

New chiral phases of superfluid ³He stabilized by anisotropic silica aerogel

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SI.1. GINZBURG-LANDAU THEORY

The Ginzburg-Landau (GL) free-energy functional for *p*-wave, spin-triplet pairing of ³He quasiparticles in a homogeneous isotropic medium is defined in Ref. [SI1] in terms of the 3×3 matrix order parameter, $A_{\mu j}$, which transforms as a vector under spin rotations with respect to index μ , separately as a vector under orbital rotations with respect to index j,

$$\Delta\Omega_{\mathsf{GL}}[A] = \int_{\mathsf{V}} d^3r \left\{ \bar{\alpha}(T) \operatorname{Tr} AA^{\dagger} + \bar{\beta}_1 \mid \operatorname{Tr} AA^{\operatorname{tr}} \mid^2 + \bar{\beta}_2 (\operatorname{Tr} AA^{\dagger})^2 \right. \\ \left. + \bar{\beta}_3 \operatorname{Tr} AA^{\operatorname{tr}} (AA^{\operatorname{tr}})^* + \bar{\beta}_4 \operatorname{Tr} (AA^{\dagger})^2 + \bar{\beta}_5 \operatorname{Tr} AA^{\dagger} (AA^{\dagger})^* \right\}.$$
(SI1)

The material coefficient, $\bar{\alpha}(T) \simeq (T - T_{ca})\bar{\alpha}'$, depends on the single spin density of states at the Fermi surface, N_F , the pairing interaction, and the effects of scattering by the random impurity potential, all of which determine the transition temperature, T_{ca} . The prime denotes differentiation with respect to temperature. The fourth-order material parameters, $\{\bar{\beta}_i | i = 1...5\}$, which are also sensitive to impurity scattering, determine condensation energy and the structure and residual symmetry of the superfluid condensate.

The homogeneous isotropic scattering model (HISM) of Thuneberg *et al.* [S11] was extended by Sauls and Sharma in Ref. [S12] to incorporate the effect of correlations between silica strands and clusters. In this model, the aerogel correlation length, ξ_a , limits the pairing transition for small superfluid coherence lengths, $\hbar v_F/2\pi k_B T_{ca} < \xi_a$ (high pressures), while the mean free path, λ , determines the transition when the superfluid coherence length is larger than the aerogel correlation length. This condition is achieved at low pressures, and always in the limit $T_{ca} \rightarrow 0$. Indeed this limit can be used to determine λ from the critical pressure at which $T_{ca}(P_c) = 0$. The two pair-breaking length scales define an effective pair-breaking parameter, $\tilde{x} = x/(1 + \zeta_a^2/x)$, $\zeta_a = \xi_a/\lambda$ where $x = \hbar v_F/2\pi k_B T\lambda$ is the depairing parameter in the HISM [SI3] and v_F is the pressure dependent Fermi velocity of ³He [SI4]. The superfluid transition temperature in aerogel, T_{ca} , is determined by the condition $\bar{\alpha}(T_{ca}) = 0$ in the following equation,

$$\bar{\alpha}(T) = \frac{1}{3} N_F \left[\ln(T/T_c) - 2\sum_{n=0}^{\infty} \left(\frac{1}{2n+1+\tilde{x}} - \frac{1}{2n+1} \right) \right],$$
 (SI2)

where T_c is the transition temperature of pure ³He. In the limit $\tilde{x} = 0$, Eq. SI1 reduces to the case for pure ³He, namely $\alpha = (1/3)N_F \ln(T/T_c)$.



FIG. SI1. Determination of Scattering Parameters Aerogel correlation length (red circles) and standard deviation of ξ_a as a function of λ (blue squares). The minimum in the standard deviation corresponds to $\lambda = 113$ nm and $\xi_a = 39$ nm.

The correlation length is a pressure independent length scale of the aerogel microstructure. We have used Eq. 2 to fit our measurements of $T_{ca}(P)$ in the stretched aerogel (Fig. 1c) with the constraint that ξ_a be pressure independent. From this fit we find $\lambda = 113$ nm and $\xi_a = 39$ nm. In Fig. SI1 we present the standard deviation of $\xi_a(P)$ as a function of λ . Using these same values of λ and ξ_a we have calculated the square of the axial state order parameter amplitude in aerogel from the GL theory,

$$\Delta_{Aa0}^2 = \Delta_{A0}^2 \frac{T_{ca}}{T_c} \frac{\bar{\alpha}'(T_{ca})}{\alpha'(T_c)} \frac{\beta_A}{\bar{\beta}_A}$$
(SI3)

In Eq. SI2 Δ_{A0}^2 is the value for the pure A-phase [SI5, SI6], and β_A ($\bar{\beta}_A$) is the combination of β -parameters appropriate to pure (aerogel) ³He-A, *i.e.* $\beta_A = \beta_2 + \beta_4 + \beta_5$ ($\bar{\beta}_A = \bar{\beta}_2 + \bar{\beta}_4 + \bar{\beta}_5$). The pure β -parameters were taken from Ref. [SI7]. For calculating the aerogel $\bar{\beta}$ -parameters, we have extended the results of Thuneberg *et al.* [SI1] for the HISM, to include the combined effects of the aerogel correlation length and mean-free path using the correlated scattering model of Ref. [SI2].

SI.2. NMR LINE SHAPE ANALYSIS

We have performed an analysis of the NMR line shape to obtain the distribution of the chiral axis, $\vec{\ell}$, in our stretched sample. A distribution in the direction of $\vec{\ell}$ relative to the external magnetic field, \vec{H} , will lead to a distribution in the intrinsic superfluid dipole field

and hence a distribution of frequency shifts in the NMR spectrum. If $\theta = \cos^{-1}(\hat{\ell} \cdot \hat{H})$ is the angle between the chiral axis and the magnetic field, then for small tip angle, β , the frequency shift in the A-phase reduces to [SI8],

$$\Delta\omega_A(\theta) = -\frac{\Omega_A^2}{2\omega_L}\cos(2\theta).$$
(SI4)

For example, if the chiral axis orientation is unique and the normal state lineshape is given by $F_n(\omega - \omega_L)$ then the lineshape in the superfluid, F_s , is unchanged except for a constant shift, *i.e.* $F_s = F_n(\omega - \omega_L - \Delta\omega_A)$ where $\Delta\omega_A$ is given by Eq. SI3. However, for an angular distribution of $\vec{\ell}$, $P(\theta)$, there is a distribution of frequency shifts, $P(\omega)$, determined by Eq. SI3. This results in a more complex NMR lineshape that is given by the convolution product of the normal state line and $P(\omega)$,

$$F_{s}(\omega) = \int P(\omega')F_{n}\left(\omega - \omega_{L} - \omega'\right) d\omega'.$$
(SI5)

Since the normal state and superfluid spectra are obtained experimentally, $P(\theta)$ can be determined by fitting the spectrum in the superfluid phase with Eq. SI4.

In general an inversion problem, like this one, requires constraints which make it difficult to determine the uniqueness of the distribution. We choose the model that the $\vec{\ell}$ -distribution can be represented by a sum of gaussian functions with adjustable position, θ_i , width, w_i , and relative weight, A_i ,

$$P(\theta) = \sum_{i} A_{i} e^{\left(\frac{\theta - \theta_{i}}{w_{i}}\right)^{2}},$$
(SI6)

and $\int P(\theta) d\theta = 1$. In Fig. 4a we present NMR spectra (bold green curves) and corresponding fits (dashed black curves) for temperatures above (left panel) and below (right panel) the disorder transition T_d with the results for $P(\theta)$ shown in Fig. 4b. For $T_d < T < T_{ca}$ we find that the spectrum is well-represented with a distribution centered at $\theta = 90^\circ$, corresponding to the dipole-locked configuration, with a spread of $17.8^\circ \pm 3.6^\circ$. For $T < T_d$, we find that the ℓ -distribution is composed of domains as described in the text. Approximately 1/3 of the sample remains with ℓ perpendicular to \vec{H} with a spread $\leq 4^\circ$ as shown in Fig. SI2. where we present the squared residual of the fit as a function of the width of the dipole-locked peak while keeping the other parameters fixed. The remaining $\sim 2/3$ of the superfluid has $\theta = 43.8^\circ \pm 0.1^\circ$ with a width of $5.0^\circ \pm 0.8^\circ$ as shown in Fig. SI3. This distribution fits our spectrum well for $T < T_d$. To check its uniqueness we have attempted



FIG. SI2. Squared residual versus the width of the dipole-locked peak for $T < T_d$.



FIG. SI3. Squared residual versus the a) position, b) percent weight, and c) width of the dipoleunlocked peak for $T < T_d$.

to add an additional peak with variable relative weight, w_v , at the position $\theta = 67^{\circ}$, with a width of $\theta = 6^{\circ}$ and we find that the best fit to the spectrum corresponds to $w_v = 0$ as shown in Fig. SI4a. To emphasize this point, in Fig. SI4b we show the lineshape that results for the case $w_v = 15\%$ that clearly adds substantial weight in the spectrum which is inconsistent with our experiment. We infer, therefore, that the disordered state has an $\vec{\ell}$ -distribution which is non-monotonic with three principal components as shown in Fig. 4b.



FIG. SI4. Attempt to add an additional peak a) Squared residual versus the weight of an additional peak at $\theta = 67^{\circ}$. b) Fit (dashed black curve) to the NMR spectrum below T_d (bold green curve) produced with $w_v = 15\%$. The normal state line is presented as the fine red curve and the fit is performed using Eq. SI4. Significant discrepancy between the best fit and the NMR spectrum indicates that extra weight in the ℓ -distribution between 44° and 90° can be ruled out.

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