Thermal History of Solid $^4\text{He}$ Under Oscillation

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We have studied the thermal history of the resonant frequency of a torsional oscillator containing solid $^4\text{He}$. We find that the magnitude of the frequency shift that occurs below $\sim 100$ mK is multi-valued in the low temperature limit, with the exact value depending on how the state is prepared. This result can be qualitatively explained in terms of the motion and pinning of quantized vortices within the sample. Several aspects of the data are also consistent with the response of dislocation lines to oscillating stress fields imposed on the solid.

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I. INTRODUCTION

Over the last several years the torsional oscillator (TO) technique has become a popular method to study solid $^4\text{He}$. The measurements, which involve monitoring the resonant frequency $f$ of a high quality factor torsion pendulum filled with $^4\text{He}$, have revealed a "transition" below an onset temperature $T_O$ of $\sim 200$ mK. The results of several control experiments indicate that the sudden increase in the frequency is due to a superfluid-like decoupling of a fraction of the $^4\text{He}$ mass. The apparent nonclassical rotational inertia fraction (NCRIF), proportional to the frequency shift $\delta f$, is independent of the maximum oscillation speed $v_0$ of the TO provided that it is smaller than a critical value. Above this critical velocity $v_C$, the magnitude of NCRIF is attenuated. The value of $v_C$ corresponds to several quanta of circulation $\kappa$, suggesting that the important excitations in the system are vortices. Anderson has proposed a vortex liquid model that qualitatively captures a number of experimental results.

In multiple solid $^4\text{He}$ samples slowly grown within an annulus of radius, $r = 5$ mm, and width, $t = 0.95$ mm, a value of $v_C = 10 \mu\text{m s}^{-1}$ was found (corresponding to $\kappa = 2\pi r v_0 \approx 3h/m$, where $h$ is Planck's constant and $m$ is the bare $^4\text{He}$ mass). NCRIF decreased almost linearly with $\ln[v_0]$ as the speed was increased further. This attenuation, according to Ref. 13, is attributable to the nonlinear susceptibility of a vortex liquid phase, which consists of an entanglement of many thermally activated vortices. The ability of the vortices to move counter to the time-dependent superflow (relative to the cell's oscillation) results in the screening of supercurrents. As the temperature is lowered the motion and number of vortices is reduced so that NCRIF becomes finite. The accompanying dissipation that peaks in the middle of the transition is due to a matching of $f$ and the optimal rate at which vortices respond to changes in the velocity field. Residual $^3\text{He}$ atoms may cling to vortices, thereby slowing vortex motion and ultimately enhancing the onset temperature.

One prediction of Anderson's model is that $T_O$, and perhaps NCRIF, for a particular sample will decrease as the measurement frequency is lowered. Aoki et al. performed measurements in a double oscillator that operated in either an antisymmetric (high $f$) or a symmetric (low $f$) mode and found that for the same $^4\text{He}$ sample, $T_O \approx 240$ mK at 1173 Hz and $T_O \approx 160$ mK at 496 Hz. However, NCRIF became independent of frequency below $\sim 35$ mK. In addition, irreversible changes in NCRIF below $\sim 45$ mK occurred upon variation of the oscillation speed between $10 \mu\text{m s}^{-1}$ and $800 \mu\text{m s}^{-1}$.

Nonsuperfluid explanations of the TO experiments have also been proposed, in which some sort of freezing process or glass transition takes place. Although the former was only discussed qualitatively, the pinning of dislocation lines within solid $^4\text{He}$ is one possible realization of such a "freezing" transition. The mechanism for dislocation pinning is likely related to interactions with isotopic impurities. It is known that dislocations or $^3\text{He}$ atoms are mobile in solid $^4\text{He}$ below $\sim 1$ K, but the strong coupling between them in the low temperature limit can immobilize them both. In a TO study of many solid $^4\text{He}$ samples we found that the dependence of $T_O$ on the $^3\text{He}$ concentration $x_3$ is quantitatively consistent with this impurity-pinning model.

Recently Day and Beamish observed a dramatic increase (between 5% and 20%) in the shear modulus $c_{44}$ of solid $^4\text{He}$ ($x_3 \approx 0.3$ ppm) below $\sim 250$ mK, with the temperature dependence of $c_{44}$ strongly resembling that of $f$ in TO experiments. The authors also reported hysteresis in $c_{44}$ upon adjusting the stress amplitude of the measurement at $T \approx 20$ mK, similar to what is seen in the increase in the shear modulus is very likely due to the stiffening of the dislocation network via the immobilization of individual lines. It was suggested by the authors of Ref. 23 that an increase in $c_{44}$ will result in a stronger coupling between the $^4\text{He}$ sample and its container and therefore mimic an enhanced mass loading of a TO, causing $f$ to decrease. However, any enhancement to the overall rigidity of the system actually leads to a higher resonant frequency. We have carried out a finite element method (FEM) calculation employing realistic parameters of the TO and solid helium in order to confirm this result. The positive correlation between $c_{44}$ and $f$, their resemblances in the temperature dependence, and the hysteresis common to both, all suggest that either the
frequency shift is a direct consequence of the enhanced modulus and can be explained without invoking supersolidity or that the microscopic mechanism responsible for NCRI also affects the elastic properties of the solid (e.g., a recent suggestion is given in Ref. 25). The results presented below appear to favor the latter.

Sec. II presents our experimental observations in a general manner by simply describing the dependencies of the observed frequency shifts and dissipation signals on temperature (see Figs. 1 and 2), oscillation speed (see Fig. 3), and growth method. Two different interpretations of the data are given in Secs. III and IV. In the former we attempt to describe the experiments in the context of a supersolid that is permeated by vortices. Since vortex lines interact with velocity fields of one of the important parameters in this model is \( v_0 \). In Sec. IV we discuss the consequences of attributing the entire frequency shift to an increase in the shear modulus of solid \(^4\text{He}\). It is assumed in this picture that the rigidity of the solid is strongly affected by the motion of dislocation lines under the applied stress \( \sigma \). In anticipation of these two interpretations, we have simultaneously plotted the data in Fig. 3 as a function of \( v_0 \) and \( \sigma \). In addition, in both Figs. 1 and 2 we have plotted the frequency shift in terms of NCRI and \( \delta c_{44}/c_{44} \).

II. EXPERIMENTAL RESULTS

A recent study of samples grown at constant temperature and pressure (CT/CP) at a single point on the solid-liquid coexistence curve found the onset of the frequency shift to be sharp and reproducible, which is consistent with the expected formation of single crystals. In contrast, wide variations in \( \delta f \) and \( T_0 \) were observed when employing the blocked capillary (BC) method, which typically yields polycrystalline samples or highly strained crystals due to the pressure drop during freezing. In this paper we examine in detail the different thermal histories of \( \delta f \) in BC and CT/CP samples that follow from changes in oscillation speed above and below \( T_0 \). We have measured the temperature dependence of \( \delta f \) in 30 samples, with the bulk of our work being carried out on “isotopically pure” \((x_3 \approx 1 \text{ ppb of } ^4\text{He})\) samples at \( 1 \mu\text{m s}^{-1} < v_0 < 100 \mu\text{m s}^{-1} \). To overlap with Ref. 7 we also studied commercially pure \((x_3 \approx 0.3 \text{ ppm})\) samples up to speeds of \( 880 \mu\text{m s}^{-1} \).

Figure 1 shows the temperature dependence of \( \delta f_{\text{He}} \) for a BC sample before and after annealing, and one CT sample. For all samples \( x_3 \approx 1 \text{ ppb} \), data was obtained with the beryllium-copper TO from Ref. 10. Freezing \( (T_F) \) and annealing \( (T_A) \) temperatures are given for each sample. All data was taken during cooling scans in which the temperature was successively lowered in controlled steps. For any one sample the measured frequency at \( T > T_0 \) is identical for all values of \( v_0 \). For easy comparison we have set \( \delta f = 0 \) at \( T = 175 \text{ mK} \) by subtracting 1071.007 Hz, 1071.011 Hz, 1071.035 Hz, and 1071.898 Hz from the data of BC-12/10/06, BC-12/10/06 post-annealing, CT-01O507, and the empty TO. Thus, the effective mass loading for each of these samples is 891 mHz, 887 mHz, and 863 mHz.

FIG. 1: Temperature dependence of \( \delta f \) for a BC sample before and after annealing, and one CT sample. For all samples \( x_3 \approx 1 \text{ ppb} \). Data was obtained with the beryllium-copper TO from Ref. 10. Freezing \( (T_F) \) and annealing \( (T_A) \) temperatures are given for each sample. All data was taken during cooling scans in which the temperature was successively lowered in controlled steps. For any one sample the measured frequency at \( T > T_0 \) is identical for all values of \( v_0 \). For easy comparison we have set \( \delta f = 0 \) at \( T = 175 \text{ mK} \) by subtracting 1071.007 Hz, 1071.011 Hz, 1071.035 Hz, and 1071.898 Hz from the data of BC-12/10/06, BC-12/10/06 post-annealing, CT-01O507, and the empty TO. Thus, the effective mass loading for each of these samples is 891 mHz, 887 mHz, and 863 mHz.
we find \( \mu_1 \approx \frac{v_0}{T} \) and \( \delta Q_{H_e}^{-1} \). These results are plotted in Fig. 2. There is considerable dissipation in rapidly grown BC samples even at the lowest temperatures. After annealing sample BC-12/10/06 the high temperature tail of \( \delta f_{H_e} \) (and thus \( T_O \)), the low temperature limiting value of \( \delta f_{H_e} \), and the magnitude of \( \delta Q_{H_e}^{-1} \) are all reduced. Thus it appears that annealing affects the temperature dependence of \( \delta f_{H_e} \) much in the same way as driving the TO at high speed (see Fig. 2). One difference, however, is that the results from annealing are permanent.

In Fig. 3 we have plotted the low temperature value of \( \delta f_{H_e} \) measured at different \( v_0 \), which again demonstrates that the data converge at high speeds. One interesting difference from previous TO studies is the lack of saturation in \( \delta f_{H_e} \) even below 10 \( \mu m/s^{-1} \). This is most evident in two BC samples (01/25/07 and 12/10/06) (see Secs. III and IV below.

Using the following protocol, the thermal history of \( \delta f_{H_e} \) was investigated in crystals grown under different conditions. First, the temperature dependence of \( \delta f_{H_e} \) was measured while cooling at low speed (\( v_0 <3 \mu m/s^{-1} \)). At \( T \approx 20 \) mK \( v_0 \) was slowly increased. Final velocities were usually \( \sim 20 \mu m/s^{-1} \), but in some cases much greater (e.g., several hundred microns per second). Multiple thermal cycles were then performed in succession, indicated by the arrows in Fig. 3. Complete equilibration was purposely avoided while cycling between 30 mK and 60 mK in order to observe multiple metastable states. The measurement culminated with a high velocity cooling trace that began well above \( T_O \).

FIG. 3: Dependence of the low temperature value of \( \delta f_{H_e} \) on oscillation speed and applied stress. The inertial stress is approximated by \( \sigma \approx \pi \rho \omega v_0/2 \), for a given helium density \( \rho \). Values of \( \delta f_{H_e} \) were extracted from cooling scans that began well above \( T_O \). Samples are labeled according to growth method and date. The data are plotted versus a linear abscissa in the inset.

### III. THE VORTEX LIQUID MODEL

As stated in Sec. I, the TO experiments to date are qualitatively consistent with many features of Anderson's vortex liquid model. The present work, however, demonstrates that below 60 mK the system does not act like a free vortex liquid as originally proposed. In fact, below 30 mK it exhibits severe pinning. With this picture in mind we can explain the temperature dependence of NCRIF (\( \propto \delta f_{H_e} \)) in Fig. 3, the velocity dependence in Fig. 5, and the history dependence in Figs. 4 and 5.

First we consider the temperature dependence. As the system is cooled through the transition some number of free vortices are produced, which is proportional to \( v_0 \). When \( T << T_O \) the mobility of the existing vortices diminishes and it is unfavorable to create additional vortex lines. The inability of vortices to screen supercurrents leads to a sizeable NCRIF in the low temperature limit. When the system is driven at high speeds, not only is NCRIF smaller but the high temperature tail of NCRIF is reduced. This suggests that vortex pinning is very weak at \( T \geq 60 \) mK. It is also likely that vortices are more easily excited as \( T_O \) is approached.

The velocity dependence in Fig. 3 reflects the distri-
The measured value $= 0.08\%$ for CP-11/21/06 (see Fig. 5), resulting in a much larger NCRIF = 0.9\% for BC-12/10/06 (see Fig. 4) and NCRIF is reduced by a metastable state. For instance, the difference is $\sim 50\%$ at low speed and the “equilibrium” value obtained upon cooling from above $T_\text{c}$ is reached. The speeds of $v_0 = 1.3\,\mu\text{m s}^{-1}$ and $21\,\mu\text{m s}^{-1}$ correspond to $\sigma = 2.0\,\text{mPa}$ and $32\,\text{mPa}$.

In the context of superconductivity or magnetism (or superfluidity), the system is prepared differently if (velocity) field-cooling or zero field-cooling are employed. The hysteresis shown in Figs. 4 and 5 is a consequence of what the authors of Ref. 4 have called vortex glass behavior. If the state of the system is prepared at low $v_0$ there is only a “small” number of vortices frozen into the solid. When $v_0$ is gradually increased while $T < T_\text{c}$, NCRIF becomes either unstable (see Fig. 4) or metastable (see Fig. 5). In the former case, NCRIF decays until it reaches a metastable state. For $T > T_\text{c}$, NCRIF is reduced by the enhanced mobility of vortices.

The low temperature metastability seems to depend on the difference between the initial NCRIF prepared at low speed and the “equilibrium” value obtained upon cooling at the higher speed. For instance, the difference is NCRIF = 0.9\% for BC-12/10/06 (see Fig. 4) and NCRIF = 0.08\% for CP-11/21/06 (see Fig. 5), resulting in a much more stable value for the latter. The measured value for BC-12/10/06 immediately drops by about 40\% of its magnitude following the increase in $v_0$, while there is no change for CP-11/21/06. The behavior of CP-11/21/06 is similar to that in Ref. 4, which was compared with the Meissner effect (i.e., a robust NCRIF due to the inability of vortices to enter into the solid). We note that the metastability up to 800 $\mu\text{m s}^{-1}$ in Ref. 4 may be related to the small NCRIF $\approx 0.1\%$ of the sample. In comparison, upon raising $v_0$ to 800 $\mu\text{m s}^{-1}$ we observe, for a 0.3 ppm sample with NCRIF $> 1\%$ ($\delta f_{\text{He}} > 8\,\text{mHz}$), an abrupt drop in the signal that is followed by a gradual decay. In fact, in this extreme limit of $v_0$, the decay rate is essentially insensitive to temperature steps (warming and cooling) between 20 mK and 45 mK. Only upon warming above 60 mK does the decay rate increase further.

In 1 ppb samples, the stability is reduced as the temperature is raised above 30 mK. Fragments of data from scans of CP-11/21/06 are displayed in Fig. 6. The data at any $T < 60\,\text{mK}$ cannot be fit satisfactorily using a simple mathematical formula. For example, the decay at 40 mK in Fig. 6a) (between points A and B) begins immediately, but slows down abruptly after only 1 h. In Fig. 6b), NCRIF barely changes in the first hour at the same temperature (data prior to point G). Irregular rates are even more apparent at 50 mK. The decay slows down dramatically when NCRIF crosses (e.g., points E and H) from the region above the low velocity cooling trace to the region below it. This is most obvious in the Fig. 6(b), where the sample was warmed to 50 mK rapidly to result in a larger initial NCRIF. The very different decay rates in these two regions, and the identical temperature dependence in warming and cooling scans, both suggest that there is physical significance to the re-
producible data obtained in the low velocity limit. Similar behavior could not be observed in BC samples because of higher decay rates, i.e., it was impossible to exceed the low velocity trace upon warming (see Fig. 1).

Interestingly, the decay in CP-11/21/06 at 60 mK (data following point I) is different from that at 40 mK and 50 mK in that it can be well fit to an exponential form, which yields a time constant of 2.25 h. The smooth decay across the low velocity trace may indicate that the small tail of NCRIF for \( T \geq 60 \text{ mK} \) does not denote the same “boundary” as seen at lower temperatures. This observation is complemented by the enhanced decay rate at 60 mK that we find in 0.3 ppm samples as the temperature is lowered we find NCRIF to “jump” up to a steady value (see the data in Fig. 5 following point F). If we naively estimate the frequency shift due to the presence of a single vortex line using the expression\(^{25}\),

\[ f_{\text{shift}} = \frac{2\pi m}{\hbar} \frac{\Delta \sigma}{\rho} \]

we get \( 0.04/0.33 = 11\% \) for CP-11/21/07).

Although our experimental results can be qualitatively interpreted within the framework of Anderson’s vortex liquid model, very few quantitative comparisons are possible\(^ {29}\). One of the present experimental results that is difficult to understand in the vortex picture is depicted in Fig. 3. In two samples the observed frequency shift continues to increase when \( v_0 \) is reduced to 1 \( \mu \text{ms}^{-1} \), such that even the peak to peak amplitude of 2 \( \mu \text{ms}^{-1} \) is less than that which corresponds to a single quantum of circulation (~3.5 \( \mu \text{ms}^{-1} \)).

**TABLE I: Approximate stress amplitudes corresponding to \( v_C \) in various cells.** Values are estimated by ignoring the effects from the top and bottom surfaces of the cell and calculating two different expressions for the torque: \( I_\alpha = r A \sigma \). Here, \( \alpha \) is the angular acceleration and \( A \) is the area at the rim of the cell. The stress exerted in an annular geometry differs from that in a cylindrical one by the approximate factor, \( (2f) \lambda_{\text{ann}}/(rf)_{\text{cyl}} \).  

| Cell       | \( f \) [Hz] | \( v_C \) [\( \mu \text{m}s^{-1} \)] | \( \sigma_C \) [mPa] |
|------------|--------------|-------------------------------------|---------------------|
| Cylinder\(^a\) | 1072         | < 3.5                               | < 5.4               |
| Cylinder\(^b\) | 1173         | 15                                  | 12                  |
| Cylinder\(^b\) | 496          | 15                                  | 28                  |
| Annulus\(^c\) | 912          | 3-40                                 | 1.6-22              |
| Annulus\(^d\) | 912          | 10                                   | 5.4                 |
| Annulus\(^e\) | 874          | 40                                   | 3.3                 |

\( ^a \) Present set of measurements.

\( ^b \) Data from Ref. 6.

\( ^c \) Data from Ref. 7.

\( ^d \) Data from Ref. 2.

\( ^e \) Data from Ref. 4.

\( I_{\text{vort}} = \pi \rho r^4 \text{NCRIF}/\ln(r/a_0) \), we calculate a shift corresponding to \( 25\% \) of the observed NCRIF for a vortex core radius\(^ {29} \) of \( a_0 = 0.1 \) nm. This value is of the same order of magnitude to what we observe (e.g., from Fig. 5 we get 0.04/0.33 = 11\% for CP-11/21/07).

**IV. ANOMALOUS ELASTICITY OF SOLID \(^4\)He**

Due to the similarities discussed in Sec. 1 between the observed frequency shifts and the results of Ref. 23, we are compelled to interpret the data from Sec. 1 in terms of the response of the dislocation network to oscillating stress fields imposed on the solid.

Here we report our preliminary estimates of changes in the shear modulus of solid \(^4\)He. By calculating\(^ {24}\) the resonant frequency of the torsion mode for different \( c_{44} \) values we extracted the percentage change in the modulus that corresponds to a small frequency shift of the TO. The change is given by \( \delta c_{44}/c_{44} = (d\ln f/d\ln c_{44})^{-1}(\delta f_{\text{He}}/f) \), where the derivative term comes from the FEM calculation and frequency shift comes directly from our TO measurements. The results of our calculations are shown in Figs. 4 and 5 since the low temperature change in the resonant frequency is very small (parts per million), it scales linearly with the change in the shear modulus.
a result the inferred temperature dependence of $\delta c_{44}/c_{44}$ is identical to $\delta f_{He}$. It is therefore natural that the qualitative features of the data in Ref. 23 be very similar to that from TO studies.

However, the magnitude of the relative change in $c_{44}$ appears to be different. In order to account for the observed $\delta f_{He}$ in TO measurements the required $\delta c_{44}/c_{44}$ values among the samples studied are between two and five times larger than those seen in Ref. 23. Moreover, the changes are greater than the theoretical upper limit of 30% that can result from the pinning of dislocation lines. We have carried out similar calculations for some of the TO's in the literature and found that even larger values of $\delta c_{44}/c_{44}$ are necessary to account for the observed $\delta f_{He}$. For instance, typical NCRIF values in Refs. 24,52 all translate into an increase in $c_{44}$ by approximately a factor of 10. For the double oscillator from Ref.7, we find $c_{44}$ to increase by factors of 5 and 10 in the anti-symmetric and symmetric modes, respectively. This implies that the effect is very dependent on frequency, contrary to the findings of Day and Beamish. Furthermore, it is difficult to apply the dislocation pinning picture to the experiments in porous Vycor glass, in which the dimensions of helium (i.e., the pore size) are smaller than typical dislocation loop lengths in bulk crystals. This, and the fact that the crystalline phase of the confined $^4$He is body-centered-cubic rather than hexagonal-close-packed, would lead one to expect very different behavior in the response to torsional oscillations, counter to what is observed.

Day and Beamish found the measured shear modulus to be a function of stress amplitude. Below a critical stress $\sigma_C$ the magnitude of $c_{44}$ saturates at a maximum value, as does $\delta f_{He}$ in some samples (see Fig. 3). This critical value, $\sigma_C = 300$ mPa, is smaller than the estimated breakaway stress of 4 Pa for a $^3$He-$^3$He separation of 5 nm (a typical loop length) along a dislocation, but closely matches the stress at which $\delta f_{He}$ tends to zero in Fig. 3. Such strong nonlinearity between the drive and response, also seen in other low frequency acoustic measurements, occurs at “critical” speeds that are 100 to 1000 times less (e.g., ~50 nm s$^{-1}$ in Ref. 23) than in TO studies. It was concluded in Ref. 23 that $\sigma_C$ is a more fundamental quantity than $v_C$ due to it being independent of the measurement frequency. However, this is inconsistent with Ref.7 and implies that the two types of measurements, although clearly related on some level, are distinct.

V. CONCLUSIONS

We have studied a number of solid $^4$He samples grown within a torsional oscillator at constant temperature and pressure, as well as with the blocked capillary method, and found that the resonant frequency of the system possesses a very strong thermal history. The magnitude of the apparent NCRIF measured in the low temperature limit is reproducible when obtained upon cooling the sample from temperatures well above the transition. Modulation of the oscillation speed below the onset temperature reveals the existence of many metastable states.

We find that these properties are qualitatively consistent with two different interpretations: the response of vortices to velocity fields and/or the response of dislocation lines within the solid to oscillating stress fields. The mobility of either of these entities becomes very limited with decreasing temperature, resulting in hysteretic behavior below 60 mK. Extreme pinning takes place below 30 mK (1 ppb) or 45 mK (0.3 ppm), such that their motion is essentially frozen out.

At present there remain discrepancies between the experiments and either of the two interpretations. In the vortex liquid picture, the most notable problem is the very low critical velocity found in this study. In regard to the elastic properties of solid $^4$He, we find that the majority of the frequency shifts observed are appreciably larger than upper limit expected from the pinning of dislocations.

Acknowledgments

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We note that Paalanen et al. previously reported a 40% change in the shear modulus, but this included high temperature data near the melting point where there are likely other stiffening processes to account for (e.g., the presence or absence of thermally activated vacancies).
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