Inhomogeneous Stripe Phase Revisited for Surface Superconductivity

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We consider 2D surface superconductivity in high magnetic fields parallel to the surface. We demonstrate that the spin-orbit interaction at the surface changes the properties of the inhomogeneous superconducting Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) state that develops above fields given by the paramagnetic criterion. Strong spin-orbit interaction significantly *broadens* the range of existence of the LOFF phase, which takes the form of periodic superconducting stripes running along the field direction on the surface, leading to the anisotropy of its properties. Our results provide a tool for studying surface superconductivity as a function of doping.

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There has been renewed experimental and theoretical interest in the properties of metallic states localized at a surface and surface superconductivity (SSC). Surface states (a.k.a. Tamm's levels) are well known from the physics of semiconductors. Numerous angle-resolved photoemission spectroscopy (ARPES) [1,2] data and STM studies of Friedel oscillations [3] have now proven the existence of 2D metallic bands even at surfaces of metals. The bands are well separated from the bulk and possess clear-cut 2D Fermi surfaces. For heavy enough elements, such Fermi surfaces are split further by strong spin-orbit (SO) interactions. For example, SO energy for electrons at the Fermi level for Au is estimated at 0.1 eV [1], while for Li-doped surfaces of Mo and W its value increases up to 0.13 and 0.5 eV, correspondingly [2].

Islands of a surface superconducting phase were also observed for the surface-doped tungsten bronzes, WO_3:Na, at $T_c = 91.5$ K [4]. The ARPES results mentioned above suggest that SSC may actually be a rather widespread phenomenon.

It is noteworthy that the bulk WO_3 is an insulator at low doping [5]. SSC with high T_c was also induced by both electronic and hole doping of films of the prototype cuprate material, $CaCuO_2$ [6], in the field-effect transistor (FET) geometry.

All this makes us believe that the search for SSC emerges as a new and important development in studies of the properties of surfaces, especially their metallic properties. As for the SSC itself, its mechanisms are unknown and may have nothing to do with the ones in the bulk. Of a special challenge is the possibility of superconductivity at surfaces of ordinary metals, such as Cu [3] or lithium-doped Mo and W [2], and the influence of substrate on superconductivity in thin films. Thus, it is necessary to return to a more careful investigation of the low temperature properties of surfaces with absorbed atoms and the role of adsorbed atoms as dopants of carriers into the metallic surface zone (e.g., see Ref. [2]) like it was discovered for WO_3:Na [4].

The major challenge to such a program lies in discerning SSC and studying its properties. Even for insulators where doping by FET, ideally, provides an effective control on surface properties, experimental tools to probe SSC are limited in numbers. The Meissner effect due to surface superconducting islands would probably never produce a bulk screening. Thermodynamical probes are also difficult because of the smallness of contributions from surface layers of atomic thicknesses. So far, surface superconductivity has been detected only by measuring resistivity dependence on temperature in the perpendicular-to-plane magnetic fields.

In what follows, we focus on destroying SSC by magnetic fields applied *parallel* to the surface. In high enough fields, one should expect the appearance of a 2D version of inhomogeneous superconducting state known as Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) phase [7]. It was also shown that two-dimensionality broadens the region of the LOFF state on the B-T phase diagram [8]. Motivated by experimental findings mentioned above, we investigate the peculiarities introduced into this phenomenon by the SO effect or non-s-wave pairing. Ideally, FET would be a very effective doping process. Different levels of doping would result in different T_c-s. Our results for SSC in parallel magnetic fields are expressed in terms of this T_c, providing the tool for comparing theoretical predictions with experiments by controlling the doping level. Theoretically, there is no long-range order in a 2D superconductor. However, correlations are destroyed on the exponentially large spatial scale, $R \sim \xi_0 \exp(E_F/T)$, which would exceed the size of the film.

We employ below the weak-coupling BCS-like scheme by assuming that electrons interact via a weak short-ranged interaction, $U(\boldsymbol{r}, \boldsymbol{r}')$. Then $T_c \ll \epsilon_F$, and only a narrow vicinity of the Fermi surface is involved. Thus, the interaction in the momentum representation can be taken in the form:

$$U(\boldsymbol{p}, \boldsymbol{p}') = \sum_l U_l \chi_l(\boldsymbol{p}) \chi_l(\boldsymbol{p}'), \tag{1}$$

where $\boldsymbol{p}$, $\boldsymbol{p}'$ lie on the Fermi surface; the angular dependence is expressed through a complete set of basis functions $\chi_l(\boldsymbol{p})$ (index l enumerates different representations, as in expansions over the spherical functions in a 3D isotropic model). Superconducting order parameter, the "gap," $\hat{\Delta}_{\alpha\beta}(\boldsymbol{p})$ is defined by the equation:

$$\hat{\Delta}_{\alpha\beta}(\boldsymbol{p}) = |U_l|\chi_l(\boldsymbol{p}) \int \frac{d^3p'}{(2\pi)^3}\chi_l(\boldsymbol{p}'p') \times \left[T\sum_{\omega_n} F_{\alpha\beta}(\boldsymbol{p}'; i\omega_n)\right], \quad (2)$$

where $F_{\alpha\beta}(\boldsymbol{p}'; i\omega_n)$ stands for the Fourier component of Gor'kov anomalous function:

$$\hat{F}_{\alpha\beta}(\boldsymbol{r}-\boldsymbol{r}', \tau-\tau') = -\langle T_\tau[\hat{\Psi}_\alpha(\boldsymbol{r},\tau)\hat{\Psi}_\beta(\boldsymbol{r}',\tau')]\rangle, \quad (3)$$

and $U_l < 0$ is a constant in Eq. (1) corresponding to the selected pairing channel. When the field operators for electrons are rewritten in momentum space, $\hat{\Psi}_\alpha(\boldsymbol{r},\tau) = \sum_{\boldsymbol{p}} \hat{\Psi}_\alpha(\boldsymbol{p},\tau)e^{i\boldsymbol{p}\cdot\boldsymbol{r}}$, Eq. (2) becomes:

$$\hat{\Delta}_{\alpha\beta}(\boldsymbol{p}) = |U_l|\chi_l(\boldsymbol{p}) \int \frac{d^3p'}{(2\pi)^3}\chi_l(\boldsymbol{p}')\langle\hat{\Psi}_\alpha(\boldsymbol{p}',\tau)\hat{\Psi}_\beta(-\boldsymbol{p}',\tau)\rangle. \quad (4)$$

The two operators inside brackets in Eq. (4) anticommute. In the presence of the center of inversion (CI) the behavior of $\chi_l(\boldsymbol{p}')$ at $\boldsymbol{p}' \to -\boldsymbol{p}'$ alone determines the symmetry of $\hat{\Delta}_{\alpha\beta}(\boldsymbol{p})$ [even (singlet) vs odd (triplet) parity pairing]. For an s or d pairing the order parameter below T_c has the form:

$$\hat{\Delta}_{\alpha\beta}(\boldsymbol{p};\boldsymbol{q}) = \Delta(\boldsymbol{q},T)(i\sigma_y)_{\alpha\beta}\chi_l(\boldsymbol{p}), \quad (5)$$

where the momentum $\boldsymbol{q}$ stands for the spatial dependence of the gap amplitude, $\Delta(\boldsymbol{q},T)$.

Surface always breaks the CI symmetry due to the difference between the "top" and the "bottom." The direction bulk-to-surface determines $\boldsymbol{n}$, a unit vector normal to the surface. *Qualitative* changes in the surface electronic spectrum come about from the well-known Rashba term [9]:

$$\hat{h}_{\rm SO} = \alpha(\boldsymbol{\sigma}\times\boldsymbol{p}\cdot\boldsymbol{n}), \quad (6)$$

which specifies SO interactions at a surface. Equation (6) lifts the twofold spin degeneracy for band electrons. The electron spectrum now consists of two branches with two Fermi surfaces:

$$\epsilon_\pm(\boldsymbol{p}) = v_F(p - p_{F\pm}); \qquad p_{F\pm} = p_F\left(1 \pm \frac{\alpha}{v_F}\right). \quad (7)$$

Even though SO splitting, $2\alpha p_F$, may be on a scale of tenths of eV [1,2], we assume that $2\alpha p_F \ll \epsilon_F$. Below we speak of a strong or weak SO meaning the relative values of $2\alpha p_F$ and T_c.

Some theory issues regarding superconductivity without CI due to the presence of the SO term Eq. (6) were first considered in Ref. [10] and more recently in Ref. [11]. The Gor'kov function (in the momentum representation), $F_{\alpha\beta}(\boldsymbol{p},+0) = -\langle\hat{\Psi}_\alpha(\boldsymbol{p})\hat{\Psi}_\beta(-\boldsymbol{p})\rangle$, that stands under the integral in Eq. (4) represents the wave function for Cooper pairs in the condensate. In the presence of CI symmetry, the latter can be classified according to the parity:

$$F_{\alpha\beta}(\boldsymbol{p};+0) = \begin{cases} i(\sigma_y)_{\alpha\beta}f(\boldsymbol{p}); & [f(\boldsymbol{p})\ \text{even}] \\ i[(\boldsymbol{d}(\boldsymbol{p})\cdot\hat{\boldsymbol{\sigma}})\hat{\sigma}_y]_{\alpha\beta}; & [\boldsymbol{d}(\boldsymbol{p})\ \text{odd}]. \end{cases} \quad (8)$$

With CI broken by nonzero SO term Eq. (6), the pairing wave function becomes a mixture of even and odd terms. It is important to realize that while this mixing changes physical properties of the SC phase, the gap order parameter, $\hat{\Delta}_{\alpha\beta}(\boldsymbol{p};\boldsymbol{q})$, preserves its singlet form Eq. (5). For instance, s pairing indeed induces the nonzero triplet component Eq. (8), as shown in Refs. [10,11]. However, the latter does not automatically generate a "triplet" gap, $\hat{\Delta}^t_{\alpha\beta}(\boldsymbol{p};\boldsymbol{q})$. Indeed, rewriting the integration over $\boldsymbol{p}'$ in Eq. (4) as $d^3p' \to dS'_F d\xi$, where $\xi = v_F(p - p_F)$, we notice that the triplet F component is odd in particle-hole transformation, $\xi \to -\xi$, and, hence, the integrals of the form Eq. (4) would give only small terms of order $(\alpha p_F/\epsilon_F) \ll 1$. In other words, while SO interaction may significantly change spin structure of the normal and anomalous Green functions, the gap, $\hat{\Delta}_{\alpha\beta}(\boldsymbol{p},\boldsymbol{q})$, Eq. (2) preserves its usual form Eq. (5) with $\chi_l(\boldsymbol{p}) = \text{const}$ for isotropic pairing and $\chi_l(\boldsymbol{p}) \propto (p_x^2 - p_y^2)$ for the d-wave pairing.

We have calculated T_c numerically for the 2D superconductor as a function of magnetic field and spin-orbit interaction. The result is shown in Fig. 1 for the magnetic field strictly parallel to the surface (to exclude diamagnetic currents) [8].

A 1st order phase transition was initially expected between superconducting and normal states, defined by comparing their free energies: $F_s(T) = F_n - \chi_N\frac{B^2}{2}$, which would determine the so-called paramagnetic critical field, $H_{\rm par}$ [12] (χ_N—the spin susceptibility in the normal phase). The transition from normal to superconducting state is actually (at lower temperatures) a second order transition into the LOFF state. The details of the phase diagram in the vicinity of the $H_{\rm par}$ were studied numerically in Ref. [13]. The LOFF phase boundary is determined by Eq. (2) or Eq. (4), linearized in $\Delta_{\alpha\beta}(\boldsymbol{q})$ at an extremal $\boldsymbol{q}$. The corresponding expression for anomalous function linear in $\Delta_{\alpha\beta}(\boldsymbol{q})$ is obtained by solving the proper Gor'kov equations:

$$F_{\alpha\beta}(\boldsymbol{p},\boldsymbol{q};i\omega_n) = -\hat{G}^{(0)}_{\alpha\nu}(\boldsymbol{p};i\omega_n)\hat{\Delta}_{\rho\nu}(\boldsymbol{p},\boldsymbol{q}) \times \hat{G}^{(0)}_{\beta\rho}(-\boldsymbol{p}p+\boldsymbol{q};-i\omega_n), \quad (9)$$

where $\hat{G}^{(0)}_{\alpha\beta}(\boldsymbol{p};i\omega_n)$ is the normal state Green function at nonzero $\hat{h}_{\rm SO}(\boldsymbol{p})$ of Eq. (6), together with the Zeeman term, $\mu_B(\boldsymbol{\sigma}\cdot\boldsymbol{B})$:

$$[i\omega_n - \xi - \hat{h}_{\rm SO}(\boldsymbol{p}) - \mu_B\hat{\boldsymbol{\sigma}}\boldsymbol{B}]\hat{G}^{(0)}_{\alpha\beta}(\boldsymbol{p};i\omega_n) = \hat{1}. \quad (10)$$

The spin Hamiltonian on the left side, $\hat{H} = \hat{h}_{\rm SO}(\boldsymbol{p}) + \mu_B\hat{\boldsymbol{\sigma}}\boldsymbol{B}$, may be easily diagonalized:

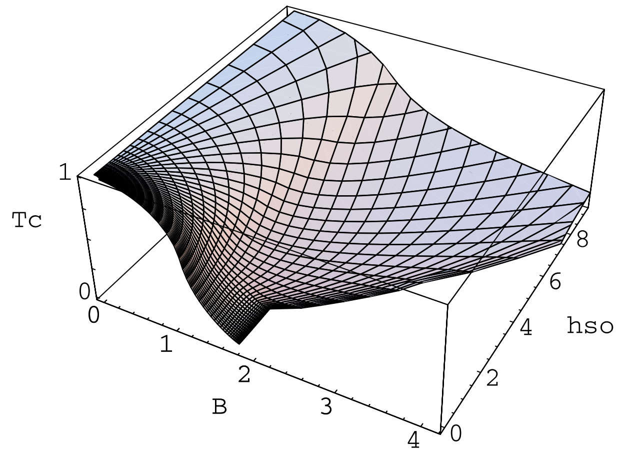

FIG. 1 (color). Calculated T_c as a function of magnetic field and spin-orbit interaction αp_F for a 2D surface superconductor. All values are in units of T_{c0}, T_c for the 2D superconductor in the absence of magnetic field. Thus, $B \to \mu_B B/T_{c0}$, $h_{SO} \to \alpha p_F/T_{c0}$, and $T_c \to T_c/T_{c0}$.

$$\tilde{\epsilon}_\lambda(p) - \xi = -\lambda\sqrt{\alpha^2 p_F^2 + 2\alpha p_{Fy}\mu_B B + (\mu_B B)^2} \equiv -\lambda\tilde{\epsilon}(p) \quad (11)$$

($\lambda = \pm 1$ for the two branches, p_{Fy} is the y-axis projection of the Fermi momentum, and $B \parallel x$) with the eigenfunctions, spinors $\eta^\lambda(\boldsymbol{p})$ of the form:

$$\eta^\lambda(\boldsymbol{p}) = \frac{1}{\sqrt{2}}\left\{\begin{matrix} 1 \\ \frac{\mu_B B - ie^{i\varphi(p)}p_F\alpha}{\lambda\tilde{\epsilon}(p)} \end{matrix}\right\}. \quad (12)$$

Substitution of Eq. (9) into Eq. (2) making use of Eq. (5) results in a rather cumbersome expression which generalizes the corresponding Eq. (7) of Ref. [8]. We sketch, therefore, only a few results for the low-T part of the phase diagram in Fig. 1. Below we discuss the main changes in the shape of the phase diagram, as introduced by SO coupling or anisotropy. As for the structure of the LOFF phase itself, we assume that the numerical analysis done in Ref. [13] remains applicable; i.e., the order parameter in the LOFF state has the structure of periodic stripes.

We state in more detail our results for the limiting cases of strong and weak SO interaction, which significantly simplify all calculations.

(a) Strong SO: $\alpha p_F \gg \Delta(0)$; subsequent analysis, which we do not provide here, leads after short calculations to our final results:

$$\boldsymbol{q} \perp \boldsymbol{B}, \qquad |\boldsymbol{q}| = \frac{2\mu_B H_{c2}}{v_F};$$
$$\mu_B H_{c2} = \sqrt{2\Delta(0)\alpha p_F} \equiv \sqrt{\frac{2\pi}{\gamma}T_{c0}\alpha p_F}. \quad (13)$$

One sees that SO interaction not only enhances the value of H_{c2} in comparison with the LOFF for 2D model of Ref. [8] , but it also fixes the direction of the structure vector. The resulting stripe structure is parallel to the magnetic field direction and has the perpendicular space periodicity $L = \sqrt{\pi\gamma/(2T_{c0}\alpha p_F)}v_F$.

According to [11], in the limit of strong SO interaction the spin susceptibility for parallel fields in superconducting state $\chi_S(T)$ is nonzero and equal to $\frac{1}{2}\chi_N$. This increases the critical paramagnetic field [12] only by a factor of $\sqrt{2}$, $\mu_B H_{\text{par}} = \Delta(0)$. Comparing this with Eq. (13), one sees that strong SO significantly increases the area occupied by the LOFF state by a factor of $\sqrt{\alpha p_F/T_{c0}} \gg 1$. Strong SO scattering by defects also enhances H_{c2} [14], but the LOFF state does not exist in the presence of disorder. Analytical expressions can also be obtained at low temperatures. For the dependence of the transition temperature on magnetic field, $T_c(B)$, one obtains, at $T \ll T_{c0}\sqrt{T_{c0}/(\alpha p_F)}$:

$$T_c(B) = 5.784(\sqrt{2\pi T_{c0}\alpha p_F/\gamma} - \mu_B B)^3/(\alpha p_F)^2, \quad (14)$$

and

$$T_c(B) = \pi T_{c0}^2/(2\gamma\mu_B B),$$
$$T_{c0}\sqrt{T_{c0}/(\alpha p_F)} \ll T \ll T_{c0}. \quad (15)$$

The expression for $T_c(B)$ also simplifies for small magnetic fields near T_{c0}:

$$\frac{T_{c0} - T_c}{T_{c0}} = \frac{7\zeta(3)}{8\pi^2}\frac{(\mu_B B)^2}{T_{c0}^2}. \quad (16)$$

Note that while the spin-orbit interaction splits the Fermi surface, quasiparticles with the same band spin index Eq. (11) form Cooper pairs in the superconducting state, so that spin-orbit interaction alone does not change T_c.

(b) Weak SO: $\alpha p_F \ll \Delta(0)$; unlike in the case of strong SO, the Cooper pair is formed mainly by pairing of electrons from the Fermi surfaces with different spin indices. The LOFF phase in a 2D superconductor with no SO interaction was first analyzed in Refs. [8,13]. According to Ref. [8], $\mu_B H_{c2} = \Delta(0) = \sqrt{2}H_{\text{par}}$, $v_F q = 2\mu_B H_{c2}$.

This result is zero order in spin-orbit interaction. The direction of $\boldsymbol{q}$ is *not fixed* with respect to B. Analysis to the second order in αp_F results in an anisotropy term which again fixes the vector $\boldsymbol{q}$, as in strong SO case Eq. (13), perpendicular to the direction of the magnetic field. Indeed, for the critical field as a function of the angle β between $\boldsymbol{q}$ and $\boldsymbol{B}$, we find:

$$\mu_B H_{c2} = \Delta(0) - \frac{\alpha^2 p_F^2}{2\Delta(0)}\cos^2\beta, \quad (17)$$

i.e., the maximum for H_{c2} is reached for $\beta = \pm\frac{\pi}{2}$.

For small magnetic fields near T_c, we get

$$\frac{T_{c0} - T_c}{T_{c0}} = \frac{7\zeta(3)}{4\pi^2}\frac{(\mu_B B)^2}{T_{c0}^2}. \quad (18)$$

A quadratic dependence on B remains valid for fields $\mu_B B \sim \alpha p_F$. However, note the factor two difference between Eqs. (16) and (18): suppression of T_c by magnetic field turns out to be slower in the case of strong spin-orbit interaction than in the case when the spin-orbit interaction is weak.

Although a more complicated LOFF periodic superstructure is possible, the energy considerations of Ref. [7] have shown in 3D that the stripe phase is energetically more favorable. A detailed study done numerically in 2D [13] has shown a more complicated than just a sinusoidal shape of the order parameter. We assume that the results of Ref. [13] remain valid in all cases considered above, so that the LOFF state preserves its striped order parameter form.

The major role of SO is in fixing of the LOFF superstructure. Anisotropy fixes the orientation of the LOFF stripes as well (see below). A 1st order reorientation transition may be expected corresponding to an abrupt change in the direction of the superconducting stripes (similar to a spin-flop transition) as the magnetic field is rotated in the 2D plane.

In the above discussion so far we have neglected any anisotropy at all. Meanwhile, the anisotropy is, of course, important. We address the issue of the effect of pinning stripe direction to the particular crystal axis only for the d-wave order parameter, since the latter is intrinsically anisotropic. Stripes may orient themselves along the directions of the gap maximums. It can be easily shown that H_{c2} for the $d_{x^2-y^2}$ superconducting order parameter takes the form $\mu_B H_{c2} = \frac{\pi}{\gamma} e^{1/4} T_{c0}$. The critical field for the LOFF phase of d wave is somewhat higher than for s wave [without the SO term Eq. (7)], as superconducting stripes get pinned to the crystal axes by the form of the order parameter and the direction of the magnetic field. Reorientation transition/twinning is also expected for any other cause.

In summary, we have shown (i) that inhomogeneous state in parallel fields extends considerably the low temperature phase diagram of surface superconductivity with increased spin-orbit interaction; (ii) all SC characteristics of the phase diagram in the (B, T) plane can be expressed in terms of T_{c0}, the critical temperature in the absence of the field, which may also serve as a method to extract the value of the SO interaction; (iii) LOFF state properties are strongly anisotropic in the plane with respect to field direction—this, for example, can be seen by measuring the anisotropy of the ac susceptibility signal; (iv) the indispensible feature of the LOFF state must be the reorientation transitions at the field rotation in the plane caused by locking of the LOFF order parameter by anisotropy.

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