Characterization of fractals with an adsorbed superfluid film

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Abstract

The tortuosity of a capillary-condensed film of superfluid ^{4}He adsorbed on 91%-porous silica aerogel has been measured, with transverse sound, as a function of helium coverage. Complementary data from ^{4}He adsorption isotherms and small-angle X-ray scattering have also been used for substrate characterization. The tortuosity is found to be roughly inversely proportional to the volume fraction of the liquid phase of helium.

Key words: superfluidity; aerogel; tortuosity; fractal

1. Introduction

We report an attempt to develop a new tool to gain information on the microscopic structure of porous materials. The idea is to measure how the tortuosity of an adsorbed liquid film changes with the liquid volume fraction. Thin films follow all irregularities of the tortuous substrate, while with thicker films the short length scale irregularities are shunted by the liquid capillary condensed in small pores. The scaling of the tortuosity with the size of the biggest filled pore or volume of adsorbed liquid could work as an independent fractal characteristic of the material.

We studied a sample of 91%-porous silica aerogel made by Airglass, Sweden. Small angle X-ray scattering revealed a range of mass fractal correlations between the scattering vectors $q = 0.02\,\text{Å}^{-1}$ and $0.06\,\text{Å}^{-1}$ (i. e. between length scales of order $\sim \pi/q = 50$ Å and 150 Å) with the fractal dimension of 2.0.

The tortuosity, α, is the geometric factor in the solution of the equation for flow of liquid along a tortuous flow path [1,2]. For superfluid ^{4}He in porous materials it is usually expressed by the "drag factor" [3], $\chi = 1 - \alpha^{-1}$, and can be used to characterize the morphology of capillary-condensed films [4].

We consider sufficiently thick films for which the chemical potential μ is dominated by the surface tension. On the surface of a film of curvature $2/r$,

$$\delta\mu \equiv \mu_b - \mu = \frac{2\sigma v_4}{r} = -k_B T \ln(\frac{p}{p_0}), \qquad (1)$$

where $\sigma = 0.3$ erg/cm^2 is the ^{4}He surface tension, $v_4 = 27.5\ \text{cm}^3/N_A$ is the ^{4}He atomic volume, μ_b is the chemical potential of bulk liquid and p_0 is SVP at T. Hence,

$$r(p) = -\frac{2\sigma v_4}{k_B T \ln(p/p_0)}. \qquad (2)$$

When (while filling up) the gas pressure increased up to p, all pores of radius smaller than $r(p)$ are full, while the bigger pores are only covered by a thin van der Waals film. The majority of the liquid is in the filled pores, and the overall density of helium scales with r between 25 Å and 75 Å (Fig. 1).

2. Experimental results and discussion

Transverse sound resonance in a thin slab of aerogel [5] was used to extract the overall coupled density of helium $\rho_\nu(T)$ from the resonant frequency $\nu(T)$. All adsorbed helium is divided into two parts: the inert

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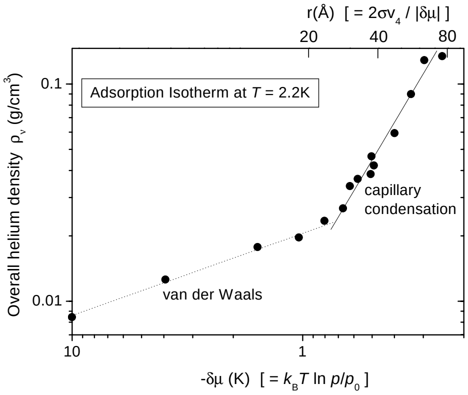

Fig. 1. ^{4}He adsorption isotherm at $T = 2.2$ K: overall density of adsorbed ^{4}He vs. its chemical potential. The cross-over from the van der Waals to the capillary condensation regime is seen at overall density 0.025 g/cm^3. For the latter regime the chemical potential corresponds to the "radius of the largest filled pores" $r(p)$ (top axis). Straight lines guide the eye.

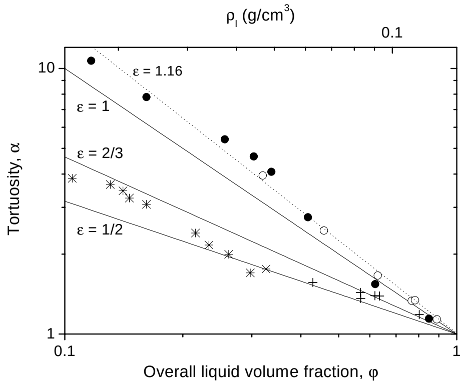

Fig. 2. Tortuosity vs liquid filling fraction: (●) our data; (○) data for 93.5%-porous aerogel [7]. Saturated samples of different porosities: ($*$) fused-glass bead samples [2], ($+$) packed powder samples [8]. Solid lines – effective-medium theory for saturated porous media [1]: ($\epsilon = 1$) self-similar needles perpendicular to flow, ($\epsilon = 2/3$) self-similar needles randomly oriented, ($\epsilon = 1/2$) self-similar spheres. Dotted line – fit to our data with $\epsilon = 1.16$.

solid layer of overall density $\rho_0 = 0.010$ g/cm^3 (using the " critical coverage" value of $n_c = 36$ μmol/m^2 for 91%-porous aerogel [6]) and the liquid part with overall density $\rho_l = \phi\rho_b$, where ϕ is the volume fraction of the liquid part and $\rho_b = 0.146$ g/cm^3 is the bulk ^{4}He density. At $T = 2.2\,$K all the helium mass is coupled to the oscillating aerogel, while at $T = 0.5\,$K all the liquid part is superfluid and coupled to the aerogel only through the tortuosity of the film. Hence,

$$\alpha = \left(1 - \frac{\rho_\nu(0.5\,\mathrm{K}) - \rho_0}{\rho_\nu(2.2\,\mathrm{K}) - \rho_0}\right)^{-1}, \tag{3}$$

$$\phi = \frac{\rho_\nu(2.2\,\mathrm{K}) - \rho_0}{\rho_b}. \tag{4}$$

In Fig. 2 we plot our values of tortuosity $\alpha(\phi)$ for the capillary-condensed regime when $\rho_\nu(2.2\,\mathrm{K}) > 0.025$ g/cm^3 (i. e. for $\phi > 0.1$) as well as some of the data obtained by Dolesi *et al.* [7] for ^{4}He at $T = 1.25$ K in 93.5%-porous silica aerogel from the same manufacturer. The latter were re-analyzed to account for the inert layer using the value $\rho_0 = 0.007$ g/cm^3 obtained from our value of 0.010 g/cm^3 by scaling with the aerogel density. Both data sets are in good agreement and follow the law $\alpha = \phi^{-\epsilon}$ with $\epsilon \approx 1.16$. For comparison, the results for ^{4}He saturated in non-fractal porous media (fused-glass beads [2] and packed powders [8]) of various porosities are shown too. It is clear that the tortuosity of films on aerogels differs markedly from that of helium in non-fractal porous media.

The solid lines in Fig. 2 show the calculated values of α for liquid in a model medium constructed by nested infinitesimal increments of density of self-similar spheres or needles [1] predicting the exponents between $\epsilon = 1/2$ and $\epsilon = 1$. Other theories for the density dependence of the tortuosity or conductivity of random or fractal networks exist, usually predicting $\epsilon \geq 1$. However, much smaller substrate-dependent values of ϵ for the tortuosity of a capillary-condensed film on surface fractals have been predicted as well [9,10]. We would like to know whether or not the power law with the $\epsilon \approx 1.16$ is universal for different types of aerogel or other porous mass fractals. Further experiments and model calculations are under way.

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References

[1] D. L. Johnson, P. N. Sen, Phys. Rev. B**24** (1981) 2486.

[2] D. L. Johnson *et al.*, Phys. Rev. Lett. **49** (1982) 1840.

[3] J. B. Mehl, W. Zimmermann, Jr., Phys. Rev. **167** (1968) 214.

[4] A. Golov *et al.*, J. Low Temp. Phys, **110** (1998) 591.

[5] I. B. Berkutov *et al.*, these Proceedings.

[6] P. A. Crowell *et al.*, Phys. Rev. B**55** (1997) 12620.

[7] R. Dolesi *et al.*, J. Low Temp. Phys. **118** (2000) 219.

[8] M. Kriss, I. Rudnick, J. Low Temp. Phys. **3** (1970) 339.

[9] P. G. de Gennes in Physics of Disordered Materials edited by D. Adler *et al.* (Plenum 1985)

[10] S. M. Cohen *et al.*, Phys. Rev. B**34** (1986) 6522.