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Absence of low temperature anomaly on the melting curve of ^{4}He

I. A. Todoshchenko[1)], *H. Alles, H. J. Junes, A. Ya. Parshin*[+], *V. Tsepelin*[*]

Low Temperature Laboratory, Helsinki University of Technology, FIN-02015 HUT, Finland

[+]*P. L. Kapitza Institute for Physical Problems RAS, 119334 Moscow, Russia*

[*]*Department of Physics, Lancaster University, Lancaster, LA1 4YB, United Kingdom*

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We have measured the melting pressure and pressure in the liquid at constant density of ultra-pure ^{4}He (0.3 ppb of ^{3}He impurities) with the accuracy of about 0.5 μbar in the temperature range from 10 to 320 mK. Our measurements show that the anomaly on the melting curve below 80 mK which we have recently observed [23] is entirely due to an anomaly in the elastic modulus of Be-Cu from which our pressure gauge is made of. We thus conclude that the melting pressure of ^{4}He follows the T^4 law due to phonons in the whole temperature range from 10 to 320 mK without any sign of a supersolid transition.

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Recent experimental results obtained by Kim and Chan [1, 2] have revived great interest to the problem of supersolidity which was first discussed almost 40 years ago [3 – 5]. The supersolid state of matter is characterized by the coexistence of crystalline order and superfluidity. In helium crystals, according to Andreev and Lifshitz [3] and Chester [4], quantum delocalization of point defects (most probably – vacancies) might decrease their activation energy to zero. Bose condensation of such defects can lead then to superfluidity in a crystal, that is, supersolidity. During 1970s and 1980s many experimental groups tried to detect this possible supersolid state by various methods, but unsuccessfully (see [6] for a review).

One possible manifestation of supersolidity would be so called nonclassical rotational inertia (NCRI) – a reduction in the rotational inertia of a solid at low temperatures [5], which can be detected by the torsional oscillator measurements on a helium crystal, similarly to the famous Andronikashvili experiment [7]. In 1981, Bishop, Paalanen and Reppy [8] have carried out such measurements at temperatures down to 25 mK with carefully annealed helium samples. With $\sim 5{\cdot}10^{-6}$ sensitivity to the superfluid fraction ρ_s/ρ they found no evidence of NCRI. However, recently Kim and Chan (KC) have observed the effect of NCRI below 0.2 K at different pressures, from melting pressure up to 140 bar, with $\rho_s/\rho \sim 0.01$ in the low temperature limit. They interpreted the onset of NCRI at 0.2 K as an indication of the transition of solid helium to the supersolid phase.

To date, several experimental groups have confirmed the KC observations [9 – 11]. Rittner and Reppy [9] have also found that ρ_s/ρ is not a universal characteristic of solid helium but can be reduced below a detectable level through annealing of the sample. Quite recently, they even have been able to produce highly disordered helium samples, where ρ_s/ρ reached much higher magnitudes, up to 0.2 [12]. However, other groups [10, 11, 13] have not confirmed the annealing effect.

In the absence of a consistent explanation of all available data on the annealing effect, the interpretation of torsional oscillator measurements in terms of a superflow in a ^{4}He crystal remains controversial. Note in this connection that the observations of KC could be explained, at least in principle, by classical mechanism of dislocation-induced plasticity of solid helium. With appropriate temperature dependence of the dislocation mobility both a reduction in the rotational inertia and a temperature peak in the damping of oscillations can be obtained, as in the Granato-Lücke theory [14]. Similar interpretations have been suggested in [15, 16].

At the moment, no other evidence for superflow in bulk solid helium has been found. Recent searches for a pressure driven superflow have given null results [17, 18]. On the other hand, Sasaki et al. [19] have detected a superflow presumably at grain boundaries in polycrystalline solid helium in contact with the superfluid phase. They observed this phenomenon even at temperatures as high as 1.1 K, which is too high compared to the supposed supersolid transition temperature of 0.2 K (and is not very far from the superfluid transition temperature in the bulk liquid). Thus it is not clear yet whether

[1)]e-mail: todo@boojum.hut.fi

this interesting observation is relevant to the KC experiments.

If the observed onset of NCRI really is a manifestation of a phase transition in the bulk solid, the equilibrium thermodynamic properties of the solid should also display an anomaly. The magnitude of this anomaly in the case of Bose condensation of vacancies can be estimated as $\delta S \sim R\rho_s/\rho$, where δS is the excess entropy due to vacancies just above the transition temperature T_c and R is the gas constant. Below T_c the excess entropy should drop to zero. With $\rho_s/\rho \sim 10^{-3} \div 10^{-2}$ it gives very large δS, which certainly has been ruled out by heat capacity measurements [6]. However, this naive estimate is valid only in the case of weakly interacting Bose gas. As an alternative, Anderson et al. [20] have suggested a model of the supersolid ground state with a number of strongly correlated vacancies and interstitials. In this model, there are no soft modes (in contrast to the Andreev-Lifshitz scenario), and δS may be very small even with relatively large superfluid fraction. Another model of a ground state with a low density of strongly correlated vacancies/interstitials was suggested by Dai et al. [21].

Thus at present there is no theoretical consensus on possible magnitude of the entropy change associated with the supposed supersolid transition. As for the experimental data, a few of earlier measurements have revealed deviations from conventional Debye behavior below 0.5 K, but later experiments have not reproduced any of such anomalies [6]. The most recent studies [22] have found a small excess heat capacity at temperatures down to 80 mK, which corresponds to $\delta S \sim 10^{-7}R$ at 300 mK, however, no indication of a phase transition near 200 mK has been found. The annealing effect has not been studied, and the nature of the observed anomaly has remained unclear. Thus it seems very important to look for any anomaly in the entropy of high-quality ^{4}He crystals around 0.2 K.

Recently we have reported on our direct high-precision measurements of the melting pressure of ^{4}He of regular purity ($\approx$ 80 ppb of ^{3}He impurities) in the range from 10 to 400 mK [23]. The melting pressure showed the expected T^4 dependence, and the coefficient was in excellent agreement with available data on the sound velocity in the liquid and the Debye temperature of the solid ^{4}He. However, we have observed an anomaly below about 100 mK, where T^4 dependence changed to much weaker, almost linear dependence. It was not clear at that time, what was the origin of the low temperature anomaly: influence of ^{3}He impurities, effects due to change in the crystal shape, or some other reason.

In this Letter we present a new set of high-precision measurements on the melting curve of ^{4}He with ultrapure (0.3 ± 0.08 ppb of ^{3}He impurities)[2)] helium where we have used an interferometer [24] to monitor the crystal shape. In addition, we have used a cryogenic valve to close the cell containing only liquid ^{4}He and measured the temperature dependence of the sensitivity of the pressure gauge. We have found out that the low temperature anomaly which we have observed in our previous experiments with ^{4}He of regular purity [23] is also present for high purity ^{4}He sample, and that it is not due to the change in the crystal shape. However, the same anomaly was observed for the pressure which was measured in the liquid at a constant volume. Thus our results prove that the reason for the observed anomaly is the small ($2 \cdot 10^{-7}$) decrease of the spring constant of our pressure gauge membrane below 100 mK. After applying the correction on the observed temperature dependence of the sensitivity of the gauge, the measured variation of the melting pressure of ^{4}He below 320 mK does not deviate from the pure phonon T^4 law with the accuracy of about 0.5 μbar. This sets the upper limit of $\sim 5 \cdot 10^{-8}R$ for a possible excess entropy in the solid ^{4}He below 320 mK.

Our capacitive pressure gauge, of a standard Straty-Adams design [25], is made of beryllium bronze (Be-Cu) and has the sensitivity $dC/dp = 41$ pF/bar at the melting pressure of 25.31 bar. The capacitance of the gauge was measured by the commercial Andeen-Hagerling 2500 A bridge yielding the accuracy of about 0.5 μbar after ~ 1 min averaging. Temperature was measured by a ^{3}He melting curve thermometer anchored to the sample cell. The ^{3}He melting pressure was converted to temperature according to the Provisional Low Temperature Scale, PLTS-2000 [26].

Crystals were nucleated and grown at constant temperature by slowly increasing the pressure in the cell. In order to create crystals in the field of view, we have used a capacitive nucleator which was operated with high voltage. During measurements on the melting pressure at different temperatures, crystals were imaged with a low-temperature Fabry-Pérot interferometer [24]. Crystals were typically grown to about 1 cm in diameter, and during cooling or warming their height and curvature did not change remarkably, ensuring that no hydrostatic or capillary corrections to the measured pressure were needed.

[2)]We are grateful to K.O. Keshishev for providing us with a sample of this ultra-pure helium. The analysis of the sample purity was made by the US Bureau of Mines.

The results of the measurements on the melting pressure with ultra-pure ^{4}He were similar to the results obtained with ^{4}He of regular purity [23], see Figs.1 and 2.

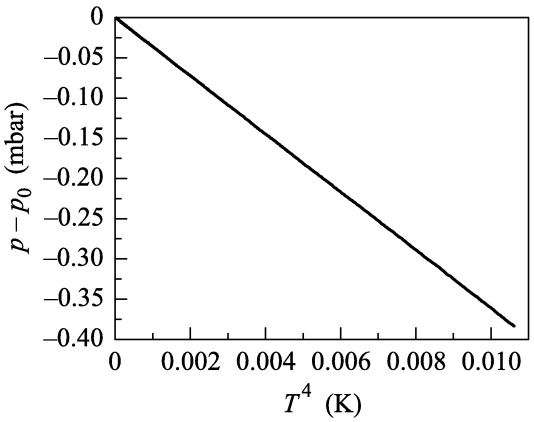

Fig.1. Melting pressure of ultra-pure ^{4}He below 320 mK. The presented curve consists of 1000 raw data points

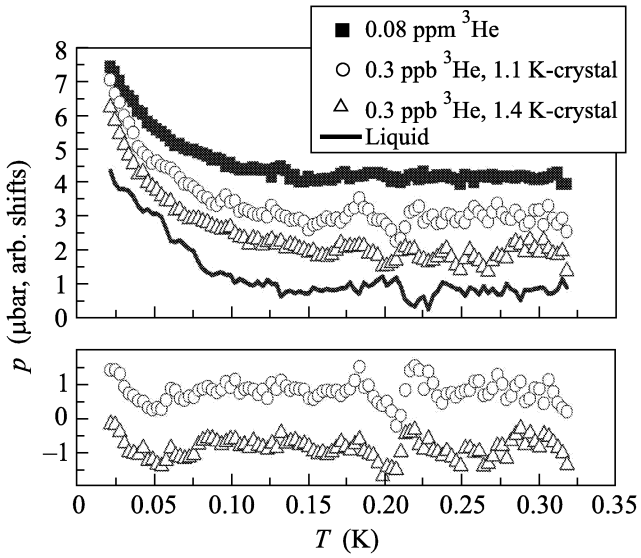

Fig.2. Upper part. Deviation of the measured pressures from the corresponding T^4 fits: melting pressure of ^{4}He of regular purity (■), melting pressure of ultra-pure ^{4}He measured with crystal grown at 1.1 K (○) and with crystal grown at 1.4 K (△), pressure in the liquid at a constant volume (solid line). Lower part. Deviation of the difference between the melting pressure and the pressure in the liquid at a constant volume from the best T^4 fit. Symbols refer to the same crystals as in the upper part. All curves are offset for clarity

The sample crystals were nucleated and grown at 10 mK, at 1.1 K where melting pressure is 80 mbar higher than at low temperatures, and at 1.4 K where melting pressure is 700 mbar higher than at low temperatures. The sensitivity of our pressure gauge is high enough to check (1) the effect of non-hydrostatic stresses [27] produced by the change of the melting pressure when the crystals grown at 1.4 K were cooled down and (2) the effect of disorder due to possible plastic deformation of such crystals. However, after subtracting off the best T^4 fit to the data, no other reproducible contribution to the melting pressure is seen above 100 mK with the accuracy of $\sim 0.5\,\mu$bar. It means that non-hydrostatic stresses in samples grown at 1.4 K had been effectively annealed during the first cooldowns of such samples. The low temperature anomaly below 100 mK is present for all ultra-pure and normal purity samples which proves that it is not due to ^{3}He impurities.

Another possible reason for the anomaly could be the corresponding temperature dependence of the pressure gauge sensitivity. The flexible membrane of the pressure gauge is made of beryllium bronze, widely used material for low temperature experiments because of its high tensile strengh and low losses on mechanical deformations. However, it is known that some types of beryllium bronze may have low temperature anomaly in the heat capacity [28] and Young modulus [29, 30]. The anomaly we have observed in the melting pressure of ^{4}He below 100 mK would correspond to a very small, $\sim 2 \cdot 10^{-7}$, relative decrease of the Young modulus of the bronze (which is still several orders of magnitude higher compared to conventional metals). The only way to detect so small change in the pressure gauge sensitivity is to measure the temperature dependence of the capacitance of the gauge at a constant pressure, or, at a pressure which depends on temperature in a known way.

We have measured the pressure in liquid ^{4}He just below the melting curve at a constant volume. Our cryogenic valve, placed on the mixing chamber plate, was found to keep the filling line of the cell closed with very small ($< 0.5\,\mu$bar/day) leakage, thus making careful measurements of the pressure in the liquid possible. The variation of the pressure at a constant volume is due to thermal expansion of the liquid and can be expressed as

$$\left(\frac{\partial p_L}{\partial T}\right)_V = -\frac{\rho}{V}\left(\frac{\partial S}{\partial \rho}\right)_T .$$

In the low temperature limit the thermodynamics of the liquid is dominated by phonons, $S \propto T^3/c^3$, which gives $(\partial p_L/\partial T)_V = 3uS/V$, where $u = (\rho/c)(\partial c/\partial \rho)_T$ is the Grüneisen constant. As a result, the pressure of the liquid at a constant volume varies as T^4.

Indeed, in the range of 100 ... 320 mK the measured pressure in the liquid obeys T^4 law, while below 100 mK we again observed the same low temperature anom-

aly (see Fig.2), apparently due to the temperature-dependent sensitivity of our pressure gauge. The measured capacitance C of the gauge can be written in the form $1/C = 1/C_0 - Ap/Y$, where Y is the Young modulus of the bronze and A is a constant which depends on the gauge design. Thus a relative change $\delta Y/Y$ in the Young modulus results in the equal relative change $-\delta p/p$ in the measured pressure.

Below 100 mK, where T^4 contribution to the pressure due to thermal expansion of the liquid is only about $0.5\,\mu$bar, the change of the sensitivity by about $5\,\mu\text{bar}/25\,\text{bar} = 2 \cdot 10^{-7}$ is obvious. However, there might be some small variation of the sensitivity also above 100 mK which would be mixed with the real pressure change. The correct way to eliminate the contribution of the pressure gauge to the measured pressure is to subtract the data measured in the liquid from the data measured on the melting curve and look for a non-phonon contribution in the residual. The results of the subtractions are shown in Fig.2 (lower part). With the accuracy of $0.5\,\mu$bar, the measured melting pressure of ^{4}He can be described by the T^4 dependence due to phonons and no sign of any phase transition is seen.

To summarize, we have carried out high-precision measurements on the melting pressure of ultra-pure ^{4}He down to 10 mK with several samples grown at different pressures. The shape and height of the crystals were carefully controlled by the interferometric imaging so that no corrections for the Laplace pressure and hydrostatic pressure were needed. All samples were single crystals, without any signs of grain boundaries or other macroscopic defects. The growth thresholds for crystals were less than $1\,\mu$bar, which guarantees low density of dislocations (less than $10^2\,\text{cm}^{-2}$). We have calibrated the sensitivity of our pressure gauge with the accuracy of $2 \cdot 10^{-8}$ by measuring the variation of the pressure in the liquid ^{4}He at a constant volume. This calibration allowed us to eliminate the low temperature anomaly observed below 100 mK [23]. As a result, we have found that the melting pressure of ^{4}He does not deviate from the T^4 law due to phonons with the accuracy of $0.5\,\mu$bar and there is no sign of a supersolid transition down to 10 mK.

In conclusion, we would line to stress that our measurements do not rule out the possiblity of a supersolid transition in high-quality ^{4}He crystals. They only set the upper limit of $\sim 5{\cdot}10^{-8}R$ for the non-phonon entropy in such crystals at the melting pressure below 320 mK.

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