

## Prediction and description of a chiral pseudogap phase

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We point out that a system which supports chiral superconductivity should also support a chiral pseudogap phase: a finite temperature phase wherein superconductivity is lost but time-reversal symmetry is still broken. This chiral pseudogap phase can be viewed as a state with phase incoherent Cooper pairs of a definite angular momentum. This physical picture suggests that the chiral pseudogap phase should have definite magnetization, should exhibit a (nonquantized) charge Hall effect, and should possess protected edge states that lead to a quantized thermal Hall response. We explain how these phenomena are realized in a Ginzburg-Landau description, and comment on the experimental signatures of the chiral pseudogap phase. We expect this work to be relevant for all systems that exhibit chiral superconductivity, including doped graphene and strontium ruthenate.

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Chiral superconductors feature pairing gaps that wind in phase around the Fermi surface (FS), breaking time-reversal symmetry (TRS).<sup>1-4</sup> They realize topological superconductivity<sup>5-8</sup> and exhibit a host of fascinating and technologically useful properties, such as protected edge states, a quantized thermal Hall effect, and unconventional zero modes in vortices.<sup>1-4</sup> Chiral *p*-wave superconductivity is believed to have been found in strontium ruthenate,<sup>9</sup> and chiral *d*-wave superconductivity has been established to be the leading weak-coupling instability in strongly doped graphene.<sup>10,11</sup> Systems that may display *d + id* superconductivity have also been discussed in Refs. 12 and 13. However, all theoretical work to date has focused on chiral superconductors at low temperatures. In this work, we argue that much of the exotic phenomenology associated with chiral superconductivity can be exhibited even at high temperatures, when the superconductivity is absent. In particular, we predict the existence of a chiral pseudogap phase with a rich phenomenology, including a magnetic dipole moment, nonquantized charge Hall effect, protected edge states, and quantized thermal Hall effect. Since this pseudogap phase does not require low temperatures or exceptionally clean systems (unlike chiral superconductivity), we expect it to be advantageous for nanoscience applications.

Emery and Kivelson have pointed out that phase incoherent Cooper pairs can form at temperatures much higher than the characteristic temperature for onset of superconductivity.<sup>14</sup> This “preformed Cooper pairs” picture has been invoked as a possible explanation for the pseudogap phase of the cuprate high- $T_c$  materials. The nature of the pseudogap phase of the cuprates remains controversial, not least because the preformed Cooper pairs picture for the cuprates does not lead to many sharp testable predictions. However, the situation is markedly different for a chiral superconductor, where a pseudogap phase with phase-incoherent preformed Cooper pairs can still break time-reversal symmetry. The resulting chiral pseudogap phase has a rich and distinctive phenomenology, which should be readily testable experimentally.

In this paper, we provide a Ginzburg-Landau description of the physics of a chiral superconductor that both demonstrates that a chiral pseudogap can exist, and also elucidates the phenomenology of the chiral pseudogap phase. We work for simplicity in the continuum, assuming continuous rotation symmetry, although we expect our results to also apply for

lattice systems. We work in two spatial dimensions, since most materials where chiral superconductivity is expected to arise are either two-dimensional, or layered three-dimensional materials. We comment at the end on the likely implications of our results for systems that are expected to exhibit chiral superconductivity, such as strontium ruthenate and doped graphene.

The broad picture is summarized in Fig. 1. At the lowest temperatures  $T < T_c$ , the system is a chiral superconductor. At high temperatures  $T > T_*$ , the system is a Fermi liquid. At intermediate temperatures  $T_c < T < T_*$  there arises a chiral pseudogap phase with phase-incoherent Cooper pairs of a definite angular momentum (Fig. 2). This chiral Cooper pairing produces a magnetization which may be detected using torque magnetometry. Moreover, the chiral pseudogap phase inherits the topological properties of the chiral superconductor, and in particular has protected edge states that lead to a quantized thermal Hall response. Finally, at finite temperature the chiral pseudogap phase also has a (nonquantized) bulk charge Hall response, even in the clean, dc limit. Such a response is forbidden in the chiral superconductor (which cannot support a voltage gradient), and thus the phenomenology of the chiral pseudogap state is in some ways even richer than that of the chiral superconductor.

We note that a system with a  $U(1) \times Z_2$  symmetry, and an intermediate phase with broken  $Z_2$  symmetry only, was also discussed in Ref. 15. However, the physics of these phases is very different, since in this case,<sup>15</sup> the  $Z_2$  symmetry was not associated with time reversal, and nor was the system coupled to an electromagnetic gauge field.

### I. GINZBURG-LANDAU THEORY

We consider a two-dimensional electron system with continuous rotation symmetry, and with a doubly degenerate Cooper instability. Cooper pairing occurs in a channel with angular momentum  $l$ , and the Cooper pair wave functions that vary around the Fermi surface as  $\psi_1 = \eta_1 \cos l\varphi$  and  $\psi_2 = \eta_2 \sin l\varphi$ , respectively, are assumed to have the same energy. Here the angle  $\varphi$  parametrizes position on the two-dimensional Fermi surface. The degeneracy of the two pairing functions follows from rotation symmetry. An analogous situation is believed to arise in strontium ruthenate (with  $l = 1$ )<sup>9</sup> and also

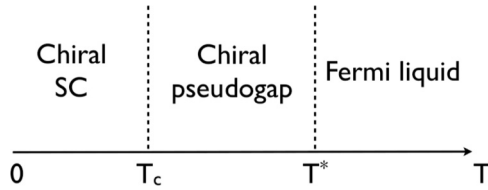


FIG. 1. Schematic phase diagram of material exhibiting chiral superconductivity. At high temperatures, the system is a Fermi liquid. At low temperatures, it is a chiral superconductor. At intermediate temperatures, there arises a chiral pseudogap phase, marked by phase-incoherent Cooper pairs of definite angular momentum. The chiral pseudogap phase breaks time-reversal symmetry, has a definite magnetization, and inherits the topological structure of the chiral superconductor, possessing protected edge states that lead to a quantized thermal Hall response. In addition, the chiral pseudogap also exhibits a (nonquantized) dc charge Hall effect, which is forbidden in the chiral superconductor.

in doped graphene (with  $l = 2$ ).<sup>10</sup> We focus on the spin-singlet case for simplicity, although we expect our results to also apply to spin-triplet systems. The gap function behaves as

$$\Delta(\mathbf{r}, t, \varphi) = \eta_1(\mathbf{r}, t) \cos l\varphi + \eta_2(\mathbf{r}, t) \sin l\varphi. \quad (1)$$

We wish to construct a Ginzburg-Landau free energy functional out of these gap wave functions. The free energy functional should contain all symmetry allowed terms. Fortunately, the restriction to a system with continuous rotation symmetry greatly restricts the allowed terms in the free energy functional. We also assume that the free energy functional should respect time-reversal symmetry and inversion symmetry. To quartic order in the gap functions, the static part of the free energy,  $F_0$ , takes the form

$$F_0 = (T - T_{MF})(|\eta_1|^2 + |\eta_2|^2) + K_1(|\eta_1|^2 + |\eta_2|^2)^2 + K_2|\eta_1^2 + \eta_2^2|^2, \quad (2)$$

where  $T_{MF}$  is the temperature for onset of Cooper pairing, and  $K_{1,2} > 0$ . The positivity of  $K_2$  means that this system favors coexistence of the two orders with relative phase  $\pm i$ .

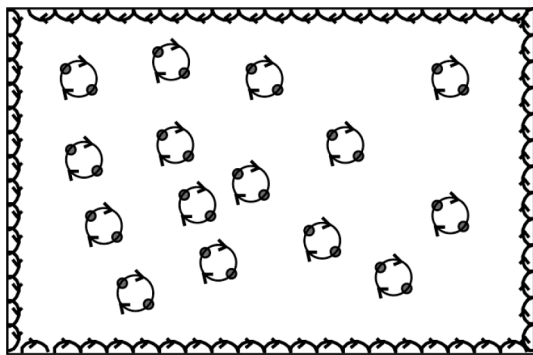


FIG. 2. The chiral pseudogap phase has chiral, phase-incoherent Cooper pairs in the bulk, which have a definite angular momentum. This picture suggests that there should be edge states associated with skipping orbits for the Cooper pairs, as shown above. This intuition manifests itself mathematically through a Chern number for the quasiparticle excitations of the chiral pseudogap phase. The orbiting Cooper pairs also produce a magnetization, which couples directly to an external magnetic field, and also to vortices.

As a result, the order parameter takes the form  $\Delta(\mathbf{r}, t, \varphi) = \Delta(\mathbf{r}, t) \exp(\pm il\varphi)$ .

To obtain the physics we are interested in, we must examine the gradient terms in the free energy. Since we are interested in finite temperature physics, we neglect temporal fluctuations, and consider a time-independent Ginzburg-Landau description of the problem, with  $\eta_{1,2}(\mathbf{r}, t) = \eta_{1,2}(\mathbf{r})$ . Note that since we are dealing with a charged system, we must take into account the coupling to the electromagnetic field. The electromagnetic field is introduced by minimal coupling, through  $i\partial \rightarrow iD = i\partial + 2eA$  (the factor of 2 arises because we have Cooper pairs). We define  $\mathbf{D}$  to be a two-component vector,  $\mathbf{D} = (i\partial_x + 2eA_x, i\partial_y + 2eA_y)$ . At second order in gradients, the free energy has the form

$$\begin{aligned} F_{\nabla} &= J_1(\mathbf{D}\eta_1)^* \cdot \mathbf{D}\eta_1 + J_1(\mathbf{D}\eta_2)^* \cdot \mathbf{D}\eta_2 \\ &\quad + J_2(\mathbf{D}\eta_1)^* \times \mathbf{D}\eta_2 \cdot \hat{\mathbf{z}} - J_2(\mathbf{D}\eta_2)^* \times \mathbf{D}\eta_1 \cdot \hat{\mathbf{z}} \\ &= J_1(\mathbf{D}\eta_1)^* \cdot \mathbf{D}\eta_1 + J_1(\mathbf{D}\eta_2)^* \cdot \mathbf{D}\eta_2 + J_2\eta_1^*(\mathbf{D}^* \times \mathbf{D}) \cdot \hat{\mathbf{z}}\eta_2 \\ &\quad - J_2\eta_2^*(\mathbf{D}^* \times \mathbf{D}) \cdot \hat{\mathbf{z}}\eta_1, \end{aligned} \quad (3)$$

where  $J_{1,2}$  are phenomenological constants, and in going from the first line to the second we have performed an integration by parts and have ignored possible boundary terms. We have used the notation  $(\mathbf{D}^* \times \mathbf{D}) \cdot \hat{\mathbf{z}} = [(-i\partial + 2eA) \times (i\partial + 2eA)] \cdot \hat{\mathbf{z}} = \partial \times \partial - 2ei(\partial_x A_y - \partial_y A_x) = \partial \times \partial - 2eiB_z$ , where  $B_z$  is the magnetic field transverse to the plane.<sup>16</sup>

We now write  $\eta_{1,2} = \Delta_0 \exp(i\theta_{1,2})$ , and we neglect (massive) fluctuations in the magnitude  $\Delta_0$ . We define new variables  $\theta_+ = \frac{1}{2}(\theta_1 + \theta_2)$  and  $\theta_- = \theta_1 - \theta_2$ , which are  $2\pi$  periodic (a  $2\pi$  shift in either leaves the physical state unchanged). Recalling that  $\partial \times \partial\theta_{1,2} = n_v$ , where  $n_v$  is the vorticity, we find that the free energy takes the simple form  $F = F_0(|\Delta_0|) + |\Delta_0|^2(F_+ + F_- + F_{+-})$ , where

$$F_+ = 2J_1 [(i\partial\theta_+ + eA) \cdot (i\partial\theta_+ + eA)], \quad (5)$$

$$F_- = \frac{1}{2}[J_1(\partial\theta_-)^2 + 8J_2B_z \sin \theta_-] + 2K_2|\Delta_0|^2 \cos 2\theta_-, \quad (6)$$

$$F_{+-} = 2J_2 \sin \theta_- n_v. \quad (7)$$

We now interpret these equations. First, note that the anisotropy term in Eq. (6) favors  $\sin \theta_- = \pm 1$ . Next, note that  $\sin \theta_-$  couples to an external magnetic field in the same way as a magnetization, and can be interpreted as the magnetization associated with the orbital angular momentum of the chiral Cooper pairs. We point out that  $\sin \theta_-$  serves as an Ising order parameter for time-reversal symmetry breaking, and couples to the vortex density (which is expected since the vortices carry magnetic flux). We also note that the  $\theta_-$  sector (at  $B_z = 0$ ) describes an XY model with Ising anisotropy. The Ising anisotropy is known to be a relevant perturbation<sup>17</sup> and thus at low enough temperatures the phase  $\theta_-$  will be pinned to  $\sin \theta_- = \pm 1$ , for arbitrarily weak  $K_2$ . The  $\theta_-$  order will be destroyed by thermal fluctuations at a temperature  $T_*$ , above which the time-reversal symmetry will be restored.

Meanwhile, the  $\theta_+$  sector is the action of a superconductor. It will have a Higgs phase, where the photon is massive, and  $\theta_+$  is locked, and it will also have a trivial phase, where  $\theta_+$  is disordered, with the phase transition occurring at a

temperature  $T_c$ . The disordering transition will be associated with proliferation of vortices in  $\theta_+$ . The magnetization  $\sin \theta_-$  imbalances the number of vortices and antivortices, so the vortex proliferation transition falls into the universality class of  $XY$  models in a magnetic field studied in Ref. 18, and not the usual Kosterlitz-Thouless universality class.

Finally, we note that the elementary vortices in the theory carry magnetic flux  $hc/e$  (instead of the more usual  $hc/2e$ ), and involve a simultaneous  $2\pi$  winding in  $\theta_1$  and  $\theta_2$ . This is because a vortex in  $\theta_1$  or  $\theta_2$  alone leaves the phase  $\theta_-$  misaligned everywhere around it, and hence carries a large anisotropy energy cost. As a result, the loss of superconductivity at  $T_c$  is associated with a proliferation of double vortices, carrying magnetic flux  $hc/e$ , in an external magnetic field.

Now the temperatures  $T_c$  and  $T^*$  are in principle independent. We assume that  $T^* > T_c$  for the purposes of this paper. Since  $Z_2$  symmetry breaking is more robust against thermal fluctuations than  $U(1)$  symmetry breaking, we believe  $T^* > T_c$  represents the generic case for a chiral superconductor.

## II. EXPERIMENTAL SIGNATURES OF THE CHIRAL PSEUDOGAP PHASE

We now discuss experimental signatures of the chiral pseudogap state. We note that since the chiral pseudogap state gaps out the full Fermi surface, there should be a gap to quasiparticle excitations. However, since the chiral pseudogap state only forms at finite temperature, there will always be some thermal excitation across the gap. These thermally excited quasiparticles will see the magnetic field from the chiral Cooper pairing, and will give rise to a (classical) charge Hall effect (and thermal Hall effect), in complete analogy with Hall effect in an external magnetic field. Note that the chiral pseudogap phase displays a bulk charge Hall effect even in the dc limit, with a clean sample, whereas the chiral superconductor cannot display a Hall conductance in this limit,<sup>19</sup> because a superconductor cannot sustain a voltage drop. Similar effects should also arise from the effect of the magnetic field on the charged Cooper pairs. However, none of these effects will be quantized or topological.

### A. Topological properties and signatures in transport

The physical picture of the chiral pseudogap state Fig. 2 suggests there should be edge modes associated with “skipping orbits” for the Cooper pairs, which should also give rise to some kind of quantized Hall effect. A good way to understand this is as follows: the (fermionic) quasiparticle spectrum of the chiral superconductor is gapped, and consists of bands that carry a Chern number. This is the origin of the quantized thermal Hall response of the chiral superconductor.<sup>20</sup> We are assuming that we are in the weak pairing regime, where the chiral superconductivity has topological properties.<sup>21</sup> Crucially, the sign of the Chern number depends only on the sign of the time-reversal symmetry breaking (i.e., whether  $\theta_- = \pm\pi/2 \pmod{2\pi}$ ), and is independent of  $\theta_+$ . When we disorder  $\theta_+$  to go from the chiral superconductor into the chiral pseudogap phase, the fermion spectrum remains gapped, and continues to carry a Chern number. Thus, the edge states and quantized thermal Hall response exhibited by the

chiral superconductor should also be exhibited by the chiral pseudogap phase.

While the thermal Hall conductance coming from the edge states is quantized, experimentally, one measures the total Hall conductance, obtained by summing over topological and quasiparticle contributions, and the contribution from thermally excited quasiparticles is not quantized. However, we could imagine isolating the “topological” Hall effect from the classical Hall effect if the width of the pseudogap region is sufficiently large. The gap to quasiparticle excitations is of order  $T^*$ . Therefore, if  $T_c \ll T^*$  (i.e., if the chiral pseudogap region is sufficiently wide), then the fermionic excitations can be suppressed arbitrarily strongly by working deep in the pseudogap regime, at  $T_c < T \ll T^*$ . The theoretically clean limit involves taking  $T_c \rightarrow 0$ , whereupon there will be a truly quantized thermal Hall response.

If the quasiparticles of the chiral pseudogap state carry any quantum numbers, the chiral pseudogap state will display the corresponding quantized Hall effects in addition to the quantized thermal Hall effect. For example, the quasiparticles of the  $d+id$  pairing state carry spin, and the  $d+id$  superconductor displays a spin quantum Hall effect.<sup>20</sup> The chiral pseudogap phase produced by thermally disordering the  $d+id$  superconductor should inherit this topological response, and should display a spin quantum Hall effect in addition to a thermal quantum Hall effect.

A provocative question is whether it is possible for the chiral pseudogap state to have a charge quantum Hall response—i.e., whether it is possible to have a direct second-order transition from a chiral superconductor to a quantum anomalous (charge) Hall state. While this is clearly not the generic case, there seems to be no reason why such a transition should be impossible, and we believe it is a fruitful topic for further investigation.

### B. Mechanical and optical signatures

The chiral pseudogap phase has a net magnetization, which may be directly measured through magnetic force magnetometry (see, e.g., Ref. 22). Moreover, since the chiral pseudogap phase has a Hall conductivity (at finite temperature), it should exhibit a Kerr effect (rotation of the polarization angle of reflected light). It was demonstrated in Ref. 23 that the Kerr rotation arising from reflection from a single two-dimensional sheet of Hall conductance  $\sigma_{xy}$  placed on a substrate of dielectric constant  $n$  is

$$\theta_K \approx \frac{8\pi \operatorname{Re} \sigma_{xy}}{c(n^2 - 1)}. \quad (8)$$

The Kerr signal arising due to a charge Hall response should be much stronger than the Kerr signal in the absence of a charge Hall response, and thus should be easier to detect experimentally. One can also consider a layered system with multiple layers of chiral pseudogap phase. It is simplest to consider the limit when there is no tunneling of quasiparticles or Cooper pairs between layers, so that the different layers are coupled only through the electromagnetic field. In this situation, the magnetic dipole interactions between layers should align the layers so that the same sense of chiral pseudogap phase is obtained in each layer, i.e.,  $\sin \theta_- = +1$  in all layers, or else  $\sin \theta_- = -1$  in all layers. The Kerr

response from this system is analogous to the Kerr response of a ferromagnet, discussed in Ref. 24.

### C. Domain structure

We note that we have thus far assumed that the Ising sector ( $\sin \theta_-$ ) has long-range order. However, an Ising system can have domains, and the chiral pseudogap state will indeed have domains with  $\sin \theta_- = \pm 1$ . In the absence of magnetic disorder, the domain size will be determined by the competition between the gradient energy cost of domain formation, and the long-range magnetic dipole interactions between domains, in analogy with the electronic microemulsion phases from Ref. 25. If the characteristic domain size is larger than the system size (or the laser spot size for Kerr effect measurements), then the domain structure can be neglected, otherwise it will suppress the experimental signal by a factor of  $\sqrt{N}$ , where  $N$  is the number of domains.

### III. RELEVANCE FOR EXPERIMENTAL SYSTEMS

The only material which is known to be a chiral superconductor is strontium ruthenate. A chiral pseudogap phase may form in this material, although it will be more complicated than the chiral pseudogap phase discussed above because the pairing in strontium ruthenate is spin triplet. This complicates the analysis because vortices in spin-triplet chiral superconductors have Majorana zero modes, which endow them with non-Abelian statistics. A detailed treatment of spin-triplet superconductors is beyond the scope of the present work. However, we note that there is numerical evidence suggesting that disordering a spin-triplet chiral superconductor can produce a state with quantized thermal Hall effect,<sup>26</sup> which is a key prediction for the chiral pseudogap phase.

A chiral pseudogap phase with spin-singlet pairing (which is what we discussed in this paper) may arise in graphene doped to a Van Hove singularity in the density of states. This system is expected to exhibit spin-singlet,  $d$ -wave chiral superconductivity if interactions are sufficiently weak.<sup>10,11</sup> Here, too, there should be a chiral pseudogap phase, without any of the complications arising from spin-triplet pairing.

The separation of scales between  $T_*$  and  $T_c$  is largest for systems with small carrier densities.<sup>14</sup> For strongly doped graphene, the carrier density will be very high, so that  $T_c$  is likely to be quite close to  $T_*$ . In this case, the chiral pseudogap phase will not be very relevant. However, if the carrier density is strongly depleted, for example by localization on disorder, then the separation between scales might become more substantial. Since all known methods of doping graphene to the Van Hove point introduce large amounts of disorder, it remains possible that the chiral pseudogap phase might be discovered in doped graphene. A phase that shares some of the phenomenology of the chiral pseudogap phase was also proposed for a cobalt-based material in Ref. 27.

Of course, the best-studied superconducting materials are probably the cuprates. While we are not aware of any compelling scenario for chiral superconductivity (or chiral pseudogaps) in the cuprates, it is amusing to note that there are numerous experiments suggesting both time-reversal

symmetry breaking and a net vorticity in the pseudogap phase of the cuprates.<sup>28,29</sup> Clearly, the mechanism outlined in this paper for generating a chiral pseudogap phase by disordering a chiral superconductor does not apply to the cuprates. However, the experiments<sup>28,29</sup> make it tempting to speculate that a chiral pseudogap phase may still be forming in these materials. A more detailed analysis of these experiments is beyond the scope of the present work.

### IV. OUTLOOK

The paucity of known chiral superconductors makes it difficult to find materials that exhibit chiral pseudogaps. However, given the intense current interest in searching for realizations of chiral superconductivity, it seems inevitable that such materials will eventually be found. At that point, a chiral pseudogap phase will likely be found also. Moreover, we know from the existing high- $T_c$  materials that pseudogap phases can survive to much higher temperatures than superconductivity. Since the chiral pseudogap phase exhibits much of the exotic phenomenology of the chiral superconductor, and may survive to significantly higher temperatures, we expect that the chiral pseudogap phase will not only be of intrinsic theoretical interest, but will also be highly beneficial for nanoscience applications.

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### APPENDIX: FIRST-PRINCIPLES DERIVATION OF THE BOSONIC GINZBURG-LANDAU THEORY

In this Appendix, I sketch how the bosonic Ginzburg-Landau theory presented in Eqs. (2)–(4) may be derived starting from a fermionic description. For maximum simplicity I assume a rotation-invariant system (as in the main paper). Most of the manipulations are standard and follow the discussion in Ref. 30. We start by writing the partition function  $Z$  in the form of a functional field integral. Thus we have  $Z = \int D[\bar{\psi}, \psi] \exp[-S(\bar{\psi}, \psi)]$ , where  $\bar{\psi}$  and  $\psi$  are Grassmann-valued fields that represent fermions, and  $S(\bar{\psi}, \psi)$  is the imaginary time action. In the Matsubara frequency representation, the action takes the form

$$\begin{aligned}
 S = T \sum_{\omega_n} \int_{\mathbf{k}} \bar{\psi}_{\omega_n, \mathbf{k}, \sigma} (-i\omega_n + \varepsilon_{\mathbf{k}} - \mu) \psi_{\omega_n, \mathbf{k}, \sigma} \\
 + T^2 \sum_{\omega_n, \omega'_n, \omega_m} \int_{\mathbf{k}, \mathbf{k}', \mathbf{q}} g_{\mathbf{k}, \mathbf{k}', \mathbf{q}}^{\sigma \sigma'} \bar{\psi}_{\omega_n + \omega_m, \mathbf{k} + \mathbf{q}, \sigma} \bar{\psi}_{-\omega_n, -\mathbf{k}, \sigma'} \\
 \times \psi_{\omega'_n + \omega_m, \mathbf{k}' + \mathbf{q}, \sigma} \psi_{-\omega'_n, -\mathbf{k}', \sigma'}, \quad (\text{A1})
 \end{aligned}$$

where  $T$  is the temperature,  $\omega_n = (2n + 1)\pi T$  is a fermionic Matsubara frequency,  $\mathbf{k}$  is a two-dimensional momentum, and  $\sigma, \sigma'$  are spin labels (repeated spin indices are summed over). The dispersion is  $\varepsilon_{\mathbf{k}} = k^2/2m$  for the special case of an isotropic system considered in this paper, and  $\mu$  is the chemical potential, which controls the Fermi wave vector. The action



above has been written down in the absence of any vector potential. We suppress the vector potential to avoid clutter, but we remind the reader that it may be reintroduced at any time through minimal substitution  $\mathbf{k} \rightarrow \mathbf{k} - e\mathbf{A}$ .

Meanwhile, the interaction  $g_{\mathbf{k},\mathbf{k}'}$  in Eq. (A1) is assumed to have an attractive component in two independent channels,

both of which have angular momentum  $l$ . We define the two functions  $d_1$  and  $d_2$  which vary with  $\mathbf{k}$  as  $d_1 = \cos l\varphi$  and  $d_2 = \sin l\varphi$ , respectively, where  $\varphi$  is the angle between the wave vector  $\mathbf{k}$  and the  $k_x$  axis. We also assume that  $l$  is even, so the pairing is spin singlet (for specificity). The quartic term in Eq. (A1) can then be written as

$$\lambda_{\mathbf{k}} \sum_{\alpha=1,2} (\bar{\psi}_{\omega_n+\omega_m, \mathbf{k}+\mathbf{q}, \uparrow} d_{\alpha} \bar{\psi}_{-\omega_n, -\mathbf{k}, \downarrow}) (\psi_{\omega'_n+\omega'_m, \mathbf{k}'+\mathbf{q}, \uparrow} d_{\alpha} \psi_{-\omega'_n, -\mathbf{k}', \downarrow}),$$

where all frequencies are summed over and momenta are integrated over.

We decouple the quartic term by means of a Hubbard-Stratanovich transformation, introducing the new bosonic fields  $\eta_1 = 2\lambda \langle \psi d_1 \psi \rangle$  and  $\eta_2 = 2\lambda \langle \psi d_2 \psi \rangle$ . This leads to a new expression for the partition function,  $Z = \int D[\eta_1, \eta_2, \bar{\psi}, \psi] \exp(-S(\eta_1, \eta_2, \bar{\psi}, \psi))$ . The action  $S$  is given by

$$S_{\omega_n, \mathbf{k}} = T \sum_{\omega_n} \int \frac{d^2k}{(2\pi)^2} \bar{\psi}_{\omega_n, \mathbf{k}, \sigma} (-i\omega_n + \varepsilon_{\mathbf{k}} - \mu) \psi_{\omega_n, \mathbf{k}, \sigma} + \frac{1}{2\lambda_{\mathbf{k}}} (|\eta_1(\omega_n, \mathbf{k})|^2 + |\eta_2(\omega_n, \mathbf{k})|^2) \\ + T^2 \sum_{\omega_n, \omega'_n} \int \frac{d^2k d^2q}{(2\pi)^2} \eta_1^*(\omega'_n, \mathbf{q}) [\psi_{\omega_n+\omega'_n, \mathbf{k}+\mathbf{q}, \uparrow} d_1 \psi_{-\omega_n, -\mathbf{k}, \downarrow}] + \eta_2^*(\omega'_n, \mathbf{q}) [\psi_{\omega_n+\omega'_n, \mathbf{k}+\mathbf{q}, \uparrow} d_2 \psi_{-\omega_n, -\mathbf{k}, \downarrow}] + \text{c.c.} \quad (\text{A2})$$

Close to the critical temperature, where time-independent Ginzburg-Landau theory is justified, we may restrict ourselves to the zero-frequency component of the bosonic fields  $\eta_1$  and  $\eta_2$ . However, the possible spatial variation of the bosonic fields (i.e., the wave-vector dependence) should be taken into account. After switching to a Nambu spinor representation and integrating out the fermions in the manner of Ref. 30, one obtains a purely bosonic action  $S[\eta_1, \eta_2]$ , where

$$S = \int \frac{d^2k}{(2\pi)^2} \frac{1}{2\lambda_{\mathbf{k}}} (|\eta_1(\mathbf{k})|^2 + |\eta_2(\mathbf{k})|^2) + \text{Tr} \ln \mathcal{G}. \quad (\text{A3})$$

Here the Green function  $\mathcal{G}$  is a  $2 \times 2$  matrix in Nambu spinor space, and is defined by the relation

$$\mathcal{G}^{-1}(\omega_n, \mathbf{k}, \mathbf{k}') = \begin{pmatrix} [i\omega_n - (\varepsilon_{\mathbf{k}} - \mu)]\delta(\mathbf{k} - \mathbf{k}') & \eta_1(\mathbf{k} - \mathbf{k}')d_1(\mathbf{k}) + \eta_2(\mathbf{k} - \mathbf{k}')d_2(\mathbf{k}) \\ \eta_1^*(\mathbf{k} - \mathbf{k}')d_1(\mathbf{k}) + \eta_2^*(\mathbf{k} - \mathbf{k}')d_2(\mathbf{k}) & [i\omega_n + (\varepsilon_{\mathbf{k}} - \mu)]\delta(\mathbf{k} - \mathbf{k}') \end{pmatrix}, \quad (\text{A4})$$

where  $\delta(\mathbf{k} - \mathbf{k}')$  is a Dirac  $\delta$  function. If we expand the bosonic action (A3) in powers of  $\eta_{1,2}$  up to quartic order in the bosonic fields, and neglect any spatial variation (i.e., retain the zero wave-vector component of  $\eta_{1,2}$  only), then we obtain the Landau free energy (2). For the particular fermionic model considered here we obtain  $K_2 = \frac{1}{2}K_1$ , but this result is not generic, and different values of  $K_2$  would be obtained starting from different fermionic models.

To obtain the gradient terms in Eqs. (3) and (4), we only need to work to quadratic order in the bosonic fields  $\eta_{1,2}$ , but we must retain the nonzero wave-vector components (we must retain the spatial variation). The terms proportional to  $J_1$  in Eqs. (3) and (4) may be obtained by means of a standard gradient expansion, as discussed in Ref. 30. However, a standard gradient expansion misses the terms proportional to  $J_2$  in Eqs. (3) and (4). Nonetheless, these terms are symmetry allowed, and so should be present in the action.

Intuitively, the  $J_2$  terms describe the effect of the intrinsic orbital magnetization associated with chiral Cooper pairing—they lead to a coupling between the intrinsic magnetization

and external magnetic fields, and also between the intrinsic magnetization and the magnetic flux in vortices. One way of determining  $J_2$  is to determine the net orbital magnetization arising from chiral Cooper pairing. This approach has been discussed in Ref. 31. However, ideally we would like to derive this term starting from the action (A3).

To obtain the  $J_2$  terms from Eq. (A3) using a gradient expansion, one must take into account the weak particle-hole asymmetry of the spectrum. The origin of these terms is closely related to the chiral anomaly, and is missed by a simple-minded gradient expansion. How to compute the coefficient  $J_2$  using a gradient expansion has been discussed at great length in Ref. 32, however, the calculations required are lengthy and subtle. The magnitude of  $J_2$  is of order  $Nl\mu_B$ , where  $\mu_B$  is the Bohr magneton, and  $N$  is the mean Cooper pair density, as established in Refs. 31 and 32. Since the precise value of  $J_2$  is not important for the present work, we refer the reader to Ref. 32 for more details on how to calculate  $J_2$ . For the present work, we need only the fact that the terms proportional to  $J_2$  are symmetry allowed.

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