Migration of ^{3}He impurities along dislocation lines in ^{4}He single crystals

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Abstract. We have studied acoustic resonances in ^{4}He single crystals in the temperature region $20 < T < 300$ mK where a large softening occurs due to the unbinding of dislocations from ^{3}He impurities. Here we present an analysis of the dependence of resonance frequencies on the sound amplitude, that is on the acoustic stress in the crystal. Above a threshold of order 10^{-6} bar, this stress is able to unbind dislocations from impurities. Our results indicate that ^{3}He impurities diffuse along dislocations at 60 mK but not at 23 mK.

It is now well accepted that the softening of solid ^{4}He in the temperature domain from 50 to 200 mK is due to the unbinding of ^{3}He impurities from dislocations [1 - 4]. As for the relation between this 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 [5]. In order to better understand the role of dislocations, we have studied the softening in ultrapure oriented ^{4}He single crystals as a function of stress. We have observed a large softening as a function of the stress applied in agreement with the unbinding model. In addition, this temperature dependent effect suggests that atoms diffuse along dislocation lines at 60 mK and not at 23 mK as expected if their motion is thermally activated above energy barriers. This behavior looks different from their motion through the lattice where it is known to proceed by coherent quantum tunneling [6].

The acoustic 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. The two sides of the copper plate are closed by two sapphire windows. The rectangular hole is tilted so that helium enters via the top left corner through a 0.1 mm capillary. At the bottom of the acoustic cavity, two piezoelectric transducers (PZT 5500, thickness 1.0 mm, diameter 4.0 mm) are used to excite and detect acoustic waves. A sinusoidal voltage (V = 2 to 100 mV rms) is produced by a synthesized function generator. It is applied to the driving transducer and produces a longitudinal displacement δz at its front face ($\delta z = d_{33}$V $\simeq 1.6 \times 10^{-3}$ to 0.08 $\mathring{A}$) which leads to a strain and a stress field in the whole crystal. The stress applied on the second transducer generates an electric charge on the face of the stressed electrode. This charge is amplified as a current by an ultra-low noise current preamplifier and measured with a digital lock-in amplifier. The frequency of the fundamental mode in the cavity depends on all 5 elastic coefficients of the hcp single crystal (c_{11}, c_{12}, c_{13}, c_{44}, c_{66}). By looking at the stress field of the fundamental mode in numerical simulations, we have observed that the maximum stress is on the walls of the cavity (see Fig 2). Thus the maximum stress at resonance is proportional to the current measured by

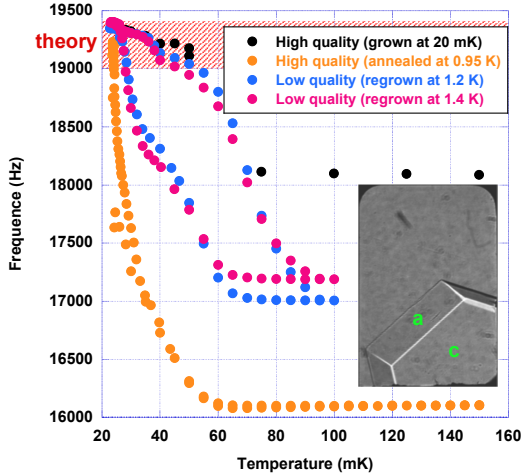the detection transducer ($\sigma_{det} \simeq 0.02$ to 2 Pa).

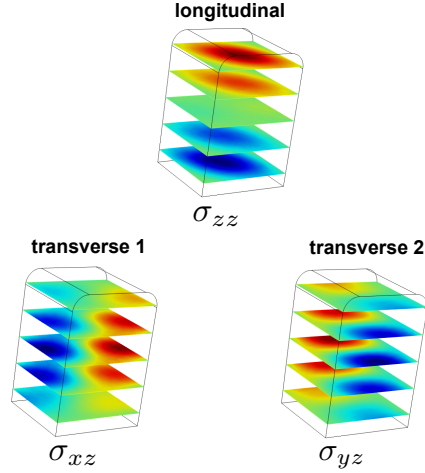**Figure 1.** Softening for ^{4}He crystals having different qualities but the same orientation

Figure 2. Components σ_{zz}, σ_{xz}, σ_{yz} of the stress tensor at resonance by Finite Element Method. High stress regions are in blue and red.

We have studied the softening for crystals with different qualities but the same orientation (Fig 1). Details of the growth method are described in [4]. The c-axis orientation of the crystal (θ=78°, ϕ=80°) is calculated from photographs taken during rapid growth where facets are visible (see inbox Fig 1). By using the orientation of the crystal and the elastic constants measured by Greywall [7] above 1K, we have done two numerical simulations (Finite Difference Method and Finite Element Method) in order to find the fundamental mode of the cavity. They give approximately the same resonant frequency for the fundamental mode (19200±200 Hz). It agrees with all samples at low temperature (Fig 1). This is consistent with the idea that above 1K and below 20 mK, dislocations are not free to move. Above 1K, the dislocations motion is damped by thermal phonons whereas below 20 mK, dislocations are pinned by ^{3}He impurities. We assume that the softening is due to the gliding of dislocations in the basal plane as usual in hcp structures. Since c_{44} is the only coefficient related to shear stress in the basal plane, we attribute all the softening to the c_{44} decrease in temperature. The string model of Granato-Lücke applies to ^{4}He crystals [8] explains the softening effect by the unbinding of ^{3}He impurities from dislocation as the following. Thermal fluctuations exert an effective stress on dislocations, the resulting force applied on bound ^{3}He, $F = (8/\pi)b\sigma L$, is proportional to the stress σ, to the free length L of the dislocation, and to the Burgers vector b. $L = L_i L_n/(L_i + L_n)$ depends on the distance L_i between two ^{3}He impurities and on the distance L_n between two nodes in the dislocation network. When F becomes larger than the binding force, impurities unbind from dislocations which can glide freely and decrease the effective c_{44}.

This model qualitatively explains the observed hysteresis between warming and cooling in low quality ^{4}He single crystals (see Fig 1). At high temperature, as dislocations are not pinned by ^{3}He impurities, they are free to move and the free length L is large. Since the thermal fluctuations are applied on large lengths, the resulting force applied on a possible bound ^{3}He is large as well. On cooling, one needs to reach a lower temperature to start binding ^{3}He atoms than for the reverse on warming. During further cooling, more ^{3}He bind to dislocations and c_{44} increases. At low temperature, the crystal is stiff because dislocations are full of impurities. When warming the crystal from low temperature, the free dislocation length is small, the thermal

fluctuations are applied on short lengths and the resulting force on ^{3}He impurities is also small so that one needs to reach a higher temperature to start unbinding dislocations. Once ^{3}He impurities unbind from dislocations they do not bind again because thermal fluctuations are applied on larger lengths. c_{44} decreases. The observed hysteresis is also enhanced by a long relaxation time for the unbinding effect compared to the time for measurements which are made in typically 7 hours for a full temperature cycle).

In another series of measurements, we followed the upper curve of Fig.1 by warming the low quality single crystal (regrown at 1.2 K) from 20 mK to 3 different temperatures 23 mK, 60 mK and 200 mK. At each temperature we measured the resonant frequency as a function of the drive voltage. Fig 3 shows c_{44} - which we obtained from the resonant frequency and the numerical simulation - as a function of the maximum stress at resonance calculated as described above. Because of the existence of a relaxation time for the unbinding effect, we did all measurements in the same way. We did cycles in amplitude, increasing successively the drive voltage from 2 mV to 100 mV then decreasing it. We have spent the same amount of time for each data point. Fig.3 shows a plot of c_{44} vs σ. Numbers 1 to 4 correspond to four successive situations at 60 mK (see Fig 3). **(1)** The stress field is inhomogeneous in the cell. As we increase the stress, it unbinds dislocations where it is high enough. Part of the dislocations are free to move and the mean c_{44} decreases. **(2)** During cooling, no ^{3}He binds back to the dislocations at this temperature because the free length is still large. c_{44} does not change. **(3)** At sufficiently low stress, ^{3}He migrate along dislocations and repopulate the most strongly excited regions of the crystal. The number of ^{3}He atoms is constant in the dislocation network, c_{44} remains nearly constant. **(4)** Because of the migration, it is possible to unbind some more ^{3}He in the highly stressed regions. c_{44} decreases as a function of stress with a steep slope.
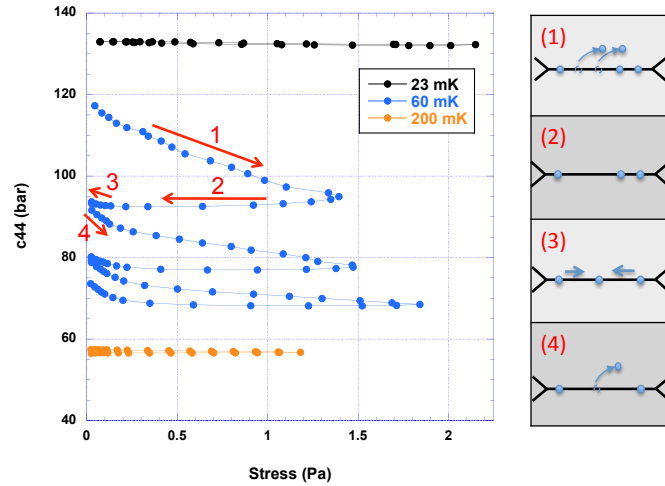

Figure 3. Stress dependence of c_{44} in the low quality crystal regrown at 1.2 K

Iwasa and Susuki[8] calculated the binding force of a ^{3}He atom to a dislocation and found $F_m \simeq 10^{-14}$N. The critical stress one needs to apply in order to unbind a ^{3}He impurity from a dislocation is $\sigma_c = \frac{\pi}{8} \frac{F_m}{b \, L_i}$. It is inversely proportional to the free length L which is very small at low temperature, of order the interatomic distance a. As a consequence, the critical stress for unbinding is very large at low temperature. This explains why Fig 3 does not show any stress dependence in c_{44} at 23 mK. We do not observe any stress dependence in c_{44} at 200 mK neither because all ^{3}He atoms have unbound from dislocations at this temperature. More interesting is the progressive decrease in c_{44} after each stress cycle at 60 mK. We did 3 cycles in amplitude at

this temperature and found that c_{44} decreases more after the first cycle than after the following ones, meaning that the system tends towards a stable state which has to be close to the lower curve on Fig.1 that had been obtained during warming. The most important remark on Fig 3 is the change in slope between situations (3) and (4). We understand it as an indication that ^{3}He impurities are able to migrate along dilsocation lines at 60 mK.

In order to check this interpretation we have repeated the stress cycles at lower temperature. Because it is not possible to unbind dislocations at 23 mK in low quality single crystals where L_n is too small, we have used high quality ones with the same orientation.
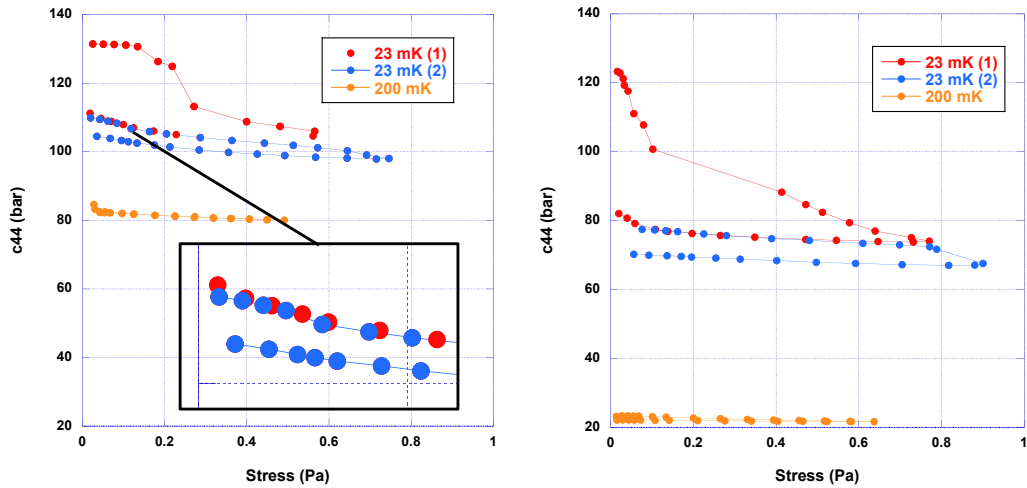

Figure 4. Stress dependence of c_{44} in high quality crystals, one grown at 25 mK (left) and one regrown at 0.95 K (right)

These high quality crystals have shown a much larger sensitivity to the applied stress σ at low temperature than low quality ones (see Fig 4). This is probably because the dislocation network is denser in low quality crystals, so that the critical stress at low temperature is larger. After two cycles in amplitude at 23 mK, we have observed that the stress is sufficient to unbind ^{3}He from dislocations. However, in both cases, there is no change in slope between the end of the first cycle and the beginning of the second one. Apparently, ^{3}He atoms do not migrate along dislocations at 23 mK.

In conclusion our results are consistent with a migration of ^{3}He atoms along dislocations at 60 mK not at 23 mK, as expected if their motion is thermally activated above energy barriers. The origin for these energy barriers might be related to the existence of a distribution in binding energies[3]. The resulting diffusion of ^{3}He atoms along dislocation lines appears different from the coherent tunneling in the bulk[6].

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