Anomalous Softening of ^{4}He Crystals

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We have measured the elasticity of high quality ultrapure ^{4}He single crystals in the low temperature region where supersolidity is supposed to occur. At 20 mK, our results are consistent with elastic coefficients previously measured at 1.2 K. As the temperature increases from 20 to 100 mK, a large softening occurs because dislocations unpin from ^{3}He impurities. In the absence of ^{3}He impurities, dislocations are free to move down to 20 mK; the crystals are soft. The large magnitude of this anomalous softening shows that dislocations form a mobile mosaic structure. It illustrates the remarkable quantum plasticity of ^{4}He crystals.

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Soon after the discovery of supersolid behavior in solid ^{4}He [1,2] by Kim and Chan, Day and Beamish have shown that the transition from the normal to a possibly supersolid state is accompanied by an increase in the shear modulus [3]. They interpreted this "stiffening" as due to the binding of mobile dislocations to ^{3}He impurities, as first proposed by Iwasa [4] and Paalanen [5]. Day and Beamish studied samples grown at constant volume which should be polycrystalline [6]. Here, we present measurements on oriented single crystals with a very low ^{3}He concentration and very low dislocation density. Our three main conclusions are: (1) The rigidity of solid ^{4}He is not anomalously *large* at low temperature but anomalously *small* in the intermediate temperature range between very low temperatures ($T < 20$ mK) where dislocations are probably pinned by ^{3}He impurities and high temperatures ($T > 1$ K) where their motion must be damped by thermal fluctuations [7]. This softening is consistent with a change in mobility of the dislocations, not with a proliferation of vortices near a supersolid transition [8]. For some reason which needs to be investigated, pinned dislocations may allow supersolid behavior while mobile dislocations may not [9]. (2) This anomalous softening is even larger in high quality single crystals than in polycrystals. If one attributes it to a change in the elastic coefficient c_{44} only, c_{44} should nearly vanish above the transition because dislocations form a mosaic structure and are able to move very easily in the basal plane [10–12]. (3) High quality crystals grown at 25 mK should be free of any impurity [13]. They are soft. If there is a roughening transition of dislocations in the absence of ^{3}He [14], it has to occur below 20 mK.

Our cell is a rectangular hole 1.213×1.766 cm^2 in a 1.1 cm thick copper plate attached to the mixing chamber of a dilution refrigerator with optical access [see Fig. 1]. All crystals were grown at constant temperature T and pressure $P_m(T)$ from liquid ^{4}He containing 0.4 ppb ^{3}He.

Crystals could be melted down to a 1 mm seed and regrown either slowly (<1 μm/s) at 25 mK to obtain high quality crystals or above 1.2 K to make low quality single crystals. Their orientation is determined from growth shapes [Fig. 4].

Inside a copper plate at the bottom of the cell, piezoelectric transducers are used to excite and detect acoustic waves. Figure 2 shows a recording of acoustic resonances in the helium sample. They shift to low frequency as T increases. For most recordings, we used a low excitation
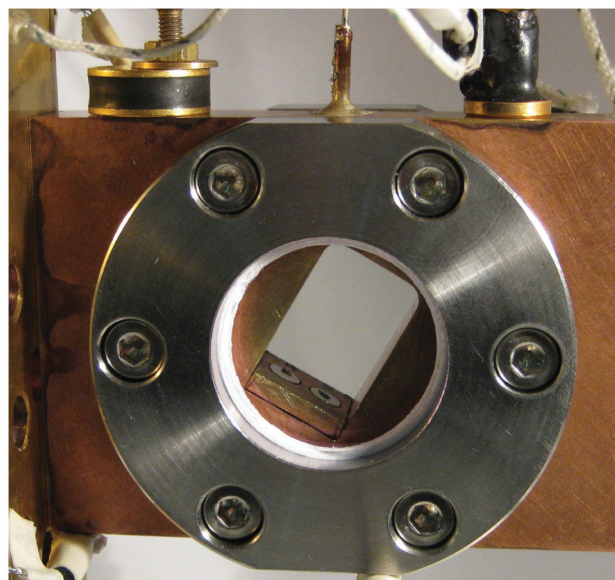

FIG. 1 (color). The helium cell is a rectangular hole in a 1.1 cm-thick copper plate. It is closed by two sapphire windows, Indium rings, and stainless steel clamps. It is tilted so that helium enters via the top left corner through a 0.1 mm capillary. At the bottom, two piezoelectric transducers are glued with stycast epoxy inside a thin cooper plate.
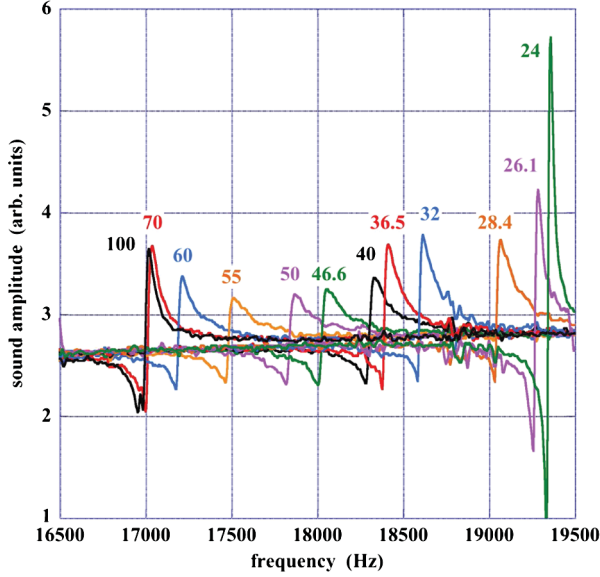
FIG. 2 (color). Sound amplitude as a function of frequency for crystal $X5c$. Resonances are labeled by their temperature in mK.
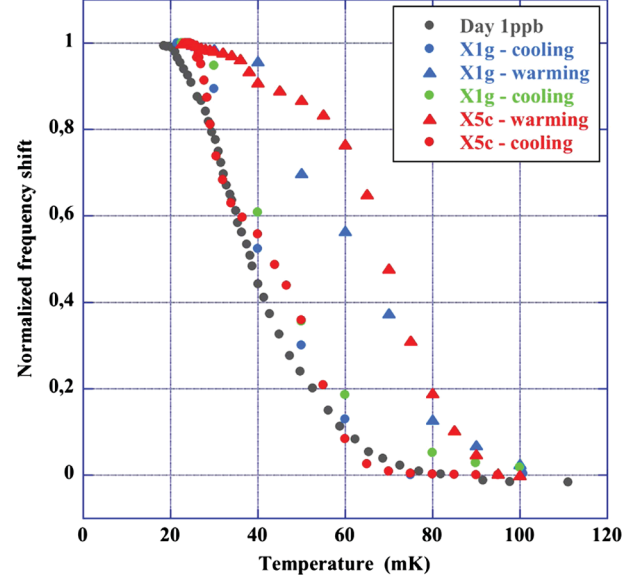

FIG. 3 (color). Resonance frequency shift $\{f(T) - f(120\text{ mK})\}/[f(20\text{ mK}) - f(120\text{ mK})]$ for the low quality crystals $X1g$, $X1h$, and $X5c$. Recordings during cooling agree with shear modulus measurements of a 1 ppb polycrystal by Day *et al.* [3].

voltage $V = 4$ mV rms. We estimate the maximum corresponding strain in the cell $\epsilon_{\max} = d_{33}V\sqrt{2}(S_{\text{piezo}}/S_{\text{cell}})Q\pi/L$ as 1.1×10^{-8} since the surface ratio $S_{\text{piezo}}/S_{\text{cell}} = 0.096$, $d_{33} = 0.8$ Å/Volt (1/5 of its room temperature value [15]), the maximum quality factor $Q = 1000$ and the cell width $L = 1.213$ cm. This is below the threshold for nonlinear elastic behavior that we found at 2×10^{-8} for low quality single crystals [16], in agreement with the value 3×10^{-8} found by Day and Beamish in polycrystals [3].

We first prepared a low quality single crystal by growing it at $T = 1.45$ K. During cooldown, the cell remains at $P_m(T)$ till various thin spaces in the cell crystallize. Since P_m decreases by about 1 bar [17], stresses induce a large disorder [18]. Figure 3 shows that during cooldown the disordered single crystal $X1g$ has the same temperature dependence as a polycrystal grown by Day and Beamish using ^{4}He with a similar purity (less than 1 ppb). We conclude that the change in elasticity cannot be due to the properties of grain boundaries which are not liquid [6]. It must be the elasticity of each grain which changes. During warming, the stiffness shows a definite hysteresis which depends on warming rate, a behavior which needs to be further studied because it was not observed in polycrystals [3].

After this, we prepared high quality single crystals by growing them slowly (<1 μm/s) at 25 mK. Under these growth conditions, Ruutu *et al.* [19] have shown that the dislocation density is between 0 and 100 cm^{-2}. By growing crystals slowly from the superfluid liquid around 1 K, Syshchenko and Beamish found 7.5×10^3 dislocations per cm^2 [20]. We assume that our best crystals have a small density of dislocations of order 100 cm^{-2}. As concerns

their purity, Pantalei *et al.* [13] have shown that the equilibrium ratio X_3^h/X_3^L of the respective ^{3}He concentrations in the hcp crystal h and in the liquid L is less than 10^{-21} at 25 mK. Since we use ultrapure ^{4}He to start with, all ^{3}He impurities should be rejected in the liquid during growth at 25 mK and they should remain there as long as the cell is not warmed up. In the presence of dislocations, there should be bound states on dislocations where the energy is 0.73 ± 0.45 K below that in the bulk solid [21] but definitely above that in the liquid. As a consequence, the concentration X_3^d on dislocations has to be smaller than $X_3^L = 4 \times 10^{-10}$, so that the distance $L_i = a/X_3^d$ between ^{3}He impurities is larger than 1 m, hundred times the cell size (the interatomic distance a is about 0.3 nm). In other words, even in the presence of dislocations, crystals fresh from growth at 25 mK must be nearly free of any impurity (at most 1% of the dislocations might have one ^{3}He bound to them).

Figure 4 shows that such a crystal—$X5a$—is anomalously soft. Indeed, its fundamental resonance is at 17 650 Hz while, according to the elastic coefficients c_{ij} which had been measured at 1.2 K [22,23], it should be 19 200 $\pm$ 200 Hz. This value was calculated numerically since the wave is nowhere purely longitudinal nor transverse so that the resonance frequency depends on all coefficients c_{ij} and on the crystal orientation. Aleinikava *et al.* [14] had predicted that, due to the influence of the lattice potential and even in the absence of impurities, the dislocations should present a crossover from a rough high temperature state to a smooth low temperature state at a temperature of order the kink energy. According to our
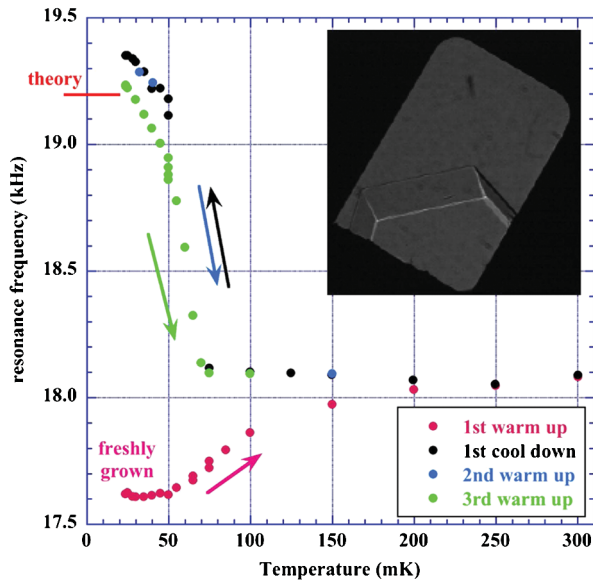
FIG. 4 (color). Resonance frequency vs temperature for crystal *X5a*. The photograph shows its orientation.

observations, this crossover occurs only below 20 mK if it exists, or perhaps the smooth dislocation lines experience a dynamic roughening at extremely low stress amplitude.

During the first warm-up after growth, *X5a* stiffens (its resonance frequency increases). This behavior is found for all freshly grown crystals. It is the opposite of the softening measured usually. When the crystal is cooled down afterwards, its stiffness increases again, and reaches 19 400 Hz. Given the 1% uncertainty in the elastic coefficients of solid ^{4}He [22], the agreement with the calculated value (19 200 ± 200 Hz) is good. We understand this behavior as follows. *X5a* is always in contact with a liquid where all ^{3}He impurities have been trapped during growth. Since the equilibrium concentration ratio X_3^h/X_3^L reaches 10^{-3} at 120 mK (even more at higher *T*), some ^{3}He atoms must leave the liquid, enter the solid, and bind to some of the dislocations during warming up to 300 mK. As a result, the resonance frequency increases up to 18 100 Hz, a value which is still less than the calculated value. Since Syshchenko [20] showed that ^{3}He atoms have a wide distribution of binding energies to dislocations, this is perhaps because ^{3}He occupy deep potential wells only, possibly at the nodes of the dislocation network. When cooling down again, more ^{3}He atoms bind to dislocations and kill their mobility so that the crystal recovers its normal stiffness. According to Schratter *et al.* [24], ^{3}He atoms are ballistic quasiparticles moving by coherent tunneling. They can be bound either to dislocations or in the liquid, apparently more easily to dislocations because impure liquid regions are small. Since ^{3}He atoms do not seem to move along dislocations, they need to escape in the bulk crystal and travel without meeting other dislocations in order to reach the liquid. The escape probability is small at low temperature. Careful studies on very long time

scales show that the stiff state at 20 mK is not absolutely stable. It relaxes to the soft state with a very long relaxation time of order 100 hours. This relaxation is apparently shorter in the presence of mechanical vibrations as we shall see below.

During further temperature cycles, the resonance frequency varies from 19 300 ± 100 Hz at 20 mK to 18 100 Hz at 300 mK. The latter value being intermediate between 17 650 and 19 400, we believe that the ^{3}He concentration in the solid is roughly stable, given by a thermodynamic equilibrium at 300 mK. One could check this interpretation by applying temperature cycles of variable amplitude. Moreover, warming up to 300 mK may produce some work hardening because the melting pressure P_m decreases by 300 μbar [25] and the plasticity threshold [3] could be reached.

After the above study between 20 and 300 mK, we warmed *X5a* up to 0.95 K and cooled it down again, becoming *X5b*. Figure 5 shows that its resonance frequency is now stable near 15 900 Hz above 60 mK and that it may increase up to the normal value 19 200 Hz at 20 mK. We have realized that the stiffness depends on the cooling rate and on the vibration level in the cell, something which should be considered in all further studies of anomalies in helium crystals. Nearly all dilution refrigerators have a so-called "1 K pot" filled with ^{4}He and pumped down to a few mbar. This 1 K pot needs to be filled thanks to a fill line coming from the ^{4}He bath at 4.2 K. If the liquid in the 1 K pot fill line is not well thermalized before it enters, some instabilities produce mechanical vibrations there. This was our case and we could record mechanical noise with our piezoelectric detector. This
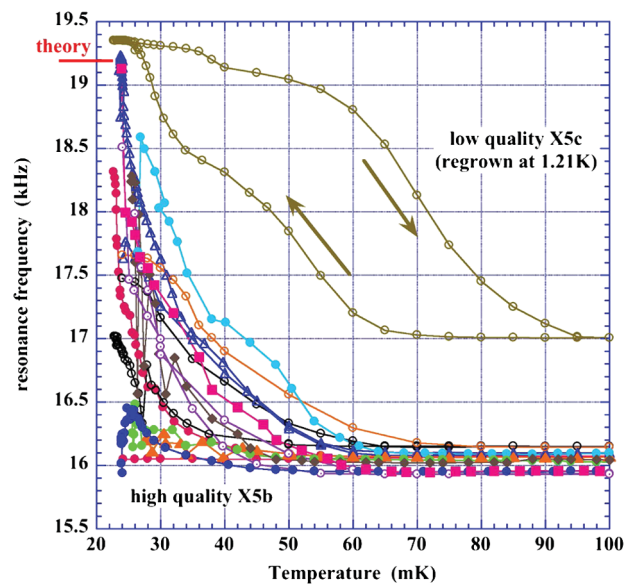

FIG. 5 (color). Resonance frequency vs temperature for crystal *X5b*. Different rates of cooling or warming, with or without noise from the 1 K pot, lead to different behaviors. The low quality crystal *X5c* does not soften as much as *X5b*.

noise looked small but it was large enough to decrease the sound velocity of high quality single crystals by 20%. It could be easily suppressed by closing a valve at the entrance of the fill line. Only high quality crystals were sensitive to this noise. The different curves on Fig. 5 correspond to different rates of cooling or warming with or without noise from the 1 K pot.

The value 15 900 Hz is very low. If attributed to a reduction in the coefficient c_{44} only, our numerical calculation shows that c_{44} nearly vanishes: with $c_{44} = 0$, the resonance frequency would be 14 500 Hz. 15 900 Hz corresponds to $c_{44} = 17$ bar, and a decrease from 19 400 Hz corresponds to a reduction of c_{44} by 86%. c_{44} determines the velocity $v_t = (c_{44}/\rho)^{1/2}$ of transverse waves traveling perpendicular to the basal plane [22,23]. It is the coefficient that should be reduced if dislocations glide in this basal plane [3]. A very large reduction in c_{44} implies that dislocations cooperate by forming the low angle grain boundaries of a mosaic structure [10–12]. If dislocations formed a network of single lines, the reduction in c_{44} should not exceed 5%. We have checked that, in order to reach 15 900 Hz, it is necessary to warm up the crystal above 0.8 K. Above this temperature, the dislocation network is apparently able to reorganize itself. Maris and Balibar [26] have shown that for a polycrystal the reduction in shear modulus should be smaller than for a single crystal. Note that for a dislocation density $\Lambda \approx 100$ cm^{-2} and a deformation $\epsilon = 10^{-8}$, we find that dislocations should move at 16 kHz macroscopic distances of order $\epsilon/a\Lambda \approx 30$ μm, that is at velocities up to 3 m/s. This is a spectacular aspect of quantum plasticity. It would be interesting to see if it is related to the possible splitting of dislocations into partials [27].

Figure 5 also shows a hysteresis cycle corresponding to the crystal $X5c$ obtained by regrowing $X5b$ at 1.21 K. Its softening is large but not as much as $X5b$. The softening occurs at higher temperature, probably because the dislocation density is larger so that a larger stress [4] or larger thermal fluctuations are needed to unbind the ^{3}He impurities. The ^{3}He concentration may also be higher.

We finally studied the resonance frequency as a function of the sound amplitude at 24 mK [16]. Starting with a high quality crystal in a stiff state, we found that applying a large sound amplitude (100 mV) irreversibly lowered the frequency as if ^{3}He atoms progressively unbound and reached their ground state in the small liquid regions [16]. A similar study with the disordered crystal $X5c$ showed only a small variation of the stiffness with amplitude, as found by Day *et al.* [28] in polycrystals. Apparently, disordered crystals with a short interdislocation distance L_N are much less sensitive to the amplitude of the applied stress.

In conclusion, our observations are consistent with a softening of ^{4}He crystals due to the unbinding of dislocations from ^{3}He impurities. High quality crystals show a transition from a normal stiff state below 20 mK to an anomalous soft state at higher temperature that is very sensitive to traces of ^{3}He impurities and to very small stresses. As for the relation between the softening and the possible transition to a supersolid state, it has been suggested that the core of dislocations is supersolid only if they are pinned [9]. This suggestion should be checked by studying the rotational inertia of crystals similar to those analyzed in the present Letter.

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