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Elementary Excitations in Solid and Liquid ^{4}He at the Melting Pressure

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Abstract

Recent discovery of a nonclassical rotational inertia (NCRI) in solid ^{4}He below 0.2 K by Kim and Chan has revived great interest in the problem of supersolidity and initiated intensive study on the properties of solid ^{4}He. A direct proof that the onset of NCRI corresponds to the supersolid transition would be the observation of a corresponding drop of the entropy of solid ^{4}He below the transition temperature. We have measured the melting pressure of ultrapure ^{4}He in the temperature range from 0.01 to 0.45 K with several single crystals grown at different pressures and with the accuracy of 0.5 μbar. In addition, supplementary measurements of the pressure in liquid ^{4}He at constant volume have been performed, which allowed us to eliminate the contribution of the temperature-dependent properties of the pressure gauge from the measured melting pressure data. With the correction to the temperature-dependent sensitivity of the pressure gauge, the variation of the melting pressure of ^{4}He below 320 mK obeys the pure T^4 law due to phonons with the accuracy of 0.5 μbar, and no sign of the transition is seen (Todoshchenko et al. in JETP Lett. 85:454, 2007). This sets the upper limit of $\sim 5 \cdot 10^{-8} R$ for a possible excess entropy in high-quality ^{4}He crystals below 320 mK. At higher temperatures the contribution from rotons in the superfluid ^{4}He has been observed. The thermal expansion coefficient of the superfluid ^{4}He has been measured in the range from 0.01 to 0.7 K with the accuracy of $\sim 10^{-7}$ 1/K, or by two orders of magnitude better than in previ-

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ous measurements. The roton contributions to the melting pressure and to the pressure in liquid at a constant volume are consistent and yield the value of 6.8 K for the roton gap, which is very close to the values obtained with other methods. As no contribution due to weakly interacting vacancies to the melting pressure of ^{4}He has been observed, the lower limit of about 5.5 K for their activation energy can be set.

Keywords Quantum solids · Supersolids · Excitations in quantum systems

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1 Introduction

In 2004 Kim and Chan [2, 3] have observed about 1% reduction of the rotational inertia of the solid ^{4}He sample below $T_r \approx 0.2$ K and they have interpreted it as the onset of supersolidity in solid ^{4}He. To date, several experimental groups have confirmed the effect of NCRI [4–7], but its physical nature yet remains unclear.

Originally, supersolidity in quantum crystals was suggested by Andreev and Lifshitz [8] and Chester [9] as a result of Bose-Einstein condensation (BEC) of weakly interacting vacancies (or interstitials). Due to quantum delocalization, the activation energy Φ of vacancies may become zero. In such a case the equilibrium concentration of vacancies is finite down to the absolute zero (zero-point vacancies), and at low enough temperature these vacancies condense to the ground state resulting in superfluid behavior of the crystal. However, different experiments have revealed that the activation energy of vacancies in solid ^{4}He is finite at all densities. On the other hand, sufficiently high non-equilibrium concentrations of vacancies even with $\Phi > 0$ may probably be created at low temperatures by some way, also resulting in BEC [8]. Nevertheless, even in this case there must be a huge drop in the entropy—of the order of $(\rho_s/\rho)R$—below T_r which has never been observed in thermodynamic measurements, and thus this possibility does not have any experimental ground yet (see [10] for a review).

The annealing effect, discovered by Rittner and Reppy [4], indicates that the observed NCRI should be attributed, at least partially, to some kind of quenched disorder [7, 11–13]. Clark, West and Chan [12] reported on observation of a small NCRI fraction even in carefully annealed possibly single crystals of ^{4}He. However, the annealing procedure cannot guarantee elimination of all quenched defects, and thus no convincing conclusion on supersolidity of a perfect helium crystal can be done. In the theory, various explanations of the observed NCRI have been presented, both with [14–18] and without quenched disorder involved [19–22].

A search for thermodynamic signature of the onset of NCRI recently yielded observation of a small peak in the heat capacity of solid helium below 200 mK [23–25]. Similarly to the NCRI fraction, the amplitude of this peak decreases upon annealing, but still remains noticeable even with ultrapure, best quality crystals. Here again the question is whether this peak originates from some remnant disorder or it is an inherent property of a perfect crystal.

In the present work we report on our very accurate measurements on the melting pressure of ^{4}He in the range from 0.01 to 0.45 K aimed to look for any sign of the additional entropy in solid and of the possible supersolid transition. The important feature of our measurements is that the crystal is always in equilibrium with liquid phase, thus providing the best possible conditions for annealing defects. Perfect faceting during growth and low facet growth thresholds (less than 1 μbar) guarantee extremely low degree of disorder (density of dislocations less than 10^2 cm^{-2}) [26] of our crystals.

In order to separate the entropy of solid from the entropy of liquid and from the possible effects of the temperature-dependent sensitivity of the pressure gauge, we have measured also the pressure in the liquid ^{4}He at constant density close to solidification. We did not find any sign of the supersolid transition down to 10 mK. At the same time, a contribution from weakly interacting vacancies to the melting pressure has not been observed either. In contrast, T^8 term was found to be consistent with the heat capacity measurements by Gardner et al. [27], but on the limit of our accuracy. Thermal expansion coefficient of liquid ^{4}He has been measured with the accuracy of $\sim 10^{-7}$ 1/K, by two orders of magnitude better than it has been measured before, and the obtained results differ significantly from the earlier data [28, 29].

2 Experimental Details

We have performed our measurements in the optical cell [30] which allowed us to control the crystal shape and thus exclude the possible capillary and hydrostatic contributions on the measured melting pressure. Our capacitive pressure gauge is of a standard Straty-Adams design [31]. The sensitive element is a Be–Cu membrane to which one of the electrodes is glued. The sensitivity of our pressure gauge is $dC/dp = 41$ pF/bar at the ^{4}He melting pressure. Temperature was measured by the ^{3}He melting curve thermometer anchored to the sample cell. We have also a cryogenic valve with very small leakage, less than 0.5 μbar/day, which allowed us to isolate the cell containing only liquid ^{4}He and make accurate measurements on thermal expansion of liquid ^{4}He.

Besides commercially pure ^{4}He (0.08 ppm of ^{3}He impurities), we also used ultrapure ^{4}He (0.4 ppb of ^{3}He).[1] Our solid samples were grown at low temperatures ($p_m = 25.3$ bar), at 1.1 K ($p_m = 25.4$ bar) and at 1.4 K ($p_m = 26.0$ bar). No systematic difference between the data measured with samples grown from liquid helium of different purity and at different pressures has been observed.

3 Results

The melting pressure of ^{4}He has been measured in the range from 0.01 to 0.45 K with the accuracy of $\approx$0.5 μbar. To exclude the effects caused by temperature-dependent

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[1] We are grateful to K.O. Keshishev for providing us with a sample of this ultrapure helium. The analysis of the sample purity was made by Institut für Umweltphysik, Universität Bremen.

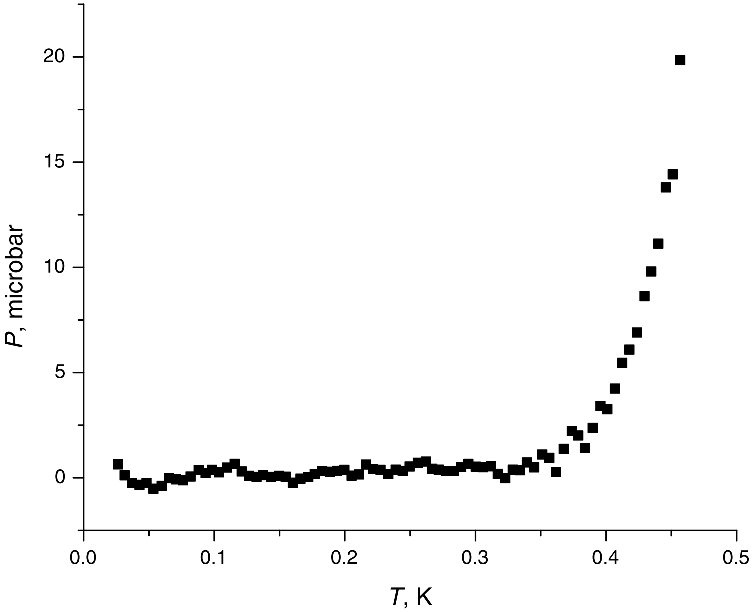

Fig. 1 Difference between the measured melting pressure and pressure in the liquid at constant volume; best T^4 fit below 0.32 K is subtracted. Note that the subtracted T^4 term is about 500 μbar at 0.32 K

properties of the Be–Cu membrane of the pressure gauge, we have also measured the pressure at a constant density in the liquid ^{4}He just below the melting curve and subtracted it from the measured melting pressure. At low temperatures, the contribution to the melting pressure from the phonons in solid and in liquid is proportional to T^4.

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, $S \propto VT^3/c^3$, which gives $(\partial p_L/\partial T)_V = (3u+1)S/V$, 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 also varies as T^4.

After subtracting the pressure measured in the liquid from the measured melting pressure, the data at low temperatures fits with the T^4 dependence very well, the residual being less than 1 μbar, see Fig. 1. Above 0.32 K the result of subtraction starts to deviate from T^4 behavior indicating additional entropy in the liquid. This must be the contribution from rotons to the entropy of the liquid. To confirm that, we first fitted the data measured in the liquid at constant volume with T^4 term plus roton

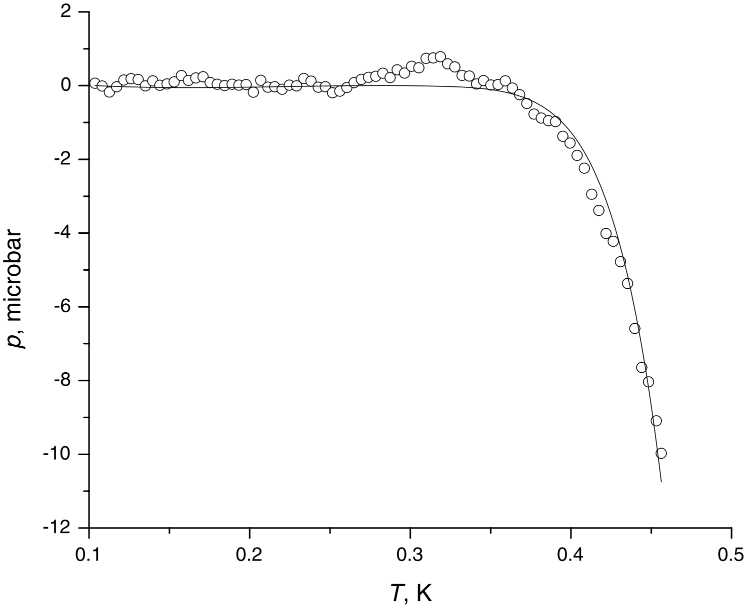

Fig. 2 Measured pressure in the liquid at constant density just below the melting curve. *Solid line* is the fit with the roton contribution (see text). T^4 term is subtracted. Deviation near the melting curve minimum of ^{3}He, $T_{\min} = 0.315$ K, is because of poor sensitivity of ^{3}He melting curve thermometer in this range

contribution,

$$p_{L,rot} = \frac{\partial F}{\partial V}\left(\frac{\Delta}{k_B T}\right)^{-3/2} \exp\left(-\frac{\Delta}{k_B T}\right) \\ - F\frac{\partial \Delta}{k_B \partial V}\left[\left(\frac{\Delta}{k_B T}\right)^{-1/2} + \frac{1}{2}\left(\frac{\Delta}{k_B T}\right)^{-3/2}\right]\exp\left(-\frac{\Delta}{k_B T}\right), \qquad (1)$$

where the prefactor $F = 2Vk_B m_0^{1/2} p_0^2 \Delta^{1/2}/(2\pi\hbar^3)^{3/2}$ and the derivatives were taken from Greywalls measurements [32, 33], so that the roton gap Δ was the only fitting parameter.

As it is seen from Fig. 2, the fit is rather good, giving the value of 6.8 K for the roton gap, which is quite close to the values obtained by other methods, being slightly smaller. The reason might be in the T^5 and T^7 terms in the entropy of the liquid, which are not extremely small above 0.4 K [32, 33]. Then we may consider (1) as just the form of presentation of all non-T^4 contributions to the pressure of liquid with the value of Δ as a fitting parameter. This form is physical because the dominating contribution is still due to rotons, but the value of the gap could be found slightly different from the real one.

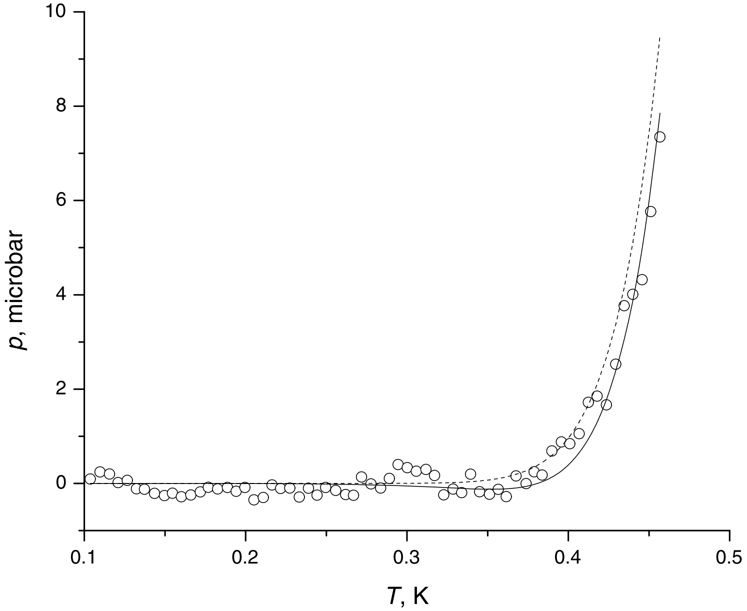

Fig. 3 Measured melting pressure after subtraction of the T^4 term. *Dashed line*—contribution from rotons in the liquid with the gap value $\Delta = 6.8$ K which was obtained from the data on pressure in the liquid. *Solid line*—contribution from rotons plus T^8 term taken from measurements by Gardner et al. [27] (see text). Lines are not fits!

Then we used the obtained value of the roton gap to calculate the contribution from rotons to the melting curve,

$$p_{mc,rot} = \frac{F}{\Delta V_{ls}} \frac{\Delta}{k_B} \left(\frac{\Delta}{k_B T}\right)^{-3/2} \exp\left(-\frac{\Delta}{k_B T}\right),$$

and compared the result with the measured melting pressure, Fig. 3. We see that the roton contribution with the gap value taken from the liquid data, the dashed curve in Fig. 3, explains most of the deviation of the melting curve from T^4 law, but goes slightly above the experimental points. The difference could be attributed to T^8 term which corresponds to the T^7 term in the heat capacity of solid ^{4}He observed by Gardner et al. [27], $C_7 = 11.4\ \mathrm{mJ/(mole\ K^8)}\ T^7$. Adding the corresponding term to the melting pressure, $p_{mc,8} = -(1/56)(C_7/\Delta V_{ls})\ T = -860\ (\mu\mathrm{bar/K^8})\ T^8$, we obtained almost exact coincidence with the measured data points (the solid line in Fig. 3). Nevertheless, the observed T^8 term is on the limit of our accuracy, being about 2 µbar at 0.45 K.

We can conclude that the melting curve of ^{4}He in the range 0.01 . . . 0.45 K is explained by phonons in both phases, rotons in the liquid and T^7 term in the entropy of solid with the accuracy of 0.5 µbar. The absence of the contribution from free vacancies allows us to set the lower limit for their activation energy. Free vacancies

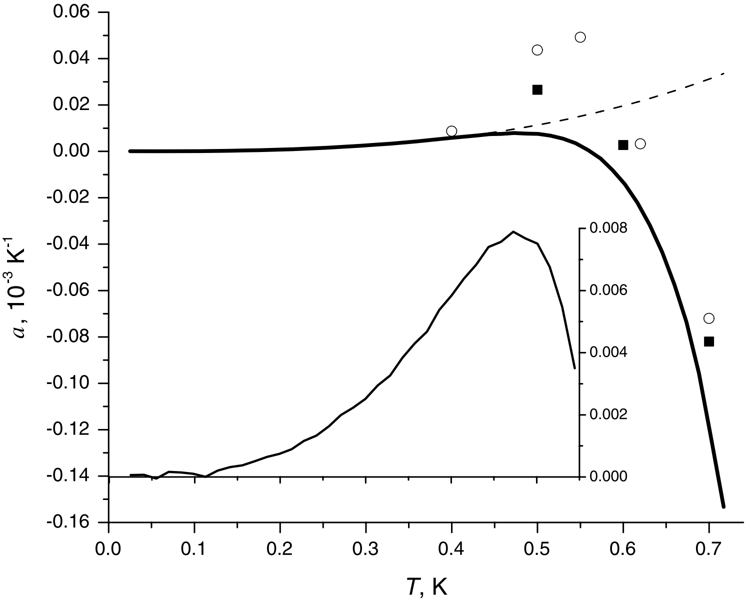

Fig. 4 Thermal expansion coefficient of liquid ^{4}He at 25.1 bar. *Solid line*—present work, *circles*—measurements by Grilly [28], *squares*—measurements by Mills and Sydoriak [29], *dashed line*—calculation in the phonon limit using sound velocity data [36], see text. Insert is the expanded along the Y-axis view of our data

should contribute

$$p_{mc,free\ vac} = \frac{V_s}{\Delta V_{ls}}\left(\frac{M}{2\pi\hbar^2}\right)^{3/2}(k_B T)^{5/2}\exp\left(-\frac{\Phi}{k_B T}\right) \tag{2}$$

to the melting pressure, where M is the effective mass of vacancies. At high temperatures the activation energy Φ measured by different methods shows drastic reduction with the decrease of density down to 6–8 K close to melting. At low temperatures the density of the solid at melting pressure is the lowest possible, and one may expect even smaller values of Φ [34]. However, our measurements do not show much smaller value of the activation energy. According to (2), with the activation energy $\Phi = 4$ K and the mass M equal to ^{4}He atomic mass, the contribution from vacancies to the melting pressure would be as large as 40 μbar at 0.45 K. In fact, this is the lower limit for the contribution, because different calculations give the value of the bandwidth B from 1 to 10 K [35] which corresponds to the effective mass $M \approx 4\hbar^2/(Ba^2) = 1\ldots 10\, m_4$.

As the dependence on Φ is strong, we can set quite accurate lower limit for the activation energy of free vacancies to be 5.5 K, which would result in the contribution of 1.5 μbar at 0.45 K.

As there is the lack of the available accurate data on the thermal expansion of the liquid ^{4}He, we have extended our measurements on the pressure in the liquid ^{4}He at constant density up to 0.72 K. Our accuracy on the thermal expansion coefficient $\alpha = -(1/\rho)(\partial\rho/\partial T)_p$ is about 10^{-7} 1/K (see Fig. 4) which is two orders of magnitude better than in the well-known measurement by Grilly [28]. Unexpectedly, we have found large deviation from the previous measurement by Grilly and by Mills and Sydoriak [29]. First, we have located a maximum in thermal expansion at about 0.47 K instead of 0.53 K, measured by Grilly. Second, the value of α at maximum is by an order of magnitude smaller than in measurements by Grilly and by Mills and Sydoriak. To check our data independently, we have evaluated the thermal expansion coefficient using available data on the sound velocity in liquid ^{4}He. At low temperatures, where phonons dominate, thermal expansion can be expressed as

$$\alpha = \beta(1+3u)\frac{S}{V} = \beta\frac{2\pi^2}{45}\frac{k_B^4}{\hbar^3 c^3}(1+3u)\times T^3, \tag{3}$$

where $\beta = (1/\rho)(\partial\rho/\partial p)_T$ is the isothermal compressibility of the liquid. Substituting $c = 366$ m/s, $u = 2.21$ [36], and $\beta = 4.5 \cdot 10^{-3}$ bar^{-1}, we obtained $\alpha = 9.0 \cdot 10^{-5} T^3/\mathrm{K}^4$, and the corresponding line lies exactly on our measured data points below 0.4 K (Fig. 4, dashed line).

4 Discussion

Our measurements on the melting pressure of ^{4}He up to 0.45 K show the T^4 contribution from phonons in solid and in liquid, the contribution from rotons in liquid with the gap value of 6.8 K and the T^8 term corresponding to T^7 term in the entropy in solid (the latter is on the limit of our accuracy). We set the upper limit of $\sim 5 \cdot 10^{-8} R$ for other contributions to the entropy of solid ^{4}He. No sign of any transition has been observed down to 0.01 K. Absence of the contribution from weakly interacting vacancies indicates that their activation energy is as large as at high temperature, 6–8 K, and does not decrease remarkably at low temperatures (low densities), the lower limit of 5.5 K can be set by our measurements.

On the other hand, free vacancies do not contribute to the thermodynamics of ^{4}He crystals also at higher temperatures. Gardner et al. have found only T^3 and T^7 terms in heat capacity up to 1.2 K, while simple estimate using

$$C_{vac} = V_s k_B \left(\frac{M\Phi}{2\pi\hbar^2}\right)^{3/2} \left(\frac{\Phi}{k_B T}\right)^{1/2} \left[1 + 3\frac{k_B T}{\Phi} + \frac{15}{4}\left(\frac{k_B T}{\Phi}\right)^2\right] \exp\left(-\frac{\Phi}{k_B T}\right)$$

with $\Phi = 8$ K gives 50 mJ/(mol K), which is larger than the measured T^7 term, $C_7 = 39$ mJ/(mol K) [27] and about 20% of total heat capacity at 1.2 K. Thus the model of free weakly interacting vacancies in the solid ^{4}He seems not to explain all available experimental data.

The nature of the large T^7 term in entropy of solid ^{4}He is still unclear. It has been shown recently by Maris and Balibar [37] that this term may still be explained by the phonon dispersion, rough estimates of which give the right order of magnitude. Of course, it is very difficult to explain the absence of T^5 term while T^7 term is about 20% of the total heat capacity. On the other hand, Anderson et al. [20] attribute T^7 term to the strongly-correlated vacancies, and have found the same order of magnitude for this term by fitting the data on the lattice constant, which is very intriguing.

Further experiments at even lower temperatures and with higher accuracy are still needed as well as more developed theories to understand the nature of solid ^{4}He.

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