SAXS Studies of Absorption of Helium into Silica Aerogel Near the Liquid Vapor Critical Point

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Introduction

Phase transitions of fluids in porous media have been the subject of intense experimental and theoretical interest in order to understand the effects of quenched randomness on critical phenomena. In the case of binary fluids imbibed within porous media, Brochard and de Gennes have predicted that the critical behavior should be described within the context of the Random Field Ising Model (RFIM).[1] It is expected that critical fluids should show similar behavior. Earlier thermodynamic experiments probing the liquid-vapor critical point of He in aerogel, however, found critical exponents more similar to the bulk values than those for the RFIM.[2] It seems likely that the critical behavior of a fluid in aerogel may depend on the detailed structure of the random environment and the corresponding structurally induced correlations in the helium. Indeed, helium in aerogel displays a wide range of remarkable behavior, which seems difficult to explain by simply considering the aerogel as a featureless random environment.[3] Up to the present, however, no experiments have probed the critical behavior of this system on a microscopic scale. We report here small angle x-ray scattering measurements of the density correlations induced in He vapor by aerogel near the liquid-vapor critical temperature.

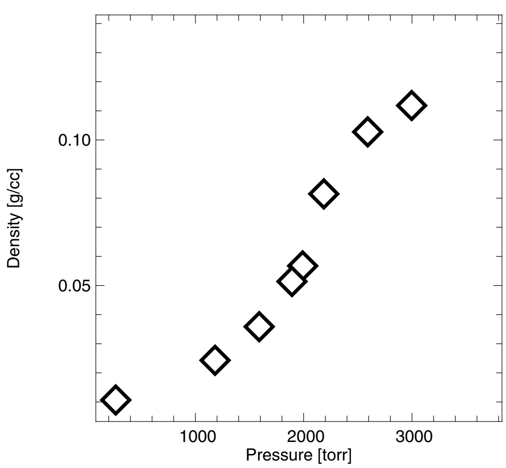

FIG. 1. Adsorption isotherm for He into 98% porous aerogel at 5.5 K.

Materials and Methods

The sample consisted of a 98% porous aerogel grown into a cylindrical plug of silver sinter, pressed into a copper cell, and held within a vapor-cooled cryostat equipped with Be x-ray windows. To permit x-ray passage, a 3-mm-diameter hole was drilled through the side of the cylindrical plug prior to the growth of the aerogel. Isotherms were performed by dosing He gas from a reservoir of known volume at room temperature into the cell through a narrow capillary. Figure 1 shows a typical adsorption isotherm measured at 5.5 K .

Scattering measurements were performed at IMMW-CAT's beamline 8-ID-E at the Advanced Photon Source (APS) and employed undulator x-ray radiation with a photon energy of 7.66 keV. Small-angle scattering was measured in transmission mode using a Princeton Instruments SCX-TE CCD camera coupled to a GdOS(Tb) phosphor by a 2.3:1 capillary taper.

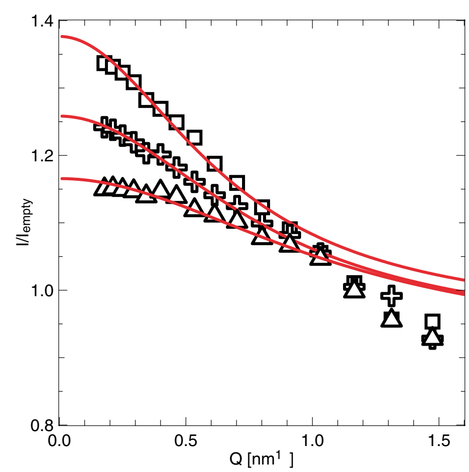

FIG. 2. Ratio of SAXS from He-dosed aerogel to empty aerogel for various helium densities. Triangles: 0.10 g/cc; crosses: 0.08 g/cc; square: 0.05 g/cc.

Results and Discussion

Small-angle scattering data as a function of adsorbed dose are shown in Fig. 2. The data have been normalized by the scattering from the empty aerogel to accentuate the deviations in the scattering due to the He vapor. Note that, in spite of its low electron density, the scattering from the helium significantly increases the total scattering. Over the range shown, the largest excess scattering is seen to occur at the lowest vapor densities. The scattering was modeled using a form based on that of Frisken, Cannell, Lin, and Sinha.[4] The predicted intensity ratio is given by $I/Ie = |A_1 + A_2 G|^2$ with $A_1 = (n_{SiO2}-n_{He})/ n_{SiO2}$, $A_2 = n_{He}/ n_{SiO2}$, n_{He} the bulk electron density for helium, n_{SiO2} the bulk electron density for silica, and $G = S/(1 + Q^2 D^2)$, with S a susceptibility and D a correlation length. The model provides a good description of the data for small Q but does not predict the reduced scattering intensity at large Q. This may result from the presence of a compact solidlike layer of helium directly adjacent to the aerogel that is not accounted for by this model. Fit results are shown in Table I. It is found that the susceptibility and correlation length both decrease with increasing vapor density. Measurements have also been carried out over a range of temperatures between 4.5 K and 5.8 K. A more complete analysis of the data is currently in progress.

Table I. Fit results corresponding to the model described in the text for the data shown in Fig. 2.

n_{He}	S	D
0.05	8.2 ± 0.1	14.3 ± 0.5
0.08	4.35 ± 0.03	11.7 ± 0.3
0.10	2.72 ± 0.04	8.6 ± 0.4

Acknowledgments

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