Nematic disclination in the ground state of solid $^4$He confined to nanoporous materials

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The ground state of solid $^4$He in a cylindrical nanopore hosts a topological linear defect which can be viewed as a nematic-type Frank’s disclination. The associated singular strain (or, rather, splay) may cause partial melting around the line to create a superfluid core of the disclination. The resulting phase, compactified supersolid (CSS), is studied by ab initio Monte Carlo simulations and by a simple model explaining its main feature – a gradual decrease of the superfluid response with pressure observed in vycor. The CSS is found to transform into insulating compactified solid (CS) by a first order transition with very wide hysteresis.

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In the last decade, the interest in solid $^4$He has been revived by the claims of observing supersolidity in the torsional oscillator (TO) experiments [1–6]. Although the alternative explanation in terms of the dislocation plasticity [7] gains momentum, the reports of the direct superflow through the solid $^4$He [8], which is consistent with the presence of dislocations with superfluid cores [9–10], make the subject even more fascinating.

One of the longstanding open questions is the nature of solid $^4$He confined to a mesoscopic geometry, such as, e.g., vycor or gelsil glass. A number of recent experiments aim at observing one-dimensional flow through porous materials with small diameters [11–13] in an attempt to answer this question. The recent structural study [14] finds that solid $^4$He at low pressure is likely to be in the hcp phase and transforms into hcp only at higher pressure. In fact, the original claim of the supersolid response was about $^4$He in vycor [1–6].

Superfluidity of $^4$He in vycor persists at a pressure $P$ as high as 10–20 bar above the melting pressure $P_m \approx 25$ bar in macroscopic samples. Several models have been proposed to explain this effect [15–15], including the conjecture that $^4$He remains liquid close to vycor wall with the solid forming away from the wall [17]. The picture of a persistent superfluid layer was supported by Monte Carlo simulations of about 200 $^4$He atoms [19]. A variational study of $^4$He in a cylindrical pore [20] has found that $^4$He forms shells concentric with the pore wall. These shells are hexagonal layers rolled into cylinders which are also superfluid. An ab initio Monte Carlo study [21] of $^4$He confined to a cylinder at saturated vapor pressure has found the shell structure as well, with, however, no intra-shell structural order. While for a pore with a diameter $R_0 = 2.9\,\text{Å}$ is insulating, a pore with $R_0 = 14\,\text{Å}$ demonstrates weak superfluidity.

In this letter we show that the key to understanding the nature of confined phases of solid $^4$He is the topology of the local hcp axis: its winding induced by the confined geometry and by the strong attraction of $^4$He atoms to the wall creates a topological defect which closely resembles a nematic disclination with Frank index 1 (see Ref. [22]). This disclination precipitated by the densest (hexagonal) 2D layer formed at the wall produces a long-range strain (splay) which may partially melt the solid in close analogy to the strain induced supersolidity in dislocation cores in macroscopic samples [23]. There is, though, a significant difference between the two: in contrast to dislocations, the nematic-type disclination is a part of the ground state of solid $^4$He compactified into a nanopore. We also note a crucial difference between the true nematic disclination and the defect we discuss here: while the first one is unstable in nematic liquid crystals (see Ref. [22]), the roton feature of the spectrum of $^4$He induces the layered structure [24] and, thus, stabilizes the defect — in a manner similar to smectic-A liquid crystals.

We find the following states of $^4$He inside the nanopore: i) A surface phase featuring a few surface layers (shells) adsorbed on the wall, with pore bulk being empty. These layers can be insulating and superfluid (SF); ii) Bulk SF phase. It consists of the crystalline surface shells coexisting with a bulk superfluid, characterized by the roton-induced radial density modulation. This modulation becomes stronger with increasing pressure $P$ and undergoes a crossover to the CSS state. iii) The bulk CS which features hexagonal layers rolled into concentric shells occupying the whole pore. In contrast to the claim made in Ref. [20], the CS shows no superfluid response. There is a first order phase transition between the CS and CSS, which is characterized by wide hysteresis – much wider than that in the case of the standard liquid-solid transition in macroscopic samples of $^4$He.

Model. We consider a grand-canonical ensemble of $N \sim 600–2000$ $^4$He atoms, depending on external chemical potential $\mu$, at temperature $T = 0.2\,\text{K}$ and con-
where in the cylindrical geometry $\xi$ pore wall and geometry. The precise shape of $V$ wall. We intentionally ignore the wall dynamics as well.

The potential $\frac{\hbar^2}{2m} \sum_{i=1}^{N} \nabla_i^2 + \sum_{i<j} V_{\text{Aziz}}(r_{ij}) + \sum_{i} V_{\text{sub}}(r_i)$, \( i-th \) $^4$He atom located at $\vec{r}_i$; $V_{\text{Aziz}}(r_{ij})$ is the standard central Aziz-potential \[25\], with $r_{ij} = |\vec{r}_i - \vec{r}_j|$. The potential $V_{\text{sub}} = \frac{D}{2} \left( \frac{b^2}{\xi^2} - 3 \frac{b^3}{\xi^3} \right)$, $b = 2.0 \text{Å}$, $D = 80 \text{K}$, between the core and $^4$He atoms is the so called 3-9 potential \[21\], where in the cylindrical geometry $\xi = R_0 - r > 0$, with $R_0 = 25.8 \text{Å}$ defining the radial position of the hard wall. We intentionally ignore the wall dynamics as well as its roughness in order to reveal the role of the confined geometry. The precise shape of $V_{\text{sub}}(r_i)$ does not change anything qualitatively (cf. Ref. \[26\]) as long as its depth $D = 80 \text{K}$ is much bigger than that ($\approx 11 \text{K}$) of $V_{\text{Aziz}}(r_{ij})$.

Results of the simulations. We have conducted \textit{ab initio} MC simulations by the Worm Algorithm \[27\] [28]. The particle number $N$ as a function of $\mu$ is shown in Fig. 1. At $\mu < -50 \text{K}$, the outermost shell becomes populated and forms a superfluid. It solidifies into a hexagonal (insulating) shell at $\mu \approx -30 \text{K}$. This stage is not reflected in Figs. 1,2. The second shell forms in the range $-12 < \mu < -7 \text{K}$. It is a low density superfluid which exhibits no visible structural order (see Fig.1 of the Suppl. Mat. \[29\]). Accordingly, the curves in Figs. 1,2 show linear dependencies with $\mu$ in this range. During this stage the pore bulk remains empty. At $\mu \approx -7 \text{K}$ $^4$He undergoes a dimensional crossover marked by the jumps in the plots $N$ vs $\mu$, Fig. 1 and $\rho_s$ vs $\mu$, Fig. 2.

FIG. 1. (Color online) The number of particles as a function of chemical potential for two phases CSS and CS. The both sided arrow indicates the ending of the hysteresis at $\mu \approx 3.2 \text{K}$. Inset: a columnar view along the cylindrical axis of a typical atomic configuration of CSS (left) and CS (right) both at $\mu = 7.1 \text{K}$. The solid (red) circle outlines the pore boundary at $R = R_0$. The pore length is 30 Å. The quasi-disordered region in the CSS is the superfluid core.

FIG. 2. (Color online) Superfluid stiffness $\rho_s$ vs $\mu$ of $^4$He in the nanopore. The solid red line is the fit by Eq. 2. The two-sided arrow indicates closing of the hysteresis at $\mu \approx 3.2 \text{K}$ as in Fig. 1.

FIG. 3. (Color online) Atomic, $n(r)$, and condensate, $c$-map, densities along the radial direction in the samples corresponding to $\mu = 7.1$ and $30 \text{K}$. 

$\rho_s[\text{a.u.}]$
correspond to filling the whole pore by $^4$He, so that the bulk region can be characterized as a low density superfluid. In this phase, while only two outer shells are clearly defined and possess the hexagonal order, the weak radial density modulations, as a precursor of other shells, can also be detected in the pore bulk. These modulations, shown in Fig. 2 of the Suppl. Mat. [29], are the consequence of the roton minimum in the spectrum of liquid $^4$He [24].

The CS begins as a metastable phase at $\mu \approx 3.2\text{K}$ as shown in Fig. 1. The shells (we observed eighth) of a CS are well-defined and exhibit hexagonal order consistent with the whole hcp crystal being compactified (see the Suppl. Mat. [29] for details). There is also a central (insulating) core hosting $^4$He atoms along a very narrow straight line coinciding with the cylinder axis. The CS phase is characterized by zero superfluid response as shown by the data $\rho_s = 0$ for $\mu > 3.2\text{K}$, Fig. 2. A weak dependence of $N$ vs $\mu$ of the CS, Fig. 1, indicates that doping is still possible in this insulating state. However, the extra particles (or vacancies) do not form a superfluid. Instead, they create regions of phase separated vacancies or interstitials—very similar to the case of macroscopic samples studied in Ref. [30]. Lowering $\mu$ below $\mu \approx 3.2\text{K}$ results in a jump-like melting of CS into the bulk SF. This indicates closing of the hysteresis at its low end as marked by the double sided arrows in the curves $N$ vs $\mu$, Fig. 1, and in $\rho_s$ vs $\mu$, Fig. 2.

While the CS is metastable at $3.2\text{K} < \mu < 7 - 10\text{K}$, the CSS is stable in this region and becomes metastable above $\mu \approx 7 - 10\text{K}$. Due to a very wide hysteresis more accurate finding of the transition point turned out to be a very challenging task. As Fig. 1 indicates, the upper end of the hysteresis—where the metastable CSS transforms into the stable CS—could not be determined: CSS persisted at $\mu$ as high as $38\text{K}$. This situation should be contrasted with the results of our simulations of $^4$He confined into a slab with flat smooth wall and periodic boundary conditions along the wall (so that no disclination exists) producing the same 3-9 potential $V_{\text{sub}}$: while the lower end of the hysteresis of the SF-solid transition found to be at $\mu \approx 3.0\text{K}$ is close to that in the nanopore ($\mu \approx 3.2\text{K}$), the upper end is at $\mu \approx 7.0\text{K}$.

A typical atomic configuration of the CSS is shown in the left inset in Fig. 1. It features well defined outer shells (3 of them at $\mu = 7.1\text{K}$), each with slightly distorted hexagonal order (shown in Figs. 4.5 in the Suppl. Mat. [29]), as well as the superfluid core which is visibly disordered within the radius $R \sim 12 - 15\text{A}$. Most of the superfluid response comes from this core. Despite being apparently fully disordered, there are distinct radial density $n(r)$ as well as superfluid density (represented by the so called condensate map, c-map [31]) modulations observed in the core, Fig. 3.

Increasing $\mu$ in the CSS phase leads to the compression of the superfluid core and to a corresponding gradual suppression of the superfluid stiffness $\rho_s$ as seen in Fig. 2. The core compression can also be recognized in Fig. 3. The c-map of the $\mu = 30\text{K}$ sample is more concentrated in the center of the pore than that of the $\mu = 7.1\text{K}$ sample. The corresponding mechanism behind such compression, which we consider as the main cause of the gradual suppression of $\rho_s$ with increasing pressures ($\mu$ in the simulations), will be discussed below. On top of this overall suppression, there are additional peaks and dips in $\rho_s$ vs $\mu$, Fig. 2 which may be related to structural fluctuations caused by the proximity to the CS phase.

We have also determined the structure factor of the pore averaged over its orientations, Fig. 4. It features three main peaks in the region of the momenta $\sim 2.0 - 2.2\text{A}^{-1}$—one strong and two satellites reminiscent of the three main Bragg peaks of hcp solid. The higher order peaks are washed out by quantum fluctuations and are "hidden" under the wide shoulder at high momenta. Thus, despite being compactified, the hcp-type geometry of the atomic arrangement can be determined in X-ray scattering experiments.

**Nematic disclination with superfluid core.** The CS and CSS phases are characterized by winding of the local $C_6$ axis around the pore axis. It is induced by a strong attraction to the pore wall which adsorbs 2D hexagonal layers with the local $C_6$ axis oriented along the radial direction $\hat{r}$. This can be described by the director field $\hat{C} \sim \hat{r}$ in the same manner as in a nematic disclination with Frank index $n = 1$ (see, e.g., Ref. [22]). The associated energy density (of splay) has a singularity $\sim (\nabla \cdot \hat{C})^2 \propto 1/r^2$, where $r$ is the radial distance from the disclination core located along the cylinder axis. The emergence of the splay singularity can be understood as
follows. Like in smectic-A liquid crystals, the orientation of the director is perpendicular to the layers determined by the coarse grained density profile $\rho(\vec{r})$ as $\vec{C} \propto \vec{\nabla} \rho$ (see [22]). In the concentric geometry of the shells $\vec{\nabla} \rho(\vec{r}) \propto \hat{r}$, that is, $(\vec{\nabla} \vec{C})^2 \sim (\vec{\nabla}^2 \rho)^2 \propto 1/r^2$.

It is important that the splay singularity cannot be removed by allowing $\vec{C}$ to smoothly reorient along the cylinder axis (as in the case of a non-singular nematic disclination solution) because of the layered geometry – such a transformation costs a large surface energy (as explained in detail in the Suppl. Mat. [29]). Instead, the singularity can be healed by partial melting of inner shells within some radius $R$ around the disclination line. This, on one hand, gains the splay energy $\sim \int d^2r (\vec{\nabla} \vec{C})^2 \sim \ln R$, and, on the other, costs the bulk $\sim (P - P_m)R^2 > 0$ as well as the surface tension $2\pi \sigma R$ energies, where $\sigma$ stands for the surface tension between CS and CSS. In this simplified approach we ignore the surface energy. Then, the balance of the splay and bulk energies gives the radius of the superfluid core as $R \propto 1/\sqrt{P - P_m}$. Thus, as $P$ increases, the core radius shrinks and so does its superfluid response $\rho_s \propto R^2$,

$$\rho_s \propto \frac{1}{P - P_m}.$$  

(2)

The red line in Fig. 2 is the fit of the numerically found $\rho_s$ by the dependence \eqref{eq:rho_s}, where we have chosen $\mu = \mu_m = 3.5K$ (taken from the simulations of a $^4$He in the slab) as corresponding to the melting pressure $P_m$, so that $(P - P_m)/P_m = (\mu - \mu_m)/\mu_m$, and also we consider the limit $R \ll R_0$.

Thus, the CSS phase is, actually, the CS where several central shells have lost part of their hexagonal structure as well as of the radial contrast and acquired superfluid order instead. In the simulations we associate the CSS to CS transition with vanishing of the superfluidity. Understanding of this transition in terms of the Landau description by the variables typical to liquid crystals, crystals and superfluids, as well as the consideration of the role of disorder of the pore walls, are left for future work.

Discussion. Our analysis, simulations and conclusions about the compactified nature of solid and supersolid $^4$He in nanopores are directly relevant to pores with radii below a threshold, $R_{\text{max}} \sim 300\text{Å}$ (as estimated in the Suppl. Mat. [29]), which is well above typical radii in vycor or gelsil glass. We consider it as a lower bound because the CS or CSS may exist as metastable phases in much larger pores due to the geometrical (macroscopic) energy barrier between the compactified and the standard hcp solids.

The mechanism we have identified should be compared with the wetting model \cite{15,17}, where $^4$He at the wall remains liquid until pressure overcomes the surface tension nucleation barrier. This model has trouble explaining the observed gradual decrease of the superfluid response with pressure \cite{17} because the nucleation mechanism implies an abrupt solidification and, accordingly, an abrupt drop of the superfluidity. In contrast, as we have found, the CSS is characterized by gradual shrinking of the superfluid core, that is, by gradual decrease of its superfluid response with pressure. Our observations are also in a contrast to the persistent liquid layer model \cite{19}, and to the variational results \cite{20} predicting that the solid in a nanopore is always a supersolid.

In the recent study \cite{14}, $^4$He in vycor was found to be in bcc phase at $P < 98$ bar and $T \approx 0.5 – 0.7K$, whereas the transformation to a hcp solid takes place at higher pressure. The structure factor for CS found in our simulations, Fig. 4 is strikingly similar to that found in Ref. \cite{14} at high pressure. In future work it would be important, on the one hand, to repeat the experiment \cite{14} at lower temperatures, and, on the other, to perform the MC simulations at temperatures higher than $T = 0.2K$ considered here. One possibility is that there is a non-trivial transition line in the $P - T$ plane where the compactified hcp solid becomes a compactified bcc.

An interesting and important question in the context of the experiments of Ref. \cite{5} is how the nematic disclination ends at the interface between a pore and macroscopic bulk of solid $^4$He.

Conclusion – We have revealed the nature of superfluidity and of the compactified geometry of $^4$He in cylindrical nanopores: there is a topological defect inherently present in the ground state—a nematic-type disclination. The crucial role of the roton feature is in seeding the concentric shells in the pore bulk so that the splay singularity becomes stable and could only be relieved by local melting away from the wall—around the disclination line. This creates a compactified supersolid core which can undergo a first order phase transition into a compactified solid (insulator) above the thermodynamical solid-liquid transition in macroscopic samples. Such a core can also persist in a metastable states at pressures significantly exceeding the spinodal for the over pressured superfluid in macroscopic samples of $^4$He. This finding offers a compelling explanation for the physics of $^4$He confined to restricted geometries where the local $C_6$ axis, playing the role of the nematic director, can not be uniquely defined everywhere. Thus, in the multiple-connected geometry of nanoporous materials confining $^4$He, the superfluid response at high pressure is controlled by a network of nematic-type disclinations.

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Supplemental Material

In the Supplemental Material we illustrate graphically typical atomic configurations, the density profile of the surface layer phase and of the pore filled with superfluid. We also look in more detail into the compactification process by computing the interparticle distances, the strain field and comparing the energies of the compactified and non-compactified structures.

THE SURFACE AND THE LOW DENSITY PHASES

At low $\mu$, at most, the first two outer shells are formed. This phase is called the surface phase (i). The columnar view of a typical atomic configuration at $\mu = -9.375\text{K}$ is shown in Fig. 5. In this phase the superfluid response comes from the second shell (farthest from the wall). Accordingly, the first shell is ordered and the second one is disordered. The bulk phase (ii) begins at $\mu \geq -7\text{K}$. It can be viewed as two outer shells coexisting with the low density superfluid filling the pore bulk. The bulk density $n(r)$ and the superfluid density (shown by the c-map) are both modulated in the radial direction at the wavelength corresponding to the roton. These modulations observed in a sample $\mu = -3.0\text{K}$ are shown in Fig. 6. These are the precursors of the shells which eventually form the CS and CSS.

THE CS VS NON-CS GEOMETRIES

A possible fitting of the $hcp$ structure into a cylinder with least of the bulk strain is shown in the top panel of Fig. 7. In this structure, the strong attractive wall potential creates several (here we show two) outmost hexagonal shells wrapped around the wall. Since being closely packed in 2D, such shells minimize the surface energy. The $C_6$ axis in these shells is oriented radially with respect to the pore axis. In the inner part of the pore, however, the $C_6$ axis is aligned with the cylinder axis similarly to the director in the non-singular nematic disclination solution (see in Ref. [22]). Simulations of pores with radii $< 15\text{Å}$ [20, 21] as well as our present work with the pore of almost twice that radius show that this configuration is not realized at least in pores with radii less than $\sim 30\text{Å}$. The preferred configuration is the compactified $hcp$ solid shown in the bottom panel in Fig. 7. This configuration hosts the singular Frank’s disclination of index $n = 1$, with its axis coinciding with the central core $r = 0$. Atoms there are arranged along a very narrow line. As we will estimate below, the compactified configuration, CS, has lower energy than the standard $hcp$ in pores with radii, at least, up to $R_0 \sim 300\text{Å}$.

Let’s consider in detail the ideal compactified $hcp$ geometry. In order to produce minimal residual strain, the A-B hexagonal (basal) planes of the standard $hcp$ structure should be rolled into concentric cylinders along the
FIG. 7. (Color online) The columnar views along the pore axis of two possible configurations. Top panel: the non-compactified hcp solid. The \(C_6\) axis is in the radial direction in the outer shells (purple dots) and it becomes along the cylinder axis (perpendicular to the page plane) in the inner part of the pore (blue dots). Bottom panel: The compactified hcp solid. The \(C_6\) axis is along the radial direction (in the page plane) from the cylinder axis. It winds by \(2\pi\) in a manner similar to the director in nematic disclination [22].

The number of unit cells in the \(N\)th shell with radius \(R_N\) is given by \(M(N) = 2\pi R_N/a(N)\), where the length of the unit cell \(a(N)\) may vary from shell to shell. The next shell has radius \(R_{N+1} = R_N + a_z(N)\), where \(a_z(N)\) is the radial distance between the \(N\)th and \((N+1)\)th shells. In a perfect hcp crystal \(a_z = \sqrt{2/3}a\), where \(a \approx 3.6 - 3.7\, \text{Å}\) is the unit cell length in the basal plane. In the compactified hcp solid this relation needs to be relaxed to \(a_z(N) = \gamma(N)\sqrt{2/3}a(N)\) with \(\gamma(N) \approx 1\) in order to minimize the strain. Thus, the radius of the \(N\)-th shell becomes \(R_N = \sum_{N'=1}^{N} \gamma(N')\sqrt{2/3}a(N')\). Expressing \(a(N)\) in terms of the integer number \(M(N)\), we obtain the equation for the shell radii

\[
R_N = 2\pi \sqrt{\frac{2}{3}} \sum_{N'=1}^{N} \gamma(N')\frac{R_{N'}}{M(N')}. \tag{3}
\]

This equation has a solution \(M(N) = 5N\), \(\gamma(N) = 5/(2\pi \sqrt{\frac{2}{3}}) \approx 0.975\). Thus, \(a_z(N)\) is compressed (radia-
ally) by about 2.5% if compared with the standard $\text{hcp}$ crystal.

In addition to the radial compression, there is a shear strain of one shell with respect to its neighbor. The "quantization" rule $M(N) = 5N$, Eq. (3), implies that the circumference of each shell is broken into $5$ equal angular segments, each subtended by an angle $2\pi/5$. Within each segment, the smallest distance between two atoms from neighboring shells reaches the minimum $\sqrt{3}a/2 \approx 0.87a$ along one radial line. [There are five of such lines forming the $C_5$ symmetric pattern.] Thus, this strain can be estimated as $\approx 1 - 0.87 = 0.13$ at its maximum, and about $0.13/2 \approx 0.065$ on average for the whole sample. In simulations we have observed that such strain has been relaxed to about $0.04$ by the static angular modulation of atomic displacement about $0.5\text{Å}$ along the pore axis with the angular period $2\pi/5$. Fig. 8 shows this pattern (see also Fig. 9).

Thus, the CS structure can be characterized by the 0.025 compression strain and by about 0.04 of the shear strain. We estimate the resulting energy change as being due to the elastic energy $\delta E_{el} \sim (0.025^2 + 0.04^2)E_D$, where $E_D$ is determined by elastic constants defining the Debye energy $\sim 30\text{K}$ in of solid $^4\text{He}$. Thus, the compactification costs about extra $0.07\text{K}$ per particle. In other words, the non-compactified $\text{hcp} \ ^4\text{He}$ of the same average density represented in the upper panel in Fig. 7 has less energy $\sim 0.07KR_0^2$ if one ignores the boundary. The boundary between the ideal $\text{hcp}$ and the outer shells are characterized by maximal possible misfit — the $C_6$ axis must rotate by $90^\circ$ in order to be aligned with the cylinder axis. We estimate the energy of such misfit as being larger than $\sim 0.1E_D \sim 3\text{K}$. Thus, the total excess energy of the CS can be written as $\sim -3K \cdot 2\pi(R_0/a) + 0.07K \cdot \pi(R_0/a)^2$. It becomes larger than that of the non-compactified structure at radii larger than $R_0 \approx 90a$. For typical values of $a$ this estimate gives about $R_0 \approx 300\text{Å}$.

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