THE OSMOTIC PRESSURE OF ^3He – ^4He MIXTURES ALONG THE PHASE SEPARATION CURVE

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The osmotic pressure of ^3He – ^4He mixtures was measured along the phase separation curve at temperatures up to 500 mK by balancing it with the fountain pressure of pure ^4He . The usefulness of the secondary osmotic pressure thermometer was reinvestigated.

Measurement of the osmotic pressure of ^3He in the superfluid phase in the mixing chamber of a dilution refrigerator may give information about the temperature of the liquid or the concentration of ^3He in the superfluid phase during circulation (provided the other of these parameters is known). This can be seen from the working definition [1] of the osmotic pressure π :

$$\pi(x, T) V_4(T) = \mu_4(0, T) - \mu_4(x, T), \quad (1)$$

T is the temperature, V_4 the molar volume of pure ^4He , x the concentration of ^3He and μ_4 the partial chemical potential of ^4He in the mixture.

It was suggested by London et al. [2] that during operation this concentration x equals the equilibrium concentration x_- of the lower phase separation curve. Hence the osmotic pressure depends only on the temperature and would therefore provide an easy and reliable secondary thermometer. However, the circulation of ^3He or ^4He in the mixing chamber should, in principle, give rise to non-equilibrium concentrations in the superfluid phase, preventing correct temperature determination from osmotic pressure measurements. Values of the osmotic pressure at the phase separation curve were reported by London et al. [2], Varoquaux and co-workers [3–5], at zero external pressure and by Landau et al. [1] and recently by Pennings et al. [6] under pressure.

The present investigations were made with a conventional dilution refrigerator at zero external pressure. The experimental set-up is schematically shown in fig. 1. Four heat exchangers are used between the still and the mixing chamber. The cylindrical mixing chamber, inside dimensions: height 23 mm and diameter 34 mm, is made of a single piece of electrolytic copper, out-

Fig. 1. Scheme of the dilution refrigerator. Arrows indicate the ^3He flow. 1 and 2 are continuous heat exchangers; 3 and 4 step heat exchangers.

gassed at 850°C for several hours before assembling. Two cells A and B with a semicircle cross-section are milled in the copperblock. Cell A is used for circulating the ^3He . Cell B, the non-circulating side, is connected with cell A by lowering the separating wall by a few mm. The height of this wall is 20 mm. The osmotic pressures in both cells are recorded by the method proposed by London [2]. Two bulbs are connected to the bottom of cell A and B by capillaries having a length of 1 m and an inner diameter of 0.35 mm. Heaters can increase the temperatures T_A and T_B of the bulbs until

the fountain pressures compensate the osmotic pressures in the connected cells. The heating powers can be varied between 2 and 80 μW without observable change (<0.2 mK) in the bulb temperatures. The middle of the capillaries are thermally anchored to a point between the first two heat exchangers, counting from the still. The bulbs and the mixing chamber are in the same horizontal level, hence hydrostatic corrections can be neglected. T_A and T_B are measured with carbon resistors calibrated against the ^4He vapour pressure scale. They vary between 1.3 and 1.8 K.

The temperature T_M of the mixture inside cell B also is measured with a carbon resistor calibrated with CMN in situ. The calibration was checked with an NMR thermometer [7]. The temperature T_M of the mixture is varied by heating the third heat exchanger, counting from the still. In this way temperature differences in the mixing chamber are minimized. Additional information about the performance of the refrigerator can also be obtained from the ratio of the temperature of the last heat exchanger and T_M . In our case this value always exceeded 2.5 below 0.2 K.

The fountain pressure tables of Radebaugh [8] are used for easy comparison with the existing literature. Temperature variations of the mixture are synchronously recorded by the bulb thermometers in the entire temperature interval. At higher heating powers the bulbs can be brought into equilibrium within 15 min. Results for the osmotic pressure obtained with slow variations of T_M , e.g. less than 5 mK/min, agreed within experimental accuracy with those measured in a stationary state, i.e., T_M is constant [5].

Below 0.5 K the osmotic pressure in cell B, where no circulation takes place, can be approximated by

$$\pi = 16.9 + 640 T_M^2, \quad (2)$$

(π in torr, T_M in K). Small deviations from this expression can be read from fig. 2 together with the results presented in the literature. The osmotic pressure in cell A, with circulation, never exceeded those values in cell B. Either the osmotic pressures in A and B were equal, confirming the calibration of the thermometers, or the osmotic pressure in cell A was substantially lower. In fig. 3 a typical run of experiments is shown. At the lowest temperatures no difference in osmotic pressure is observed. At a higher value of T_M , in this case 88 mK, the osmotic pressure π_A in cell A becomes gradually lower than π_B . The magnitude of

Fig. 2. Plot of $\pi_{\text{exp}} - \pi_{\text{cal}}$ versus T . π_{cal} is obtained from (2). The numbers correspond to the following authors: 1: London [2]; 2: Landau [1]; 3: Pennings [6]; 4: Varoquaux [5]; 5: Radebaugh [8]; 6: this work.

$\pi_B - \pi_A$ seems to depend on the position of the phase separation level in cell A relative to the ends of the capillaries for in and outflowing ^3He (see fig. 1).

This phenomenon could be observed, both with increasing and decreasing T_M . It is not yet completely understood why and how the position of the phase separation level plays a role. Further investigations are still in progress.

As can be seen from (1) a non-zero value of $\pi_B - \pi_A$ results from either temperature or concentration differences. Because of the width of the slit between cell A and B we believe there is good thermal contact and a uniform temperature exists throughout the total mixing chamber. This conclusion is strengthened by the fact that it was possible to obtain equal values of π_A and π_B throughout the whole of the investigated temperature range. Furthermore it may be mentioned that, assuming concentration equilibrium, the osmotic pressure differences lead to temperature differences between cell A and B as indicated in fig. 3. It seems, however, highly unlikely that these temperature differences would increase with temperature, while heat conductivity also increases.

We therefore suggest that the main part of $\pi_B - \pi_A$ is caused by a concentration difference Δx . This necessarily implies that the superfluid connection between the cells is broken or at least cannot transport sufficient ^4He in order to cancel the occurring Δx .

Fig. 3. The difference $\pi_B - \pi_A$ in osmotic pressure versus T_M (top), and the corresponding ΔT_M assuming concentration equilibrium.

due to the ^3He circulation. This assumption will be investigated by connecting the superfluids in cell A and B via an extra capillary containing a superleak. By regulating the temperature of this superleak the assumed gradient in μ_4 between cell A and B can be cancelled or maintained. A rough estimate of Δx can be made with the help of [8]. For the run shown in fig. 3 Δx varies linearly between 0 at 88 mK to 6×10^{-3} at 160 mK. It should be noted that a lower concentration than the equilibrium concentration acts as a heat load on the mixing chamber [9].

It is strongly suggested by the data that the results obtained in cell B give the osmotic pressure at the lower phase separation curve. Unfortunately only London's results [2] were measured under comparable conditions. Varoquaux' data [3–5] were obtained in a separate cell in thermal contact with the mixing chamber. The values of Pennings et al. [6] and Landau et al. [1] are taken under external pressure, the first with ^4He circulation, the latter in a static state. Landau's values at 0 K indicate that linear extrapolation of π ($p = 2.8$) and π ($p = 1.4$) to zero pressure would lead to a value 1.3 torr higher than the actual value of π ($p = 0$). When linear extrapolation is also erroneous at higher temperatures it would at least ex-

plain part of the discrepancies between Pennings' and our results. Systematic errors of a few millikelvin in the temperature scales used might be responsible for the rest.

A simple analytical relation between the temperature of the bulb and the mixture in the entire interval from 0 to 500 mK has not yet been obtained. In the range of 70 to 500 mK, however, an almost linear dependence between T_B and T_M with a slope of 1.4 is observed. This indicates that sufficient resolution for accurate determination of T_M exists.

In conclusion it can be said that measurement of the osmotic pressure at the phase transition curve can be realized in a mixing chamber and is a quick and reliable method for determining the temperature of the mixture.

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