Chiral superconductivity from repulsive interactions in doped graphene

Rahul Nandkishore¹, L. S. Levitov¹ and A. V. Chubukov²*

Chiral superconductivity, which breaks time-reversal symmetry, can exhibit a wealth of fascinating properties that are highly sought after for nanoscience applications. We identify doped graphene monolayer as a system where chiral superconductivity can be realized. In this material, a unique situation arises at a doping where the Fermi surface is nested and the density of states is singular. In this regime, d-wave superconductivity can emerge from repulsive electron-electron interactions. Using a renormalization group method, we argue that superconductivity dominates over all competing orders for generic weak repulsive interactions. Superconductivity develops simultaneously in two degenerate d-wave pairing channels. We argue that the resulting superconducting state is of chiral type, with the phase of the superconducting order parameter winding by π around the Fermi surface. Realization of this state in doped graphene will prove that superconductivity can emerge from electron-electron repulsion, and will open the door to applications of chiral superconductivity.

The search for solid-state realization of chiral superconductivity has a long history. Spin-triplet p-wave chiral superconductivity (ρ, ρp state) has probably been found in Sr₂RuO₄ (ref. 25), which represents an analogue of superfluid ³He (ref. 9), but the spin-singlet d + id state has not yet been observed experimentally. Such a state was once proposed as a candidate state for high-Tc cuprate superconductors⁹, but later gave way to a more conventional TRS-preserving d-wave state. The key difficulty in realizing a d + id state is that the interactions that favour a d-wave state usually have strong momentum dependence and hence distinguish between dₓ²−ᵧ² and dₓᵧ pairing. However, in graphene the dₓ²−ᵧ² and dₓᵧ pairing channels are degenerate by symmetry⁸, opening the door to formation of a d + id superconducting state.

How can superconductivity be induced in graphene? Existing proposals for superconductivity in undoped graphene rely on the conventional phonon-mediated BCS mechanism⁷, which leads to an s-wave superconductivity with low Tc values for realistic carrier densities owing to the vanishing density of states of relativistic particles. However, there is an alternative route to superconductivity, wherein repulsive microscopic interactions give rise to attraction in a d-wave channel. This alternative route becomes viable when graphene is doped to the M point of the Brillouin zone corresponding to 3/8 or 5/8 filling of the π band (pristine graphene corresponds to 1/2 filling). At this filling factor, a logarithmic Van Hove singularity originates from three inequivalent saddle points, and the Fermi surface also exhibits a high degree of nesting, forming a perfect hexagon when third-neighbour (and higher) hopping effects are neglected. The combination of a singular DOS and a near-nested Fermi surface strongly enhances the effect of interactions, allowing non-trivial phases to emerge at relatively high temperatures, even if interactions are weak compared with the fermionic bandwidth W. Relevant doping levels were recently achieved experimentally using calcium and potassium dopants. Also, a new technique that employs ionic liquids as gate dielectrics allows high levels of doping to be reached without introducing chemical disorder.

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Competing orders

In systems with near-nested Fermi surface, superconductivity has to compete with charge-density-wave (CDW) and spin-density-wave (SDW) instabilities. At the first glance, it may seem that a system with repulsive interactions should develop a density-wave order rather than become a superconductor. However, to analyse this properly, one needs to know the susceptibilities to the various orders at a relatively small energy, $E_0$, at which the order actually develops. The couplings at $E_0$ generally differ from their bare values because of renormalizations by fermions with energies between $E_0$ and $W$. At weak coupling, these renormalizations are well captured by the renormalization group technique.

Interacting fermions with a nested Fermi surface and logarithmically divergent DOS have previously been studied on the square lattice using renormalization group methods, where SDW fluctuations were argued to stimulate superconductivity. The analysis also revealed near degeneracy between superconductivity and SDW orders. The competition between these orders is decided by a subtle interplay between deviations from perfect nesting, which favour superconductivity, and subleading terms in the renormalization flow, which favour SDW. In contrast, the renormalization group procedure on the honeycomb lattice unambiguously selects superconductivity at leading order, allowing us to safely neglect subleading terms. The difference arises because the honeycomb lattice contains three saddle points, whereas the square lattice has only two, and the extra saddle point tips the balance seen on the square lattice between magnetism and superconductivity decisively in favour of superconductivity. A similar tipping of a balance between magnetism and superconductivity decisively in favour of superconductivity. A similar tipping of a balance between superconductivity and SDW in favour of superconductivity has been found in theoretical studies of Fe-based superconductors.

In previous works on graphene at the M point, various instabilities were argued to stimulate superconductivity. The analysis of interactions, driving the system into a chiral superconducting state, was performed for the square lattice using renormalization group methods. The extra saddle point tips the balance seen on the square lattice, which contains three saddle points, whereas the square lattice has only two, allowing us to safely neglect subleading terms in the renormalization group procedure on the honeycomb lattice unambiguously selecting superconductivity at leading order.

The model

We follow the procedure developed for the square lattice and construct a patch renormalization group that considers only fermions near three saddle points, which dominate the DOS. There are four distinct interactions in the low-energy theory, involving two-particle scattering between different patches, as shown in Fig. 2.

The system is described by the low-energy theory

$$
\mathcal{L} = \sum_{\alpha=1}^3 \psi^\dagger_{\alpha}(\partial_t - \epsilon_\alpha + \mu)\psi_{\alpha} - \frac{1}{2}g_{1}\psi^\dagger_{\alpha}\psi^\dagger_{\beta}\psi_{\beta}\psi_{\alpha} - \sum_{\alpha\neq\beta} \frac{1}{2}[g_{2}\psi^\dagger_{\alpha}\psi^\dagger_{\beta}\psi_{\alpha}\psi_{\beta} + g_{3}\psi^\dagger_{\alpha}\psi^\dagger_{\beta}\psi_{\alpha}\psi_{\beta} + g_{4}\psi^\dagger_{\alpha}\psi^\dagger_{\beta}\psi_{\alpha}\psi_{\beta}] 
$$

(1)

where summation is over patch labels $\alpha, \beta = M_1, M_2, M_3$. A spin sum is implicit in the above expression, with the spin structure for each of the four terms being $\gamma, \delta, \delta, \gamma$, where $\gamma$ and $\delta$ label the spin up and down states. Here $\epsilon_\alpha$ is the tight binding dispersion, expanded and mean-field theory. Ref. 4 considered the instability to d-wave superconductivity, ref. 5 considered a charge 'Pomeranchuk' instability to a metallic phase breaking lattice rotation symmetry, and refs 6–8 considered a SDW instability to an insulating phase. Within the framework of mean-field theory, used in the above works, all of these phases are legitimate potential instabilities of the system. However, clearly graphene at the M point cannot be simultaneously superconducting, metallic and insulating. The renormalization group analysis treats all competing orders on an equal footing, and predicts that the dominant weak coupling instability is to superconductivity, for any choice of repulsive interactions, even for perfect nesting. Further, the Ginzburg–Landau theory constructed near the renormalization group fixed point favours the $d + id$ state.
and \( \Pi_{\text{pp}}(0) = v_0 \ln(A/\max(T, \mu)) \) when \( A \) is our ultraviolet cutoff (Fig. 1) and \( T \) is the temperature. The density of states at a saddle point is \( v_0 \ln(A/\max(T, \mu)) \) per spin projection. The second log factor in \( \Pi_{\text{pp}}(0) \) arises in a conventional way owing to the divergence in the Cooper channel. The second log factor in \( \Pi_{\text{pp}}(Q_{\alpha \beta}) \) arises from nesting of the Fermi surface, and is cut in the infrared by any term that spoils the nesting, such as third-neighbour hopping \( t_3 \) or doping \( \mu \) (ref. 4). At weak coupling the Cooper log is large, and we assume that \( \max(t_3, \mu) \ll A \) so that the nesting log is also large. In this limit, \( \Pi_{\text{pp}}(0) \) and \( \Pi_{\text{pp}}(Q) \) are parametrically larger than \( \Pi_{\text{ph}}(0) \) and \( \Pi_{\text{pp}}(Q) \), so that the renormalization group is dominated by the double log divergent susceptibilities.

**Renormalization group equations**

The renormalization group equations are obtained by extending the approach developed for the square lattice problem to a number of patches \( n > 2 \). The number of patches matters only in diagrams with zero net momentum in fermion loops, as it is only there that we get summation over fermion flavours inside the loop. The only zero-momentum loop with a log divergence is in the Cooper channel. Moreover, only the \( g_3 \) interaction changes the patch label of a Cooper pair, therefore, the number of patches affects only diagrams where two \( g_3 \) interactions are combined in the Cooper channel. With logarithmic accuracy, using \( y = \Pi_{\text{pp}}(k = 0, E) = (v_0/4) \ln^2(A/E) \) as the renormalization group time, we obtain the \( \beta \)-functions

\[
\frac{\text{d}g_1}{\text{d}y} = 2d_1g_1(g_2 - g_1), \quad \frac{\text{d}g_2}{\text{d}y} = d_1(g_2^2 + g_3^2),
\]

\[
\frac{\text{d}g_3}{\text{d}y} = -(n-2)g_2^2 - 2g_3g_4 + 2d_1g_4(2g_2 - g_1),
\]

(2)

Here \( d_1(y) = d\Pi_{\text{pp}}(Q)/Q \approx \Pi_{\text{pp}}(Q)/\Pi_{\text{pp}}(0) \) is the ‘nesting parameter’\(^{31,34} \). This quantity equals one in the perfectly nested limit. For non-perfect nesting, \( d_1(y) \) has the asymptotic forms \( d_1(y = 0) = 1, \quad d_1(y \gg 1) = \ln(A/t_3)/\sqrt{y} \) and interpolates smoothly in between. Because the renormalization group equations flow to strong coupling at a finite scale \( y_c \), we treat \( 0 < d_1(y_c) < 1 \) as a parameter in our analysis.

The \( \beta \)-functions, equation (2), reproduce the two-patch renormalization group from ref. 31 when we take \( n = 2 \) and neglect subleading \( O(\log) \) divergent terms \( (d_2, y) \) from ref. 31, and also reproduce for \( n = 2 \) the renormalization group equations for the Fe-pnictides\(^ {35} \). Graphene near the Van Hove singularity, however, is described by \( n = 3 \).

We note from inspection of equation (2) that \( g_1, g_2, \) and \( g_3 \) must stay positive (repulsive) if they start out positive. This follows because the \( \beta \)-function for \( g_1 \) is positive definite, and the \( \beta \)-functions for \( g_2 \) and \( g_3 \) vanish as the respective couplings go to zero. However, \( g_3 \) decreases under the renormalization group, eventually changing sign and becoming negative. As we will see, \( g_3 = g_2 \) becomes large and positive under the renormalization group, driving an instability to a superconducting phase. However, the positive \( g_2 \) coupling penalizes \( s \)-wave superconductivity, so pairing occurs in a higher angular momentum \( (d \text{-wave}) \) channel.

We integrate our renormalization group equations with \( n = 3 \), starting from \( g_3 = g_2 = 0.1 \) (these values are chosen for illustration) and modelling \( d_1 \) as \( d_1(y) = 1/\sqrt{1+y} \). The results are plotted in Fig. 3. Similar results are obtained if we just treat \( d_1 \) as a constant. The couplings diverge at a scale \( y_c \sim 1/g_0 \), corresponding to a critical temperature and ordering energy scale

\[
T_c, E_0 \sim A\exp(-A/\sqrt{g_0v_0})
\]

(3)
repulsive bare couplings leads to the same limiting trajectory (see equations, which may be solved for the coefficients \( \gamma_i \)). Substituting into equation (2), we obtain a set of polynomial equations for the coefficients \( \gamma_i \).

Figure 3 | Flow of couplings with renormalization group scale \( y \), starting from repulsive interactions. Note that the coupling \( g_4 \) changes sign and becomes attractive, leading to a (superconducting) instability at the energy scale \( y_c \) (equation (3)). Inset: Critical couplings \( G_i \) (equation (4)) near \( y_c \) as a function of the nesting parameter at the ordering energy scale, \( d_1(y_c) \).

Here \( A \) is a non-universal number that depends on how we model \( d_1(y) \). For \( d_1 = 1 \) (perfect nesting, corresponding to zero third-neighbour hopping \( t_3 \)), we obtain \( A = 1.5 \). An RPA-type estimate of \( g_0 \) is outlined in the Supplementary Information. Although \( T_c \) and \( E_0 \) are exponentially sensitive to \( g_0 \), introducing a considerable uncertainty to our estimate, a strong enhancement of characteristic energy scales relative to the BCS result is evident from equation (3).

A similar \( \sqrt{T_c} \) dependence arises in the treatment of colour superconductivity in the analysis of the pairing near quantum-critical points in 3D, in the presence of a \( T_c \) that is strongly enhanced compared with the standard BCS result, \( T_c \sim \exp(-A/g_0/\hbar) \). It should be noted that the enhancement of \( T_c \) in equation (3) arises from weak-coupling physics. It is distinct from the high-\( T_c \) superconductivity that could arise if the microscopic interactions were strong.

Returning to our renormalization group analysis, we note that near the instability threshold, \( g_1, g_2, g_3 \to \infty \) and \( g_4 \to -\infty \), with \( -g_4 > g_3 > g_1 > g_4 \). This observation may be made precise by noting that close to \( y_c \), the interactions scale as

\[
g_i(y) \approx \frac{G_i}{y_c - y}
\]

one test vertex for each patch. The renormalization of the test vertices is governed by the equation

\[
\frac{\partial}{\partial y} \begin{bmatrix} \Delta_1 \\ \Delta_2 \\ \Delta_3 \end{bmatrix} = -2 \begin{bmatrix} g_4 & g_5 & g_3 \\ g_3 & g_4 & g_3 \\ g_3 & g_3 & g_4 \end{bmatrix} \begin{bmatrix} \Delta_1 \\ \Delta_2 \\ \Delta_3 \end{bmatrix}
\]

which can be diagonalized by transforming to the eigenvector basis

\[
\Delta_x = \frac{\Delta}{\sqrt{2}} (0, 1, -1), \quad \Delta_y = \sqrt{3} \Delta \left( 1, -\frac{1}{2}, -\frac{1}{2} \right)
\]

Here \( \Delta_x \) is an \( s \)-wave order, whereas \( \Delta_y \) and \( \Delta_z \) correspond to order parameters that vary around the Fermi surface as \( \Delta \cos(2\phi) \) and \( \Delta \sin(2\phi) \), where \( \phi \) is the angle to the \( x \) axis (Fig. 4). Such a dependence describes \( d \)-wave superconducting orders (SCd), as the gap changes sign four times along the Fermi surface. In 2D notation, the two order parameters \( \Delta_x \) and \( \Delta_y \) correspond to \( d_{xy} \) and \( d_{x^2-y^2} \) superconducting orders respectively.

Notably, we find that the \( s \)-wave vertex \( \Delta_x \), equation (7), has a negative eigenvalue and is suppressed under renormalization group flow (equation (5)). This is to be expected, given that we started out with repulsive microscopic interactions. At the same time, the \( d \)-wave orders \( \Delta_y \) and \( \Delta_z \) have the (identical) eigenvalue \( g_3 - g_1 \), which may be negative at the bare level but becomes positive under the renormalization group, indicating an instability in the \( d \)-wave channel. We solve equation (5) for the \( d \)-wave orders, by...
substituting the scaling form of the interactions (equation (4)), and find that the \( d \)-wave susceptibility diverges near \( \gamma \), as

\[
\chi_{SCD}(y) = \frac{\Delta_{G,F}(y)}{\Delta_{G,F}(0)} \sim (y_c - y)^{2G_i - G_d} \tag{8}
\]

where we recall, \( G_i - G_d > 0 \).

The divergence of the SCd susceptibility indicates an instability to \( d \)-wave superconductivity under the renormalization group, with the \( \Delta \cos(2\varphi) \) and \( \Delta \sin(2\varphi) \) order parameters having identical susceptibility. The degeneracy of the two \( d \)-wave orders is guaranteed, because the \( d_{x^2-y^2} \) and \( d_{xy} \) functions belong to the same 2D irreducible representation of the lattice point group\(^{3,26}\). However, this does not guarantee that \( d \)-wave superconductivity will develop, because the SCd instability must compete against the tendency for density-wave formation.

To investigate density-wave formation, we introduce test vertices representing pairing of particles with holes on a different patch. The particles and holes may pair in the charge channel, forming CDW, or in the spin channel, forming SDW. We compute the renormalization of the pairing vertices under the renormalization group, and find that the CDW vertex is suppressed by interactions, but the SDW vertex \( \Delta_{SDW} \) is enhanced, similar to ref. 31. The SDW susceptibility \( \chi_{SDW} \) diverges near \( \gamma \), as

\[
\chi_{SDW} = \frac{\Delta_{SDW}(y)}{\Delta_{SDW}(0)} \sim (y_c - y)^{-2G_i + G_d}/j(\gamma) \tag{9}
\]

This describes a potential instability towards SDW formation, which will compete with the SCd instability. The SDW instability arises provided there is at least partial nesting (that is, the nesting parameter \( d_i(y_c) \neq 0 \). However, as \(-G_i > G_d\) for all \( 0 < d_i(y_c) \leq 1 \) (Fig. 3 inset), it follows from comparison of equations (9) and (8) that the SCd susceptibility diverges faster than the SDW susceptibility, for all values of nesting. At perfect nesting \((d_i = 1)\), the SCd susceptibility diverges as \((y_c - y)^{-1.5}\), whereas the SDW susceptibility diverges only as \((y_c - y)^{-1}\). As we move away from perfect nesting, the SCd susceptibility diverges faster, and the SDW susceptibility diverges more slowly, so that SCd is the leading instability for all values of nesting, within validity of the renormalization group. This is in contrast to the square lattice\(^{31}\), where at perfect nesting the SCd and SDW instabilities have the same exponent under the renormalization group, with subleading terms lifting the degeneracy in favour of SDW, which is in turn overtaken by SCd at some \( d_i < 1 \).

We also considered the phonon-mediated attraction in the pairing channel. This interaction could induce \( s \)-wave superconductivity provided that it overwhelms the electronic repulsion in the \( s \)-wave channel at the Debye frequency scale, \( \omega_D < \Lambda \). In our renormalization group flow, the \( s \)-wave coupling \((2g(3) + g(4))\) remains positive and initially drops, in complete analogy with the McMillan regime in conventional superconductors. However, it eventually changes trend and becomes more repulsive at larger renormalization group scales. The conventional \( s \)-wave pairing mediated by phonons may occur if the initial suppression is strong enough at the renormalization group scales corresponding to \( \omega_D \).

Finally, we considered the possibility of ordering in a channel exhibiting only a \( \log^3 \) divergence, for example the Pomeranchuk ordering. However, we found that such orders cannot compete with superconductivity (see Supplementary Information).

### Competition of \( d \)-wave orders below \( T_c \)

We now investigate the competition of the \( d_{x^2-y^2} \) and \( d_{xy} \) superconducting orders (equation (6)) below \( T_c \). In this regime, the system may either develop one of these two orders, or a linear combination of the two. The ordered state that minimizes the free energy wins. The hexagonal lattice point group symmetry dictates that the free energy below \( T_c \) must take the form\(^{45}\)

\[
F = \frac{1}{2}(T - T_c)(\Delta_1^2 + \Delta_2^2) + K_1(\Delta_1^2 + \Delta_2^2)^2 + K_2|\Delta_1^2 + \Delta_2^2| + O(\Delta^4) \tag{10}
\]

with \( K_1 > 0 \). This free energy allows for two possible superconducting phases. If \( K_2 < 0 \) then a \( d_{x^2-y^2} \) or a \( d_{xy} \) superconducting state would arise, whereas if \( K_2 > 0 \) then the \( d_{x^2-y^2} \) and \( d_{xy} \) orders can co-exist\(^{41}\). We now calculate \( K_2 \) (an alternative but equivalent microscopic treatment is provided in the Supplementary Information).

We begin by writing the free energy as the sum of the free energy on three patches

\[
F = F(\Delta_1) + F(\Delta_2) + F(\Delta_3) \tag{11}
\]

where the free energy on a patch is given by the standard Landau expansion

\[
F(\Delta_j) = \alpha(T - T_c)|\Delta_j|^2 + K|\Delta_j|^4, \quad K > 0 \tag{12}
\]

In this expression, it is essential to realize that \( \Delta_1, \Delta_2 \), and \( \Delta_3 \) are not independent, but must be expressed in terms of the two parameters \( \Delta_1 \) and \( \Delta_2 \) (equation (6)). Rewriting equations (11) and (12) in terms of the two independent variables \( \Delta_{xy} \), we obtain equation (10) with \( K_1 = \frac{1}{2}K > 0 \) and \( K_2 = \frac{1}{4}K > 0 \). This implies the co-existence of \( d_{x^2-y^2} \) and \( d_{xy} \) orders. Minimization of the free energy (10) with \( K_2 > 0 \) leads to \( |\Delta_1| = |\Delta_2| \) and \( \text{Arg}(\Delta_1/\Delta_2) = \pi/2 \). This order parameter can be rewritten as a three-component vector in the patch basis, which takes the form

\[
\Delta_j = d \Delta_j = (1, e^{2\pi i/3}, e^{-2\pi i/3})
\]

In this state, the superconducting gap varies around the Fermi surface as \( \Delta \exp(\pm 2\pi i/3) \). Such an order parameter corresponds to \( d_{xy}(y_j) \) (or \( d + id \)) superconductivity (Fig. 4), and is a spin-singlet analogue of the \( p + ip \) state that has been predicted for \( \text{Sr}_x\text{RuO}_4 \).

### Outlook

The robustness of \( d + id \) superconductivity in the weak coupling limit, demonstrated by the above analysis, leads us to believe that graphene-based chiral superconductivity can be realized experimentally. There are several questions pertaining to the behaviour of realistic systems which should be clarified by future work. Determination of the phase structure for interactions of moderate strength and of long-range character remains an open problem, as does an accurate estimate of \( T_c \), and the role of disorder, against which \( d \)-wave superconductivity is not protected. Graphene-based \( d + id \) superconductivity, if realized in experiment, will play a vital role in the development of technology designed to exploit topological superconductivity.

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Author contributions

R.N., I.S.I. and A.V.C. jointly identified the problem, performed the analysis and wrote the paper.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturephysics. Reprints and permissions information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to A.V.C.