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Experiments on ^{3}He – ^{4}He Mixtures in Aerogel

W. J. McRae, G. Lawes, A. Golov[1], E. N. Smith, N. Mulders[2], J. M. Parpia

Laboratory of Atomic & Solid State Physics, Cornell University
Ithaca, NY 14853 USA
[1] *Now at Dept. of Physics and Astronomy, Univ. of Manchester*
Manchester M13 9PL, UK
[2] *Dept. of Physics and Astronomy, Univ. of Delaware*
Newark, DE 19716 USA

We describe experiments on ^{3}He-^{4}He mixtures in 98% open aerogel grown by cold-deposition ($T \leq 80mK$) of ^{4}He followed by the deposition of ^{3}He or alternatively by cooling down a homogenous mixture. The two approaches led to different ^{4}He film morphologies. We will also describe the observation of migration of ^{4}He from or toward the cell below 100 mK. This migration can lead to the exclusion of ^{4}He (other than the localized surface layers) for ^{4}He concentrations between 4% and 11% in the cell.

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1. INTRODUCTION

In aerogel the phase diagram of ^{3}He-^{4}He mixtures is significantly altered from that of the bulk. At low ^{4}He concentration, $x_4 < 0.1$, no phase separation is observed down to the lowest temperatures[1,2], and the coexistence curve is detached from the lambda line. The details of this unusual behavior are poorly understood, especially in the context of our experiments where we have a silver sinter of area comparable to that of the aerogel in contact with the mixture.

We have identified key features in our experiments which affect the morphology and concentration of the ^{4}He-rich phase in aerogel: 1. the interplay of gravitational and interfacial energies between two different porous materials, the aerogel and a sintered silver heat exchanger. 2. gravitational effects that induce capillary condensation. 3. preparation history of the sample –

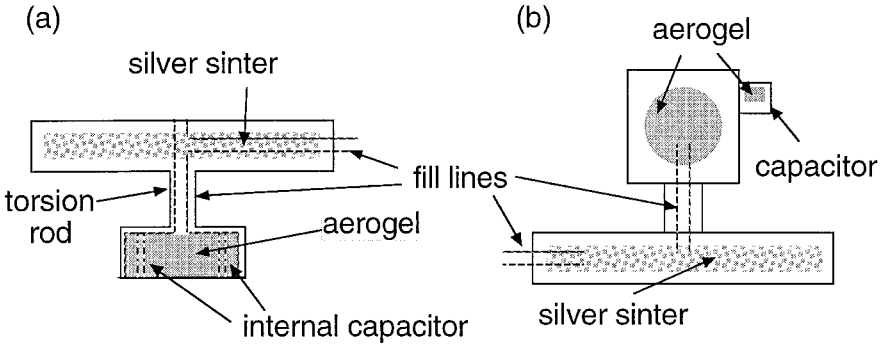

Fig. 1. Schematic of the two experimental cells.

whether it was cold-deposited (below 0.2K) or cooled from a homogeneous mixture starting from a high temperature (1K). 4. the migration of ^{4}He induced by interaction of the mixture with high temperature parts of the dilution refrigerator.

2. EXPERIMENTAL TECHNIQUE

We studied the helium 4 concentration in two similar aerogel cells (see Figure 1): The first set-up (cell 1, configuration A) consists of a torsional oscillator with its head almost completely filled with aerogel of 98% porosity (0.4cm^3 open volume and 9.3m^2 surface area). There is a sintered silver heat exchanger (0.4cm^3 open volume and 4.2m^2 surface area). The aerogel is grown in situ within (and surrounding) a parallel-plane capacitor. Measuring the capacitance allows us to determine the absolute value of the fraction of ^{4}He to better than 1%.

A second, similar cell (cell 2, configuration B) was constructed for the observation of low frequency sound. It has the identical sintered silver heat exchanger to cell 1, however this cell is oriented so that the sinter is below the aerogel. The aerogel sample is a disc of 3mm thickness and 1cm diameter oriented so that the flat face of the disc is vertical. The capacitor for measuring the ^{4}He fraction is mounted on the side of the disc and contains a small amount of the same aerogel grown in situ.

In both cases the cells were mounted on a nuclear demagnetization stage connected by a superconducting heat switch to a dilution refrigerator. The

fill line going into the heat exchanger and cell was thermally anchored to the mixing chamber, and to higher temperature heat exchangers. The aerogel and silver sinter were separated by 1cm vertically and were approximately 10cm below the mixing chamber of the dilution refrigerator. Results are obtained at a pressure p = 21.6 bar.

3. RESULTS AND DISCUSSION

3.1. Sample Preparation

We have studied a range of ^{4}He content in configuration A (aerogel below the sinter) ranging up to 34%. The first 0.4mmole of ^{4}He corresponded to the localized layer[1,3] of 28μmole/m^2 adsorbed on the total surface of aerogel and sinter. If we attempt to increase the ^{4}He fraction x_4 in the aerogel by increasing the ^{4}He-^{3}He ratio at T = 1K, with cooling, most of this ^{4}He preferentially moves into the sinter rather than the aerogel. However, we found that x_4 in the aerogel could be increased without having a high overall ^{4}He content, providing we first introduced pure ^{4}He to the empty cell at a temperature below 0.1 K. The ^{4}He coated the surfaces of the cell (including the strands of aerogel) with a thick ^{4}He film. This "film state" (corresponding to a distribution different from that achieved by cooling a mixture from 1K) persisted even after the remaining volume was filled with ^{3}He and the cell brought up to a pressure of 21.6 bar. The ^{4}He film-state with $x_4 > 12\%$ was found to be stable (for 14mmole of ^{4}He in the entire sample) providing the temperature was not raised above 0.2 K[2]. If the cell was warmed above phase separation (to 1K), the system undergoes a remixing transition and results in a pronounced hysteresis in the amount and distribution of the ^{4}He in the aerogel.

3.2. Expulsion of ^{4}He from Aerogel

If x_4 is small (below 4% - x in Fig. 2) this corresponds to the aerogel being coated with two layers of "localized" solid ^{4}He. Once achieved, this concentration is stable.

We find in configuration A that when we started with a concentration between 4% and 10% (+ in Fig. 2), x_4 decreases, and ^{4}He is expelled from the cell till the stable 4% concentration is achieved. This behavior has been noted on several trials and likely reflects the interplay between gravity, surface energy, and the geometries of the two porous media. At face value, the observation can be interpreted in one of three ways: a) The ^{4}He rich

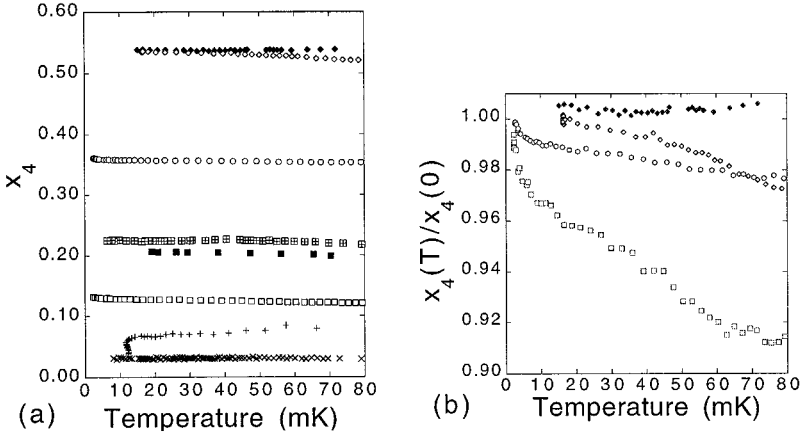

Fig. 2. (a) The ^{4}He fraction x_4 is plotted against temperature for a variety of concentrations. The filled diamonds and the open diamonds are cell 2, configuration B; all other symbols are cell 1, configuration A. (b) Four of the runs from (a) are shown scaled by the low temperature value of x_4. Symbols are the same as in (a).

phase does not wet the ^{4}He localized layer but instead migrates to the silver sinter. b) The shape of the interface between the ^{3}He rich phase and the ^{4}He rich phase is quite different in the sinter and in the aerogel. If the ^{4}He-rich phase were to stay in the aerogel it would have a large interface area compared to the configuration achieved if the same amount of ^{4}He were to occupy the sinter. The concavities in the sinter lead to a rapidly diminishing interface area with the addition of ^{4}He. Thus the energetics favor the penalty of gravitational potential energy compared to the interfacial area. Presumably, as more of the sinter is occupied by the ^{4}He rich phase, eventually the gravitational free energy cost is too great and it is energetically favorable for the ^{4}He-rich phase to occupy the aerogel (for x_4 above 10%). However, we have not been able to trace the development of the ^{4}He rich phase in this important concentration region. c) Since no phase separation is observed when the mixture is cooled to below 100 mK from T=1K for $x_4<10\%$, the interface between the ^{3}He and the ^{4}He rich phase may be diffuse and there may be a continuous distribution of densities of ^{4}He with distance from the wall. The solubility of the ^{4}He in ^{3}He is very small at these temperatures[4], and this diffuse interface costs free energy and is avoided by raising the gravitational potential energy of the dilute mixture. We consider possibility (a) to be unlikely as the ^{4}He-rich film should wet the localized solid almost perfectly in contrast to the ^{3}He. We plan to conduct experiments

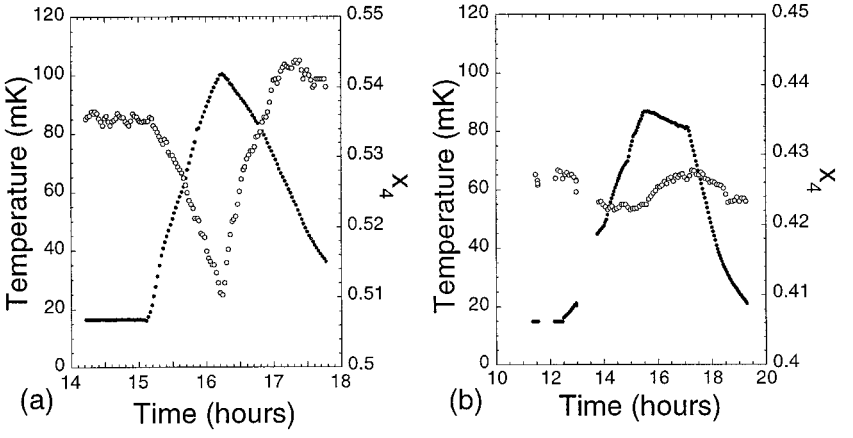

Fig. 3. Cell temperature (filled circles) and ^{4}He fraction (open circles) are plotted simultaneously versus time. (a) (on left) shows the cold-deposited film state before remixing. (b) (on right) shows the same sample after remixing.

that will allow us to vary x_4 quasi-continuously in the region around 10%. By studying the evolution of x_4 as ^{4}He is added, we should be able to observe if there is an abrupt or gradual transition from x_4=4% to 10% and this should determine which of the possibilities (b) or (c) is correct.

3.3. Variation of ^{4}He Concentration with Temperature

For cold deposited film states in cell 1, configuration A and $x_4 > 12\%$, we observe a reproducible temperature variation of x_4 as the temperature is lowered (Fig. 2). Thus, if we start from 12% ^{4}He at 100 mK, the final low temperature concentration is approximately 13.5%, (squares with dots in Fig. 2), and the same behavior is seen for higher x_4.

When we cold deposited a sample into cell 2, configuration B, we observed a similar variation in x_4 (open diamonds in Fig. 2) with the cell and mixing chamber connected. However when we isolated the nuclear stage from the dilution refrigerator and heated the stage while maintaining the dilution column below 20mK, no temperature variation was observed (closed diamonds in Fig. 2). We conclude that the temperature dependent variation in x_4 is affected by thermal gradients in the dilution column.

3.4. Gravitational Collapse

In cell 1 configuration A, we found that irrespective of preparation (cold deposited or cooled from a homogenous mixture) we could not achieve an x_4 great than 40% without inducing an invasion of the aerogel by the ^{4}He rich component. We interpret this as the onset of capillary condensation, beginning in the lowest parts of the cell (in aerogel) due to gravitational variation in film thickness. In contrast in cell 2 configuration B with the cold deposited film we found x_4 to be initially 54%, increasing gradually over several cycles of warming and cooling (never going above 120mK) to 60%, showing no evidence of a gravitational collapse (Fig. 3a). Upon cooling from the remixed state without altering the mixture, x_4 was found to be 42% and showed little variation with temperature, with or without the connection to the mixing chamber enabled (Fig. 3b). This suggests that the higher x_4 values obtained before remixing represented a metastable state which underwent some redistribution of ^{4}He on each cycle of warming to 100mK.

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