Finite size Berezinski-Kosterlitz-Thouless transition at grain boundaries in solid $^4$He and role of $^3$He impurities

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We analyze the complex phenomenology of the Non-Classical Rotational Inertia (NCRI) observed at low temperature in solid $^4$He within the context of a two dimensional Berezinski-Kosterlitz-Thouless transition in a premelted $^4$He film at the grain boundaries. We show that both the temperature and $^3$He doping dependence of the NCRI fraction (NCRIF) can be ascribed to finite size effects induced by the finite grain size. We give an estimate of the average size of the grains which we argue to be limited by the isotopic $^3$He impurities and we provide a simple power-law relation between the NCRIF and the $^3$He concentration.

The report of a Non Classical Rotational Inertia (NCRI) in solid $^4$He [1, 2] has opened an intense debate in the physics community about its possible “super-solids” (SS) nature. Although the observation of NCRI has been confirmed by other groups [3, 4, 5], its phenomenology presents strong discrepancies with a simple supersolid phase, so that the precise origin of this phenomenon is still unclear. On one hand the supposed SS transition appears to be anomalously broad in temperature [1, 2]. On the other hand the NCRI strongly depends on the external conditions [1, 2, 4, 5]. In Refs. [1, 2] for instance annealing was shown to reduce and even to make disappear the NCRI fraction (NCRIF). Moreover, using a different set-up, in Ref. [4] it was shown that the mass flow, associated with a SS phase, occurred only in the presence of grain boundaries (GB), and it was absent when GBs were not detected. This observation gives rise then to an alternative hypothesis to the SS phase, namely, that a liquid phase is confined at the GBs and that the mass flow is related to superfluidity of the liquid component, similarly to a Rollin film. Partial wetting of GBs was experimentally observed in Ref. [10], and the possible superfluid (SF) ordering was investigated in Refs. [11, 12, 13]. Interestingly enough, a change of the shear modulus has also been observed at the low temperature, with a similar dependence on annealing and on $^3$He concentration as the NCRIF [14]. The connection between these two quantities is thus worth to be further investigated.

Quite puzzling is also the dependence of the NCRI phenomenology on the $^3$He concentration $x_3$. The first report of NCRI [1] was observed in commercial $^4$He, which contains generally a low concentration $x_3 \sim 0.3$ ppm. Further investigations showed that the critical temperature increases monotonically with $x_3$ whereas NCRIF increases with $x_3$ only up to an optimal doping at $x_3 \sim 300$ ppb, after which the magnitude decreases [15].

In this paper we propose that, due to the strong confinement on the grain boundaries, the SF transition of the premelted liquid component can be described in terms of a two dimensional Berezinski-Kosterlitz-Thouless (BKT) superfluid transition where the grain size gives rise to finite size effects. We propose also a simple model where the concentration of $^3$He impurities rules the grain size, and hence the finite size effects. We show that this framework can explain in a natural way, for $x_3 \leq 300$ ppb, the broadness of the SF transition and the dependence of the NCRIF on the $^3$He impurity concentration.

In the following we shall model the polycrystal $^4$He samples in terms of spherical grains with radius $R$ and probability distribution function $P(R)$. Premelting effects, as discussed in Refs. [10, 11, 12, 13, 16], is expected to give rise to a thin liquid film with thickness $d$. Partial wetting [11, 12], reducing the liquid amount covering the grain, can be also considered but it will not change our results. We assume that $d \ll R$, so that the liquid helium system confined on the GB surface can be regarded as two-dimensional. This kind of model was employed by Kotsubo and Williams (KW) to explain the behavior of SF $^4$He films on different substrates [17]. The important ingredient within this context is that the size $R$ of the grain provides an intrinsic finite size cut-off which makes the BKT transition to be smooth. The broadness of the NCRI transition can be thus employed to estimate the average size $R_0$ of the GBs.

The BKT self-consistent equations on a spherical geometry were discussed in Ref. [17] by KW. We can define the energy $U_0(\theta)$ of an isolated vortex-antivortex pair at angular distance $\theta$, in units of $k_B T$, as

$$U_0(\theta) = 2U_c + \int_{2\theta_c}^{\theta} \frac{\pi K_0}{\tan[(\theta' - \theta_c)/2]} \, d\theta', \tag{1}$$

where $K_0 = \hbar^2 a_s^0 / m^2 k_B T$ is related to the bare areal SF density $a_s^0$, $m$ is the $^4$He mass, $U_c$ is the vortex core energy, and $\theta_c = a_0/R$ is the minimum vortex-antivortex angular distance given by the vortex core size $a_0$. In the presence of screening effects due to vortex pair polariz-
tion, we can generalize Eq. (1) as

\[ U(\theta) = 2U_c + \int_{0}^{\theta} \frac{\pi K_0}{c_0(\theta)} \tan \left[ (\theta' - \theta_c)/2 \right] d\theta', \quad (2) \]

where the static dielectric constant \( \epsilon_0(\theta) \) can be evaluated in a self-consistent way as:

\[ \epsilon_0(\theta) = 1 + \frac{4\pi^3 K_0}{2(2\pi)} \int_{0}^{\theta} d\theta' \theta'^2 \sin \theta' \exp[-U(\theta')]. \quad (3) \]

Eqs. (1)-(3) can be evaluated self-consistently for all \( \theta \leq \pi \) to obtain the observable SF density \( \sigma_s(T) = \sigma_s^0/\epsilon(\theta = \pi) \) as function of temperature. Eqs. (1)-(3) can be also generalized to the dynamical case by introducing the dynamic dielectric constant \( \epsilon(\theta, \omega) \) which depends on the diffusion constant of the vortices \( D \) and on the angular frequency \( \omega \) through the parameter \( r_D = \sqrt{2D/\omega} \). In the physical range of the experimental setup \( r_D, a_0 \gg 1 \) and the evaluation of the SF density \( \sigma_s \) in the dynamical regime is practically indistinguishable from the static one. The introduction of the dynamical analysis permits however the evaluation as well of the change of quality factor \( \Delta Q^{-1} \).

Let us now apply the above analysis to our polycrystal spherical-grain model. For a single grain of size \( R \), assuming \( d \ll R \), we can estimate the temperature dependent NCRIF \( n_s(T, R) \) as

\[ n_s(T, R) \simeq \frac{4\pi R^2 \sigma_s(T)}{4\pi \rho R^3/3} = \frac{3\sigma_s}{\rho R}, \quad (4) \]

(where \( \rho \) is the solid \( ^4 \)He density), and a mean \( n_s(T) = \int dR P(R) n_s(T, R) \). In the following we shall show that the SF temperature profile is mainly ruled by the mean grain size value \( R_0 \). In this case we can roughly estimate the zero temperature NCRIF \( n_s \simeq 3\sigma_s^0/\rho R_0 \). Note also that in the \( R_0 \gg a_0 \) limit the areal SF density \( \sigma_s^0 \) is roughly proportional to the Berezinski-Kosterlitz-Thouless temperature \( T_{BKT} \) where \( \sigma_s(T) \) drops to zero. Within this context thus the effect of finite GB size \( R \) is mainly to give rise to a significant broadness of the transition whereas the temperature profile of the transition is only weakly affected. If we assume \( \sigma_s^0 \simeq \sigma_s(T_{BKT}) = (2m^2/\pi h^2) k_B T_{BKT} \), and we estimate \( T_{BKT} \) from the temperature \( T_{50} \) at which \( n_s(T) \) drops to its 50 % value of \( n_s(T = 0) \), we can thus obtain a free fitting parameter estimate of the GB size:

\[ R_0 \simeq \frac{6m^2 k_B T_{BKT}}{\pi \rho h^2 \sigma_s}. \quad (5) \]

We would like to stress that, because of the simplicity of this model and of the slight approximations in the estimates of \( T_{BKT} \) and \( \sigma_s^0 \), Eq. (5) is simply meant to give the order of magnitude of \( R_0 \).

We now apply our model to the specific case of NCRI in solid \( ^4 \)He. We consider a \( ^4 \)He density \( \rho = 0.2 \text{ g/cm}^3 \), which corresponds to a molar volume \( \sim 20 \text{ cm}^3 \) and to a pressure 41 bar. We set also typical values for the vortex core size \( a_0 = 50 \text{ Å} \) and energy \( U_c = 2.5K_0 \). We consider for the moment two extreme probability distribution functions, namely a single value \( P(R) = \delta(R-R_0) \) and a flat \( P(R) = 1/R_0 \) for \( R_0 \leq R \leq R_0 + \Delta R_0 \), with \( \Delta R_0 = R_0/2 \). With this choice of parameters, the overall profile of the NCRIF \( n_s(T) \) is uniquely determined by the only two free parameters, namely \( R_0 \) and \( \sigma_s^0 \), where \( \sigma_s^0/R_0 \) rules the magnitude of \( n_s \) at \( T = 0 \) while \( R_0 \) is related to the broadness of the SF transition.

In order to show the feasibility of this approach to reproduce the experimental results, we compare in Fig. 1 the \( n_s \) vs. \( T \) data for the 133 ppb \( ^4 \)He Penn State (TOP) sample with our best fit, which gives \( \sigma_s^0 = 0.26 \times 10^{-9} \text{ g/cm}^2 \) and \( R_0 = 130 \text{ Å} \), and with a flat \( ^4 \)BKT transition for large grains \( R \gg a_0 \). Note that there is only a slight difference between the \( \delta \)-like and the flat \( P(R) \). As a matter of fact, we have checked that different distribution functions \( P(R) \) do not affect qualitatively our results, so that from now on we shall consider for simplicity a simple \( \delta \)-like distribution. The nice agreement between the experimental data and our results suggests that the NCRI broad transition is not related to inhomogeneities of the samples but it stems from finite size effects due to the finiteness of the \( ^4 \)He grain. It is important to underline that, while the fitting procedure gives a refinement of the GB size, the order of magnitude of \( R \) is essentially given by the relation (5), so that the good agreement in the broadness of the transition is not a result of the fit but it can be considered as an independent check of the validity of our analysis.

The estimate size \( R_0 \simeq 130 \text{ Å} \) of the grains can appear quite puzzling especially considering that \( ^4 \)He at these conditions is thought to solidify in a polycrystal form with macroscopic size of grains. However, on one hand the crystallographic evidence cannot exclude mosaics of small-angle grain boundaries. On the other hand, the presence of such small grains can naturally account for the high sensitivity to annealing and to preparation and freezing procedures, and the similarities with the NCRI phenomenology in Vycor glasses [21]. Nevertheless, the
physical origin of such an extremely small grain size and its dependence on $^3\text{He}$ concentration requires to be explained. We address this issue in the second part of this Letter, where we relate the origin of GB to the presence of liquid melted bubbles induced by $^3\text{He}$ impurities.

In order to gain a further insight on this point, let us discuss before the experimental dependence of the NCRI phenomenology on the $^3\text{He}$ concentration and on the growth/measurement condition. In particular, we analyze two samples [15] in the highly dilute limit $x_3 \simeq 1$ ppb, one belonging to the University of Florida (TOF) family, grown with the blocked capillary method, and one still belonging to the Penn State University but grown at constant pressure (CP). For both these samples measurements of the change of the quality factor $\Delta[Q^{-1}]$ were also available [15].

The first thing to observe is that, despite both samples are at same $x_3$, the signals are significantly different (Fig. 2). For the TOF sample, the value of $n_s$ is almost one order of magnitude smaller than for the CP sample. Its variation as a function of the temperature is also different. In fact, while the $n_s$ presents a quite smooth behavior in the CP sample, the drop of $n_s$ at $T \simeq 0.03$ K is much sharper followed by an additional tail. Similar features are observed in the behaviors of the quality factor $\Delta[Q^{-1}]$ which is quite broad in the CP sample compared to the sharp peak at $T \simeq 0.03$ K in the TOF sample (note that the longer tail in the $n_s$ is not observed in $\Delta[Q^{-1}]$, suggesting that this features in the $n_s(T)$ behavior could have a spurious origin).

We can see now that our model permits to understand in a very natural way also the $x_3$ dependence of the NCRI phenomenology. We first note that, although the two samples have different $n_s(T = 0)$ of a order of magnitude, their $T_{\text{BKT}}$, defined for instance as $T_{\text{BKT}} = T_{\text{BKT}}^0$, are of a similar magnitude. A simple analysis, using again Eq. \ref{eq:Q1}, would point out thus an average radius $R_0$ of the GBs in the TOF sample one order of magnitude larger than in the CP one. As a consequence, finite size effects are expected to give rise to a much sharper transition in the TOF than in the CP case, in agreement with the experimental observation. These simple considerations are corroborated by a more detailed analysis. In Fig. 2 we show our best fits for both the TOF and CP samples, compare with the experimental data. Estimates for $R_0$ and $\sigma_0$, in these cases, are respectively $R_0 = 790$ Å, $\sigma_0 = 0.105 \times 10^{-9}$ g/cm$^2$, and $R_0 = 100$ Å, $\sigma_0 = 0.216 \times 10^{-9}$ g/cm$^2$. For the evaluation of the quality factor $\Delta[Q^{-1}]$ we have used respectively $r_D = 1.2 \times 10^4$ Å and $r_D = 0.3 \times 10^4$ Å, which give $r_D/R \gg 1$. We remind that in such quasi-static regime the parameter $r_D$ simply acts as a scale factor on $\Delta[Q^{-1}]$ while it does not affect significantly the NCRI. The reliability of such limit is confirmed by Ref. [16] where the magnitude of $\Delta[Q^{-1}]$ was shown to be sensibly affected by changing the set-up frequency while the NCRI was essentially untouched.

We can see that the finite GB size BKT theory can naturally account not only for the broad NCRI transition but also for the experimental height and broadness of the $\Delta[Q^{-1}]$ factor. This latter point is not surprising since in the finite size BKT framework, the broadness of the $n_s$ transition and of the $\Delta[Q^{-1}]$ peak are related. Although our theory can account for the difference of the signals in a simple manner, the long tail of $n_s$ for $T > T_{\text{BKT}}$ in the TOF sample remains unexplained. As mentioned above, however, it should be noted that this feature of the NCRI has no counterpart in $\Delta[Q^{-1}]$, which is sharply located at $T \approx T_{\text{BKT}} \leq 15$. This suggests that the long tail in $n_s(T)$ might be due in this sample to different physics not related to the superfluid GB transition.

After assessed the robustness of our analysis in explaining the NCRI phenomenology as function of $x_3$ and growth conditions, in the last part of this Letter we address the origin itself of such $x_3$-dependence. In particular we propose that the main effect of $^3\text{He}$ impurities is to provide an intrinsic maximum length scale for the growth of grains, favoring thus the presence of grain boundaries, and hence to sustain a superfluid NCRI on the GB surfaces. It should be clear, on the other hand, that additional sources of disorder, favoring the GB formation, can be present, as shown by the different phenomenology for different growth conditions and by the annealing dependence [1, 6]. We assume for the moment that the only source of GB is the presence of $^3\text{He}$ impurities. This assumption is probably valid for the TOF samples, which show the smallest NCRIFs for similar $x_3$, and it is corroborated by the sharp drops of $n_s$ at $T_{\text{BKT}}$, suggesting quite large grains. We consider, for dilute $^3\text{He}$ concentrations $x_3 \lesssim 1$ ppm, a uniform distribution of the $^3\text{He}$ impurities within the sample, with an average distance $d_{\text{He}}$ between $^3\text{He}$ atoms $d_{\text{He}} \approx x_3^{-1/3}$. In Ref. [16] it was shown that $^3\text{He}$ impurities, due to the stronger quantum fluctuations of zero point motion, induce a local melting of the host $^3\text{He}$ even at low temperature much smaller than the bulk $^3\text{He}$ melting. The presence of local liq-

FIG. 2: (Color online) Comparison between theory and experimental NCRIIF and $\Delta[Q^{-1}]$ data taken from Ref. [15] for the 1 ppb TOF (upper) and CP (lower) samples.
A simple estimate gives for instance $R_{\text{ter}} 2$ in the last analysis, an average value of the grain diameter $2R_0$ as larger than the vortex size $a_0 = 50$ A. In this regime the reliability of the mean-field analysis is strongly questionable.

In conclusion, in this Letter we showed that the complex NCRI phenomenology is fully compatible with a superfluid Berezinski-Kosterlitz-Thouless transition induced in the thin premelted liquid film at the grain boundaries. Note by the way that the two dimensional BKT transition is characterized by the lack of specific heat anomalies, in agreement with the heat-capacity measurements in solid $^3$He. Both the temperature and $x$ dependence are shown to be ascribable to finite grain size effects. We propose also a simple picture where $^3$He impurities are directly related to the maximum size of the grains and we predict a simple scaling relation between the NCRIF $n_s$ and the $^3$He impurity concentration.

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FIG. 3: (Color online) (a) schematic sketch of GB structure of $^4$He samples in the presence of $^3$He impurities. Light blue areas represent solid $^4$He, white spots $^3$He impurities and dark blue regions premelted $^4$He at the GBs and around $^3$He impurities. (b) $n_s$ vs. $x_3$ plot of different samples (symbols) taken from Ref. [15] compared with our model prediction [Eq. (6)] (solid line), which is expected to fail in the gray region where $R \approx 2 - 3a_0$ and the mean-field theory breaks down.

Water spots, which we remind survive also at virtually zero temperature, around the $^3$He impurities poses a strong constraint on the growth of the grain size in the freezing process. The average distance $d_{^4\text{He}}$ provides thus the maximum length scale for the growth of grain size and, in the last analysis, an average value of the grain diameter $2R_0 \approx d_{^4\text{He}}$ (see Fig. 3) for a sketched picture. This simple estimate gives for instance $R_0 \approx 1600$ A for $x_3 = 1$ ppb and $R_0 \approx 313$ A for $x_3 = 133$ ppb, 2-3 times larger than the actual fit estimates. Taking into account that these figures are purely indicative since other sources of disorder are always present further limiting the size of the grains, such estimates are not bad and provide the order of magnitude of the size of the grains. The most convincing probe of such picture is the power-law dependence of the behavior of $n_s$ as function of $x_3$, $n_s \approx x_3^{1/3}$. Assuming that a $p_s$ fraction of the thin liquid film of thickness $d$ at the GBs undergoes a superfluid BKT transition, and approximating the liquid $^4$He density with the solid one, we have an areal SF density at the GBs $\sigma_s = p_s d\rho_s$, and, from Eq. (4),

$$n_s \approx 6 p_s d / d_{^4\text{He}} \approx 6 p_s d x_3^{1/3} / a,$$

where we have assumed $d \ll R_0$ and $R_0 \approx d_{^4\text{He}} / 2$. where $a = 3.2$ A is the solid He-He distance at the pressure here considered. Such power-law behavior, assuming for instance $d = 1.2$ A and $p_s = 0.01$, is shown in Fig. 3b in qualitative agreement with the experimental data for the TOF samples and the lowest $x_3$ TOF samples [15] where other sources of disorder are thought to be small. Note that the unknown quantities $p_s$, $d$, in the log-log plot of Fig. 3a, determine only the vertical off-shift of the $n_s - x_3$ behavior but not its slope which is uniquely determined by geometrical considerations. In CP samples on the other hand additional limiting mechanisms on the grain size are probably operative conceal-

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