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# Chiral Superconductivity in the Flat bands of Rhombohedral Graphene

**Signatures of Chiral Superconductivity in Rhombohedral Graphene**
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*Recommended with a Commentary by Ya-Hui Zhang (Johns Hopkins University) and Ashvin Vishwanath (Harvard University)*

**Background:** There are several compelling reasons to search for chiral superconductors, where superconductivity coexists with significant time-reversal symmetry breaking. Firstly, in most solids the energy scales associated with electron pairing are much smaller than typical kinetic energies, so the emergence of superconductivity relies on the degeneracy of the electron dispersion: $E(k) = E(-k)$. Such a condition, reminiscent of nesting, is ultimately governed by symmetries like time reversal or inversion, which enables even relatively weak attractive interactions to have a profound effect. Consequently, the observation of superconductivity in the *absence* of such symmetry, strongly suggests the presence of novel physics.

Secondly, the search for chiral superconductors is closely connected to the pursuit of topological superconductivity, a holy grail in condensed matter physics. A two dimensional superconductor with a spinless single-component Fermi surface is likely to exhibit time reversal symmetry breaking $p+ip$ pairing. This type of superconductivity is linked to the presence of Majorana zero modes in vortices and at the edges, which are a key resource for topological quantum computing. Such p-wave pairing is thought to be realized in superfluid $\mathrm{He}_3$ and in composite fermions at $\nu = \frac{5}{2}$ in quantum Hall systems[1], giving rise to the Moore-Read Pfaffian state. In principle, similar $p + ip$ pairing could be realized for electrons, leading to a chiral superconductor. To achieve such a state in real materials, a single-component Fermi surface and a pairing instability are both required. The first condition is not hard to achieve eg. by spin polarization, but meeting the second condition simultaneously has been challenging.

The **featured reference** provides compelling evidence for the emergence of chiral superconductivity ($T_c \approx 0.3$K) in an exceptionally simple materials system composed entirely of carbon. The material platform is a stack of 4 or 5 sheets of graphene. Additionally, three further conditions must be met - (i) the graphene sheets are in the rhombohedral stacking arrangement, (ii) a strong vertical electric field (also called a displacement field) $D$ is applied to the stack to create flat bands, and (iii) a small density of electrons, a fraction of $10^{12}$ per square centimeter is introduced.

Interestingly, this setup is almost identical to those discussed in two previous Journal Club articles [2, 3, 4], on the integer and fractional quantum anomalous Hall effects in multilayer rhombohedral graphene. In those studies, a weak moiré potential was applied to the electrons through an aligned boron nitride substrate on the opposite side of the graphene stack. In contrast, in the featured reference, no noticeable moir’e potential is present. The observed phenomena also differ: superconductivity in the current study, as opposed to the quantum anomalous Hall effects reported previously. However, a common feature is the spontaneous breaking of time reversal symmetry, which is also present in the moiré-less limit, pointing to chiral superconductivity reported in the featured reference.

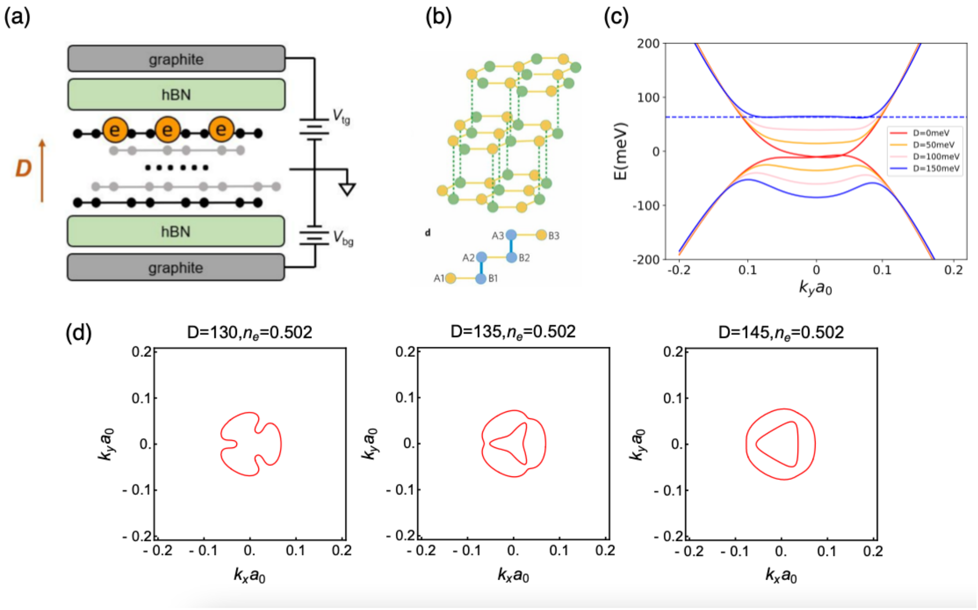

Figure 1: (a) Illustration of the experimental system. Both top and bottom hBN layers are misaligned so there is no detectable moiré. (b) Illustration of ABC stacking for $n = 3$. Low energy band is dominated by $A_1$ and $B_n$. (c) Illustration of bands at valley $K$ for different displacement fields $D$ (in units of meV) in tetra-layer graphene. $D$ opens a gap and makes a portion of the band bottom quite flat. (d) Fermi surface evolution at fixed $n$ under $D$ for one spin-valley flavor. $n$ is in units of $10^{12}$ cm$^{-2}$. (c)(d) are calculated using parameters in Ref. [5]. (Figure (a) taken from the featured reference).

**Rhombohedral multilayer graphene:** consists of $n$ graphene layers in the specific ABC stacking. Although for $n > 2$, the most stable configuration is in ABA stacking, recent experiments have successfully fabricated samples in ABC stacking for $n \geq 3$, as shown in figure 1(b). For large $n$ such as $n = 4, 5$, and strong displacement fields $D$, the free electron dispersion is shown in Figure 1c,d. In a single valley, the band bottom of the conduction band is quite flat and separated from the valence band by a large gap (Figure 1c). We note that for large $D$, the conduction band bottom is roughly similar for $n = 4, 5, 6, 7$, so theoretically we expect only weak dependence on the number of layers. The band structure in the opposite valley is related by time reversal symmetry.

In both the featured reference and the previous experiments by the same group, [4] with a moiré superlattice, the sample is dual gated, so one can tune the density $n$ and displacement field $D$ separately. Along a stripe of $(n, D)$ space, the Fermi energy is expected to be around the flat band bottom. In the previous aligned sample with the moiré superlattice, quantum anomalous Hall effects were observed in this region. In contrast, in the current moiré-less sample, a portion of the same $(n, D)$ region is replaced by superconductors.

**Experimental data on chiral superconductor:** In Fig. 2(a)(b) we reproduce the phase diagram for both tetra-layer and penta-layer samples from the featured reference. Both samples show three superconductor regions: SC1, SC2 and SC3. We will focus on SC1 and SC2 which are chiral. The critical temperature ($T_c$) is around $200 - 300$ mK. Additionally, the single most striking piece of evidence for time reversal breaking is that in the normal state, the Hall angle from $\tan\theta_H = \frac{R_{xy}}{R_{xx}}$ can be *as large as* 0.1. To our best knowledge, this is the first superconductor whose normal state has such a large Hall angle.

The experimental data strongly suggests that both SC1 and SC2 have *spontaneous* valley polarization and emerge from a fully spin-valley polarized metallic normal state. Of course this scenario is consistent with the large anomalous Hall effect in the normal state. Furthermore, there are clear quantum oscillations (see Fig. 2(c)) in a nearby metallic phase on the right side of SC1, which implies that at least the *nearby* phase is a fully spin-valley polarized quarter-metal (QM) phase*. The next question is whether SC1 is also spin-valley polarized. We can get an indirect answer to this question. First, consistent with a fully spin-valley polarized superconductor, both SC1 and SC2 are robust up to the largest in-plane magnetic field $B_{\parallel} = 5$ T. Also, the evolution of phase boundaries under out-of-plane magnetic field $B$ shown in Fig. 2 (d). If SC1 is not fully spin-valley polarized, then the spin and valley Zeeman coupling will lower the energy of the QM phase compared to SC1, so we expect that SC1 phase would shrink and gave away to QM. However, the boundary between SC1 and QM stays independent of $B$, suggesting that they have equal valley polarization. On the other hand, the left phase boundary of SC1 moves to even SC1. All of these suggest that SC1 is also a fully spin-valley polarized phase.

**A Second Metallic Phase:** In the normal state at $T = 480$ mK, i.e. above $T_c$, there seems to be a 'phase boundary' corresponding to sudden resistivity drop, which separates the QM phase and an undetermined metal (UM) phase (See Fig. 2(e)). There are anomalous Hall signals in both UM and QM, so UM likely also has a valley polarization, A reasonable guess for the UM phase could be a fully spin-valley polarized quarter-metal, but with *annular* Fermi surface. Such an evolution of Fermi surface shape is expected on applying a displacement field (Figure 1(d)). While further experiments are needed to fully understand the nature of this UM phase, one may speculate that SC1 and SC2 emerge from the simply connected (QM) and annular Fermi surface (UM) respectively.

**Critical Field and BCS-BEC regime:** Given the experimental evidence for a fully spin-valley polarized superconducting state and the absence of inversion symmetry in these

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[*] A direct confirmation of spin-valley polarization via measuring quantum oscillations is, unfortunately, not available since the experiment does not observe clear quantum oscillations above $T_c$ for SC1 and SC2,. Presumably, this is due to the low mobility because of large effective mass.

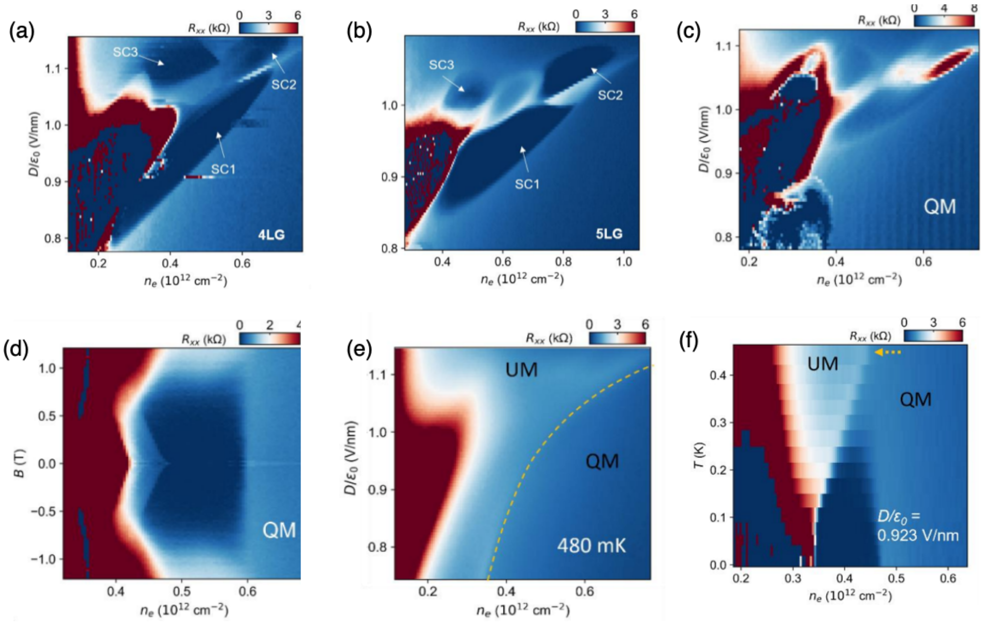

Figure 2: (a)(b) Phase diagram in $n = 4$ and $n = 5$ system. (c) Tetra-layer sample under a magnetic field $B_{\perp} = 1$ T. From quantum oscillation, the nearby phase in the right side of SC1 and SC2 is a quarter-metal (QM) with single spin-valley polarized Fermi surface. (d) Under out of plane magnetic field $B$ for the tetra-layer sample, the SC1 phase stays robust in the right side and even expands in the left side. Measurements are taken at base temperature with $T = 7$ mK. (e) At $T = 480$ mK above $T_c$, there is a phase boundary (or maybe crossover) separating the QM phase and an undetermined metal (UM) phase. (f) At lower temperature, QM expands and UM shrinks. At $T = 0$ limit, the whole SC1 region seems to be inside the QM region. (Taken from the featured reference).

structures resulting in $\epsilon(\mathbf{k}) \neq \epsilon(-\mathbf{k})$, this setup inherently precludes a conventional weak-coupling pairing instability. The emergence of superconductivity in this scenario is therefore remarkable and likely rooted in a strong-coupling mechanism. It should not come as a surprise then that the out-of-plane magnetic field required to kill superconductivity is anomalously large: $B_{\perp;c} = 1.4$T. Further insight can be gleaned from the Ginzburg-Landau coherence length, $\xi_{\mathrm{GL}} = \sqrt{\frac{\Phi_0}{2\pi B_{\perp;c}}}$, where $\Phi_0$ is the magnetic flux quantum. Comparing $\xi_{\mathrm{GL}}$ with the inter-particle distance, $d = 1/\sqrt{n}$, one finds that in the samples SC1 and SC2, the coherence length is only marginally larger than the inter-particle distance, suggesting proximity to the BCS-BEC boundary, albeit still within the BCS regime. This contrasts sharply with SC3, which exhibits a significantly larger coherence length, indicating a more conventional BCS-like superconducting state.

**Topological superconductor?** Independent of the microscopic pairing mechanism, we can ask whether the observed superconductivity is topological. It is well known [1] that a $p_x + ip_y$ superconductor with single Fermi surface is topological in the weak-pairing limit but is converted into a trivial superconductor on increasing the strength of pairing. We emphasize that even within the weak-coupling regime, a *trivial* chiral superconductor is possible, in the annular Fermi surface case. As illustrated in Fig. 1(d), a Lifshitz transition in the Fermi surface topology is observed with increasing $D$. For $D < D_c$, a single Fermi surface is present, suggesting the potential for a topological superconductor upon weak pairing. However, for $D > D_c$, a *trivial* superconductor is obtained even with weak $p + ip$ pairing[5, 6]. Therefore, while this general setting is a promising venue for topological superconductivity, further experiments are essential to definitively establish the presence of topology and Majorana zero modes within this system.

**Pairing mechanism and PDW?** While it is hard to make assertions regarding the microscopic mechanism, if we focus on a single flavor model, it is plausible that the pairing arises predominantly from Coulomb interactions. Notably, theoretical calculations employing the random-phase approximation (RPA) have yielded $p+ip$ pairing[5, 6, 7, 8]. Strong pairing strengths were observed despite the condition $\epsilon(\mathbf{k}) \neq \epsilon(-\mathbf{k})$. However, it is important to acknowledge that RPA calculations likely overestimates pairing strengths.

Because the band bottom is at $K$ corner of the graphene Brillouin zone, even a zero-momentum pairing ($\tilde{Q} = 0$) relative to $\mathbf{K}$ can be interpreted as a *pair-density-wave* (PDW) superconductor. More interestingly, the presence of the trigonal warping term can result in a finite and incommensurate Cooper pair momentum, $\tilde{Q} \neq 0$ relative to $\mathbf{K}$ [5]. This additionally results in $C_3$ rotation breaking or a charge density wave (CDW) order depending on the form of the condensate.

**Interplay with crystalline and quantum Hall orders:** A crucial aspect overlooked in current RPA theories is the emergence of integer and even fractional quantum anomalous Hall states upon the introduction of a weak moiré superlattice potential $V_M$ within a similar $(n, D)$ parameter range [2, 3, 4]. Previous Hartree-Fock calculations demonstrate that even in the $V_M = 0$ limit, anomalous Hall crystals can form [9, 10, 11], where a spontaneously generated electron crystal gives rise to a narrow $C = 1$ Chern band. One possible scenario is that $V_M = 0$ corresponds to a chiral superconductor, which transition to an anomalous Hall crystal under the introduction of a small $V_M$. It is then conceivable that the proximate anomalous Hall crystal phase plays a significant role in the emergence of the chiral superconducting state. Bringing together the various aspects of rhombohedral graphene - crystalline order, chiral superconductivity and quantized anomalous Hall effects - into a unified and coherent framework is an exciting direction for future research.

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