Cryptoferromagnetism in superconductors with a broken time-reversal symmetry

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Abstract. The cryptoferromagnetic (CFM) state (the state with intrinsic domain structure) in superconducting ferromagnets subjected to external magnetic field is studied theoretically. Ferromagnetism (broken time-reversal symmetry) originates either from electron spin or from orbital moment of Cooper pairs (chiral $p$-wave superconductors like Sr$_2$RuO$_4$). We suggest the phase diagram, where the CFM state is present together with the Meissner and the mixed states. The size of domains in the CFM state is roughly of the order of the London penetration depth and in contrast to normal ferromagnets does not depend on either shape or size of the sample. At the scales much larger than the London penetration depth the spontaneous magnetization is absent in average, therefore the state is called cryptoferromagnetic. In contrast to the fully diamagnetic Meissner state or the partially diamagnetic mixed state, the CFM state is paramagnetic. The CFM state can be detected experimentally with detailed measurements of magnetization curves.

In recent years numerous experimental evidences of coexistence of superconductivity with ferromagnetism (broken time-reversal symmetry) in various materials were reported [1, 2, 3, 4]. Two types of such coexistence are possible: (i) The phase transitions to the ferromagnetic and the superconducting states occurs at different temperatures, so the coexistence starts below the lower from the two transitions. Rutheno-cuprates [1] belong to this type: the superconductivity onset occurs at the temperature much lower than the temperature of the magnetic transition. Normally different elements of the crystal structure are responsible for ferromagnetism and superconductivity, and spontaneous magnetization (FM order parameter) is related to spin. Later we call them spin superconducting ferromagnets (spin SFs). (ii) The magnetic and the superconducting transitions occur simultaneously. This can take place in unconventional superconductors with triplet Cooper pairing. An example of them is strontium ruthenate Sr$_2$RuO$_4$ [2, 3, 4]. The microscopic theory connects spontaneous magnetization in this material not with spin but with the intrinsic orbital moment of the $p$-wave Cooper pair with the wave function in the momentum space proportional to $p_x + ip_y$ (chiral $p$-wave superconductivity) [5]. We shall call them orbital superconducting ferromagnets (orbital SFs).

Whereas proof of superconductivity in SF materials is quite straightforward, a clear-cut detection of the FM order parameter is much more problematic. The internal magnetic field is screened out by the Meissner currents and can be present only near sample borders and defects, in particular, domain walls (DWs). This strongly suppresses the stray magnetic fields around the sample, which are most convincing evidence of ferromagnetism. In particular, Kirtley et al. [6] could not detect any stray field from DWs or edges in Sr$_2$RuO$_4$ samples. This is a challenge for the theory and for the very scenario of chiral $p$-wave pairing. Among possible explanations why experimentalists cannot see stray fields from DWs is the presence of domain
structure with a period essentially smaller than a distance between a sample surface and a probe used by experimentalists. In SF materials one must discern two possible types of equilibrium domain structure [7]. The first one is well known for normal ferromagnets: The domain structure results from competition between the energy of DWs and the magnetostatic energy of stray fields generated by the magnetic flux exiting from the sample surface. The period of the structure depends on the shape and the size of the sample going to infinity when the sample size grows. One can call these domains extrinsic FM domains. Since in SFs the Meissner effect exerts the magnetic field, it is impossible to benefit from decreasing the bulk magnetostatic energy in comparison with the DW energy, and extrinsic domains cannot appear at equilibrium [8]. But also long ago there was known another type of domains, which decrease the bulk magnetostatic energy at the expense of destroying the Meissner state [9]. The size of these domains is roughly of the order of the London penetration depth $\lambda$ and does not depend on either shape or size of the sample. Strictly speaking the state with this domain structure at the macroscopic scales is not FM but antiferromagnetic though with a rather large period. We shall call such a state cryptoferromagnetic (CFM). All previous theoretical investigations of the domain structure in SFs were restricted with the case of zero external magnetic field. We obtained the full phase diagram of both spin and orbital SFs cryptoferromagnetism in superconductors with broken TRS.

Here we present the analysis of the intrinsic domain structure (CFM state) in nonzero external magnetic field. We obtained the full phase diagram of both spin and orbital SFs with the phase transitions from the CFM state to the Meissner and to the mixed states. We calculated the magnetization curves, which demonstrated the absence of any diamagnetism in the CFM state. We argue that measurements of magnetization curves can provide evidences of cryptoferromagnetism in superconductors with broken TRS.

Let us consider a stripe domain structure with $180^\circ$ DWs in a sample subjected to external magnetic field $\mathbf{H}_0 = (0, H_0, 0)$. The DWs are parallel to the $yz$-plane separating domains with alternating magnetization $\mathbf{M} = (0, \pm M_0, 0)$ along the $+y$ or $-y$ direction. Since the $\mathbf{H}_0$ orientation is preferable the width $d_\uparrow$ of domains with the magnetization $\mathbf{M}$ parallel to $\mathbf{H}_0$ ($\uparrow$-domains) exceeds the width $d_\downarrow$ of the domains with $\mathbf{M}$ antiparallel to $\mathbf{H}_0$ ($\downarrow$-domains). We restrict ourselves to the simplest case when the London penetration length $\lambda$ exceeds the coherence length and the DW thickness. Then the surface energy $\sigma$ and the internal structure of DW is not affected by fields and currents at scales of $\lambda$. The Gibbs potential inside domains is

$$\mathcal{G} = \int d^3x \left( \frac{\hbar^2}{8\pi} + \frac{2\pi\lambda^2}{c^2} j^2 - \mathbf{h} \cdot \mathbf{M} - \frac{\mathbf{h} \cdot \mathbf{H}_0}{4\pi} \right),$$

where $\mathbf{h}$ is the magnetic field, and the electric current $\mathbf{j}$ is connected with the magnetic field $\mathbf{h}$ via the Maxwell equation $\nabla \times \mathbf{h} = (4\pi/c)\mathbf{j}$. Variation of the Gibbs potential yields the magnetic field $\mathbf{h}_{\uparrow,\downarrow} = (0, h_{\uparrow,\downarrow}, 0)$ in the $\uparrow$-domains and $\downarrow$-domains $h_{\uparrow,\downarrow} = (H_0 \pm 4\pi M_0) \cosh (x/\lambda - \xi_{\uparrow,\downarrow})/ \cosh \xi_{\uparrow,\downarrow}$, where $x$ is the distance from the DW and $\xi_{\uparrow,\downarrow} = d_{\uparrow,\downarrow}/2\lambda$ are reduced domain widths. All magnetic fields are low compared to the upper critical field $H_{c2}$. Substituting $\mathbf{h}_{\uparrow,\downarrow}$ into Eq. (1), adding the surface energy $\sigma$ of DWs, and averaging over the domain-structure period $d = d_\uparrow + d_\downarrow$ we arrive to the following expression for reduced energy density $\mathcal{E} = \mathcal{G}/2\pi M_0^2 V$ ($V$ is the sample volume):

$$\mathcal{E} = |2w - (1 + h_0)^2 \tanh \xi_\uparrow - (1 - h_0)^2 \tanh \xi_\downarrow| \xi_{\uparrow,\downarrow}^{-1}.$$

Here $h_0 = H_0/4\pi M_0$ and $w = \sigma/4\pi M_0^2 \lambda$ are dimensionless parameters. If $h_0 = 0$ and $\xi_\uparrow = \xi_\downarrow$ Eq. (2) coincides with the free energy density of Krey [9].

Minimization of energy density Eq. (2) with respect to $\xi_{\uparrow,\downarrow}$ yields the system of two nonlinear equations for $\xi_{\uparrow,\downarrow}$:

$$h_0 = \tanh \frac{\xi_\uparrow - \xi_\downarrow}{2} \tanh \frac{\xi_\uparrow + \xi_\downarrow}{2}, \quad w = \frac{\sinh 2\xi_\uparrow + \sinh 2\xi_\downarrow - 2(\xi_\uparrow + \xi_\downarrow)}{(\cosh \xi_\uparrow + \cosh \xi_\downarrow)^2}.$$

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The magnetic induction \( \mathbf{B} = \langle \mathbf{h} \rangle \) is determined by reduced magnetic induction \( b = B/4\pi M_0 \):

\[
b = \frac{1}{2} \frac{\partial \mathcal{E}}{\partial h_0} = [(1 + h_0) \tanh \xi_\uparrow - (1 - h_0) \tanh \xi_\downarrow](\xi_\uparrow + \xi_\downarrow)^{-1}.
\]

(4)

Fig. 1 shows the phase diagram in the plane \( w^2 - h_0 \). The area of the CFM state is restricted by two lines where the phase transition between the CFM and the Meissner states occurs:

1. The line \( \mathcal{E} = 0 \) (line 1 in Fig. 1), which corresponds to the limit \( \xi_{\downarrow,\uparrow} \to \infty \). The values of \( w \) and \( h_0 \) on this line are connected by the relation \( w = 1 + h_0^2 \).

2. The line on which domains with magnetization opposite to the external magnetic field vanish, \( \xi_\downarrow = 0 \) (line 2 in Fig. 1). The equation describing this line is \( w_c = h_c^{1/2}(1 + h_c) - (1 - h_c^2)\xi_{c\downarrow}/2 \), where \( \xi_{c\downarrow} = \ln(1 + h_c^{1/2}) - \ln(1 - h_c^{1/2}) \) is the critical size of the \( \uparrow \)-domain. The magnetic induction on the critical line is \( b_c = 2h_c^{1/2}\xi_{c\downarrow}^{-1} \).

Let us consider the left lower corner of this diagram where \( w \ll 1 \) and \( h_0 \ll 1 \). The critical parameters on the line \( \xi_\downarrow = 0 \) as functions of \( w \) are \( h_c = (3w)^{2/3}/4 \) and \( \xi_{c\downarrow} = (3w)^{1/3} \). Aside from the critical line Eqs. (3) yield: \( 4h_0 = \xi_\uparrow - \xi_{c\downarrow}^2 \), \( 3w = \xi_\uparrow + \xi_{c\downarrow}^2 \). For small \( h_0 \ll h_c \) these equations can be solved analytically: \( \xi_{c\downarrow} = 4^{1/3}h_c^{1/2}(1 - 4^{-4/3}h_0^2h_c^{-2}) \pm 4^{-1/3}h_0h_c^{1/2} \).

The linear magnetic permeability in the CFM state is determined from two relations connecting \( \mu \) and \( w \) with the period \( \xi = \xi_\uparrow + \xi_\downarrow \approx 2\xi_\uparrow \):

\[
\mu = \frac{db}{dh_0} = \coth \xi/\xi, \quad w = \tanh \xi - \frac{\xi}{\cosh \xi}.
\]

(5)

In the limit \( w \to 0 \) the magnetic permeability is divergent: \( \mu \approx (2/3w)^{2/3} \). The whole magnetization curves \( b(h_0) \), which were calculated numerically, are shown in Fig. 2.

We should also consider the transition to the mixed state. The reduced free energy of the mixed state with respect to the energy of the Meissner state is \( \mathcal{E}_m = -(1 + h_0 - H^*/4\pi M)^2 = -(1 + h_0 - h_{c1}w^2)^2 \). The field \( H^* \) inside the mixed state differs from the first critical magnetic field \( H_{c1} = (\Phi_0 \ln \kappa)/4\pi \lambda^2 \) by another logarithm factor, but we neglect it assuming \( H^* \approx H_{c1} \). Here \( \kappa = \lambda/\xi_0 \) is the ratio of \( \lambda \) to the coherence length \( \xi_0 \) and the reduced first critical field was introduced: \( h_{c1} = H_{c1}/4\pi M_0w^2 = \Phi_0M_0^2\ln \kappa/\sigma^2 \). The phase transition to the mixed state may occur either (i) from the Meissner state being determined by the condition \( \mathcal{E}_m = 0 \) (the second-order transition, line 3 in Fig. 1), or from the CFM state crossing the critical line on which \( \mathcal{E}_m = \mathcal{E} \) (the first-order transition, line 4 in Fig. 1). At zero external field \( h_0 \) and small \( w \) the phase transition between the mixed state and the CFM state occurs at \( w_m \approx \sqrt{3}(2h_{c1})^{-3/4} \).

Thus whatever large \( h_{c1} \) is, in the left lower corner of the phase diagram there is always the spontaneous vortex phase, i.e., the mixed state without external magnetic field. The CFM state disappears from the phase diagram at \( h_{c1} < 0.5 \).

Now let us analyze the phase transformations in the process of cooling down below the SC critical temperature. This process is different for spin and orbital SFs. In the case of spin SFs, when the magnetic transition occurs at much higher temperature, one may neglect temperature dependence of \( M_0, \sigma, \) and \( h_{c1} \). Then only \( w^2 \propto 1/\lambda^2 \propto \tau \) depends on relative temperature difference \( \tau = 1 - T/T_c \). On the phase diagrams of Figs. 1 the state moves along straight lines parallel to the horizontal axis \( w^2 \). From these figures it is evident that just below the critical temperature the system enters the mixed state. At further cooling down the system crosses to the Meissner state either directly or through the area of the CFM state. For orbital SFs the cooling process occurs differently. In this case the “magnetization” \( M_0 \propto \lambda^2 \propto \tau \), and the DW surface energy is a product of the condensation energy \( H^2_2(\tau) \sim [\Phi_0/\lambda(\tau)\xi_0(\tau)]^2 \) and the coherence length \( \xi_0(\tau) \sim \xi_0/\sqrt{\tau} \): \( \sigma \sim \tau^{3/2}[\Phi_0^2/\lambda_0^2\xi_0^2] \). Here \( \lambda_0 \) and \( \xi_0 \) are the penetration depth and the coherence length at zero temperature. Then the parameters \( w^2 \sim h_{c1}^1 \sim (\lambda_0/\xi_0)^2 \) do
not depend on temperature whereas the reduced magnetic field does: \( h_0 = H_0 / 4\pi M_0 \propto 1 / \tau \). Thus in the field-cooling process the state moves along vertical lines on the phase diagrams in Fig. 1. However, as pointed out above, the CFM state can compete with the mixed state only if \( h_{c1} \) is high enough. Since \( h_{c1} \sim (\xi_0 / \lambda_0)^2 \), this requires \( \lambda_0 \) not large compared to \( \xi_0 \). According to [6] the ratio of \( \lambda_0 = 190 \text{ nm} \) to \( \xi_0 = 66 \text{ nm} \) is not too high indeed. But this means that the DW thickness is not so small compared to \( \lambda \) as assumed in our analysis. Therefore for orbital SFs our analysis can provide only a qualitative but still credible picture of the phase transformations.

In conclusion, we analyzed the phase diagram of superconductors with a broken time-reversal symmetry, in which superconductivity competes either with spin ferromagnetism or with ferromagnetism originated from the intrinsic angular momentum of Cooper pairs (chiral \( p \)-wave superconductors like \( \text{Sr}_2\text{RuO}_4 \)). The phase diagram includes the state with the intrinsic domain structure, which has no FM order at macroscopic scales and therefore called cryptoferromagnetic. The CFM state is not diamagnetic, either fully (as the Meissner state), or partially (as the mixed state). The state can be detected experimentally with detailed measurements of magnetization curves.

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Figure 1. Phase diagram for the value of the reduced lower critical field \( h_{c1} = 2 \). The lighter (yellow) shaded area is the CFM state. The darker (blue) shaded area is the mixed state. The rest is the Meissner state.

Figure 2. Magnetization curves are shown at different values of specific surface energy of the domain wall \( w \). Vertical lines correspond to critical fields above which the intrinsic domain structure collapses.

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