

## Nonunitary multiorbital superconductivity from competing interactions in Dirac materials

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Unconventional superconductors represent one of the most intriguing quantum states of matter. In particular, multiorbital systems have the potential to host exotic *nonunitary* superconducting (NU SC) states. While the microscopic origin of nonunitarity is not yet fully solved, competing interactions are suspected to play a crucial role in stabilizing such states. The interplay between charge order and superconductivity has been a recurring theme in unconventionally superconducting systems, ranging from cuprate-based superconductors to dichalcogenide systems and even to twisted van der Waals materials. Here, we demonstrate that the existence of competing interactions gives rise to a nonunitary superconducting state. We show that the nonunitarity stems from a competing charge-ordered state whose interplay with superconductivity promotes multiorbital order. We establish this mechanism both from a Ginzburg-Landau perspective and from a fully microscopic self-consistent solution of a multiorbital Dirac material. Our results put forward competing interactions as a powerful mechanism for driving nonunitary multiorbital superconductivity.

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Materials with competing interactions represent a paradigmatic playground to engineer novel electronic states of matter [1,2]. Generically, the interplay of different interaction channels, typically attractive electron-phonon coupling and repulsive Coulomb interactions, can give rise to competing states. Such competitions lead to especially rich physics in the presence of many active electronic orbitals, where the existence of additional degrees of freedom substantially enlarges the space of emergent symmetry-broken states [3–5]. Several materials show charge order coexisting with unconventional superconductivity, such as cuprate superconductors [6–8], two-dimensional materials [9,10], and twisted van der Waals heterostructures [11,12]. The latter have recently attracted much attention for their diverse and often tunable interplay between symmetry-broken states [13,14].

Unconventional superconducting states [15] are intensely pursued for their potential topological and exotic properties [16]. Among them, nonunitary superconducting states hold a special place [17–20] because they feature spin and/or orbital polarization in their pairing wave function. In the microscopic description of multiorbital systems, nonunitary superconductivity is characterized by a nontrivial interorbital pairing matrix  $\hat{\Delta}$  with  $\hat{\Delta}^\dagger \hat{\Delta} \propto \mathbb{1}$ . Generic mechanisms that ensure the emergence of these nonunitary states are so far not well understood, yet tuning competing interactions between different orbitals has been shown to be a promising route to

stabilize complex interorbital pairing channels [21–26]. Finding and understanding minimal multiorbital models realizing this mechanism still remains an open problem in correlated quantum matter [5,25,27–32].

In this Letter, we demonstrate that different interactions can cooperate to generate nonunitary multiorbital superconductivity. We first use a Ginzburg-Landau theory to show the cooperative effect between charge order and superconductivity (cf. Fig. 1). Second, we demonstrate this effect using a microscopic model with both repulsive and attractive interactions and show how two distinct orders in the ground state of a Dirac system favor the onset of superconductivity with nonunitary pairing. Interestingly, the emergence of the nonunitary superconducting state from the cooperative charge and superconducting orders can be probed by a gap opening away from the chemical potential, providing a simple experimentally observable signature of nonunitary superconductivity.

We first address how the cooperative effect between a charge density wave (CDW) and superconductivity (SC) can be captured using a symmetry analysis of the Ginzburg-Landau free energy of a multiorbital system. While the following argument does not depend on the existence of a Dirac crossing in the electronic spectra, this situation will be of particular interest when addressing a microscopic model realizing this phenomenology. In the following, we consider a material with a bipartite lattice structure with two sublattices *A* and *B*. The corresponding point group *G* contains symmetry operations  $G' \subset G$  that preserve the two sublattices, as well as symmetry operations  $G \setminus G'$  that exchange them. We assume that below the (bare) critical temperature  $T_c^{\text{CDW}}$ , this bipartite sublattice symmetry is broken by a CDW with the order parameter  $m = n_A - n_B$ , where  $n_A$  ( $n_B$ ) are electronic densities in sublattice *A* (*B*). The Ginzburg-Landau free-energy density

near such a phase transition is

$$f_{\text{CDW}} = \alpha(T)m^2 + \beta m^4, \quad (1)$$

where  $\alpha(T) = \alpha'(T - T_c^{\text{CDW}})$  and  $\beta > 0$  for stability. The order parameter transforms according to the one-dimensional irreducible representation (1D irrep) of  $G$ , which has characters  $+1$  ( $-1$ ) on the conjugacy classes which preserve (exchange) the sublattices.

We consider the typical case of a material close to a  $s$ -wave superconducting phase transition with the (bare) critical temperature  $T_c^{\text{SC}}$ . In the absence of the charge density wave, we would be tempted to use a single complex-valued order parameter  $\Delta$  to describe the phase transition with the Ginzburg-Landau free-energy density  $f_{\text{SC}} = a(T)|\Delta|^2 + b|\Delta|^4$ , where  $a(T) = a'(T - T_c^{\text{SC}})$  and  $b > 0$ , and  $\Delta$  transforms according to a 1D irrep of  $G$ . However, the presence of a nearby CDW phase necessitates two SC order parameters  $\Delta_{A,B} = |\Delta_{A,B}|e^{i\varphi_{A,B}}$ , one per sublattice. The modified free-energy density is [33]

$$f_{\text{SC}} = a_1(T)(|\Delta_A|^2 + |\Delta_B|^2) + a_2|\Delta_A - \Delta_B|^2, \quad (2)$$

where the second term will minimize the phase difference between  $\Delta_A$  and  $\Delta_B$ , allowing us to choose  $\varphi_A = \varphi_B = 0$ .

Crucially, we take into account that the SC and CDW order parameters are generally not independent here, but instead couple through

$$f_{\text{CPL}} = \lambda m(|\Delta_A|^2 - |\Delta_B|^2). \quad (3)$$

The coupling  $f_{\text{CPL}}$  can naturally lead to  $|\Delta_A| \neq |\Delta_B|$ , which, as we will see later, is a minimal sufficient condition to have a nonunitary pairing matrix [cf. Eq. (8)]. We thus introduce the unitary SC order parameter  $\Delta = (\Delta_A + \Delta_B)/2$  and the nonunitary (NU) SC order parameter  $\delta = (\Delta_A - \Delta_B)/2$ , and state our system's total free-energy density  $f = f_{\text{SC}} + f_{\text{CDW}} + f_{\text{CPL}}$  to leading order in  $m$ ,  $\Delta$ , and  $\delta$ , i.e.,

$$f = 2a_1\Delta^2 + (\delta \quad m) \begin{pmatrix} \frac{a_1}{\lambda\Delta} + a_2 & \lambda\Delta \\ \lambda\Delta & \alpha \end{pmatrix} \begin{pmatrix} \delta \\ m \end{pmatrix}. \quad (4)$$

In what follows, we discuss the stable phases of  $f$  and the phase transition from the unitary SC phase ( $\Delta \neq 0$ ,  $m = \delta = 0$ ) to a nonunitary phase with both SC and CDW order ( $\Delta, m, \delta \neq 0$ ), using the linearized Ginzburg-Landau equations [34], i.e., we find the two eigenvalues of the matrix in Eq. (4).

Generally, as shown in Fig. 1, we find that the coupling  $f_{\text{CPL}}$  between NU SC ( $\delta$ ) and the CDW ( $m$ ) tends to be cooperative, i.e., SC and CDW favor each other. In Fig. 1(a), we compare the free-energy density for the SC phase in the absence of the CDW ( $\Delta \neq 0$ ,  $\delta = m = 0$ ) and the NU SC phase ( $\Delta, \delta, m \neq 0$ ). We find that in the latter case, the local minimum is lower and shifted towards larger magnitudes, indicating that the coupling to the CDW amplifies and stabilizes the SC order parameter  $\Delta$ . In Fig. 1(b), we show a phase diagram for temperature  $T$  versus critical temperature  $T_c^{\text{CDW}}$  of the CDW. For  $T_c^{\text{SC}} > T_c^{\text{CDW}}$ , the system is in the normal metal state above  $T_c^{\text{SC}}$ . By lowering the temperature below  $T_c^{\text{SC}}$ , the system transitions into a unitary SC phase ( $\Delta \neq 0$ , but  $\delta = m = 0$ ). A further reduction below  $T_c$  leads to a second phase transition into NU SC order ( $\Delta, \delta, m \neq 0$ ).

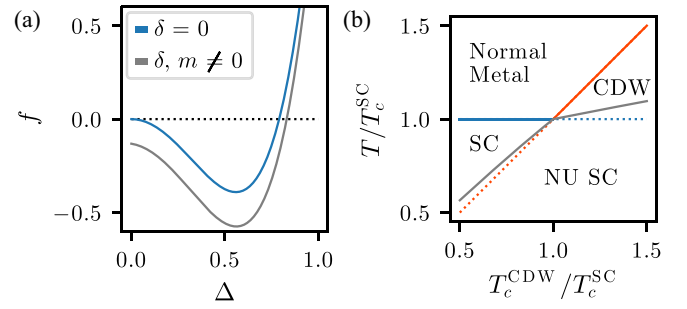


FIG. 1. Ginzburg-Landau theory for coupled order parameters  $\Delta$  of unitary superconductivity (SC),  $\delta$  of nonunitary superconductivity (NU SC), and  $m$  of a charge density wave (CDW) in a bipartite system. (a) Free-energy density  $f$  as a function of the SC order parameter  $\Delta$ , illustrating that the presence of a charge order ( $\delta, m \neq 0$ ) lowers the condensation energy. (b) Phase diagram featuring regions where NU SC and a CDW coexist. Shown are phase transition lines between a normal metal and a SC (CDW) state in the absence of a CDW (SC) state in blue (red), as well as the transition line (gray) into the nonunitary phase where SC and the CDW coexist. We also show the case when the order parameters are uncoupled (dotted lines) [cf.  $\lambda = 0$  in Eq. (3)].

We observe that the coupling between NU SC  $\delta$  and the CDW  $m$  increases the transition temperature from  $T_c^{\text{CDW}}$  to  $T_c > T_c^{\text{CDW}}$ , which again signals that the coupling between order parameters mutually stabilizes SC and CDW phases. Similarly, for  $T_c^{\text{CDW}} > T_c^{\text{SC}}$ , the system transitions from the normal metal phase first to a pure CDW phase and then to the NU SC phase.

After having observed the cooperative mechanism between CDW and SC from a phenomenological point of view, we move on to demonstrate how this behavior is realized in a microscopic model. As a minimal example featuring the mechanism of coupled order parameters, we consider a honeycomb lattice as shown in Fig. 2(a). Each unit cell contains two atoms with sublattice labels  $\alpha \in \{A, B\}$ , which we assume to be of the same type. The point group is then  $G = D_{6h}$  and, when sublattice symmetry is broken, the symmetry elements reduce to  $G' = D_{3h}$ . In reciprocal space, the first Brillouin zone is also hexagonal and has high-symmetry points  $\Gamma$ ,  $K$ ,  $K'$ , and  $M$ ; see Fig. 2(b).

We can describe the electronic properties in the tight-binding approximation with the Hamiltonian

$$H_0 = t \sum_{\langle iA, jB \rangle, s} c_{iAs}^\dagger c_{jBs} + \text{H.c.} - \mu \sum_{i, \alpha, s} n_{ias}, \quad (5)$$

where  $t$  is the hopping amplitude between nearest-neighbor orbitals, and  $c_{ias}$  ( $c_{ias}^\dagger$ ) destroys (creates) an electron on sublattice  $\alpha$  with spin  $s \in \{\uparrow, \downarrow\}$  in unit cell  $i$ . The second term accounts for the chemical potential  $\mu$  ( $\mu = 0$  at half filling  $\nu = 0.5$ ), where  $n_{ias} = c_{ias}^\dagger c_{ias}$  is the local electron density operator. In Fig. 2(c), we see the electronic band structure of Hamiltonian (5) along a high-symmetry path. It shows a characteristic Dirac crossing near the  $K$  and  $K'$  points. We assume that the chemical potential is slightly below half filling ( $\nu = 0.5$ ) such that the lower electronic band is partially empty (here,  $\nu = 0.49$ ) with circular Fermi surfaces around the  $K$  and  $K'$  points.

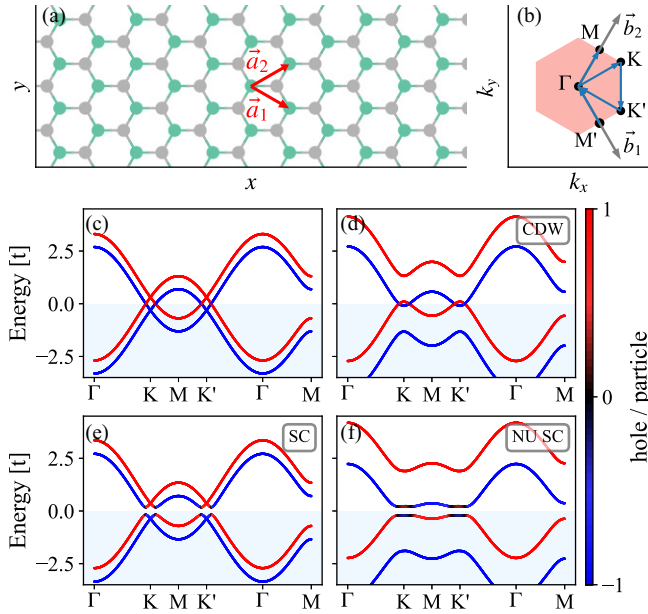


FIG. 2. Honeycomb lattice and electronic band structures with and without mean-field interactions. (a) Real-space structure with lattice vectors  $\mathbf{a}_1$ ,  $\mathbf{a}_2$  and sublattices  $\alpha = A, B$  (green, gray) and (b) reciprocal-space first Brillouin zone with high-symmetry points  $\Gamma$ ,  $M$ ,  $K$ , and  $K'$  and high-symmetry path. (c)–(f) Energy bands along this high-symmetry path: (c) in the absence of interactions, (d) in a regime dominated by nearest-neighbor repulsion  $V$  (CDW), (e) in a regime dominated by on-site attraction  $U$  (SC), and (f) in an intermediate regime with both sizable interactions  $U$  and  $V$  (NU SC) (cf. stars highlighted in Fig. 3). The band color distinguishes particle-hole branches (red and blue) that are required to treat particle-particle pairing.

We now include effective electron-electron interactions in the model to promote the conventional local  $s$ -wave channel and a CDW. We consider short-range interactions of strength  $U < 0$  (attractive) between electrons on the same site and strength  $V > 0$  (repulsive) between nearest neighbors, i.e.,

$$H_{\text{int}} = U \sum_{i\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + V \sum_{\langle iA, jB \rangle, ss'} n_{iAs} n_{jBs'}. \quad (6)$$

Note that in real materials, our choice of  $U < 0$  and  $V > 0$  may result from strong renormalization due to screening and exchange effects from additional atomic orbitals and/or electron-phonon coupling. Another possible regime that we will not explore here would be  $U > 0$  and  $V < 0$ , which would require a careful analysis of the cooperation or competition between the spin-density wave, spin-singlet superconductivity, and spin-triplet superconductivity physics [23,24].

In the mean-field approximation, we find that the Hamiltonian in Eq. (5) obtains Hartree-Fock corrections  $t \rightarrow \bar{t}_{ss'}(G)$  and  $\mu \rightarrow \bar{\mu}_{\alpha s}(G)$  through mean fields  $G_{i\alpha, j\beta}^{ss'} = \langle c_{i\alpha s}^\dagger c_{j\beta s'} \rangle$  and  $F_{i\alpha, j\beta}^{ss'} = \langle c_{i\alpha s}^\dagger c_{j\beta s'}^\dagger \rangle$ , where  $\langle \cdot \rangle$  is the ground-state expectation value of the corresponding mean-field Hamiltonian  $\bar{H}_0$ ; see the Supplemental Material [35] for details. The approximation also introduces particle-particle pairing terms, of which we

include the on-site spin-singlet contribution,

$$\bar{H}_P = \frac{U}{2} \sum_{i, \alpha, s, s'} \Delta_{i\alpha, i\alpha}^{0*} c_{i\alpha s}^\dagger (-i\sigma_y)_{ss'} c_{i\alpha s'}^\dagger + \text{H.c.}, \quad (7)$$

where  $\Delta_{i\alpha, i\alpha}^0(F)$  is the local singlet pairing amplitude. The corresponding local pairing matrix,

$$\hat{\Delta}_i^0 = \frac{\Delta_A^0 + \Delta_B^0}{2} (-i\tau_0\sigma_y) + \frac{\Delta_A^0 - \Delta_B^0}{2} (-i\tau_z\sigma_y), \quad (8)$$

is unitary only if  $|\Delta_A^0| = |\Delta_B^0|$ , in which case  $\hat{\Delta}_i^0 (\hat{\Delta}_i^0)^\dagger \sim \tau_0\sigma_0$ . We note that here NU SC corresponds to a sublattice-polarized pairing wave function. We used Pauli matrices  $\tau_i$  ( $\sigma_i$ ) for the sublattice (spin) basis.

For our microscopic model, we now solve the self-consistency relation for the mean fields numerically (see Supplemental Material [35]). We assume that the ground state preserves the original translational symmetries, but can break other space-group symmetries. We consider the mean-field electronic bands of  $\bar{H}_0 + \bar{H}_P$  (cf. Fig. 2), as well as the order parameters (cf. Fig. 3).

The nearest-neighbor repulsion ( $V > 0$ ) alone favors the formation of a CDW [36–39]. It reduces the symmetry group ( $D_{6h} \rightarrow D_{3h}$ ) by breaking the sublattice symmetry, and is characterized by the same order parameter  $m = n_A - n_B$  that we introduced earlier. As a consequence, the Dirac cones in the electronic spectrum acquire a mass term, leading to a band gap as shown in Fig. 2(d). If we instead consider the case without nearest-neighbor repulsion, the attractive on-site interaction alone ( $U < 0$ ) allows pairs with  $s$ -wave orbital symmetry and singlet-spin configuration to form. This is described by the pairing order parameter  $\Delta \equiv \sum_\alpha |\Delta_\alpha^0|$  and leads to a (topologically trivial) hybridization gap between the electron and hole branches in the spectrum; see Fig. 2(e). The hybridization gap is small even for sizable interaction strengths, which is a result of the vanishing density of states near the Dirac point in the electronic spectrum.

When both nearest-neighbor repulsion ( $V > 0$ ) and on-site attraction ( $U < 0$ ) are sizable, we find a third phase with both a CDW and  $s$ -wave singlet SC with the mean-field electronic band structure shown in Fig. 2(f). This phase is characterized by the order parameter  $\delta \equiv ||\Delta_A^0| - |\Delta_B^0||$ , which describes a sublattice imbalance in the  $s$ -wave pairing amplitude. We see that the enhanced density of states near the band edges favors the formation of pairs. In what follows, we will investigate the phase diagram and the mechanism with which the CDW induces sublattice symmetry-broken pairing correlations (NU SC).

In Fig. 3(a), we see the mean-field phase diagram showing the three symmetry-breaking phases we just introduced: the CDW when  $V > 0$  dominates (red), the unitary  $s$ -wave singlet SC when  $U < 0$  dominates (blue), and a mixed phase with NU SC due to broken sublattice symmetry both in the charge and the pairing sectors (gray) when both interactions are sizable. We find that the coexistence phase occupies the majority of phase space and always appears together with finite order parameters for the other phases. Furthermore, stronger on-site interaction  $U < 0$  lowers the critical nearest-neighbor interaction  $V_c$  at which a CDW occurs (red transition line).

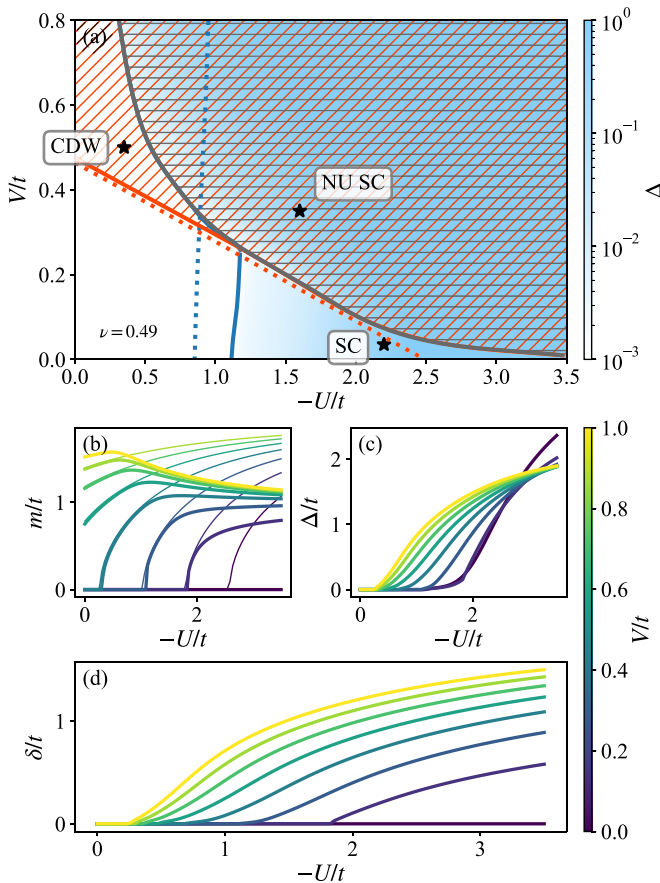


FIG. 3. Mean-field analysis of the interplay between charge density wave and spin-singlet  $s$ -wave SC in the honeycomb lattice for on-site attraction ( $U < 0$ ) and nearest-neighbor repulsion ( $V > 0$ ). (a) Phase diagram showing the SC order parameter  $\Delta_0$  (blue), regions with CDW order (red, striped), and regions with NU SC (gray, striped). The transition lines (solid lines) indicate where the respective order parameter exceeds  $10^{-3}t$ . Transition lines when CDW order is artificially suppressed (blue, dotted) and when SC is suppressed (red, dotted) are also shown. (b)–(d) Order parameters for varying on-site attraction  $U$  at discrete values of nearest-neighbor repulsion (indicated by line colors). (b) The CDW order parameter  $m$ , (c) the average  $\Delta$  of the  $s$ -wave parameters in sublattices  $A$  and  $B$ , and (d) the order parameter  $\delta$  for the pairing imbalance between the sublattices. The thin lines in (b) show the order parameter when SC is artificially suppressed. The numerical calculations use  $10^4$  points in reciprocal space, at filling  $\nu = 0.49$  and temperature  $k_B T = 10^{-3}t$ .

Increasing the nearest-neighbor interaction  $V < V_c$  (below the onset of a CDW) disfavors the SC phase (blue transition line). In Figs. 3(b)–3(d), we find that (within our numerical resolution) the onset of the CDW and NU SC orders emerge as second-order phase transitions.

To investigate the nature of the coexistence phase, we repeat the self-consistent mean-field iteration without allowing for the CDW. We achieve this by enforcing  $n_A = n_B$  in each step. In Fig. 3(a), we see that this constraint leads to a significantly modified phase diagram: while unitary  $s$ -wave pairing still occurs (blue dashed transition line), the sublattice-asymmetric nonunitary pairing no longer does, i.e.,  $\delta = 0$ . This means that the NU SC relies on the presence of the charge density wave. We also see that suppressing the CDW results in a shift in the (blue) transition line of the unitary SC order parameter  $\Delta$  shown in Fig. 3(a). For small  $V$ , the constraint shifts the transition towards a smaller critical threshold  $U_c$ , which we attribute to a renormalized Fermi velocity (e.g., reduced density of states). At larger  $V$ , the threshold  $U_c$  shifts towards larger values, which is in accordance with the observations made in the effective Ginzburg-Landau free-energy densities, where we studied the coupling between the order parameters  $m$  and  $\delta$  [cf. Eq. (3)]: The CDW significantly enhances SC, allowing sizable pairings at much smaller on-site interactions  $U < 0$  (e.g., at  $U_c \sim -0.6t$  for  $V \sim 0.6t$ ).

To summarize, we have shown that the existence of competing interactions in Dirac systems naturally leads to nonunitary multiorbital superconducting states. In particular, we have shown that the interplay between superconductivity and charge order fluctuations leads to a cooperative effect in which the appearance of nonunitary superconducting order dramatically enhances the superconducting gap. Our results put forward competing interactions as a compelling mechanism giving rise to nonunitary superconductivity, and establish Dirac materials as a paradigmatic system to realize unconventional multiorbital superconductivity.

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