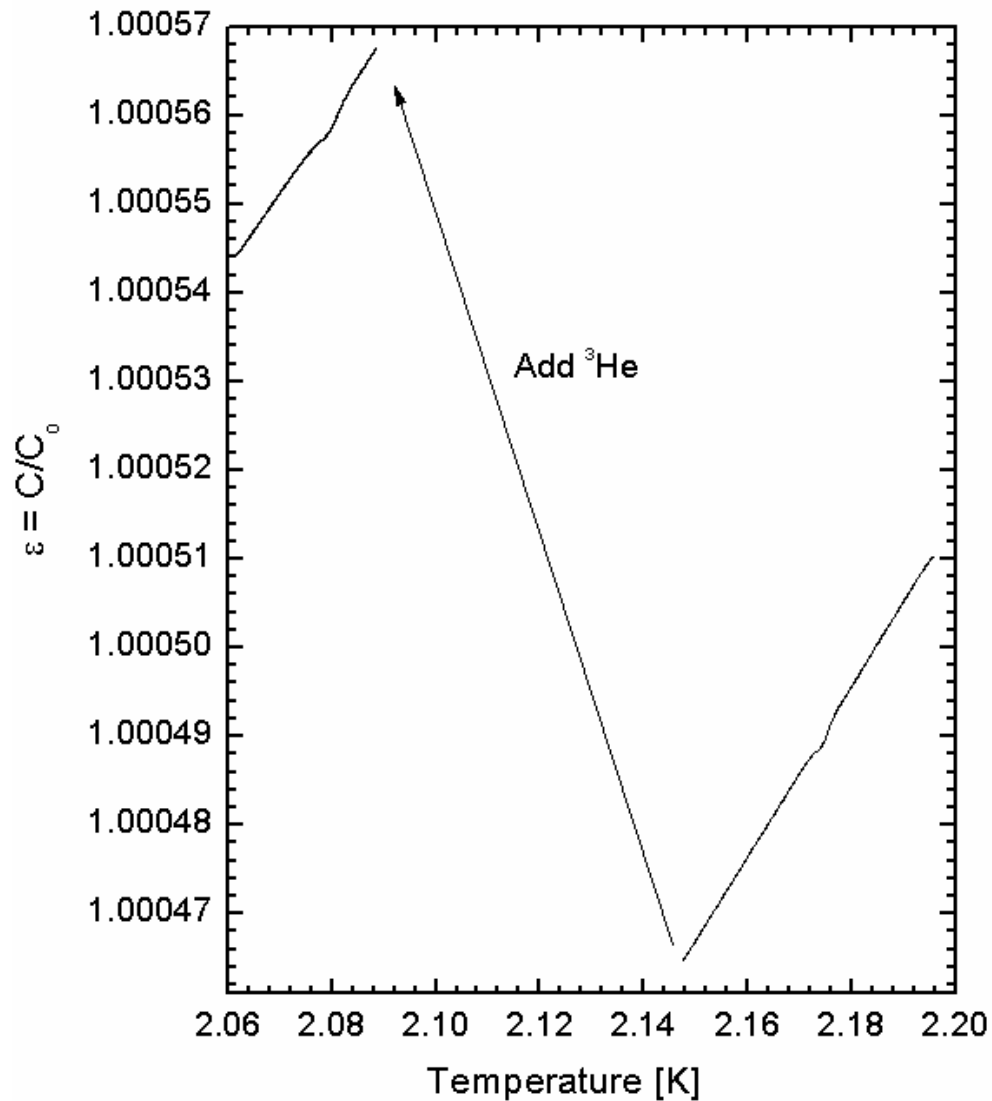
# Summary on superfluid film.

- 1) Critical Casimir effect, due to the spatial confinement of the fluctuating order parameter near the critical point is observed helium films near the lambda point.
- 2) The force near the superfluid transition is attractive, leading to a thinning of the adsorbed film. This indicates the superfluid order parameter of the adsorbed film satisfies Dirichlet boundary condition both at the solid and the vapor interfaces. Our experiment quantitatively confirm the prediction of finite size scaling.
- 3) The superfluid film is thinner than normal film due to Casimir force associated with Goldstone mode and surface fluctuations.  
[Golestanian and Kardar; Zandi, Rudnick and Kardar]
- 4) Recent Monte Carlo simulation by Hucht reproduces the observed scaling function.
- 5) The superfluid transition temperature occurs below the  $T$  where the minimum in film thickness is found.

# Casimir effect in $^3\text{He}$ - $^4\text{He}$ mixture

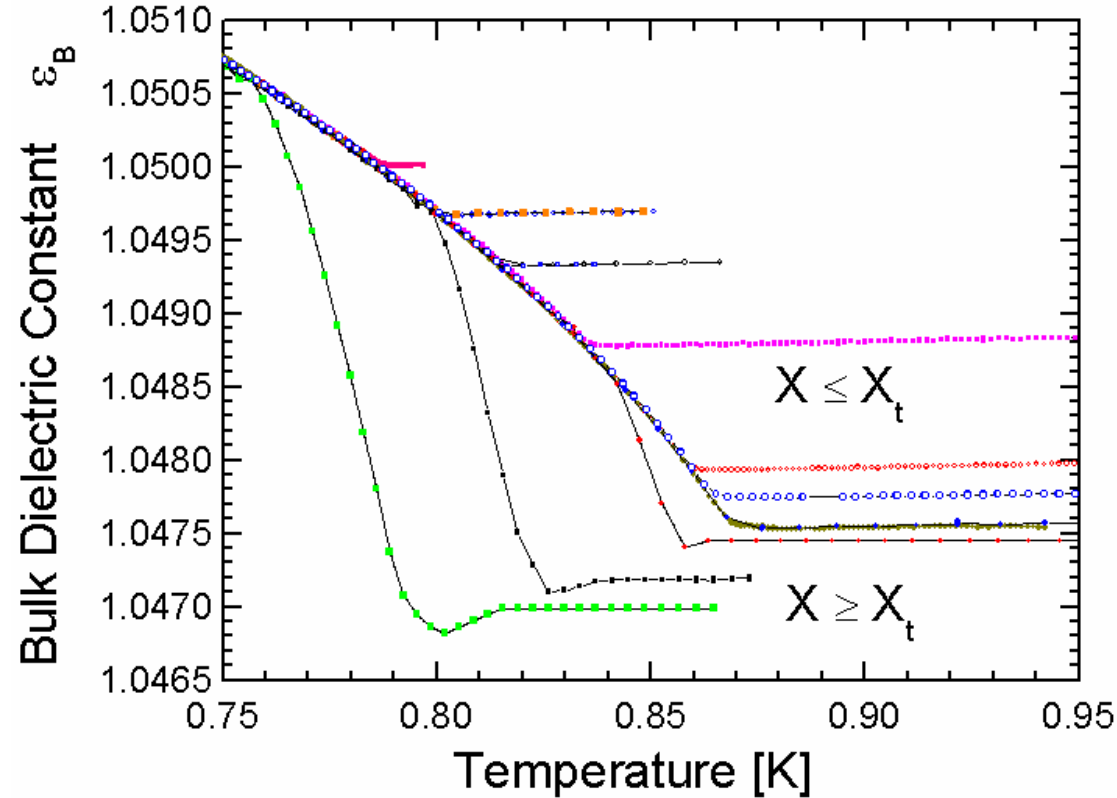
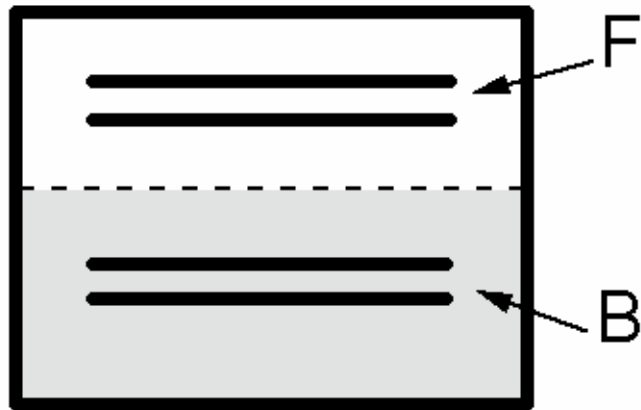
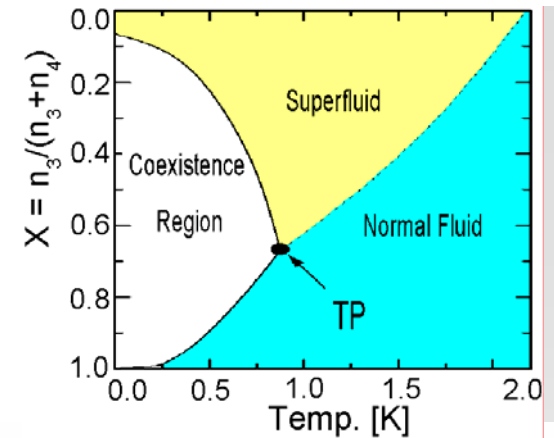


# Dilute Mixtures

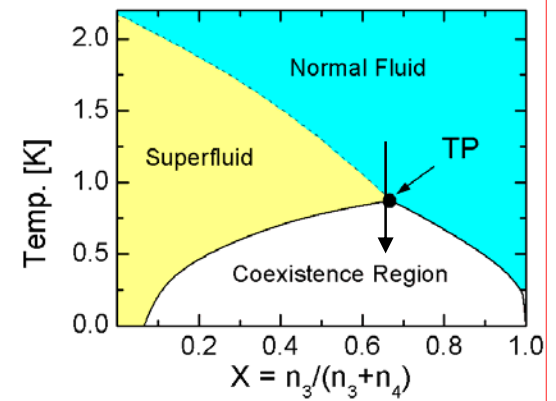
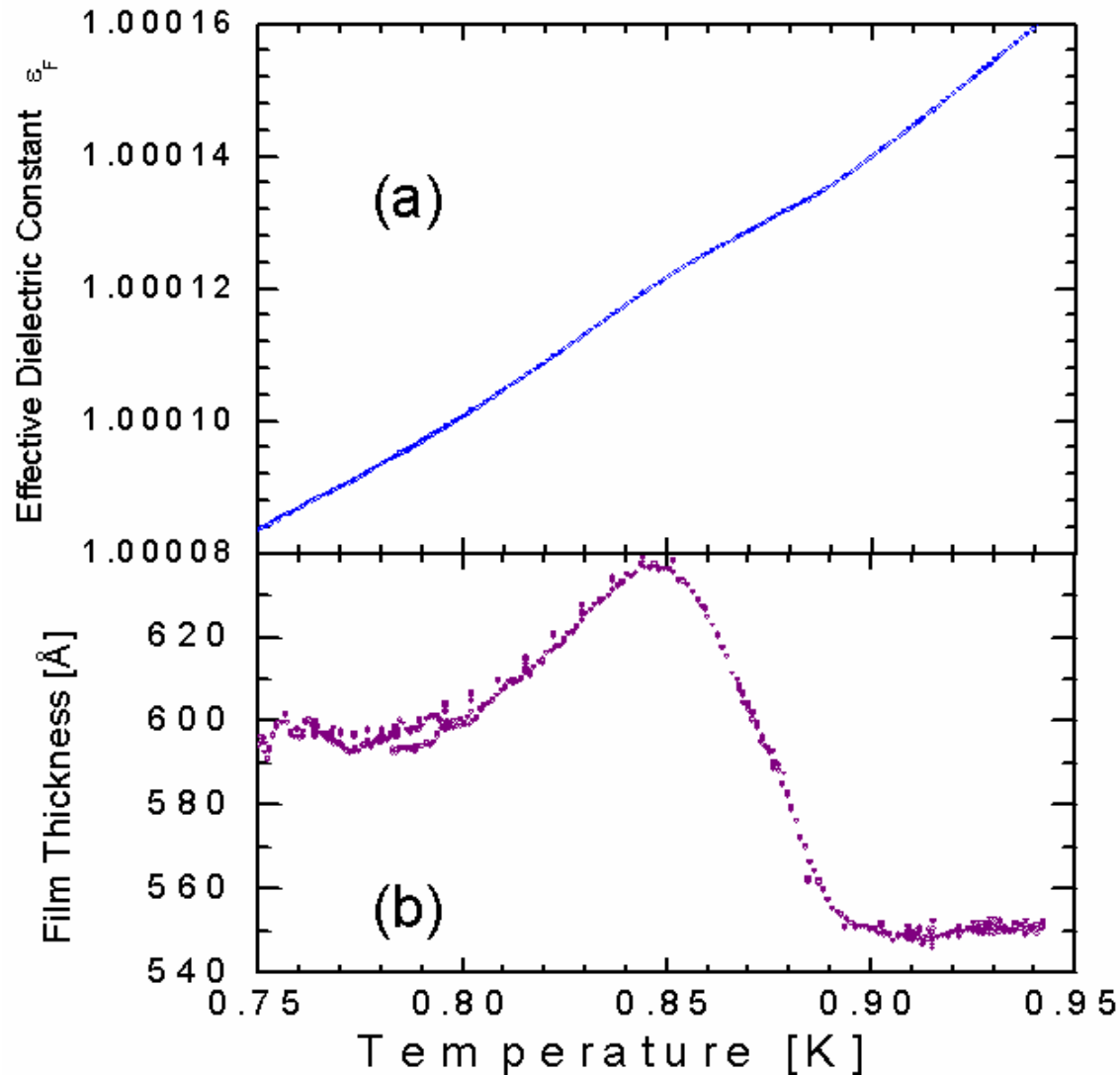


# Casimir effect near the tricritical point

- Diffusion of mixture components
- Need to determine tricritical point and phase separation temperature

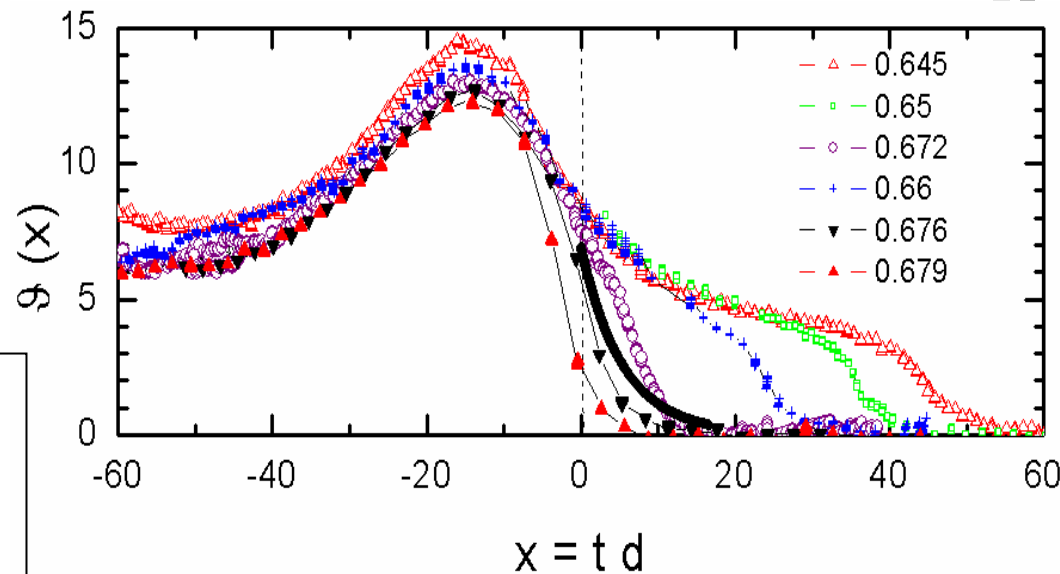
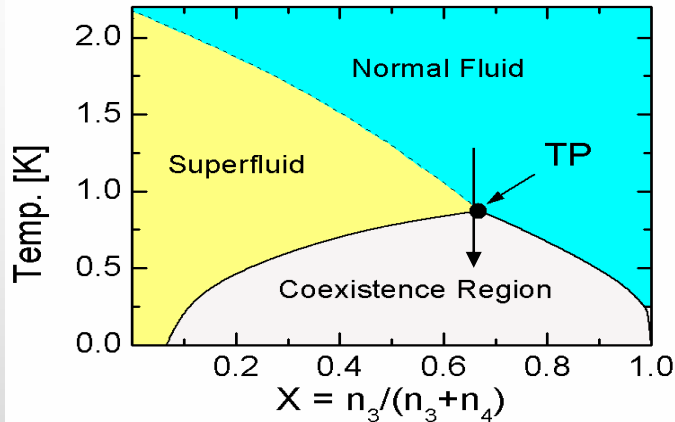


# $\epsilon(T)$ and $d(T)$ close to tricritical point



- The observed thickening is consistent with a repulsive critical Casimir force.

A repulsive Critical Casimir force ( $\vartheta > 0!$ ) is observed at the tricritical point!



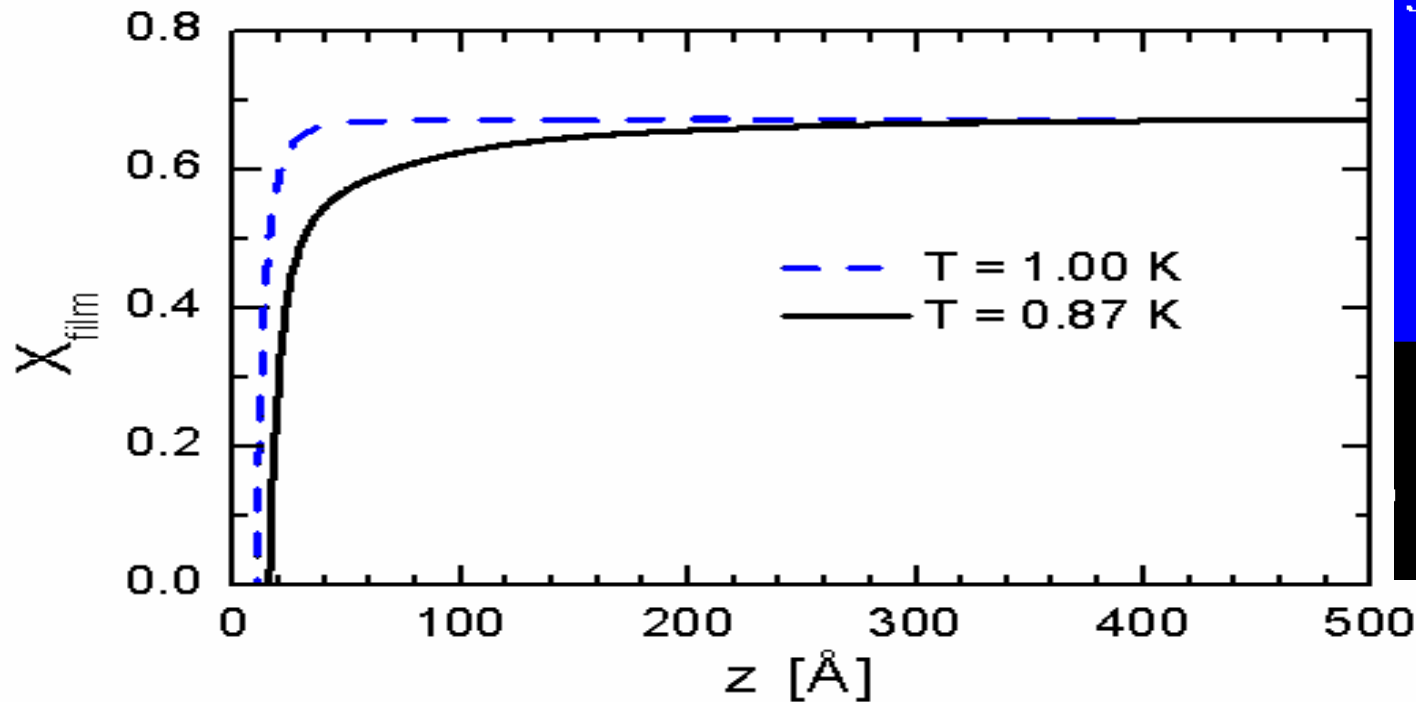
Repulsive force predicted by J. O.Indekeu, J. Chem. Soc. Faraday Transactions II 82, 1835 (1986); solid curve due to M. Krech.

More thorough study by A. Maciolek and Dietrich, Europhysics Lett. 74, 22(2006);

also see Maciolek, Gambassi and Dietrich, PRE 76, 031124(2007)

$$\frac{\gamma}{d^3(1+d/\lambda)} + \frac{k_B T_C \bar{V} \vartheta(d/\xi)}{d^3} = \bar{m}gh$$

# Concentration gradients in mixture films near tricritical point



$^3\text{He}$  same as bulk

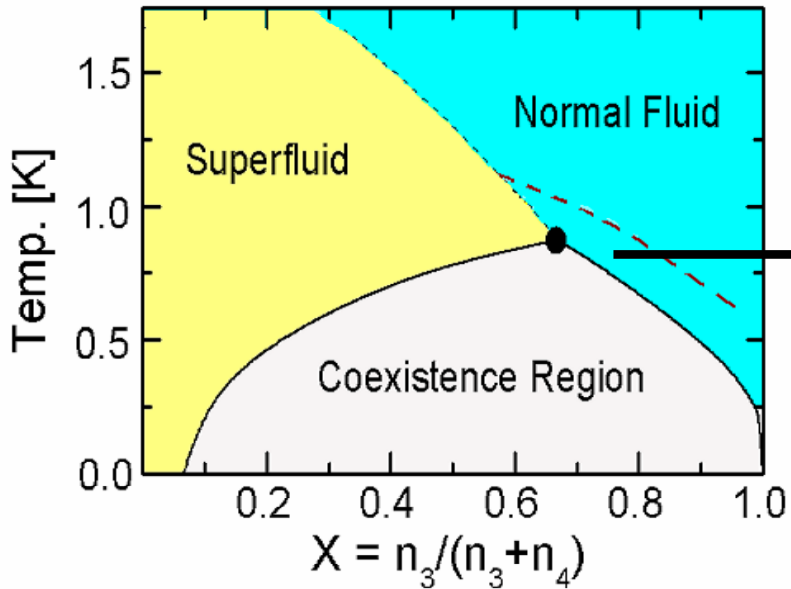
high  $^4\text{He}$

substrate

$$dX = \left( \frac{V_3}{\bar{V}} - 1 \right) \frac{du}{1 - X} \left( \frac{\partial X}{\partial \phi} \right)_{T,P}$$

G. Goellner et al., JLTTP 13,  
113 (1973)

# The tricritical point in $^3\text{He}$ - $^4\text{He}$

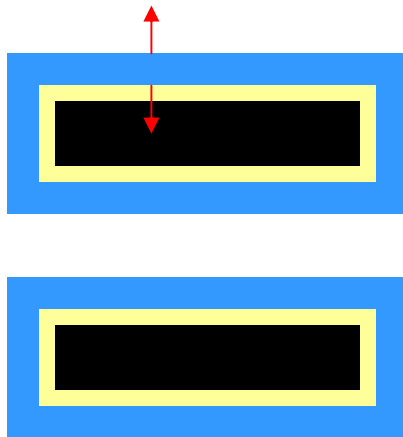


J.-P. Lahuerte et al., Phys. Rev. B 15, 4214 (1977)

Below this dotted line, a thin layer close to the substrate is superfluid

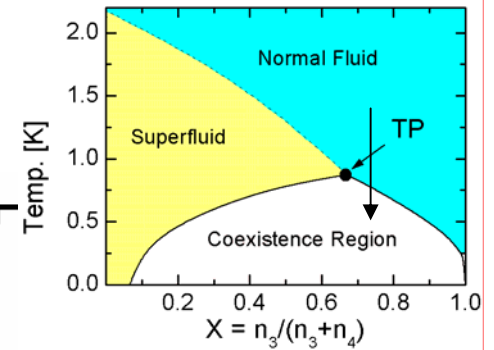
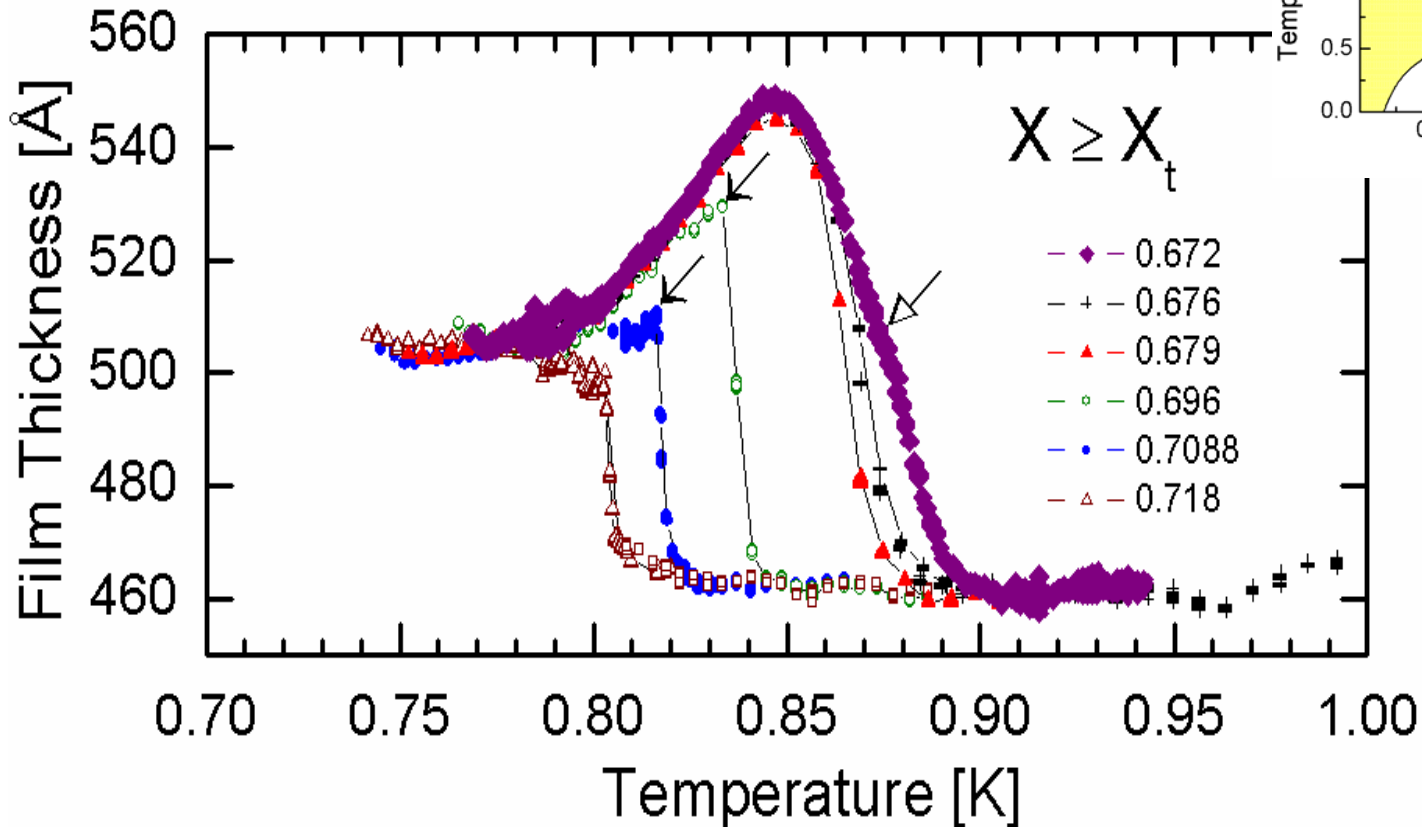
This superfluid layer forces the O.P. in the fluid above it to assume a different B.C. close to the substrate

⇒ **Repulsive critical Casimir force**



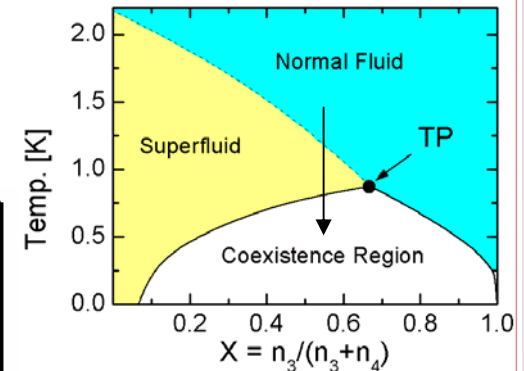
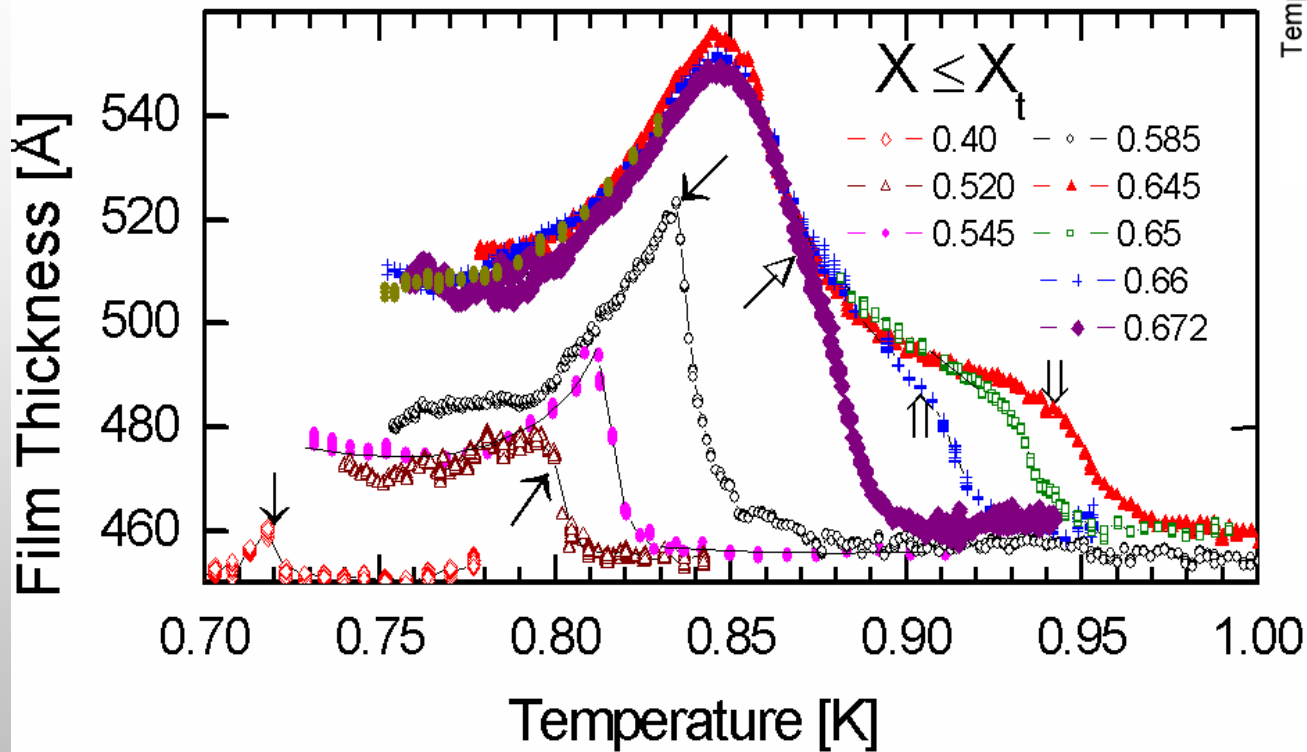


$d(T)$  for  $X \geq X_{tri}$

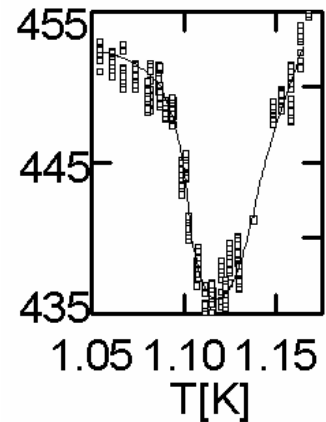


A thickening occurs as we approach  $T_{sep}$ . The curves deviate from “universal” curve just below  $T_{sep}$ .

$d(T)$  for  $X \leq X_{tri}$



$X = 0.585$



A thickening occurs close to  $T_{sep}$  and also close to  $T_\lambda$  for  $X \geq 0.60$  but for  $X=0.59$  there is a dip similar to the one seen more dilute mixtures.

# Summary of tricritical point results

- Repulsive Casimir force is seen in adsorbed mixture film near the tricritical point. A consequence of asymmetric order parameter at the solid and vapor interfaces
- The scaling function is well described by theoretical calculations.

Physical Review Letters 83, 1187 (1999)

Physical Review Letters 88, 086101 (2002)

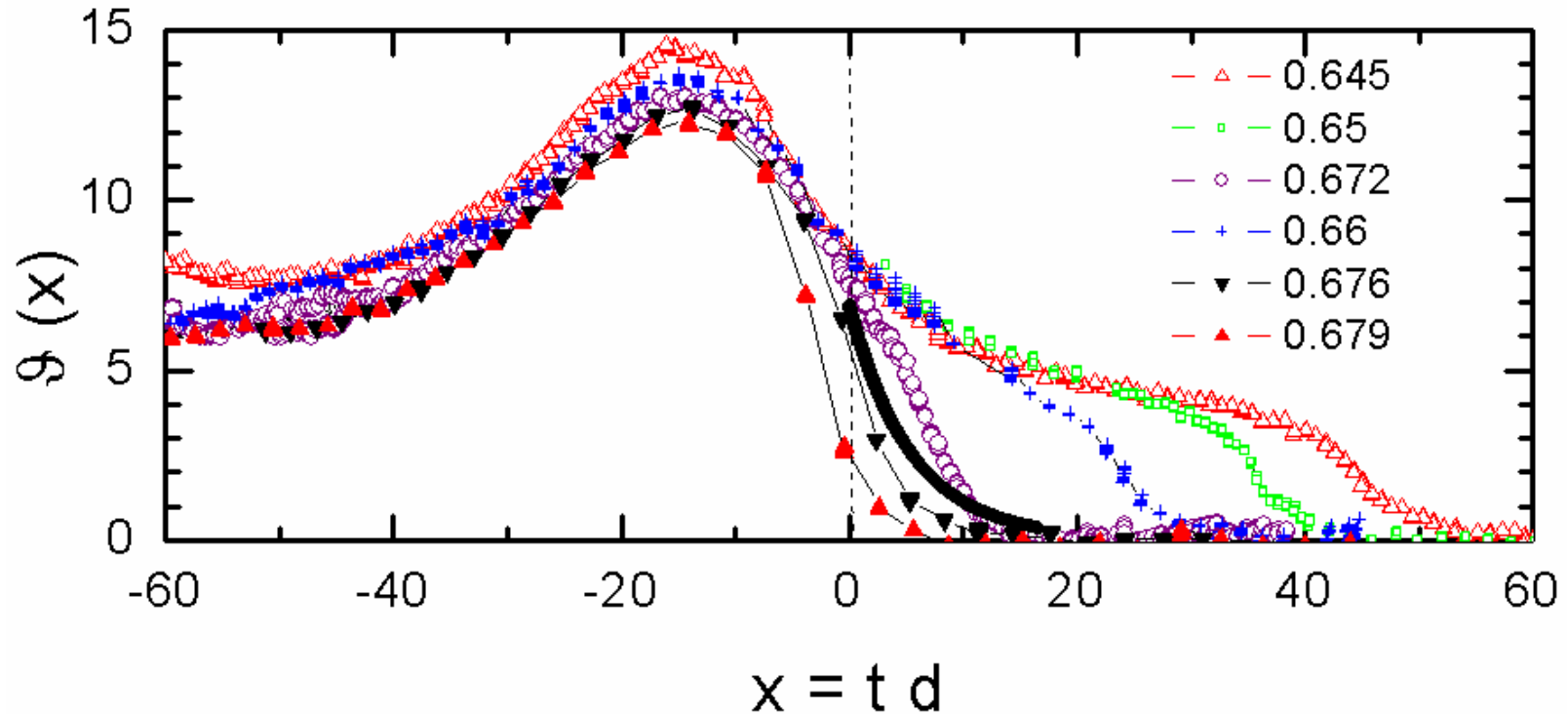
Physical Review Letters 97, 075301 (2006)

J. of Low Temp. Phys. 134, 527 (2004)



# Scaling function $\mathfrak{G}(x)$

$$\frac{\gamma}{d^3(1+d/\lambda)} + \frac{k_B T_C \bar{V} \mathfrak{G}(d/\xi)}{d^3} = \bar{m}gh$$



- Solid curve is a theoretical calculation assuming the layer near the substrate is superfluid but there is no superfluid at the vapor interface.