Mobile solid $^3$He on carbon nanotube promoted by topological frustration

Low-dimensional $^3$He on a nanotube displays dimerization and delocalization of topological defects in its ground state.

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Abstract

Low dimensional fermionic quantum systems are exceptionally interesting because they reveal distinctive physical phenomena, including among others, topologically protected excitations, edge states, frustration, and fractionalization. Two-dimensional $^3$He has indeed shown a remarkable variety of phases of matter including the unusual quantum spin liquid. Our aim was to lower the dimension of the $^3$He system even more by confining it on a suspended carbon nanotube. We demonstrate that $^3$He on a nanotube merges both fermionic and bosonic phenomena, with a quantum phase transition between solid 1/3 phase and a fluid-like solid. The bosonic dimer fluid-like solid contains topology-induced vacancies which are delocalized owing to large zero-point motion. We thus observe a quantum phase transition from fermionic $^3$He crystal in to a bosonic one in quasi-1D geometry.
Physics of quantum liquids and solids is eminently rich and often counterintuitive. Bosonic liquid \(^4\)He possesses superfluidity, e.g. ability to flow without resistance, due to Bose-Einstein condensation of particles below 2.1 K \([1, 2]\). Its fermionic counterpart, liquid \(^3\)He displays superfluidity below 2.5 mK due to paring of atoms into bosonic dimers (so-called Cooper pairs) \([3, 4]\). Moreover, a notion has been put forward that bosonic quantum solid could behave like a fluid, due to mobile delocalized defects such as vacancies. Even more, those vacancies, if present at low enough temperature, must Bose-condense, and the crystal must then behave as a superfluid, keeping at the same time its translational symmetry \([5, 6, 7]\). Search for this spectacular supersolid state has been ongoing since 1980s, and Kim and Chan announced hints of superflow in solid \(^4\)He in 2004 \([8, 9]\). However, later it was established that the equilibrium concentration of vacancies is exponentially small at low temperatures \([10, 11]\), and the observed superflow was most likely through superfluid inside cores of dislocations or on grain boundaries \([12]\). Thus, it is extremely hard to observe a superflow even in “the most quantum” crystal, solid helium, in three dimensions.

Lowering the dimensionality, however, opens up the possibility for topologically stabilized vacancies. In particular, carbon nanotube (CNT), which can be viewed as a rolled-and-glued narrow strip of graphene (a sheet of most symmetric c-plane of graphite), has generally such chirality that its symmetry differs from the regular commensurate-solid superlattice composed of adsorbed helium atoms on graphite. This mismatch ensures a finite amount of defects/vacancies in adsorbed solid helium even at zero temperature, i.e. so-called zero-point vacancies, which give their hallmark on the ground state properties, and which have not yet been ever found in the bulk 3D helium crystals. In the planar geometry of graphite, where carbon atoms are assembled into a plain hexagonal lattice, only the famous 1/3 solid phase (every third carbon hexagon is occupied by helium atom), is well established \([13, 14]\), although Greywall also reported a sign of commensurate dimer phases \([15]\). The dimer commensurate phases should be
even more stable on a curved nanotubes due to a larger distance between adsorption cites. The vacancies in such bosonic dimer crystal are also bosonic, and due to small mass and the associated strong zero-point motion, they delocalize at low enough temperature to procure a mobile, fluid-like solid phase as we demonstrate in this work. The relevance of this kind of topological frustration has recently been demonstrated also in nanomagnetism \cite{16}. Our defect-rich system differs distinctly from one-dimensional Bose-Einstein condensates in an optical lattice where a supersolid has recently been created using coupling to additional cavities \cite{17}.

At low helium coverages the periodic CNT lattice potential is not able to localize the adsorbed atoms, hence fluid and gas phases are formed. On graphite, a uniform fluid $^3$He phase has been observed to form at coverages $\rho < \rho_{LS} \approx 4.2 \text{ nm}^{-2}$ \cite{13, 18}. In 2012 Sato \emph{et al.} observed that at extremely small densities, $\rho < 0.8 \text{ nm}^{-2}$, the liquid forms clusters while the rest of substrate is covered with gas phase \cite{19}. As our present experimental work demonstrates, such coexisting phases of liquid clusters and a rarefied gas also appear in adsorbed $^3$He on CNT.

Extraordinary electrical and mechanical properties of suspended CNTs facilitate their use as ultra-sensitive detectors for force sensing \cite{20} and for surface physics \cite{21}. Helium atoms provide additional mass $\delta m$ and an additional spring constant $\delta k$ for the nanotube (due to change in tension \cite{22}). The relative frequency shift $\delta F/F = \delta k/2k - \delta m/2m$ of the mechanical resonance of the tube thus provides information of the involved energies and the nature of the phases formed by the adsorbed helium.

In our experiments, low-coverage $^3$He layer on CNT displayed a "clustered liquid + dilute gas" to "uniform fluid" transition at temperatures 0.1 ... 0.5 K. This transition manifested itself as an abrupt drop of the CNT resonant frequency $F_0(T)$ with increasing temperature $T$, due to redistribution of helium mass from the ends to uniform coverage over the tube (see Fig. 1A). At larger coverages helium is distributed uniformly, and observed jumps in $F_0(T)$ traces are identified as changes in a stiffness of the oscillator due to structural phase transitions, as illus-
Figure 1: **Phase transitions at different areal densities $\rho$.** (A) Resonant frequency $F_0$ vs. $T$ across the transition from "clustered liquid - gas" coexistence to "uniform fluid". $F_0$ drops owing to redistribution of $^3$He atoms from clusters near ends of the nanotube to a uniform layer. (B) Transition from normal solid to fluid. Resonant frequency is higher in the solid phase because localized $^3$He atoms increase the tension in the CNT (see text); the panel also illustrates the determination of the jump $\delta F_0$. (C) Transition from "gliding" solid to fluid. Absence of jump of $F_0$ across the transition indicates that $^3$He atoms are delocalized in this new solid phase and do not produce additional tension of the tube. (D) Transition from gliding to normal solid manifested by an abrupt increase of tension due to localization of atoms, which increases the tension of the tube. The second transition from normal solid to fluid is seen as a drop of the resonant frequency due to decrease of tension, as in (B). The linear $F_0(T)$ at the lowest $T$ (see also Fig. 4A) can be extrapolated to the fluid phase, with no jump, as in the panel (C).
trated in Fig. 1B–D. As seen in the $F_0(T)$ traces in Fig. 1, there are abrupt frequency changes by $\delta F_0$, either positive or negative, depending on the stiffness modification across the critical temperature $T_c$. Basic $^3$He phases on graphite are illustrated in the insets of Fig. 2 and in fig. S3.

The phase diagram, constructed from $F_0(T)$ sweeps, is displayed in Fig. 2. The transition temperature $T_c$ has a maximum around $\rho = 1 - 2$ nm$^{-2}$, above which it decreases very rapidly with increasing coverage. This can be explained by 2D van der Waals (vdW) approach for liquid-gas coexistence (see the supplementary materials, IV). A fit using the 2D vdW phase separation equation to the measured transition temperatures is illustrated by the parabola-looking curve in Fig. 2 at densities below 4.2 nm$^{-2}$. The calculated magnitude of the jump $\delta F_0$ (Eq. 4 in the supplementary materials) is in perfect agreement with our experiment, as shown in Fig. 3.

When the liquid state fully overtakes the gas phase at 4.2 nm$^{-2}$, the jump $\delta F_0$ approaches zero, but with further increase of coverage there emerges again a large frequency drop (Figs. 1B and 3). This frequency decrease indicates a transition from solid to fluid, as localized solid helium phase enhances tension in the tube (see the supplementary materials, V). We note that the origin of the increase of tension is due to localized $^3$He whose zero point motion in anharmonic adsorption potential leads to additional force between carbon atoms. The coverage 6.4 nm$^{-2}$ corresponds to the 1/3 commensurate phase on graphite (see the insert in Fig. 2). The most symmetric 1/3 phase evidently maximizes the additional tension and thereby the frequency jump $\delta F_0$, as seen in Fig. 3. The melting temperature of the 1/3 phase on CNT, $T_c \approx 0.1$ K in Fig. 2 is strongly suppressed in comparison with the $T_c \approx 3$ K on graphite [13]. This is a clear signature of larger vertical motion of $^3$He atoms on a CNT, which smears the carbon lattice potential.

The frequency change $\delta F_0$ across the melting temperature disappears at coverages $\rho \approx 6.7$ nm$^{-2}$ (Fig. 1C), which means that the additional tension due to the solid helium phase vanishes. We attribute such a ”soft” solid state to a mobile solid phase which is formed by a gas of
Figure 2: **Phase diagram of adsorbed $^3$He on a CNT.** The individual points on the phase diagram were obtained from $F_0(T)$ traces illustrated in Fig. 1. Circles mark transitions from the phase-separated "clustered liquid + gas" system to "uniform fluid"; squares are transitions from the mixture "1/3 solid plus liquid" to uniform fluid. Diamonds mark the transition from gliding solid to fluid (below 8.2 nm$^{-2}$) and to the high-$T$ solid. Solid squares denote the transition from solid to liquid at high coverages. Different colors represent different mechanical resonances. The solid curve depicts the fit of the liquid-gas phase separation curve using the van der Waals equation of state. The transition occurs at temperature $T_c(\rho)$ where the line $\rho = \text{const}(T)$ crosses the phase separation curve below critical temperature $T_{cr}$ as illustrated in the insert. Vertical arrows indicate the coverage of the principal commensurate solid phases of which 3/8 and 2/5 phases are composed from dimers (see fig. S3). The insets illustrate, from left to right, liquid+gas coexistence, 1/3, and 3/8 phases.
Figure 3: **Frequency drop across phase transitions.** Difference $\delta F_0$ between the resonant frequency in the low-temperature phases and in the high-temperature uniform fluid phase, determined as illustrated in Fig. 1B. Circles: transition from "clustered liquid + gas" two-phase system to a "uniform fluid". The difference $\delta F_0$ vs coverage dependence is described very well by Eq. 4 of the supplementary materials (solid curve) assuming that liquid clusters are concentrated symmetrically near ends of the tube. Squares: transition from solid to fluid. The jump is due to the loss of the additional tension provided by solid helium. The dashed lines indicates the concentration of the 1/3 commensurate phase. Diamonds: jump from the gliding solid to fluid phase ($\rho > 8.1 \text{ nm}^{-2}$ extrapolated through normal solid phase). Absence of the change of the resonant frequency indicates that there is no additional tension provided by gliding solid phase. Insert: $F_0(T \to 0)$ vs coverage. The line shows general softening trend due to increase in the mass. The bump at $4.2 < \rho < 6.4 \text{ nm}^{-2}$ is due to additional stiffness caused by solidification of helium. The abrupt softening of solid helium at $6.4 \text{ nm}^{-2}$ implies quantum phase transition from normal to soft liquid-like solid.
delocalized vacancies. This solid with mobile vacancies is expected to produce only small additional tension, like a liquid does, because in this phase helium atoms are delocalized, in contrast to regular solids. This interpretation is corroborated by the observation of two transitions at higher coverage $\gtrsim 8.4 \text{ nm}^{-2}$ shown in Fig. 1D: the solid-induced tension appears abruptly at $T \approx 0.1 \text{ K}$ ("gliding" solid to "high-$T$" solid transition), and then disappears again with melting of solid at $T = 0.13 \ldots 0.25 \text{ K}$ depending on the coverage. The frequency jump $\delta F_0$ between 1/3 phase and the gliding solid is observed down to the lowest temperatures, as shown in the insert in Fig. 3, which indicates a quantum phase transition in the adsorbed film as a function of coverage.

We interpret the gliding solid phase as a bosonic dimer solid with delocalized topologically stabilized vacancies which provides fast mass transport. According to Andreev [23], in a magnetically-disordered fermionic solid $^{3}\text{He}$ a vacancy, due to its magnetic moment, forms a strongly localized magnetic polaron. Indeed, the monomer fermionic 1/3 phase shows no signs of mobile vacancies through mismatch-induced defects. However, when the coverage increases further, bosonic dimer commensurate solids such as 2/5 [15] and the here-introduced 3/8 phases become possible (see fig. S3C and the insert in Fig. 2).

The temperature dependence of the resonant frequency $F_0(T)$ was also analysed to understand the behavior across the quantum phase transition as a function of $^{3}\text{He}$ coverage (see Fig. 4A). Change of $F_0$ reflects the change in helium pressure $P$ as explained in the supplementary materials, VI. The power-law fitting of the data, $F_0(T) - F_0(0) \propto T^\beta$ gave $\beta = 2.1 \pm 0.1$ in the normal solid below $7.3 \text{ nm}^{-2}$, corresponding to 1D phonons. In the gliding solid phase, we find the exponent $\beta = 1.00 \pm 0.05$ (fig. S5A). The observed linear $P(T)$ dependence implies ideal gas type of behavior with a fixed number of particles which can be related to delocalized zero-point vacancies [5]. Truly, the nanotube as a seamlessly rolled graphene sheet works as a template for a superlattice of $^{3}\text{He}$ dimers. However, helium superlattice with a clearly larger
Figure 4: Quantum phase transition between monomer 1/3 phase and gliding dimer solid. (A) Temperature dependence of resonant frequency $F_0$ at different coverages indicated by the values on the right within the frame; the numbers point to curves in the corresponding order from top to bottom. We find $F_0(T) - F_0(0) \propto T^\beta$ with $\beta \approx 2$ at coverages $< 7.0 \text{ nm}^{-2}$, which corresponds to a solid with 1D phonon excitations (see text). The quadratic behavior changes abruptly to linear at $\rho \approx 7.0 \text{ nm}^{-2}$, which persists till complete monolayer coverage. Linear temperature dependence can be obtained with constant number of weakly interacting excitations, such as vacancies induced by topological mismatch. The quadratic fit for the highest density 6.7 nm$^{-2}$ of normal solid, and the linear fit for the lowest density 7.3 nm$^{-2}$ of gliding solid are shown by black curves. (B) Illustration of high-coverage $^3$He superlattice on a CNT with mismatch-induced defects. The defects are produced when, for example, the dimerized 3/8 superlattice on graphene (see fig. S3B) is rolled around the horizontal axis to form a CNT. The red line marks the connection of top and bottom edges of the graphene “stencil”.


period cannot generally be matched with the CNT lattice, and the dimer lattice will contain plenty of defects (see Fig. 4B). The system of point defects is thus topologically protected due to the mismatch, and no new defects appear with increasing temperature as the corresponding activation energy is much higher.

The observed quantum state of weakly interacting delocalized vacancies is truly extraordinary because, in contrast to Cooper pairs stabilized by phonons, pairing of fermionic $^3$He atoms on CNT into bosonic dimers is promoted by carbon lattice. Owing to topological frustration, the dimer phase contains defects and acquires properties reminiscent of bosonic supersolid in its ground state. Equally intriguing would be to study the nuclear spin system of the dimer solid phase because this phase, combining the paired $^3$He and frustrated 2D lattice, would facilitate a combination of exotic magnetic phenomena of $p$-wave-paired superfluid and quantum spin liquid.

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SUPPLEMENTARY MATERIALS

Materials and Methods

Supplementary Text

Figs. S1 to S5

References (24–38)
Supplementary Materials for
Mobile solid \(^3\)He phase on carbon nanotube

I. Materials and methods

Fabrication of nanotube samples: The CNTs were synthesized in the gas phase with the floating catalyst chemical vapour deposition growth method (FC-CVD) followed by the direct thermophoretic deposition onto prefabricated chips \(^{24}\). For contact material we employed molybdenum-rhenium/palladium (MoRe/Pd) bilayer. The 150 nm MoRe layer was first co-sputtered on top of a strongly doped Si wafer with 285 nm thick SiO\(_2\) layer. In a second step a 5 nm Pd layer was sputtered on top of the existing MoRe. Optical lithography and reactive ion etching (RIE) were used to pattern 119 source drain electrode pairs per chip. Clean FC-CVD synthesized nanotubes with diameters of \(1.6 \pm 0.2 \text{ nm}\) were deposited on the clean chip. In the last fabrication step, the chip was annealed in vacuum at 220 °C for 10 min to increase contact transparency \(^{25}\). No superconductivity was observed in the measured samples. The Coulomb energy of CNTs amounted approximately to 8 meV. The doped substrate acted as back gate with capacitance \(C_g \simeq 0.4 - 0.5 \text{ aF}\) to the tube.

All source-drain pairs were checked regarding room temperature resistance of the CNT using an automatic probe station. Two devices with a room temperature resistance of \(\sim 100 \text{ k}\Omega\) were chosen and bonded to a PCB in a parallel configuration. Scanning electron microscope images confirmed \(L = 700 \text{ nm}\) for the length of the suspended part of one nanotube device after all measurements were completed, while the total length of the tubes varied around 2-3 \(\mu\text{m}\). The other device had detached before imaging, but according to nearly equal mechanical frequency, we estimate \(L \simeq 700 \text{ nm’s}\) for its suspended part as well.

Biasing: The bias point of the nanotube for detection of mechanical motion was deter-
mined by measuring its differential conductance $dI/dU_{bias}$ and the so-called transconductance $dI/dU_{gate}$. Both these values should be large enough to provide sufficient current for sensitive detection. In Fig. S1A we show the transconductance at different bias and gate voltages, while Fig. S1B displays DC I-V characteristics at different gate voltages. On the basis of these measurements, the working point $[U_{gate}, U_{bias}]$ was chosen to have large amplification for mechanical motion and low Joule heating.

**Preparation of $^3$He sample:** After electrical characterization of the CNT sample along with its mechanical resonance properties in vacuum, $^3$He atoms were gradually added to the sample chamber. As a single nanotube can adsorb only very little amount of $^3$He, we used a grafoil sample of surface area 17.6 m² as a ballast. Using grafoil ballast, we were able to control the coverage of the adsorbed $^3$He with an accuracy of 0.05 nm⁻²; the calibration of our coverage scale is discussed below.

After each addition we waited for 10-20 hours and subsequently performed temperature sweeps up and down at a rate of 10 ... 30 mK/h. Typically, the $T$ sweep at each coverage was repeated at least once, often several times. Most of the data were collected on two resonances with vacuum frequencies 325.35 and 348.55 MHz, see Fig. S2. Both lines also displayed quite similar coverages, which hints that these two signals must originate from very similar CNTs. The similarity of the two nanotubes also confirmed by equal resistance at room temperature and by nearly equal Coulomb energies (see Fig. S1A).

**Mechanical resonance detection:** When a RF voltage $\Delta U = U_{RF} \cos \Omega t$ is applied on top of the DC gate voltage $U_{gate}$, the tube starts to oscillate because of the oscillating electrical field $\Delta E$. The oscillating electrical force $F = Q \cdot \Delta E$ will produce the mechanical motion of the tube; here $Q = C_g(U_{gate} + \Delta U)$ denotes the total charge on the nanotube with capacitance $C_g$ to the gate. The equation of motion for the vertical position $z$ of a CNT with radius $r$ at height $H$
above the gate electrode becomes \( C_g U_{\text{gate}} U_{RF} \cos \Omega t / (r \log 2H/r) = M \partial^2 z / \partial t^2 + \gamma \partial z / \partial t + k z \), where \( M \) is the effective mass of the tube, \( k \) is a spring constant, and \( \gamma \) denotes dissipation. The conductivity \( \sigma(E) \) of the tube also oscillates at the same frequency, as it is field-dependent. As a result, the current through the tube \( I = \Delta U \sigma \propto \cos^2 \Omega t \) obtains an additional constant term which is proportional to the amplitude of mechanical oscillations,

\[
I_{DC} \propto \frac{C_g U_{\text{gate}}^2 U_{RF}^2 \sigma'_{E}}{M} \frac{1}{\Omega_0^2 - \Omega^2 + 2\pi i \Gamma \Omega},
\]

where \( \sigma'_{E} \) denotes the derivative of the conductance with respect to the electric field \( E \), \( \Omega_0 = 2\pi F_0 \), and \( \Gamma \) are the central frequency and the width of the mechanical resonance of the tube, respectively. In logarithmic approximation, the tube capacitance per unit length can be written as \( C_g / L = 2\pi \varepsilon_0 / \log (2H/r) \).

Most of our data was obtained using the so-called frequency modulation method [26]. When the driving RF frequency \( \Omega \) is modulated over \( \Omega \pm \delta \) at an audio frequency \( \omega \sim 1 \text{ kHz}, \Omega(t) = \Omega + \delta \cos \omega t \), the nanotube will produce a mixing current component at frequency \( \omega \) proportional to the mechanical response:

\[
\langle I \rangle \propto \frac{d}{d\Omega} \frac{1}{\Omega_0^2 - \Omega^2 + 2\pi i \Gamma \Omega}.
\]

This audio frequency signal can be recorded accurately using a sensitive low-frequency lock-in amplifier. Originally, after adjusting the detection phase, only the real part of the differentiated Lorentzian in Eq. (2) was required in the analysis [26]. In the general case, however, there appears a weakly temperature-dependent phase shift between the voltage and the conductance that needs to be accounted for. Therefore, the phase of the Lorentzian becomes a fitting parameter, and the real and imaginary parts need to be considered when analysing the measured response curves, see examples of the fit in Fig. S2.

\(^1\sigma'_{E} \) can also be written as \( d^2 I / dU_{\text{bias}} dU_{\text{gate}} \).
Coverage scale of adsorbed $^3$He atoms: Owing to substantial tension $F_0$, the mechanical resonant frequency of our samples can be approximated by $F_0 = (1/2L)\sqrt{F_0/\mu}$, where $\mu$ denotes the mass per unit length. The coverage $\rho$ was calculated from the frequency shift in the uniform fluid phase where helium is distributed uniformly over the tube and does not contribute appreciably to the tension of the tube:

$$\frac{\Delta F}{F_0} = -\frac{M_{He}}{2M_C} = -\frac{\rho_{He}}{8\rho_C}$$  \hspace{1cm} (3)

where $M_{He}$ is the total mass of the adsorbed $^3$He, $M_C$ and $\rho_C = 38.2$ nm$^{-2}$ denote the mass and the areal density of carbon atoms in the nanotube, respectively, and $F_0$ is the resonant frequency of the nanotube in vacuum. This yields the relationship $\rho_{He} = -305.4$ nm$^{-2}\Delta F/F_0$ for the helium coverage. For calculations of frequency shifts with non-uniform mass distribution $\mu(x)$ along the nanotube ($x$-axis), the mode displacement $z$ of the oscillating nanotube was approximated with the cosine form $z = z_{max}(1 - \cos 2\pi x/L)/2$ which differs less than 10% from the exact oscillating shape [27]. The modification of the kinetic energy due to helium mass was calculated by integrating $\frac{1}{2}\mu(x)\dot{z}(x)^2$ over the nanotube.

Even though the frequency of the fitted peaks can be determined with very high accuracy, our coverage scale has an uncertainty due to frequency shifts with temperature. This shift influenced in particular the measurements of ”clustered liquid + gas” to “uniform liquid” transition temperature. The coverage was derived from the frequency shift determined at 0.25 K according to Eq. (3). The errors come from the fact that 0.25 K is rather close to ”liquid + gas” to ”uniform fluid” transition temperature (see Fig. 1A of the Main text), and the data were extrapolated from higher temperatures. Small shifts due to the Coulomb spring effects and background charge variation [28][29][30] were eliminated by tuning the gate and bias voltages of the working point.

Temperature: Noise thermometry was employed for measurement of temperature over the
whole experimental range. The noise thermometer was manufactured by Physical-Technical Institute (Physikalisch-Technische Bundesanstalt) in Berlin [31].

II: Electrical conductivity of the nanotube (Fig. S1)

Owing to the Coulomb gap, the conductance $\sigma$ of the nanotube varies over several orders of magnitude depending on the applied gate voltage $U_{\text{gate}}$ and on the bias voltage $U_{\text{bias}}$. A useful quantity of the differential conductance is the so-called transconductance $dI/dU_{\text{gate}}$ which can be measured by applying a small AC voltage to the gate. The working point in our measurements $[U_{\text{gate}}, U_{\text{bias}}]$ was located near one of the largest and widest peaks seen in Fig. S1A. The bias voltage $U_{\text{bias}}$ should be kept small enough to avoid unnecessary Joule heating $IU_{\text{bias}}$ (see Fig. S1B for typical IV-curves). However, in our sample the heating was primarily deposited on the contacts on electrodes, owing to the ballistic nature of the conductance of the tubes. The working point was optimized by measuring $dI/dU_{\text{gate}}(U_{\text{gate}}, U_{\text{bias}})$ and $I(U_{\text{gate}}, U_{\text{bias}})$ manifolds and readjusting the values in order to compensate for the drift due to background charge variation.

III: Quantum diffusion of the adsorbed helium at low densities

$^3$He atoms on the nanotube lattice are attracted to the centers of carbon hexagons. The energy barrier separating two neighboring hexagon centers located at the distance of $d = 2.46$ Å (equilibrium distance in the planar graphite/grafito geometry) is quite large, $U = 2.9$ meV = 33.6 K [14]. The hopping time $t_h$ to the neighboring hexagon is $t_h \sim 1/(fP)$ where $P$ is the probability of tunneling through the barrier, and $f$ is the attempt frequency. At temperatures as low as 0.1 K, the probability of thermally activated tunneling $P_{\text{th}} \sim \exp[-U/(k_BT)]$ is negligible, and hopping is governed by the quantum tunneling which probability can be estimated in the
quasi-classical approximation as $P_q \sim \exp \left( -\sqrt{2m(U - E)}d/\hbar \right)$ where $E$ is the lowest energy level in the potential well [32].

The shape of the potential depends on the curvature of the carbon lattice. By neglecting the curvature and taking the first term of the Fourier expansion $U(x) = (U/2)(1 - \cos 2\pi x/d)$ for the shape of the potential, we obtain $U(d) \approx (\pi^2/d^2)Ux^2$ for its harmonic approximation. This yields for the oscillation frequency $f = \sqrt{U/(2md^2)} \sim 10^{12} \text{ Hz}$ and for the lowest energy level $E = \pi hf \approx 20 \text{ K}$. Consequently, the width of the barrier decreases to $d^* \approx 1.2 \text{ Å}$, and the quantum tunneling probability becomes as large as $P_q \sim 0.2$. The corresponding diffusion coefficient in the limit of small density can be written as $D \sim h^2 f P \sim 10^{-8} \text{ m}^2/\text{s}$. Hence, we obtain a characteristic time scale $\tau \sim (1/2)L^2/D \sim 10 \text{ µs}$ that governs the diffusion of a helium atom across the tube length of $L = 700 \text{ nm}$. Similar estimate can be made using the band model for helium atoms on graphite [33]. Using the group velocity $v_g = \hbar k/m_3$ for $k = 2\pi/L$, we obtain $t = v_g/L \approx 5 \text{ µs}$ for the traversal time. Thus, at small densities, the atoms on the tube can arrange themselves to an equilibrium configuration on time scales much less than a few minutes corresponding to the duration of our single resonance curve measurement. On the other hand, the diffusion time is much longer than the mechanical oscillation period, which prevents mass redistribution within one oscillation cycle.

IV: 2D van der Waals approach at low densities

A pressure $P$ in two dimensions is introduced as the force per unit length of the boundary, arising from the kinetic energy of the particles confined to in-plane motion on the CNT. The equation for ideal gas is identical to the 3D formula, $P = m(N/S) \left( \bar{v}_x^2 + \bar{v}_y^2 \right) = (N_a/S_m)k_B T = RT/S_m$, where the surface area $S_m$ of one mole plays the role of the molar volume in 3D, and $\bar{v}_i^2$ describes the average of square velocity of particles in the direction $i = \{x, y\}$. The vdW correction $b$ to the molar surface area $S_m$ is a constant equal to the total area occupied
by the hard cores of one mole of particles. Particles feel an attraction to the rest of the particles, which is proportional to the coverage $\rho \propto 1/S_m$. This leads to a change in the pressure at the boundary, but since the deficit of pressure is proportional to the number of particles, the correction is quadratic in coverage, $P \rightarrow P + a/S_m^2$. The functional form of the vdW equation is therefore the same as in 3D, but with the replacement of molar volume with molar surface area, $(P + a/S_m^2)(S_m - b) = RT$. The reduced form reads then universally, $[P/P_{cr} + 3(\rho/\rho_{cr})^2] \cdot (3\rho_{cr}/\rho - 1) = 8(T/T_{cr})$, where $T_{cr}$, $\rho_{cr}$, $P_{cr}$ are the critical values for temperature, density and pressure.

The above vdW form can also be applied to 2D $^3$He, a fermionic quantum system. The ideal gas law for fermions contains additional $\rho^2$ terms, while the exchange interaction yields an additional increase in pressure. However, both modifications can be included in the parameters of the vdW equation. A fit of the 2D quantum vdW equation to the transition temperature with $T_{cr} = 0.52$ K and $3\rho_{cr} = 4.2$ nm$^{-2}$ is shown in Fig. 2 of the Main text.

The validity of the vdW approach was also confirmed by calculating the frequency drop at the phase separation curve. We assume that liquid helium clusters start to form near the ends of the tube where the atomic binding energy is larger due to a surface roughness at the ends of nanotube [19]. Redistribution of $^3$He atoms from liquid puddles near the ends to a uniform fluid layer over the tube at the phase transition changes the resonant frequency according to

$$\frac{\delta F}{F_0} = \left[ -\frac{1}{6\pi} \sin \pi \frac{\rho}{\rho_0} + \frac{1}{48\pi} \sin 2\pi \frac{\rho}{\rho_0} \right] \frac{\rho_0}{\rho_C},$$

where $\rho_0 = 3\rho_{cr}$, and $\rho_C = 38.2$ nm$^{-2}$ is the areal density of carbon atoms. The frequency jump calculated from Eq. 4 plotted in Fig. 3 of the Main text as a solid curve, fits the experimental data points very well. Note that there is no fitting parameter in Eq. 4, once the critical coverage $\rho_{cr} = 1.43$ nm$^{-2}$ is determined from the phase diagram (Fig. 2 of the main text).

The largest liquid–gas co-existence coverage 4.2 nm$^{-2}$ is five times larger than the corre-
sponding coverage in $^3\text{He}$ on graphite, 0.8 nm$^{-2}$ [19]. This difference can be attributed to transverse, vertical motion of $^3\text{He}$ on the nanotube. In the variational Monte-Carlo study on $^3\text{He}$ monolayer on graphite there is no self-binding for a strict 2D system, but accounting for the vertical motion of atoms shows that $^3\text{He}$ film forms a clustered liquid with critical coverage of $\approx 2$ nm$^{-2}$ [34]. Although this effect is small on graphite [35], it is expected to be larger in our case because the number of carbon atoms binding $^3\text{He}$ is smaller on a CNT. Hence, the observed quantum vdW equation of state on a CNT is influenced also by quantum mechanical motion in the third, radial direction.

V: Commensurate solid phases of helium on a hexagonal carbon lattice (Fig. S3)

As discussed in the Main text, stability of commensurate phases of $^3\text{He}$ on graphite lattice is influenced by the curvature of the lattice in a CNT. Here we discuss briefly how the orientation of the $^3\text{He}$ structure on the curved CNT influences its stability. Basic structures of $^3\text{He}$ on a graphite lattice are illustrated in Fig. S3. For easier comparison of the structures, all frames are drawn with the same arm-chair type of top and bottom edges.

Among the possible commensurate solid helium phases on the carbon hexagonal lattice, the most symmetric and stable is the monomer 1/3 phase in which helium atoms occupy every third hexagon. The 1/3 phase has been experimentally observed on graphite by Bretz et al. at the coverage of 6.4 nm$^{-2}$ with both $^3\text{He}$ and $^4\text{He}$ [13]. At lower coverages, a mixture of 1/3 solid and fluid phases has been observed [18]. On a CNT, the true hexagonal symmetry of the 1/3 phase is broken as the equilibrium distance between helium atoms becomes direction dependent. This influences the structures in equilibrium, and it may lead to modified attributes for the 1/3 phase on a CNT, for example, reduced stability against other possible configurations.

At higher coverages, other commensurate phase structures are possible including ones composed of dimers, as illustrated in Figs. S3B and S3C, or even chains, as in Figs. S3D and S3E.
However, in the planar geometry of graphite, these phases are most likely not stable because the hard core diameter of helium atom is larger than the distance between centers of neighboring hexagons as discussed in the Main text. Nevertheless, even in the planar geometry of graphite, Greywall seemed to observe an onset of specific features at the corresponding coverages and suggested that there could exist 2/5 or 3/7 phases [15] illustrated in Figs. S3C and S3D.

In contrast to the planar geometry of graphite, CNT can facilitate additional commensurate phases: the distance between helium atoms is now larger and exceeds the hard core diameter when the orientation of dimers is inclined with respect to the axis of the tube. The dimer crystalline phases illustrated in Figs. S3B and S3C become then possible. Thus, we may conclude that CNT, due to its curvature, supports stable solid commensurate dimer phases in which the orientation of dimers is perpendicular or inclined with respect to the tube’s axis. For example, when the structures shown in Figs. S3B and S3C are wound into a CNT around their horizontal axes, as depicted in Fig. 4B in the Main text.

VI: Tension induced by adsorbed helium (Figs. S4, S5)

Similar to what has been observed with other substrates and adsorbents [22, 37], $^3$He atoms adsorbed on CNT contribute both to the mass and to the spring constant of the tube, thereby changing the resonant frequency. The actual frequency vs. mass relation depends on the location of the mass on the oscillation tube. For example, if the mass is located at the ends of the tube, the influence of the mass is small compared with the same mass located near the center. Hence, a transition between islanded film (liquid clusters near the ends of the tube) and a uniformly distributed fluid becomes quite prominent.

In constructing our coverage scale in Sect. I, we assumed that the change in the elastic state of the tube can be neglected in fluid phase at high temperatures, and that the frequency shift is
governed by the mass of helium atoms $M_{He}$ added to the mass of the carbon lattice $M_C$, see Eq. (3). At high $T$ the distance between helium atoms and the substrate is the largest, which minimizes the influence of helium atoms on the elastic properties of the CNT. Upon localization of the atoms in solid phases, the free surface energy becomes lowered, which leads to a change in the spring constant of the tube as observed in the experiments as a frequency shift.

At larger coverages $\rho > 4.2 \text{ nm}^{-2}$, the transition from uniform fluid to solid is not associated with a change in the effective mass on the tube but only with a change in the effective spring constant of the mechanical resonator. In fact, our coverage scale is corroborated by the experimental findings at 1/3 filling at $\rho = 6.4 \text{ nm}^{-2}$ coverage, at which the "$\sqrt{3} \times \sqrt{3}$" commensurate phase induces the strongest observed frequency jump between the liquid and solid phases.

The contribution of helium to the nanotube spring constant can be viewed as a change in the tension $F$, which relates directly to the mechanical resonant frequency, $F_0 = (1/2L) \sqrt{F_0/\mu} \sim 330 \text{ MHz}$; here we have used the length $L = 700 \text{ nm}$ measured with SEM after the experiment, $\mu \simeq 3.8 \cdot 10^{-15} \text{ kg/m}$ for the mass per unit length of the tube with diameter $d = 1.6 \text{ nm}$, and $F_0 \simeq 0.80 \text{ nN}$. In the solid phase, helium atoms are preferably localized above the centers of carbon hexagons, see Fig. S3. Upon localization of the atoms, there occurs a change in the surface energy which enhances the tension $F$ of CNT. The magnitude of this additional tension can be estimated approximatively using the total energy density of adsorption, $\delta F \sim \pi d \mathcal{E} \rho \simeq 60 \text{ pN}$ where we have taken coverage $1/3$ ($\rho = 6.4 \text{ nm}^{-2}$), and the adsorption energy $\mathcal{E} = -E_{1,0} = 137 \text{ K per atom}$ [14].

One can obtain a more accurate estimate for the helium-induced tension from the well-known adsorption potential for helium atom on graphite $U(z)$ [14]. Owing to the zero point motion, and due to the asymmetry of the adsorption potential, the equilibrium location of the He atom is not at the potential minimum, but at an average height about $0.2 \text{ Å}$ further from the
carbon lattice [14, 38], as it is seen in the Fig. S4A. Consequently, there is an attracting force between He and C atoms, which is balanced by the force coming from the zero point motion. This force results in a change of the unit cell size and inducing an extra tension in the lattice.

The magnitude of the force can be estimated by differentiating the potential \( U(z) \): \( f_\parallel(z) = \frac{1}{6} \frac{dU}{dz}(z) \cdot \tan \phi = \frac{1}{6} \frac{dU}{dz}(z) \cdot \left( \frac{a}{z} \right) \) where \( f_\parallel \) is the longitudinal component of the (attracting) force acting from helium atom at the average height \( z \) on a single neighboring carbon atom, see Fig. S4. With the interatomic distance in carbon lattice \( a = 1.42 \) Å and average distance from nanotube to helium atom \( z = 2.88 \) Å we find \( f_\parallel = 1.0 \) pN. Now, assuming again the 1/3 phase, we calculate the additional net tension over the cross-section of the tube \( \delta F = F_\parallel = \frac{\pi d}{3\sqrt{3}a/2}2f_\parallel = 28 \) pN (see Fig. S4), in a close agreement with the crude evaluation made in the previous paragraph. The estimated additional tension gives about 2 % for the relative change of the resonant frequency. This change is close to what we observe in the frequency across the quantum phase transition (QPT) from 1/3 phase to gliding solid state as seen in Fig. 3 of the Main text.

We note that both estimations set an upper limit for the helium-induced tension: the energy density approach disregards the mutual disposition of helium and carbon atoms, while the gradient method takes into account only nearest neighbors. With these reservations, we assert that the estimations are well in accordance with our experimental results displayed in Fig. 3 of the Main text. The additional tension is strongest in the most symmetric 1/3 phase, while it is much weaker in the fluid and in the "gliding solid" states in which helium atoms are delocalized.

We emphasize that the additional tension due to adsorbed helium atoms is inherently a quantum effect. A classical particle would remain at the minimum of the adsorption potential and would not exert a perpendicular force on the adsorbing atoms. A quantum particle, however, cannot be localized, and its average position in an asymmetric potential will be shifted away from the minimum due to zero-point oscillation.
Unlike normal solid phase, uniform fluid and gliding solid states do not produce additional net force on individual carbon atoms, as helium atoms are mobile in these phases and, on average, distributed uniformly. However, all phases possess internal pressure which also increases the tension $F$ of the nanotube. To estimate this contribution, we assume that the characteristic pressure in the 2D solid helium is of the order of bulk $^3\text{He}$ solidification pressure, 34 bar, multiplied by the thickness of helium layer, $c \sim 3$ Å, $P_{2D} = P_{\text{sol}}c \sim 0.001$ N/m. For a CNT with diameter of $d = 1.6$ nm, this contributes $\delta F = \pi d P_{2D} = 5$ pN to the tension of the tube. By comparing the additional tension $\delta F$ with the tension $F_0 \simeq 0.8$ nN of the bare tube, we find that the pressure of adsorbed helium layer can give up to 1 MHz increase of the resonant frequency.

The additional tension due to helium pressure depends both on temperature (due to thermal expansion) and on helium density of the adsorbed solid helium (due to compressibility). Indeed, in solid phases at $\rho > 4.1$ nm$^{-2}$ at $T < 0.1$ K we have observed intriguing dependence of the resonant frequency on temperature, which includes both linear and quadratic behavior (Fig. 4A of the Main text and Fig. S5A).

Our nanotube is operating mechanically in the regime where the gate voltage dominates the tension in the tube and the frequency increases with $V_g$. In this regime, the resonant frequency $F_0 = (1/2L)\sqrt{F/\mu}$ of the tube will then be changed according to

$$\frac{\delta F_0}{F_0} = \frac{1}{2} \frac{\delta F}{F}.$$  

(5)

The two most important contributions from the adsorbed helium to $F_0(F)$ of the oscillator are: the additional force $F_{||}$ in the commensurate normal solid described above, and the helium pressure $P$ which contributes directly to the tension,

$$\delta F(T) = F_{||} + 2\pi P(T).$$  

(6)

The first term is temperature-independent at low $T$, and it provides the large shift of the central frequency $F_0$ in the commensurate monomer $1/3$ phase, which disappears at the transition to
the liquid (with the increase of temperature) or to the gliding dimer solid (with the increase of density). The second term is present in all phases, and it can be estimated as proportional to $T^2$ in normal solid due to one-dimensional phonons, as we have indeed observed in the experiment (see Fig. 4A of the Main text). Transverse phonons with characteristic energies of $\hbar c/d \sim 1 \text{ K}$ are not excited at our temperatures; $c \simeq 170 \text{ m/s}$ denotes the estimated speed of sound [13].

At coverages $\rho > 7.0 \text{ nm}^{-2}$ the quadratic behavior of the resonant frequency $F_0$ as a function of temperature abruptly changes to a linear one (Fig. 4A of the Main text and Fig. S5A) pointing to a quantum phase transition to another solid phase where the excitation spectrum is quite different. The pressure $P$ which is proportional to $\delta F_0$ is now linear in temperature, corresponding to a phase with constant specific heat. One such phase could be the supersolid state, while another possible solid state with $P \propto T$ stems from the proposal by Andreev and Lifshitz on a delocalized system of weakly interacting vacancies which have an excitation spectrum $\varepsilon = \varepsilon_0 + \hbar^2 k^2 / 2m^*$ ($\varepsilon_0$ is the activation energy of vacancy) [5]. In solid $^3\text{He}$ on CNT the vacancies are topologically stable due to the mismatch of solid helium superlattice, and as such, they are attributes of the ground state and have $\varepsilon_0 = 0$. In this case, they would have exactly the same properties as an ideal gas with constant number of particles, having temperature-independent heat capacity.
FIG. S1: **Electrical characteristics of the nanotube sample.** Most of the data was measured on a parallel combination of two chips with preselected tubes: (A) Decimal logarithm of the transconductance \(dI/dU_{\text{gate}}\) in Siemens as a function of the bias and gate voltages. The twin structure of the transconductance peaks reflects the fact that there are two parallel tubes with slightly different Coulomb energies. One example of working points \([U_{\text{gate}}, U_{\text{bias}}]\) is marked by a red circle. (B) DC I-V curves at two different values of the gate voltage.
FIG. S2: **Mechanical resonance spectra.** The two mechanical resonances on which most of the data were obtained. Thin curves are fits using the differentiated Lorentzian form of Eq. [2] while the fits illustrated by the thick lines include also dephasing fluctuations. For determination of the resonance frequencies in this work, the former method without dephasing was employed. The data was measured on $^3$He coverage $\rho = 1.15 \text{ nm}^{-2}$ at the base temperature 20 mK of the dilution refrigerator.
FIG. S3: Basic commensurate solid phases. Patterns of helium atoms adsorbed on a plane hexagonal carbon lattice. (A) Monomer 1/3 phase observed on graphite [13]. (B) The introduced here most symmetric 3/8 dimer phase. (C) 2/5 dimer phase proposed by Greywall [15]; both 3/8 and 2/5 phases, together with topologically-induced defects, may form the backbone of the gliding solid. (D) 3/7 phase suggested by Greywall [15]. (E) Striped 1/2 phase. In the planar geometry this is the maximum density of helium on the first layer, above which the second layer starts to form.
FIG. S4: **Tension of the nanotube due to adsorbed helium atoms.** (A) Adsorption potential \( U(z) \) (thick curve) \(^{14}\), the ground state, and the wave function profile along \( z \)-axis. Projection of attractive force \( f_z \) acting from single carbon atom of CNT to an adsorbed helium atom is calculated as \((1/6)\) of the derivative \(-dU/dz\) of the adsorption potential. (B) Longitudinal component of the force \( f_{||} = f \sin \phi = -(1/6) dU/dz \tan \phi \). (C) Setting of the calculation of the additional tension due to the adsorbed helium in the 1/3 phase. The net force acting on the cross-section of the tube is \((\pi d/l) \cdot 2f_{||} = 4\pi df_{||}/(3\sqrt{3}a)\) \((d \text{ is the diameter of the tube, } l \text{ is the periodicity of the 1/3 superlattice marked in the picture})\).
FIG. S5: **Power law fits across quantum phase transition.** (A) Exponent $\beta$ of the power law fit $F_0(T) = F_0(0) + a_\beta T^\beta$ of the data shown in Fig. 4A of the Main text. (B) Prefactor $a_\beta$ of the fit $F_0(T) = F_0(0) + a_\beta T^\beta$ with exponents distributed around $\beta = 2.1 \pm 0.1$ for normal solid at $\rho < 7.0$ nm$^{-2}$, and with $\beta = 1.00 \pm 0.05$ for gliding solid state at $\rho > 7.0$ nm$^{-2}$.

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