

## **THERMAL EXPANSION OF HELIUM FILLED AEROGEL AT THE LAMBDA POINT**

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Under reasonable assumptions, we show that He-filled Aerogels should exhibit a strong negative thermal expansion at the  $\lambda$  point :  $\alpha = -10^{-3}/K$ . We observed an expansion smaller than  $10^{-6}/K$  across the transition. This very strong discrepancy can be related either to (i) specific elastic properties of the gels, (ii) the dependance of  $T_\lambda$  on the pore size is drastically different of what could be expected.

### **1 INTRODUCTION**

Liquid Helium in silica aerogels is known to behave very differently than in other porous media.<sup>1,2,3</sup>; this difference is currently attributed to the specific fractal geometry of these gels<sup>4,5</sup>. Although the onset temperature of the superfluidity is only slightly depressed, the exponent of the superfluid transition is significantly changed, and a violation of the Josephson scaling relation is apparently observed<sup>2</sup>.

Unlike conventional porous media, the aerogels have exceptionally small bulk moduli ( $E=10$  bars, compared to  $7 \times 10^5$  bars for bulk  $\text{SiO}_2$ ), so that we can expect a significant feed-back effect of the Helium onto the gel when crossing the lambda transition. A measurement of the thermal expansion of the gel could then provide information on thermodynamic properties of the Helium inside the gel.

## 2 THERMODYNAMICS OF THE EXPANSION

In a typical experiment, helium is free to enter in the porous matrix, the volume of which is free to adjust. The helium will adjust its density to reach the same chemical potential as outside ( $\mu_4$ ). The volume of the porous sample will then adjust to minimize the thermodynamical potential

adapted to that situation :  $\Omega = \frac{V_0 E}{2}$

$\left(\frac{\delta V}{V_0}\right)^2 + (F_4(N, T, V) - N \mu_4) + P_{\text{ext}}V$ , where  $P_{\text{ext}}$  is the pressure exercised on the sample,  $N$  the number of He atoms in the sample,  $V$  its volume,  $V_0$  the volume when there is no helium and  $P_{\text{ext}}=0$ , and  $\delta V=V-V_0$ . The first term is the elastic energy of the gel, and the second the grand-potential for the He. Using the above expression for  $\Omega$  we find

$E\left(\frac{\delta V}{V}\right) + \left(\frac{\partial F_4}{\partial V}\right)_{T, N} + P_{\text{ext}} = 0$ . We write  $F_4 = V f_4(T, \rho, \epsilon)$ ,

where  $\rho = N/V$  is the density of the helium,  $\epsilon = \frac{\delta V}{V}$  characterizes the deformation of the gel and  $f_4$  is the free energy of Helium per unit volume and

calculate  $\frac{\partial F_4}{\partial V}_{T, N} = (f_4 - \rho \left(\frac{\partial f_4}{\partial \rho}\right)_{T, \epsilon}) + \left(\frac{\partial f_4}{\partial \epsilon}\right)_{T, \rho}$ .

The second term expresses the way  $f_4$  is changed when the density of the gel varies at fixed  $\rho$ , while the first is  $-P$ , the pressure of the helium in the pores. In the experiment  $P=P_{\text{ext}}$ , and we finally obtain :

$$\left(\frac{\delta V}{V}\right) = -\frac{1}{E} \left(\frac{\partial f_4}{\partial \epsilon}\right)_{T, \rho} \text{ and } \alpha_T = \frac{\partial V}{V \partial T} = +\frac{1}{E} \left(\frac{\partial s_4}{\partial \epsilon}\right)_{T, \rho} \quad (1)$$

$s_4$  being the entropy of He per unit volume. As an order of magnitude for  $\left(\frac{\partial s_4}{\partial \epsilon}\right)_{T, \rho}$ , we assume that :

(i) the main effect of changes in  $\epsilon$  is to change the linear size of all the pores by  $1/3 \epsilon$ , (ii)

the dependance of  $s_4$  on comes from the shift in  $T_\lambda(\epsilon)$ .  $s_4(T, \epsilon) = s_4(T - T_\lambda(\epsilon))$ ; and (iii) that  $T_\lambda(\epsilon)$  is the temperature at which  $\xi(T)$  reaches some length characteristic of the porous medium. Neither of these hypotheses are obvious, but an approach similar to (ii) yields a good order of magnitude estimate for the thermal expansion of bulk Helium at  $T_\lambda$  and (iii) yields the right order of magnitude of the shifts in  $T_\lambda$  in porous media. Under these circumstances one can estimate  $\alpha_T = -\frac{c_P}{E} \cdot \frac{\partial T_\lambda}{T_\lambda \partial \epsilon}$  where

$$E \approx 10^6 \text{ Pa}, \quad c_P \approx 10^6 \text{ J/m}^3/\text{K}, \quad \frac{\partial T_\lambda}{T_\lambda \partial \epsilon} = \frac{\Delta T_\lambda}{3\nu T_\lambda} \approx +10^{-3}$$

as evaluated from the shift  $\Delta T_\lambda = 5\text{mK}$  and the exponent  $\nu$  of bulk helium. Thus, we expect  $\alpha_T \approx -10^{-3} \text{ K}^{-1}$  close to the  $\lambda$  point.

### 3 EXPERIMENTAL VALUE FOR $\alpha_T$

We measured the changes in the length  $l \approx 8.7$  mm of a neutrally reacted aerogel sample<sup>6</sup>, at a density  $0.16 \text{ g/cm}^3$ . The resolution is about  $1\text{\AA}$ . The measurements were performed with liquid helium close to the vapor pressure, for  $1.5\text{K} < T < 2.5\text{K}$ . We observed that the expansion below  $T_\lambda$ , is essentially controlled by the fountain pressure that develops in the gel when the temperature varies in time. We can anyway estimate that for  $dT/dt = 0$  we would have  $|\alpha_T| \leq 2 \cdot 10^{-6}/\text{K}$ . A more accurate determination of  $\alpha_T$  is possible but it will be difficult to interpret, since for such small values, slight changes of the properties of the first layers adsorbed on the gel contribute largely to  $\alpha_T$ <sup>7</sup>.

The measurement of  $\alpha_T$  does not allow the determination of a critical exponent, because the effect of the  $\lambda$  transition on the gel is apparently minor. This fact is in contradiction

with the analysis we have proposed. One (or more) of the hypothesis (i-iii) is thus not correct. Concerning (i), it is to be expected that the smallest pores will contract less than the largest ones. If the discrepancy observed comes from that fact, it implies a ratio of 1 to  $\approx 500$  in the deformation at different length scales. (ii) is certainly not strictly correct, but we consider it like a dimensional-analysis argument that should provide the correct order of magnitude. Finally, concerning (iii), if the nature of the transition in the gel is different than in the bulk, because of the fluctuations of the order parameter involve (for instance) vortices around pieces of  $\text{SiO}_2^7$ , these fluctuations might well be far less coupled to linear sizes of the pores than argument (iii) claims.

In any case, the measurement of  $\alpha_T$  rises a new question on the nature of the  $\lambda$  transition in the aerogels.

#### REFERENCES

1. M.H.W Chan, K.I. Blum, S.Q. Murphy, G.K.S. Wong and J.D Reppy, PRL **61**, 1950, (1988)
1. Wong, Crowell, Cho and Reppy, PRL **65**, 2410, (1990)
3. Mulders N., Mehrotra R., Goldner L. and G. Ahlers, PRL **67**, 695, (1991)
4. Vacher R., Woignier T., Pelous J., Courtens E., PRB, **37**, 11,6500, (1988)
5. We are very gratefull to Pelous J., Woignier T., Vacher R. for providing us samples of neutrally reacted gels.
6. P. Thibault et al, in progress.
7. G. Williams PRL **59**, 17, 1926, (1987)