Exotic Forms of Superconductivity

Liang Fu (MIT)

Exotic Forms of Superconductivity

- Pairing & SC from strong repulsion
- Spin-polarized SC
- Finite-momentum SC

Collaborations

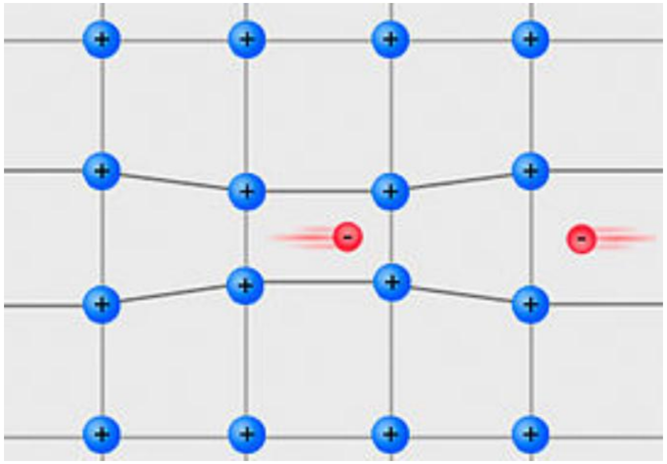
Kevin Slagle
(Caltech => Rice => Magic)

Valentin Crepel
(MIT => Flatiron)

Daniele Guerci

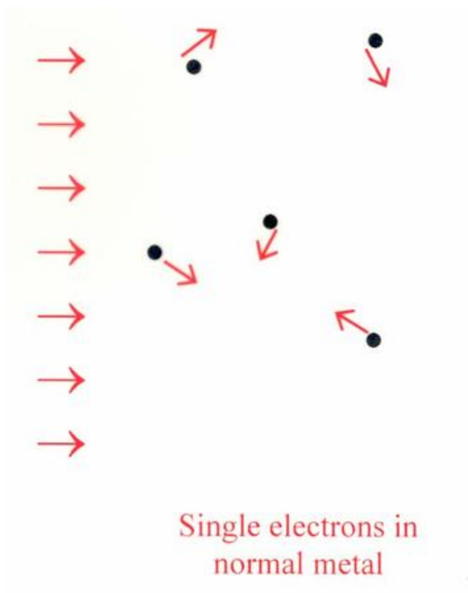
Thanks to Tommaso Cea, Paco Guinea, Max Geier and Lina Kamra
for related collaborations

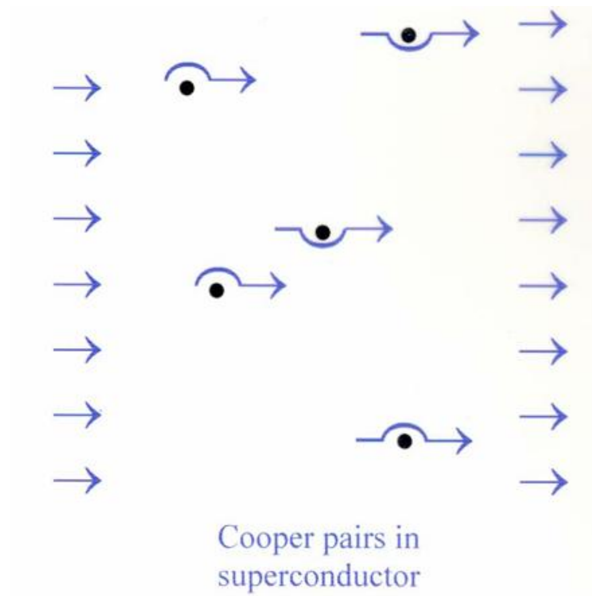
Electron-Phonon Superconductors



In most SCs, lattice vibration provides the "glue" that binds electrons into pairs.

Cooper Pairing & Superconductivity





(Leggett)

Pairs of electrons condense into same state:

$$\Psi_{\mathrm{BCS}}(\{r\}) = \mathrm{Det}[\psi(r_i, r_j)]$$

Cooper Pairing & Superconductivity

Think of Cooper pairing as a kind of marriage. Just as marriage can help two people sail through life's ups and downs by joining forces, so Cooper pairing allows electrons to travel through a conductor without getting bogged down in lots of troublesome little obstacles. [Chris Woodford, "How cool stuff works."]

Spin-Singlet Pairing

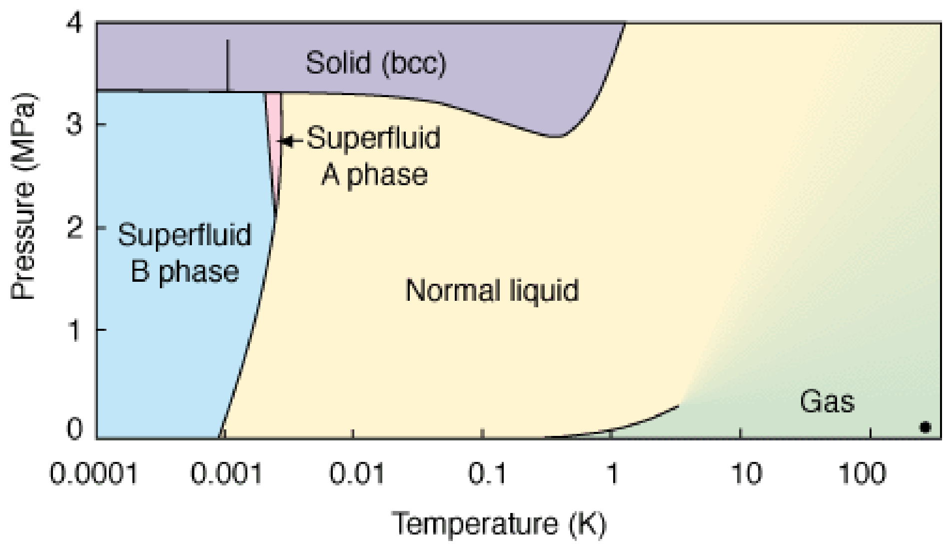
$$\psi(r_1, r_2; s_1, s_2) = \psi(r_1, r_2)|\uparrow\downarrow - \downarrow\uparrow\rangle$$

with $\psi(r_1, r_2) = \psi(r_2, r_1)$

Almost all known SCs are spin singlet.

Superfluid He-3

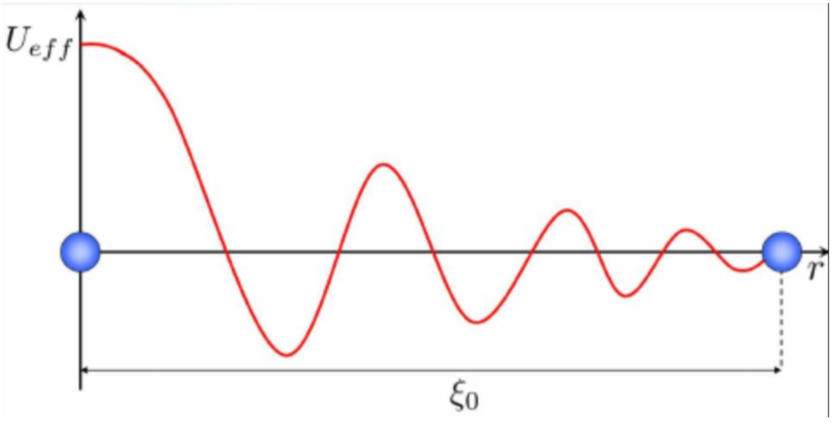
Paired state of neural atoms that strongly repel at short distance



Spin-triplet pairing:

$$\varphi(\mathbf{r}_1 - \mathbf{r}_2 : \sigma_1\sigma_2) = \varphi_{\uparrow\uparrow}(\mathbf{r}_1 - \mathbf{r}_2)\,|\uparrow\uparrow\rangle + \varphi_{\uparrow\downarrow}(\mathbf{r}_1 - \mathbf{r}_2)\,|\uparrow\downarrow + \downarrow\uparrow\rangle + \varphi_{\downarrow\downarrow}(\mathbf{r}_1 - \mathbf{r}_2)\,|\downarrow\downarrow\rangle.$$

Kohn-Luttinger Mechanism

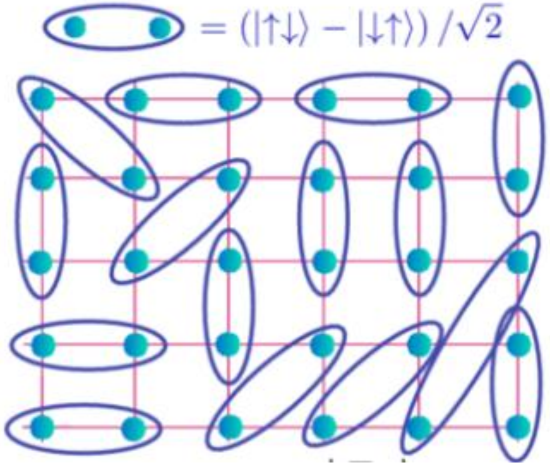


- attraction from *many-body* effect: Friedel oscillation
- controlled calculation for weak repulsive interaction yields T_C that is exponentially small.

$$T_C/T_F \sim \exp\left(-\frac{1}{g}\right), \qquad g \propto U^2 \ll 1$$

SC in Doped Mott Insulator

Shortly after the discovery of high-Tc SC, Anderson proposed that (1) the undoped Mott insulator is a quantum spin liquid composed of spin singlets; (2) doping turns singlets into mobile Cooper pairs.



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However, experiments showed that the undoped insulator is an antiferromagnet. And despite decades of theoretical and numerical studies, whether high-Tc SC occurs in doped Hubbard model remains debated.

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Challenge: lack of analytical control at strong interaction (contrast with BCS theory at small phonon frequency and electron-phonon coupling and Kohn-Luttinger theory at small repulsive interaction strength)

SC From Doping Band Insulators

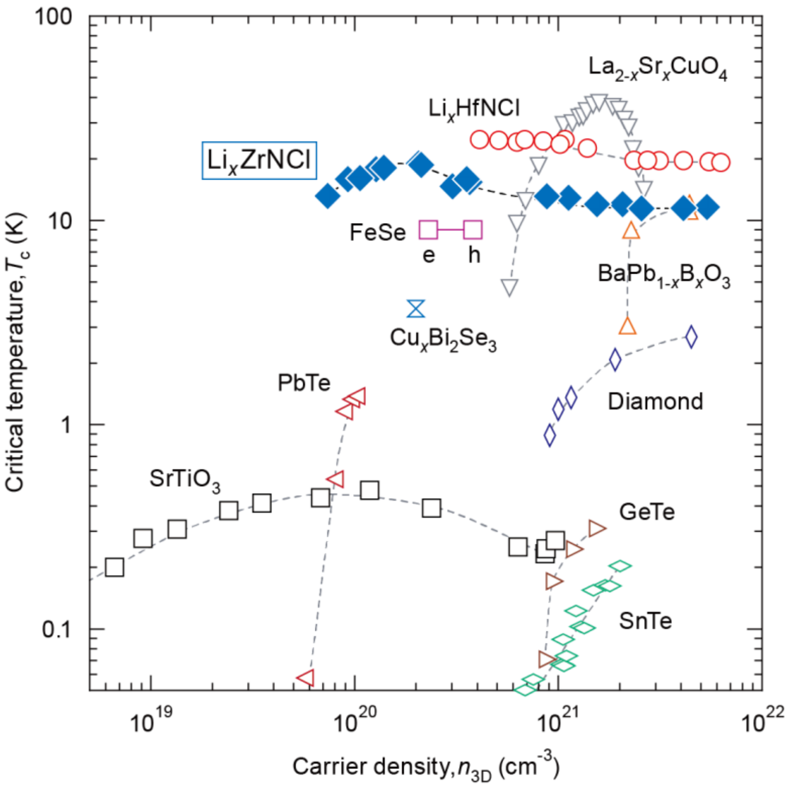
Many surprises:

- low carrier density & small E_F
- (relatively) high Tc: 20K in ZrNCl
- doped topological insulators:

  $Cu_xBi_2Se_3$, SnTe, WTe_2
- doped large-gap trivial insulators:

  ZrNCl, $SrTiO_3$

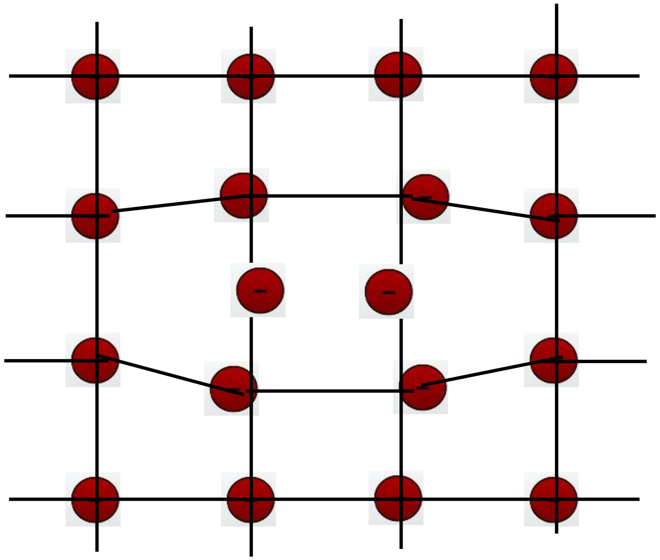
Electronic pairing mechanism ?



Iwasa et al, arXiv (2020)

Exciton Mediated Pairing

SC from particle-hole fluctuations in multiband systems:



Excitons replace phonons
to mediate pairing.

- related idea dates back to Little (1964), Bardeen (1973)...
- theoretical advance hindered by lack of small parameter
- pairing symmetry? high-Tc? competing phases?

The N+2 Problem

We first solve the problem of two doped particles in band insulators with (strong) repulsive interaction to demonstrate the possibility of bound state, i.e., attraction from repulsion.

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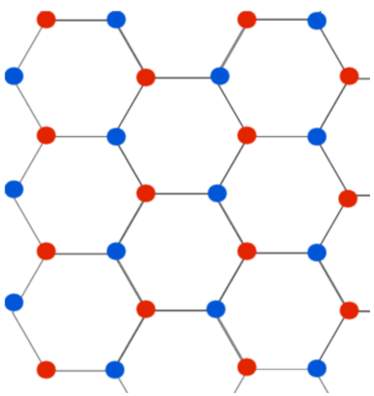
Minimal model: spinless "Hubbard" model on bipartite lattice

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_t,$$
$$\mathcal{H}_0 = V \sum_{\langle r,r' \rangle} n_r n_{r'} + \Delta \sum_{r \in B} n_r,$$
$$\mathcal{H}_t = -t \sum_{\langle r,r' \rangle} (c_r^\dagger c_{r'} + hc).$$



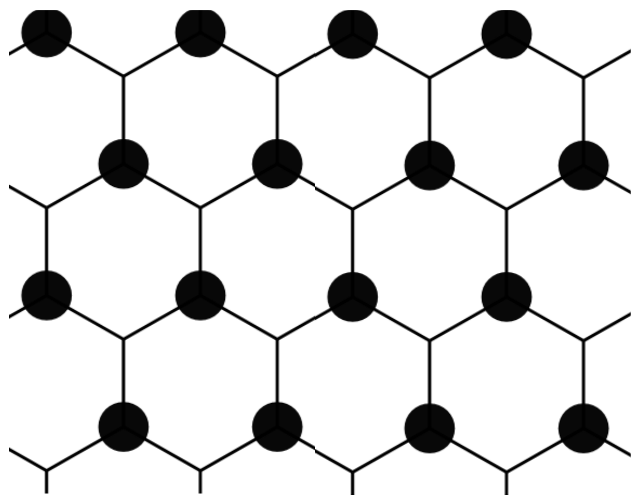
Slagle & LF, PRB (2020)

Rigorous Methods

Expansion in kinetic hopping t:

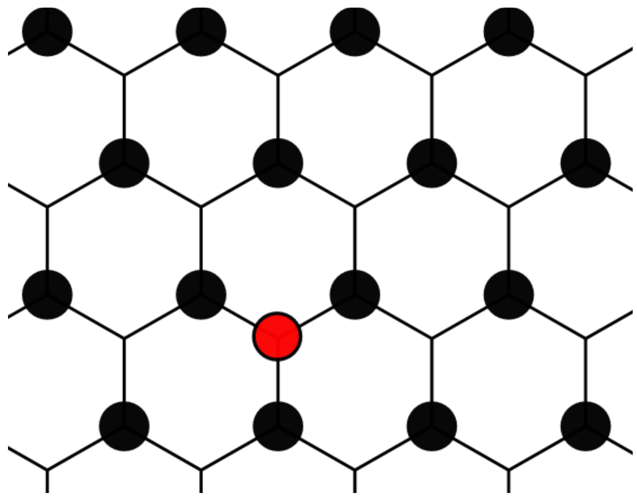
- small t/Δ Crepel & LF, Sci Adv (2021)
- small t/V Guerci & LF, 2503.05863

Strong Interaction Regime: $V \gg t$



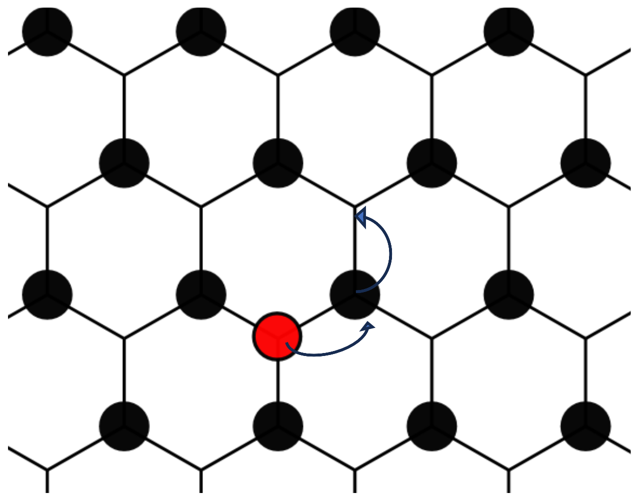
For $V \gg t$, ground state at half filling is a sublattice polarized insulator, with fermions occupying A or B sites depending on the sign of Δ.

One Doped Particle



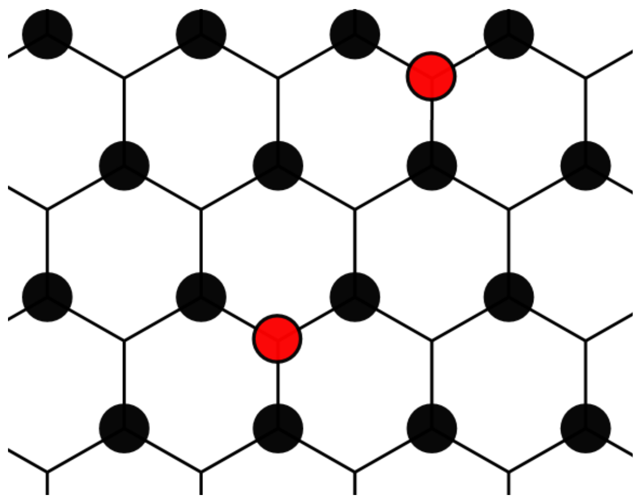
For $V \to \infty$, $E_1 = \Delta + 3V$

One Doped Particle



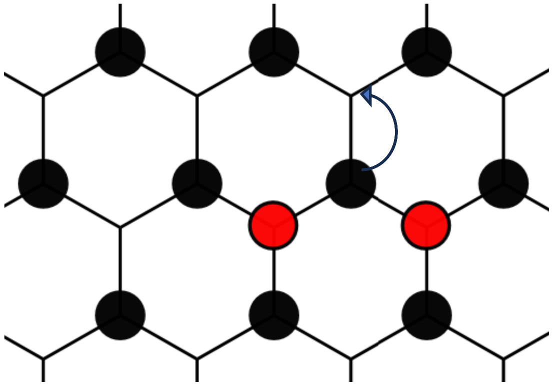
For small t/V, hopping via second-order kinetic process with amplitude $t_f = \frac{t^2}{\Delta + V}$.

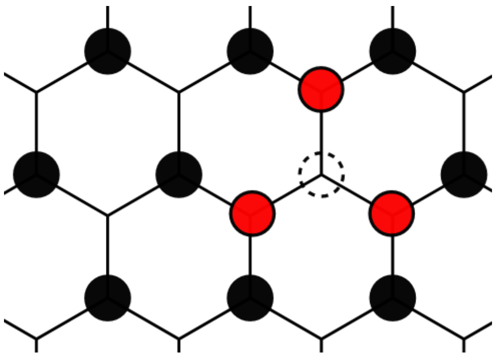
Two Doped Particles



For small t/V, $2E_1 = 2\Delta + 6V + o(t^2/V)$

Excitonic Cooper Pair

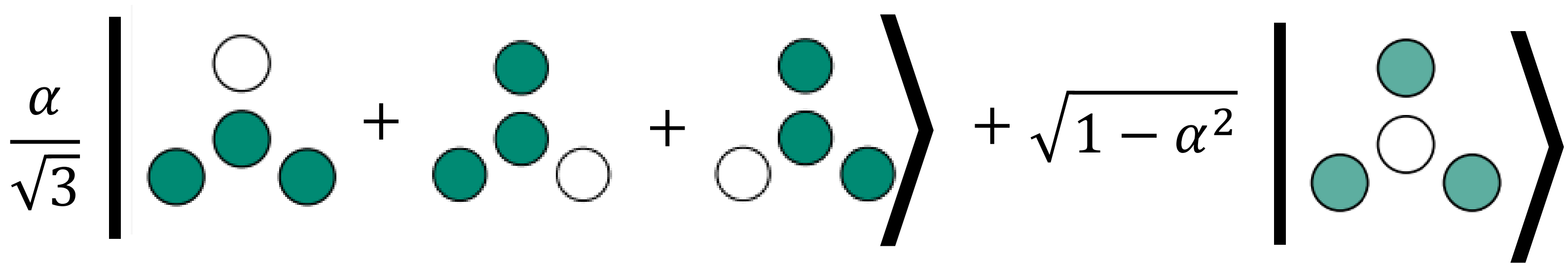




First-order kinetic process without costing interaction energy induces tight bound state of two doped particles dressed with an exciton $E_2 = 2E_1 - \epsilon_b$

Guerci & LF, 2503.05863

Excitonic Cooper Pair



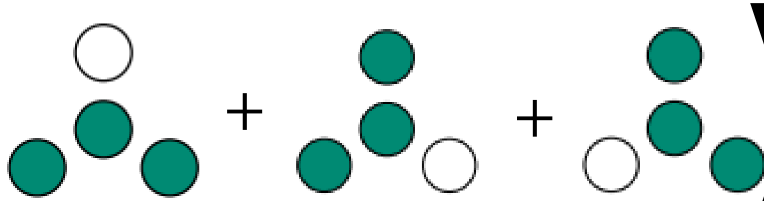
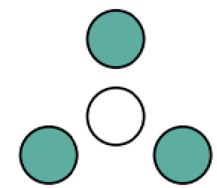
- f-wave symmetry
- hybridized state of Cooper pair and exciton: $\alpha^2 = \frac{1}{2} + \frac{1}{\sqrt{4+48t^2/\Delta^2}}$
- binding energy $\epsilon_b = \sqrt{\frac{\Delta^2}{16} + \frac{3t^2}{4}} - \frac{\Delta}{4}$

$\Delta \gg t$: $\alpha \to 1$ and $\epsilon_b \to 3t^2/\Delta$

$\Delta = 0$: $\alpha = \frac{1}{2}$ and $\epsilon_b = \sqrt{3}t/2$

large binding energy affords robustness to perturbations.

Excitonic Cooper Pair

$\frac{\alpha}{\sqrt{3}}$ |  ⟩ $+ \sqrt{1-\alpha^2}$ |  ⟩

Attraction emerges at strong repulsion !

At large V/t, pairing energy is much larger than fermion bandwidth:

$$\epsilon_b \gg W_f = \frac{9t^2}{\Delta + V}$$

Therefore, low-energy charge carriers are charge-2e bosons.

Guerci & LF, 2503.05863

Three-body problem: pairing induced by third electron !

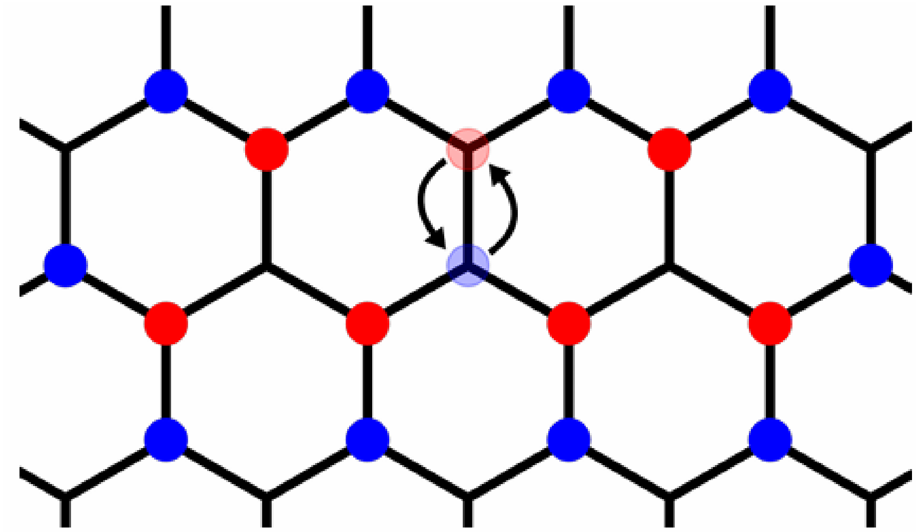
Excitonic Cooper Pairs at Finite Density

Unlike weak pairing BCS theory, pairing in general does NOT guarantee superconductivity.

At finite doping density, Bose condensation of mobile electron pairs leads to a BEC superconductor. Alternatively, they can phase separate into a high-density region and an undoped region.

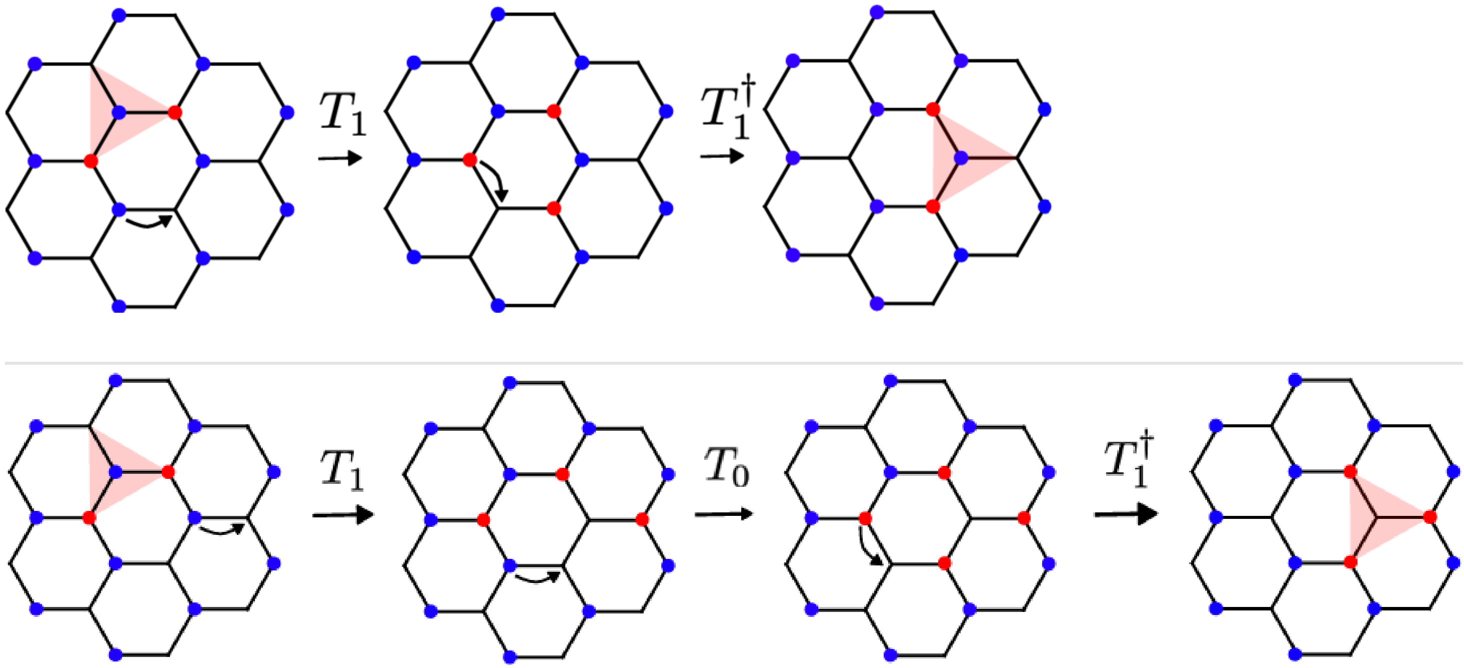
Excitonic SC competes with phase separation!

Phase Separation



At $V \to \infty$, excitonic Cooper pairs are immobile and attract at next nearest neighbor distance, leading to phase separation at small doping.

Quantum Fluctuation

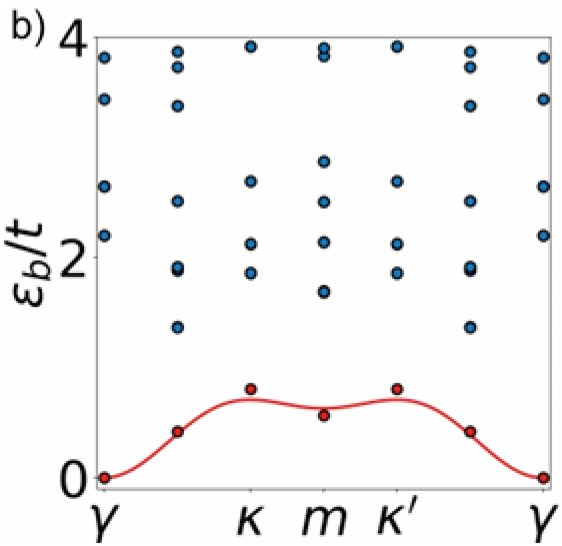


Excitonic Cooper pairs move coherently via a sequence of higher-order processes T_n that change interaction energy by nV.

Guerci & LF, 2503.05863

Mobile Electron Pair

$V = 5t, \Delta = 0$

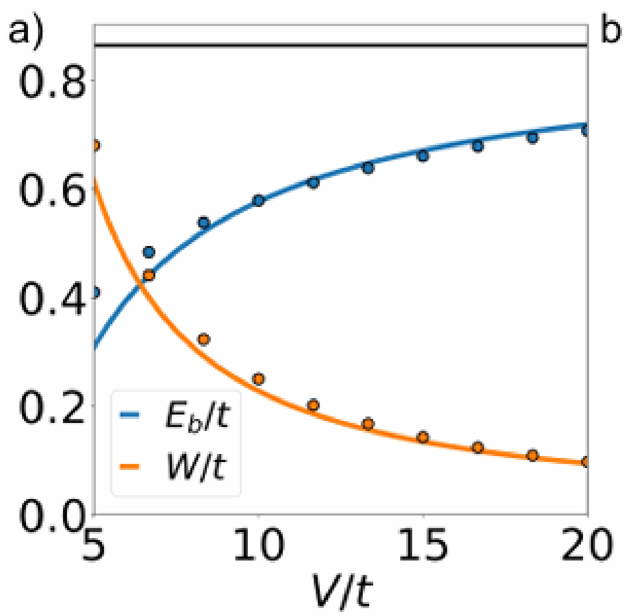


From t/V expansion to second-order:

$$t_b = \frac{t^2}{6V} + \frac{\sqrt{3}t^3}{2V^2} + \mathcal{O}\left(\frac{t^4}{V^3}\right)$$

Coherent motion of electron pair on the lattice of B sites with amplitude t_b, leading to bandwidth $W = 9t_b$.

Mobile Electron Pair



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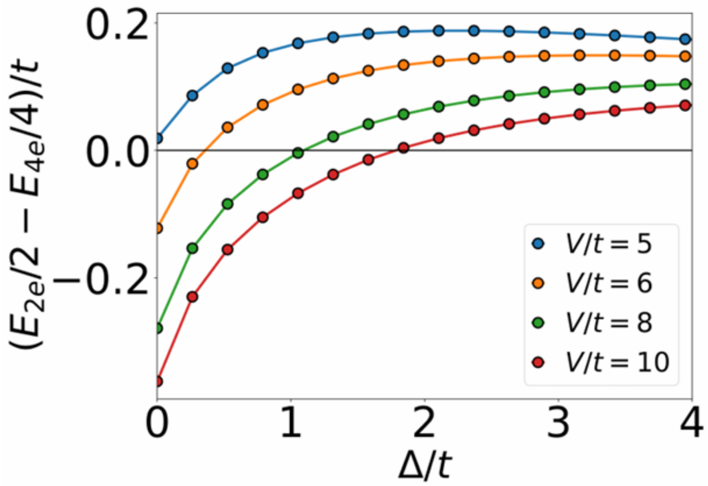
$$t_b = \frac{t^2}{6V} + \frac{\sqrt{3}t^3}{2V^2} + \mathcal{O}\left(\frac{t^4}{V^3}\right)$$

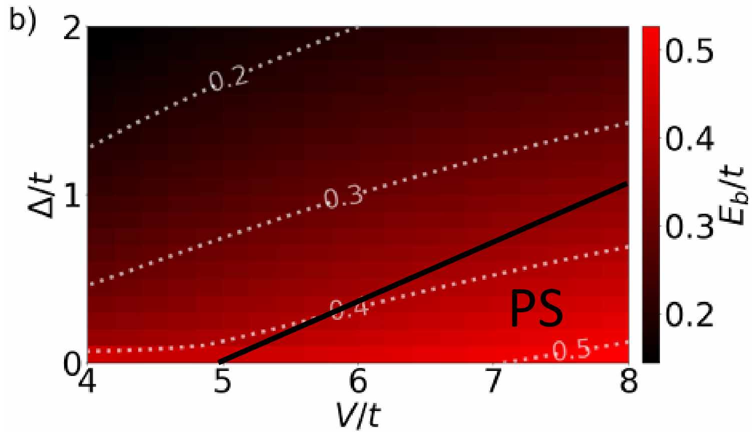
$$E_b = \frac{\sqrt{3}}{2}t - \frac{3t^2}{V} + \frac{5\sqrt{3}t^3}{8V^2} + \mathcal{O}\left(\frac{t^4}{V^3}\right).$$

As quantum fluctuation t/V increases, binding energy decreases while boson bandwidth increases.

Interaction between Electron Pairs



Interaction between electron pairs become repulsive as V reduces or Δ increases.



Optimum SC is expected near phase separation.

BEC Superconductivity

from strong repulsion

Repulsive Bose gas condenses into superfluid at low T:

$$H = \int dr\, \phi^{+} \left(\frac{-\nabla^2}{2m} - \mu \right) \phi + g|\phi|^4$$

- Quantum critical point at $\mu = 0$ with $z = 2$: g is marginal
- T_c increases linearly with doping density δ

$$k_B T_c \approx C \frac{\hbar^2 \rho}{m_b} = C \frac{W}{3\sqrt{3}} |\delta|$$

- Maximum density determined by boson size bounds T_c:

$|\delta| = \sqrt{3} a^2 / (\pi \langle r^2 \rangle)$ For $V = 8t$, $\langle r^2 \rangle \approx 1.25 a^2$ => $k_B T_c \approx 0.06t$

Large Sublattice Potential

Crepel & LF,
Sci Adv (2021)

Effective Hamiltonian for Doped Particles

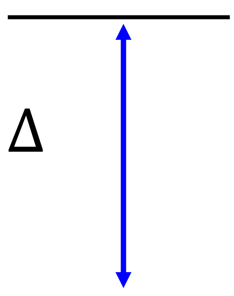
For $\Delta \gg t$, low-energy subspace: $N_A = N, N_B = N_p$

high-energy subspace: $N_A < N, N_B = N_p + (N - N_A)$

with $N_p = \delta \cdot N$ the number of doped particles.

Kinetic hopping between nearest neighbor sites changes $N_A - N_B$, thus coupling low- and high-energy subspaces.

$E_g \geq \Delta$



$$\mathcal{H}_0 = V \sum_{\langle r, r' \rangle} n_r n_{r'} + \Delta \sum_{r \in B} n_r$$

$$\equiv H_l \bigoplus H_h$$

Crepel & LF, Sci Adv (2021)

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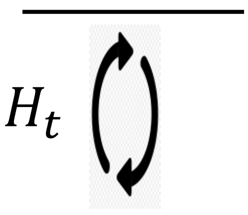
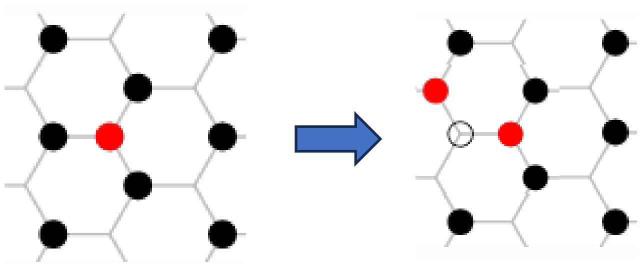


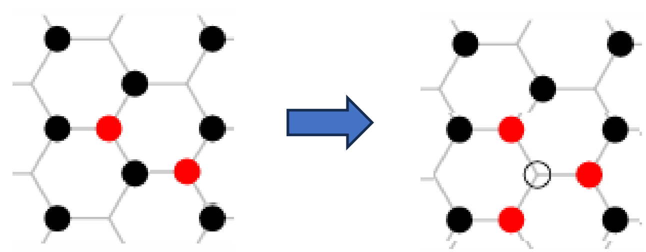
$$\mathcal{H}_t = -t \sum_{\langle r,r' \rangle} (c_r^\dagger c_{r'} + hc).$$

polaron



$N_p = 1$: $E_g = \Delta + V$

trimer



$N_p = 2,$: $E_g = \Delta$

Effective Hamiltonian for Doped Particles

Decouple low- & high-energy manifold by Schrieffer-Wolf transformation

$$\mathcal{H}' = e^{iS}\mathcal{H}e^{-iS} \qquad H_l' = H_l + \sum_n \left(\frac{t}{\Delta}\right)^n \delta H_n$$

After performing SW, off-diagonal part is removed and diagonal part acquires corrections that can be calculated perturbatively.

$$\mathcal{H}' = \sum_{\langle i,j \rangle} t_f (f_i^\dagger f_j + hc) + V_f n_i n_j + \sum_{(ijk)\in\triangle} \lambda (f_i^\dagger n_j f_k + P_{ijk}) + U_3 n_i n_j n_k.$$

$$t_f = \frac{t^2}{\Delta + V}, \qquad V_f = -\frac{t^2}{\Delta} + \frac{4t^2}{\Delta + V} - \frac{3t^2}{\Delta + 2V},$$

$$\lambda = \frac{t^2}{\Delta} - \frac{t^2}{\Delta + V} \qquad U_3 = \frac{3t^2}{\Delta} - \frac{6t^2}{\Delta + V} + \frac{3t^2}{\Delta + 2V}.$$

Effective Hamiltonian for Doped Particles

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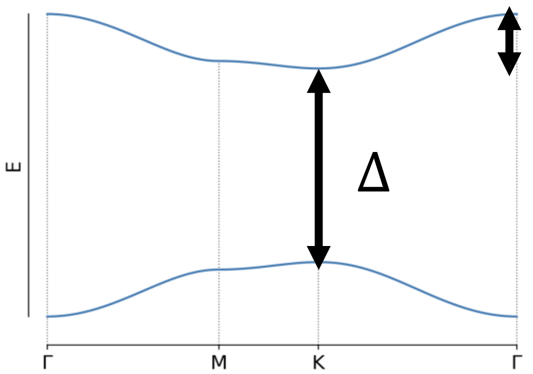
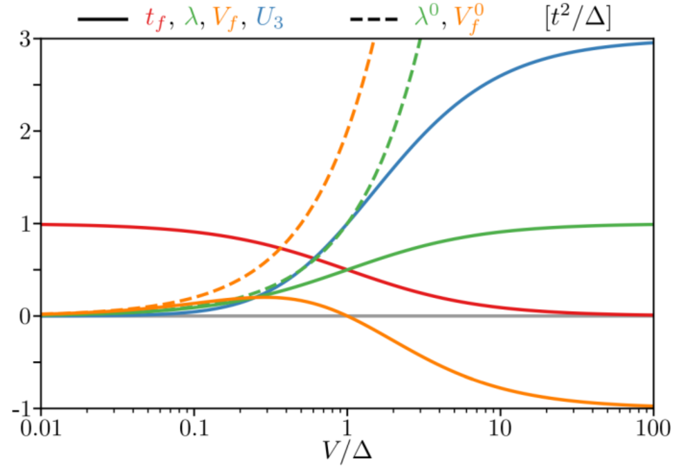
- exact at large Δ/t for <u>any doping</u> & interaction
- effective interaction is <u>instantaneous</u>

Interband Screening

Noninteracting bands

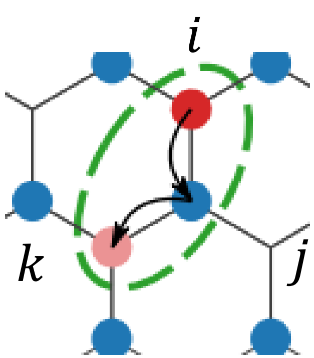


$$W = \frac{9t^2}{\Delta}$$

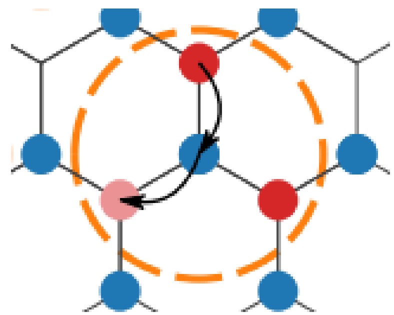


- conduction band minima at $\pm K$: valley degeneracy
- effective interaction is *strongly* modified by interband processes
- interaction projected to partially filled band misses the key!

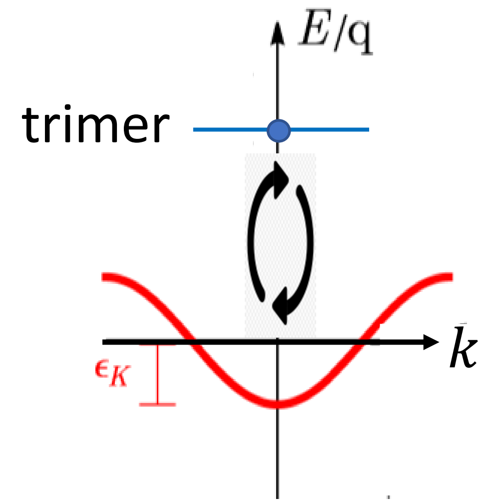
Trimer Mediated Attraction



$E_g = \Delta + V$



$E_g = \Delta$



Transition amplitude of a doped particle from site i to k depends on the occupation of nearby site j.

single-particle hopping: $t_f(f_i^\dagger f_j + hc)$

$$t_f = \frac{t^2}{\Delta + V},$$

correlated hopping: $\lambda(f_i^\dagger n_j f_k + P_{ijk})$

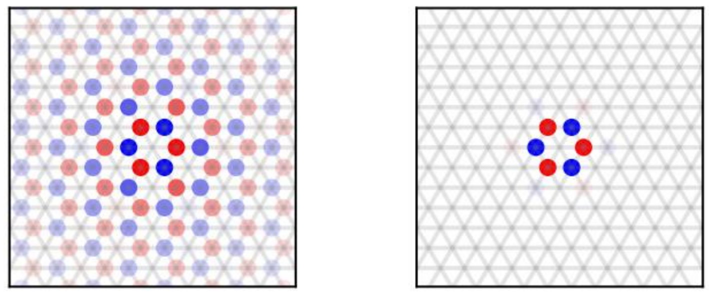
$$\lambda = \frac{t^2}{\Delta} - \frac{t^2}{\Delta + V}$$

Two-Particle Bound State

from **exact** solution of low-energy model

f-wave pairing

a)



$$\mathcal{H}' = \sum_{\langle i,j \rangle} t_f (f_i^\dagger f_j + hc) + V_f n_i n_j \; + \sum_{(ijk)\in\triangle} \lambda (f_i^\dagger n_j f_k + P_{ijk})$$

- For large V/Δ, only virtual transition to trimer occurs, leading to tight bound pair resonating within a triangle (f-wave).
- For small V/Δ, H' reduces to attractive Fermi gas with valley degeneracy.

Continuum Theory at Low Density

$$\mathcal{H}' = \sum_{\langle i,j \rangle} t_f (f_i^\dagger f_j + hc) + V_f n_i n_j \ + \sum_{(ijk)\in\triangle} \lambda (f_i^\dagger n_j f_k + P_{ijk}) + U_3 n_i n_j n_k.$$

Low-energy modes around band bottom at $\pm K$

$$\widetilde{\mathcal{H}} = \int \mathrm{d}x \sum_{\tau=\pm} \psi_\tau^\dagger \left[\frac{-\nabla^2}{2m} \right] \psi_\tau + g \psi_+^\dagger \psi_+ \psi_-^\dagger \psi_-,$$

with $m = 2/(3t_f a^2), \quad g = 6a^2(V_f - 2\lambda) < 0.$

Pseudospin-1/2 Fermi liquid with attractive contact interaction:

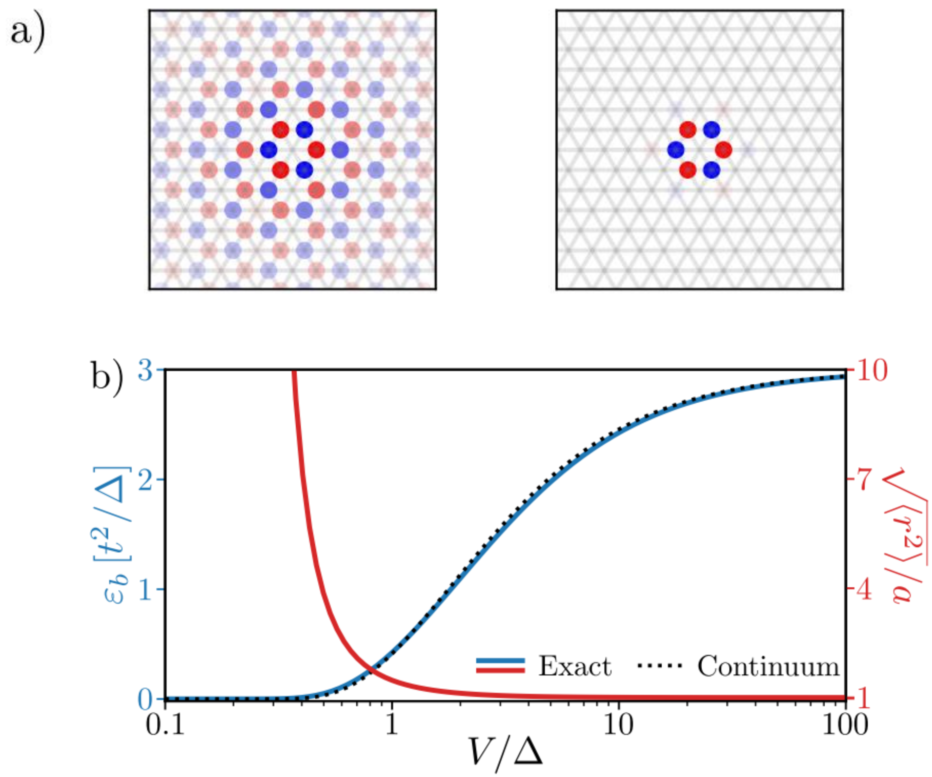
- BCS-BEC crossover tuned by interaction and density
- s-wave valley-singlet = f-wave pairing on the lattice

Two-Particle Bound State

from **exact** solution of effective model H'

f-wave pairing

Pair binding energy ϵ_b



Pair size

$$\epsilon_b \propto \frac{9W}{\pi}\left(e^{1/g_0} - 1\right)^{-1}$$

with $W = \frac{9t^2}{\Delta + V}$, $g_0 = \frac{6V^2}{\pi\Delta(\Delta + 2V)}$

nonperturbative in V

Non-BCS 2D Superconductor

Attractive Fermi liquid at $E_F \gg \epsilon_b$ (degenerate regime, weak-coupling)

$$T_c, \Delta \sim \sqrt{E_F \epsilon_b}$$

Randeria, Duan, Shieh (1993) …

Our model of SC in doped insulator

$$T_c, \Delta \propto \sqrt{\delta}\, W e^{-\frac{1}{2g_0}}$$

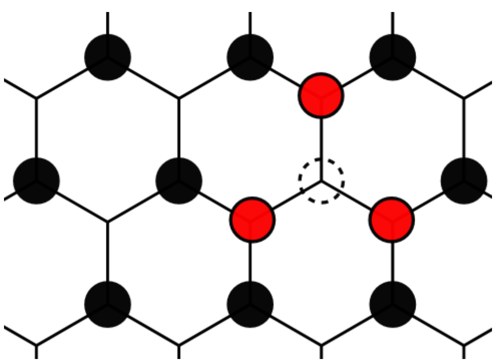
- accurate for $T_c \leq 0.1 E_F$ (weak-coupling, non-BCS)
- $\frac{\Delta}{k_B T_c} \approx \mathbf{4.79}$ (1.764 in phonon SC with Debye cutoff)
- $T_c \propto \sqrt{\delta}$ despite constant density of states

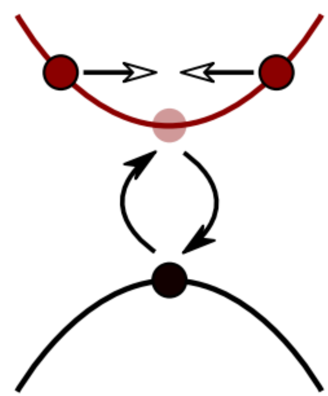
BCS-BEC crossover for $E_F \sim \epsilon_b$ (very low density, strong-coupling)

Summary

Excitonic mechanism for SC from repulsive interaction:

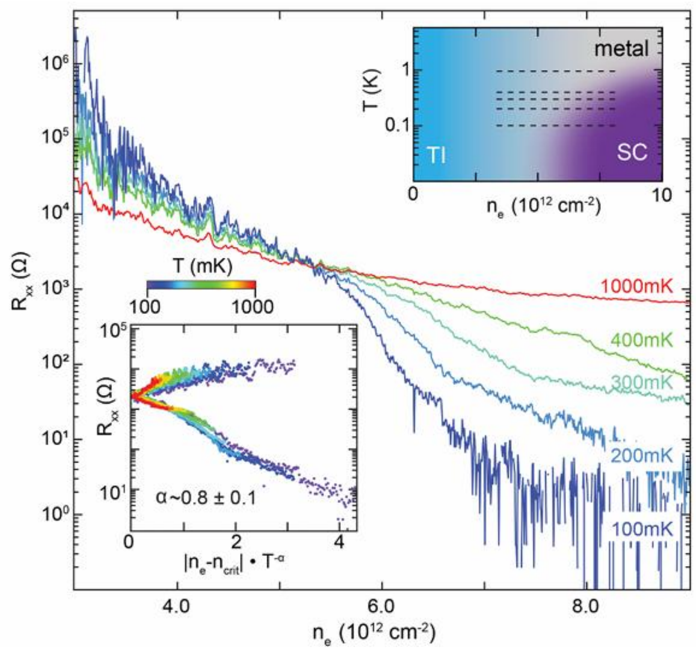
- two key parameters: band gap and interaction strength
- strong interaction:
  excitonic Cooper pair, phase separation & BEC
- large band gap:
  exciton mediated attraction between conduction electrons



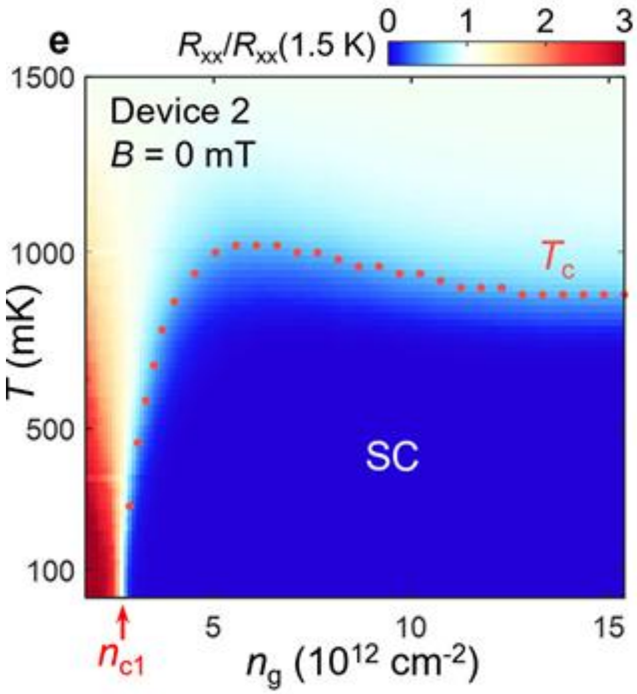


SC in 2D Materials

Gate-induced SC in band insulators: monolayer WTe_2



Sajadi et al, Science (2018)
Fatemi et al, Science (2018)



Sanfeng Wu et al, 2501.16699

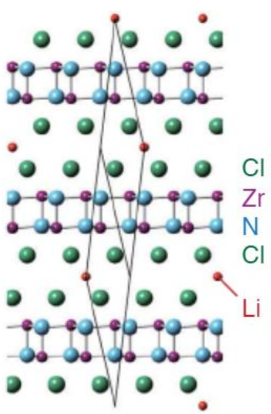
SC in 2D Materials

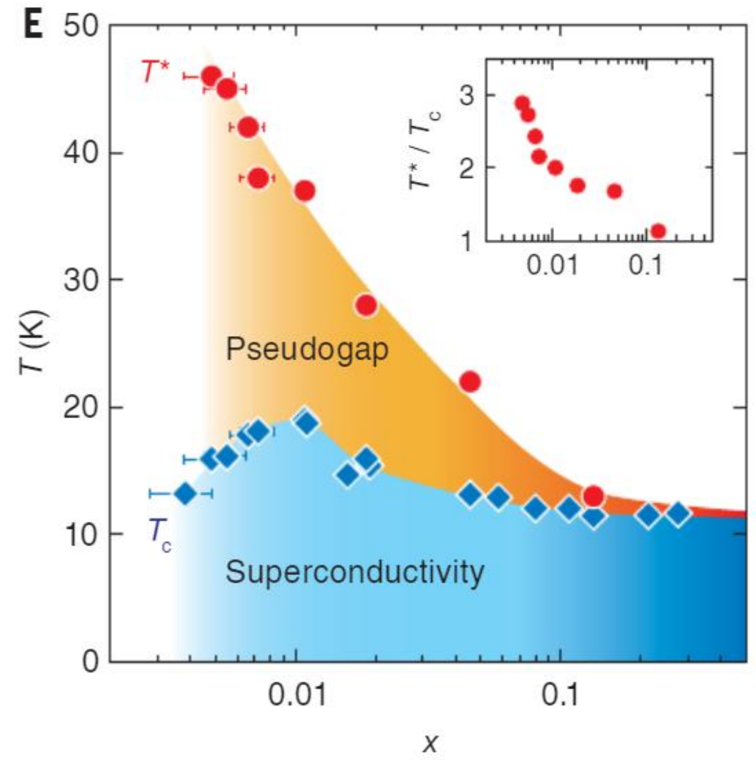
Gate-induced SC in band insulators: ZrNCl (band gap 2.5eV)

SUPERCONDUCTIVITY

Gate-controlled BCS-BEC crossover in a two-dimensional superconductor

Yuji Nakagawa[1,2], Yuichi Kasahara[3], Takuya Nomoto[1], Ryotaro Arita[1,4], Tsutomu Nojima[5], Yoshihiro Iwasa[1,2,4 *]

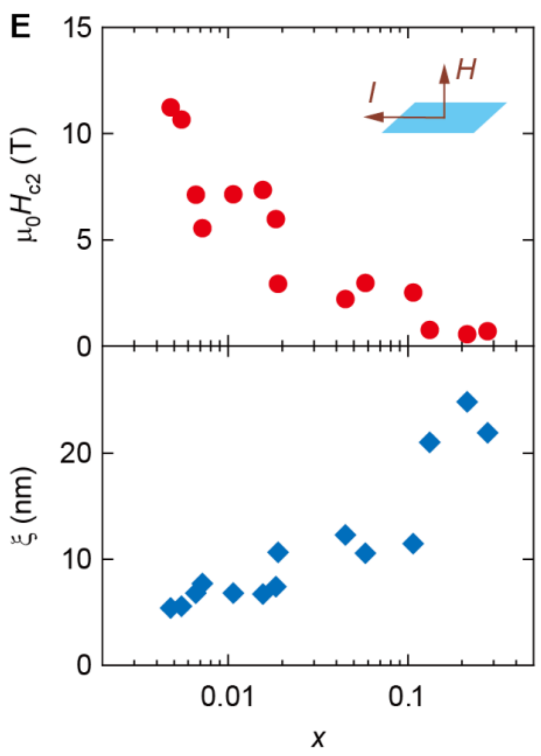




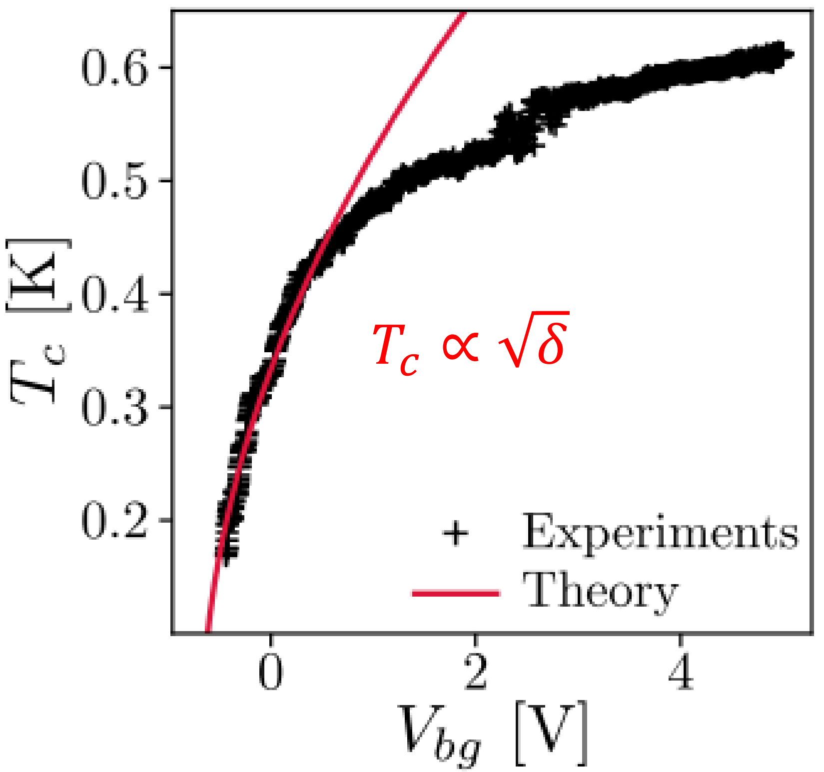
Non-BCS SC

At low doping: $\Delta \sim \sqrt{E_F \epsilon_b}$ $\quad \mathrm{v_F} \sim \sqrt{E_F/m}$

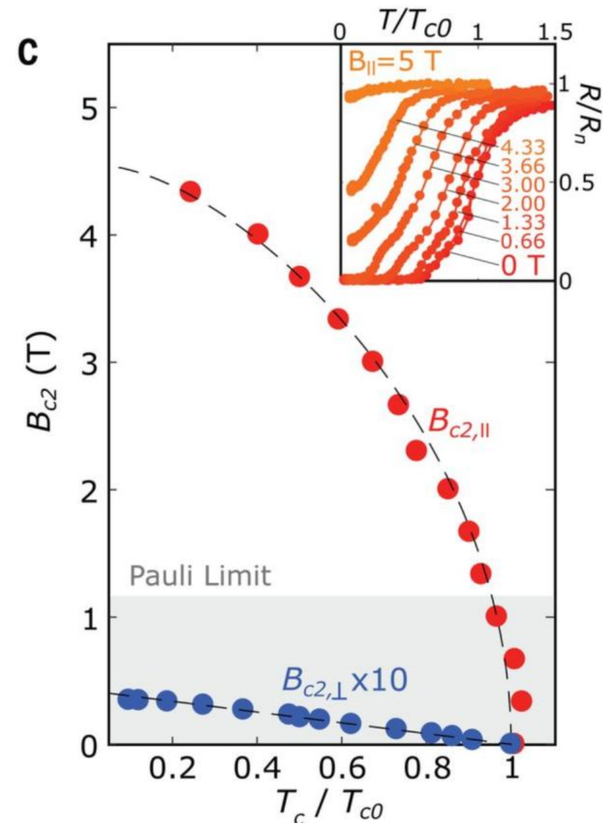
Coherence length $\xi = \mathrm{v_F}/\Delta$ is independent of doping!



Application to WTe_2

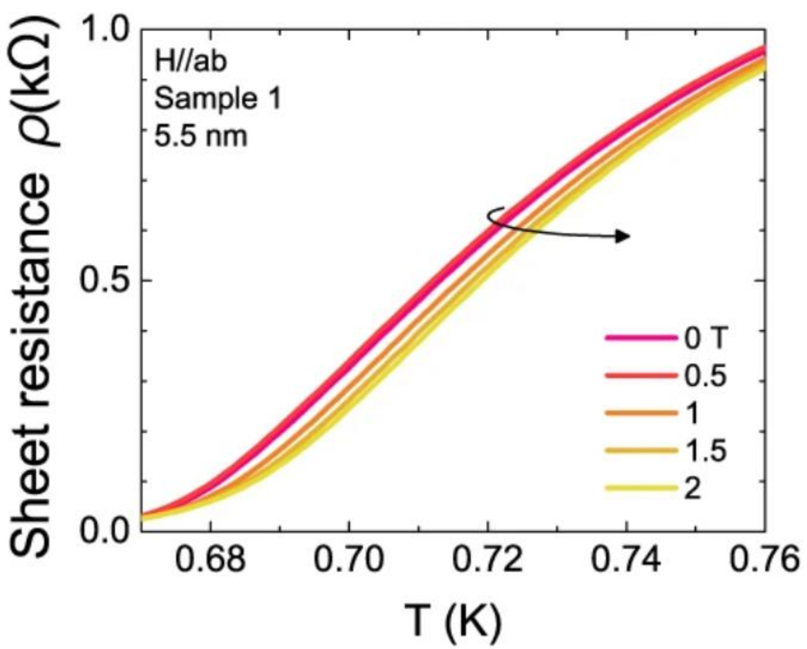


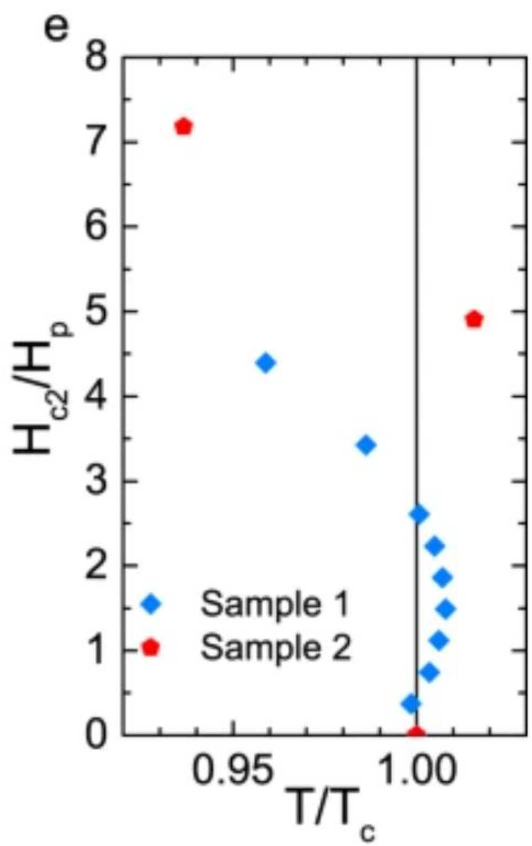
large violation of Pauli limit



Crepel & LF, PNAS (2022)

Magnetic Field Enhances Tc!





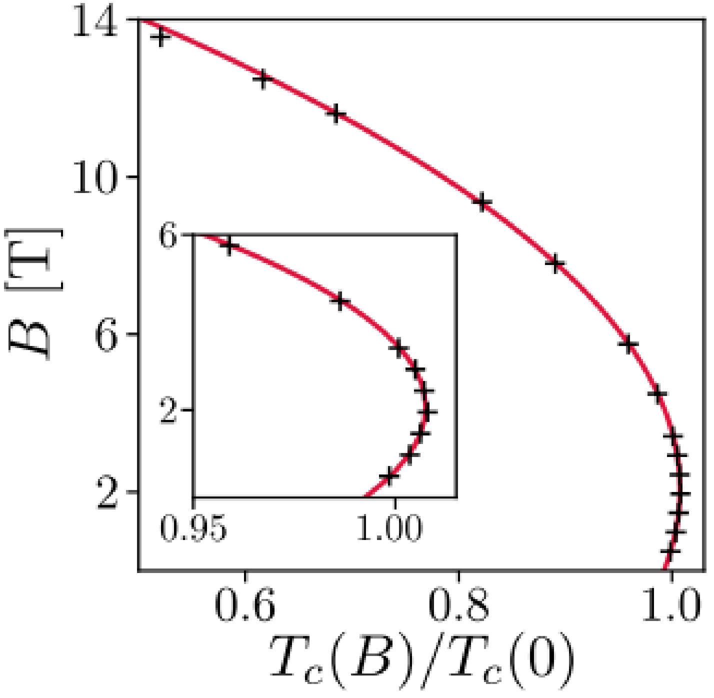
Lu Li, Sci. Rep. (2018)

Prediction: Spin-Triplet SC in WTe_2

- Spin-triplet, valley-singlet pairing parameterized by d-vector
- Zeeman effect on triplet pairs

$$F = \alpha(\mathbf{d}\cdot\mathbf{d}^*) + \mu\mathbf{B}\cdot(i\mathbf{d}\times\mathbf{d}^*) + \eta|\mathbf{B}\cdot\mathbf{d}|^2 + \chi B^2(\mathbf{d}\cdot\mathbf{d}^*),$$

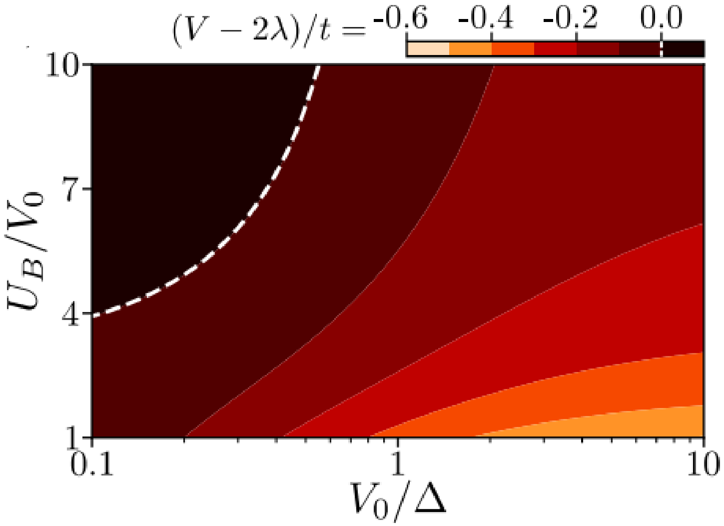
$$\Delta T_c^B = \mu B - \chi B^2$$

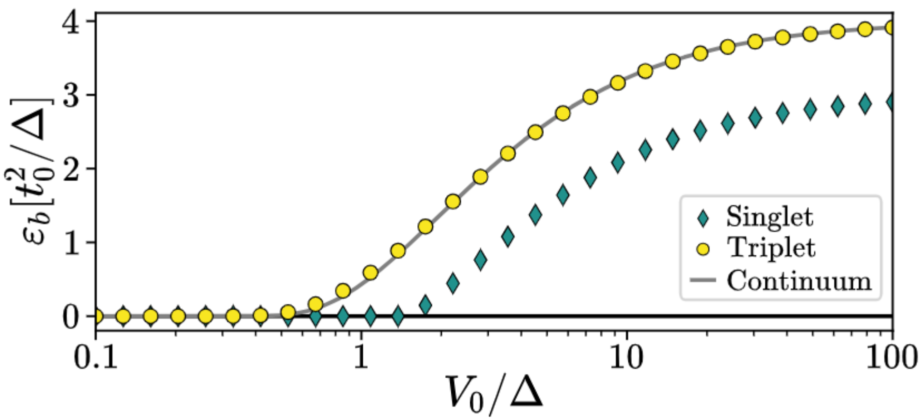


Crepel & LF, PNAS (2022)

f-Wave Spin-Triplet & Valley-Singlet Pairing

Spinful Hubbard model with U and V





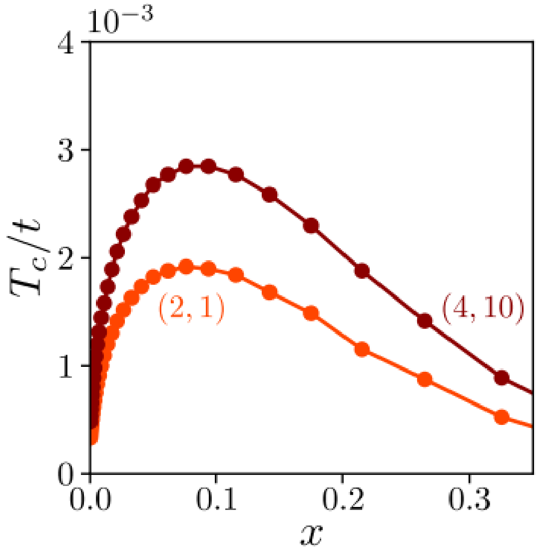
Continuum model: spin + valley

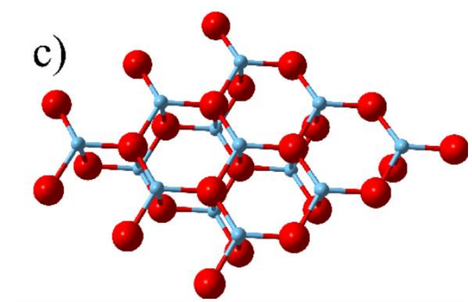
$$\tilde{\mathcal{H}}_i = \int \mathrm{d}x \; g_0(\rho_{K\uparrow}\rho_{K\downarrow} + \rho_{K'\uparrow}\rho_{K'\downarrow}) + g_1 \rho_K \rho_{K'} + g_2 \mathbf{s}_K \cdot \mathbf{s}_{K'}$$

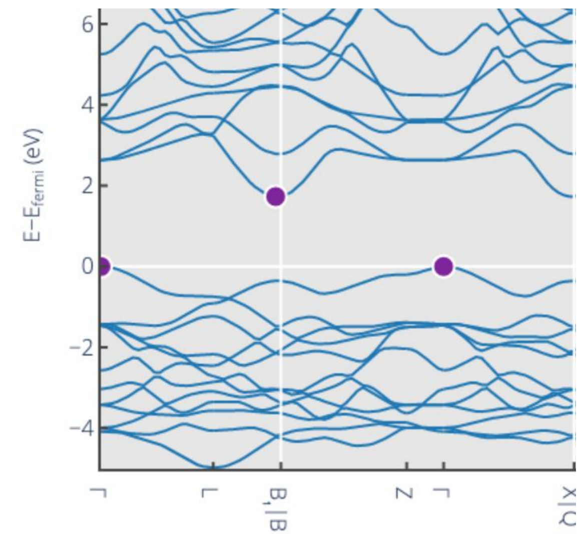
Crepel & LF, PNAS (2022)

Application to ZrNCl

Crepel & LF, PNAS (2022)







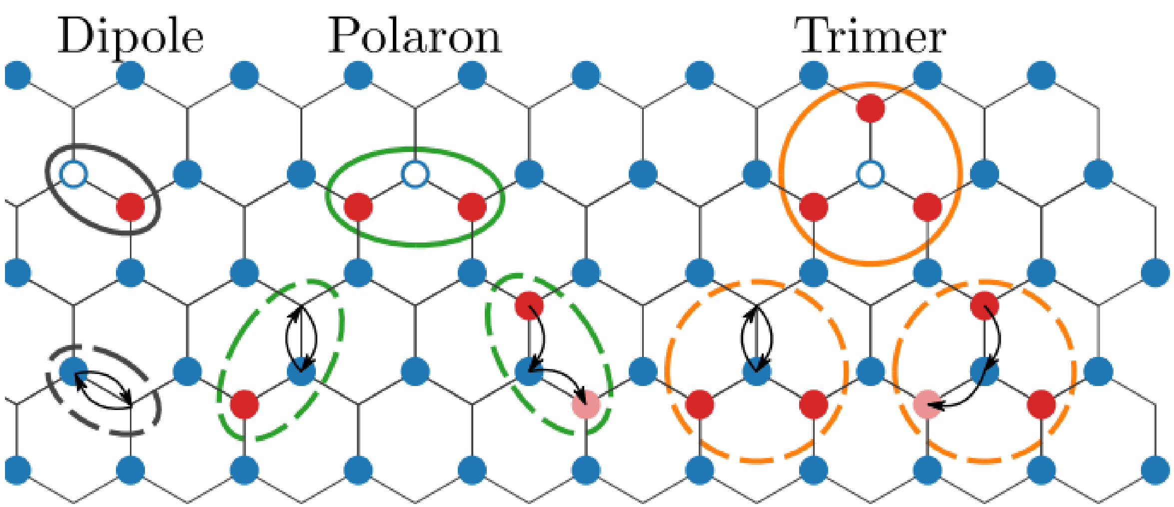
the effective mass at the K point $m = 0.580 m_e$ [51, 52], we estimate $t \sim 0.65\,\mathrm{eV}$. With the parameters $U_B = 4V_0 = 4\Delta$ of Fig. 4, we obtain a critical temperature $T_c \sim 0.002t \simeq 15\,\mathrm{K}$ for 9% doping, which lies very close to the experimentally measured value.

Prediction: ZrNCl at low electron doping is a f-wave spin-triplet SC.

Strong-Coupling Expansion $t \ll \Delta$

$$\mathcal{H}_0 = V \sum_{\langle r,r' \rangle} n_r n_{r'} + \Delta \sum_{r \in B} n_r, \qquad \mathcal{H}_t = -t \sum_{\langle r,r' \rangle} (c_r^\dagger c_{r'} + hc).$$

- hopping of electron from A to B site increases energy.
- low-energy physics governed by second-order processes



- intermediate states: dipoles, polarons and **trimers** cost energies $E_d = \Delta + 2V$, $E_p = \Delta + V$, $\boldsymbol{E_t = \Delta}$ respectively.

Equal-Spin Pairing

$\psi(x,y)c_{x\uparrow}^{+}c_{y\uparrow}^{+}$ with $\psi(x,y) = -\psi(y,x)$

Search for Spin-Triplet Superconductor

Lesson from Sr_2RuO_4 :

Letter | Published: 17 December 1998

Spin-triplet superconductivity in Sr_2RuO_4 identified by ^{17}O Knight shift

Letter | Published: 23 September 2019

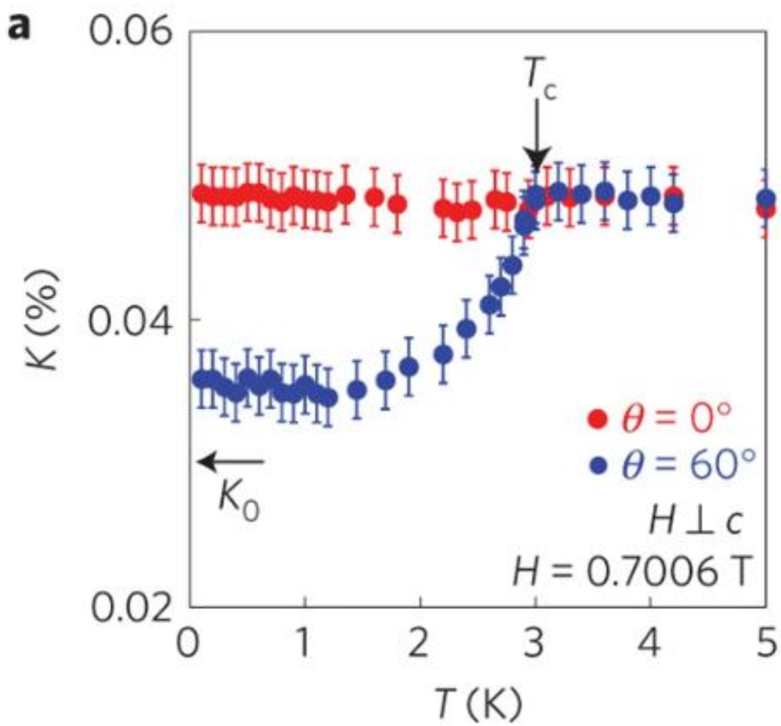
Constraints on the superconducting order parameter in Sr_2RuO_4 from oxygen-17 nuclear magnetic resonance

Perspective | Published: 11 November 2024

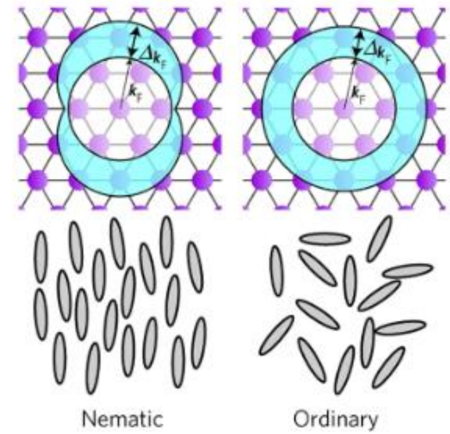
Thirty years of puzzling superconductivity in Sr_2RuO_4

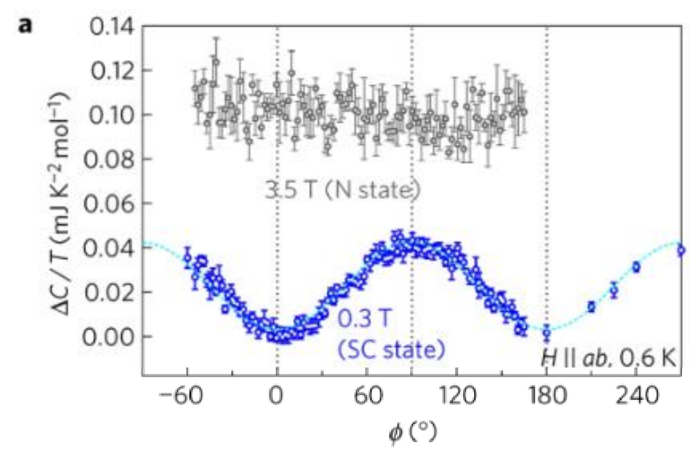
Spin-Triplet Nematic Superconductor

$Cu_xBi_2Se_3$



Matano … Zheng, Nat. Phys. (2016)





Yonezawa … Maeno, Nat. Phys. (2017)

Spin-Polarized Superconductors

- Full spin polarization (lecture 1 & 2)
  - rhombohedral graphene
  - pairing mechanism
  - topology & Majorana

- Partial spin polarization (lecture 3)
  - finite momentum pairing
  - beyond pairing

Unconventional Superconductivity

PHYSICAL REVIEW B **81**, 224505 (2010)

Superconductivity in the repulsive Hubbard model: An asymptotically exact weak-coupling solution

S. Raghu,[1] S. A. Kivelson,[1] and D. J. Scalapino[1,2]

PHYSICAL REVIEW B **85**, 024516 (2012)

Effects of longer-range interactions on unconventional superconductivity

S. Raghu,[1,2] E. Berg,[3] A. V. Chubukov,[4] and S. A. Kivelson[1]

Electronic & Superconducting Liquid Crystals

VOLUME 85, NUMBER 10 PHYSICAL REVIEW LETTERS 4 SEPTEMBER 2000

Quantum Theory of the Smectic Metal State in Stripe Phases

V. J. Emery,[1] E. Fradkin,[2] S. A. Kivelson,[3,4] and T. C. Lubensky[5]

Article | Published: 13 September 2009

Charge-4*e* superconductivity from pair-density-wave order in certain high-temperature superconductors

Erez Berg, Eduardo Fradkin & Steven A. Kivelson

Chiral Superconductivity, Pair Density Wave and Majorana Fermions in Rhombohedral Graphene

Liang Fu

SIMONS FOUNDATION

Collaboration

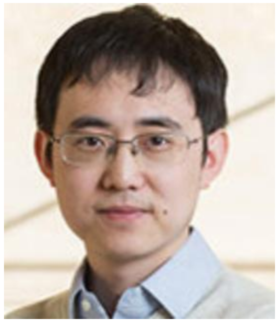
Max Geier

Margarita Davydova
(=>Caltech)

Filippo Gaggioli

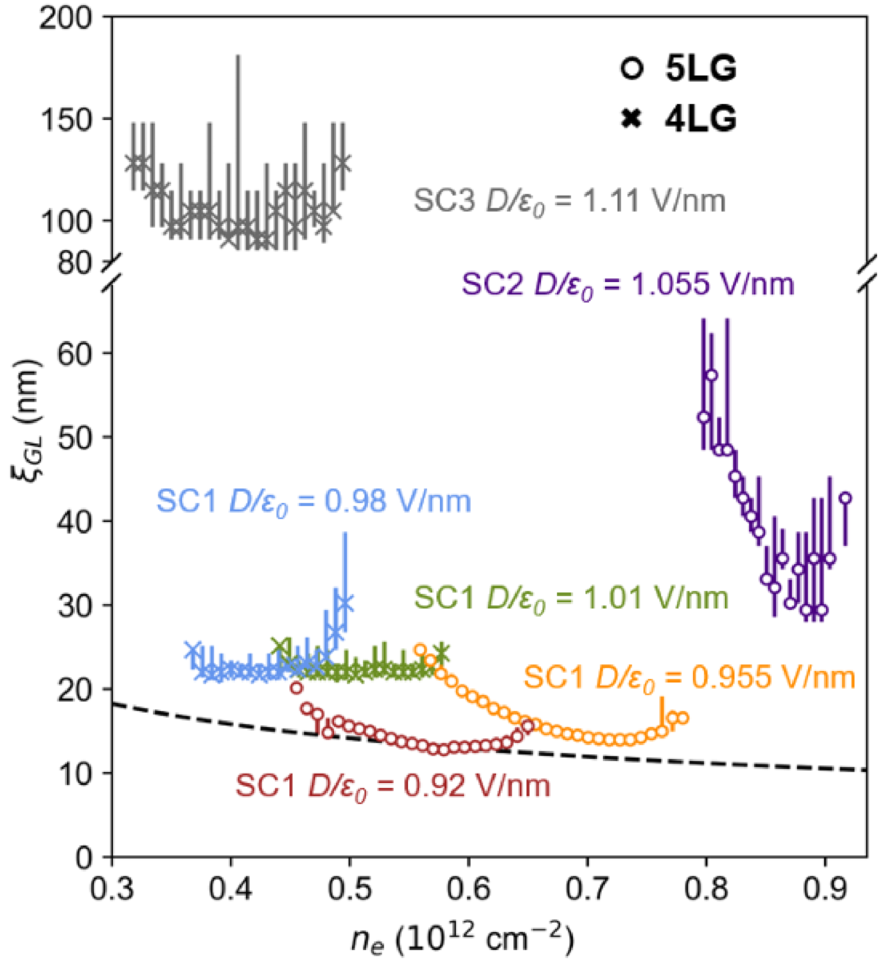
Daniele Guerci

Experiment:



Long Ju

Tonghang Han



5LG
4LG
SC3 D/ε_0 = 1.11 V/nm
SC2 D/ε_0 = 1.055 V/nm
SC1 D/ε_0 = 0.98 V/nm
SC1 D/ε_0 = 1.01 V/nm
SC1 D/ε_0 = 0.955 V/nm
SC1 D/ε_0 = 0.92 V/nm
ξ_{GL} (nm)
n_e (10^{12} cm^{-2})
200
150
100
80
60
50
40
30
20
10
0
0.3
0.4
0.5
0.6
0.7
0.8
0.9

Pairing from Repulsion

Electronic Correlation Effects and Superconductivity in Doped Fullerenes

SUDIP CHAKRAVARTY, MARTIN P. GELFAND, STEVEN KIVELSON

Pair binding in small Hubbard-model molecules

Steven R. White
Department of Physics, University of California at Irvine, Irvine, California 92717

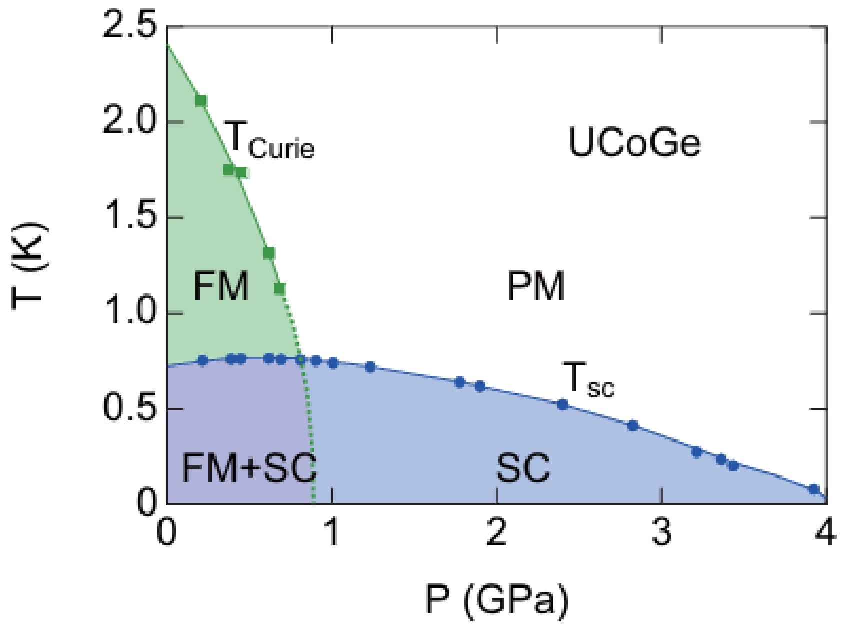
Sudip Chakravarty, Martin P. Gelfand, and Steven A. Kivelson
Department of Physics, University of California at Los Angeles, Los Angeles, California 90024

Optimal inhomogeneity for superconductivity: Finite-size studies

Wei-Feng Tsai,[1,2] Hong Yao,[2] Andreas Läuchli,[3] and Steven A. Kivelson[2]

Pair Binding Energy: $\Delta = 2E_0(N+1) - E_0(N+2) - E_0(N)$

Spin-Polarized Superconductivity



Pairing Competes with Magnetization

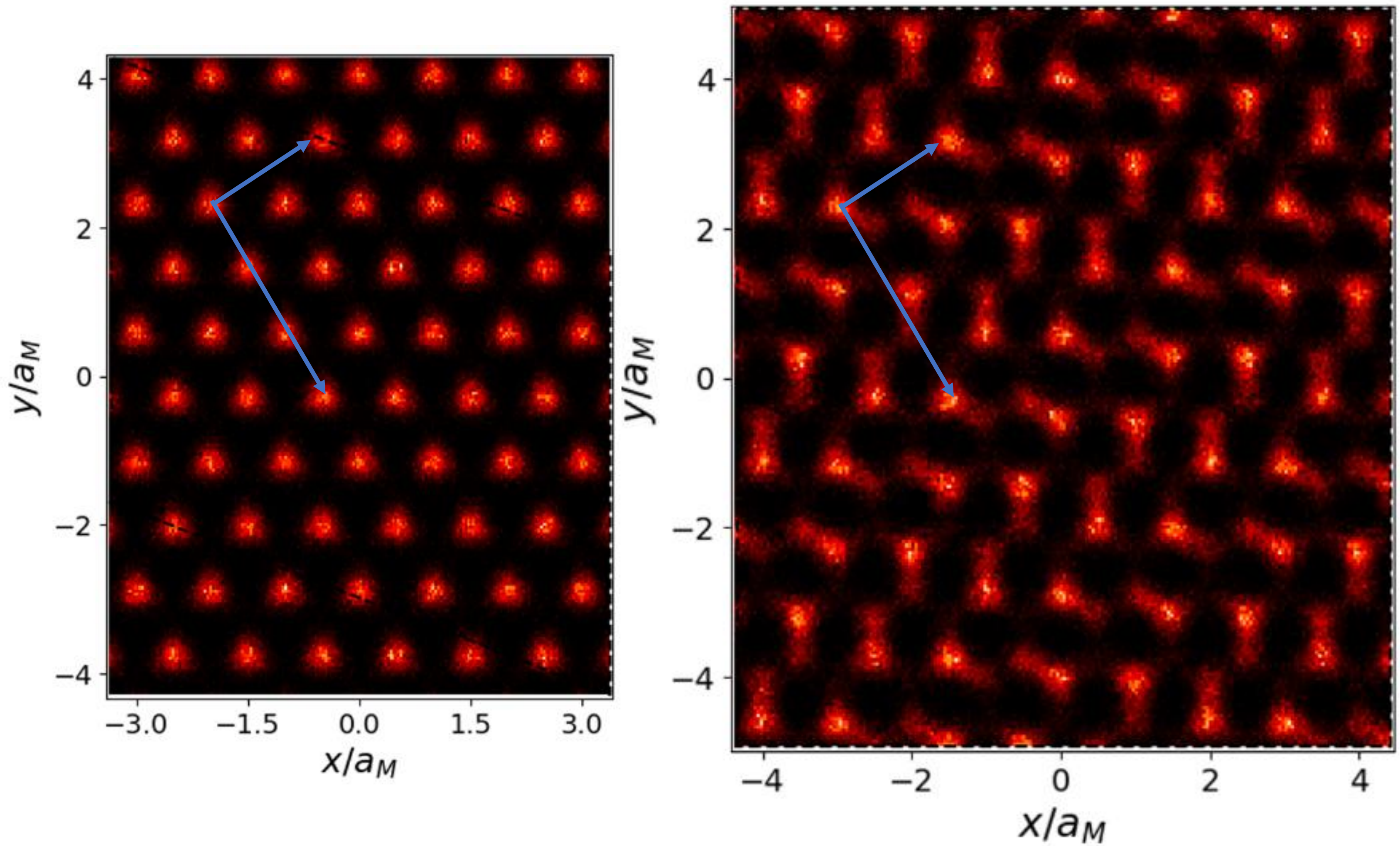
Spin-singlet superconductors have zero total spin and exhibit a finite energy gap to spin-flip excitations. Increasing Zeeman field drives a superconductor-to-normal transition at $\mu_0 B_P \approx 1.86 T_c$.

Spin-Polarized Superconductivity

- Topology & Majorana Fermion
- Rhombohedral Graphene
- Magnonic Cooper pair

AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
UNITED STATES AIR FORCE
SIMONS FOUNDATION

SC in Graphene Systems


4
2
0
−2
−4
y/aM
−3.0
−1.5
0.0
1.5
3.0
x/aM
4
2
0
−2
−4
y/aM
−4
−2
0
2
4
x/aM