Improved Capacitive Melting Curve Measurements

Alexander Sebedash\textsuperscript{1,2}, Juha Tuoriniemi\textsuperscript{1}, Elias Pentti\textsuperscript{1}, and Anssi Salmela\textsuperscript{1}

\textsuperscript{1} Low Temperature Laboratory, Helsinki University of Technology, PO Box 5100, FI-02015 TKK, Finland
\textsuperscript{2} Kapitza Institute for Physical Problems RAS, Kosygina 2, Moscow 119334, Russia

E-mail: sebedash@boojum.hut.fi

Abstract.

Sensitivity of the capacitive method for determining the melting pressure of helium can be enhanced by loading the empty side of the capacitor with helium at a pressure nearly equal to that desired to be measured and by using a relatively thin and flexible membrane in between. This way one can achieve a nanobar resolution at the level of 30 bar, which is two orders of magnitude better than that of the best gauges with vacuum reference. This extends the applicability of melting curve thermometry to lower temperatures and would allow detecting tiny anomalies in the melting pressure, which must be associated with any phenomena contributing to the entropy of the liquid or solid phases. We demonstrated this principle in measurements of the crystallization pressure of isotopic helium mixtures at millikelvin temperatures by using partly solid pure $^4\text{He}$ as the reference substance providing the best possible universal reference pressure. The achieved sensitivity was good enough for melting curve thermometry on mixtures down to 100 $\mu$K. Similar system can be used on pure isotopes by virtue of a blocked capillary giving a stable reference condition with liquid slightly below the melting pressure in the reference volume. This was tested with pure $^4\text{He}$ at temperatures 0.08–0.3 K. To avoid spurious heating effects, one must carefully choose and arrange any dielectric materials close to the active capacitor. We observed some 100 pW loading at moderate excitation voltages.

1. Introduction

The slopes of the melting curves of helium liquids tend to zero with decreasing temperature, so that melting curve thermometry at ever lower temperatures calls for increased sensitivity of the pressure transducers. To be applicable at the lowest temperatures reached in helium liquids—of order 0.1 mK—the required pressure resolution is in the range of nanobars [1]. We describe a simple scheme, which enabled us to improve sensitivity of pressure measurements by two orders of magnitude in comparison with the traditional capacitive method. This principle was successfully demonstrated recently in our measurements on dilute $^3\text{He}$–$^4\text{He}$ solutions [2]. In this contribution we present melting curve measurements on pure $^4\text{He}$ and discuss the feasibility of extending melting curve thermometry on pure $^3\text{He}$ down to 0.1 mK.

The idea is based on a straightforward differential gauge principle implemented to a capacitive transducer. The traditional design is often referred to as the Straty-Adams type; for details, see a comprehensive review by Adams [3]. In short, a pressure difference over a membrane causes a deflection, which changes the free gap of a capacitor and thus of its capacitance. The success of this method relies on very precise devices available for detecting very small changes in capacitance [4].
Consider a parallel plate capacitor with vacuum between the plates and the working pressure $p$ pushing the plates closer to each other. Its capacitance $C$ is obtained as $C(p) = C_0 \cdot (1 - p/p_s)^{-1}$, where $C_0$ is the nominal value at $p = 0$ and the plates become shorted at $p = p_s$, i.e., the capacitance diverges. One can express the resolution for pressure, $\delta p$, in terms of the smallest observable change in capacitance, $\delta C$, as $\delta p = (\frac{dC}{dp})(\frac{1}{C}) \cdot (\frac{dC}{dp}) \cdot (p_s - p)$. So, making $(p_s - p)$ smaller improves the sensitivity. In principle, any such device becomes arbitrarily sensitive, if the working pressure can be arranged sufficiently close to $p_s$. In practice, however, it is not possible to work too close to such an instability, because an increase of losses and a local electric breakdown will occur before physical touching of the electrodes. The largest reliable working capacitance with dimensions of order 1 cm is not more than 200 pF. To gain more sensitivity within these constraints, one must decrease the value of $p_s$. This is achieved at the expense of dynamic range of the transducer, which cannot be much larger than $p_s$. To use working pressures considerably higher than that, one must apply counter-pressure to the opposite side of the membrane, denoted hereon as the reference pressure $p_r$, by which the working pressure is measured in relative terms, $p \rightarrow \Delta p = p - p_r$. For a conventional Straty-Adams gauge $p_r = 0$.

The gauge we describe here was designed to be used for helium mixtures and it had $p_s = 41$ kPa. Since the saturated solution has 34 kPa higher crystallization pressure than pure $^4$He at $T = 0$ [5], the working pressure in this case was at most 7 kPa less than $p_r + p_s$ (with $p_r = p_{m4}$, the melting pressure of $^4$He). As the absolute range was about 2.5 MPa, we gained roughly two orders of magnitude sensitivity by this differential scheme. When this particular gauge is used for pure $^4$He, we have $p_s = 20$ kPa, but there is no impediment for making this much smaller, say 100 Pa, if the design was aimed for measurements on $^4$He specifically.

2. Experimental procedure

Our gauge and its operation for measuring osmotic pressure of helium mixtures has been described in Ref. [5]. Here we give further details that are important for obtaining the best results. Two separate volumes, reference and main, were built one inside another with a flexible membrane (the first capacitor electrode) as part of the separating wall. The rest of the capacitor was built into the reference volume, consisting of an opposing fixed capacitor electrode and a grounded ring as an electric guard. All structures, except the membrane, were rigid to sustain the working pressure without deformation. The membrane electrode with the diameter $D = 30$ mm and thickness $\delta = 0.2$ mm had the desired flexibility due to its high geometrical ratio $D/\delta$. To better maintain the parallel plate geometry, the fixed electrode had a clearly smaller diameter (8 mm) than the flexing membrane electrode.

The fixed capacitor electrode and the ground ring were glued together and polished to produce a flat and smooth surface. The facing capacitor electrodes were chemically plated with thin layer of silver and polished again. The gap in between was determined by a thin mylar film ring with thickness 23 $\mu$m. Special care was devoted to precise centering and alignment. Several small holes permitted liquid flow into the capacitor gap. These were covered by filters from cigarette paper to prevent any dirt from coming into the gap. The construction was such that minimum amount of potentially lossy dielectric materials were in the electric field of the capacitor. The capacitance of the unloaded gauge was $C_0 = 8$ pF, while the dissipative fraction indicated by the capacitance bridge was less than $10^{-6}$ with the neutral membrane position (at $p = p_r$).

Steady reference pressure was created by exploiting the shallow minimum of the $^4$He melting curve: solid helium plug isolated the experimental chamber from the warmer environment, where the conditions were changing, for example, due to the liquid level in the dewar. The scheme is illustrated in Fig. 1. A short tube with the inner diameter 2 mm served as a volume in which solid was formed by controlling its temperature. It was in contact with the still plate through a weak thermal link, thus giving possibility to overheat that volume. The connecting capillary to below had inner diameter less than 0.1 mm. The pressure increased along its length downwards.
due to the hydrostatic pressure. Over its entire length this was, by chance, about the same as the depth of the minimum in the melting curve, \( \sim 0.8 \) kPa. The melting pressure in such thin capillary is somewhat higher than in the bulk: the difference is estimated to be \( \sim 40 \) Pa, judged on the basis of the radius of curvature of solid restricted to such geometry. This is important, since then the solid plug does not extend to the thin capillary even if the reference volume becomes slightly compressed due to the deflection of the gauge membrane.

3. Results and discussion
Most ideal substance for testing purposes is probably pure \(^4\)He. Its melting curve flattens quickly according to a well established power law [6] at easily accessible temperature range. When we used our gauge for measurements on helium mixtures, we had partly solid \(^4\)He in the reference volume, whereas now there was only liquid \(^4\)He there. Otherwise, no pressure difference to be measured would have existed with partly solid \(^4\)He also in the main volume. When there is no solid phase accommodating to the tiny compression due to the membrane displacement, the gauge calibration has to be scaled down somewhat because of the back elastic reaction of the reference fluid. This is analyzed in the following.

The displacement of the membrane continues to be proportional to the pressure change \( \Delta p \) in the main volume, but with modified elastic coefficient: 
\[
k_{\text{eff}} = \frac{k_0}{1 + k_0/k_{\text{Ref}}},
\]
where \( k_0 \) describes the membrane itself and \( k_{\text{Ref}} \) is due to the compression of the liquid in the reference volume. In the linear approximation with almost constant volume \( V_{\text{Ref}} \), we have 
\[
k_{\text{Ref}} = 3 \cdot V_{\text{Ref}} \cdot K/S,
\]
where \( K \) is compressibility of the liquid and \( S \) is the working surface area of the membrane. By making \( V_{\text{Ref}} \) sufficiently large, this effect can be diminished to any level desired. We made two runs with different volumes, \( V_{\text{Ref}} = 3 \) and \( 18 \) cm\(^3\), respectively. In the first run [7] the observed sensitivity was 28\% of the nominal value, as deduced from the slope of the \(^4\)He melting curve [6]. This value corresponds to \( k_0/k_{\text{Ref}} = 2.6 \). The second run [5] yielded 70\% of the full sensitivity, indicating \( k_0/k_{\text{Ref}} = 0.43 \). The ratio of these values is exactly as expected on the basis of the used volumes in the two cases. This comparison is illustrated in Fig. 2.

![Figure 1. Filling line blocking diagram. Solid helium was formed in the small volume attached to the still, whereas other parts of the reference system had liquid only. The pressure along the length of the connecting capillary (shown as the line with a kink) varied due to the hydrostatic pressure. Most of the length was physically below \( T = 0.1 \) K—therefore the seeming kink in the line plotted versus the \( T \) -axis. Solid helium did not extend to the thin capillary, since the melting pressure in such a narrow channel (\(< 0.1 \) mm) is somewhat higher for natural reasons.](image-url)
The data in Fig. 2 are shown as measured by the capacitance bridge [4] without any averaging. Observed noise level was about ±5 mPa. This value is two times more than the electric noise of the capacitance bridge itself. The excess noise, most probably, was owing to the blocking conditions, because the noise amplitude didn’t depend on temperature of the gauge volumes.

Precise determination of the melting pressure of pure $^3$He is important as a thermometric standard. By conventional means the applicability is limited to above 0.5 mK [8]. By our scheme such measurements can be extended to 0.1 mK without any further adaptation, while maintaining precision of order 10%. A simple modification can be envisioned, however, which would allow even more sensitivity and better control over the reference condition. This would consist of a divided cell with very thin membrane in between, only one common filling line from room temperature, and an inter-connecting capillary maintaining the pressure in balance during filling. The connecting capillary would have a small blocking volume in the middle, which can be overheated enough to form a plug there disconnecting the two sides of the cell once the conditions are suitable. Then it is enough to control the pressure effectively in just one volume and the reference may be linked to that at will. Further complications are avoided by operation at such low temperatures, that the temperature dependence of liquid $^3$He density has become negligible.

In conclusion, we have demonstrated the prospects of increasing sensitivity of the capacitive method for melting curve measurements. The performance of our pressure gauge was checked by measuring the melting curve of pure $^4$He at temperatures 0.08–0.3 K. It was possible to resolve less than 10 mPa pressure difference. We utilized a simple scheme for blocking the filling line. The sensitivity was limited by noise of the capacitance bridge and temperature fluctuations of the plug. The design can easily be adapted for investigations of pure isotopes’ melting properties.

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