Magnetic Response of Odd-Frequency *s*-Wave Cooper Pairs in a Superfluid Proximity System

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We investigate the magnetic response of a dirty-normal-Fermi-liquid–spin-triplet-superfluid proximity system consisting of liquid ^{3}He and aerogel. In contrast to bulk superfluids, Pauli spin susceptibility in the proximity system exceeds its normal-state value locally around the interface. This enhanced Pauli paramagnetism originates from odd-frequency s-wave pairing arising due to spatial inhomogeneity. A characteristic observable signature of the paramagnetic effect can be found in the spin susceptibility temperature dependence.

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In general, Cooper pairing states can be classified into even- and odd-frequency symmetry classes [1,2]. Because of their frequency symmetry, odd-frequency pairs do not have equal-time correlation. Interestingly, such an exotic pairing may arise in normal metals attached to an even-frequency superconductor as a result of the combined effect of orbital parity mixing at the interface and the penetration of Cooper pairs [3,4]. When the normal metal is a ferromagnet [5,6] or the interface has magnetic properties [7], the singlet-triplet pair mixing due to the magnetism provides another mechanism of odd-frequency-pair creation.

Although odd-frequency paring is possible in bulk materials through a strong retardation effect in two-particle interactions [1,2], conclusive evidence has not yet been observed to date. It is therefore of great interest to detect odd-frequency pairs existing ubiquitously in a variety of proximity systems. Of particular interest are those with s-wave orbital symmetry because they are robust against impurity scattering and can diffuse deeply into normal metals. The long-range penetration of the odd-frequency s-wave pairs has been demonstrated using a strong ferromagnet in Josephson devices [8,9], but the long-range effect itself is not due to odd-frequency symmetry but to the s-wave orbital structure.

From recent theoretical studies of the proximity effect, it is becoming clear that odd-frequency s-wave pairs exhibit an anomalous response to external fields. The Meissner response is predicted to be paramagnetic [10], which is in stark contrast to conventional Meissner diamagnetism of even-frequency superconductors. A relevant peculiarity in the electromagnetic response has also been predicted [11]. In this Letter, we show that nontrivial behavior due to the odd-frequency s-wave pairing manifests itself in Pauli spin magnetism. We propose a method for detecting the proximity-induced odd-frequency s-wave pairs via measurements of the spin susceptibility.

To investigate the spin magnetism, we consider the superfluid proximity system proposed in Ref. [12] and shown in Fig. 1. Aerogel, a highly porous material [13–20], is partly immersed in liquid ^{3}He, a Fermi liquid that undergoes a superfluid transition at $T_c \sim 1$ mK to a spin-triplet p-wave state [21]. The aerogel introduces impuritylike disorder into the liquid ^{3}He and destroys any non-s-wave Cooper pairs. The pair-breaking effect in bulk superfluid ^{3}He has already been studied in detail using silica aerogel with a ~98% porosity [13–20]. The mean free path l in the aerogel is comparable to the superfluid coherence length ξ_0, and thus the aerogel induces strong pair breaking. Because of the pressure dependence of ξ_0, the pair breaking is stronger at lower pressures and even completely suppresses the non-s-wave superfluidity of ^{3}He below a critical pressure P_c [15,17,18]. Thus, at low pressures below P_c, odd-frequency s-wave pairs dominate the superfluidity induced in the aerogel layer by the proximity effect.

Since liquid ^{3}He in the bulk is intrinsically pure and its superfluid properties are theoretically well understood [21], the superfluid proximity structure serves as a

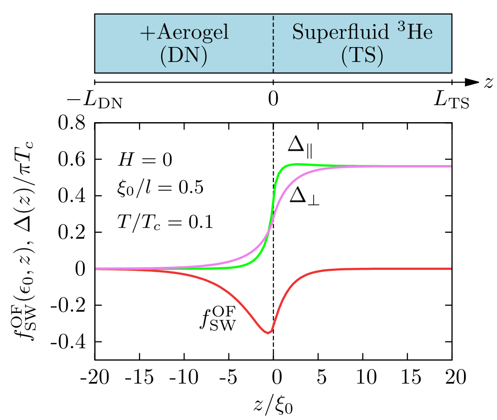

FIG. 1 (color online). Dirty-normal-Fermi-liquid–spin-triplet-superfluid (DN/TS) proximity system consisting of liquid ^{3}He and aerogel. Liquid ^{3}He fills the entire space $-L_{\rm DN} < z < L_{\rm TS}$, and the aerogel is embedded into the portion $-L_{\rm DN} < z < 0$. The plot shows the spatial dependence of the p-wave gap functions $\Delta_{\parallel,\perp}$ and the odd-frequency s-wave pair amplitude $f^{\rm OF}_{\rm SW}$ at the lowest Matsubara frequency in the case for which liquid ^{3}He is in the superfluid B phase. Impurity scattering by the aerogel suppresses the p-wave gap functions.

well-defined model system to investigate the proximity effect. Moreover, it presents a unique testing ground for exploring the spin magnetism of proximity-induced odd-frequency states because liquid ^{3}He does not show orbital magnetism such as Meissner effect and there is no intrinsic magnetic field such as in ferromagnets. In what follows, our attention is mainly focused on the magnetic response of the odd-frequency s-wave pairs induced in the aerogel layer.

Our theory for the superfluidity is based on the quasiclassical Green's function method [22]. Since the coherence length of superfluid ^{3}He is much longer than the Fermi wavelength, quasiclassical theory gives a quantitative and reliable description of superfluid phenomena [22]. The quasiclassical Green's function $\hat{g}$ is defined as a 4×4 Nambu-space matrix and is a function of $(\hat{p}, \epsilon_n, z)$, where $\hat{p}$ is the unit vector specifying the direction of the Fermi momentum, and $\epsilon_n = \pi T(2n+1)(n = 0, \pm 1, \pm 2, \ldots)$ is the Matsubara frequency at a temperature T. In the present system, $\hat{g}$ obeys

$$iv_F \hat{p}_z \partial_z \hat{g} = [\hat{g}, \hat{\epsilon}(\hat{p}, \epsilon_n, z) - \hat{\sigma}_{\mathrm{imp}}(\hat{p}, \epsilon_n, z)], \qquad (1)$$

where v_F is the Fermi velocity, $\hat{\sigma}_{\mathrm{imp}}$ is the impurity self-energy in the aerogel layer, and $\hat{\epsilon}$ is an energy matrix of the form

$$\hat{\epsilon}(\hat{p}, \epsilon_n, z) = \begin{pmatrix} i\epsilon_n - v(\hat{p}, z) & \Delta(\hat{p}, z) \\ \tilde{\Delta}(\hat{p}, z) & -i\epsilon_n - \tilde{v}(\hat{p}, z) \end{pmatrix}. \qquad (2)$$

Equation (1) is supplemented by the normalization condition $\hat{g}^2 = -1$. In Eq. (2), v is a perturbation including the Fermi liquid correction, Δ is a spin-triplet p-wave gap function, and $\tilde{X}$ denotes the transformation $\tilde{X}(\hat{p}) = X(-\hat{p})^*$. The perturbation of interest here is Zeeman coupling, $-\mu_0 \boldsymbol{H} \cdot \boldsymbol{\sigma}$ + (the Fermi liquid correction), where μ_0 is the magnetic moment of ^{3}He, $\boldsymbol{H}$ is the external magnetic field, and $\boldsymbol{\sigma}$ is the Pauli matrix. The matrix structure of $\hat{g}$ can be parametrized as

$$\hat{g} = \begin{pmatrix} g & f \\ \tilde{f} & \tilde{g} \end{pmatrix}. \qquad (3)$$

The spin-space matrices g and f have symmetries of

$$g(\hat{p}, \epsilon_n, z) = g(\hat{p}, -\epsilon_n, z)^\dagger, \qquad (4)$$

$$f(\hat{p}, \epsilon_n, z) = -f(-\hat{p}, -\epsilon_n, z)^T, \qquad (5)$$

where the superscript T denotes the transpose.

The general symmetry relation of Eq. (5) can be used to classify the Cooper-pair amplitude. The matrix decomposition $f = (f_0 + \boldsymbol{f} \cdot \boldsymbol{\sigma}) i\sigma_2$ defines the spin-singlet (f_0) and spin-triplet ($\boldsymbol{f}$) pair amplitudes. The latter is related self-consistently to the spin-triplet gap function $\Delta(\hat{p}, z) = \boldsymbol{d}(\hat{p}, z) \cdot \boldsymbol{\sigma} i\sigma_2$ through

$$\boldsymbol{d}(\hat{p}, z) = -N(0)\pi T \sum_n \langle v_{\hat{p},\hat{p}'} \boldsymbol{f}(\hat{p}', \epsilon_n, z) \rangle_{\hat{p}'}, \qquad (6)$$

where $N(0)$ is the density of states at the Fermi level in the normal state, $v_{\hat{p},\hat{p}'} = -3v_1 \hat{p} \cdot \hat{p}'$ is the p-wave pairing interaction, and $\langle \cdots \rangle_{\hat{p}}$ denotes the average over the Fermi surface. The even-frequency (EF) and odd-frequency (OF) pair amplitudes are defined by

$$\left. \begin{array}{l} f^{\mathrm{EF}}(\hat{p}, \epsilon_n, z) \\ f^{\mathrm{OF}}(\hat{p}, \epsilon_n, z) \end{array} \right\} = \frac{1}{2}[f(\hat{p}, \epsilon_n, z) \pm f(\hat{p}, -\epsilon_n, z)]. \qquad (7)$$

From $g = g_0 + \boldsymbol{g} \cdot \boldsymbol{\sigma}$, we can calculate the spin magnetization as

$$\boldsymbol{M}(z) = \chi_N \boldsymbol{H} - \frac{\chi_N}{\mu_0} \pi T \sum_n \langle \boldsymbol{g}(\hat{p}, \epsilon_n, z) \rangle_{\hat{p}}. \qquad (8)$$

Here, $\chi_N = 2N(0)\mu_0^2/(1 + F_0^a)$ is the spin susceptibility in the normal state with $F_0^a \simeq -0.7$ [23] being a Landau parameter associated with the Fermi liquid correction to the Zeeman coupling. The normalization condition $\hat{g}^2 = -1$ requires the following relation to hold:

$$\boldsymbol{g} = -\frac{1}{2g_0}[\tilde{\boldsymbol{f}} f_0 - \boldsymbol{f} \tilde{f}_0 + i(\boldsymbol{f} \times \tilde{\boldsymbol{f}})]. \qquad (9)$$

As we shall see later, using Eqs. (8) and (9), we can derive a formula [Eq. (15) along with Eqs. (16) and (17)] that explicitly relates the spin susceptibility to the pair amplitudes in the aerogel layer.

Sharma and Sauls [19] employed the above quasiclassical theory to develop a linear response theory for the impurity effect on the spin susceptibility of superfluid ^{3}He in an infinitely large aerogel, and the theory was found to give good agreement with the experimental results [20].

To study the magnetic response of the superfluid proximity system, we calculated $\hat{g}$ numerically using the Riccati parametrization technique [12,24,25]. The following simple but realistic model system was considered. Liquid ^{3}He is assumed to be in the superfluid B phase, a typical example of even-frequency spin-triplet states in which odd-frequency-pair creation occurs at the interface [12] and surface [25]. The layer widths, L_{DN} and L_{TS}, are much larger than $\xi_0 = v_F/2\pi T_c$, which is at most ~80 nm. The numerical results presented here are those for L_{DN}, $L_{\mathrm{TS}} \to \infty$. For simplicity, the magnetic field is applied perpendicular to the interface; the system then preserves the rotational symmetry around the interface normal. The aerogel-superfluid interface is characterized by a discontinuous change in the impurity scattering rate $1/\tau$ at $z = 0$; i.e., $1/\tau$ takes a constant finite value for $z < 0$ but is zero for $z > 0$. We then evaluated the impurity self-energy using the standard self-consistent Born approximation, $\hat{\sigma}_{\mathrm{imp}} = -\langle \hat{g} \rangle_{\hat{p}}/2\tau$.

The mean free path $l = v_F \tau$ in a typical aerogel with a 98% porosity is 100–200 nm. Thus, the aerogel provides moderate disorder $\xi_0/l \lesssim 1$ that is sufficient enough to destroy the superfluidity of ^{3}He, but the dirty-limit condition $\xi_0/l \gg 1$ is not fulfilled. Nonetheless, dirty-limit behavior, as expected from Usadel theory [26] for superconducting alloys, can be seen in the numerical results we present here.

In the aerogel-superfluid ^{3}He-B system, the d vector takes the form [27–30]

$$\boldsymbol{d}(\hat{p}, z) = \Delta_\parallel(z)(\hat{p}_x \boldsymbol{e}_1 + \hat{p}_y \boldsymbol{e}_2) + \Delta_\perp(z)\hat{p}_z \boldsymbol{e}_3$$
$$= \Delta_\parallel(z)\sqrt{1-\hat{p}_z^2}\boldsymbol{e}_1'(\phi) + \Delta_\perp(z)\hat{p}_z \boldsymbol{e}_3, \qquad (10)$$

where $\phi = \arctan(\hat{p}_y/\hat{p}_x)$ is the azimuthal angle of $\hat{p}$, and $\boldsymbol{e}_1'(\phi) = \cos\phi \boldsymbol{e}_1 + \sin\phi \boldsymbol{e}_2$. The spin-triplet pair amplitude $\boldsymbol{f}$ has the self-consistent structure

$$\boldsymbol{f} = f_\parallel(\hat{p}_z, \epsilon_n, z)\boldsymbol{e}_1'(\phi) + f_\perp(\hat{p}_z, \epsilon_n, z)\boldsymbol{e}_3. \qquad (11)$$

In the aerogel, the pair amplitude is expected to be nearly isotropic (independent of $\hat{p}$) owing to impurity scattering. According to Usadel theory [26], this implies that the $f_\parallel \boldsymbol{e}_1'(\phi)$ term, which does not have an s-wave component, decays rapidly in the aerogel layer with a proximity distance of order l, while the $f_\perp \boldsymbol{e}_3$ term can penetrate to a distance $\sim(v_F l/6\epsilon_n)^{1/2} \propto T^{-1/2}$. It is also suggested from Usadel theory that $f_\perp$ in the aerogel layer is composed mainly of s-wave (SW) pairs and subdominant p-wave (PW) pairs:

$$f_\perp(\hat{p}_z, \epsilon_n, z) \simeq f_{\rm SW}^{\rm OF}(\epsilon_n, z) + 3\hat{p}_z f_{\rm PW}^{\rm EF}(\epsilon_n, z). \qquad (12)$$

In the present system, the s-wave component $f_{\rm SW}^{\rm OF}$ is generated by parity mixing at the aerogel-superfluid interface [12]. In Fig. 1, we show the z dependence of $f_{\rm SW}^{\rm OF}(\epsilon_0, z)$ and $\Delta_{\parallel,\perp}(z)$. The penetration of $\Delta_\perp(z)$ shows the manifestation of $f_{\rm PW}^{\rm EF}(\epsilon_n, z)$ in the aerogel layer. We can see that the proximity range of both $f_{\rm SW}^{\rm OF}(\epsilon_0, z)$ and $\Delta_\perp(z)$ is larger than that of $\Delta_\parallel(z)$. The difference in the proximity range is also demonstrated in Fig. 2(b).

Turning to the magnetic response, the magnetization $\boldsymbol{M}(z)$ in the system is along the z direction, as would be expected from the geometry, which can be shown explicitly from the fact that g_0 and f_0 are independent of ϕ because of the rotational symmetry about z. We are therefore interested in $M_z(z)$ or the local spin susceptibility $\chi^{zz}(z) = M_z(z)/H$ given by

$$\chi^{zz}(z) = \chi_N + \frac{\chi_N}{\mu_0 H}\pi T \sum_n \left\langle \frac{\tilde{f}_\perp f_0 - f_\perp \tilde{f}_0}{2g_0} \right\rangle_{\hat{p}}. \qquad (13)$$

Here, the arguments of the Green's functions are omitted for brevity. We note that Eq. (13) has a physically appealing structure; the product of the singlet (f_0) and triplet ($f_\perp$) pair amplitudes, which are mixed by the applied magnetic field, determines the magnitude of the deviation of the spin susceptibility from its normal-state value.

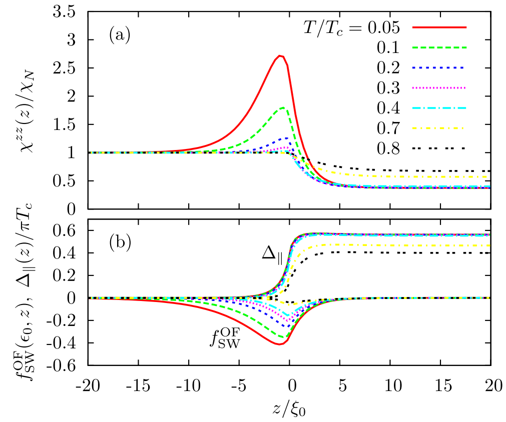

FIG. 2 (color online). Spatial dependence of (a) $\chi^{zz}(z)/\chi_N$ and (b) $\Delta_\parallel(z)/\pi T_c$ and $f_{\rm SW}^{\rm OF}(\epsilon_0, z)$ at various reduced temperatures T/T_c. In the numerical calculations, we take $\xi_0/l = 0.5$, $\mu_0 H/\pi T_c = 0.005$, and $F_0^a = -0.7$.

In Fig. 2(a), the susceptibility $\chi^{zz}(z)/\chi_N$ obtained from Eq. (13) is plotted as a function of z/ξ_0. The pair-breaking parameter ξ_0/l is the same as that in Fig. 1. The magnitude of the magnetic field is taken to be $\mu_0 H/\pi T_c = 0.005$, which corresponds to $H = 20$–50 mT (the range is due to the pressure dependence of T_c). This is such a weak magnetic field that $\Delta_{\parallel,\perp}(z)$ and $f_{\rm SW}^{\rm OF}(\epsilon_n, z)$ are almost unperturbed. As the temperature decreases from T_c, the bulk superfluid value of $\chi^{zz}(z)/\chi_N$ decreases from unity. However, the interface value exceeds unity at low temperatures below $\sim 0.4T_c$, and at the same time, a peak appears in the aerogel layer. This peak has a rather long tail on the aerogel side. The proximity distance increases with decreasing temperature even below the temperature at which the bulk superfluid susceptibility is already well saturated.

In Fig. 2(b), the z dependence of $f_{\rm SW}^{\rm OF}(\epsilon_0, z)$ is shown for the same parameters as those in Fig. 2(a). For reference, the gap function $\Delta_\parallel(z)$ is also plotted as a typical quantity with a proximity distance of order l. A clear correlation is seen between the odd-frequency s-wave pair amplitude and the peak structure of the local susceptibility.

To express this relationship more definitively, we analyze Eq. (13). In a dirty system, an approximation similar to that in Eq. (12) can be applied to the spin-singlet pair amplitude f_0;

$$f_0(\hat{p}_z, \epsilon_n, z) \simeq f_{\rm SW}^{\rm EF}(\epsilon_n, z) + 3\hat{p}_z f_{\rm PW}^{\rm OF}(\epsilon_n, z). \qquad (14)$$

The applied magnetic field induces the singlet pair amplitudes $f_{\rm SW}^{\rm EF}$ and $f_{\rm PW}^{\rm OF}$ in the triplet superfluid characterized by $f_{\rm SW}^{\rm OF}$ and $f_{\rm PW}^{\rm EF}$. For a weak magnetic field, the total pair amplitude f is dominated by the odd-frequency s-wave component $f_{\rm SW}^{\rm OF}$. When the magnitude $|f_{\rm SW}^{\rm OF}|$ is sufficiently smaller than unity [Fig. 2(b)], we can replace g_0 by the normal-state value $s_n i$ with $s_n = {\rm sgn}(\epsilon_n)$. Then, from Eq. (13) we obtain the following formula for $\chi^{zz}(z)/\chi_N$ in dirty systems:

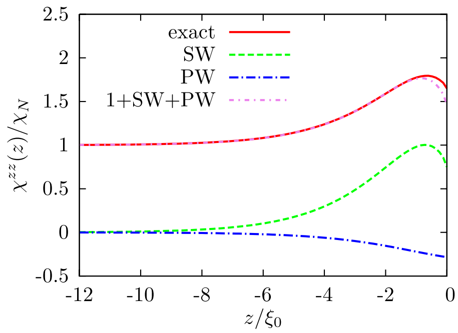

FIG. 3 (color online). Contributions to $\chi^{zz}(z)/\chi_N$ from the s-wave (SW) and p-wave (PW) pairs in the aerogel layer. The results are obtained from Eqs. (16) and (17) for $T/T_c = 0.1$, $\xi_0/l = 0.5$, $\mu_0 H/\pi T_c = 0.005$, and $F_0^a = -0.7$. The full numerical result $\chi^{zz}(z)/\chi_N$ in the aerogel layer is well approximated by $1 + \mathrm{SW} + \mathrm{PW}$, i.e., Eq. (15).

$$\frac{\chi^{zz}(z)}{\chi_N} = 1 + \frac{\chi^{zz}_{\mathrm{SW}}(z)}{\chi_N} + \frac{\chi^{zz}_{\mathrm{PW}}(z)}{\chi_N}, \tag{15}$$

where

$$\frac{\chi^{zz}_{\mathrm{SW}}(z)}{\chi_N} = \frac{\pi T}{\mu_0 H}\sum_n s_n \mathrm{Im}[f^{\mathrm{OF}}_{\mathrm{SW}}(\epsilon_n, z)^* f^{\mathrm{EF}}_{\mathrm{SW}}(\epsilon_n, z)], \tag{16}$$

$$\frac{\chi^{zz}_{\mathrm{PW}}(z)}{\chi_N} = -\frac{3\pi T}{\mu_0 H}\sum_n s_n \mathrm{Im}[f^{\mathrm{EF}}_{\mathrm{PW}}(\epsilon_n, z)^* f^{\mathrm{OF}}_{\mathrm{PW}}(\epsilon_n, z)]. \tag{17}$$

As demonstrated in Fig. 3, Eqs. (15)–(17) give a good description of the local spin susceptibility in the aerogel layer. The deviation from the full theory is apparent only in the vicinity of the interface where any partial wave components of the pair amplitude can have a comparable amplitude [12]. Figure 3 shows clearly that the peak in the local susceptibility comes from $\chi^{zz}_{\mathrm{SW}}(z)$, which describes the change in $\chi^{zz}(z)$ due to the formation of odd-frequency s-wave pairs in the original unperturbed state of the system. Interestingly, $\chi^{zz}_{\mathrm{SW}}(z)$ has the opposite sign to $\chi^{zz}_{\mathrm{PW}}(z)$, as determined by the characteristic product $(f^{\mathrm{EF}}_{\mathrm{PW}})^* f^{\mathrm{OF}}_{\mathrm{PW}}$ of conventional even-frequency odd-parity superfluids.

For experimental purposes, it is useful to consider the interface susceptibility as defined by

$$\Delta\chi^{zz} = \Delta\chi^{zz}_A + \Delta\chi^{zz}_S, \tag{18}$$

where

$$\Delta\chi^{zz}_A = \int_{z<0}\frac{dz}{\xi_0}[\chi^{zz}(z) - \chi^{zz}(-\infty)], \tag{19}$$

$$\Delta\chi^{zz}_S = \int_{z>0}\frac{dz}{\xi_0}[\chi^{zz}(z) - \chi^{zz}(\infty)]. \tag{20}$$

In Fig. 4(a), we show the temperature dependence of the interface susceptibility $\Delta\chi^{zz}$ for $\xi_0/l = 0.5$, 0.75, and 1.0. The individual components $\Delta\chi^{zz}_A$ and $\Delta\chi^{zz}_S$ along with the total are shown for $\xi_0/l = 0.5$ in Fig. 4(b). The results are shown down to a temperature of $T = 0.05T_c$, which is somewhat below the lowest experimentally accessible

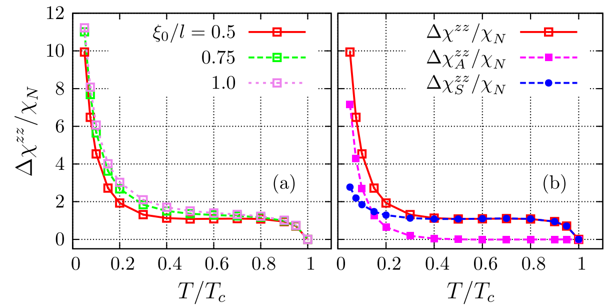

FIG. 4 (color online). Temperature dependence of the interface susceptibility $\Delta\chi^{zz} = \Delta\chi^{zz}_A + \Delta\chi^{zz}_S$ for $\mu_0 H/\pi T_c = 0.005$ and $F_0^a = -0.7$. (a) $\Delta\chi^{zz}$ for $\xi_0/l = 0.5$, 0.75, 1.0. (b) $\Delta\chi^{zz}$, $\Delta\chi^{zz}_A$, and $\Delta\chi^{zz}_S$ for $\xi_0/l = 0.5$.

temperature of $\sim 0.1T_c$ [31]. The contribution from the superfluid layer $\Delta\chi^{zz}_S$ becomes nearly independent of T for temperatures below T_c after an initial increase because the superfluid healing length decreases with T, while the interface value of the local susceptibility increases relative to the bulk value. At low temperatures where the local susceptibility has a peak, the contribution from the aerogel layer $\Delta\chi^{zz}_A$ gives rise to a steep increase in $\Delta\chi^{zz}$.

In summary, we have studied the magnetic response of the odd-frequency s-wave Cooper pairs induced by the proximity effect in the aerogel-superfluid ^{3}He system as depicted in Fig. 1. Clear-cut evidence for the penetration of the odd-frequency s-wave pairs is provided by the "chair-shape" temperature dependence of the interface susceptibility $\Delta\chi^{zz}$, as shown in Fig. 4(a). Two effects cause the characteristic low-temperature increase of $\Delta\chi^{zz}$. One is the increase in the proximity distance. As a consequence of this effect, $\Delta\chi^{zz}$ continues to increase, at least until the proximity distance reaches the system size. The other effect is the local susceptibility enhancement over the Pauli paramagnetic value χ_N. We have shown that this anomaly is due to the formation of odd-frequency s-wave pairs. In the present work, we have focused on a dirty system in which superfluidity is dominated by odd-frequency s-wave pairs. We note that local enhancement of the spin susceptibility has also been predicted to occur at the nodes of the spatially oscillating gap function of a Fulde-Ferrell-Larkin-Ovchinnikov superconducting state [32] and at the surfaces of a superfluid ^{3}He film [29]. These systems present the opportunity to investigate the spin magnetism of non-s-wave odd-frequency pairs arising due to spatial inhomogeneity and to discuss whether the enhanced Pauli paramagnetism is a universal property of odd-frequency paring states.

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