Supplementary Material for

Phase Diagram of the Topological Superfluid \(^{3}\)He Confined in a Nanoscale Slab Geometry

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Phase diagram of topological superfluids confined in a nano-scale slab geometry

Materials and Methods

1 Experimental Set-up

A schematic diagram of the experimental arrangement is shown in Fig. S1. The cell is supported on a structure fabricated from the machineable ceramic, Macor. The NMR transmitter saddle coil is supported on a Macor former. The NMR static field coil is located in the helium bath; measurements were performed in a magnetic field of 32 mT. The SQUID NMR spectrometer developed for this work has been described elsewhere (20).

The silver cold-plate is attached to the top plate of a copper nuclear adiabatic stage through a silver rod. This allows the temperature of the silver plate to be conveniently swept using the heater shown. The cell construction has been described in detail elsewhere (33). The cavity is created by oxidisation, patterning and standard optical lithography of a silicon wafer. The $^3$He confined in the cell was cooled through the column of $^3$He within the fill line, which leads to a sintered silver heat exchanger mounted on a silver plate attached to a copper nuclear adiabatic demagnetisation stage. This method exploits the high thermal conductivity of liquid $^3$He at low temperatures, and is analogous to cooling an electronic sample “through the leads”. The temperature of the silver plate, and hence the $^3$He sample, was measured by a platinum NMR thermometer. The bulk superfluid transition temperature $T_{c}^{\text{bulk}}$ was conveniently observed through the NMR response of a small volume of bulk liquid at the

![Fig. S1. Experimental set-up. The cell, sintered silver heat exchanger, Pt NMR thermometer and heater are mounted on the silver plate, thermally connected to the nuclear demagnetisation stage. Also shown are the receiver NMR coil, the transmitter NMR coil that slides over the cell, and the directions of fields $B_{\text{rx}}$ and $B_{\text{tx}}$, and of the static NMR field $B_0$.](image-url)
mouth of the fill line. The $^3$He pressure was regulated with a precision better than 1 mbar.

The nominal cavity height is determined by a profilometer scan before bonding. The silicon wafer is anodically bonded to Hoya SD-2 glass. For these experiments we chose a 10×7 mm unsupported cavity, with 3 mm thick walls to limit the pressure dependence of cavity height. The cavity height profile was determined at room temperature by optical interferometry using laser light (at wavelengths 665 nm and 532 nm) in a refinement of the method described in (33). Measurements of cavity height at low temperatures are required since this is the key control parameter. Measurements were performed at room temperature, 77 K and below 7 K, using a small cryostat with optical access. Spectral analysis was performed on a reflected collimated white light beam of diameter 0.3 mm. Results at room temperature are consistent with the single wavelength technique. We observe some bowing of the cavity, down to 20 K, arising from differential thermal contraction between the glass, silicon and epoxy used to attach the fill line. Cavity height profiles are shown in Fig. 1 of the paper. Measurements show that the pressure dependence of the cavity height is temperature-independent with a coefficient 28 nm/bar at the centre of the cavity.

2 Interpretation of NMR Spectra

We performed pulsed NMR in a static field $B_0 = 32$ mT normal to the plane of the slab. For $^3$He with gyromagnetic ratio $\gamma = -2\pi \times 32$ MHz/T this field corresponds to the Larmor frequency $f_L = \gamma B_0 / 2\pi = 1$ MHz. The NMR spectra were obtained by taking the Fourier transform of the free induction decay (FID), following a small-angle tipping pulse (in the range 2-5°). We represent these spectra by the amplitude of the Fourier transform. We multiplied the FID by the apodisation function $f(t) = \exp(-t/10\text{ ms})$ prior to taking the Fourier transform to improve sensitivity. This 'T$_2$' filter was not applied in the initial imaging experiments, described in section 2.1.

2.1 Identification of Cavity and Bulk Signals by NMR Imaging in the Normal State

We can unambiguously identify the NMR signal from the $^3$He film confined within the cavity. For $^3$He in the normal state the NMR spectra have a double-peak structure shown in Fig. S2. In the normal state the resonance occurs at the Larmor frequency, $f_L = \gamma B_0 / 2\pi$, and this line shape is a result of inhomogeneity of the static magnetic field $B_0$ caused by magnetism of materials used in the experimental setup, for example the epoxy used to glue in the fill line. We perform simple one-dimensional NMR imaging by applying static field gradients, either perpendicular or parallel to the cavity surface normal, which add position-dependent shifts to the Larmor frequency, and observing shifts and broadening of the peaks in the NMR spectra. This allows us to clearly attribute the origin

Fig. S2. Identification of cavity and bulk signals in the normal state using a 1D imaging technique. NMR spectrum at 10 mK, 0 bar shows a double-peak structure. By performing NMR in a field with externally applied gradients $\partial B_z / \partial x'$ (the direction $x'$ of the gradient lies in the plane of the slab turned about 30° away from $x$, defined in Fig. 1) and $\partial B_z / \partial z$ (the gradient normal to the slab) we infer that the left hand peak originates from the slab of confined helium and the right hand peak is due to a volume of bulk liquid at the mouth of the fill line, shown red in Fig. 1 of the paper.
of the resolved peaks to the cavity and to the small volume of bulk liquid around the mouth of the fill line. The cavity signal is sensitive to the in-plane gradients, but remains unchanged in gradients along the surface normal, since the height of the cavity is so small. The bulk signal is shifted by the in-plane gradients as expected. The application of a gradient along the surface normal, $\partial B_z/\partial z = 4 \text{ mT/m}$, largely cancels the field inhomogeneity present in the environment and thus sharpens the bulk peak. This gradient is applied throughout the measurements.

2.2 NMR Spectroscopy of Superfluid $^3\text{He}$

Nuclear magnetic resonance continues to be applied as a probe of unconventional superconductors (34). There are however important differences between its application in these systems and in superfluid $^3\text{He}$, which we briefly outline here.

In a normal metal the nuclei are sensitive to the conduction electrons via the Knight shift and the spin-lattice relaxation time, arising from coupling between the nuclei and electrons via the hyperfine interaction. In a superconducting metal the electrons form Cooper pairs, and the magnetic susceptibility of these superconducting electrons is sensitive to the pairing state and nodal structure of the gap. This susceptibility is not directly measurable because of the Meissner effect. Nevertheless the Knight shift determined by NMR can probe the pairing state, through its dependence on the local spin susceptibility. The pairing state, and gap symmetry also determines the quasiparticle spectrum and hence the temperature dependence of the spin-lattice relaxation time in the superconducting state. In type II superconductors the field distribution arising from a vortex lattice can also be measured by NMR spectroscopy.

By contrast in superfluid $^3\text{He}$ the pairs are formed from $^3\text{He}$ quasiparticles with spin $S = 1/2$ arising from the nuclear spin $S = 1/2$ of $^3\text{He}$ atoms. Therefore NMR constitutes a direct probe of the pairs, via their (nuclear) spin degrees of freedom (1,2). In normal liquid $^3\text{He}$ the NMR precession occurs at the Larmor frequency $f_L = \gamma B_0/2\pi$. The tiny dipole interactions between $^3\text{He}$ nuclei are responsible for the relaxation of this precession but have a negligible effect on its frequency. In the presence of the superfluid order, the coherent orbital motion of the paired atoms results in a dramatic amplification of the dipole interaction. The density of dipole energy $F_D \sim \lambda_0 N_F \Delta^2$, where $N_F$ is the energy density of states at the Fermi level, $\Delta$ is the superfluid energy gap, so $N_F \Delta^2$ represents the condensation energy per unit volume, and $\lambda_0 \sim 10^{-6}$ sets the relative scale of the dipole energy (2). Unlike the condensation energy the weak dipole energy is not invariant under the relative rotations of the spins of the Cooper pairs with respect to the orbital degrees of freedom of the order parameter, providing the spin-orbit coupling. The important consequence is that such rotations in the NMR experiment result in an additional dipole torque on the spins which shifts the frequency $f$ of precession of the magnetisation away from the Larmor frequency $f_L$,

$$f^2 - f_L^2 \sim \gamma^2 F_D / \chi,$$

where $\chi$ is the magnetic susceptibility. The sign and magnitude of the shift is specific to the particular superfluid phase, its orientation prior to the NMR pulse and the power of the pulse (tipping angle $\beta$), and in every case can be modelled by solving the Leggett equations (1,2). In particular for small $\beta$

$$f^2 - f_L^2 = \frac{\gamma^2}{4\pi^2 \chi} \left[ \frac{\partial^2 F_D}{\partial \theta_x^2} + \frac{\partial^2 F_D}{\partial \theta_y^2} \right],$$

where $\theta_x$ and $\theta_y$ describe infinitesimal rotations of the Cooper pair spins about axes $x$ and $y$, chosen to be the two principal axes of the $\partial^2 F_D / \partial \theta_x \partial \theta_y$ tensor, perpendicular to the static field $B_0(1)$. It is clear that a positive/negative shift is expected in a configuration that minimizes/maximizes the dipole energy.

In the main text of the paper and some sections of this supplementary material we discuss the shifts of the frequency itself $\Delta f = f - f_L$ rather than the frequency squared. Since the shifts are small compared to the Larmor frequency, there is a straightforward correspondence $2f_L \times \Delta f = f^2 - f_L^2$.
Predictions for the Confined Sample

The orientation of the A phase is usually parametrised with a pair of unit vectors \( \hat{I} \) and \( \hat{d} \) (2). The inhomogeneity of the planar distortion (the given configuration. (B dynamics in the B phase with a uniform planar distortion (the given text of the paper corresponds to \( \hat{I} = \hat{z} \) and \( \hat{d} = \hat{y} \). The directions of \( \hat{I} \) and \( \hat{d} \) are determined by competing effects of the magnetic field, boundaries and dipole energy.

In a magnetic field exceeding the characteristic dipolar field \( B_D \sim 5 \text{ mT} \), determined by a competition of the Zeeman and dipole energies, \( \hat{d} \) is locked perpendicular to the field, and the small tipping angle NMR frequency shift is determined (25) by the angle between the field \( B_0 \) and \( \hat{I} \):

\[
f^2_\perp - f^2_\parallel = \frac{\gamma^2 \lambda_D N_F}{5\pi^2 \chi} \Delta^2 \left[ 1 - 2(\hat{I} \cdot B_0)^2 \right],
\]

Here \( \Delta \) is the energy gap maximum in the momentum space (reached at \( \hat{p} \perp \hat{I} \)). \( \chi \) is the magnetic susceptibility of the normal state and equal-spin-paired A phase, \( B_0 \) is the unit vector in the direction of the field. The strong surface boundary condition orients \( \hat{I} \) perpendicular to the walls of the container, and in the geometry of our experiment \( (\hat{I} \cdot B_0)^2 = 1 \). This ‘dipole-unlocked’ orientation, \( \hat{I} \perp \hat{d} \), maximises the dipolar energy leading to a negative shift

\[
f^2_\perp - f^2_\parallel = -\frac{\gamma^2 \lambda_D N_F}{5\pi^2 \chi} \Delta^2.
\]

The quantitative interpretation of the A phase frequency shifts taking the spatial distribution of the gap \( \Delta(z) \) into account is discussed below in Section 5.

The effect of boundaries on \( \hat{I} \) only persists over distances of order dipolar length \( \xi_D \sim 10 \mu \text{m} \) (2). In bulk, and in the absence of other orienting effects such as flow, \( \hat{I} \) is oriented parallel to \( \hat{d} \) by the dipole energy, \( \hat{I} \cdot B_0 = 0 \), leading to a positive shift of the same magnitude as the one given above.

The spin-orbit orientation of the B phase is described by the rotation \( R \) that needs to be applied to the spins of the state \( \Delta(\hat{p}) = \Delta_\parallel(-\hat{p}_x + i\hat{p}_y)|\uparrow\uparrow\rangle + \Delta_\parallel|\downarrow\downarrow\rangle + \Delta_\perp|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \) to yield the given configuration. \( R \) is parametrised via rotation axis \( \hat{n} \) and angle \( \theta \). The existing model of spin dynamics in the B phase with a uniform planar distortion (28) qualitatively accounts for both positive (B+) and negative (B-) frequency shifts we observe in this phase.\(^1\) The positive shift

\[
f^2_{B+} - f^2_\perp = \frac{\gamma^2 \lambda_D N_F}{5\pi^2 \chi} (\Delta^2 - \Delta^2_\perp)
\]

arises from the orientation with \( \hat{n} \parallel \hat{z} \) and \( \theta = \arccos(-\Delta_\perp/4\Delta_\parallel) \), which in the \( \Delta_\perp = \Delta_\parallel \) limit corresponds to the orientation the bulk B phase is found in (1,2). The order parameter corresponding to this orientation is \( \Delta(\hat{p}) = \Delta_\parallel e^{-i\theta}(-\hat{p}_x + i\hat{p}_y)|\uparrow\uparrow\rangle + \Delta_\parallel|\downarrow\downarrow\rangle + \Delta_\perp|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \), slightly more complicated than the expression given in the main text of the paper which represents \( \theta = 0 \), but also composed of pairs with \( L_z = -S_z \). The negative shift

\[
f^2_{B-} - f^2_\perp = -\frac{\gamma^2 \lambda_D N_F}{5\pi^2 \chi} (\Delta^2 + 2\Delta^2_\perp).
\]

corresponds to the orientation with \( \hat{n} \perp \hat{z} \) and \( \theta = \pi \). The different directions of \( \hat{n} \) in the \( xy \) plane are degenerate, \( \hat{n} = \hat{x} \) yields \( \Delta(\hat{p}) = \Delta_\parallel(-\hat{p}_x + i\hat{p}_y)|\downarrow\downarrow\rangle + \Delta_\parallel|\uparrow\uparrow\rangle + \Delta_\perp|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \), presented in the main text of the paper. Unlike \( B_+ \), here the pairs have \( L_z = S_z \). The predicted inhomogeneity of the planar distortion (24,30) is expected to modify the above equations for \( f_{B+} \) and \( f_{B-} \). Further work, such as NMR with large tipping pulses, is required to observe this effect.

\(^1\)In the language of Ref. (28) \( B_+ \) and \( B_- \) correspond to the ‘BS’ and ‘HPD2’ modes of precession respectively, both in the relevant small tipping angle limit, \( \beta \to 0 \), see Eqs. 4.5 and 4.10 therein; the bulk B phase Leggett frequency in Eq. 4.4 is substituted with \( B^D_\parallel = 3\gamma^2 \lambda_D N_F \Delta^2_\parallel/\chi_B \). Notice that the authors of Ref. (28), who are concerned with the orientation with respect to magnetic field, use the notation \( \Delta_\parallel \) and \( \Delta_\perp \) in the opposite sense to us and other researchers working on \(^{3} \text{He} \) confined in slab geometry.
2.3 NMR Signatures of Superfluid Transition in Cavity and Bulk

In the external field of 32 mT the magnetic environment of the cell is temperature-independent below 20 mK, hence the frequencies of the slab and bulk lines are constant from 20 mK down to transitions $T_c^{slab}$ and $T_c^{bulk}$, where they develop shifts due to the onset of superfluidity, Fig. S3.

The bulk liquid near the mouth of the fill-line provides a convenient ‘marker’ for the bulk transition temperature, $T_b^{bulk}$. This marker, observed at each of the pressures studied, allows an accurate calibration of the thermometry. It is immediately apparent from these NMR signals that $T_c$ is suppressed in the cavity and that different superfluid states are stabilised in bulk and slab near $T_c$: the dipole-unlocked A phase with a negative shift in the cavity, and the B phase with a textural positive frequency shift in the bulk (2,25).

We infer the frequencies of the lines from the position of peaks in the modulus of the NMR spectrum. Below the transition the bulk peak reduces in size and within 0.1 mK below $T_b^{bulk}$ it disappears in the noise of the NMR spectrometer. This is attributed to fast NMR relaxation due to superfluid spin transport between the bulk volume and liquid inside the fill line not excited by the NMR pulse.

2.4 NMR Signatures of A-B Transition in Cavity

At pressures above 4 bar and low temperatures the sample is found in the B phase, where two possible NMR signatures are observed, $B_+$ and $B_-$, as shown in Fig. S4.

Each time the sample is cooled into the B phase either of the two lines or a combination of them is obtained stochastically. The transition between the A and B phases occurs over a finite temperature range where the phases coexist, as shown in Fig. S5. Hysteresis is observed between the cooling and warming behaviour.
Fig. S5. NMR spectra through the A-B transition at $P = 5.5$ bar with ‘diffuse’ walls. On cooling the data show the gradual disappearance of the A phase peak and the simultaneous growth of a positively shifted peak from the planar distorted B phase; here in the B$_+$ configuration. The reverse is observed on warming. The vertical grey lines show the Larmor frequency; spectra are offset vertically for clarity.

In order to track the amount of the A phase in the sample we define the A phase peak size as

$$\Sigma = \int_{f_{\min}}^{f_{\max}} |G(f)| \, df,$$

where $G(f)$ is the spectrum, and the integration window encompasses the A phase peak in the spectrum at all temperatures. This quantity, presented in Fig. 3F in the paper and Fig. S6, has a property of taking a constant value in the A phase and normal state, and dropping as soon as the B phase signatures appear in the spectrum, justifying its use as an indicator of the A-B transition. Depending on pressure and boundary conditions one of four typical situations (see Fig. S6) is observed and temperatures of

Fig. S6. Peak size $\Sigma$ of the A phase NMR line and evaluation of the A-B transition at a range of pressures with ‘diffuse’ walls. At 2.2 bar (A) no A-B transition is observed, $\Sigma$ is constant from the lowest temperatures up into the normal state, consistent with an equal spin pairing state. At 4.0 (C) and 5.5 bar (D) the transition is observed between $T_{AB}$ and $T'_{AB}$. Above $T_{AB}$ the cell is completely filled with the A phase, below $T'_{AB}$ there is no A phase. Hysteresis is observed between warming and cooling. At 4.0 bar $\Sigma$ follows a reproducible hysteresis loop, at 5.5 bar the growth of the A phase on warming is reproducible, but stochastic supercooling of the A phase is observed on cooling. At 3.0 bar (B) the A-B coexistence (grey) extends to the lowest temperatures investigated, so only $T_{AB(\text{warming})}$ and $T_{AB(\text{cooling})}$ are measured. Observations with ‘specular’ walls are qualitatively similar, except that reproducible behaviour on cool-downs extends up to 5.5 bar (see Fig. 3F in the paper) and the hysteresis is more pronounced.
up to four features of the A-B transition are identified. Determination of the A-B transition line on the phase diagram from these data is discussed in detail in Section 4.

Figure S7 shows the NMR spectra over the whole temperature range where superfluidity is observed at 2.2 and 5.5 bar. This figure contains a subset (about 4%) of the large dataset used to produce Fig. 2, which typically includes a spectrum every 0.01 mK.

**Fig. S7.** Galleries of NMR spectra at 2.2 and 5.5 bar over the temperature range from the lowest investigated temperature up into the normal state, illustrating the measurements compiled into Fig. 2. The vertical grey lines show the Larmor frequency in the slab; the bulk peak is observed close to \( T_\text{c} \) (see Fig. S3 for more detail); spectra are offset vertically for clarity. (A) Warm-up through the A phase at 2.2 bar. (B) Warm-up from the B\(^-\) state at 5.5 bar; at \( T = 0.6T_\text{c} \) a small fraction of the sample undergoes an orientational transition into B\(^+\) state, which can be traced together with B\(^-\) all the way up to \( T_\text{AB} = 0.7T_\text{c} \). This transition is a manifestation of the metastable nature of the B\(^-\) state, which has higher dipole energy than B\(^+\). (C) Cool-down into B\(^+\) state at 5.5 bar.

### 3 Tuning Surface Scattering from Diffuse towards Specular by \(^4\text{He}\) Preplating

#### 3.1 Background

At low temperatures a layer of solid helium forms on the walls of a container filled with liquid helium, due to the van der Waals attraction. Depending on both the pressure of the liquid and the surface of the container this layer can be one to several atoms thick. In the case of \(^3\text{He}\) this solid exhibits para- or ferromagnetism, with a magnetic susceptibility much larger than the liquid. Rapid exchange between the liquid and the solid layer results in a combined NMR line, and the frequency shifts due to superfluidity are difficult to interpret (35).

\(^4\text{He}\) atoms are more tightly bound to surfaces than \(^3\text{He}\), so if \(^4\text{He}\) is added to the system, it displaces \(^3\text{He}\) from the surface, and eventually the solid layer consists entirely of \(^4\text{He}\), eliminating the surface magnetism (36). The solubility of \(^4\text{He}\) in \(^3\text{He}\) is exponentially small below \( \sim 100 \) mK, and at low temperatures a further admission of \(^4\text{He}\) creates a liquid film between the solid \(^4\text{He}\) and the liquid \(^3\text{He}\). This liquid \(^4\text{He}\) layer develops superfluidity below a thickness-dependent transition temperature (37). Decorating the surface with \(^4\text{He}\) has a strong effect on the scattering of \(^3\text{He}\) quasiparticles from the surfaces (38), and there is evidence that a sufficiently thick \(^4\text{He}\) film produces close to specular scattering (35,39).
3.2 Preplating Procedure in This Experiment

The procedure to preplate the surfaces with $^4$He was as follows: the cell is loaded with a uniform $^3$He-$^4$He mixture at 1.5 K up to a pressure of 1 bar, then the $^4$He deposits on the surfaces as the mixture phase-separates on cooling the cryostat to several mK.

An ideal condition for NMR studies of superfluid $^3$He in a slab with diffuse scattering at the walls is when the $^4$He coverage is just enough to displace the $^3$He solid layer, leaving a solid $^4$He layer at the wall. In our experiments with ‘diffuse’ boundaries the mixture contained 0.5% $^4$He. No change to the sample magnetisation is observed between 4.2 and 1.6 mK, placing an upper bound of 2 $\mu$mol/m$^2$ on the density of solid $^3$He, an order of magnitude lower than the typical density of a solid helium layer on surfaces (37).

In order to enhance the specular scattering we need to stabilise a sufficiently thick superfluid $^4$He film at the surfaces of the cavity. The boundary condition we denote as ‘specular’ was obtained with a 2.2% $^4$He admixture. The measurements of $T_c$ suppression (Fig. 3E) and low temperature gap suppression (Fig. 4C) suggest that full specularity was not achieved. The specularity remained unchanged with the addition of up to five times higher $^4$He concentrations. This observation suggests a limit to the $^4$He film thickness obtainable on the cavity surface and is explained by the following model. A thick superfluid $^4$He film may flow away from the cell and heat exchanger along the surface of the fill line, driven by the thermo-mechanical effect (40). If the film thickness is such that it remains superfluid in the sections of the fill line above some characteristic temperature $T^* \sim 100$ mK, where the solubility of $^4$He in $^3$He becomes non-negligible, $^4$He dissolves into bulk $^3$He in the fill line. The depletion of $^4$He film in the cell continues until a film surface density is reached, such that the temperature $T_{KT}$ of the superfluid transition in the film is of order $T^*$. This condition sets an upper limit on the $^4$He film thickness on the cavity surface, which is reached for all of the experiments on ‘specular’ boundaries. This effect will be eliminated in future by the installation of a superleak-tight cold valve interrupting the cell fill line at $T < 20$ mK.

4 Phase Diagram of Superfluid $^3$He in a Slab: Discussion

4.1 Coherence Length, Reduced Thickness and Effective Confinement

Within the framework of Ginzburg-Landau theory (2), valid close to $T_c$, the properties of superfluid $^3$He confined to a cavity of height $D$ are completely determined by the reduced thickness $D/\xi(T,P)$.

In the literature a number of definitions of coherence length $\xi(T,P)$ are used, differing by a constant prefactor, the most common being the ‘transverse’ coherence length introduced in (12),

$$\xi_{\text{tr}}(T) = \left[\frac{7\zeta(3)}{20}\right]^{1/2}\frac{\xi_0}{1 - \frac{T}{T_c}}^{-1/2}.$$ 

Here $T_c$ is the bulk superfluid transition temperature, $\xi_0 = \hbar v_F/2\pi k_B T_c$ is the Cooper pair diameter and $\zeta$ is the Riemann zeta function, $\zeta(3) \approx 1.20$.

It is convenient to extend the definition of reduced thickness beyond the temperature range $T \rightarrow T_c$, as a simple measure of effective confinement at all temperatures. As discussed in the paper, we

![Fig. S8. Comparison of coherence length $\xi_\Delta$ used in this work with the quantity $\xi_{\text{tr}}$ defined within the Ginzburg-Landau theory](image-url)
introduce the coherence length, related within a numerical factor to that proposed in (41),

\[ \xi_\Delta(T) = \frac{\hbar v_F}{\Delta_B(T)\sqrt{10}}, \]

where \( \Delta_B(T) \) is the weak coupling B phase gap (42). This has the property that \( \xi_\Delta(T) \to \xi_{tr}(T) \) as \( T \to T_c \) and \( \xi_\Delta(0) = 1.13\xi_0 \). A comparison of these quantities is shown in the Fig. S8.

In the Ginzburg-Landau regime, phase transitions are predicted to occur at universal values of reduced thickness independent of \( D \) (43). The critical values obtained using Ginzburg-Landau parameters derived from weak-coupling theory are presented in Table S1. The paper cited above also discusses the shift in critical thickness due to strong-coupling effects.

<table>
<thead>
<tr>
<th>Phase Transition</th>
<th>Boundary</th>
<th>( D/\xi_{tr} )</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>normal to A</td>
<td>specular</td>
<td>0</td>
<td>(29)</td>
</tr>
<tr>
<td></td>
<td>diffuse</td>
<td>( \pi )</td>
<td>(29)</td>
</tr>
<tr>
<td>A to B</td>
<td>specular</td>
<td>( \pi \sqrt{6} \approx 7.7 )</td>
<td>(41)</td>
</tr>
<tr>
<td></td>
<td>diffuse</td>
<td>( \approx 7.1 )</td>
<td>(43)</td>
</tr>
</tbody>
</table>

Table S1. Critical reduced thickness of phase transitions in a slab in Ginzburg-Landau regime with weak coupling parameters.

In the present experiment the A-B transitions occur well outside the Ginzburg-Landau regime, due to the strong confinement. However the theoretical prediction for the A-B transition temperature under confinement from quasiclassical theory (24, 30, 41), is well represented by \( D/\xi_\Delta \approx \text{constant} \), particularly over the temperature range for which the transition is observed in the present work (see Fig. S9). Therefore we use \( D/\xi_\Delta \) as our measure of reduced thickness (or effective confinement) to correct the measured phase diagram for the effects of the measured cavity height distribution.

Fig. S9. Predicted A-B phase boundary, from quasiclassical theory in the weak coupling limit (30, 41), replotted as a function of the measure of effective confinement, \( D/\xi_\Delta \), used in this work. The plot shows the weak dependence on effective confinement for both specular and diffuse scattering, and agreement with the G-L calculations (Table S1) in the limit \( T \to T_c \). The superfluid transition temperature for diffuse walls (29) is also shown.

4.2 Determination of A-B Phase Boundary and Superfluid Transition Temperature

4.2.1 Correcting for Measured Cavity Height Distribution in This Experiment

At all sample pressures studied, the cavity height distribution has been determined (see Fig. 1C of the paper). The consequent variation in effective confinement across the cavity accounts for the finite temperature range over which the A-B phase transition occurs.

Fig. S10 shows that the thickness distribution can account well for the overall A-B transition width. Therefore we attribute \( T_{AB} \) features (Figs. 3F and S6), clearly identified at all pressures...
Fig. S10. Thickness distribution as origin of the A-B transition width. The left panel shows the A phase peak size $\Sigma$ observed on warming from the B phase at $P = 5.5$ bar with ‘diffuse’ boundaries (red solid line) compared with a fraction of the cell where $D/\xi_\Delta(T) < D_{\text{max}}/\xi_\Delta(T^\text{AB(warming)}) = 13.1$ (black dashed line). Both quantities mainly vary between $T^\text{AB(warming)}$ and $T^\text{AB(warming)}$ justifying assigning $T^\text{AB(warming/cooling)}$ to the maximum cavity height. Other panels show at a number of temperatures the regions of the cavity satisfying the condition $D/\xi_\Delta(T) < 13.1$, predicted to be occupied by the A phase, leaving the rest of the cavity (white) to the B phase.

where the A-B transition is observed, to the maximum height $D_{\text{max}}$ in the slab and use these values to infer the reduced critical thickness of the A-B transition on cooling and warming shown in Fig. 3 of the paper. The transition width observed is somewhat smaller than the prediction based on the thickness distribution, with a profile not exactly as predicted. This is attributed to the fact that different regions of the cavity contribute with different weight to the A phase signal size, due to the position-dependent sensitivity of the NMR receiver coil and spatial dependence of the static magnetic field. At 5.5 bar with ‘diffuse’ walls the A phase stochastically supercooled (see Fig. S6), in this case the highest temperature where the A phase peak size was observed to drop was identified with $T^\text{AB(cooling)}$.

On the other hand, at the superfluid transition a single feature is observed, the onset of a frequency shift of the slab NMR line. Therefore the reduced thickness of this transition is evaluated for $D = (D_{\text{min}} + D_{\text{max}})/2 \pm (D_{\text{max}} - D_{\text{min}})/2$.

In Fig. 3D,E of the paper the $T = 0$ line is plotted using $D_{\text{max}}$, representing the phase diagram in the thickest part of the slab. In these coordinates the phase boundaries are extrapolated along straight lines. For the ‘diffuse’ walls

$$\frac{1}{2}(D_{\text{min}} + D_{\text{max}})/\xi_\Delta(T^\text{slab}) = 3.74,$$

$$D_{\text{max}}/\xi_\Delta(T^\text{AB(cooling)}) = 9.20 + 0.76P/\text{bar},$$

$$D_{\text{max}}/\xi_\Delta(T^\text{AB(warming)}) = 9.00 + 0.76P/\text{bar}.$$  

For ‘specular’ walls

$$\frac{1}{2}(D_{\text{min}} + D_{\text{max}})/\xi_\Delta(T^\text{slab}) = 2.00,$$

$$D_{\text{max}}/\xi_\Delta(T^\text{AB(cooling)}) = 9.83 + 0.87P/\text{bar},$$

$$D_{\text{max}}/\xi_\Delta(T^\text{AB(warming)}) = 9.37 + 0.87P/\text{bar}.$$  

The same lines are mapped into the $P - T$ plane to provide the extrapolated phase boundaries in Fig. 3A,B, i.e. making the simplifying assumption that $D/\xi_\Delta(T^\text{AB})$ is a temperature-independent constant at each pressure. The use of the precise weak coupling theoretical A-B boundary [Fig. S9] would lead to an upward revision of the pressure at which $T^\text{AB} = 0$ of between 0.5 and 1 bar. The accuracy of the extrapolation of the A-B boundary is also constrained by the influence of strong coupling on the theoretical phase diagram given in Fig. S9, which is presently unknown.

Strong coupling effects appear in Ginzburg-Landau theory as deviations of the $\beta$-coefficients of the fourth order G-L free energy terms from their well-established weak coupling values (44). These effects are pressure-dependent. Qualitatively this is predicted to increase the critical values of reduced thickness (43); for a quantitative comparison the calculation in (43) needs to be repeated using the experimentally obtained $\beta$-coefficients (44).
\[ D / \xi_{\Delta} \] vs pressure is a good representation of influence on the phase diagram of strong-coupling effects. The clear discrepancy between the measured A-B phase boundary and the predictions of weak-coupling quasi-classical theory demonstrates that strong coupling effects are present at all pressures.

### 4.2.2 Measurements on \(^3\text{He}\) Confined Within Stacks

The first experiments on \(^3\text{He}\) confined between Mylar sheets, spaced by 4 µm, performed in Helsinki (25) were designed to provide a well-defined geometry to control the superfluid texture for NMR experiments; however, a small suppression of \(T_{AB}\) due to confinement was observed. In subsequent work at Cornell \(^3\text{He}\) was confined within a stack of typically 1000 Mylar sheets of nominal spacing 300 nm, only the A phase was observed; no A-B transition was observed at any pressure (21, 35). Similar NMR studies on a stack of \(^3\text{He}\) slabs each of nominal thickness 1.1 µm, were subsequently carried out at Osaka City University (45). In this case the distribution of thickness across the stack was measured, Fig. S11A, revealing the significant limitation of this approach to studies of confined helium. The raw continuous wave NMR spectrum showed three peaks, one of which was attributed to the signal from within the stack. Evidence for the A-B transition was reported, and is compared with our results in Fig. S11B. No coexistence of A and B phases was reported in this work, leading to a significant uncertainty in the reduced thickness of the transition. A decrease in the reduced thickness of the A-B transition with \(^4\text{He}\) preplating was also reported, in contradiction to our result and also to the theoretical prediction.

![Graph A](image1)

![Graph B](image2)

**Fig. S11.** (A) Broad distribution of thickness in a stack of slabs used at Osaka (45), compared to the maximum thickness variation observed in our experiment achieved at \(P = 5.5\) bar. (B) Reduced thickness at the A-B transition. The error bars on the Osaka data reflect the uncertainty in cavity thickness. Data from our measurements, at low pressures, with a linear extrapolation.

More recently measurements on a stack of 0.8 µm slabs were reported (46) by the same group. Here kinks were observed in the frequency shift, rather than jumps expected to accompany the first order A-B transition, as observed in our experiment. Therefore we believe these results cannot be reliably interpreted as an observation of the A-B transition. Ambiguities in these data may be attributed to a combination of several factors: large static magnetic field inhomogeneity (the NMR linewidth at 717 kHz was reported to be 2.5 kHz, ten times higher than in our work); limited sensitivity of the conventional NMR spectrometer; presence of solid \(^3\text{He}\) at surfaces; significant thickness distribution comparable to that measured in the earlier work of this group.

### 4.2.3 Suppression of the Superfluid Transition Temperature in Films

This section provides further discussion of the observed suppression of the superfluid transition in the cavity, relative to bulk. Although the suppression of the A phase energy gap in the cavity with ‘diffuse’ boundaries agrees well with the prediction for diffusely scattering walls, the \(T_{c}^{\text{lab}}\) is found to
Fig. S12. Comparison of suppression of the superfluid transition temperature with theoretical predictions and previous measurements. The inset shows the detail of our measurements near $T_c$. The coordinates are chosen so constant confinement in Ginzburg-Landau regime, $D/\xi_{tr} = \text{const}$, is represented by straight lines. Also shown, solid red line, is the prediction of quasiclassical theory (29). Our data (RHUL) are presented for ‘diffuse’ boundaries, other measurements are for unsaturated pure $^3$He films. Measurements at Purdue University (19) show the weakest $T_c$ suppression. Slightly higher suppression was observed at Queen’s University (17), following the extrapolation of Ginzburg-Landau prediction $D/\xi_{tr} = \pi$. We observe even higher suppression, which together with recent measurements at RIKEN (47) follows $D/\xi_{tr} = 3.74$.

be suppressed more strongly than given by the same theory. This is shown in Fig. S12, compare also with Table S1.

Fig. S12 compares the suppression of the transition observed in our experiment with prior work on flow in unsaturated films. Since the free surface is assumed to be specular, the effective film thickness is double the actual film thickness. The early measurements (17, 19) show weaker $T_c$ suppression than ours, close to the theoretical prediction, however more recent studies at RIKEN (47) are in good agreement with our work. In all four of these experiments the temperature of the sample of confined helium is inferred from a thermometer coupled to bulk $^3$He or metal substrate used for cooling down the sample, therefore a systematic error in thermometry due to overheating of the sample cannot be ruled out. This would reduce the measured sample temperature and might lead to the observed disagreement with the theoretical prediction. However the close match between our measurements and those done at RIKEN suggests that the disagreement between these measurements and the theory may be real. The apparent agreement between the earlier measurements and the theory may arise from systematic errors in determining $^3$He film thickness. More experiments and theory are required to resolve this issue.

5 Gap Suppression in Confined A Phase Inferred from NMR Frequency Shift

As shown above in section 2.2 the NMR frequency shift exhibited by the A phase in the slab perpendicular to the static NMR field ($\hat{l} \parallel B_0$, $\hat{d} \perp B_0$) is exactly opposite to the shift observed in the bulk ($\hat{l} \parallel \hat{d} \perp B_0$), the magnitude of these shifts being

$$ |f^2 - f_z^2| = \frac{\gamma^2 \lambda_D N_F}{5\pi^2 \chi_N} \Delta^2. $$

When diffuse scattering is present at the walls, the energy gap is predicted to develop a spatially-dependent suppression $\Delta(z)$ (see Fig. 4B of the paper, where the gap profile for diffuse walls represents
calculations (24) at $T/T_{c}^{\text{bulk}} = 0.9$; for partially specular walls it is shown schematically, that the suppression is weaker than for diffuse walls (23), and the frequency shift evaluated locally takes different values across the slab. The method used for understanding NMR response from spin and orbital glass states in the A phase of $^3$He in aerogel (48) can be applied to a slab of thickness $D$ short compared to the dipolar length $\xi_D \sim 10 \mu m$ (2). Under this condition the NMR precession is uniform and the frequency shift is determined by the spatially averaged dipole energy

$$|f^2 - f_s^2| = \frac{\gamma_2 \lambda_D N_F}{5\pi^2 \chi_N} (\Delta^2(z)).$$

The $\gamma_2 \lambda_D N_F / 5\pi^2 \chi_N$ coefficient only depends on the normal state properties of $^3$He and at a given pressure it takes the same temperature independent value in bulk and in a slab with surfaces of any kind. This makes the frequency shift a powerful probe of the energy gap amplitude.

Close to $T_c$ in the Ginzburg-Landau (GL) regime (2) the squared energy gap is linear with temperature, this behaviour is reflected in the frequency shift (see Fig. 2 of the paper). To characterize the average gap suppression close to $T_c$ we determine the slope $S_{GL}$ as a coefficient in the straight-line fit

$$|f^2(T) - f_s^2| = S_{GL} \left( 1 - \frac{T}{T_{c}^{\text{slab}}} \right), \quad \text{over } 0.97T_{c}^{\text{slab}} < T < T_{c}^{\text{slab}}.$$

The values of $S_{GL}$ obtained for ‘diffuse’ and ‘specular’ walls are presented in Fig. S13. The temperature range is chosen to be large enough to obtain reasonably precise values of $S_{GL}$ but small enough that the temperature dependence of the frequency shift is nearly linear, so $S_{GL}$ reflects the initial slope (IS)

$$\text{IS} = \frac{\partial |f^2 - f_s^2|}{\partial (1 - T/T_{c}^{\text{slab}})}, \quad \text{at } T = T_{c}^{\text{slab}} - 0.$$ 

The initial slope of the bulk A phase has been measured at pressures down to 5 bar (49) and at this pressure (Fig. S13) it is in agreement with the interpolation of $S_{GL}$ measured in our experiment for ‘specular’ walls. These experimental data are fit by

$$S_{GL}^{\text{specular}}(P) = 0.14 \times 10^{10} \text{ Hz}^2 + 0.32 \times 10^{10} \text{ Hz}^2/\text{bar} \times P.$$ 

Given this agreement we hypothesise that the measured GL slope in the slab with ‘specular’ walls equals that of the bulk A phase at all pressures. We use this slope to scale both ‘specular’ and ‘diffuse’ data and compare them with the theoretical predictions.

In Fig. S14, we show the mean square energy gap in a slab with diffuse walls, calculated within the framework of quasi-classical theory in the weak-coupling limit (24). This is compared to the bulk energy gap, equivalent to a slab with specular walls, also in the weak-coupling limit (42). This plot also shows the temperature dependence of the slope $\partial \langle \Delta^2 \rangle / \partial (1 - T/T_{c}^{\text{slab}})$. Thus we emphasise that $S_{GL}$, evaluated from experiment and theory, is sensitive to the temperature range chosen. We evaluate a theoretical GL slope over the same reduced temperature range as chosen for the experimental data:

$$\langle \Delta^2 \rangle = S_{GL}^{\Delta} \left( 1 - \frac{T}{T_{c}^{\text{slab}}} \right), \quad \text{over } 0.97T_{c}^{\text{slab}} < T < T_{c}^{\text{slab}}.$$

The slopes $S_{GL}^{\Delta \text{diffuse}}$ and $S_{GL}^{\Delta \text{specular}}$ inferred from the theoretical calculations, Fig. S14, can be used to make a prediction for the initial slope with diffuse walls $S_{GL}^{\Delta \text{diffuse}} = S_{GL}^{\Delta} \times S_{GL}^{\Delta \text{diffuse}} / S_{GL}^{\Delta \text{specular}}$ shown with a red line in Fig. S13. The top right panel of the Fig. S13 shows the $S_{GL}^{\Delta \text{diffuse}} / S_{GL}^{\Delta \text{specular}}$ ratio directly. This procedure confirms that measured gap suppression for ‘diffuse’ boundaries near $T_c$ is in good agreement with the predictions of quasiclassical theory, supporting the above assumption.

The comparison between the experiment and theory, made over the whole temperature range, is shown in Fig. 4C in the paper, for data at 2.2 bar. Both the experimental and theoretical data are scaled by the initial GL slope for specular scattering. The experimental value of $S_{GL}^{\Delta \text{specular}}$(2.2 bar) = $0.62 \times 10^{10}$ Hz$^2$ is used to scale the experimental frequency shifts and $S_{GL}^{\Delta \text{specular}} = 0.29 \times 2\pi k_B T_{c}^{\text{bulk}}$ for the theoretically calculated gap (see Fig. S14). At this pressure the slab thickness distribution is characterised by the mean and standard deviation of $D/\xi_0 = 12.1 \pm 0.2$, with range $D_{\min} = 11.4\xi_0$ and $D_{\max} = 12.3\xi_0$. For the theoretical calculation $D = 12.0\xi_0$ is used.
**Fig. S13.** The left-hand panel shows the slope $S_{GL}$ of the A phase frequency shift near $T_c$ observed in the slab with the ‘diffuse’ and ‘specular’ boundaries and the initial slope observed in the bulk (49). See text for definition of $S_{GL}$. The low pressure region where our data were taken is shown in more detail in the bottom right panel. In the slab with ‘specular’ boundaries $S_{GL}$ is found to depend linearly on pressure. The ‘specular’ $S_{GL}$ agrees with the bulk initial slope at 5 bar and can be continued smoothly (although not linearly) to the bulk values at higher pressures. A straight line fit through $S_{GL}^{specular}(P)$ shown with a blue line is used to normalise all measurements, presented in the top right panel. The red line is a theoretical prediction of $S_{GL}$ obtained for fully diffuse walls under assumption $S_{GL}^{specular}$ corresponds to the bulk value at all pressures. See text and Fig. S14 caption for more details.

**Fig. S14.** Modelling the A phase in a slab with fully diffuse walls close to $T_c$. Left: the averaged squared energy gap $\langle \Delta^2 \rangle$ near $T_c$, based on (24, 42). Right: the derivative of $\langle \Delta^2 \rangle$ with respect to temperature reveals considerable non-linearity near $T_c$. Here $T_c$ stands for $T_c^{slab}$ and $T_c^{bulk}$ for the slab and bulk calculations respectively. For comparison with experiment a straight line is fitted through $\langle \Delta^2 \rangle$ vs $T$ in the $0.97T_c < T < T_c$ range. See text for more details.

**Supplementary References**

References and Notes


22. Materials and methods are available as supplementary materials on *Science* Online.


