Spontaneous spin-triplet exciton condensation in ABC-stacked trilayer graphene

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We study the neutral filling ABC-stacked trilayer graphene with a long-range Coulomb interaction. The extremely large density of states near Dirac points leads to more plausible formation of spin ferromagnets, and our calculation indicates that the spin split energy approximates to 32 meV. Electron and hole carriers coexist in this spontaneous ferromagnetic system. The Coulomb interaction between carriers with opposite charge leads to triplet exciton condensation, namely, a spin superconductor. Using a BCS-type self-consistent calculation, we demonstrate the exciton condensation energy gap is about 7.8 meV. Further, we consider the thermal fluctuations and determine the Berezinskii-Kosterlitz-Thouless critical temperature to be around 11 K.

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I. INTRODUCTION

Recent experimental realization of monolayer and multilayer graphene have opened the possibility of exploring their intriguing electronic properties,[1] which depend dramatically on the number of layers and the stacking sequence. Under low-energy approximation, the ABC-stacked trilayer graphene possesses two cubic bands while that of ABA-stacked trilayer graphene possesses two massless bands and two massive bands around half filling.[2–4] Previous studies have shown that the system is unstable toward the formation of weak ferromagnetic states in graphene multilayers with long-range Coulomb interactions.[5] Such arguments rely on a variational calculation of the ground-state energy.[5] In ABC-stacked trilayer graphene, the density of states (DOS) is extremely large around half-filling,[4] which more plausibly leads to the formation of ferromagnetic states with electron and hole pockets. Our results show that the spin splits energy is approximately about 32 meV.

Researchers have shown that the exciton condensation could be realized in systems with both electrons and holes carrier.[6–8] The Coulomb interaction between spatially separated holes and electrons guarantee the exciton condensation and superfluid states present in such systems. There are also exciton condensations in bilayer electron systems in the quantum Hall regime.[9] Recently, many studies have focused on the exciton condensation in biased graphene bilayers separated by an insulating layer.[10–14] The excitons in these systems are spin unpolarized. By contrast, our former studies have demonstrated the spin-triplet exciton states exist in ferromagnetic (FM) graphene.[15] In FM graphene, the energy and spin difference between electrons and holes prevent the electron-hole (E-H) recombination and ensure the stability of spin-triplet excitons. At low temperature, such a system exhibits spin superconductivity. Accordingly, a crucial issue is to find a ferromagnetic gapless system with large spin split.

Our study indicates ABC-stacked trilayer graphene is a good candidate for spin superconductivity. In this work we show that a noninteracting spin degenerate state is fragile to the longe-range Coulomb interaction. Due to the large DOS around the nodal points, the energy bands can easily spilt into spin ferromagnetic bands. For the neutral filling case, the electron and hole pocket sizes are deduced by a variational method. The spin split energy approximates to 32 meV in this system. Furthermore, we consider the exciton condensation formed by electron and hole carriers with opposite spin configurations. We carry out a BCS-type calculation and obtain the condensation energy gap, which is about 7.8 meV. At low temperature $T < T_C$, the spin-triplet exciton condensation leads to spin superconductivity.[15] Further, we consider the thermal fluctuations and calculate the Berezinskii-Kosterlitz-Thouless (BKT) critical temperature T_{BKT}.

The paper is organized as follows. In Sec. II the model is introduced. In Sec. III we carry out the variational calculation of electron-hole pocket sizes and estimate the superconductivity energy gap by BCS-type formulas. The calculation of BKT temperature is also given in Sec. III. The conclusion is in Sec. IV. Some mathematical details are included in the appendixes.

II. MODEL HAMILTONIAN

The lattice structure for ABC-stacked trilayer graphene is depicted in Fig. 1. For simplicity, we consider the nearest-neighbor coupling between carbon atoms in one layer and only take into account the vertical atoms coupling between the neighboring layers. The low-energy Hamiltonian can be written as: $H_{\mathbf{k}} = \sum \Psi^{+}_{\mathbf{k},\sigma} H(\mathbf{k}) \Psi_{\mathbf{k},\sigma}$, in which $\Psi_{\mathbf{k},\sigma} = (a_{1\mathbf{k},\sigma}, b_{1\mathbf{k},\sigma}, a_{2\mathbf{k},\sigma}, b_{2\mathbf{k},\sigma}, a_{3\mathbf{k},\sigma}, b_{3\mathbf{k},\sigma})$ and $a_{i\mathbf{k},\sigma}(b_{i\mathbf{k},\sigma})$ annihilates an electron with momentum $\mathbf{k}$ on sublattice $a(b)$ in the ith layer ($i = 1,2,3$) with spin σ. The low-energy Hamiltonian for ABC-stacked trilayer graphene around the Dirac point $\overrightarrow{K}$ reads[16]

$$H(\mathbf{k}) = \begin{bmatrix} 0 & v_F|k|e^{i\phi_{\mathbf{k}}} & 0 & 0 & 0 & 0 \\ v_F|k|e^{-i\phi_{\mathbf{k}}} & 0 & -\gamma_1 & 0 & 0 & 0 \\ 0 & -\gamma_1 & 0 & v_F|k|e^{i\phi_{\mathbf{k}}} & 0 & 0 \\ 0 & 0 & v_F|k|e^{-i\phi_{\mathbf{k}}} & 0 & -\gamma_1 & 0 \\ 0 & 0 & 0 & -\gamma_1 & 0 & v_F|k|e^{i\phi_{\mathbf{k}}} \\ 0 & 0 & 0 & 0 & v_F|k|e^{-i\phi_{\mathbf{k}}} & 0 \end{bmatrix}. \quad (1)$$

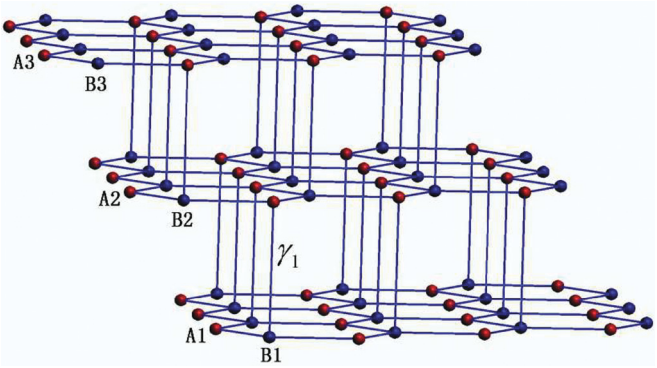

FIG. 1. (Color online) Schematic lattice structure of ABC-stacked trilayer graphene.

Here $\tan\phi_{\mathbf{k}} = -\frac{k_y}{k_x}$, $\gamma_1 \simeq 0.35$ eV is the interlayer coupling strength, $v_F = \frac{3}{2}at$ is the Fermi velocity in Eq. (1), and $t \approx 3$ eV is the nearest-neighbor coupling strength in same layer, the lattice constants $a = 0.142$ nm.

The low-energy dispersion can be obtained by a unitary transformation: $\Psi_{\mathbf{k}} = M_{\mathbf{k}}\Phi_{\mathbf{k}}$, and the explicit form of $M_{\mathbf{k}}$ is given in Appendix A. After the unitary transformation, the Hamiltonian becomes diagonalized and the six energy bands are depicted in Fig. 2. From now on, we focus on the low-energy bands 1 and 2 (labeled by the same index as shown in Fig. 2) around half filling. The energy differences between bands 1 and 2 and bands 3–6 is approximately γ_1.

Around the half filling, the low-energy band denotes the states that mostly occupy sites A on layer 1 and sites B on layer 3. The low-energy Hamiltonian has been given formerly as[4,16]

$$H_{a_1b_3}(\mathbf{k}) = \begin{pmatrix} 0 & \frac{v_F^3(k_x - ik_y)^3}{\gamma_1^2} \\ \frac{v_F^3(k_x + ik_y)^3}{\gamma_1^2} & 0 \end{pmatrix}. \tag{2}$$

As shown by Zhang *et al.*,[4,17] a trilayer graphene is the chiral generalization of a monolayer graphene and a bilayer graphene with $N = 3$ and the density of states $D(E) \sim E^{\frac{2-N}{N}}$ diverges when E approximates to zero. In according with the Stoner criteria, such nearly flat bands make the gapless system very fragile to the electron-electron interaction and susceptible to

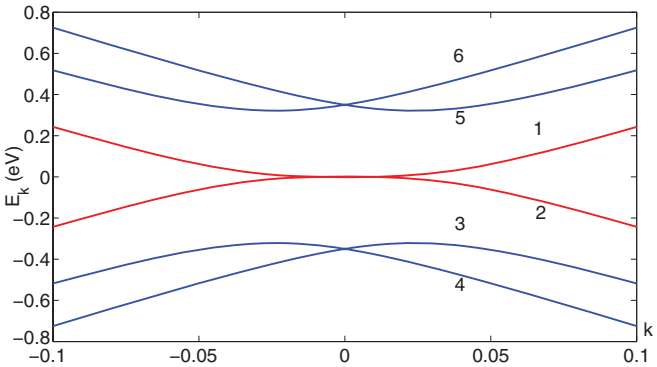

FIG. 2. (Color online) Band dispersion of ABC-stacked trilayer graphene. The bands are labeled by 1–6 corresponding to labels in the context.

form spin split bands. Under the low-energy approximation, the electrons are mostly located on layer 1 and layer 3. The Coulomb interaction can be conveniently written in terms of intralayer and interlayer Coulomb interaction denoted by layer densities in $\mathbf{k}$ space as follows:

$$\rho_i(\mathbf{q}) = \sum_{k,\sigma}\left(\Psi^{a+}_{\mathbf{k}+\mathbf{q},i,\sigma}\Psi^{a}_{\mathbf{k},i,\sigma} + \Psi^{b+}_{\mathbf{k}+\mathbf{q},i,\sigma}\Psi^{b}_{\mathbf{k},i,\sigma}\right). \tag{3}$$

Under the long wave limit, the Coulomb term can be expressed as

$$H_{\text{Coul}} = \frac{1}{2S}\sum_{\mathbf{q}\neq 0}\sum_{i,j}\rho_i(\mathbf{q})\,V_{ij}(\mathbf{q})\,\rho_j(-\mathbf{q}). \tag{4}$$

Here $i,j = 1,3$ denotes the layer index and $V_{ij}(\mathbf{q}) = \frac{2\pi e^2}{\epsilon_0 q}e^{-\alpha_{ij}qd}$ denotes the Coulomb interaction in $\mathbf{k}$ space with $\alpha_{11} = \alpha_{33} = 0$ and $\alpha_{13} = \alpha_{31} = 2$, $d \approx 0.34$ nm is the layer distance. In the following section, we will give out detailed proof that in the neutral filling case the ground state will be ferromagnetic. For convenience, we express the density operators in the diagonalized basis

$$\rho_i(\mathbf{q}) = \sum_{p,\sigma,m,n}\Phi^{+}_{\mathbf{p}+\mathbf{q},m,\sigma}X^{i}_{mn}(\mathbf{p}+\mathbf{q},\mathbf{p})\,\Phi_{\mathbf{p},n,\sigma}, \tag{5}$$

where i denotes the layer index and m,n denote the column indexes corresponding to bands in Fig. 2. Thus the Coulomb interaction can be expressed as[5]

$$H_{\text{Coul}} = \frac{S}{2}\iint\frac{d^2\mathbf{p}d^2\mathbf{q}}{(2\pi)^4}\sum_{ij\sigma\sigma' mn}X^{i}_{mn}(\mathbf{q},\mathbf{p})\,X^{j}_{nm} \times(\mathbf{p},\mathbf{q})\,\mathbf{n}_{m,\sigma}(\mathbf{q})\,\mathbf{n}_{n,\sigma'}(\mathbf{p})\,V_{ij}(\mathbf{p}-\mathbf{q}). \tag{6}$$

Here $\mathbf{n}_{m,\sigma}(\mathbf{q})$ denotes the occupancy and the explicit form of $X^{i}(\mathbf{p},\mathbf{q})$ is given in Appendix B.

III. SPONTANEOUS SPIN SPLIT AND SPIN SUPERCONDUCTIVITY

A. Band ferromagnetism in ABC-stacked trilayer graphene

We consider the neutral filling case and represent the spin split states around each $\mathbf{K}$ point by one electron pocket with spin $\uparrow$ and one hole pocket with spin $\downarrow$. Under the band ferromagnetic condition, the variational states can be expressed as: $\mathbf{n}_{1,\uparrow}(\mathbf{k}) = \Theta(Q-k)$, $\mathbf{n}_{1,\downarrow}(\mathbf{k}) = 0$, and $\mathbf{n}_{2,\uparrow}(\mathbf{k}) = 1, \mathbf{n}_{2\downarrow}(\mathbf{k}) = 1 - \Theta(Q-k)$, as depicted in Fig. 3. The parameter Q needs to be calculated by the lowest-energy condition. With the formation of electron and hole pockets, the kinetic energy cost can be expressed by

$$\Delta E_{\text{kin}} = 2\int_0^{E(Q)}\varepsilon_k\rho(\epsilon_k)\,d\epsilon_k = \frac{Sv_F^3Q^5}{5\pi\gamma_1^2}. \tag{7}$$

Here S denotes the surface area and we assume $\hbar = 1$. The expression for the Coulomb interaction is very lengthy and we give out the leading term as follows:

$$\Delta E_{\text{Coul}} = \frac{-e^2SQ^3}{4\pi\epsilon_0(2\pi)^3}(C_1 - C_2 + C_3Q^2d^2), \tag{8}$$

where $C_1 \approx 1.33$, $C_2 \approx 0.56$, and $C_3 \approx 0.38$. It is easy to verify that $Qd \ll 1$. The explicit expressions for Coulomb

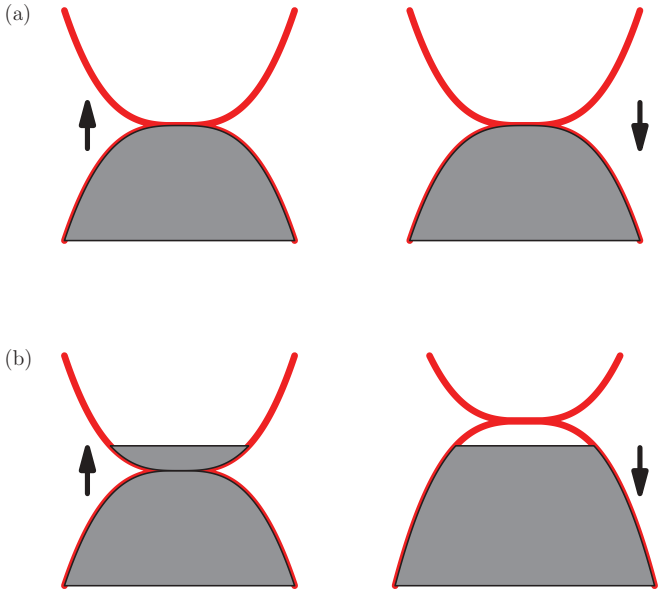

FIG. 3. (Color online) Sketch map of (a) noninteracting bands and (b) spin ferromagnetism bands.

integrals are given in Appendix B. Thus when we consider the energy change by including Eq. (7) and Eq. (8), the variational parameter Q satisfies the relation $v_F Q \sim \sqrt{\frac{3e^2(C_1-C_2)}{2\pi\epsilon_0 v_F}}\gamma_1 \approx 0.09\gamma_1 \approx 32\,\text{meV}$. We need to emphasize that such spin split pockets are much larger than those in bilayer graphene and ABA-stacked trilayer graphene.[5] This large spin split pockets lead to more carriers of spin fluid. In the previous calculation we neglect the Coulomb interaction between different Dirac points. Generally, the Coulomb interaction between different valleys can lead to the pseudospin split. However, the Coulomb interaction strength is very small and expressed by $V \sim \frac{2\pi e^2}{2|K|\epsilon_0}$, thus the pseudospin split is very small compared to real spin split.

B. Exciton condensation and spin superconductivity

Naturally, in such a system with both electron and hole carriers, there might be exciton condensation. For simplicity, we assume the critical energy scale for exciton condensation $T_c \ll v_F Q$, thus the formation of exciton condensation will not change the spin split mechanism in general. In this way, we can formulate the low-energy Hamiltonian for different spin as follows:

$$H_0 = \sum_{k,\sigma} \psi^+_{k,\sigma} \begin{pmatrix} \sigma v_F Q & v_F^3(k_x - ik_y)^3 \\ v_F^3(k_x + ik_y)^3 & \sigma v_F Q \end{pmatrix} \psi_{k,\sigma}.$$
$$U_c = \sum_{s,s';i,j;\sigma,\sigma'} U_{ij}^{ss'} n_{i\sigma}^{s} n_{j\sigma'}^{s'}, \tag{9}$$

where $\psi_{k,\sigma} = (a_{1k\sigma}, b_{3k\sigma})^T$, $s/s' = a,b$ is the lattice index, $n_{i\sigma}^s$ is the local electron number operator on sublattice s of ith site with spin σ. The $\sigma v_F Q$ term with $\sigma = \pm 1$ indicates the band spin splits. For the noninteracting Hamiltonian in Eq. (9), the low-energy bands consist of a spin $\uparrow$ band $\epsilon_{+\uparrow}$ with electron carrier and a spin $\downarrow$ band $\epsilon_{-\downarrow}$ with hole carrier as depicted in Fig. 3(b). We can regard the annihilation operator $\alpha_{k-\downarrow}$ as the creation operator of spin $\uparrow$ hole. Thus, we can define operators $\alpha_{ke\uparrow} = \alpha_{k+\uparrow}$ and $\alpha^+_{kh\uparrow} = \alpha_{k-\downarrow}$.[15] In this way, the interaction between electron and hole can be decoupled by mean-field approximation: $U \approx \sum_k \Delta_k \alpha^+_{ke\uparrow}\alpha^+_{kh\uparrow} + \text{H.c.}$ with the E-H exciton condensation parameter $\Delta_k \equiv -\sum_k U_{kk'}\langle\alpha_{k'e\uparrow}\alpha_{k'h\uparrow}\rangle$, where $U_{kk'}$ denotes the Coulomb interaction in k space.

The energy gap Δ can be estimated self-consistently as follows:

$$\Delta_k = \sum_{k'} \frac{U_{kk'}\Delta_{k'}}{2\xi_k} \tanh\left(\frac{1}{2}\beta\xi_k\right). \tag{10}$$

Here $\beta = \frac{1}{k_B T}$ and the quasiparticle dispersion $\xi_k = \sqrt{(v_F Q - \frac{v_F^3 k^3}{\gamma_1^2})^2 + \Delta_{k'}^2}$. We consider the major contribution of the on-site Coulomb interaction and thus $U_{kk'} = U\Theta(k_D - |\mathbf{k} - \mathbf{k}'|)$, where $v_F k_D \sim \gamma_1$ represents the energy cut-off for low-energy bands. The estimation of U is given out as follows. Taking the screening effect into account, we consider the short-range exciton condensation on nearby lattices, thus the Coulomb interaction can be easily represented in discrete form,

$$U^{ab}_{\mathbf{kk}'} = \sum_{j\delta} U^{ab}_{0j} e^{-i(\mathbf{k}-\mathbf{k}')(\mathbf{r}_j+\vec{\delta})} \approx \sum_{\delta} U^{ab}_{00} e^{-i(\mathbf{k}-\mathbf{k}')\vec{\delta}},$$
$$U^{aa}_{\mathbf{kk}'} = \sum_{j\delta} U^{aa}_{0j} e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{r}_j} \approx U^{aa}_{00}. \tag{11}$$

In Eq. (11) U^{ab}_{0j} and U^{aa}_{0j} represent the Coulomb interaction between carbon atoms (a,b) on site 0 and site j and $\vec{\delta}$ denote the three nearest-neighbor vectors connecting A/B sublattices. In the ferromagnetic bands with electron and hole pockets approximating to $v_F Q \sim 32\,\text{meV}$, the averaged occupancies over sites can be easily calculated as $n_f \approx 0.03$. Given the occupancy number, we obtain the on-site and nearest-neighbor Coulomb energy as $U^{aa}_{00} = \frac{n_f^2 e^2}{4\pi\epsilon_0 d_{\text{carbon}}} \approx 1.5\,\text{eV}$ and $U^{ab}_{00} = \frac{n_f^2 e^2}{4\pi\epsilon_0 a} \approx 0.1\,\text{eV}$, where $d_{\text{carbon}} \sim 10^{-11}$ m is of the order of carbon atom diameter and $a = 1.42 \times 10^{-10}$m is the lattice constant. In the self-consistent calculation, we mainly consider the on-site Coulomb energy and set $U = U^{aa}_{00}$. We solve the self-consistent equation numerically and obtain $\Delta \approx 7.8\,\text{meV}$. We need to emphasize the exciton condensation gap Δ is very small compared with the spin split energy $v_F Q$, thus the formation of excitons only slightly change the size of the spin split pockets. As for the experimental aspects, recent transportation measurements show that there is a spontaneous gap opening at about 6 meV in charge neutral ABC-stacked trilayer graphene,[3] which is consistent with our calculations.

C. Thermal fluctuations and Berezinskii-Kosterlitz-Thouless critical temperature

The BCS-type results obtained above are based on mean-field analysis. In two-dimensional (2D) systems, the thermal fluctuations can significantly induce the BKT phase transition.[12,18] In general, the order parameter can be represented by $\Delta(\tau, r_1, r_2)$. We take the assumption that the modulus of order parameter hardly differs from the mean-field value and only consider the thermal phase fluctuation (we also neglect the quantum fluctuations). Thus the order parameter can be

expressed as the mean-field order parameter multiplied by a location-dependent phase $\triangle(\tau,r_1,r_2) \approx \Delta(r_1-r_2)e^{-i2\theta(R)}$. Under this approximation, the Gor'kov Green's function can be deduced and the effective action can be obtained. The details are given in Appendix C.

The Eq. (C6) in Appendix C corresponds to 2D XY model $H = \frac{J_s}{2}\int d^2\mathbf{r}\,(\nabla\theta)^2$. The self-consistent equation for BKT temperature is readily obtained by using the relation $T_{\mathrm{BKT}} = \frac{\pi}{2}J_s$[19], which reads

$$T_{\mathrm{BKT}} = \frac{9\pi v_F^3}{4\gamma_1^2}\iint \frac{d^2\mathbf{k}k}{(2\pi)^2}\left[1-\frac{\varepsilon_k - v_F Q}{\xi_k}\tanh\left(\frac{\xi_k}{2k_BT}\right)\right] - \frac{9\pi v_F^6}{4\gamma_1^4}\iint \frac{d^2\mathbf{k}}{(2\pi)^2}\frac{k^4}{k_BT\,\cosh^2\left(\frac{\xi_k}{2k_BT}\right)}. \tag{12}$$

Here, $\varepsilon_k = \frac{v_F^3k^3}{\gamma_1^2}$ is the dispersion of ABC-stacked trilayer graphene. We consider the low-energy bands with energy cutoff $\Theta(\gamma_1-\varepsilon_k)$, and the numerical results show $T_{\mathrm{BKT}} = 11$ K. This type of spin conductivity differ from exciton BEC in biased graphene bilayers[10–14] and the experimental condition is under the ability of present techniques.

IV. CONCLUSION

In summary, we have studied the neutral filling ABC-stacked trilayer graphene with a long-range Coulomb interaction. We have shown that the noninteracting ground state is unstable towards forming a spin ferromagnet. The energy of electron and hole pockets are deduced by a variational method. The extremely large DOS near Dirac points leads to more plausible formation of spin ferromagnets. Our calculation indicates the spin split energy approximates to 32 meV. This spontaneous ferromagnetic trilayer graphene possesses both electron and hole carriers. According to a previous study,[15] the Coulomb interaction between electron and hole carriers leads to spin-triplet exciton condensation. Using a BCS-type self-consistent calculation, we have obtained the energy gap is about 7.8 meV. Furthermore, we take into account the thermal fluctuations and determine the BKT critical temperature in this system. Such spin superconductor can be detected experimentally.[15]

ACKNOWLEDGMENTS

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APPENDIX A: UNITARY TRANSFORM MATRIX

The eigenmatrix of $H(\mathbf{k})$ can be written as $M_{\mathbf{k}} = M_1(\mathbf{k})M_2M_3(\mathbf{k})M_4$, where

$$M_1(\mathbf{k}) = \mathrm{diag}[\,1\ e^{-i\phi_{\mathbf{k}}}\ 1\ e^{-i\phi_{\mathbf{k}}}\ 1\ e^{-i\phi_{\mathbf{k}}}\,] \tag{A1}$$

is a gauge transformation, and

$$M_2 = \begin{bmatrix}1&0&0&0&0&0\\0&0&1&0&0&0\\0&0&0&1&0&0\\0&0&0&0&1&0\\0&0&0&0&0&1\\0&1&0&0&0&0\end{bmatrix} \tag{A2}$$

is a redistribution of basis vectors. After unitary transformation by $M_1(\mathbf{k})M_2$, the Hamiltonian reads,

$$H_2(\mathbf{k}) = \begin{bmatrix}0&0&v_Fk&0&0&0\\0&0&0&0&0&v_Fk\\v_Fk&0&0&-\gamma_1&0&0\\0&0&-\gamma_1&0&v_Fk&0\\0&0&0&v_Fk&0&-\gamma_1\\0&v_Fk&0&0&-\gamma_1&0\end{bmatrix}. \tag{A3}$$

The matrix $H_2(\mathbf{k})$ can be decoupled into two parts: the block diagonal part H_2^I and block off-diagonal part H_2^{II}

$$H_2^I(\mathbf{k}) = \begin{bmatrix}0&0&0&0&0&0\\0&0&0&0&0&0\\0&0&0&-\gamma_1&0&0\\0&0&-\gamma_1&0&v_Fk&0\\0&0&0&v_Fk&0&-\gamma_1\\0&0&0&0&-\gamma_1&0\end{bmatrix}, \tag{A4}$$

$$H_2^{II}(\mathbf{k}) = \begin{bmatrix}0&0&v_Fk&0&0&0\\0&0&0&0&0&v_Fk\\v_Fk&0&0&0&0&0\\0&0&0&0&0&0\\0&0&0&0&0&0\\0&v_Fk&0&0&0&0\end{bmatrix}. \tag{A5}$$

Thus, the coupling between the low-energy part and the high-energy part in $H_2(\mathbf{k})$ can be transformed by Lowdin partitioning uniform transformation:[20] $e^{-S}H_2e^S \approx H_2^I + [H_2^{II},S] + H_2^{II} + [H_2^I,S]$, where

$$S = \begin{bmatrix}0&\chi\\-\chi^T&0\end{bmatrix}, \tag{A6}$$

$$\chi = \begin{pmatrix}0&-\frac{v_Fk}{\gamma_1}&0&-\frac{v_F^2k^2}{\gamma_1^2}\\-\frac{v_F^2k^2}{\gamma_1^2}&0&-\frac{v_Fk}{\gamma_1}&0\end{pmatrix}. \tag{A7}$$

Thus the unitary matrix $M_3 = e^S$ reads

$$M_3 = \begin{bmatrix}1&0&0&-\frac{v_Fk}{\gamma_1}&0&-\frac{v_F^2k^2}{\gamma_1^2}\\0&1&-\frac{v_F^2k^2}{\gamma_1^2}&0&-\frac{v_Fk}{\gamma_1}&0\\0&\frac{v_F^2k^2}{\gamma_1^2}&1&0&0&0\\\frac{v_Fk}{\gamma_1}&0&0&1&0&0\\0&\frac{v_Fk}{\gamma_1}&0&0&1&0\\\frac{v_F^2k^2}{\gamma_1^2}&0&0&0&0&1\end{bmatrix}. \tag{A8}$$

Finally, the Hamiltonian needs to recombine to form the symmetric and antisymmetric bands by

$$M_4 = \frac{1}{\sqrt{2}}\begin{bmatrix}1&1\\1&-1\end{bmatrix}\otimes I_{3\times3}.$$

Thus, the overall unitary transform matrix can be given out by the following relation:

$$M_{\mathbf{k}}=\frac{1}{\sqrt{2}}\begin{bmatrix} 1 & 1 & -\frac{v_F k}{\gamma_1} & \frac{v_F k}{\gamma_1} & -\frac{v_F^2 k^2}{\gamma_1^2} & \frac{v_F^2 k^2}{\gamma_1^2} \\ e^{-i\phi_{\mathbf{k}}}\frac{v_F^2 k^2}{\gamma_1^2} & -e^{-i\phi_{\mathbf{k}}}\frac{v_F^2 k^2}{\gamma_1^2} & e^{-i\phi_{\mathbf{k}}} & e^{-i\phi_{\mathbf{k}}} & 0 & 0 \\ \frac{v_F k}{\gamma_1} & \frac{v_F k}{\gamma_1} & 1 & -1 & 0 & 0 \\ e^{-i\phi_{\mathbf{k}}}\frac{v_F k}{\gamma_1} & -e^{-i\phi_{\mathbf{k}}}\frac{k}{\gamma_1} & 0 & 0 & e^{-i\phi_{\mathbf{k}}} & e^{-i\phi_{\mathbf{k}}} \\ \frac{v_F^2 k^2}{\gamma_1^2} & \frac{v_F^2 k^2}{\gamma_1^2} & 0 & 0 & 1 & -1 \\ e^{-i\phi_{\mathbf{k}}} & -e^{-i\phi_{\mathbf{k}}} & -e^{-i\phi_{\mathbf{k}}}\frac{v_F^2 k^2}{\gamma_1^2} & -e^{-i\phi_{\mathbf{k}}}\frac{v_F^2 k^2}{\gamma_1^2} & -e^{-i\phi_{\mathbf{k}}}\frac{v_F k}{\gamma_1} & -e^{-i\phi_{\mathbf{k}}}\frac{v_F k}{\gamma_1} \end{bmatrix}. \tag{A9}$$

APPENDIX B: COULOMB INTERACTION INTEGRAL

In Eq. (5), the matrix that associates carriers in layer 1 or layer 3 with different band indexes can be expressed as follows:

$$X^1(\mathbf{q},\mathbf{p})=M_{\mathbf{q}}^{+}\begin{bmatrix} I_{2\times 2} & 0 \\ 0 & 0_{4\times 4} \end{bmatrix} M_{\mathbf{p}} \tag{B1}$$

$$X^3(\mathbf{q},\mathbf{p})=M_{\mathbf{q}}^{+}\begin{bmatrix} 0_{4\times 4} & 0 \\ 0 & I_{2\times 2} \end{bmatrix} M_{\mathbf{p}}. \tag{B2}$$

With the variational ground states that $\triangle\mathbf{n}_{1,\uparrow}(\mathbf{k})=\Theta(Q-k)$ and $\triangle\mathbf{n}_{2\downarrow}(\mathbf{k})=-\Theta(Q-k)$, we just need to calculate the matrix elements that represent coupling between band 1 and band 2 in $X^1(\mathbf{q},\mathbf{p})$ and $X^3(\mathbf{q},\mathbf{p})$,

$$X_{12}^1(\mathbf{q},\mathbf{p})=\frac{1}{2}-\frac{e^{i(\phi_{\mathbf{q}}-\phi_{\mathbf{p}})}v_F^4 p^2 q^2}{2\gamma_1^4}, \qquad X_{12}^3(\mathbf{q},\mathbf{p})=-e^{i(\phi_{\mathbf{q}}-\phi_{\mathbf{p}})}X_{12}^1(\mathbf{q},\mathbf{p}). \tag{B3}$$

Thus the Coulomb interaction can be easily expressed as

$$\begin{aligned}\frac{E_{\mathrm{Coul}}}{S}=\frac{1}{2}\int\frac{d^2\mathbf{p}d^2\mathbf{q}}{(2\pi)^2}\big[&2\left|X_{\mathbf{pq}}^1(1,2)\right|^2\Theta(Q-p)\Theta(Q-q)V_{11}(\mathbf{q}-\mathbf{p})+2\mathrm{Re}\left|X_{\mathbf{pq}}^1(1,2)X_{\mathbf{pq}}^3(1,2)\right| \\ &\times\Theta(Q-p)\Theta(Q-q)V_{13}(\mathbf{q}-\mathbf{p})\big],\end{aligned} \tag{B4}$$

and $\tan(\phi_{\mathbf{p}})=\frac{-p_y}{p_x}$, the first integration in Eq. (B2) denotes the intralayer Coulomb interaction and can be expressed as

$$\begin{aligned}&\iint\frac{d^2\mathbf{p}d^2\mathbf{q}}{(2\pi)^4}\left[\frac{1}{4}-\cos[\phi_{\mathbf{q}}-\phi_{\mathbf{p}}]\frac{p^2q^2}{2\gamma_1^4}+\frac{1}{4}\left(\frac{p^2q^2}{\gamma_1^4}\right)^2\right]\frac{-1}{|\mathbf{p}-\mathbf{q}|}\frac{2\pi e}{\epsilon_0} \\ &=-\iiint\frac{d\theta dpdq}{(2\pi)^3}pq\left[\frac{1}{2}-\cos\theta\,\frac{p^2q^2}{\gamma_1^4}+\frac{1}{2}\left(\frac{p^2q^2}{\gamma_1^4}\right)^2\right]\frac{1}{\sqrt{p^2+q^2-2pq\cos\theta}}\frac{\pi e}{\epsilon_0}.\end{aligned} \tag{B5}$$

We assume that $\frac{p}{Q}=x$ and $\frac{q}{Q}=y$ then Eq. (B5) is expressed by the following parts:

$$-\iiint\frac{d\theta dpdq}{(2\pi)^3}pq\frac{1}{2\sqrt{p^2+q^2-2pq\cos\theta}}\approx-\frac{Q^3}{2(2\pi)^3}\times 1.33, \tag{B6}$$

$$\iiint\frac{d\theta dpdq}{(2\pi)^3}pq\frac{p^2q^2}{\gamma_1^4}\frac{\cos\theta}{\sqrt{p^2+q^2-2pq\cos\theta}}\approx\frac{Q^3}{(2\pi)^3}\frac{Q^4}{\gamma_1^4}\times 0.16,. \tag{B7}$$

$$\iiint\frac{d\theta dpdq}{(2\pi)^3}pq\left(\frac{p^2q^2}{\gamma_1^4}\right)^2\frac{1}{2\sqrt{p^2+q^2-2pq\cos\theta}}\approx\frac{-Q^3}{2(2\pi)^3}\left(\frac{Q^4}{\gamma_1^4}\right)^2\times 0.14. \tag{B8}$$

In the next step, we calculate the interlayer Coulomb interaction in Eq. (B2) denoted by integrals in Eq. (B9). Assuming the carrier pocket is small, thus we have $Qd\ll 1$, one can expand $e^{-2|\mathbf{p}-\mathbf{q}|d}$ by its Taylor series and keep the leading terms. The interlayer Coulomb interaction is given in Eq. (B9) and expressed by Eqs. (B9)–(B11).

$$\frac{1}{2}\iiint\frac{d\theta dpdq}{(2\pi)^3}pq\left[-\frac{1}{2}\cos\theta+\frac{p^2q^2}{\gamma_1^4}-\frac{1}{2}\left(\frac{p^2q^2}{\gamma_1^4}\right)^2\cos(\phi_{\mathbf{q}}-\phi_{\mathbf{p}})\right]\frac{-e^{-2|\mathbf{p}-\mathbf{q}|d}}{|\mathbf{p}-\mathbf{q}|}\frac{2\pi e}{\epsilon_0}, \tag{B9}$$

$$\frac{1}{2}\frac{Q^3}{(2\pi)^3}\int_0^{\pi}\int_0^1\int_0^1\frac{d\theta dxdy\,xy\cos\theta[1+2Q^2d^2(x^2+y^2-2xy\cos\theta)]}{\sqrt{x^2+y^2-2xy\cos\theta}}\approx\frac{1}{2}\frac{Q^3}{(2\pi)^3}[0.56-0.38Q^2d^2], \tag{B10}$$

$$-\frac{Q^3}{(2\pi)^3}\frac{Q^4}{\gamma_1^4}\int_0^\pi\int_0^1\int_0^1\frac{d\theta dx dy\, x^3y^3}{\sqrt{x^2+y^2-2xy\cos\theta}}\approx-\frac{Q^3}{(2\pi)^3}\frac{Q^4}{\gamma_1^4}\times 0.32, \tag{B11}$$

$$\frac{1}{2}\left(\frac{Q^4}{\gamma_1^4}\right)^2\frac{Q^3}{(2\pi)^3}\int_0^\pi\int_0^1\int_0^1\frac{d\theta dx dy\, x^5y^5\cos\theta}{\sqrt{x^2+y^2-2xy\cos\theta}}\approx\frac{1}{2}\left(\frac{Q^4}{\gamma_1^4}\right)^2\frac{Q^3}{(2\pi)^3}\times 0.08. \tag{B12}$$

APPENDIX C: DERIVATION OF SUPERFLUID DENSITY J_s

In general, the quantum partition function of superconductor has the formal expression after integration of the Grassmann fields[19]

$$\mathcal{Z}=\int D(\Delta^*,\Delta)\exp\left[-\frac{1}{U}\int d\tau d^2r\,|\Delta|^2+\ln\det\hat{\mathcal{G}}^{-1}\right]. \tag{C1}$$

The full Gor'kov Green's function coupled with electromagnetic potential takes the form

$$\hat{\mathcal{G}}^{-1}=\begin{pmatrix}-\partial_\tau-i\phi-\epsilon_k(-i\nabla-\mathbf{A})+\mu & \Delta e^{i2\theta}\\ \Delta e^{-i2\theta} & -\partial_\tau+i\phi+\epsilon_k(-i\nabla+\mathbf{A})-\mu\end{pmatrix}. \tag{C2}$$

After gauge transformation $\hat{U}=\left(\begin{smallmatrix}e^{-i\theta} & 0\\ 0 & e^{i\theta}\end{smallmatrix}\right)$, the Gor'kov Green's function can be expressed as

$$\hat{\mathcal{G}}^{-1}=\begin{pmatrix}-\partial_\tau-i(\phi+\partial_\tau\theta)-\epsilon_k(-i\nabla-\mathbf{A}-\nabla\theta)+\mu & \Delta\\ \Delta & -\partial_\tau+i(\phi+\partial_\tau\theta)+\epsilon_k(-i\nabla+\mathbf{A}+\nabla\theta)-\mu\end{pmatrix}. \tag{C3}$$

For cubic band dispersion case with $\epsilon_k(-i\nabla)=m|-i\nabla|^3$ and $m=\frac{v_F^3}{\gamma_1^2}$, we assume the field and its gradient are weak. Thus, one can expand the Gor'kov Green's function in Taylor series

$$\hat{\mathcal{G}}^{-1}=\underbrace{-\sigma_0\partial_\tau-\sigma_3[\epsilon_k(-i\nabla)-\mu]+\sigma_1\Delta}_{\hat{\mathcal{G}}_0^{-1}}\underbrace{-i\sigma_3(\phi+\partial_\tau\theta)+3\sigma_0 m[|-i\nabla|(-i\nabla)\cdot(A+\nabla\theta)]}_{X_1}$$
$$\times\underbrace{-\frac{9}{4}\sigma_3 m[|-i\nabla|(A+\nabla\theta)^2]}_{X_2}+\cdots. \tag{C4}$$

The system action can be expressed by the Taylor series,

$$\mathcal{S}=-\ln\det\hat{\mathcal{G}}^{-1}=-\mathrm{tr}\ln(\hat{\mathcal{G}}^{-1})=-\mathrm{tr}\ln\left(\hat{\mathcal{G}}_0^{-1}\right)+\mathrm{tr}(\hat{\mathcal{G}}_0X_1)+\mathrm{tr}\left(\hat{\mathcal{G}}_0X_2+\tfrac{1}{2}\hat{\mathcal{G}}_0X_1\hat{\mathcal{G}}_0X_1\right)+\cdots. \tag{C5}$$

In Eq. (C5), the first part denotes the superfluid action without coupling with electromagnetic fields, and the second part denotes the electronic and magnetic static coupling with superfluid and can be eliminated by static coupling with the positive charge irons. The third term in Eq. (C5) represents the Goldstone mode action. From now on, we neglect the electromagnetic field and only retain the phase fluctuation part, thus the Goldstone action can be expressed as

$$S^{[2]}=\frac{1}{2}\mathrm{tr}(\hat{\mathcal{G}}_0X_1\hat{\mathcal{G}}_0X_1)=\frac{J_s}{2}\int d\tau d^2\mathbf{r}(\nabla\theta)^2, \tag{C6}$$

and the superfluid density is

$$J_s=\frac{9v_F^3}{2\gamma_1^2}\iint\frac{d^2\mathbf{k}k}{(2\pi)^2}\left[1-\frac{\varepsilon_k-v_FQ}{\xi_k}\tanh\left(\frac{\xi_k}{2k_BT}\right)\right]-\frac{9v_F^6}{2\gamma_1^4}\iint\frac{d^2\mathbf{k}}{(2\pi)^2}\frac{k^4}{k_BT\,\cosh^2\left(\frac{\xi_k}{2k_BT}\right)}. \tag{C7}$$

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