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A sensitive capacitance thermometer at low temperature for use in magnetic fields up to 20 T

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We have established the magnetic-field independence up to 20 T of a capacitance thermometer based on the incommensurate crystal (Pb$_{0.45}$Sn$_{0.55}$)$_2$P$_2$Se$_6$ in the temperature range from 25 mK to 0.5 K. The sensitivity of the thermometer, $d \ln C/d \ln T$, is $5 \times 10^{-3}$ at low temperatures and is about an order of magnitude larger than for any other previously reported capacitance thermometer. The most sensitive temperature range of the thermometer can be tuned by varying the measuring ac frequency and voltage. The reproducibility of the thermometer upon cycling below 1 K is better than 1 mK. © 1996 American Institute of Physics. [S0034-6748(96)03807-5]

I. INTRODUCTION

Thermometry at low temperatures (below 1 K) and high magnetic fields (up to 30 or 35 T) is still a developing area since the majority of all thermometers used at low temperatures are influenced by the application of a magnetic field. On the other hand, dilution refrigeration in a field-cooled system has made much progress in recent years and the need for relatively easy-to-use, field-independent thermometers in these physical extremes has increased proportionally.

Negligible coupling to a magnetic field is expected for nonmagnetic dielectric materials. For this reason capacitance thermometers based on dielectric materials have proven to be suitable for low-temperature thermometry in moderate magnetic fields. In particular, the low-temperature dielectric behavior of amorphous materials is used as a thermometric property. On the basis of certain glasses a number of low-temperature capacitance sensors have been developed. However, most of the known glass capacitance thermometers suffer from relatively low sensitivity due to their low dielectric polarizability. An attempt to improve the sensitivity of capacitance sensors in the millikelvin temperature region has been made by using a glass-ceramic dielectric material and by substitution of Sn atoms by Pb atoms. Unfortunately, these capacitance sensors show drift, which is undesirable for accurate temperature measurements.

The temperature dependence of the dielectric constant of amorphous materials exhibits a minimum in the millikelvin region. It is well known that even high-quality ferroelectric single crystals possess this feature. The so-called glasslike properties of ferroelectric materials are even more pronounced in the disordered crystals (relaxational ferroelectrics). However, most of these materials are not very useful in low-temperature thermometry due to the instability of the ferromagnetic domain structure which causes significant drift.

A noticeable exception is a capacitance thermometer based on crystalline (Pb$_{0.45}$Sn$_{0.55}$)$_2$P$_2$Se$_6$; a Sn$_{1-y}$Pb$_y$ mixed crystal belonging to the family of Sn$_2$P$_2$Se$_6$ ferroelectric materials, which has proven to be a good and sensitive capacitance thermometer for temperatures above 1 K. The Pb$_{0.45}$Sn$_{0.55}$P$_2$Se$_6$ solid solutions with $y>0.4$ undergo a phase transition at intermediate temperatures (about 100 K) from the paraelectric state to the incommensurate phase, which extends down to very low temperatures. For the $y=0.45$ compound the paraelectric incommensurate transition temperature is approximately 120 K. The loss of translational symmetry along the $q$ vector of the incommensurate modulation and structural disorder caused by the substitution of Sn atoms by Pb atoms should render the dielectric properties of the mixed crystals (Pb$_{0.45}$Sn$_{0.55}$)$_2$P$_2$Se$_6$ close to those of amorphous materials.

For this reason, the low-temperature dielectric constant of (Pb$_{0.45}$Sn$_{0.55}$)$_2$P$_2$Se$_6$ is expected to behave similarly to that of amorphous materials. Therefore, we can hope to develop a highly sensitive and magnetic-field-independent capacitance thermometer for temperatures below 1 K on the basis of this material.

In the following we report on the low-temperature behavior of two capacitance thermometers based on (Pb$_{0.45}$Sn$_{0.55}$)$_2$P$_2$Se$_6$ in magnetic fields up to 20 T, and show that they can serve us as excellent, highly sensitive field-independent thermometers.

II. EXPERIMENT

The Pb$_{0.45}$Sn$_{0.55}$P$_2$Se$_6$ was grown using a Bridgeman technique. The crystal (typical size 10 mm in diameter and 25 mm long) was cut and polished to obtain platelets (size...
5×5×0.8 mm³) perpendicular to the [100] direction. The samples had gold sputtered electrodes and were connected to stainless-steel coaxial wiring with silver paint and a small amount of epoxy.

Two samples (I and II) were mounted in different runs inside the mixing chamber of an adapted commercial dilution refrigerator, placed in a 20 T Bitter magnet. The original metal mixing chamber had been replaced by a homemade, Kapton foil mixing chamber. To avoid eddy current heating, only plastic components for the dilution refrigerator were used inside the bore of the magnet. The silver sintered heat exchangers were located about 1 m above the field center, where the magnetic field is reduced by a factor of about 100 relative to the value at the field center.

For thermometry we used three types of thermometers in the mixing chamber: calibrated Speer 100 Ω carbon resistors, a cerium magnesium nitrate (CMN) thermometer, and a vibrating wire thermometer. In zero magnetic field we have used the carbon resistors and the CMN thermometer.

To establish the magnetic-field independence of the capacitance thermometers we have to use a field-independent temperature reference. For this purpose we chose the temperature-dependent, but magnetic-field-independent, viscosity of the saturated ⁴He–³He mixture in the mixing chamber, which was measured by the vibrating wire thermometer. Of course this method is somewhat cumbersome in practice, but it was in our case the easiest and a readily available method.

The vibrating wire thermometer was a 100-μm-diam manganine wire, shaped as a somewhat elongated semicircle with a radius of about 2 mm. This device had a mechanical resonance at 5.3 kHz, which was excited by a small alternating current through the wire in the presence of a magnetic field perpendicular to the wire. There are two contributions to the measured quality factor \(Q\) of the resonance. The first is the intrinsic damping of the wire due to friction and eddy currents and is independent of temperature (at low enough temperatures). This magnetic-field-dependent quality factor \(Q_{\text{vac}}\) was measured at 4.2 K.

The second contribution to \(Q_{\text{meas}}\) stems from the viscosity of the surrounding medium and was calculated at any field according to

\[
Q^{-1} = Q_{\text{meas}}^{-1} - Q_{\text{vac}}^{-1}(B).
\]

The thus obtained quality factor \(Q(T)\) is a measure of the viscosity and therefore of the temperature of the mixture. The viscosity of a saturated ³He–⁴He mixture is known to obey the following relation between 7 and 60 mK:

\[
\eta(T) = 7.6 \times 10^{-8} T^{-1.8}.
\]

Using this, the \(Q(T)\) for a saturated mixture below 100 mK is approximately proportional to the temperature and a monotonously rising function of the temperature up to at least 1 K.

The measurements of the capacitance at various excitation voltages at a frequency of 1 kHz were performed using an Andeen–Hagerling 2500 automatic capacitance bridge.

\[\text{FIG. 1. Capacitance and loss factor } \tan \delta \text{ vs temperature in zero field of sample I measured at 0.25 V and 1 kHz.}\]

For measurements at different frequencies we have used a General-Radio-type 1615A capacitance bridge.

III. RESULTS AND DISCUSSION

To demonstrate the pronounced temperature dependence of the dielectric constant of the \((\text{Pb}_{0.45}\text{Sn}_{0.55})_{2}\text{P}_{2}\text{Se}_{6}\) material, we have plotted in Fig. 1 the capacitance \(C\) and loss factor \(\tan \delta\) of sample I in zero field at an excitation voltage \(U\) of 0.25 V and a frequency \(f\) of 1 kHz. The straight parts of the curve to the left- and to the right-hand side of the minimum have sensitivities \(\frac{d}{dT} \ln \frac{C}{f} U^2\) and \(1 \times 10^{-5}\), respectively. Figure 1 shows that \((\text{Pb}_{0.45}\text{Sn}_{0.55})_{2}\text{P}_{2}\text{Se}_{6}\) exhibits the dielectric behavior characteristic of amorphous dielectric materials, but with much steeper slopes on either side of the \(C(T)\) minimum. We have verified that a pure crystal of \(\text{Sn}_{2}\text{P}_{2}\text{Se}_{6}\) produces no appreciable capacitance variation as a function of temperature in this temperature range, which confirms our conjecture about the origin of the dielectric behavior of this material.

An important consideration for thermometry is self-heating of the thermometer. As can be seen from Fig. 1, the capacitance curve starts to bend downward below about 12 mK. This saturation behavior is probably caused by self-heating of the sample, which can be calculated using the formula \(Q = \pi C f U^2 \tan \delta\). Taking \(\tan \delta\) from Fig. 1, we estimate a temperature rise due to self-heating for this \((\text{Pb}_{0.45}\text{Sn}_{0.55})_{2}\text{P}_{2}\text{Se}_{6}\) thermometer of about 1 mK at \(T = 10\) mK, which will decrease approximately as \(T^{-3}\) for higher temperatures. Since the \(\frac{d}{dT} \ln \frac{C}{f} U^2\) for \((\text{Pb}_{0.45}\text{Sn}_{0.55})_{2}\text{P}_{2}\text{Se}_{6}\) is quite large (almost one order of magnitude larger than for glasses), the excitation voltage can be reduced to below 100 mV without major loss of accuracy.

The magnetic-field independence of the dielectric constant of \((\text{Pb}_{0.45}\text{Sn}_{0.55})_{2}\text{P}_{2}\text{Se}_{6}\) is demonstrated in Fig. 2, where we have plotted the capacitance of sample II for four different magnetic fields (1, 5, 10, and 20 T) as a function of \(Q\), the (vacuum) corrected quality factor of the resonance of the vibrating wire thermometer, which is a field-independent measure of the temperature. In Fig. 2 the lowest \(Q\) measured...
corresponds to about 25 mK, whereas the highest Q data were taken at around 500 mK. One observes that the data for the different magnetic fields lie on the same curve. Further-
more, we have observed that the value of the capacitance at the minimum does not change from 0 to 20 T. Thus, the capacitance thermometers are shown to be field independent for magnetic fields ranging from 0 to 20 T and at least down to temperatures of 25 mK.

The reproducibility of the studied thermometers upon temperature cycling below 1 K (warming and cooling back) is better than our measurement resolution. We have, upon cycling between room temperature and 10 mK, observed slight changes in the value of the capacitance and even in $d\ln C/d\ln T$ below $T_{\text{min}}$. However, when the capacitance thermometer is slowly cooled down from temperatures above 120 K (the temperature at which the incommensurate phase sets in) good reproducibility is obtained.

To investigate the close relationship between the ferro-
lectric material $(Pb_{0.45}Sn_{0.55})_2P_2Se_6$ and amorphous dielec-
trics further, we have also measured the frequency and voltage dependence of the capacitance thermometers.

The temperature dependence of the capacitance for sample II at various frequencies at zero field is shown in Fig. 3, where with increasing frequency the minimum is seen to shift gradually to higher temperatures. As is clear from Fig. 3, the dielectric dispersion (frequency dependence of $C$) occurs in the whole temperature region studied. However, the dispersion at temperatures below the temperature where the minimum occurs ($T_{\text{min}}$) is much smaller than above $T_{\text{min}}$. The measurements of $T_{\text{min}}$ at different frequencies show the same power-law behavior $^{14,15}$ as in the case of amorphous materials and for our second sample $T_{\text{min}}$ depends on frequency approximately as $\omega^{0.30}$.

In Fig. 4 the zero field $C(T)$ dependences at various excitation voltages for sample II are shown. We observe that the temperature of the minimum in $C(T)$, $T_{\text{min}}$, depends also on the excitation voltage: $T_{\text{min}}$ shifts toward higher temperatures with increasing voltage. In fact, this behavior can also be described by a power law as $T_{\text{min}}$ is observed to be proportional to $U^{0.26}$ down to the lowest voltages. The excitation voltage variations produce the largest capacitance variations at temperatures below $T_{\text{min}}$. The power-law dependence of $T_{\text{min}}$ on $U$ has never been shown before, although qualita-
tively a voltage dependence of the capacitance of glass thermometers at very low temperatures has been observed previously. $^{16}$ Note that the frequency and voltage affect dif-
ferent parts of the $C(T)$ curve, but are described by an almost identical power law shift in $T_{\text{min}}$. At this moment we have no explanation for this behavior.

The frequency and excitation voltage dependences shown in Figs. 3 and 4 provide a useful property, namely, the tunability of the most sensitive range of the thermometers to specific temperature ranges. On the other hand it implies that the calibration of these thermometers must be specified for a
given excitation frequency and voltage, on both sides of the minimum.

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13 For pure liquid $^3$He the relative increase in the viscosity due to a 20 T magnetic field is less than 0.5% below 100 mK. For a saturated $^3$He–$^4$He mixture this number is not yet known, but is expected to be similar; see, for example, A. E. Meyerovich, in Progress in Low-Temperature Physics (Elsevier, Amsterdam, 1987), Vol. XI.

