

Condensation of helium into silica aerogels is an out-of-equilibrium phase transition

Despite decades of theoretical and experimental efforts, the condensation and evaporation of fluids inside porous media are still not fully understood. The ANR (National Research Agency) project "HEVEPOR", a collaboration led by Néel Institute and involving the LPTMC (University Pierre and Marie Curie, Paris) and the GES (Montpellier University UM2), aims to unravel the mechanisms responsible for the observed hysteresis between evaporation and condensation, and to understand their relationship to the porous media microstructure. A specific goal is to assert the influence of disorder and confinement on the liquid-gas transition. With that goal, we study a novel experimental system, helium in transparent porous glasses. Using helium as a fluid allows a direct observation of the physical phenomena at play.

We have studied condensation of helium into aerogels, very open materials of large porosity (90% and more) made of interconnected silica strands on a nanometric scale. Aerogels are an experimental realization of dilute disorder. In 2001, it was predicted by the LPTMC group that this disorder alone could explain the hysteresis between condensation and evaporation. The disorder creates a complex energy landscape with many metastable states separated by large energy barriers, which cannot be crossed on experimental timescales. Condensation then takes place by local avalanches from gas to liquid state as the pressure is increased. A specific prediction is that the avalanche size increases as the temperature is lowered, and that, at low enough disorder (large porosity), the size diverges at a critical temperature T^* below which condensation involves a macroscopic avalanche from vapor to liquid at some given pressure. This out-of-equilibrium phase transition is similar to the jump of magnetization predicted in some disordered magnetic materials when the field is increased.

In 2008, we published the first evidence that this phenomenon does indeed occur in a silica aerogel of porosity 95% synthesized in Montpellier. This year, we reproduced our former results on a different sample of the same porosity. Improvements to the experimental set-up result in an unprecedented resolution in the adsorption isotherms, allowing their detailed study. In agreement with the theory, the slope of the adsorption isotherm (the condensed quantity as a function of the pressure) steadily increases as the temperature decreases, a result which cannot be explained by classical theories of condensation into porous materials.

In contrast to the new prediction, this slope does not diverge at a finite temperature. We attribute this discrepancy to a small heterogeneity of the macroscopic avalanche pressure across the sample.

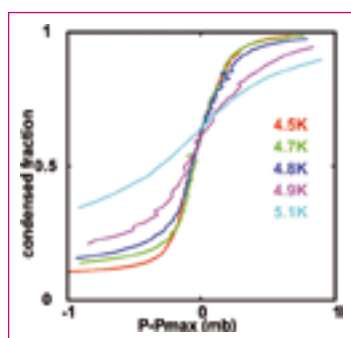


Figure 1: adsorption isotherms (condensed liquid fraction ϕ as a function of pressure) for helium in a 95% porosity aerogel: the pressure is referred to the point of maximal slope. Isotherms become steeper as the temperature is decreased.

This interpretation is supported by our optical observations. Pictures taken along the pressure quasi-plateau reveal a two-scale heterogeneity of the helium distribution. The bright region contains microscopic heterogeneities which scatter light. This region is separated by a macroscopic interface from a dark one, where the aerogel is totally filled with liquid. This interface is the locus of the macroscopic avalanche, which propagates to the center of the aerogel as condensation proceeds.



Figure 2: propagation (controlled by the helium injection) of the macroscopic avalanche at 4.7 K between a mixed gas-liquid state (white) and the liquid (dark).

A detailed analysis of the thermodynamic and optical measurements allows us to estimate a value of ~ 4.8 K for T^* , and to determine the temperature dependence of both the macroscopic avalanche pressure and the typical size of microscopic helium droplets at the avalanche threshold ; These results pave the way to a better understanding of the macro-avalanche process and its possible connection to a depinning of the microscopic interface between liquid and gas.

This work was performed by F. Bonnet, M. Melich, L. Guyon, P.E. Wolf and the late L. Puech, of the HELFA team, with the contributions of F. Despetis (GES) and E. Kierlik (LPTMC).

CONTACT

Pierre-Etienne WOLF

pierre-etienne.wolf@grenoble.cnrs.fr
Tel: +33 476 88 12 73

FURTHER READING

EVIDENCE FOR A DISORDER-DRIVEN PHASE TRANSITION IN THE CONDENSATION OF ^4He IN AEROGELS

Bonnet, T. Lambert, B. Cross, L. Guyon, F. Despetis, L. Puech et P. E. Wolf, Europhysics Letters 82 56003 (2008). *EPL Editor's choice 2008.*