

Acoustic Properties of Solid ^4He in the Limit of Zero Impurity

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Received: 30 June 2009 / Accepted: 7 September 2009 / Published online: 23 September 2009
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Abstract We have studied the elasticity of solid ^4He in relation with its possible supersolidity. For this we have measured acoustic resonance frequencies in a 1 cm^3 cell filled either with polycrystals or with single crystals of ^4He . We have observed a large stiffening at low temperature as first observed by Day and Beamish in polycrystals. The ^3He impurity content has been varied from 300 ppb to 0.4 ppb. When kept in equilibrium with liquid helium, single crystals should be impurity free. In these crystals, a large stiffening is observed, which should not be the result of the pinning of dislocations by impurities.

Keywords Solid helium 4 · Elasticity · Supersolidity

PACS 67.80.-s

1 Introduction

The existence of supersolidity is well established now. In three different types of experiments, several groups have found anomalies in the rotation properties [1–14], in the specific heat [15] and in the elastic properties [13, 16, 17] of ^4He crystals. All these anomalies are very sensitive to the disorder of the samples and to the concentration of ^3He impurities. Surprisingly, Day *et al.* [13, 16] have discovered that

This work was supported in part by the French ANR grant BLAN07-1-215296 and by the US National Science Foundation grant No. DMR-0605355.

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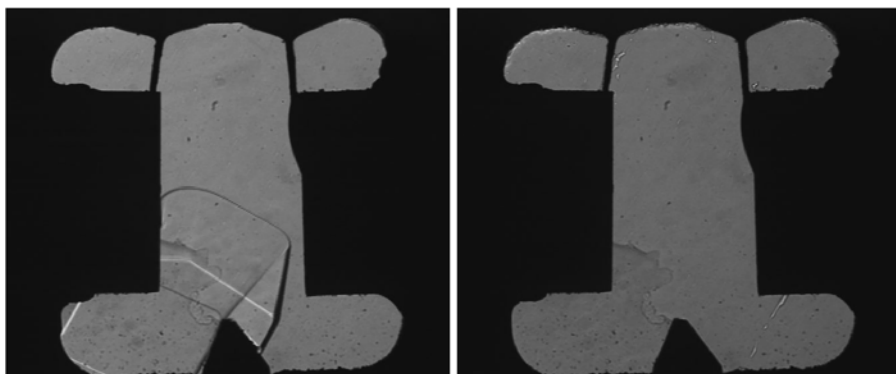


Fig. 1 Two photographs of the single crystal X6 in the 1 cm³ cell. *Left*: during growth. *Right*: after growth and in equilibrium with a few liquid droplets. The two black rectangular objects are the piezo-electric transducers with their electrical connexions on top

when supersolidity appears, the shear modulus μ of solid samples *increases*. Day *et al.* [16] have attributed this increase to ³He impurities which bind to dislocations and pin them below a temperature in the range 50 to 200 mK which depends on their concentration.

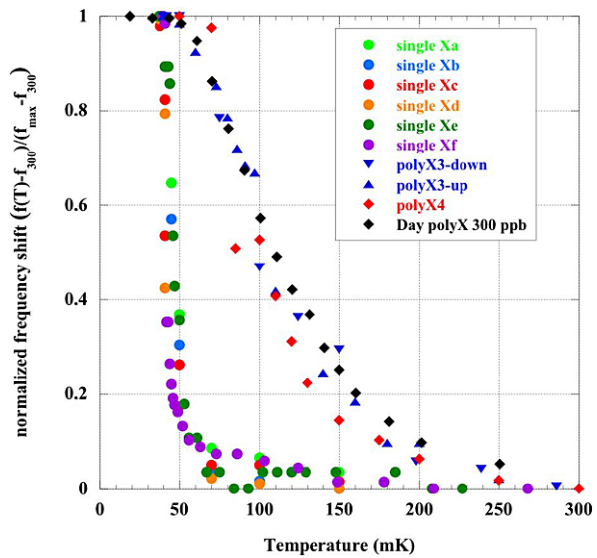
It seems somewhat surprising that an interpretation in terms of dislocations is relevant for samples which are mostly polycrystalline with a large density of grain boundaries [4, 18]. We have made acoustic measurements similar to those previously done by Day *et al.* in order to compare polycrystals to single crystals. By doing so, we have found evidence that the interpretation in terms of pinning of dislocation is not sufficient because we have observed stiffening in single crystals which are in equilibrium with liquid helium so that their ³He impurity concentration should be negligible.

2 Experimental Setup

As shown by the photographs in Fig. 1, we have placed two piezo-electric transducers in a nearly cubic cell made by cutting a square hole (11 × 11 mm²) in a 10 mm thick copper plate which is closed by two glass windows (6 mm thick). Between the windows and the copper plate, indium rings allow the cell to be leak-tight up to 65 bar at least. Thanks to large windows on the cryostat itself, helium crystals can be observed from the outside with an ordinary camera. The transducers are transverse PZT ceramics¹ providing a displacement in the vertical direction (about 0.8 Å/V at low temperature). They resonate around 500 kHz but they are used near 30 kHz to excite and detect acoustic resonances *inside the cell*. We have used very low levels of excitation, typically 1 mV, in order to stay below the threshold for non-linear elastic behavior even at resonances whose quality factor is of order 1,000. We have found

¹PZT “shear bars” made of material C5800 by Channel Industries Inc.

Fig. 2 (Color online) The shift in resonance frequency of solid samples prepared with ^4He containing 300 ppb ^3He impurities. For polycrystals (diamonds and triangles) the frequency increases as the temperature is reduced below an onset temperature of about 200 mK. For single crystals, the onset temperature is much lower, about 50 mK



that this threshold corresponds to a strain of order 10^{-8} , in agreement with Day *et al.* In classical crystals, this threshold is of order 10^{-2} , comparable to the plasticity threshold which is itself close to the fracture limit. Finding a threshold lower by 6 orders of magnitude is a strong indication that the mobility of defects in quantum crystals plays an important role in their elastic properties.

3 Poly- and Single Crystals Grown from Natural ^4He

We first used natural ^4He which is assumed to contain 300 ppb ^3He impurities. We prepared polycrystals by the “blocked capillary method” as done by Day *et al.*, that is by crystallizing a liquid sample at 63 bar and constant volume. As shown previously [18], this method ends up with a polycrystalline sample at 33 bar with grain sizes in the micrometer range. We have also prepared polycrystals by fast injection of mass at 100 mK through the fill line of the cell, in order to compare with a polycrystal at 25 bar. This type of polycrystal was shown by Sasaki *et al.* [18] to have grain sizes of order 50 μm . Both polycrystals (X3 and X4 on Fig. 2) show a stiffening at low temperature which is very similar to what has been found before by Day *et al.* The amplitude of the stiffening varies from one sample to the other but, once normalized by the maximum variation, the data fall on a universal curve. The effect is apparently independent of pressure and frequency (data are taken at 25 and 33 bar, and we work at 30 kHz while Day *et al.* work from 20 Hz to 8 kHz).

We then prepared single crystals which had to be obtained by slowly injecting mass into the cell at constant temperature and pressure. One difficulty was that the walls are preferentially wet by the liquid phase (the contact angle of the liquid-solid interface is about 45° [18, 19]) so that the fill line blocks before the liquid crystallizes in several corners. A future cell will have a fully convex geometry and the orifice of the fill line at the highest point of the cell [20] but, with the present cell, we had to fill

the cell as much as possible at 1.45 K before cooling down so that the rest of the liquid regions crystallizes. When cooling from 1.45 K to 1 K, the liquid-solid equilibrium pressure decreases from 26.3 to 25.3 bar so that stresses damage the crystal quality. If done too quickly, the crystal breaks. In most cases, the final sample is a single crystal in equilibrium with a few liquid droplets which have a typical volume of 1 mm^3 (see the picture on the right of Fig. 1).

We have studied the acoustic resonances in these low quality single crystals. Figure 2 shows the frequency shift normalized by its low temperature value. The real shift varies from sample to sample. No systematic difference in shift amplitude was observed between poly- and single crystals but it occurs at much lower temperature in single crystals: the onset is around 50 mK instead of 200 and the transition was not yet completed at the lowest temperature we could achieve (36 mK). There are at least 3 possible interpretations for this observation: either the binding energy of ^3He impurities is much smaller on *dislocations*—which may explain the anomalies in single crystals—than on *grain boundaries* which may be relevant for polycrystals, or the purity of single crystals is different from that of polycrystals, or the stiffening is not, or *not only*, due to the pinning of dislocations proposed by Day *et al.* In order to progress, we have repeated the experiment with ultrapure ^4He containing 0.4 ppb ^3He only.

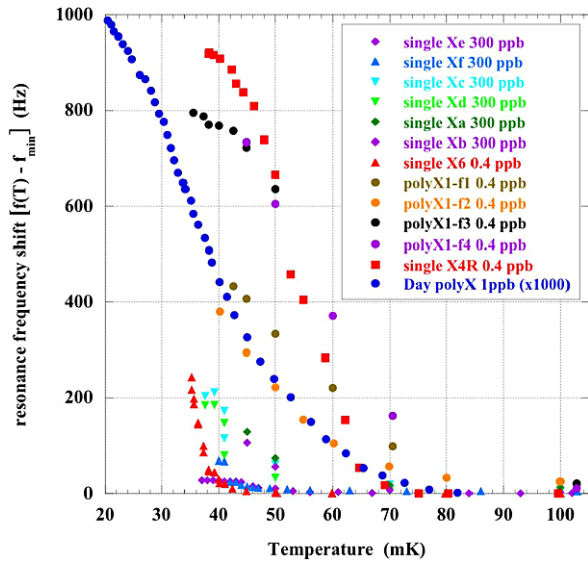
4 Ultrapure Samples

When preparing ultrapure *polycrystals*, we found a stiffening once more in agreement with the results previously obtained by Day *et al.* (this time with 1 ppb ^3He): the onset of stiffening is around 70 mK. Note that, for comparison, in Fig. 2 we have multiplied Day's normalized variation by 1,000 because, in our cell for the mode studied, we have calculated numerically that the frequency shift is one third of the modulus shift ($\Delta f/f = 0.33\Delta\mu/\mu$) so that Day's change by 10% in μ should induce a change by 1,000 Hz in our experiment where f is around 30 kHz. The ultrapure *single crystals* gave the same sharp stiffening below 40 mK which we had already observed with normal purity single crystals. At this stage, we realized that the energy of ^3He atoms is lower by more than 1 K in the liquid than in the solid. We have calculated this effect [20] by using the theory by Edwards and Balibar [21]. The ratio of the concentration in the hcp solid to the concentration in the liquid is

$$\frac{X_3^h}{X_3^L} = \frac{4.42}{T^{3/2}} \exp\left[-\frac{1.359}{T}\right] \quad (1)$$

which is 10^{-12} at 40 mK. Furthermore, the diffusion coefficient D of ^3He atoms in solid ^4He is large, especially for low X_3^h , as shown by Schratter and Allen [22] who found $DX_3^h = 2.4 \times 10^{-11} \text{ cm}^2/\text{s}$. It means that even if during growth ^3He atoms enter the solid, the impurities diffuse fast enough on a macroscopic distance (1 cm) for the equilibrium concentrations to be reached in a small time. As a consequence, in crystals in equilibrium with liquid helium, the ^3He concentration is extremely small: starting with 300 ppb (respectively 0.4 ppb), X_3^h should be below 10^{-15} (respectively

Fig. 3 (Color online) Frequency shift for ultrapure crystals prepared with 0.4 ppb ^3He content except for Day's polycrystal (1 ppb) and for single crystals Xa to Xf (300 ppb)



4×10^{-19}) as soon as a 1 mm^3 liquid drop is in contact with our 1 cm^3 single crystals, even less if the liquid volume is larger. This is probably why our single crystals show the same stiffening temperature for 300 ppb as for 0.4 ppb He gas: the final crystals are free of impurities. Our results open the way to studies with no impurities at all (one can reach 10^{-21} if the liquid volume is about the same as the solid volume in contact with it). Note that, even if ^3He atoms bind to dislocations with an energy in the range 0.3 to 0.7 K [23, 24], the dislocations should also be free of impurities below 50 mK as soon as some liquid ^4He is in contact with the solid.

We then succeeded in recrystallizing the single crystals by annealing them at 1.45 K with the fill line open. This method allowed us to study “recrystallized” samples at the melting pressure but with no apparent liquid region in equilibrium with them. Figure 3 shows that such recrystallized samples stiffen at a temperature similar to that of polycrystals with the same purity (0.4 ppb). Since crystals show a large stiffening even in the absence of impurities, the pinning of dislocations by impurities cannot be the only mechanism in all crystals.

Unfortunately, we consider the above results as preliminary because our thermometer apparently disconnects from the cell temperature below 40 mK. This is probably because either light or RF radiation are not sufficiently well filtered and warm up the thermometer (a Speer carbon resistance) when its thermal contact with the cell becomes too weak (the Kapitza resistance diverges as T tends to zero). If this is true, our observation of a stiffening below 40 mK is qualitatively correct but the transition may take place in a broader temperature domain, between 40 and 10 mK (the base temperature of our refrigerator).

In conclusion, what could happen in the absence of impurities? A first possibility is that there is a binding energy for dislocations among themselves: the dislocation network would be connected or the nodes of their network would stop moving below a certain temperature only. However, it is usually assumed that dislocations form a

connected network with fixed nodes [25–27]. It could be that there is a roughening transition temperature of order 40 mK below which dislocations stop moving [28]. A third possibility is that stiffening is due to vortex proliferation at the supersolid transition. The latter hypothesis assumes that supersolidity is an intrinsic property of ideal ^4He crystals which is only enhanced and displaced to higher temperature by the presence of disorder. This would agree with the predictions by P.W. Anderson [29] but contradict many others [30–32]. For further progress, it looks interesting to run experiments in ideal crystals containing no impurities and no structural defects, something which now appears possible [20]. One would see if ideal crystals show a stiffening at low temperature and a rotation anomaly at the same temperature. Note that, in Lin's experiment [15], a small peak in the specific heat was measured with crystals in equilibrium with the liquid. They were single crystals probably not free of dislocations but free of impurities. One also has to repeat the dc-flow experiments by Sasaki *et al.* [33, 34] which were done down to 50 mK only in crystals in equilibrium with liquid helium while the present results show that there is a transition below 40 mK for impurity free crystals.

Acknowledgements We are grateful to F. Caupin, E. Rolley, C. Guthmann, Yu. Mukharsky, E. Varoquaux, A. Braslau, J. Bossy, J. Dupont-Roc and P. Jacquier for helpful discussions.

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