Effects of surface roughness on paramagnetic response of small unconventional superconductors

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We theoretically study effects of surface roughness on the magnetic response of small unconventional superconductors by solving the Eilenberger equation for the quasiclassical Green function and the Maxwell equation for the vector potential simultaneously and self-consistently. The paramagnetic phase of spin-singlet \(d\)-wave superconducting disks is drastically suppressed by the surface roughness, whereas that of spin-triplet \(p\)-wave disks is robust even in the presence of the roughness. Such difference derives from the orbital symmetry of paramagnetic odd-frequency Cooper pairs appearing at the surface of disks. The orbital part of the paramagnetic pairing correlation is \(p\)-wave symmetry in the \(d\)-wave disks, whereas it is \(s\)-wave symmetry in the \(p\)-wave ones. Calculating the free-energy, we also confirm that the paramagnetic state is more stable than the normal state, which indicates a possibility of detecting the paramagnetic effect in experiments. Indeed our results are consistent with an experimental finding on high-\(T_c\) thin films.

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I. INTRODUCTION

Diamagnetic response to an external magnetic field is a fundamental property of all superconductors.\(^1\) The Meissner current (coherent motion of the Cooper pairs) screens a weak magnetic field at a surface of a superconductor. As a result, the phase coherence of superconducting condensate is well preserved far away from the surface. A number of experiments, however, have reported the paramagnetic response of small superconductors and mesoscopic proximity structures.\(^2\)–\(^7\)

Recent theoretical studies have suggested the existence of paramagnetic Cooper pairs in inhomogeneous superconductors.\(^8\)–\(^14\) A spatial gradient of the superconducting order parameter induces subdominant pairing correlations. The pairing symmetry of such induced Cooper pairs is different from that of principal Cooper pairs in bulk superconducting state.\(^13\)–\(^15\) For example, the principal Cooper pairs in high-\(T_c\) superconductors belong to the spin-singlet \(d\)-wave (even parity) class. In (110) direction of high-\(T_c\) cuprate, a surface acts as a pair breaker and suppresses the pair potential drastically. Simultaneously, the spin-singlet odd-parity pairs are locally induced at the surface as a subdominant correlation. A surface generates odd-parity pairing correlations from the \(d\)-wave even-parity correlation because the surface breaks inversion symmetry locally. Since the pairing correlation function must be antisymmetric under the permutation of two electrons, the induced pairs have the odd-frequency symmetry.\(^16\) To our knowledge, such induced odd-frequency pairs indicate the paramagnetic response to an external magnetic field. Odd-frequency Cooper pairs can be generated also from conventional superconductors in the presence of spin-dependent potentials.\(^17\)–\(^18\)

In a previous paper,\(^19\) we have shown that magnetic susceptibility of small enough unconventional superconducting disks can be paramagnetic at a sufficiently low temperature. Odd-frequency Cooper pairs induced by a surface are responsible for the unusual paramagnetic Meissner effect. The magnetic response of Cooper pair is well characterized by so-called “pair density” which is defined by diagonal elements of the response function to a magnetic field. Even-frequency Cooper pairs have a positive pair density, whereas induced odd-frequency pairs have a negative pair density. So far an experiment has reported the decrease of pair density at low temperature in high-\(T_c\) superconducting films on which internal surfaces are introduced by the heavy-ion irradiation.\(^20\) Thus our theoretical results are consistent with the experiment at least qualitatively. However, signs of the paramagnetic effect in the experiment is much weaker than our theoretical prediction. The discrepancy may come from sample quality at surfaces. Artificially introduced internal surfaces can be very rough in the experiment, whereas surfaces are specular in the theory. Actually, several theories have pointed out that the surface roughness affects properties of the surface Andreev bound states of a high-\(T_c\) superconductor.\(^21\)–\(^24\)

The purpose of this paper is to clarify effects of surface roughness on the paramagnetic Meissner response of small unconventional superconductors. We consider a two-dimensional superconducting disk with spin-singlet \(d\)-wave symmetry or spin-triplet \(p\)-wave one. In numerical simulation, we solve the Eilenberger equation and the Maxwell equation simultaneously and self-consistently. Surface roughness is considered through an impurity self-energy within the Born approximation. We find that the surface roughness suppresses drastically the paramagnetic response of a spin-singlet \(d\)-wave superconducting disk. On the other hand in a spin-triplet \(p\)-wave disk, the paramagnetic property is robust even in the presence of surface roughness. The induced odd-frequency
where \( \omega_n = (2n + 1)\pi T \) is the fermionic Matsubara frequency, \( n \) is an integer number, and \( T \) is a temperature. In this paper, the symbol \( \cdot \cdot \cdot \) represents a \( 4 \times 4 \) matrix structure, \( \cdot \cdot \cdot \) represents a \( 2 \times 2 \) matrix structure in spin space and \( \hat{\sigma}_0 \) is the identity matrix in spin space. A vector potential is denoted by \( \mathbf{A}(r) \). We introduced a definition \( K(r, \mathbf{k}, i\omega_n) \equiv K^*(r, -\mathbf{k}, i\omega_n) \) for all functions \( K(r, \mathbf{k}, i\omega_n) \). Effects of rough surfaces are taken into account through an impurity self-energy of a quasiparticle defined by,

\[
\hat{\Sigma}(r, i\omega_n) = \Theta(|r| - R + w) \frac{i}{2\tau_0} \int \frac{dk}{2\pi} \hat{g}(r, \mathbf{k}, i\omega_n),
\]

where \( \tau_0 \) is the life time of a quasiparticle and \( \Theta(x) \) is the Heviside step function. The mean free path of a quasiparticle is defined by \( \ell = v_F \tau_0 \) in the disordered region. The anomalous Green function \( \hat{f}(r, \mathbf{k}, i\omega_n) \) is originally defined by an average of two annihilation operators of an electron. The relation

\[
\hat{f}(r, \mathbf{k}, i\omega_n) = -\hat{f}^T(r, -\mathbf{k}, -i\omega_n),
\]

represents the antisymmetric property of the anomalous Green function under the permutation of two electrons, where \( T \) represents the transpose of a matrix.

The direction of \( \mathbf{k} \) in two-dimensional momentum space is represented by an angle \( \theta \) measured from the \( x \) axis, (i.e., \( k_x = \cos \theta \) and \( k_y = \sin \theta \)). In what follows, we consider two unconventional superconductors with different pairing symmetries. One is spin-singlet \( d \)-wave symmetry \( \Delta(r, \theta) = \Delta(r) \sin(2\theta) \hat{\sigma}_z \). The other is spin-triplet \( p \)-wave symmetry \( \Delta(r, \theta) = \Delta(r) \cos(\theta) \hat{\sigma}_1 \), where \( \hat{\sigma}_j \) for \( j = 1-3 \) are the Pauli matrices in spin space. A \( d \)-wave and a \( p \)-wave pair potentials in momentum space are shown schematically in Fig. 1(b) and (c), respectively. We do not consider any spin-dependent potentials in this paper. The matrix structure of Green function is represented by

\[
\hat{g}(r, \mathbf{k}, i\omega_n) = g(r, \mathbf{k}, i\omega_n)\hat{\sigma}_0,
\]

\[
\hat{f}(r, \mathbf{k}, i\omega_n) = f(r, \mathbf{k}, i\omega_n) \times \left\{ \begin{array}{ll} \hat{\sigma}_2 & \text{for spin-singlet} \\ -i\hat{\sigma}_1 & \text{for spin-triplet} \end{array} \right.,
\]

with scalar Green functions \( g(r, \mathbf{k}, i\omega_n) \) and \( f(r, \mathbf{k}, i\omega_n) \). Spatial dependence of \( \Delta(r) \) is determined self-consistently from the gap equation

\[
\Delta(r) = \pi N_0 g_0 T \sum_{\omega_n} \int \frac{d\theta}{2\pi} f(r, \theta, i\omega_n) V_x(\theta),
\]

where \( N_0 \) is the density of states per spin of normal metal at the Fermi level, \( g_0 \) is the coupling constant, and \( V_x \) represents attractive electron-electron interactions with \( x = p \)-wave or \( d \)-wave indicating pairing symmetries. The interaction kernel \( V_x \) depends upon pairing symmetries as

\[
V_x(\theta) = \begin{cases} 2\cos \theta & \text{for } x = p \text{-wave} \\ 2\sin(2\theta) & \text{for } x = d \text{-wave} \end{cases}.
\]
FIG. 2. Local susceptibilities of the small superconducting disks. The results of a $d$-wave and those of a $p$-wave superconductor with a clean surface (i.e., $w = 0$, $\xi/\ell = 0$) are presented in (a) and (b), respectively. The results of a $d$-wave and those of a $p$-wave superconductor with a rough surface ($w = 3\xi_0$, $\xi_0/\ell = 1.0$) are demonstrated in (c) and (d), respectively. The parameters used in the simulation are $R = 10\xi_0$, $\lambda_L = 5\xi_0$, $\omega_c = 10\Delta_0$, $H^\text{ext} = 0.01H_{c2}$ and $T = 0.1T_c$.

The constant $N_0g_0$ is determined by

$$
(N_0g_0)^{-1} = \ln\left(\frac{T}{T_c}\right) + \sum_{0 < \omega_n < \omega_c/2\pi T} \frac{1}{n + 1/2},
$$

with $T_c$ and $\omega_c$ being the transition temperature and the cut-off energy, respectively.

An electric current is represented by

$$
\bar{j}(r) = \frac{\pi ev_F N_0}{2l} T \sum_{\omega_n} \int d\theta \frac{1}{2\pi} \text{Tr} [\tilde{T}_3 k \hat{g}(r, \theta, \omega_n)],
$$

with $\tilde{T}_3 = \text{diag}[\sigma_0, -\sigma_0]$. From Eq. (12) and the Maxwell equation $\nabla \times \mathbf{H}(r) = 4\pi \overline{j}(r)$, we obtain spatial profiles of a vector potential $\mathbf{A}(r)$ and a local magnetic field $\mathbf{H}(r)$. A local magnetic susceptibility is defined by

$$
\chi(r) = \frac{1}{4\pi} \frac{H(r) - H^\text{ext}}{H^\text{ext}},
$$

where $H^\text{ext}$ is an amplitude of external magnetic field applied in the $z$ direction. By integrating the local susceptibility, we obtain a susceptibility of a whole disk $X$ as

$$
X = \frac{1}{\pi R^2} \int_{|r| \leq R} dr \chi(r).
$$

To solve the Eilenberger equation Eq. (1) in a disk geometry, we use a Riccati parametrization and a numerical method discussed in Ref. 29. Using the parametrization, the Eilenberger equation can be separated into two Riccati type differential equations. When we solve the Riccati type equation along a long enough quasiclassical trajectory, solutions of the equation do not depend on initial conditions. In this paper, the length of classical trajectories is more than 30 times of the coherence length. Solving the Eilenberger equation and the Maxwell equation, we determine the pair potential, the vector potential, and the self-energy self-consistently with one another.

III. RESULTS

Throughout this paper, we fix several parameters as $R = 10\xi_0$, $\lambda_L = 5\xi_0$, $\omega_c = 10\Delta_0$, and $H^\text{ext} = 0.01H_{c2}$. Here $H_{c2} = \hbar c/|e|\xi_0^2$ is the second critical magnetic field, $\Delta_0$ is the amplitude of pair potential at the zero temperature, and $\xi_0 = \hbar v_F/2\pi T_c$ is the coherence length. All lengths are measured in units of $\xi_0$. The current density is normalized to $J_0 = \hbar c^2/4\pi|e|\xi_0^3$. The characteristic length scale of the Maxwell equation is the penetration depth defined as $\lambda_L = (4\pi|e|^2/\hbar c^2)^{-1/2}$ and is a parameter in numerical simulations. Strength of the disorder
the $d$-wave pair potential. The magnetic susceptibility is positive (paramagnetic) near four surfaces in the $x$ and the $y$ directions. In Fig. 3(a), we show a spatial profile of the local susceptibility of Fig. 2(a) along the $x$ axis at $y = 0$. The paramagnetic response at the four surfaces is well explained by the appearance of odd-frequency Cooper pairs. In this paper, we analyze the frequency symmetries of Cooper pairs by decomposing pairing functions into a series of Fourier components. In a $d$-wave disk, the anomalous Green function is described by two components

$$\hat{f}(r, \theta, i\omega_n) = [f_{ep}(r, \theta, i\omega_n) + f_{op}(r, \theta, i\omega_n)] \hat{\sigma}_2,$$

(15)

where $f_{ep}$ is an even-parity ($d$-wave) function representing the principal pairing correlation and $f_{op}$ is an odd-parity function representing the induced pairing component at a surface. To satisfy Eq. (15), $f_{op}$ must be an odd function of $\omega_n$. We decompose pair functions $f(r, \theta, i\omega_n)$ as

$$P_l(r, i\omega_n) = 2\sqrt{C_l^2 + S_l^2},$$

(16)

$$S_l = \int \frac{d\theta}{2\pi} \text{Re} [f(r, \theta, i\omega_n)] \sin (l\theta),$$

(17)

$$C_l = \int \frac{d\theta}{2\pi} \text{Re} [f(r, \theta, i\omega_n)] \cos (l\theta),$$

(18)

where $l = 0, 1, 2$ and 3 correspond to $s$-, $p$-, $d$- and $f$-wave orbital functions, respectively. In the presence of a magnetic field, the imaginary part of $f(r, \theta, i\omega_n)$ is induced by the vector potential as analytically shown in Appendix. We focus only on the real part of $f$ to analyze pairing symmetries. Figure 4(a) indicates the spatial profile of $P_l(x, i\omega_0)$ at the lowest Matsubara frequency as a function of $x$ at $y = 0$. The pairing functions of induced Cooper pairs have $p$- and $f$-wave symmetries and their amplitudes localize near the surface. Such odd-frequency Cooper pairs shows the paramagnetic response to a magnetic field. The surface also generates spin-singlet $s$-wave correlation. Its amplitude, however, is too small to confirm at a scale of plot in Fig. 4(a). Figure 5(a) shows the spatial distribution of electric current on a $d$-wave disk. The diamagnetic current flows at the central region because of the usual Meissner effect. Near the surfaces in the $x$ and $y$ directions, however, the current flows into the opposite direction to the Meissner current. Therefore a small $d$-wave superconductor can be paramagnetic due to the induced odd-frequency Cooper pairs at its surface.

A spin-triplet $p$-wave disk also indicates the similar paramagnetic effect as shown in Figs. 2(b), 3(b) and 4(b). The results are four-fold symmetric reflecting the $p$-wave superconducting pair potential. The paramagnetic effect can be seen near the surface in the $x$ direction because of induced odd-frequency Cooper pairs. The anomalous Green function is represented by Eq. (15) with replacing $\hat{\sigma}_2$ by $-i\hat{\sigma}_1$. In the $p$-wave case, $f_{ep}$ represents induced pairing correlations and is an odd function of $\omega_n$. As shown in Fig. 4(b), $f_{ep}$ mainly consists of $s$- and $d$-wave pairing correlations.
In the end of this subsection, we summarize an important difference between the paramagnetic effect of a \textit{d}-wave superconductor and that of a \textit{p}-wave one. In a \textit{d}-wave disk, surface odd-frequency Cooper pairs have a \textit{p}- or \textit{f}-wave orbital symmetry\textsuperscript{15,18}. In a \textit{p}-wave disk, on the other hand, \textit{s}- or \textit{d}-wave odd-frequency Cooper pairs are responsible for the paramagnetic effect\textsuperscript{15,18}. In the next subsection, we will show that the paramagnetic response of a disk with rough surface depends sensitively on the orbital symmetry of induced odd-frequency pairs at the surface.

\section*{B. Disks with a rough surface}

Next, we discuss effects of the surface roughness on the magnetic response of a small superconductor. The calculated results of the local susceptibility for a \textit{d}-wave superconducting disk with rough surface are shown in Fig. 2(c), where we choose $\xi_0/\ell = 1.0$. Comparing Fig. 2(a) with (c), we can find that the surface roughness completely suppresses the paramagnetic response at the four surfaces in a \textit{d}-wave disk. The central region of the disk with the rough surface recovers the usual diamagnetic response. Such effect is demonstrated more clearly in a spatial profile of local susceptibilities at $y = 0$ in Fig. 3, where the shadowed area indicates the disordered region. Spatial profiles of decomposed pairing functions are shown in Fig. 4(c). In the disordered region, a \textit{d}-wave pairing function $P_d$ is drastically suppressed due to impurity scatterings. The disordered region can be considered as a diffusive normal metal because the spatial profile of order parameter is proportional to $P_d$. Odd-frequency Cooper pairs are also fragile in the presence of surface roughness because they have \textit{p}- or \textit{f}-wave orbital symmetry. Therefore, both the paramagnetic current and the diamagnetic one disappear in the disordered region as shown in Fig. 5(c). The magnetic property of a disk is determined by that at the clean central region where even-frequency \textit{d}-wave Cooper pairs stay and contribute to the diamagnetic response. We conclude that the paramagnetic effect in a \textit{d}-wave disk is fragile in the presence of surface roughness because odd-frequency pairs have a \textit{p}-wave or a \textit{f}-wave orbital symmetry.

A \textit{p}-wave disk indicates qualitatively different magnetic response from a \textit{d}-wave one. The local susceptibility of a \textit{p}-wave disk with rough surface is shown in Fig. 2(d) and Fig. 3(b) with a broken line. Although the surface is rough enough, a \textit{p}-wave superconducting disk still show the strong paramagnetic response. The peak of the $\chi$ in Fig. 3(b) shifts to inside of the disk in the presence of surface roughness. This suggests that the Andreev bound states appear at a boundary between the clean central region and the disordered surface region. Such Andreev bound states always accompany the paramagnetic odd-frequency Cooper pairs. In addition to this, the paramagnetic response in Fig. 3(b) suggests the penetration of odd-frequency Cooper pairs into the surface disordered region. The spatial profiles of the electric current in Fig. 3(d) shows that the paramagnetic current flows not only in the clean region but also in the disordered one. Odd-frequency pairs in a \textit{p}-wave disk survive even in the presence of surface roughness because they have \textit{s}-wave orbital symmetry. Therefore, the paramagnetic effect in a \textit{p}-wave disk is robust against the surface roughness.

\section*{C. Temperature dependence}

Here we discuss the magnetic susceptibility of a whole disk which is a measurable value in experiments. The disk susceptibility in a \textit{d}-wave superconductor and that in a \textit{p}-wave one are plotted as a function of temperature in Fig. 3(a) and (b), respectively. We present the results for several choices of the disorder $\xi_0/\ell$. In simulation, we first calculate the pair potential and the vector potential self-consistently at a temperature just below $T_c$, under an external magnetic field $H^\text{ext} = 0.01H_c^2$. Then the temperature is decreased with keeping $H^\text{ext}$ unchanged. Thus the results in Fig. 3 correspond to the susceptibility in the field cool process in experiments. In the clean
limit ($\xi_0/\ell = 0.0$), both a $d$-wave disk and a $p$-wave one show the usual diamagnetic response just below $T_c$. With decreasing the temperature, however, the sign of susceptibility changes around $T = T_p \sim 0.3 T_c$ for both cases. Here, we define $T_p$ as a diamagnetic-paramagnetic crossover temperature. Blow $T_p$, superconducting disks show the anomalous paramagnetic response. The paramagnetic effect is stronger in lower temperature because odd-frequency Cooper pairs energetically localize at the Fermi level.

In a $d$-wave disk, the reentrance is slightly suppressed in the presence of the moderate surface roughness with $\xi_0/\ell = 0.1$ as shown in Fig. 6(a). When we increase the degree of the roughness further, the paramagnetic response gradually becomes weaker. At $\xi_0/\ell = 0.5$, the response is diamagnetic and the susceptibility recovers the monotonic temperature dependence which is usually observed in large enough superconductors in experiments. In the $d$-wave superconductors, the specular Andreev reflection is necessary for forming the surface bound states at the zero-energy and for appearing odd-frequency pairs. In other words, odd-frequency pairs have $p$- or $f$-wave orbital symmetry. Therefore the rough surface brakes odd-frequency pairs and suppresses the paramagnetic response. This conclusion is totally consistent with the experiment,22 where temperature dependences of the pair density on a high-$T_c$ superconducting film show a small reentrant behavior at low temperature. But the total pair density remains positive. Actually, the experimental data are very similar to the results for $\xi_0/\ell = 0.2$ in Fig. 6(a). The experimental results can be interpreted as an appearance of a small amount of odd-frequency pairs. In the experiment, the surface roughness may partially breaks odd-frequency pairs because a number of the internal surfaces are introduced by the heavy ion irradiation.

On the contrary to a $d$-wave disks, the susceptibleness of a $p$-wave disk $X(T)$ shows the reentrance and the crossover to the paramagnetic phase at low temperature for all $\xi_0/\ell$ as shown in Fig. 6(b). It has been pointed out that the surface Andreev bound states of a $p$-wave superconductor are robust under potential disorder because of the pure chiral property of surface bound states.31 In other words, odd-frequency pairs accompanied by the Andreev bound states have $s$-wave orbital symmetry.15,18 Since $s$-wave pairs are robust under the disordered potential, the paramagnetic effect of the $p$-wave superconductors persists even in the presence of surface roughness. We conclude that the robust paramagnetic response in a small size sample is a unique property of spin-triplet $p$-wave superconductors. Such property would enable us to identify the spin-triplet $p$-wave superconductivity in experiments.

IV. STABILITY OF PARAMAGNETIC SUPERCONDUCTING STATES

Generally speaking, a superconducting phase is more stable than a normal one as far as a superconductor is diamagnetic and homogeneous. Therefore a homogeneous paramagnetic superconducting phase is usually unstable. The calculated results in Sec. III, however, show that the paramagnetic phase on a small superconducting disk is spatially inhomogeneous. In such situation, it would be worthy to check if the paramagnetic phase is a stable state at a free-energy minimum or a metastable state corresponding to a free-energy local minimum. In this section, we discuss the stability of paramagnetic phase in small unconventional superconductors by calculating the free-energy in clean superconducting disks.

The free-energy is calculated from the quasiclassical Green functions:

$$F_S - F_N = \int d\mathbf{r} \mathcal{F}(\mathbf{r}),$$

$$\mathcal{F}(\mathbf{r}) = \mathcal{F}_\Delta(\mathbf{r}) + \mathcal{F}_H(\mathbf{r}),$$

$$\mathcal{F}_H(\mathbf{r}) = \frac{\{H(\mathbf{r}) - H^\text{ext}\}^2}{8\pi},$$

$$\mathcal{F}_\Delta(\mathbf{r}) = \mathcal{F}_f(\mathbf{r}) + \mathcal{F}_g(\mathbf{r}),$$

$$\mathcal{F}_f(\mathbf{r}) = \pi N_0 \int \frac{d\omega}{2\pi} T \sum_{\omega_n} \Delta^*(\mathbf{r}, \theta) f(\mathbf{r}, \theta, i\omega_n),$$

$$\mathcal{F}_g(\mathbf{r}) = 4\pi N_0 \int \frac{d\omega}{2\pi} T \sum_{\omega_n > 0} \omega_n \times \int_{\omega_n}^{\omega_{cz}} d\omega \text{Re} \{g(\mathbf{r}, \theta, i\omega) - 1\},$$

where $\mathcal{F}_\Delta(\mathbf{r})$ is the condensation energy density of electron system and $\mathcal{F}_H(\mathbf{r})$ is the energy density of a magnetic field. We introduce an additional energy cut-off $\omega_{cz}$ to evaluate the integration in Eq. 24. In this paper, we set $\omega_{cz} = 400\Delta_0$ so that $\int d\mathbf{r} \mathcal{F}(\mathbf{r})$ reaches to a converged value. The free-energy densities normalized to
The free-energy density varies gradually from the line with $\alpha = 0$ due to the four-fold symmetry. The argument above is also valid also for a $d$-wave disk. In Appendix, we present analytical expression of difference between free-energy at $H^\text{ext} = 0$ and that at $H^\text{ext} = 0$. The results show that $\mathcal{F}_\Delta$ near surfaces for $H^\text{ext} \neq 0$ is lower than that for $H^\text{ext} = 0$. Odd-frequency pairing state in the presence of electric currents is more stable than that in the absence of electric currents. This energetic property explains the paramagnetic property of odd-frequency Cooper pairs. The argument above is valid also for a $d$-wave disk. In Appendix, we present analytical expression of difference between free-energy at $H^\text{ext} = 0$ and that at $H^\text{ext} = 0$. The results show that a magnetic field decreases the free-energy at low temperature because odd-frequency Cooper pairs have the paramagnetic property.

V. CONCLUSION

We have theoretically studied effects of surface roughness on the anomalous paramagnetic response of small...
unconventional superconducting disks by using the quasiclassical Green function method. We conclude that the paramagnetic property of $p$-wave superconductors is robust under the surface roughness because the $p$-wave superconductors host the $s$-wave odd-frequency Cooper pairs at their surface. On the other hand, the paramagnetic property in $d$-wave superconductor is fragile in the presence of the surface roughness. In this case, the odd-frequency pairs at the surface have $p$-wave orbital symmetry. We have also confirmed that the paramagnetic superconducting phase are more stable than the normal state by calculating the free-energy.

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**Appendix: Analysis in a semi-infinite $p_x$-wave superconductor**

In a semi-infinite superconductor in two-dimension, it is possible to obtain analytic expression of Green functions in the clean limit. The evaluation of an electric current and free-energy by using analytical expressions would be helpful to understand numerical results in the text.

We assume that a superconductor occupies $x \geq 0$ and uniform in the $y$ direction. An magnetic field applied in the $z$ direction and its vector potential is given by $\mathbf{A} = A(x)\hat{y}$. The Eilenberger equation in $2 \times 2$ Nambu space reads,

$$i v_F \mathbf{k} \cdot \nabla \hat{g} + [\hat{H}, \hat{g}] = 0,$$

(A.1)

$$\hat{g}(x, \mathbf{k}, i\omega_n) = \left[ \begin{array}{cc} g & f \\ \mathbf{f}^T - g & (x, k, i\omega_n) \end{array} \right],$$

(A.2)

$$\hat{H} = \left[ \begin{array}{cc} i\omega_n + ev_F \mathbf{k} \cdot \mathbf{A} & i\Delta(x, \mathbf{k}) \\ is\Delta(x, \mathbf{k}) & -i\omega_n - ev_F \mathbf{k} \cdot \mathbf{A} \end{array} \right],$$

(A.3)

$$s_p = \begin{cases} 1 & \text{even-parity} \\ -1 & \text{odd-parity} \end{cases}$$

(A.4)

where a factor $s_p$ depends on the parity of order parameter and the Green functions satisfy $g^2 + s_p f = 1$. The Green functions can be expanded with respect to the vector potential as

$$g = g^{(0)} + (-iev_F \mathbf{k} \cdot \mathbf{A})\partial_{\omega_n} g^{(0)} + \frac{1}{2}(-iev_F \mathbf{k} \cdot \mathbf{A})^2 \partial_{\omega_n}^2 g^{(0)} + \cdots,$$

(A.5)

$$f = f^{(0)} + (-iev_F \mathbf{k} \cdot \mathbf{A})\partial_{\omega_n} f^{(0)} + \frac{1}{2}(-iev_F \mathbf{k} \cdot \mathbf{A})^2 \partial_{\omega_n}^2 f^{(0)} + \cdots,$$

(A.6)

because a vector potential shifts the Matsubara frequency, where $g^{(0)}$ and $f^{(0)}$ are the Green function in...
the absence of a vector potential. In what follows, we omit \( "(0)" \) from the Green function for simplicity. We note in Eq. (A.6) that parity and frequency symmetry of the second term on the right-hand side are opposite to those of the first term because \( k \) is a odd-parity function and \( \partial_{\omega_n} f \) changes a frequency symmetry. The imaginary part of an anomalous Green function represents a pairing correlation deformed by a vector potential.

In the case of a \( p_x \)-wave superconductor, it is possible to obtain a reasonable solution of the Eilenberger equation at \( A = 0 \). When we assume the spatial dependence of the pair potential as

\[
\Delta(x, \theta) = \Delta(\theta) \tanh \left( \frac{x}{\xi} \right),
\]

with \( \xi = v_F/\Delta_0 \), the Green functions are represented by

\[
g(x, \theta, i\omega_n) = \frac{\omega_n}{\Omega} + \frac{\Delta^2(\theta)}{2\omega_n\Omega} \cosh^{-2} \left( \frac{x}{\xi} \right),
\]

\[
f_p(x, \theta, i\omega_n) = \frac{\Delta(\theta)}{\Omega} \tanh \left( \frac{x}{\xi} \right),
\]

\[
f_1(x, \theta, i\omega_n) = -\frac{\Delta^2(\theta)}{2\omega_n\Omega} \cosh^{-2} \left( \frac{x}{\xi} \right),
\]

where \( \Omega = \left[ \omega_0^2 + \Delta^2(\theta) \right]^{1/2} \), and \( \Delta(\theta) = \Delta_0 \cos(\theta) \). The Green function \( f_p \) represents the principal pairing correlation in the bulk state, whereas \( f_1 \) represents the pairing correlation induced by a surface at \( x = 0 \). They are calculated from the anomalous Green function as

\[
f_p(x, \theta, i\omega_n) = \frac{1}{2} \left( f + s_p(x, \theta, i\omega_n) \right),
\]

\[
f_1(x, \theta, i\omega_n) = \frac{1}{2} \left( f - s_p(x, \theta, i\omega_n) \right).
\]

At the deep inside of the superconductor (i.e., \( x \gg \xi \)), we obtain \( f_p = f \) and \( f_1 = 0 \).

1. Current density

\( \bar{J} \) From an expression of electric current in Eq. (12), we define a linear response function \( R_{\mu, \nu} \) by

\[
j_{\mu}(r) = -\frac{e^2}{m} R_{\mu, \nu} A_{\nu},
\]

\[
\frac{R_{\mu, \nu}}{n_e} = 4\pi T \sum_{\omega_n} \int \frac{d\theta}{2\pi} k_{\mu} k_{\nu} \partial_{\omega_n} g(r, \theta, i\omega_n).
\]

with \( k = (\cos \theta, \sin \theta) \) and \( n_e = v_F^2 N_0 m \) being an electron density in two-dimension. The diagonal elements of the response function \( R_{\mu, \mu} \) correspond to so called pair density. In the present situation, by substituting Eq. (A.8) into Eq. (A.14), we obtain

\[
\frac{R_{\mu, \nu}}{n_e} = 1 - \kappa_1 \frac{\Delta_0}{\omega_0} \cosh^{-2} \left( \frac{x}{\xi} \right),
\]

\[
\kappa_1 = \int \frac{d\theta}{2\pi} \sin^2(\theta) |\cos(\theta)| = \frac{2}{3\pi},
\]

where \( \omega_0 = \pi T \) is a low energy cut-off in the Matubara summation. Using the normalization condition, the integrand in Eq. (A.14) can be represented in an alternative way,

\[
\partial_{\omega_n} g = \left[ -f_p \partial_{\omega_n} f_p + f_1 \partial_{\omega_n} f_1 \right] / g.
\]

It is possible to confirm that \( -f_p \partial_{\omega_n} f_p / g \) corresponds to the first term in Eq. (A.15), whereas \( f_1 \partial_{\omega_n} f_1 / g \) contribute to the second term. In this way, we can confirm that induced odd-frequency Cooper pairs indicate paramagnetic response to an external magnetic field. Eq. (A.15) suggests that the paramagnetic response is stronger in lower temperature.

2. Free-energy density

Substituting Eqs. (A.8)-(A.10) into Eqs. (22)-(24), we find that a free-energy density at \( A = 0 \)

\[
F_f = N_0 \kappa_2 \Delta_0^2 \log \left( \frac{2\omega_0}{\Delta_0} \right) \tanh^2 \left( \frac{x}{\xi} \right),
\]

\[
F_g = -N_0 \kappa_2 \Delta_0^2 \log \left( \frac{2\omega_0}{\Delta_0} \right) \tanh^2 \left( \frac{x}{\xi} \right)
\]

\[
+ N_0 \Delta_0^2 \kappa_2 \left[ \cosh^{-2} \left( \frac{x}{\xi} \right) - \frac{1}{2} \right],
\]

\[
\kappa_2 = \int \frac{d\theta}{2\pi} \cos^2(\theta) = \frac{1}{2}.
\]

As a result, we obtain

\[
F_\Delta = N_0 \Delta_0^2 \kappa_2 \left[ \cosh^{-2} \left( \frac{x}{\xi} \right) - \frac{1}{2} \right].
\]

The free-energy density becomes positive at \( x = 0 \) due to appearance of odd-frequency pairs.

Contribution of a magnetic field to the free-energy can be evaluated by applying the expansion in Eqs. (A.8)-(A.10) onto Eqs. (A.11)-(A.14). The free energy density becomes positive at \( x = 0 \) due to appearance of odd-frequency pairs.

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