Supplementary Information for " Charge-4e superconductivity and chiral metal in the $45^\circ-$ twisted bilayer cuperates and similar materials "

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DERIVATION OF THE EFFECTIVE HAMILTONIAN FROM GINZBURG-LANDAU THEORY

In this section, we derive the effective Hamiltonian appearing in the Eq. (12) of the main text by expanding the Ginzburg-Landau (G-L) free energy up to the fourth-order term of the order parameters.

Symmetry

To elucidate the effect of the symmetry operations on the argument of the G-L free-energy functional, let's start from the mean-field BCS Hamiltonian:

$$H_{\text{BCS-MF}} = H_{\text{TB}} + \sum_{\mathbf{r},\delta} c^\dagger_{\mathbf{r},\text{t}\uparrow} c^\dagger_{\mathbf{r}+\delta,\text{t}\downarrow} \Gamma^{(\text{t})}(\delta)\psi_{\text{t}}(\mathbf{r}) + c^\dagger_{\mathbf{r},\text{b}\uparrow} c^\dagger_{\mathbf{r}+\delta,\text{b}\downarrow} \Gamma^{(\text{b})}(\delta)\psi_{\text{b}}(\mathbf{r}) + h.c. \tag{1}$$

Here $\mathbf{r}$ labels the center-of-mass coordinate of a Cooper pair and δ is the relative coordinate between the two electrons within a Cooper pair. $\Gamma^{(\mu)}(\delta)$ is the fixed normalized real form factor with $\mu = \text{t/b}$, and $\psi_\mu(\mathbf{r})$ is a slowly-varying "envelope" function describing the spatial fluctuation of the complex pairing amplitude at finite temperature. Each symmetry operation first acts on the c and $c^\dagger$ operators, then through a dummy-index transformation, the effect is transferred to the action of the Γ and ψ. As the Γ has simple transformation rule under the symmetry, i.e. it changes sign upon every C^1_n operation and changes or does not change upon the mirror reflection operation, the effect can be transferred purely to ψ. Therefore, we have chosen an gauge in which each symmetry operation only acts on $\psi_\mu(\mathbf{r})$.

Under $\widetilde{C}^1_{2n}$, the spatial dependent pairing amplitudes change to:

$$\psi_{\text{b}}(\mathbf{r}) \to \widetilde{\psi}_{\text{b}}(\mathbf{r}) = \psi_{\text{t}}(\widehat{P}^{-1}_{\frac{\pi}{n}}\mathbf{r}), \qquad \psi_{\text{t}}(\mathbf{r}) \to \widetilde{\psi}_{\text{t}}(\mathbf{r}) = -\psi_{\text{b}}(\widehat{P}^{-1}_{\frac{\pi}{n}}\mathbf{r}). \tag{2}$$

Under the mirror reflection operation $\widehat{P}$, it is easy to prove (we have chosen a gauge without loss of generality):

$$\psi_{\text{b}}(\mathbf{r}) \to \widetilde{\psi}_{\text{b}}(\mathbf{r}) = -\psi_{\text{b}}(\widehat{P}^{-1}\mathbf{r}), \qquad \psi_{\text{t}}(\mathbf{r}) \to \widetilde{\psi}_{\text{t}}(\mathbf{r}) = \psi_{\text{t}}(\widehat{P}^{-1}\mathbf{r}). \tag{3}$$

For convenience, we rotate the basis to $\psi_\pm = \psi_t \pm i\psi_b$ and rewrite the above transformation in the $\mathbf{k}-$space

$$\begin{aligned} \psi_+(\mathbf{k}) &\xrightarrow{\widetilde{C}^1_{2n}} e^{i\pi/2}\psi_+(\widehat{P}^{-1}_{\frac{\pi}{n}}\mathbf{k}), \qquad \psi_-(\mathbf{k}) \xrightarrow{\widetilde{C}^1_{2n}} e^{-i\pi/2}\psi_-(\widehat{P}^{-1}_{\frac{\pi}{n}}\mathbf{k}) \\ \psi_+(\mathbf{k}) &\xrightarrow{\widehat{P}} \psi_-(\widehat{P}^{-1}\mathbf{k}), \qquad \psi_-(\mathbf{k}) \xrightarrow{\widehat{P}} \psi_+(\widehat{P}^{-1}\mathbf{k}). \end{aligned} \tag{4}$$

Here we consider the $\widetilde{C}^1_{2n}$ and the mirror reflection, but neglect the time-reversal symmetry. The final effect of the time-reversal symmetry on the Hamiltonian is consistent with that obtained with only considering the $\widetilde{C}^1_{2n}$ and the mirror reflection symmetries.

With the definition $\mathbf{k}_\pm = k_x \pm ik_y$, we obtain the momentum transformation relations:

$$\widehat{P}_{\frac{\pi}{n}}\mathbf{k}_+ = e^{i\pi/n}\mathbf{k}_+, \qquad \widehat{P}_{\frac{\pi}{n}}\mathbf{k}_- = e^{-i\pi/n}\mathbf{k}_-. \tag{5}$$

The second-order G-L expansion

Up to the lowest-order expansion, the differential term in G-L free energy has the following general form in the $\mathbf{k}-$ space:

$$\begin{aligned} F_0^{(2)} &= \sum_{\mathbf{k}} \psi_+^*(\mathbf{k})\psi_+(\mathbf{k})(a_1\mathbf{k}_+^2 + b_1\mathbf{k}_-^2 + c_1\mathbf{k}_+\mathbf{k}_-) \\ &+ \sum_{\mathbf{k}} \psi_+^*(\mathbf{k})\psi_-(\mathbf{k})(a_2\mathbf{k}_+^2 + b_2\mathbf{k}_-^2 + c_2\mathbf{k}_+\mathbf{k}_-) \\ &+ \sum_{\mathbf{k}} \psi_-^*(\mathbf{k})\psi_+(\mathbf{k})(a_3\mathbf{k}_+^2 + b_3\mathbf{k}_-^2 + c_3\mathbf{k}_+\mathbf{k}_-) \\ &+ \sum_{\mathbf{k}} \psi_-^*(\mathbf{k})\psi_-(\mathbf{k})(a_4\mathbf{k}_+^2 + b_4\mathbf{k}_-^2 + c_4\mathbf{k}_+\mathbf{k}_-). \end{aligned} \tag{6}$$

Under the operation $\widetilde{C}_{2n}^1$, $F_0^{(2)}$ change to:

$$\begin{aligned} F_0^{(2)} \xrightarrow{\widetilde{C}_{2n}^1} &= \sum_{\mathbf{k}} \psi_+^*(\mathbf{k})\psi_+(\mathbf{k})(a_1e^{i2\pi/n}\mathbf{k}_+^2 + b_1e^{-i2\pi/n}\mathbf{k}_-^2 + c_1\mathbf{k}_+\mathbf{k}_-) \\ &+ \sum_{\mathbf{k}} e^{-i2\pi/2}\psi_+^*(\mathbf{k})\psi_-(\mathbf{k})(a_2e^{i2\pi/n}\mathbf{k}_+^2 + b_2e^{-i2\pi/n}\mathbf{k}_-^2 + c_2\mathbf{k}_+\mathbf{k}_-) \\ &+ \sum_{\mathbf{k}} e^{i2\pi/2}\psi_-^*(\mathbf{k})\psi_+(\mathbf{k})(a_3e^{i2\pi/n}\mathbf{k}_+^2 + b_3e^{-i2\pi/n}\mathbf{k}_-^2 + c_3\mathbf{k}_+\mathbf{k}_-) \\ &+ \sum_{\mathbf{k}} \psi_-^*(\mathbf{k})\psi_-(\mathbf{k})(a_4e^{i2\pi/n}\mathbf{k}_+^2 + b_4e^{-i2\pi/n}\mathbf{k}_-^2 + c_4\mathbf{k}_+\mathbf{k}_-). \end{aligned} \tag{7}$$

As $n = 4$ or 6, the invariance of $F_0^{(2)}$ requires only $c_1, c_4 \neq 0$ while all the other coefficients keep zero. Further more, $c_1 = c_4 = B$ is required by the mirror-reflection symmetry. Changing back to the real space, we arrive at the form of $F_0^{(2)}$ as following:

$$\begin{aligned} F_0^{(2)} &= B\int d^2\mathbf{r}[(\nabla\psi_+^*)\cdot(\nabla\psi_+) + (\nabla\psi_-^*)\cdot(\nabla\psi_-)] \\ &= B\psi_0^2\int d^2\mathbf{r}[\nabla(e^{-i\theta_t} - ie^{-i\theta_b})\cdot\nabla(e^{i\theta_t} + ie^{i\theta_b}) + \nabla(e^{-i\theta_t} + ie^{-i\theta_b})\cdot\nabla(e^{i\theta_t} - ie^{i\theta_b})] \\ &= 2B\psi_0^2\int d^2\mathbf{r}[(\nabla\theta_b)^2 + (\nabla\theta_t)^2] \\ &= 4B\psi_0^2\int d^2\mathbf{r}[|\nabla\theta_+|^2 + |\nabla\theta_-|^2] \end{aligned} \tag{8}$$

Here ψ_0 represents the amplitude of the pairing order parameter.

The fourth-order G-L expansion

According to the second order expansion of the differential term in the G-L free energy, the coefficients before θ_+ and θ_- are the same. To get different coefficients, we need expand F_0 to the fourth order with the general form as:

$$F_0^{(4)} = \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \psi_\alpha^*(\mathbf{k}_1)\psi_\beta^*(\mathbf{k}_2)\psi_\gamma(\mathbf{k}_3)\psi_\nu(\mathbf{k}_4)(\sum_{i,j=1}^{4}\alpha_{ij}\mathbf{k}_{+i}\cdot\mathbf{k}_{+j} + \beta_{ij}\mathbf{k}_{-i}\cdot\mathbf{k}_{-j} + \gamma_{ij}\mathbf{k}_{+i}\cdot\mathbf{k}_{-j} + \gamma_{ij}\mathbf{k}_{-i}\cdot\mathbf{k}_{+j}) \tag{9}$$

where $\alpha, \beta, \gamma, \nu = \pm$. It is easy to verify that $\alpha + \beta + \gamma + \nu$ should be an even integer. Since the angular momentum of $\psi_\alpha^{(*)}(\mathbf{k}_i)$ is $\pm n/2$, that of $\psi_\alpha^*(\mathbf{k}_1)\psi_\beta^*(\mathbf{k}_2)\psi_\gamma(\mathbf{k}_3)\psi_\nu(\mathbf{k}_4)$ should be an integer times n. And the angular momentum of $k_\pm$ is ± 1. The invariance of $F_0^{(4)}$ under $\widetilde{C}_{2n}^1$ requires that the total angular momentum should be zero. For $n = 4$

or 6, the restriction of zero total angular momentum dictates $\alpha_{ij} = \beta_{ij} = 0$. Then, we can simplify the general form of $F_0^{(4)}$:

$$F_0^{(4)} = \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \psi_+^*(\mathbf{k}_1)\psi_+^*(\mathbf{k}_2)\psi_+(\mathbf{k}_3)\psi_+(\mathbf{k}_4)(\sum_{i,j=1}^{4} 2\gamma_{ij}^{(1)}\mathbf{k}_i\cdot\mathbf{k}_j)$$
$$+ \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \psi_+^*(\mathbf{k}_1)\psi_-^*(\mathbf{k}_2)\psi_+(\mathbf{k}_3)\psi_-(\mathbf{k}_4)(\sum_{i,j=1}^{4} 2\gamma_{ij}^{(2)}\mathbf{k}_i\cdot\mathbf{k}_j)$$
$$+ \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \psi_-^*(\mathbf{k}_1)\psi_-^*(\mathbf{k}_2)\psi_-(\mathbf{k}_3)\psi_-(\mathbf{k}_4)(\sum_{i,j=1}^{4} 2\gamma_{ij}^{(3)}\mathbf{k}_i\cdot\mathbf{k}_j) \quad (10)$$

We now consider the first and the third term in the general form of $F_0^{(4)}$ since there is only ψ_+ or ψ_-. It is easy to verify that $\psi_\pm \to \psi_\mp^*$ under TRS. Remembering all the transformation relation in mind, the form of equation (10) can be further simplified as:

$$F_{0(1,3)}^{(4)} = \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} [\psi_+^*(\mathbf{k}_1)\psi_+^*(\mathbf{k}_2)\psi_+(\mathbf{k}_3)\psi_+(\mathbf{k}_4) + \psi_-^*(\mathbf{k}_1)\psi_-^*(\mathbf{k}_2)\psi_-(\mathbf{k}_3)\psi_-(\mathbf{k}_4)]$$
$$\cdot[a(\mathbf{k}_1^2+\mathbf{k}_2^2+\mathbf{k}_3^2+\mathbf{k}_4^2) + b(\mathbf{k}_1\cdot\mathbf{k}_2+\mathbf{k}_3\cdot\mathbf{k}_4) + c(\mathbf{k}_1+\mathbf{k}_2)\cdot(\mathbf{k}_3+\mathbf{k}_4)] \quad (11)$$

A valuable equation $(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)^2 = 0$ should be emphasized before the proceeding process. Expanding this equation, we have:

$$\sum_{i=1}^{4}\mathbf{k}_i^2 = 2(\mathbf{k}_1+\mathbf{k}_2)\cdot(\mathbf{k}_3+\mathbf{k}_4) - 2(\mathbf{k}_1\cdot\mathbf{k}_2+\mathbf{k}_3\cdot\mathbf{k}_4). \quad (12)$$

We can rewrite the first and third term:

$$F_{0(1,3)}^{(4)} = \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} [\psi_+^*(\mathbf{k}_1)\psi_+^*(\mathbf{k}_2)\psi_+(\mathbf{k}_3)\psi_+(\mathbf{k}_4) + \psi_-^*(\mathbf{k}_1)\psi_-^*(\mathbf{k}_2)\psi_-(\mathbf{k}_3)\psi_-(\mathbf{k}_4)]$$
$$\cdot[(b-2a)\cdot(\mathbf{k}_1\cdot\mathbf{k}_2+\mathbf{k}_3\cdot\mathbf{k}_4) + (c+2a)\cdot(\mathbf{k}_1+\mathbf{k}_2)\cdot(\mathbf{k}_3+\mathbf{k}_4)] \quad (13)$$

By the same method, the second term of the fourth order expansion of the differential term in G-L free energy is

$$F_{0(2)}^{(4)} = \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \psi_+^*(\mathbf{k}_1)\psi_-^*(\mathbf{k}_2)\psi_+(\mathbf{k}_3)\psi_-(\mathbf{k}_4)$$
$$\cdot\ [a'(\mathbf{k}_1^2+\mathbf{k}_2^2+\mathbf{k}_3^2+\mathbf{k}_4^2) + b'(\mathbf{k}_1\cdot\mathbf{k}_2+\mathbf{k}_3\cdot\mathbf{k}_4) + c'(\mathbf{k}_1\cdot\mathbf{k}_3+\mathbf{k}_2\cdot\mathbf{k}_4) + d'(\mathbf{k}_1\cdot\mathbf{k}_4+\mathbf{k}_2\cdot\mathbf{k}_3)] \quad (14)$$

Transforming to the real space, the total form of $F_0^{(4)}$ is:

$$\begin{aligned}
F^{(4)} &= -(b-2a)\int d^2\mathbf{r}[(\nabla\psi_+^*)^2\psi_+^2 + (\psi_+^*)^2(\nabla\psi_+)^2 + (\nabla\psi_-^*)^2\psi_-^2 + (\psi_-^*)^2(\nabla\psi_-)^2] \\
&+ (c+2a)\int d^2\mathbf{r}[\nabla(\psi_+^{*2})\cdot\nabla(\psi_+^2) + \nabla(\psi_-^{*2})\cdot\nabla(\psi_-^2)] \\
&- (b'-2a')\int d^2\mathbf{r}[(\nabla\psi_+^*)\cdot(\nabla\psi_-^*)\psi_+\psi_- + \psi_+^*\psi_-^*(\nabla\psi_+)\cdot(\nabla\psi_-)] \\
&+ (c'+2a')\int d^2\mathbf{r}[\nabla\psi_+^*\cdot\nabla\psi_+|\psi_-|^2 + |\psi_+|^2\nabla\psi_-^*\cdot\nabla\psi_-] \\
&+ (d'+2a')\int d^2\mathbf{r}[\nabla\psi_+^*\cdot\nabla\psi_-\psi_-^*\psi_+ + \nabla\psi_-^*\cdot\nabla\psi_+\psi_+^*\psi_-] \\
&= 32(b+2c+2a)\psi_0^4\int d^2\mathbf{r}|\nabla\theta_+|^2 + 16(c'+2a')\psi_0^4\int d^2\mathbf{r}|\nabla\theta_-|^2.
\end{aligned} \quad (15)$$

So, the stiffness parameters ρ and κ in the text take the form as:

$$\rho = 8B\psi_0^2 + 64(b+2c+2a)\psi_0^4, \quad (16)$$
$$\kappa = 8B\psi_0^2 + 32(c'+2a')\psi_0^4. \quad (17)$$

And the the lowest order of the real space Hamiltonian is given by:

$$H_0 = \int d^2\mathbf{r}\left(\frac{\rho}{2}|\nabla\theta_+|^2 + \frac{\kappa}{2}|\nabla\theta_-|^2\right) \quad (18)$$

MORE DETAILED RESULTS ABOUT THE RG STUDY

To compare with the phase diagrams with different initial value of the coupling parameters, we present Fig.(S1) in this section. As shown in this figure, we find the direct transition regime between chiral TSF and normal phase are enhanced with larger initial value of half-vortices couplings $g_{1,1}$. Additionally, chiral metal phase will be enlarged in the phase diagram if we increase the initial value of the coupling parameter g_4.

Our RG results indicate that the interesting phases of charge 4e SC and chiral metal can always exist with different initial coupling parameters.

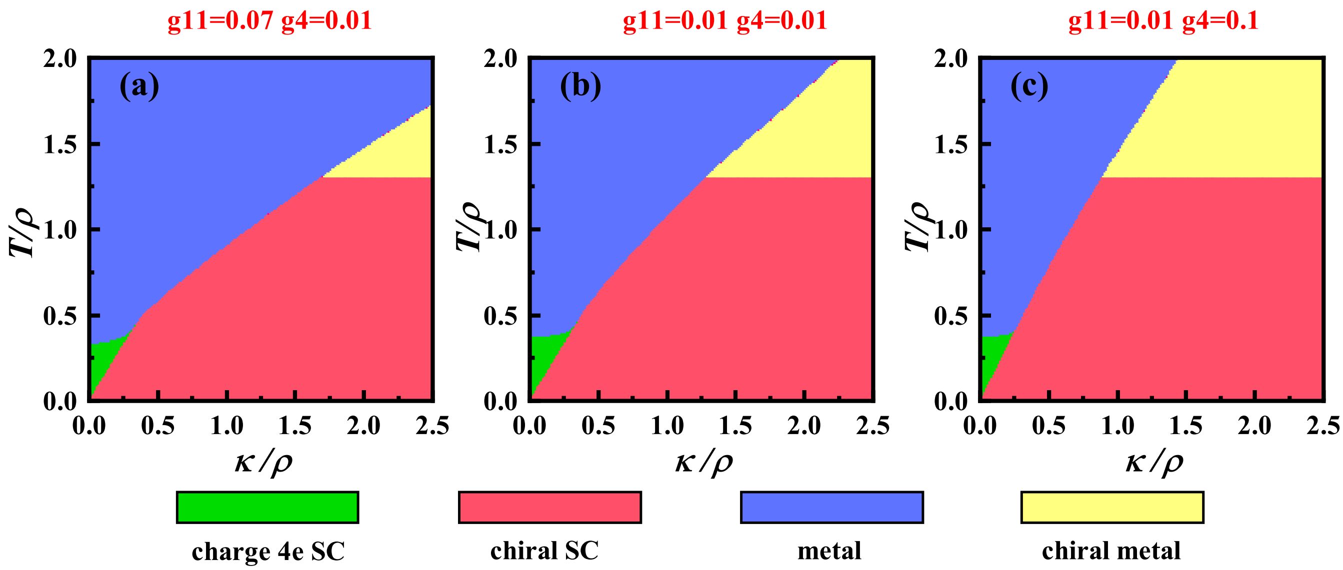

Figure S1. (Color online) Phase diagram provided by the RG approach with different initial coupling parameters. The initial values of the coupling parameters are $g_{2,0} = g_{0,2} = 0.1$, $g_{1,1} = 0.07$ and $g_4 = 0.01$ for (a), $g_{2,0} = g_{0,2} = 0.1$, $g_{1,1} = g_4 = 0.01$ for (b), and $g_{2,0} = g_{0,2} = 0.1$, $g_{1,1} = 0.01$, $g_4 = 0.1$ for (c).

MORE DETAILS RESULTS ABOUT THE MC STUDY

The properties of the correlation function of the parameter point B and C in phase diagram is shown in Fig. S2. For the parameter point B, Fig. S2(a) and (b) show the correlation functions η_+ and η_-, respectively. Both the correlation function η_+ and η_- are exponentially decay, which proves that point parameter B is the metal state. On the contrary, For the parameter point C, Fig. S2(c) and (d) show the correlation functions η_+ and η_-, respectively. The correlation function η_+ is power law decay but the correlation function η_- is a constant, which proves that parameter point D is the chiral SC.

To verify the generality of the discretized Hamiltonian, we perform the MC study with different γ to obtain the phase diagram, shown in Fig. S3 (a-c). The phase diagrams for different γ do not change qualitatively.

To verify the stability of the results, we perform the MC study with a weak first-order Josephson-coupling term added, whose coefficient is $B = 0.01\rho$. The γ is the same as that adopted in the main text. The phase diagram is shown in Fig. S3(d), which is similar with that obtained for zero B.

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[1] Meng Zeng, Lun-Hui Hu, Hong-Ye Hu, Yi-Zhuang You, and Congjun Wu, arXiv: 2102.06158.

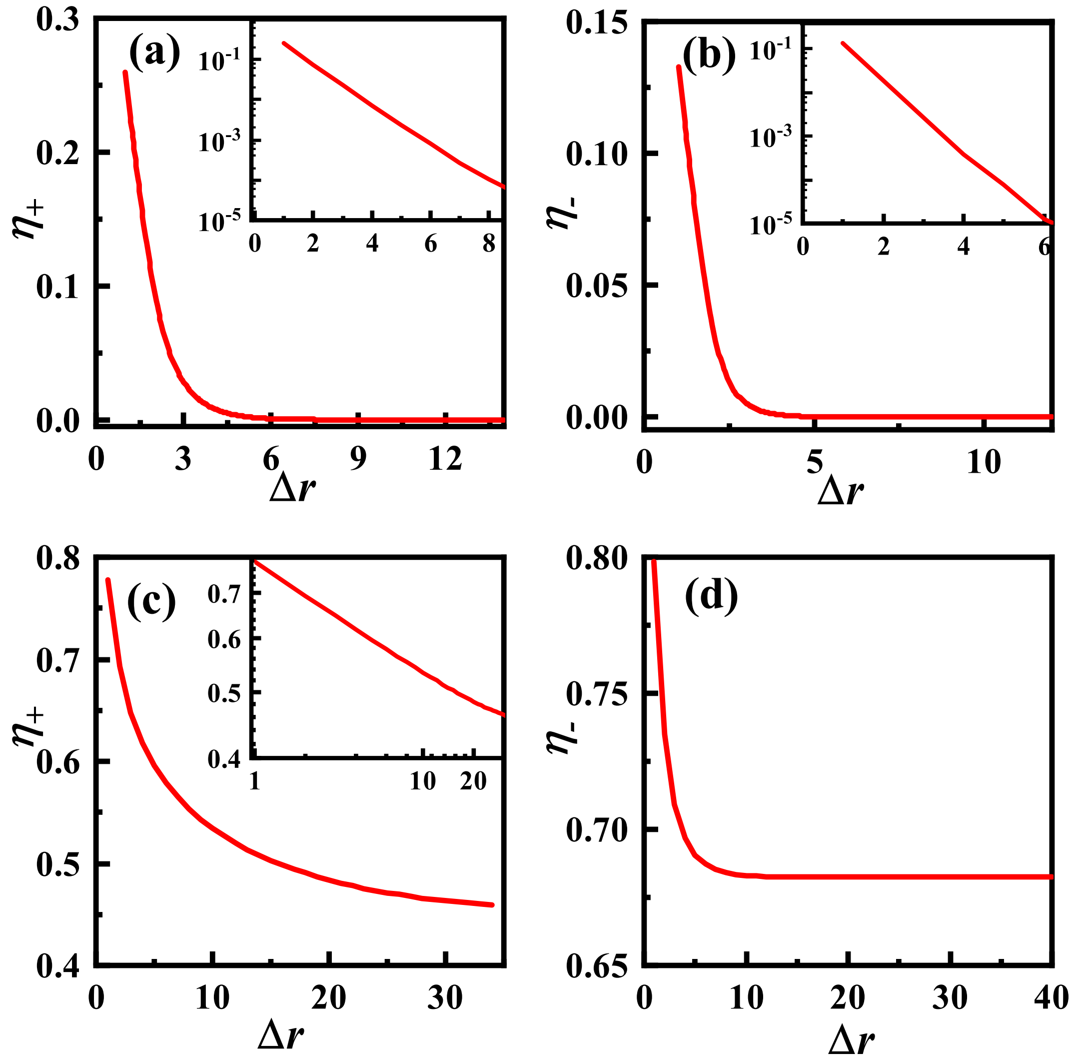

Figure S2. (Color online) (a-b) The correlation function $\eta_\pm$ for the parameter point B($\kappa/\rho = 0.6, T/\rho = 0.45$) in Fig.2(b) in the main text, respectively. The y- axes of the inset are logarithmic axes. (c) The correlation function η_+ for the parameter point C($\kappa/\rho = 1, T/\rho = 0.2$) in Fig.2(b) in the main text, both the x- and y- axes of the inset are logarithmic axes. (d) The correlation function η_- for the parameter point C in Fig.2(b) in the main text, the y- axis of the inset is logarithmic axis.

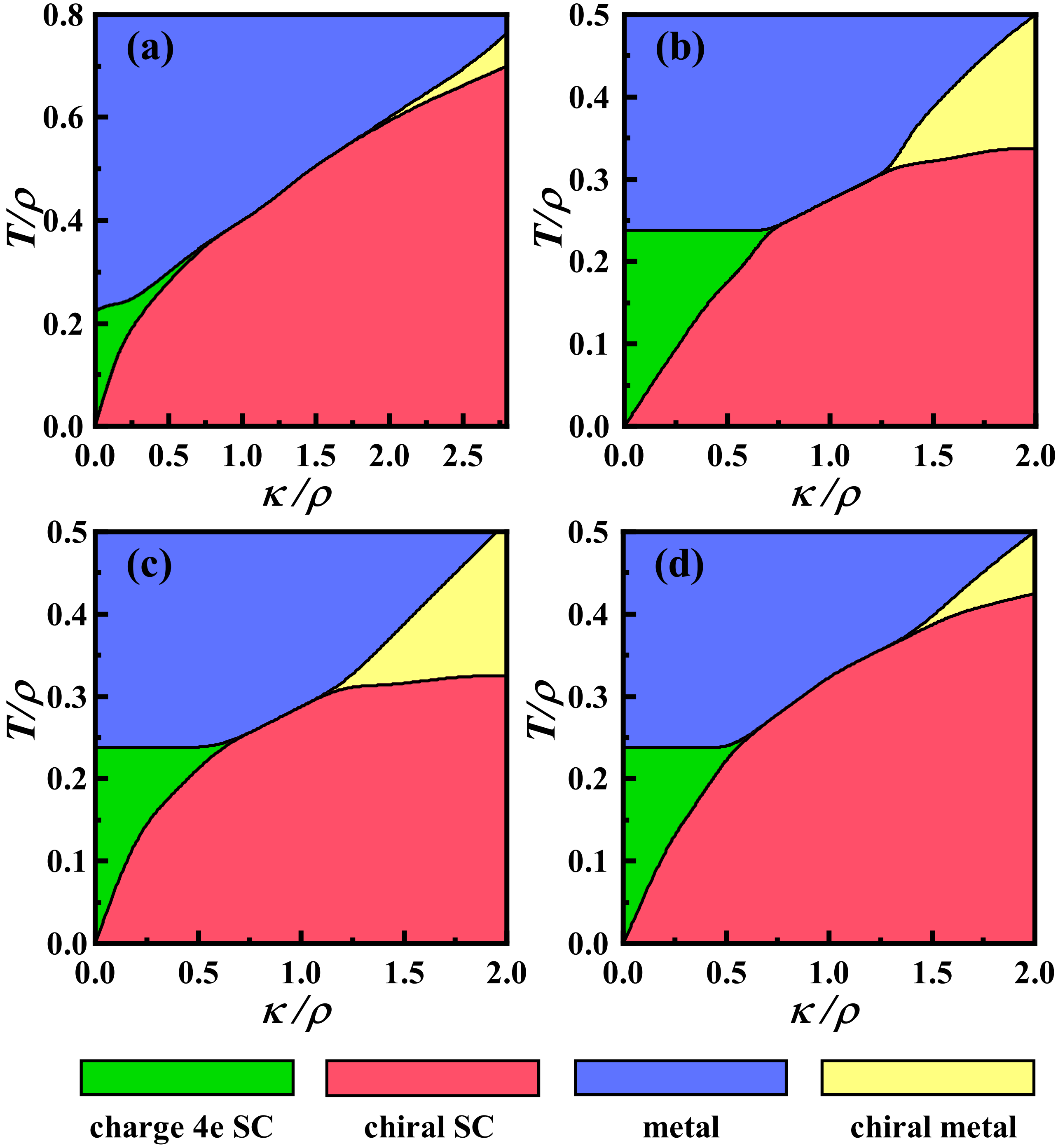

Figure S3. (Color online) Phase diagram provided by the MC study with different parameter γ and extra B term. (a) The same parameters as those in the phase diagram in the main text except that $\gamma = \rho\kappa/2(\rho+\kappa)$. (b) The same parameters as those in the phase diagram in the main text except that $\gamma = \rho\kappa/6(\rho+\kappa)$. (c) The same parameters as those in the phase diagram in the main text except that $\gamma = 0.1\rho$ is a constant. (d)The same parameters as those in the phase diagram in the main text except that a weak first-order Josephson coupling with coefficient $B = 0.01\rho$ is added.