Anomalous nonlinear attenuation of ultrasound in solid $^4\text{He}$ in a torsional oscillator below 200 mK

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Abstract. In order to elucidate the ultra-low temperature behavior of solid $^4\text{He}$, simultaneous measurements of longitudinal ultrasound (US) and torsional oscillation have been made. Changes in attenuation and velocity of US at 10 MHz have been measured in polycrystalline hcp $^4\text{He}$ samples (0.3 or 20 ppm of $^3\text{He}$ impurity) grown in a 1 kHz torsional oscillator (TO). In a 0.3 ppm $^3\text{He}$ sample, the US attenuation and velocity were found to depend on the US drive voltage at temperatures below 70 mK where the anomalies in the TO frequency and dissipation were also observed. The US attenuation at low T (10 mK) decreased monotonically as the drive voltage was decreased but then remained small and constant as the drive voltage was increased again. The US velocity change at low T was negative with respect to the high-T (400 mK) value, contrary to the positive sign expected from the known variation in the shear modulus. In a 20 ppm $^3\text{He}$ sample, both the US and TO anomalies shifted to 150 mK. The amplitude dependence and hysteresis of US attenuation were related to pinning of dislocations by $^3\text{He}$ impurities, and nonlinear spatial variations of the amplitude of US pulses were derived.

1. Introduction

Ultrasound (US) measurements in MHz-order frequency are suitable for studying mechanical properties of materials. The very first US measurements on solid helium [1] were probably made on polycrystalline samples to determine the phase diagram. Later US measurements on solid $^4\text{He}$ were mainly performed on single-crystalline samples to study anisotropic elasticity [2, 3] and dislocations [4, 5]. The effects of $^3\text{He}$ impurities in solid $^4\text{He}$ have been studied and explained by pinning of dislocations by $^3\text{He}$ impurity atoms [6]. Later a new anomaly has been observed below 200 mK in $^4\text{He}$ crystals containing $^3\text{He}$ impurities [7].

After the discovery of anomaly of the torsional oscillator (TO) containing solid $^4\text{He}$ [8, 9], the shear modulus of solid $^4\text{He}$ below kHz-order frequency has been extensively studied on polycrystalline and single-crystalline samples and with various $^3\text{He}$ concentrations including ultra-pure samples [10, 11]. Now the variation of the shear modulus is well understood as caused by dislocations and their interaction with $^3\text{He}$ impurities [12, 13]. Many of the TO anomalies can be accounted for by the shear modulus change of solid helium in the torsion rod [14].

We have made simultaneous measurements of ultrasound and torsional oscillation on polycrystalline $^4\text{He}$ with different $^3\text{He}$ concentrations [15]. Our motivation is to clarify the origin of the TO anomaly, to extend our knowledge on dislocations in solid $^4\text{He}$ and to shed light on...
the US anomaly [7]. To our knowledge, this is the first US measurement on polycrystalline $^4$He at temperatures below 1 K.

2. Experimental apparatus and procedure

Our TO incorporating 10 MHz quartz transducers has been described in detail previously [15]. Briefly, the BeCu torsion rod of 3.6 mm diameter and 9.0 mm length is attached to a bob containing sample space. A hole of 0.8 mm diameter through the torsion rod serves to introduce helium gas into the sample space. Two fins for torsional excitation and detection are mounted on the outer wall of the bob. Two 10-MHz X-cut quartz transducers of 10 mm diameter and a cylindrical spacer (10.2 mm outer diameter, 7.9 mm inner diameter, $H = 5.3$ mm height) between them are inserted into the sample space and held by springs for US measurements. Polycrystalline $^4$He is grown in the sample space by the blocked capillary method over a time interval of about two hours. After the measurements with the nominal purity $^4$He ($^3$He concentration $x_3=0.3$ ppm) have been completed, helium gas is driven out of the sample space and reloaded with $^4$He with $x_3=20$ ppm.

The resonance frequency of the empty TO at low temperature is 1009 Hz. The TO frequency is decreased by about 180 mHz when the sample space is filled with solid helium. For US measurements, 9.58 MHz RF pulses with 2 $\mu$s width and 1 kHz repetition frequency are applied to the transmitter transducer. The amplitude of the pulses can be varied by an attenuator. The propagated US signal through the sample is detected by the receiver transducer and measured by a super-heterodyne US measurement system. Changes in the US attenuation $\alpha$ and velocity $\delta v/v$ are calculated from the in-phase and out-of-phase outputs of the super-heterodyne system. The longitudinal velocity in our solid $^4$He samples is assumed to be $v = 500$ m/s.

3. Results

Results of simultaneous measurements on two samples with $x_3=0.3$ ppm and 20 ppm are shown in Fig. 1, where the data are taken as the temperature (T) is decreased down to about 10 mK.
3.1. TO anomaly
Changes in TO resonant frequency, $\delta f$, and changes in TO dissipation, $Q^{-1}$, are shown in panel (a) and (b) of Fig. 1, respectively. Effects of temperature on "backgrounds" of frequency shift and dissipation in the empty cell have been subtracted. The TO anomaly is characterized by decreases in $\delta f$ with increasing $T$ where $T < 400$ mK and peaks in $Q^{-1}$. The characteristic temperature of TO anomaly, $T_{TO}$, is the temperature of dissipation peak; $T_{TO}=70$ mK for $x_3=0.3$ ppm, and 140 mK for 20 ppm. The total anomalous shift in frequency, $\delta f(10 \text{ mK}) - \delta f(400 \text{ mK})$, is between 2 and 3 mHz.

3.2. US anomaly
Fractional changes in US velocity, $\delta v/v$, and relative changes in attenuation, $\alpha$, are shown in panel (c) and (d) of Fig. 1, respectively. US anomalies are observed around 800 mK and below 200 mK. We focus our attention to the low-T US anomaly below 200 mK. The US anomaly is characterized by decrease in $\delta v/v$ with decreasing temperature and the peak in $\alpha$ for $x_3=0.3$ ppm sample and by increase in $\delta v/v$ and decrease in $\alpha$ with decreasing temperature for $x_3=20$ ppm sample. The characteristic temperature of US anomaly, $T_{US}$, is the temperature of the attenuation peak and the temperature at which the velocity or attenuation changes on cooling. We find $T_{US}=70$ mK for $x_3=0.3$ ppm, and 150 mK for 20 ppm, which are close to $T_{TO}$ at each $^3$He concentration.

3.3. Amplitude dependent US attenuation
We have extensively studied the behavior of the sample with $x_3=0.3$ ppm. The attenuation strongly depends on the normalized US drive voltage, $D$, at $T < T_{US}$ as shown in Fig. 2. Most of the data in Fig. 2 are taken on cooling runs at different setting of $D$ except for the data $D=2.82w$ which is a warming run. $D=1$ corresponds to an excitation voltage of 89 mV peak-to-peak applied to the transmitter. The initial amplitude of US pulses excited by the transmitter should be proportional to $D$. Larger attenuation is observed for cooling runs at higher $D$.

Fig. 2 shows sharp attenuation peaks at 70 mK (low T anomaly) and 800 mK (high T anomaly). The shape of the peaks, however, is sensitive to the calibration of our US measurement.
system and the accurate height of the peaks cannot yet be determined.

Fig. 3 shows the attenuation as \(D\) is varied at about 10 mK. Squares are the data at the end of the cooling runs at various \(D\) shown in Fig. 2. It is clear that the attenuation decreases as \(D\) is decreased. When \(D\) is changed at 10 mK, hysteretic behavior is observed. In one case (diamonds), \(D\) is first decreased from 1.4 to 0.63 and then increased to 1.0. The attenuation decreases in the initial decrease in \(D\) but does not change in the increase in \(D\). In the other case (triangles), the attenuation remains at the same value on increasing \(D\) from 0.1 to 2.82. In this way, the behavior is nonlinear for decreasing \(D\) and linear for increasing \(D\).

3.4. Amplitude dependence of US velocity
The US velocity also depends on \(D\) at \(T < T_{US}\) as shown in Fig. 4. The velocity at 10 mK in this sample is smaller than the velocity at high \(T\) (400 mK), but in the other sample with \(x_3=20\) ppm we find \(v(10\ \text{mK}) > v(400\ \text{mK})\) as shown in Fig. 1(c).

3.5. Interaction between TO and US measurements
Simultaneous TO measurements while US drive voltage is changed show no change in TO characteristics although the US characteristics are changed. Similarly, simultaneous US measurements while TO excitation voltage is changed show no change in US characteristics. We, therefore, conclude that there is no measurable interaction between TO excitation and US propagation in the present TO cell.

4. Discussion

4.1. TO anomaly
Beamish et al.\cite{14} have proposed that hardening of solid \(^4\text{He}\) (i.e. increase in shear modulus) in the torsion rod is the origin of many of the observed TO anomaly. The increase in shear modulus of solid \(^4\text{He}\) below about 100 mK may be explained by dislocations being pinned by \(^3\text{He}\) impurity atoms and becoming immobile. As a result, the total torsional spring constant would increase and the TO frequency would increase. The shear modulus of solid \(^4\text{He}\) at low \(T\) is considered to be the intrinsic property of the lattice. The shear modulus would decrease at high \(T\) because dislocations become freed from pinning by \(^3\text{He}\) impurity atoms. The amount of the decrease depends on the dislocation density, distribution of network pinning length etc.

The total torsional spring constant is a sum of the contributions from the BeCu torsion rod and from solid \(^4\text{He}\) in the torsion rod. The torsional spring constant of our BeCu torsion rod is calculated to be \(K_{rod} = 92\ \text{Nm}\) and the torsional spring constant due to solid \(^4\text{He}\) in the rod at low \(T\) to be \(K_{He} = 6.4 \cdot 10^{-5}\ \text{Nm}\). If the shear modulus of solid \(^4\text{He}\) in the rod is reduced by 50\% from the low \(T\) value, the resulting frequency change is estimated to be 0.17 mHz. The observed frequency change is more than 10 times larger so that it cannot be fully explained by the hardening effect of solid \(^4\text{He}\) in the rod. Mechanisms other than the hardening effect of solid \(^4\text{He}\) in the rod must be present. A possibility is the mechanism proposed by Iwasa \cite{12, 17}, the origin of which is pinning of dislocation by \(^3\text{He}\) impurities in solid \(^4\text{He}\) in the TO, but the size of the effect is not determined theoretically. Another possibility is the motion of the quartz transducers and the spacer. Although the spacer is bonded to the inner wall of the bob with vacuum grease at room temperature, the grease may become brittle at low temperature and a gap is formed between the spacer and the pressurized bob. Then the quartz transducers and the spacer are loosely connected to the bob via springs at both ends and solid helium filling the gap acts as an additional spring which contributes to the TO anomaly.

4.2. Analysis of the amplitude dependent US attenuation
Past experiments on the temperature dependence of ultrasound propagation in solid \(^4\text{He}\) \cite{4, 5} have been carried out mostly on single crystal samples. Measured US velocity, \(v\), has been
analyzed by assuming the net fractional change in $v$ is given by $\delta v/v = \delta v_d/v + \delta v_p/v$, where $\delta v_p/v = p_0 + p_4 T^4$ is introduced to account for the change in $v$ due to phonon anharmonicity ($p_0$ and $p_4$ are fitting parameters). The dislocation line contribution $\delta v_d/v$ is written using the Granato-Lücke theory [16] which accounts for the frequency-dependent and amplitude-independent US attenuation and modulus change by treating the dislocation line as a string. The high-T US anomaly in our polycrystalline samples can be similarly analyzed by the Granato-Lücke theory as discussed in a previous report [15], where the effect of $^3$He impurities was not taken into account. It is, however, clear that the amplitude-dependent low-T anomaly below $T_{US}$ is not described by the Granato-Lücke model.

The observed amplitude dependence of US attenuation and velocity, the hysteresis of attenuation, and the dependence of $T_{US}$ on $x_3$ are similar to the characteristics of TO anomaly and those of shear-modulus anomaly in and below the kHz region, suggesting the origin of US anomaly is also pinning of dislocations by $^3$He atoms. None of the US measurements on single crystals of $^4$He grown from high purity He gas ($x_3=0.3$ ppm) have shown such an amplitude-dependent low-T anomaly, but there is an US measurement on a single crystal of $^4$He containing 30 ppm $^3$He which shows amplitude dependent velocity and attenuation at temperatures below 1 K [6].

Dislocations in solid $^4$He are strongly pinned by the network nodes and weakly pinned by $^3$He impurity atoms. The network pinning length, $L_N$, is distributed over a wide range but it is independent of temperature. The resonant frequency of a dislocation segment of length $L_N$ is given by

$$F_0 = \frac{1}{2 L_N \sqrt{\frac{C}{A}}}$$

(1)

where $A$ and $C$ are the effective mass per unit length and the line tension of the dislocation, respectively. A dislocation segment with the critical length, $L_c$, has the resonant frequency equal to the US frequency, $F$,

$$L_c = \frac{1}{2F} \sqrt{\frac{C}{A}}$$

(2)

$L_c$ slightly depends on the molar volume of solid $^4$He and is estimated to be about 9 $\mu$m for our
samples at $F = 9.58$ MHz. US attenuation at low T is mainly due to the resonant vibration of dislocation segments whose lengths are around $L_c$. The impurity pinning length is a function of $T$ and $x_3$. The average distance between pinning $^3$He atoms along the dislocation line is the average impurity pinning length, $L_i$, given by [6]

$$L_i = 3.4 \times 10^{-9}x_3^{-2/3}e^{-0.2/T}[m].$$

$L_i$ decreases with decreasing temperature and becomes equal to $L_c$ at a critical temperature, $T_c$. We estimate $T_c=94$ mK for $x_3=0.3$ ppm. The number of dislocation segments of length $L_c$ decreases at $T < T_c$ due to impurity pinning so that the attenuation is expected to decrease with decreasing temperature. However, dislocations are not necessarily pinned if the amplitude of US pulse is sufficiently large.

The US amplitude is spatially varied in our US experiment as a US pulse is excited by one transducer and propagates in the solid helium sample. The US attenuation through $H = 5.3$ mm thick solid helium is estimated to be 25 dB from the attenuation difference of the run $D=2.82w$ in Fig. 2 between 19 dB at $T > 100$ mK and -6 dB at $T < 60$ mK. Hence the amplitude of US pulse at the receiver side of the sample is about 1/18 of the incident amplitude.

The US amplitude is also temporally varied. Although the excited US pulse width is longer than the electric pulse width of 2 $\mu$s due to ringing of the transducer, it is still much shorter than the repetition interval of 1 ms.

The amplitude dependence of $\alpha$ at low T ($\sim 10$ mK) is interpreted as follows. We assume that there is a critical US amplitude, $A_c$, above which dislocation segments are not pinned even at low T and the attenuation coefficient is given by $a = 0.54$ mm$^{-1}$. On the other hand, for $A < A_c$ dislocation segments are pinned by $^3$He impurity atoms and the attenuation vanishes. The spatial variation of US amplitude is given by

$$A(x) = A_0e^{-\alpha x}$$

for $A_0 > A_c$, where $x$ is the position along the propagation direction and $A_0$ is the initial amplitude. The US pulse is attenuated as it propagates and the amplitude becomes equal to $A_c$ at $x = x_1$. Then attenuation becomes zero so that

$$A(x) = A_c$$

for $x > x_1$. According to this model of nonlinear attenuation, $A_c$ corresponds to the amplitude of received signal, $A_r$. Although $A_r$ slightly varies from run to run possibly due to ringing of the transducer, $x_1$ can be determined from known values of $A_0$ and $A_r$ for each run and $A(x)$ can be reconstructed as shown in Fig. 5. Referring to Fig. 5, when $A_0$ is decreased at low T, the received amplitude should stay at $A_r = A_c$ so that $x_1$ becomes smaller and the net attenuation is decreased. When $A_0$ is further decreased to below $A_c$, the received amplitude should become $A_r = A_0$ because of zero attenuation. Once a dislocation segment is pinned by $^3$He atoms, $L_i$ at 10 mK is so small ($L_i = 3$ nm at 20 mK for $x_3=0.3$ ppm) that the dislocation line stays in pinned state when the amplitude is increased again. Therefore, the attenuation should stay at low value and the received amplitude should increase linearly with increasing $A_0$ as observed.

4.3. Velocity change

The amplitude dependent velocity at 10 mK in Fig. 4 can be analyzed similarly as the amplitude dependent attenuation. We assume the velocity is $v = v_0$ for $A < A_c$ where dislocations are pinned and $v = v_1$ for $A > A_c$ where dislocations are not pinned. Then the measured velocity can be expressed as

$$v = v_0 \left(1 + \frac{x_1 v_1 - v_0}{H v_0} \right).$$
We obtain
\[
\frac{v_1 - v_0}{v_0} = 5.9 \times 10^{-3}
\]  
(7)
from the data in Fig. 4. We note \(v_1\) is bigger than \(v_0\) so that the US velocity decreases with decreasing temperature in the \(x_3 = 0.3\) ppm sample. In contrast, the US velocity increases in the \(x_3 = 20\) ppm sample with decreasing temperature as shown in Fig. 1(c). We expect the longitudinal velocity would increase from the known increase in the shear modulus at low T. Therefore, we need a further study to understand the negative velocity change in the \(x_3 = 0.3\) ppm sample.

4.4. US anomaly found by Ho et al.
Ho, Bindloss and Goodkind [7] reported US anomalies in their measurements on low-density single crystals of \(^4\)He containing \(^3\)He impurities. Some of their observations, such as the sharp attenuation peak at a temperature, \(T_p\), between 100 and 200 mK for samples with \(^3\)He between 10 and 30 ppm and the velocity increase around \(T_p\), are similar to our low-T anomaly. In this respect, their anomalies may possibly be related to pinning of dislocations.

5. Conclusion
We have made simultaneous measurements of longitudinal ultrasound and torsional oscillation on polycrystalline hcp \(^4\)He samples containing 0.3 and 20 ppm \(^3\)He impurity. Both US and TO measurements showed anomalous behavior at low temperatures which can be explained by pinning of dislocations by \(^3\)He impurity atoms, but no interaction was observed between TO excitation and US propagation. The frequency shift of the TO anomaly was more than 10 times larger than the expected effect from hardening of solid helium in the torsion rod. The amplitude dependence and hysteresis of US attenuation have been analyzed by the nonlinear attenuation depending on the US amplitude which varied spatially. Relationship between our low-T US anomaly and the previously observed US anomaly by Ho et al. has been suggested.

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