ECRYS 2022 : International Research School and Workshop on Electronic Crystals

August 8-20, 2022
Carges, France
(Aug 10)

# Density controlled BCS-BEC crossover in 2D superconductors

Yoshi Iwasa

Department of Applied Physics and QPEC, University of Tokyo

RIKEN Center for Emergent Matter Science

# Acknowledgements


**AP, Univ Tokyo**

**Y. Nakagawa**

T. Nojima (Tohoku)

M. Heyl (Humboldt)

K. Adachi (RIKEN)

Y. Kato (UT)

**Samples**

Y. Kasahara (Kyoto)

**DFT calculation**

R. Arita (Tokyo)

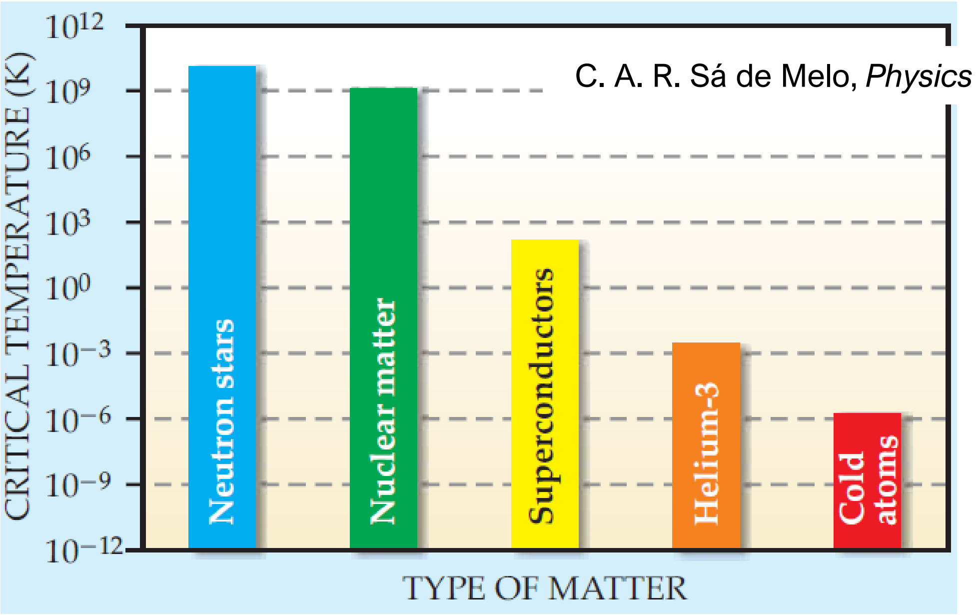
T. Nomoto (Tokyo)

# Content

1. Introduction

2. 2D BCS-BEC crossover in ion gated LixZrNCl

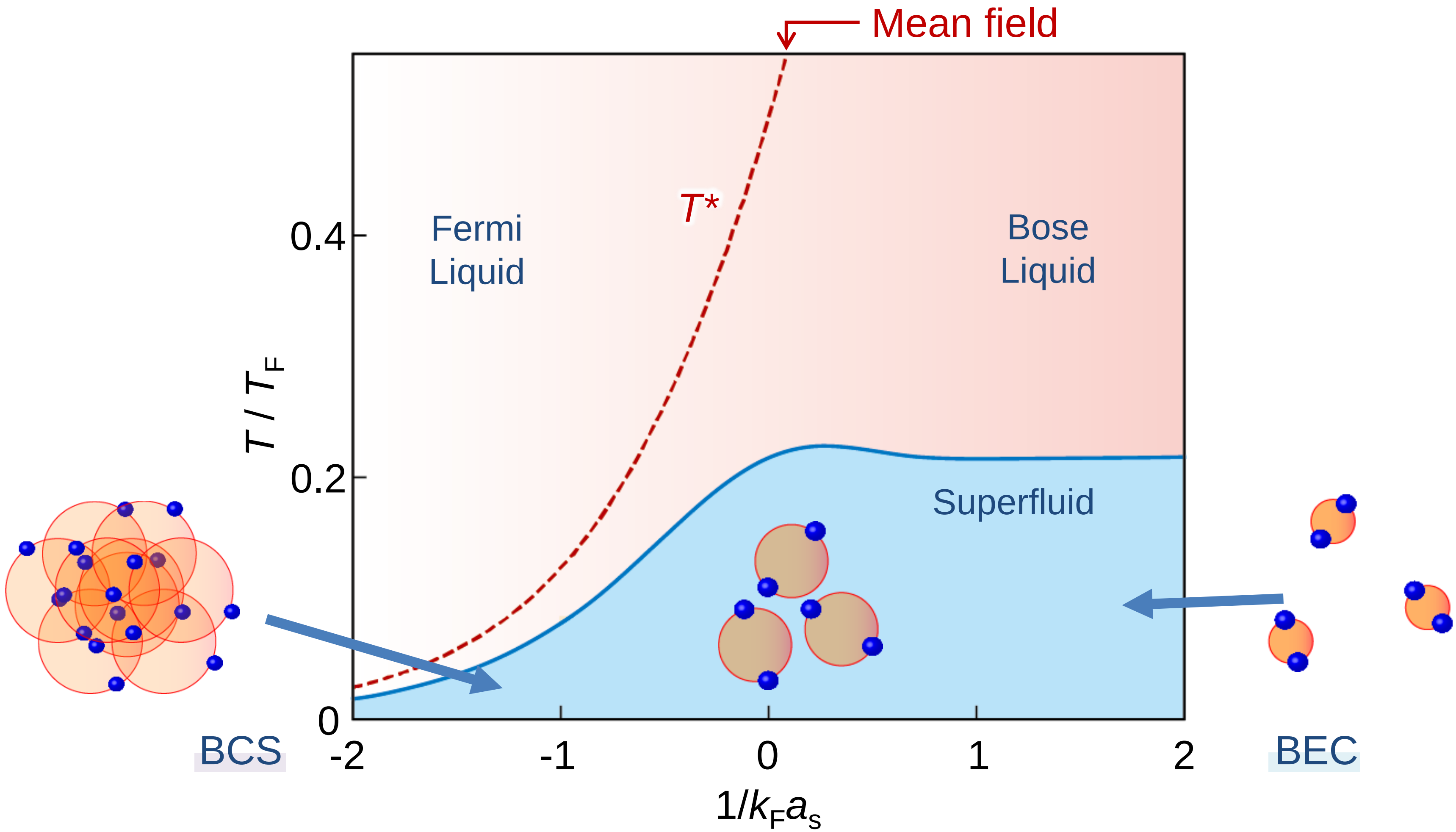
3. Vortex dynamics across the crossover




C. A. R. Sá de Melo, *Physics Today* **61**, 45 (2008).

# BCS-BEC crossover

## A theoretical phase diagram connecting BCS and BEC



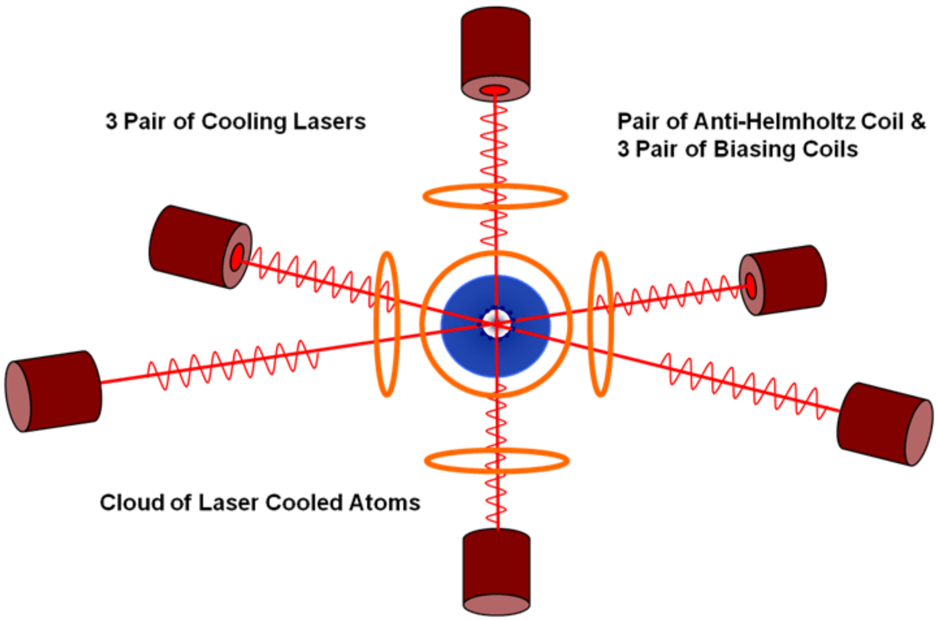

($a_s$: interaction strength), ($k_F$: Fermi wave number, carrier density)

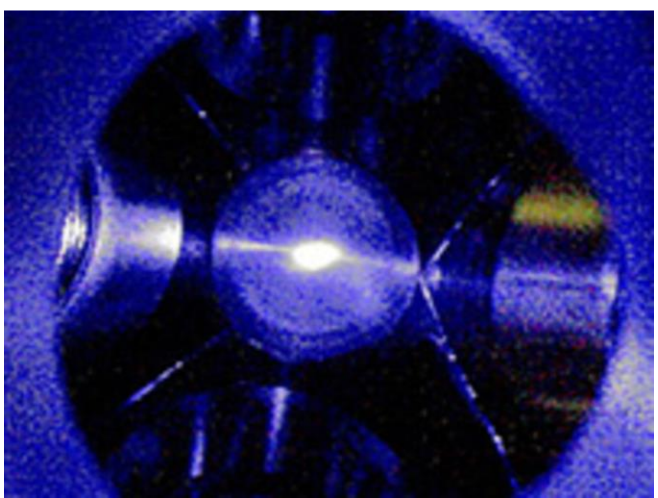

C. A. R. Sá de Melo, M. Randeria, and J. R. Engelbrecht, *Phys. Rev. Lett.* **71**, 3202 (1993).
M. Randeria and E. Taylor, *Annu. Rev. Condens. Matter Phys.* **5**, 209 (2014).

# Ultracold atoms

## Laser cooling of fermion gas

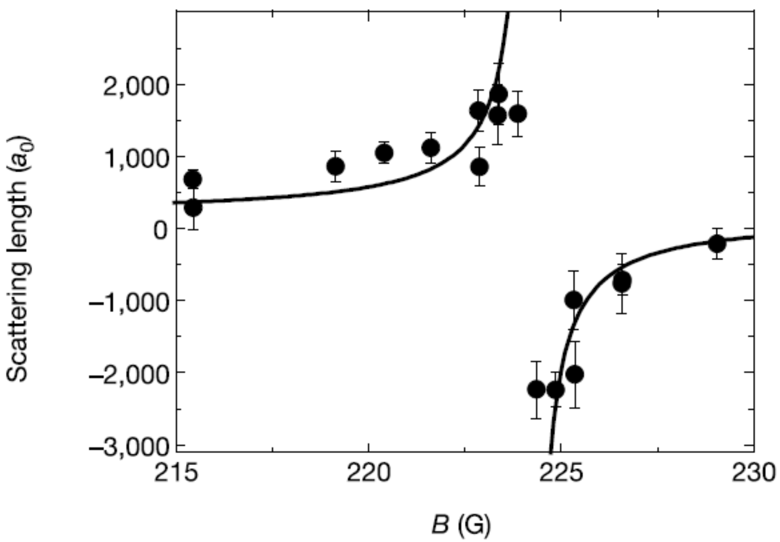
$N = 10^5 \sim 10^8$, $T < 1\ \mu K$



## Feshbach resonance

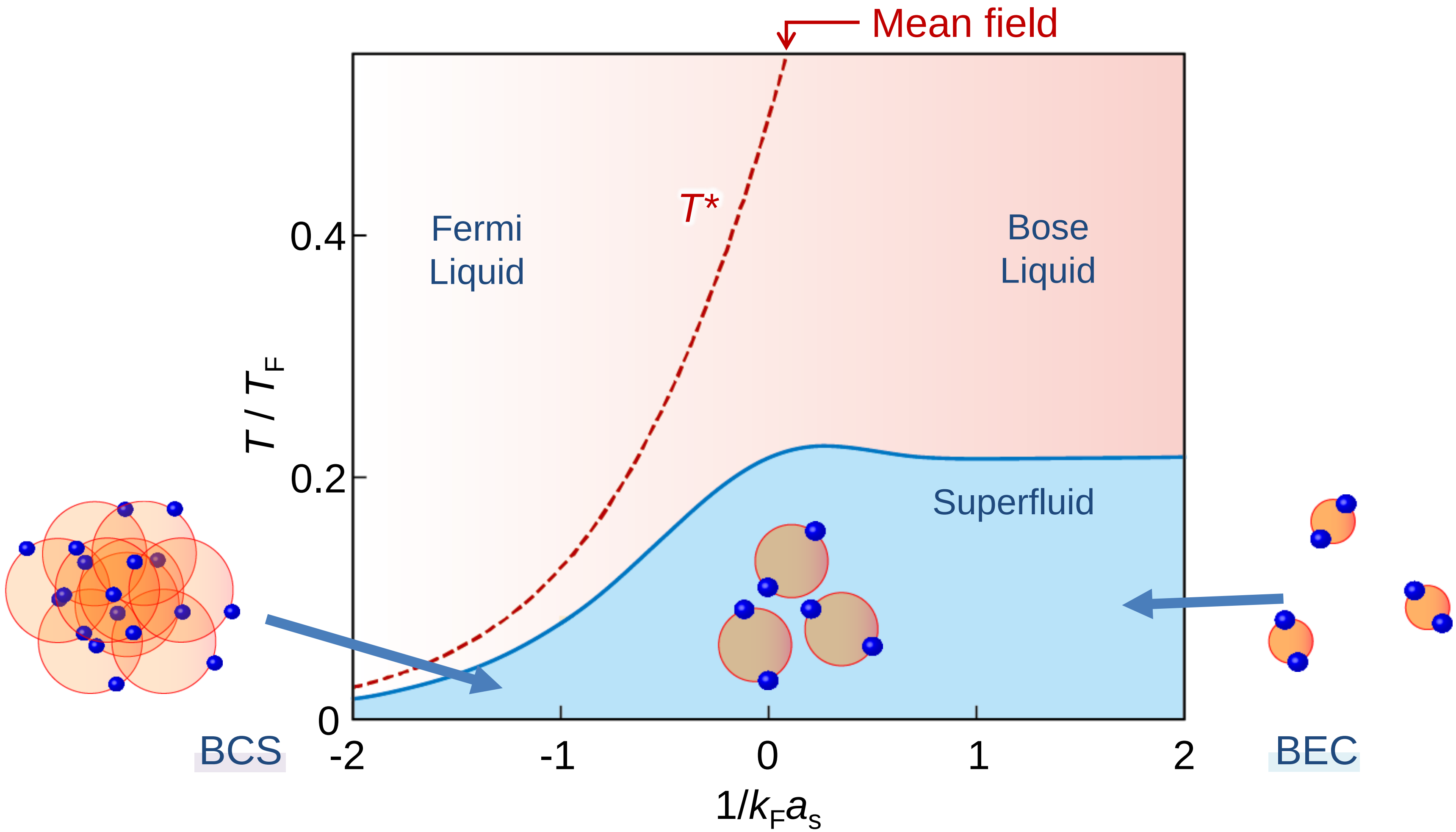
for tuning the interatomic interaction (scattering length).




C. A. Regal *et al.*, *Nature* **424**, 47 (2003).

# BCS-BEC crossover

Can BCS-BEC crossover be induced by density control?



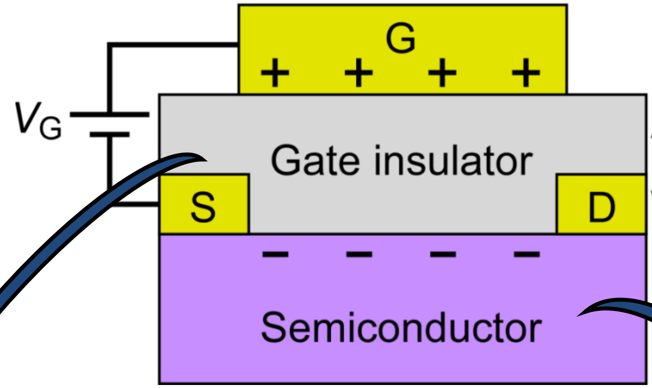

($a_s$: interaction strength), ($k_F$: Fermi wave number, carrier density)


C. A. R. Sá de Melo, M. Randeria, and J. R. Engelbrecht, *Phys. Rev. Lett.* **71**, 3202 (1993).
M. Randeria and E. Taylor,*Annu. Rev. Condens. Matter Phys.* **5**, 209 (2014).

# Density controlled 2D superconductivity: Gating

**FET**

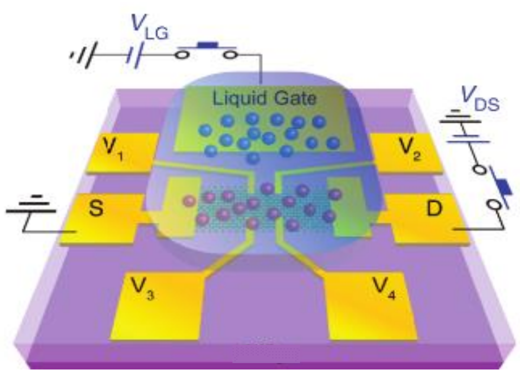
$n = CV_G/e$




**Two routes to gate-induced 2D superconductivity**

## 1. Increase *n*

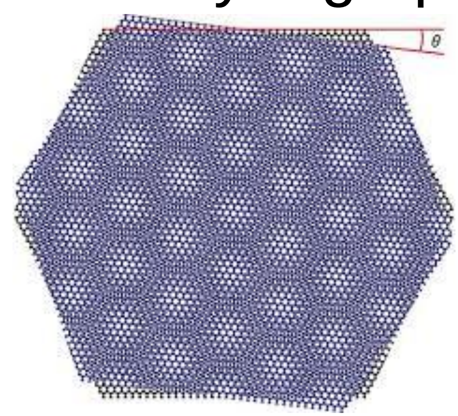
Ionic gating




K. Ueno et al., *Nat Mater*. **7**, 855 (2008)

## 2. Large lattice constant

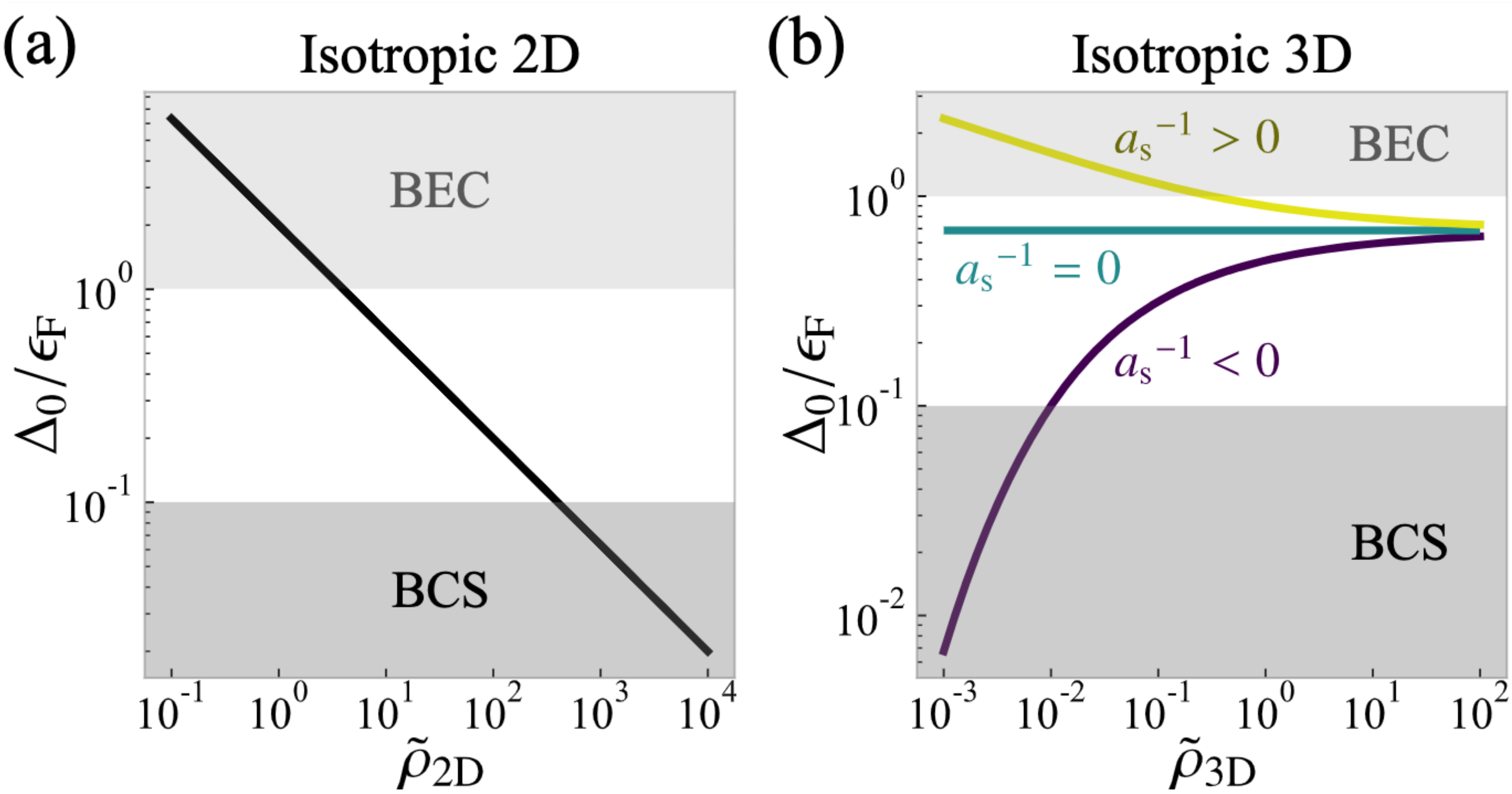
Twisted bilayer graphene




Y. Cao et al., *Nature* **556**, 80 (2018)

# Density induced BCS-BEC crossover: 2D vs 3D

Contact potential model




Courtesy of Kyosuke Adachi (RIKEN)

# Layered nitrides

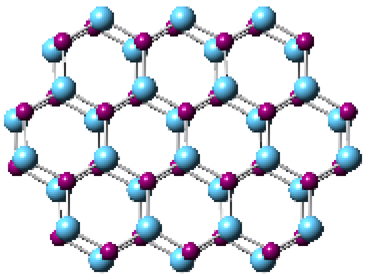
S. Yamanaka (Hiroshima)

## Semiconducting 2D materials with honeycomb structures

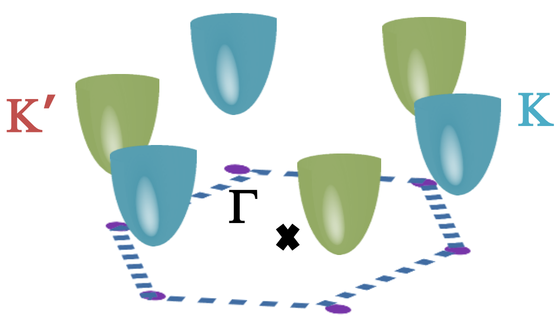
Adv Mater (1996), Nature (1998)

**Li doped ZrNCl (15 K) and HfNCl (25 K)**

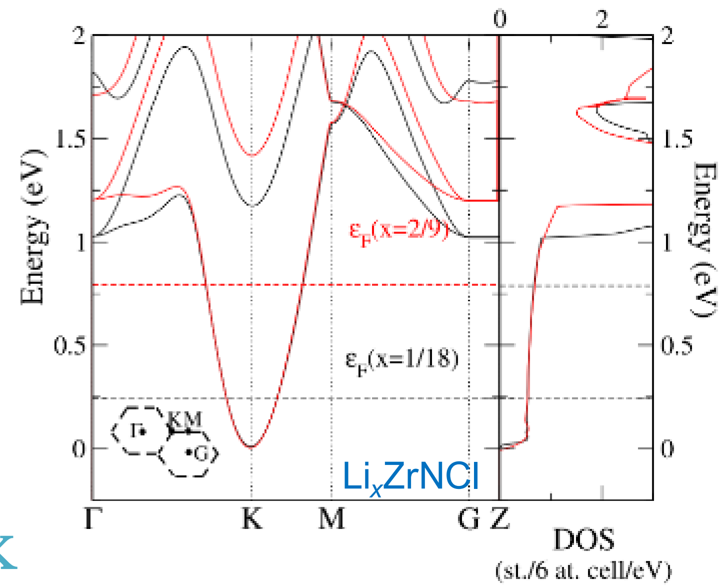
- Double-honeycomb



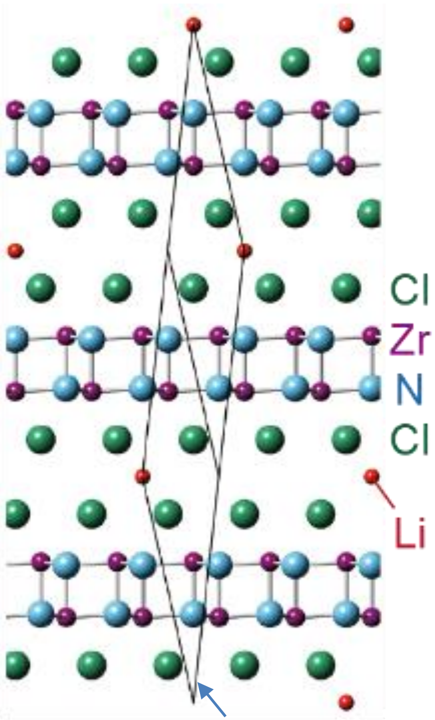
- Degenerate valleys




- Highly 2D electronic structure.



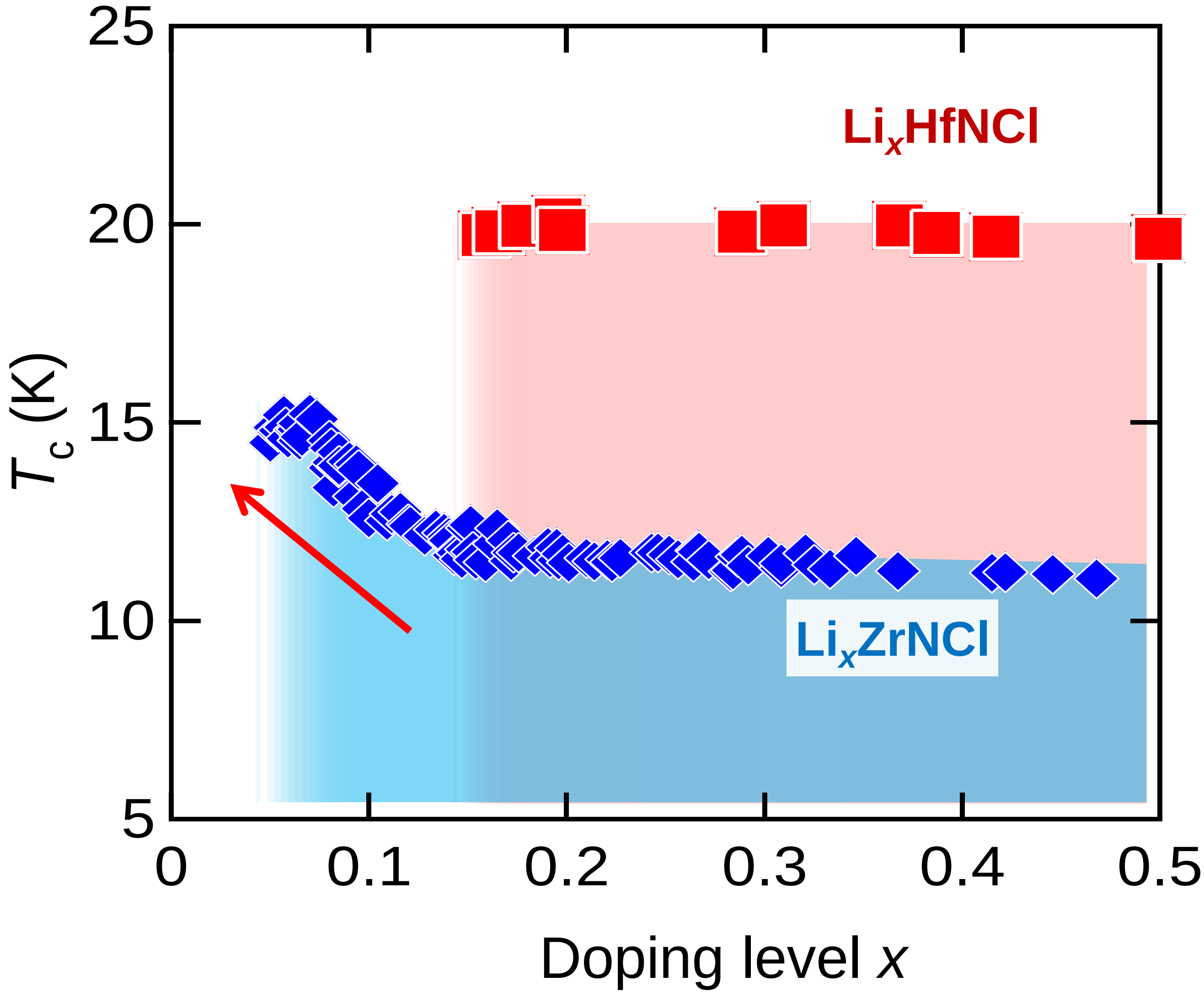

M. Calandra *et al.*, *PRL* **114**, 077001 (2015).




Rhombohedral

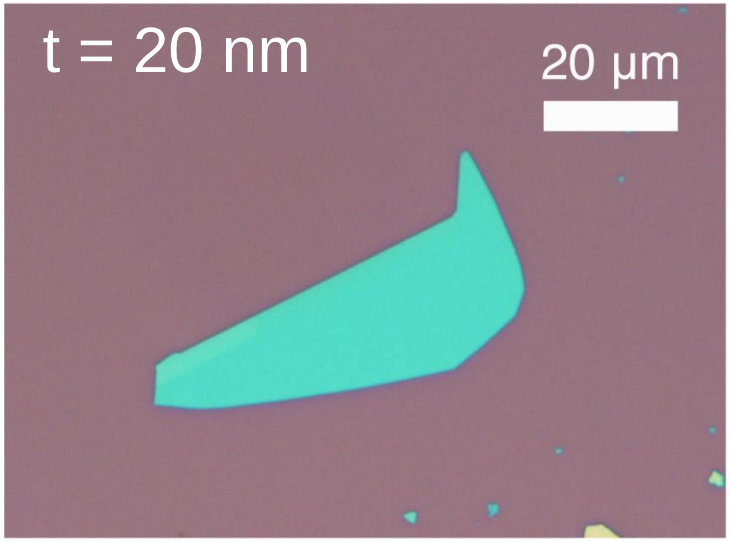
# Bulk property of layered nitrides (**ZrNCl**, **HfNCl**)

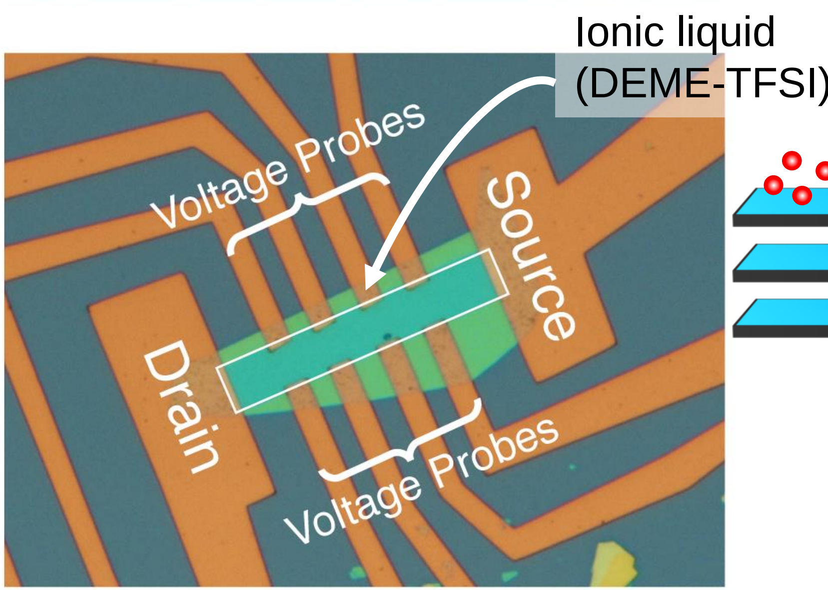
Phase diagram for bulk polycrystals

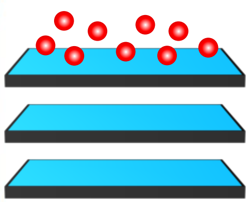


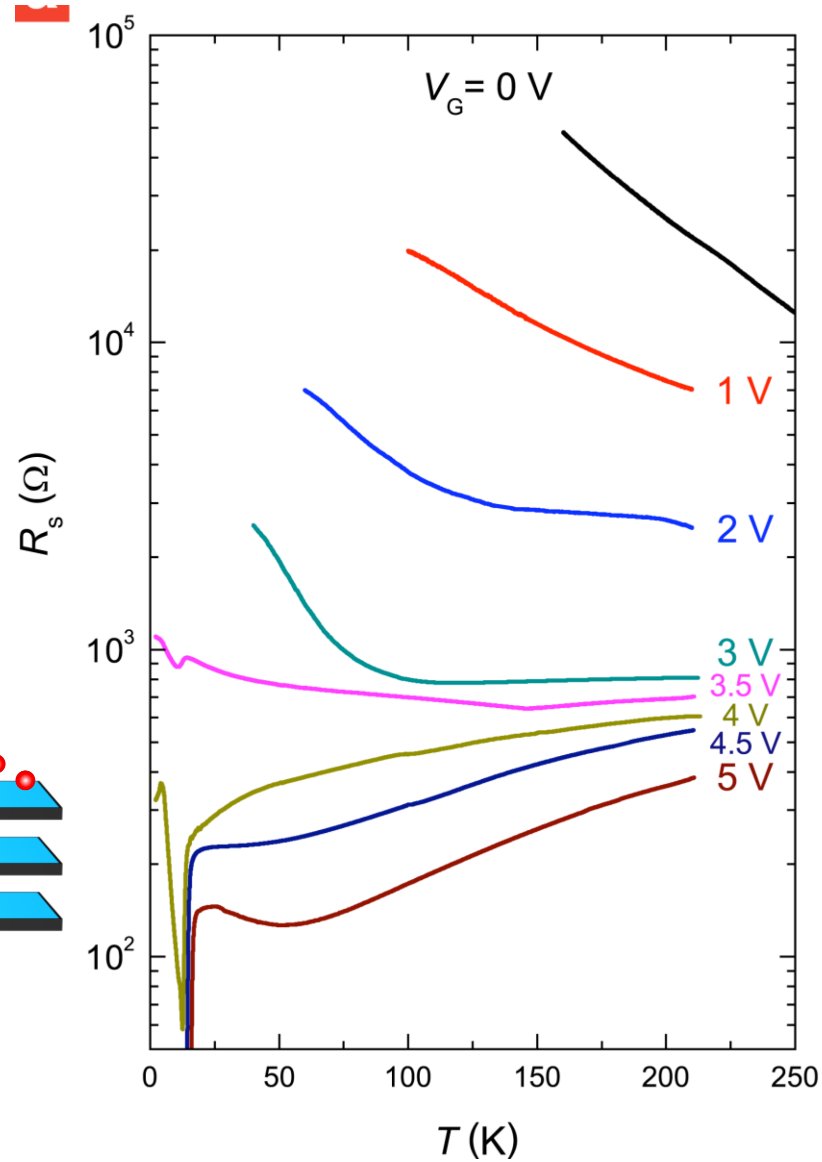

[Zr] S. Yamanaka *et al.*, *Adv. Mat.* **8**, 771 (1996); Y. Taguchi *et al.*, *PRL* **97**, 107001 (2006).
[Hf] S. Yamanaka et a., *Nature* **392**, 580 (1998). T. Takano *et al.*, *PRL* **100**, 247005 (2008).

# Gate-induced superconductivity in cleaved crystal ZrNCl





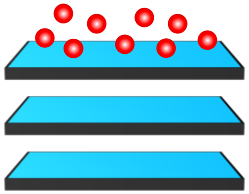

J. T. Ye *et al*. *Nat Mater* **8**, 125 (2010)

# Two modes of ionic gating

## *Electrostatic*

- Electric-double-layer transistor (EDLT)

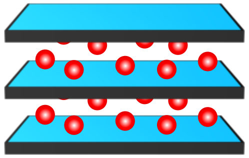
J. T. Ye *et al.*, *Science* **338**, 1193 (2012).
Y. Saito *et al.*, *Science* **350**, 409 (2015).).



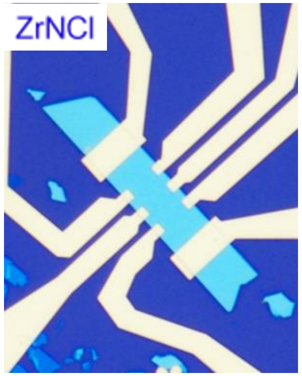
## *Electrochemical*

- Intercalation

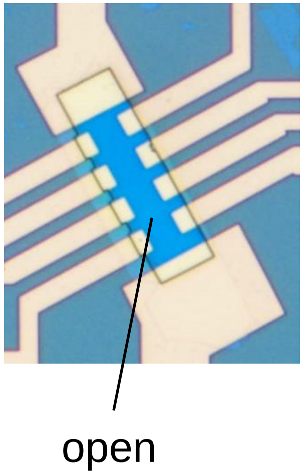
W. Shi *et al.*, *Sci. Rep.* **5**, 12534 (2015).
Y. Yu *et al.*, *Nature. Nano.* ***10***, 270 (2015).



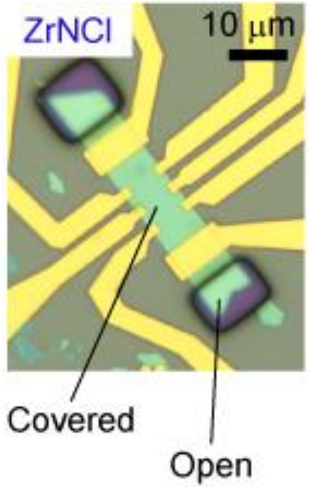

$t$ = 30-70 nm



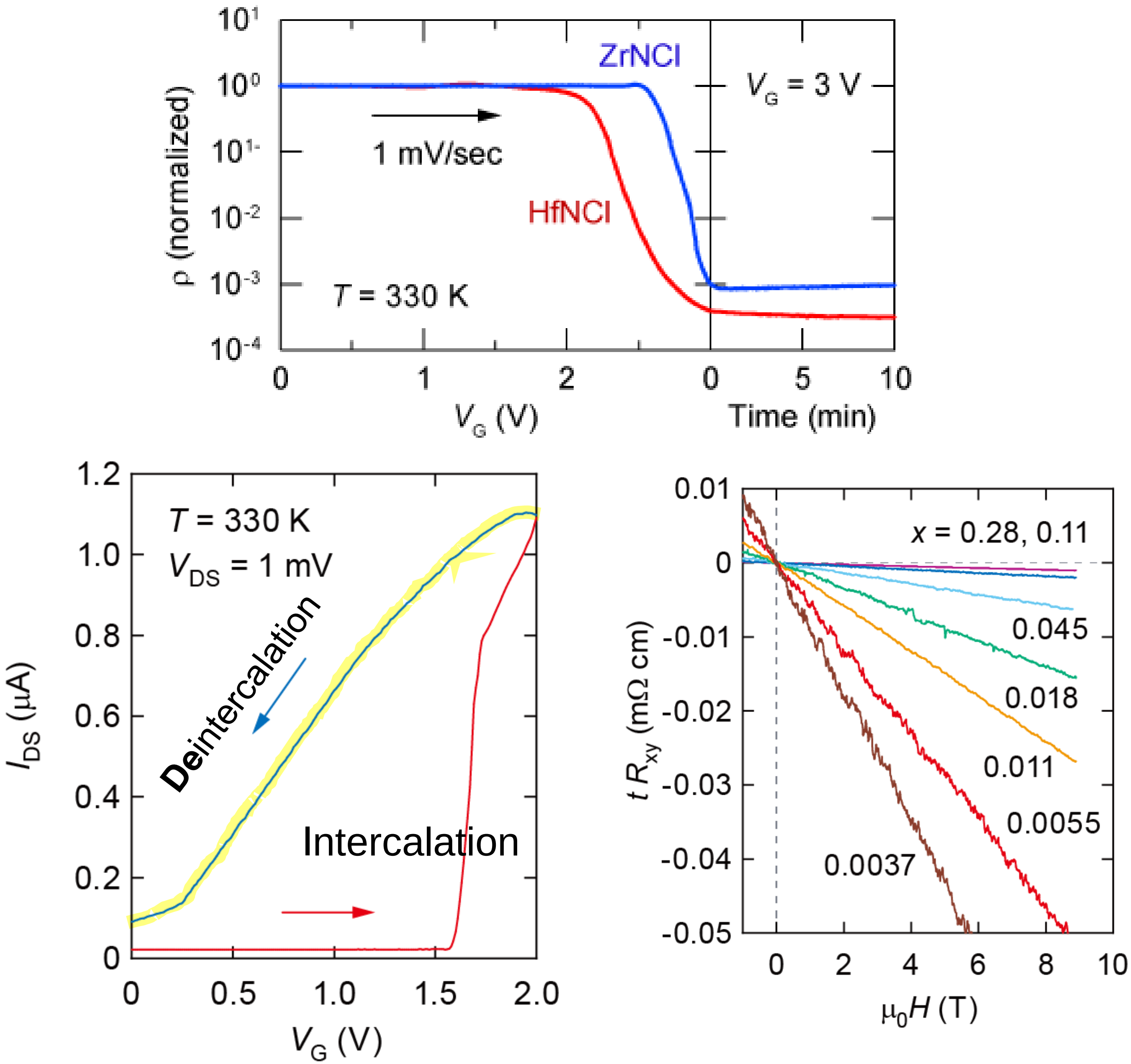

**Electrostatic**



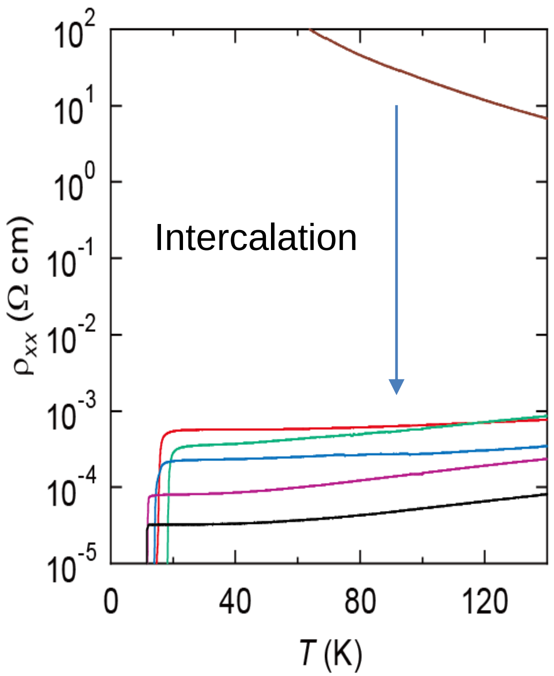

**Intercalation**

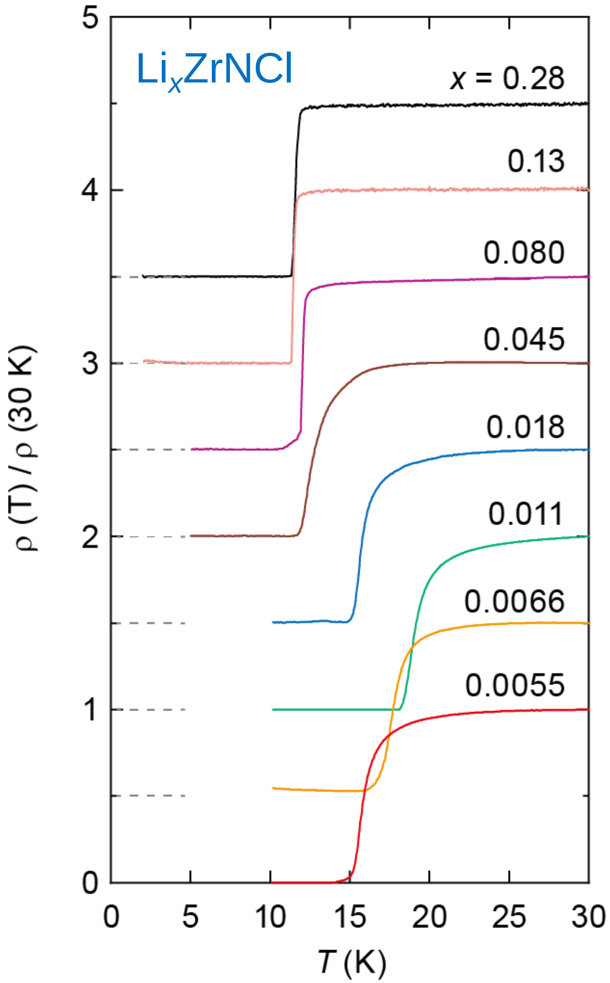


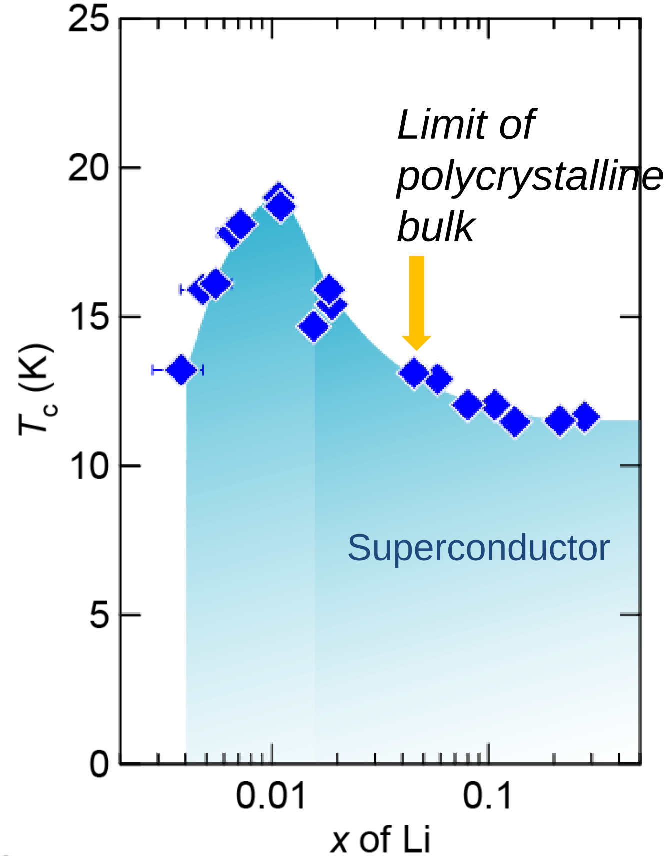
# Gate controlled ntercalation



# Phase diagram – $Li_xZrNCl$



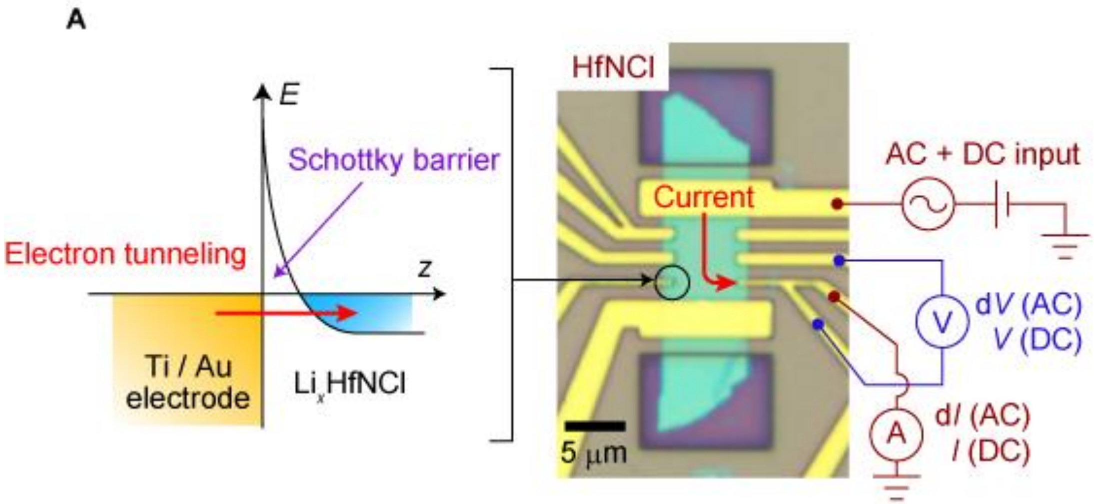



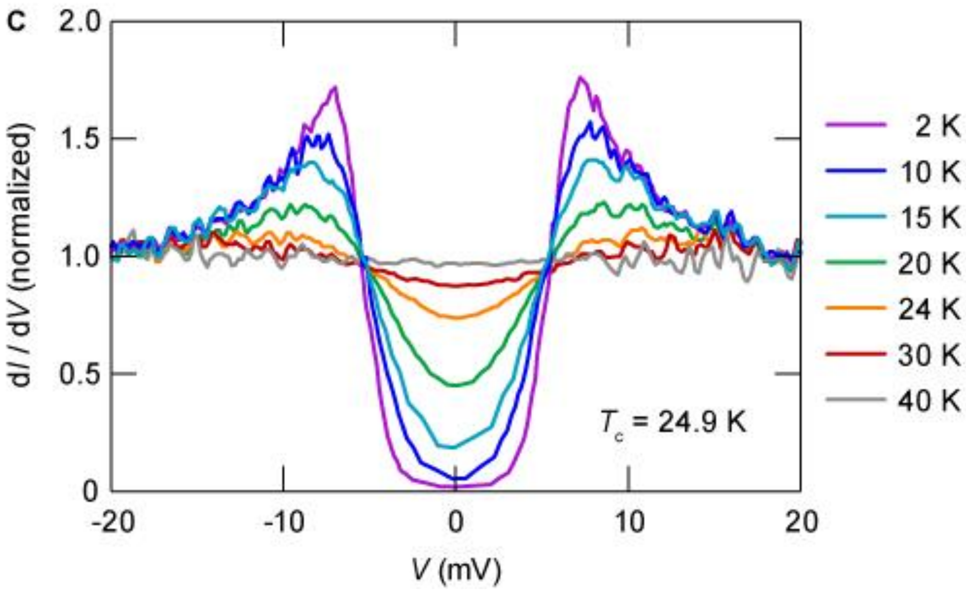




$T_c$ (max) = 19.0 K
at $x$ = 0.011 ($n_{3D} = 2 \times 10^{20}$ cm$^{-3}$)

Y. Nakagawa *et al.*, *Science* **372**, 190 (2021).

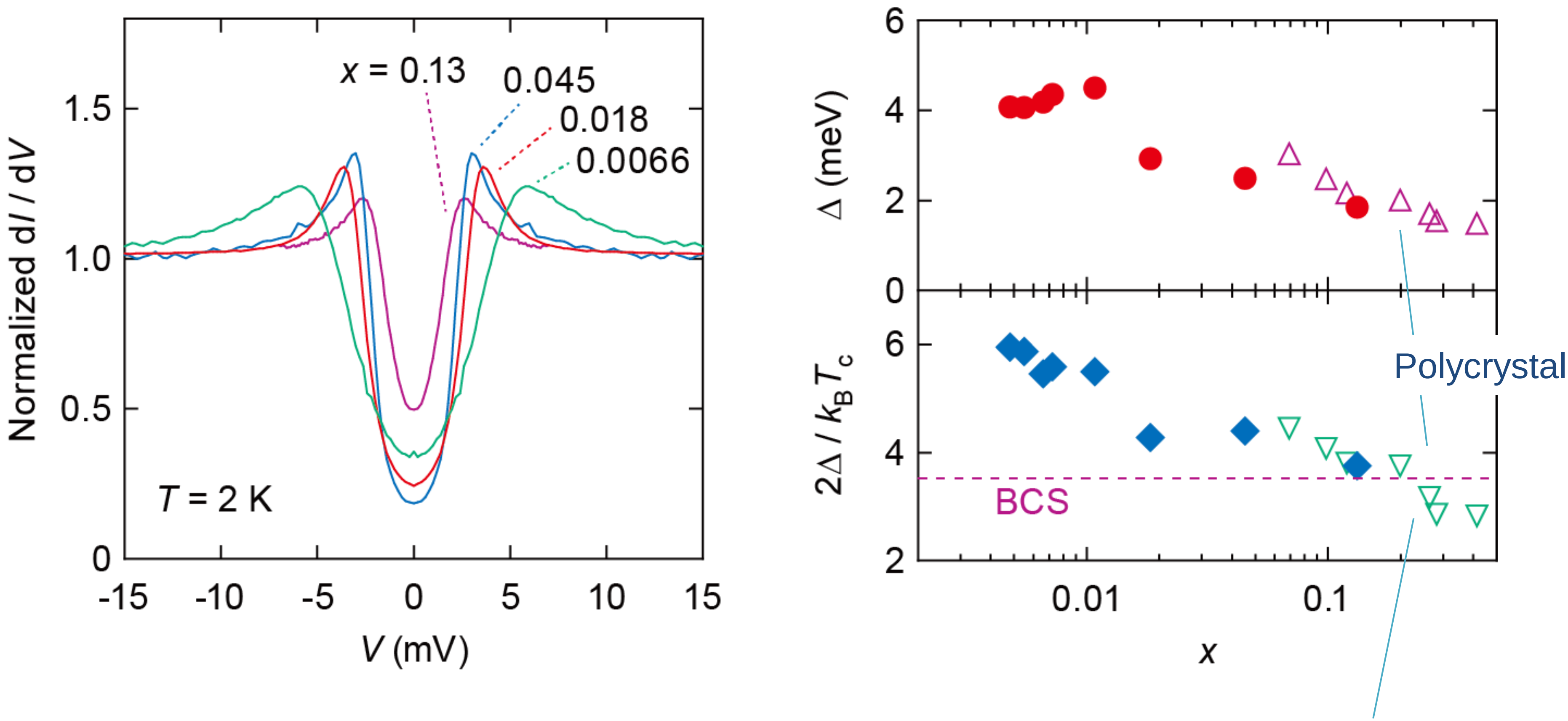
# Tunneling spectroscopy in Li doped HfNCl




Y. Nakagawa *et al.*, *PRB.* **98**, 064512 (2018).

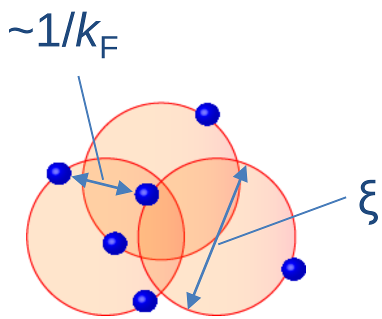
# Tunneling spectroscopy of LixZrNCl

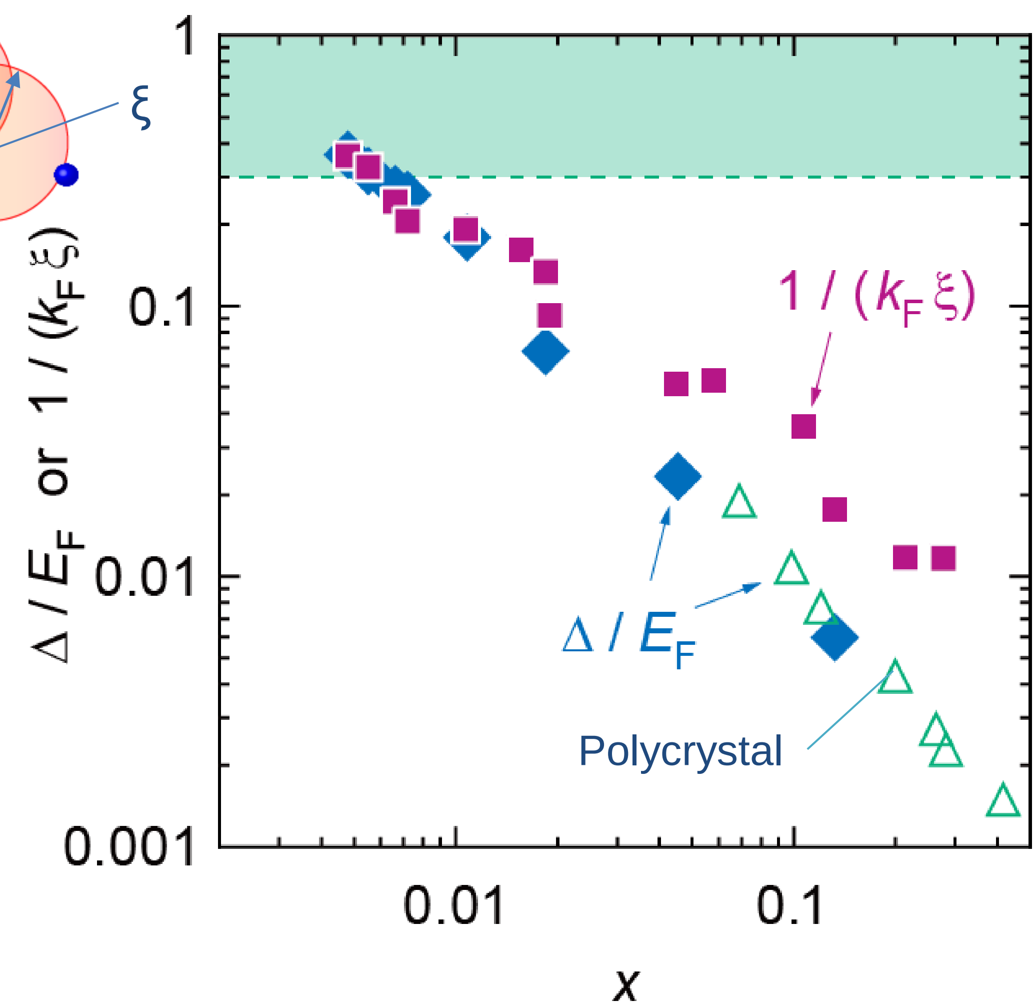


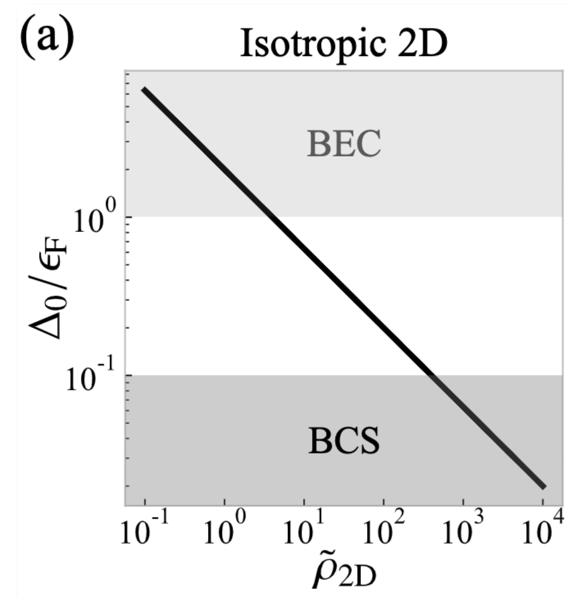

Y. Kasahara et al., PRL103, 077004 (2009).

- SC gap develops with decreasing x.
- Large gap & strong coupling at low-doping regime.

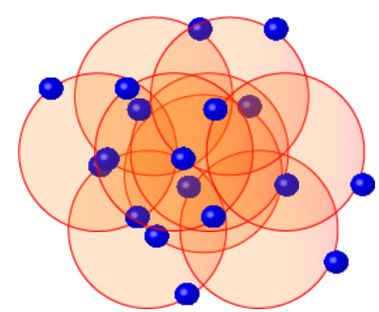
# Approaching BCS-BEC crossover



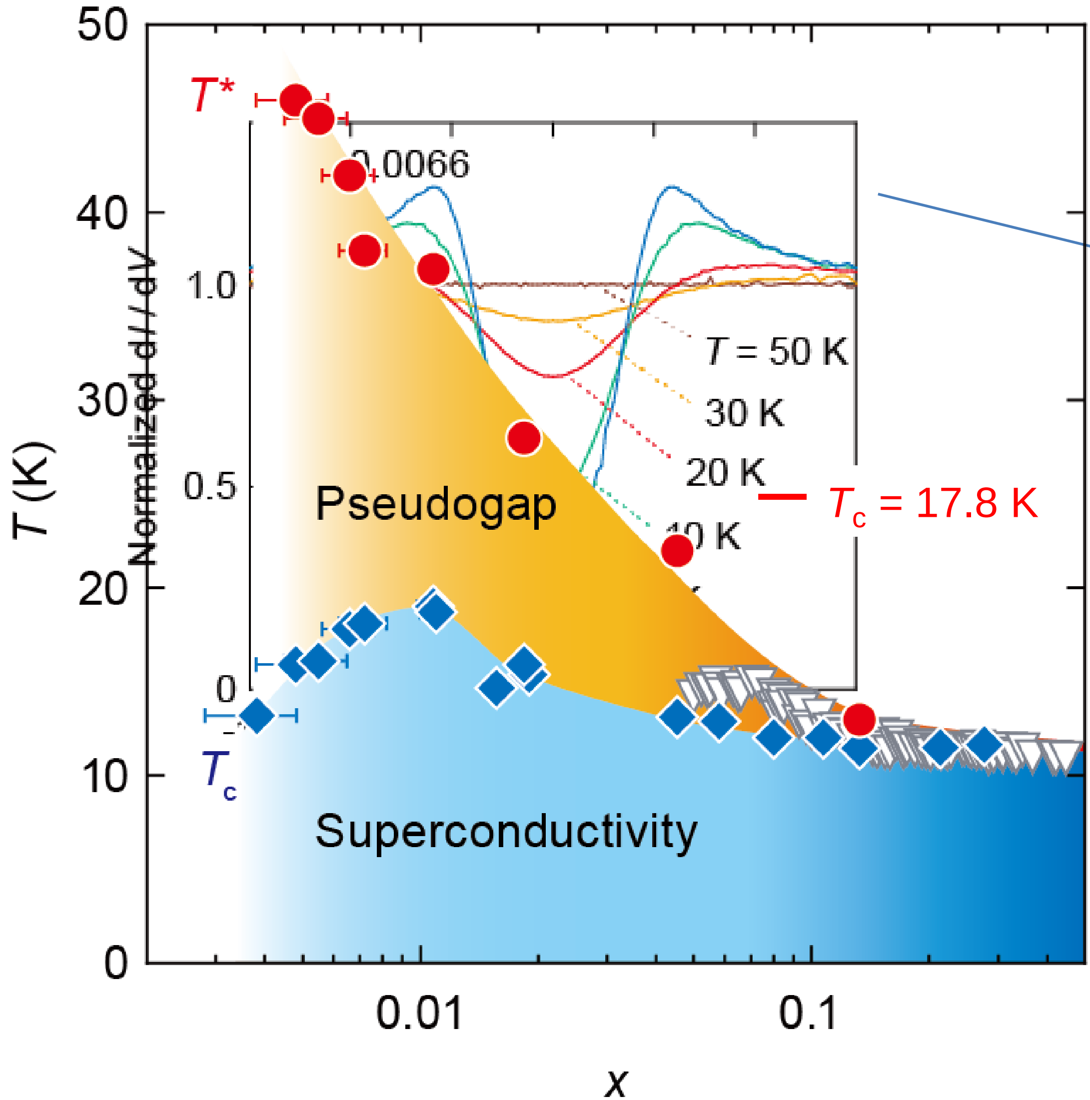



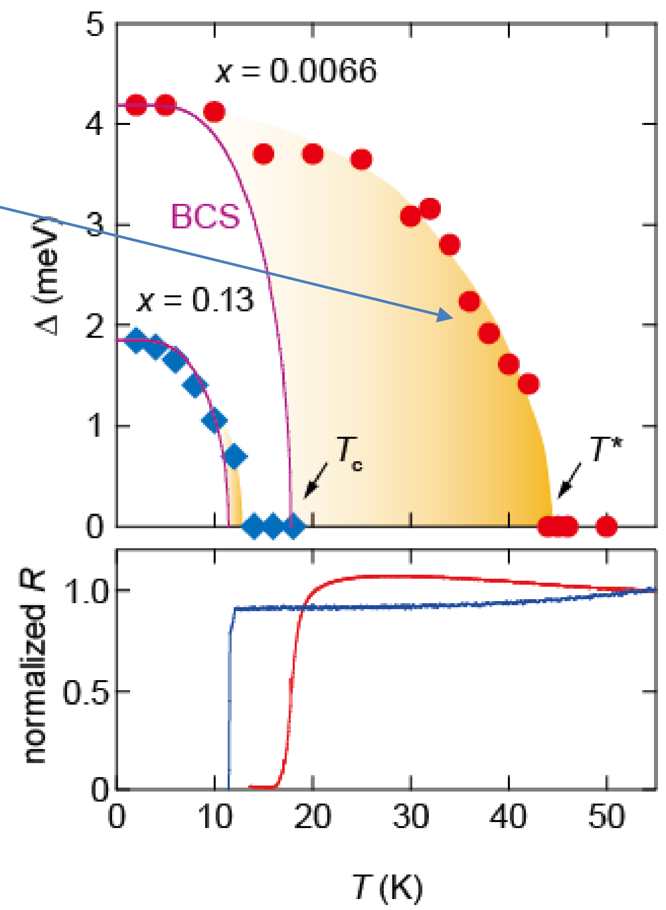




K. Adachi



# Pseudogap state

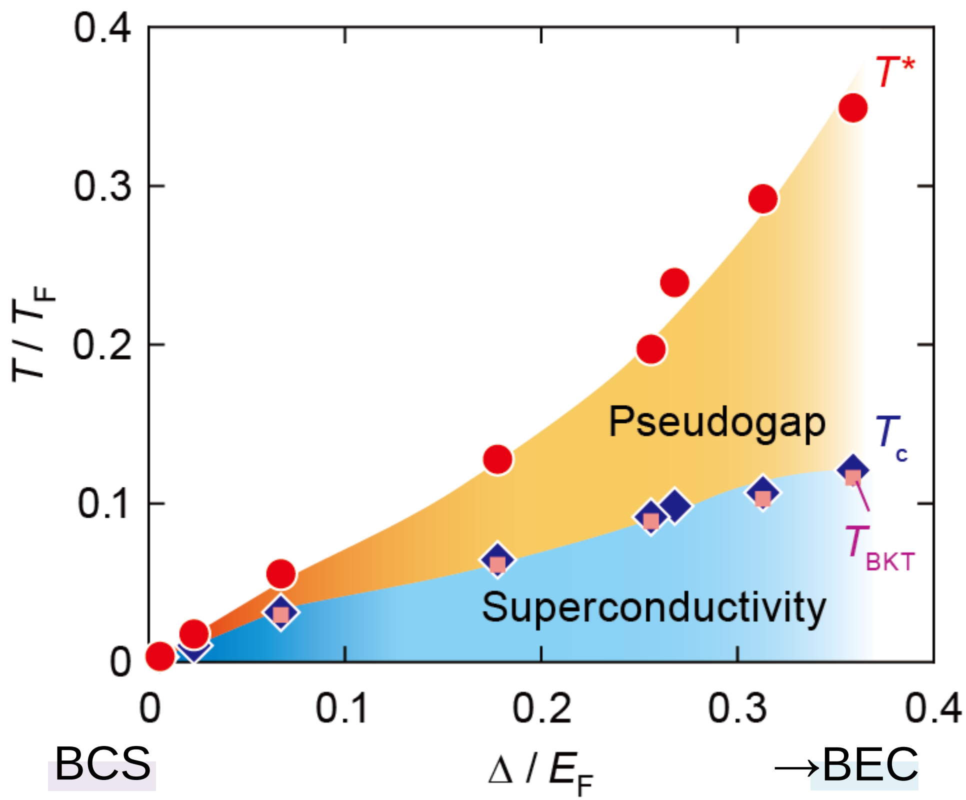


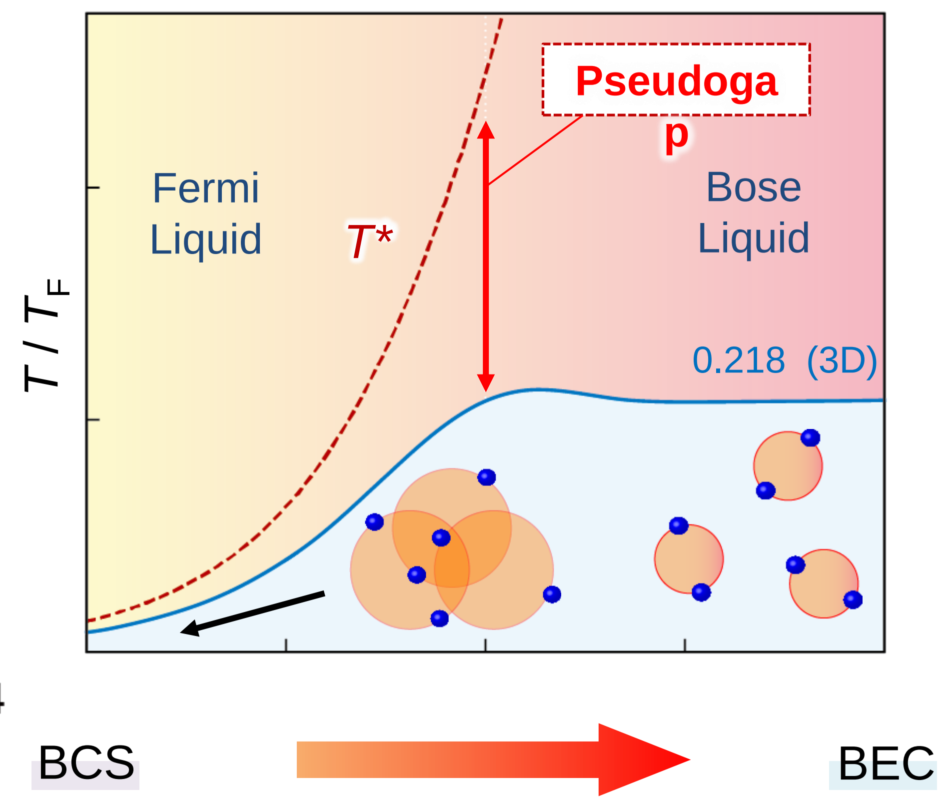

- ➢ Gap opens above $T_c$ at low doping level.
- ➢ Evolution of $T^*$ toward the insulating phase.

Y. Nakagawa *et al.*, Science 372, 190 (2021)

# BCS-BEC crossover



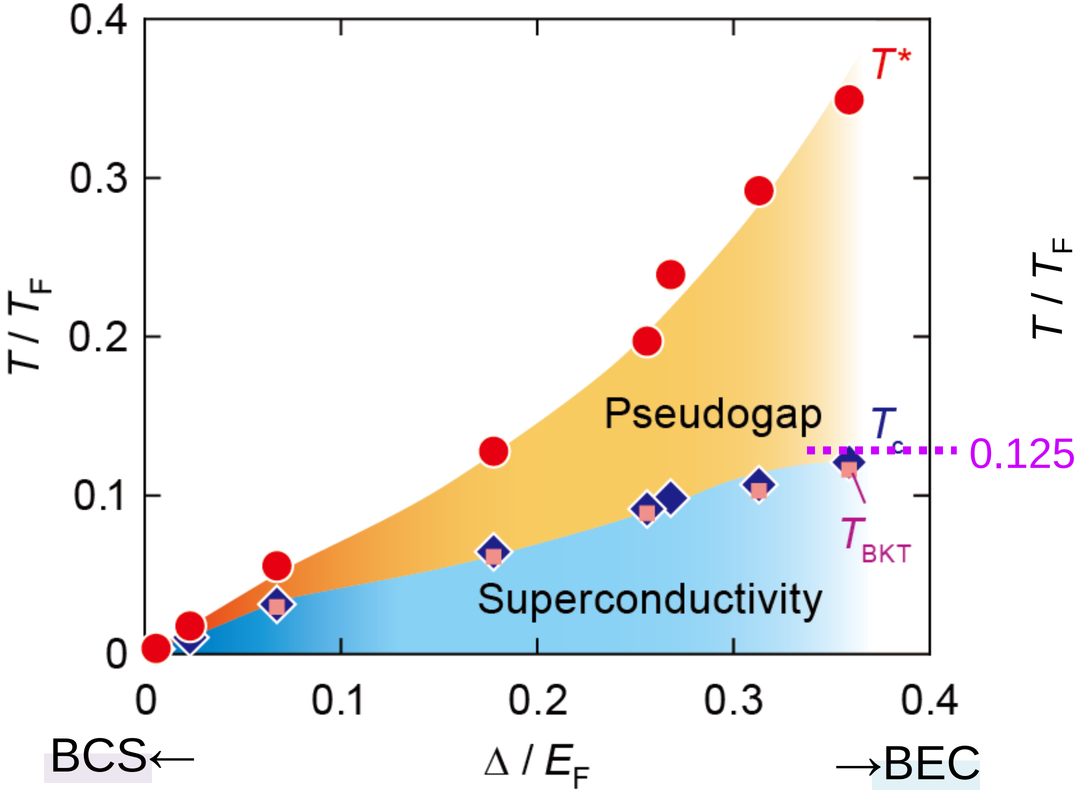

➢ Phase diagram

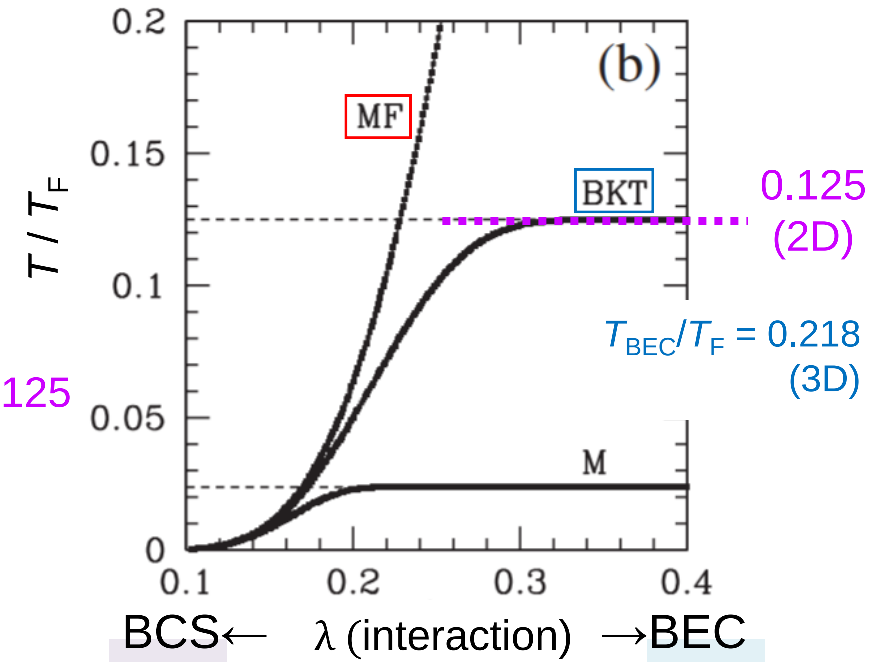



**Low density & Strong coupling**
**~Δ/E F**

➢ Pseudogap state well developed

# Comparison with theory of 2D BCS-BEC crossover




## Benchmark of 2D BCS-BEC

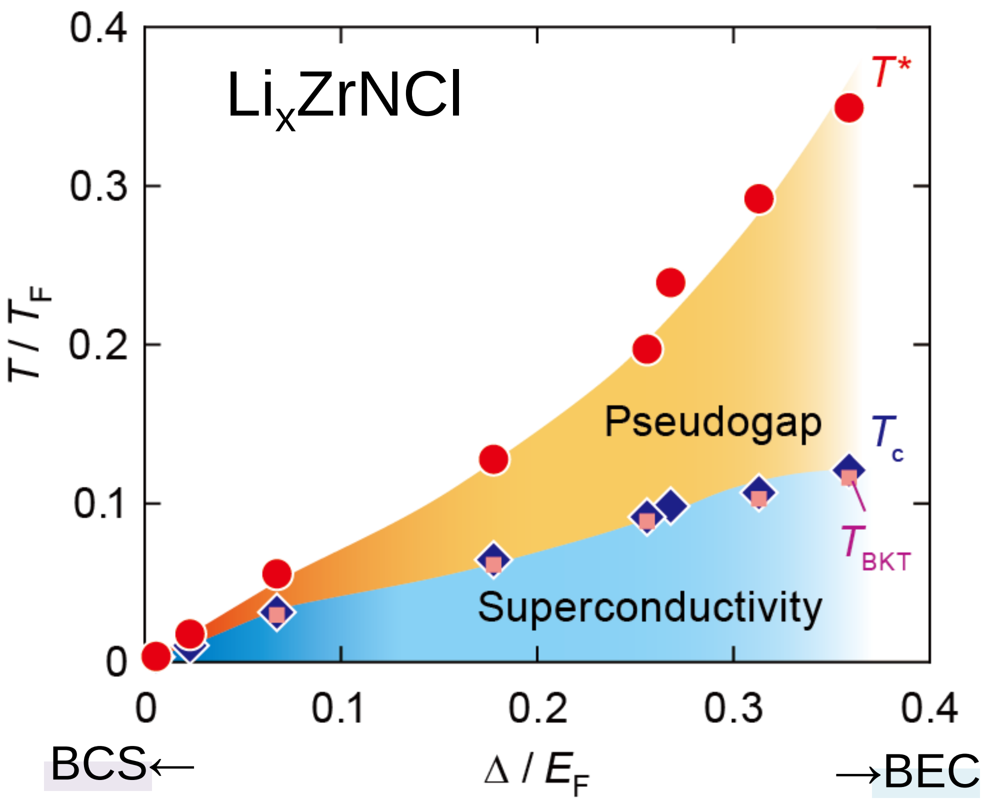
- Pesudogap
- $T_{BKT}/T_F = 1/8$

S. S. Botelho and C. A. R. Sá de Melo, *PRL* **96**, 040404 (2006).

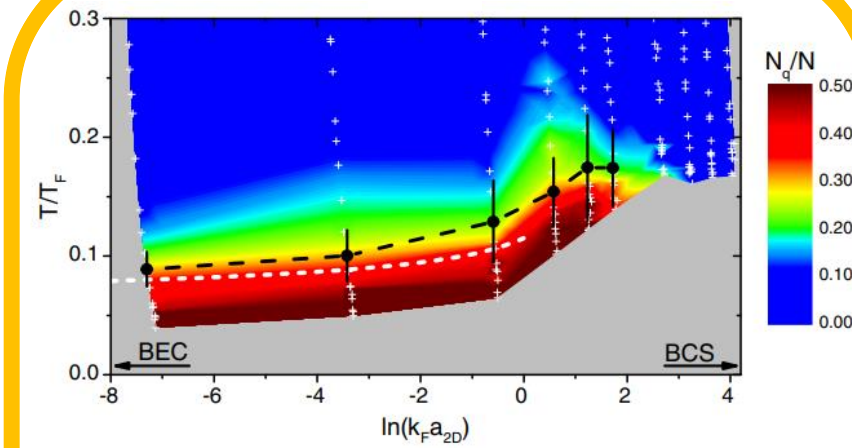
V. P. Gusynin and V. M. Loktev, J. Exp. The. Phys. **88**, 685 (1998)

M. Randeria, Science 372, 132 (2021).

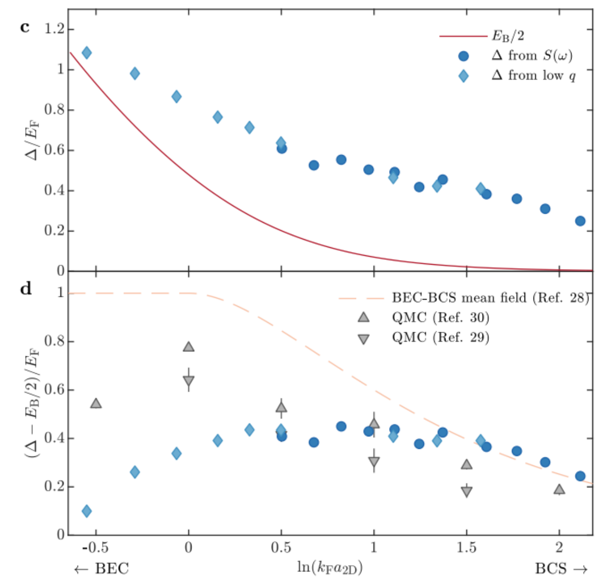
# Comparison with cold gas of $^6Li$




$^6Li$

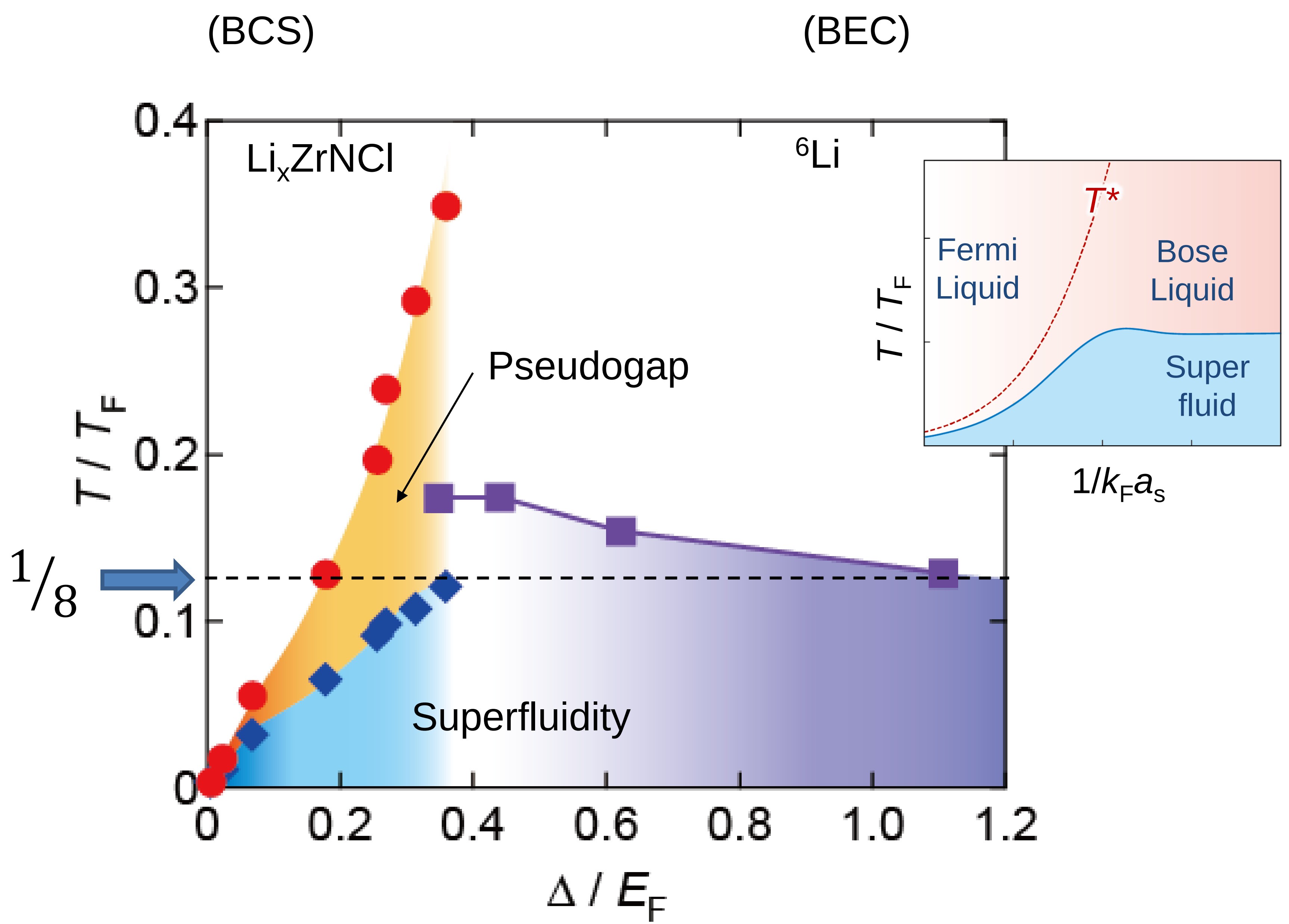



PRL 114, 230401 (2015)



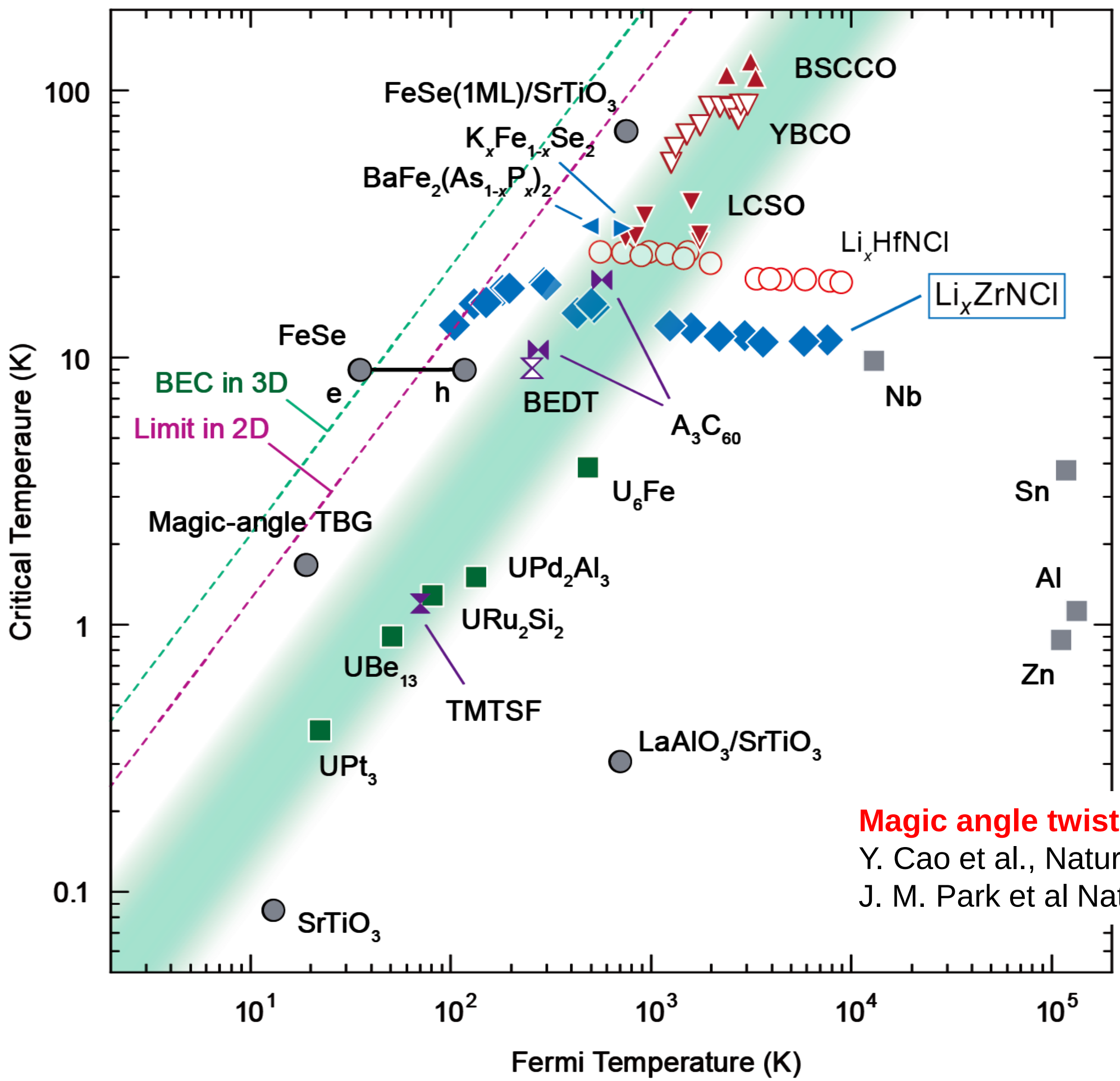

**arXiv:2106.11893 (2021)**

# Unified experimental phase diagram



# Uemura plot

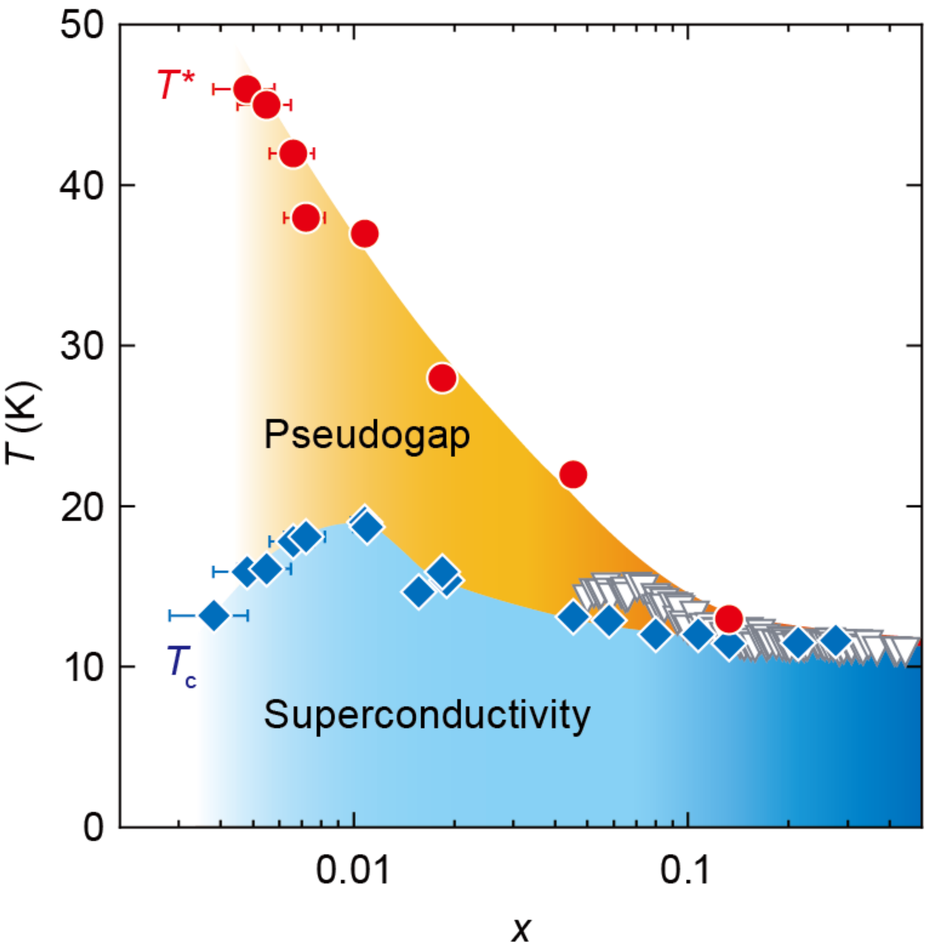
## Layered nitrides traverse from deep BCS to crossover to BEC.



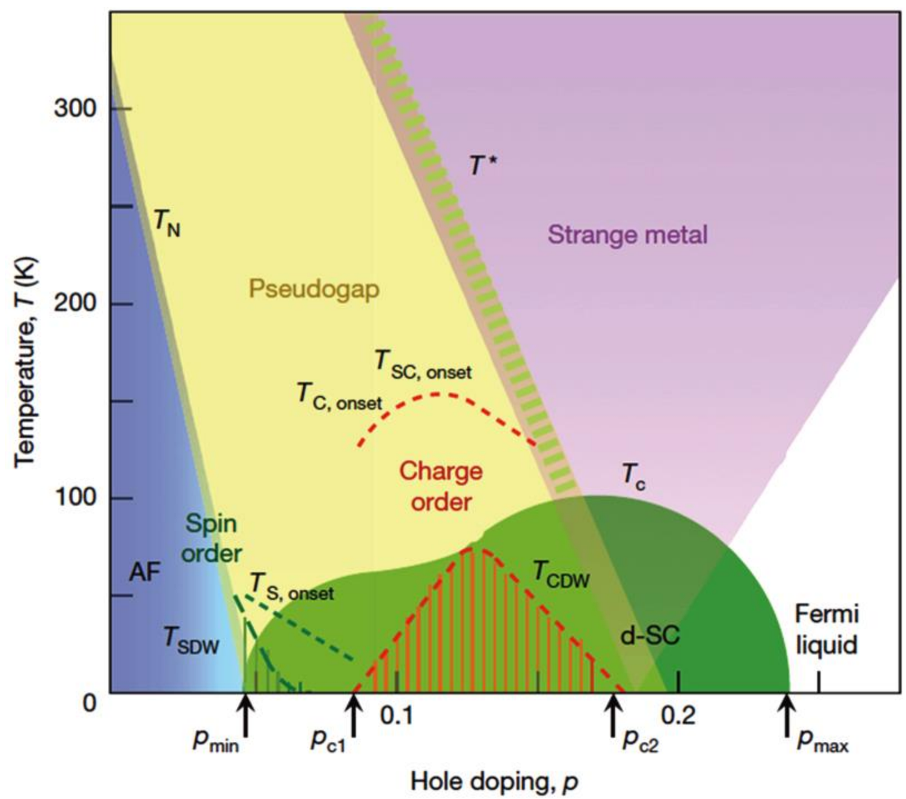

**Magic angle twisted bi & trilayer graphene**
Y. Cao et al., Nature 556, 43 (2018)
J. M. Park et al Nature 590, 249 (2021)

# Comparison with cuprates

$Li_xZrNCl$




Cuprates

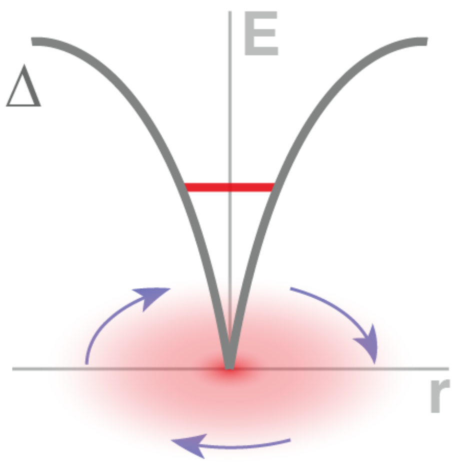



B. Keimer *et al.*, *Nature* **518**, 179 (2015).

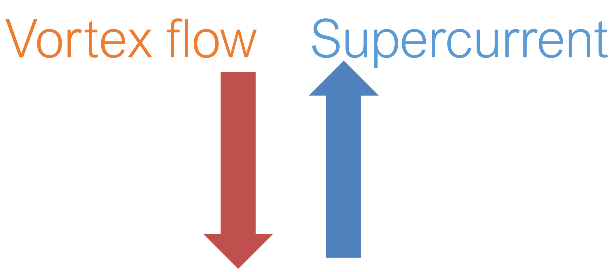
# Vortex properties in the two limits

Caroli-de Genne-Matricon quantization

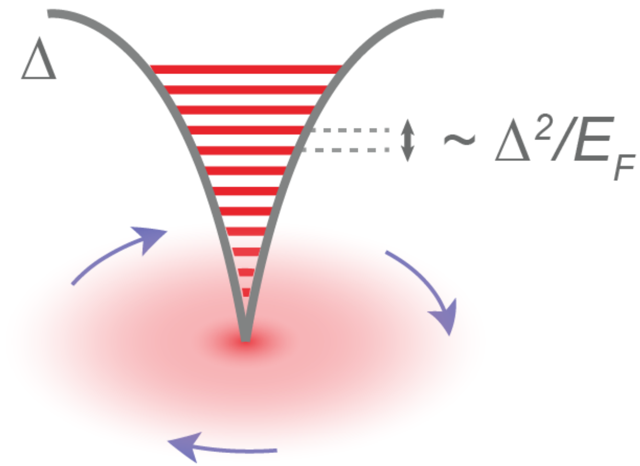
**BEC**



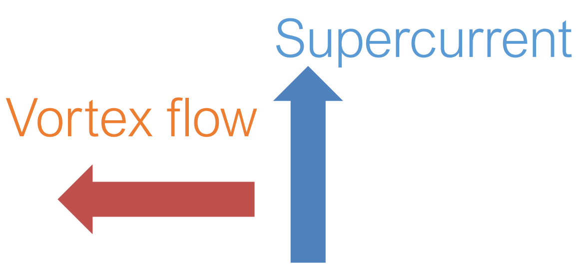

- Dissipationless core



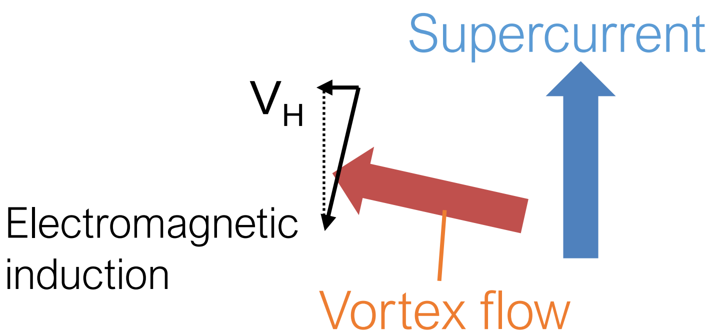

**BCS**




- Dissipative core



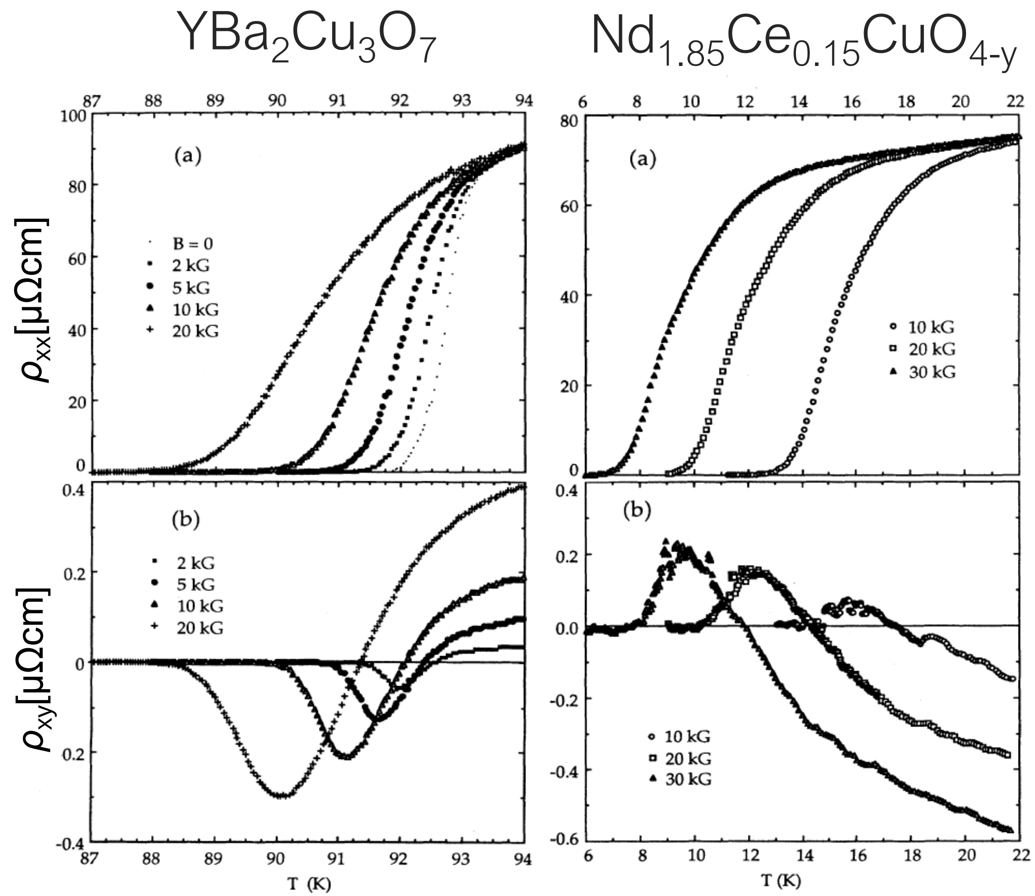
# Vortex Hall effect




Occurs in vortex liquid region

➢ important for layered superconductors

Remains to be understood.

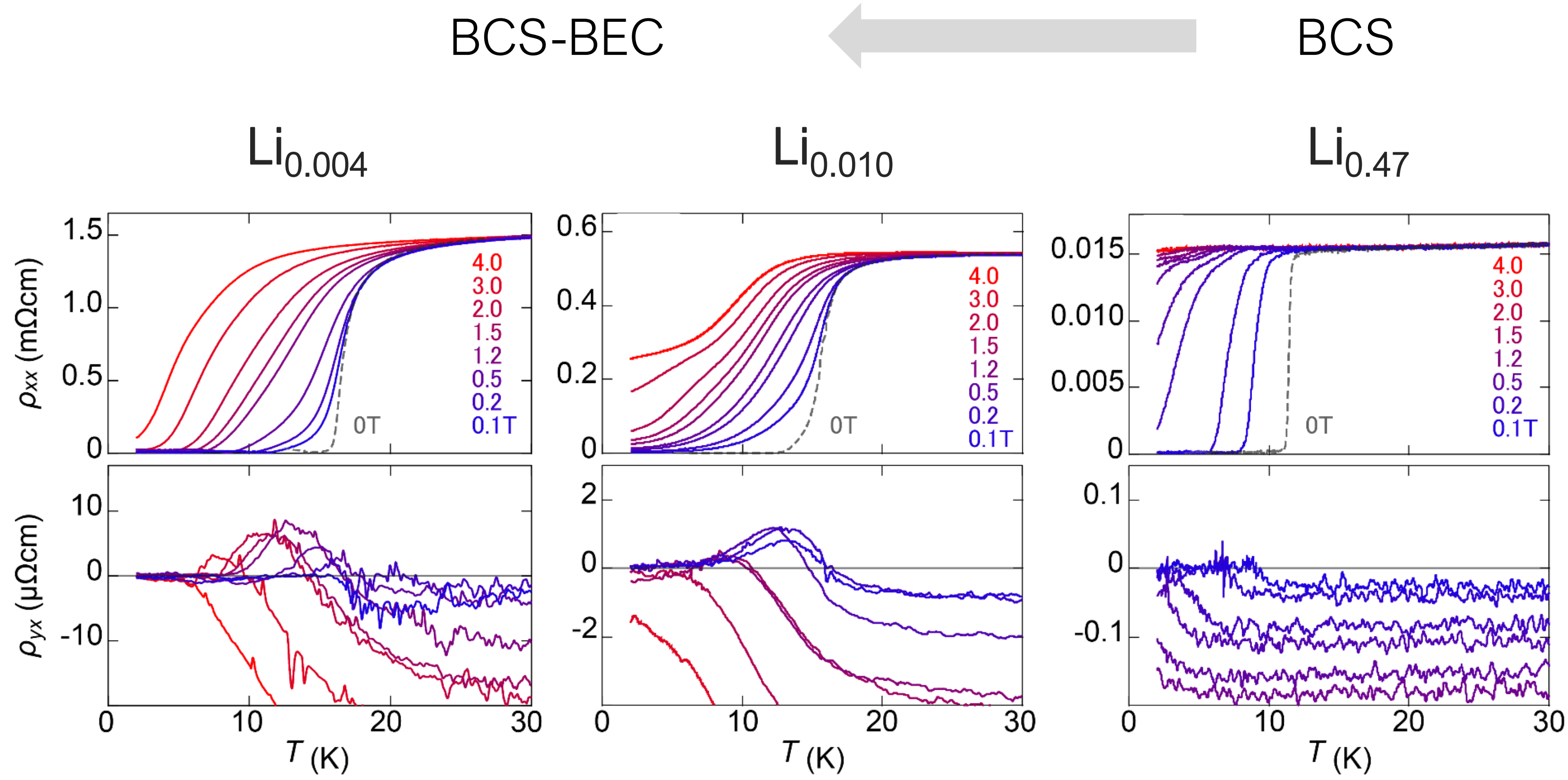



S.J. Hagen et al., PRB 47, 1064 (1993)

More VHE in cuprates:

Y. Iye, S. Nakamura, and T. Tamegai, Physica C 159, 616 (1989)
T. Nagaoka et al., PRL 80, 3594 (1998)
S. Zhao et al., PRL **122**, 247001 (2019).
R. Ogawa et al., PRB 104, L020503 (2021)

# Evolution of VHE with doping

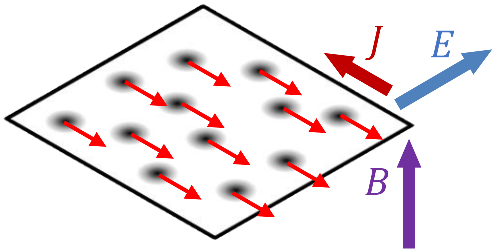


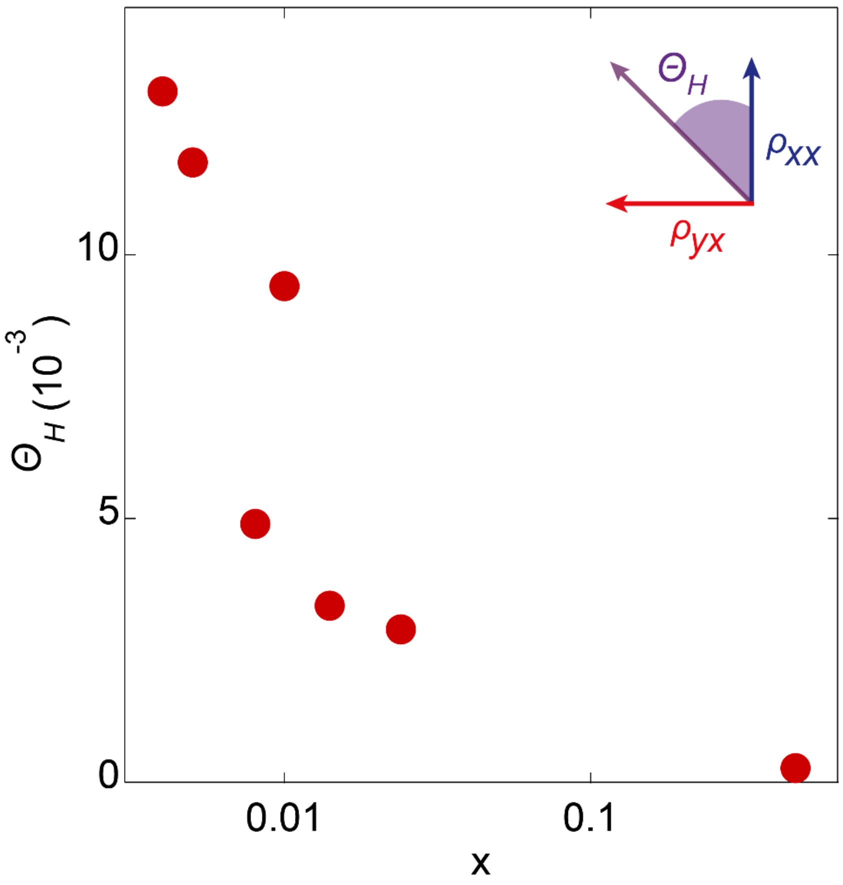

Increased doping leads to decreased VHE.

# Hall angle vs doping

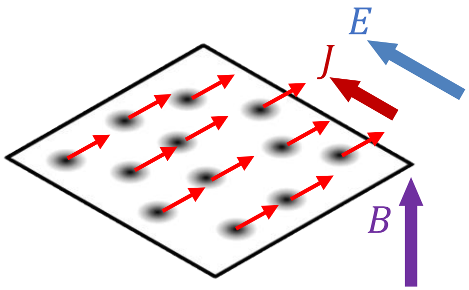
captures a trace of the large Hall angle expected in BEC-limit

BEC




BCS



# 2D time-dependent Ginzburg-Landau (TDGL) model

Kyosuke Adach RIKEN BDR, iTHEMS
Yusuke Kato, Univ Tokyo

$$(\gamma + \mathrm{i}\lambda)\frac{\partial \Delta(\boldsymbol{r},t)}{\partial t} = \left[-\frac{T-T^*}{T^*} - b|\Delta(\boldsymbol{r},t)|^2 + \xi^2\left(\boldsymbol{\nabla} + \mathrm{i}\frac{2\pi}{\phi_0}\boldsymbol{A}(\boldsymbol{r})\right)^2\right]\Delta(\boldsymbol{r},t) + \zeta(\boldsymbol{r},t)$$

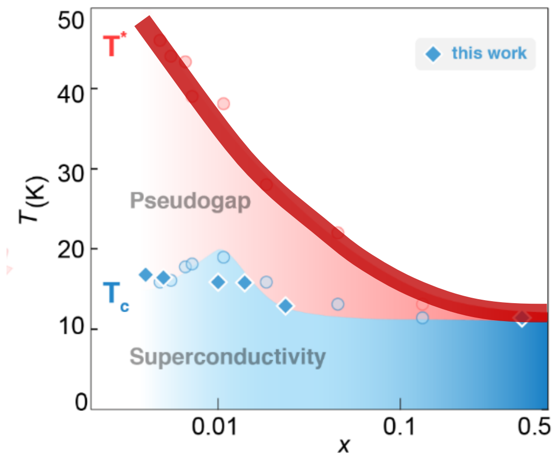
$T^*$ identified as mean-field transition temperature

$\gamma = \frac{\pi}{8T^*}$ ← For simplicity, we use the value derived for BCS region
Abrahams & Tsuneto, Phys. Rev. 152, 416 (1966)

$\lambda = -\frac{1}{2T^*}\frac{\partial T^*}{\partial E_F}$ ← Based on gauge invariance
Aronov, Hikami, & Larkin, PRB 51, 3880 (1995)

$\xi \left(= \sqrt{\phi_0/\, 2\pi B_{c2}(0)}\right)$ coherence length

$b$ parameter of fluctuation interaction

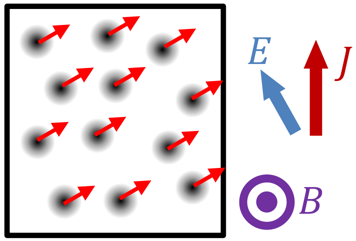



**Vortex conductivity**

Ullah & Dorsey, PRB 44, 262 (1991)

$$\sigma_{ab}^{\mathrm{V}} = \frac{1}{TS}\int_0^\infty \mathrm{d}t \int \mathrm{d}^2\boldsymbol{r}\, \mathrm{d}^2\boldsymbol{r}'\, \langle j_a(\boldsymbol{r},t) j_b(\boldsymbol{r}',t)\rangle$$

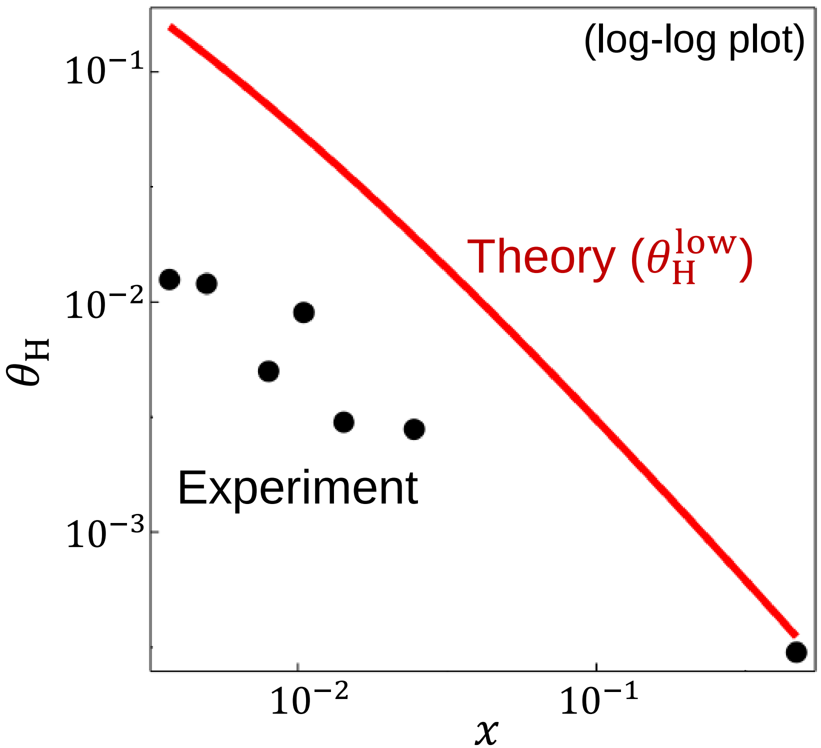
Abrahams, Prange, & Stephen, Physica 55, 230 (1971)




$$J_a^{\mathrm{V}} = \sum_{b=x,y} \sigma_{ab}^{\mathrm{V}} E_b$$

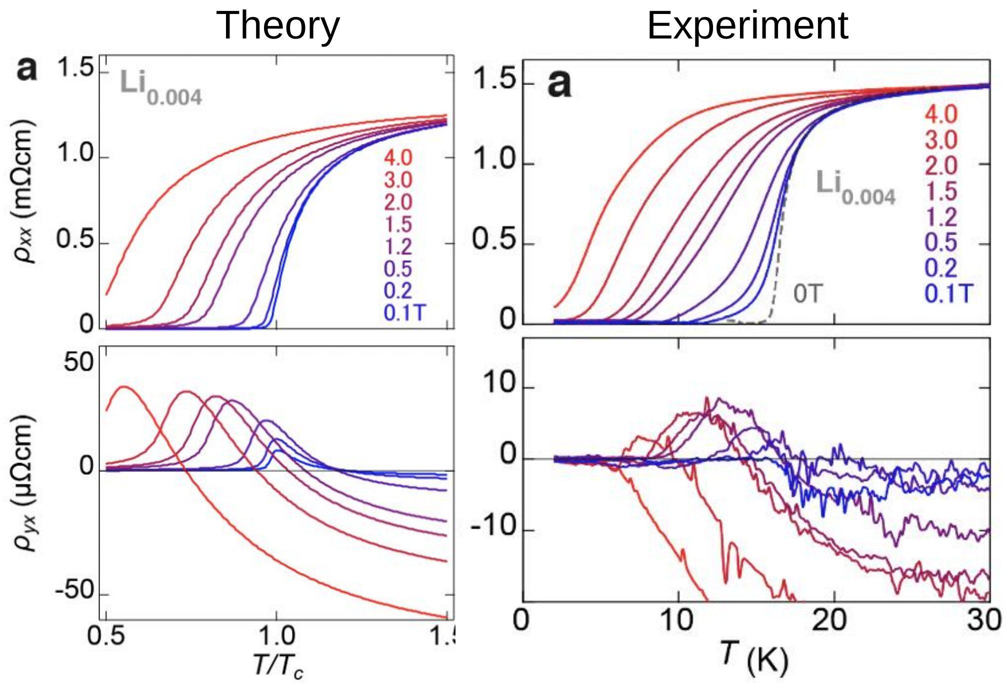
# Comparison of theory and experiments

## Hall angle vs. x



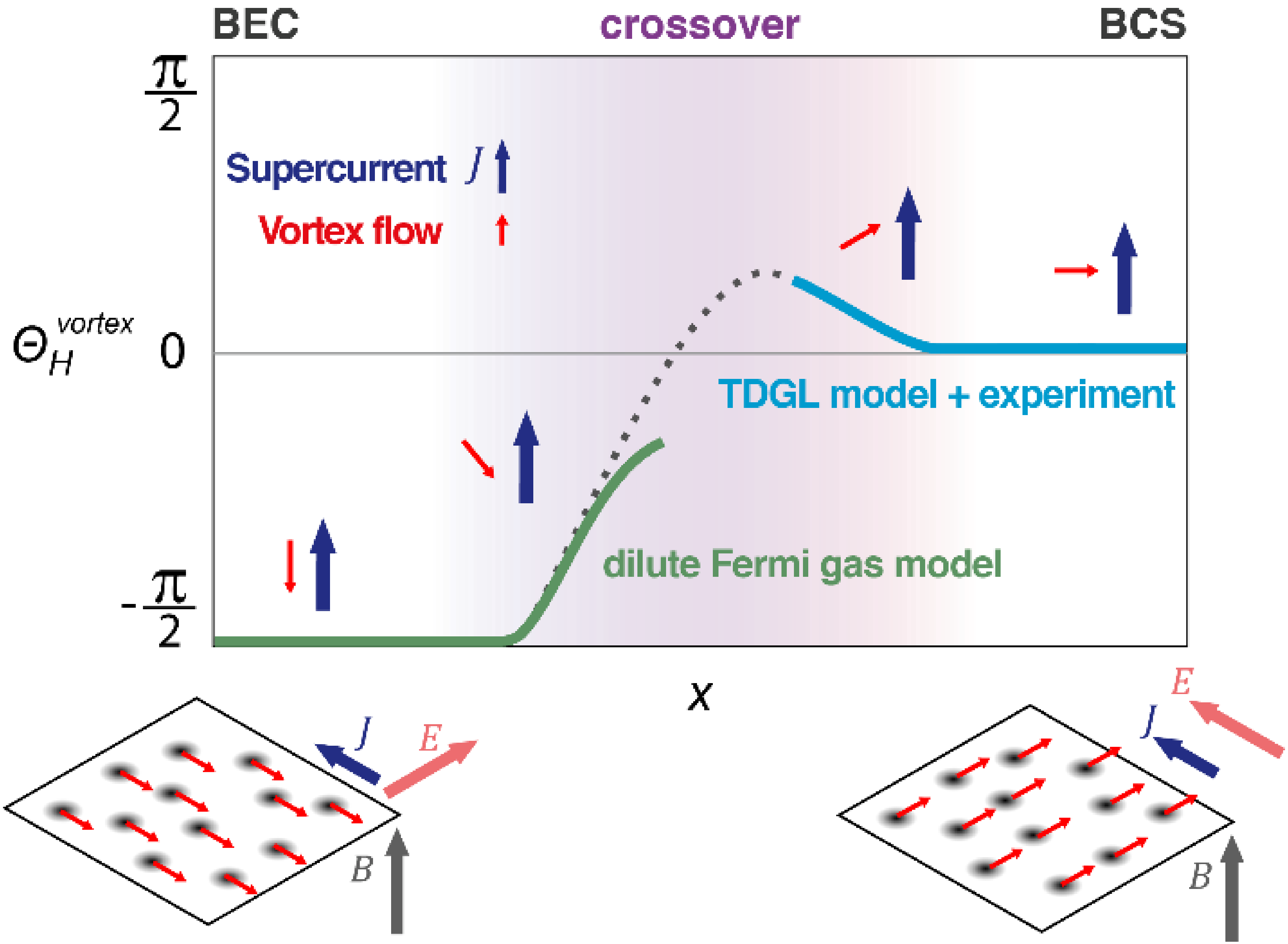

**Trend in $\theta_H$ is qualitatively captured**

## Temperature dependences



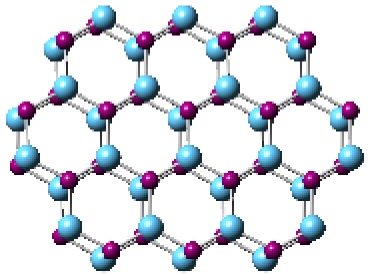

**The Hall anomaly is well reproduced**

# Evolution of vortex Hall angle through crossover



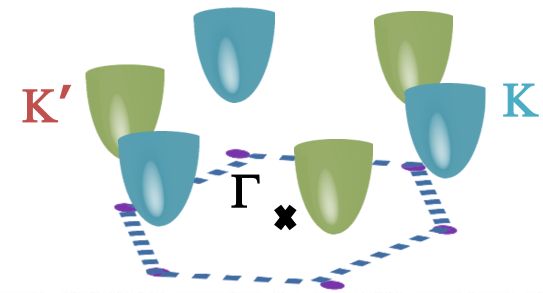
# Summary

1. Gated superconductivity of ZrNCl to the low carrier density limit

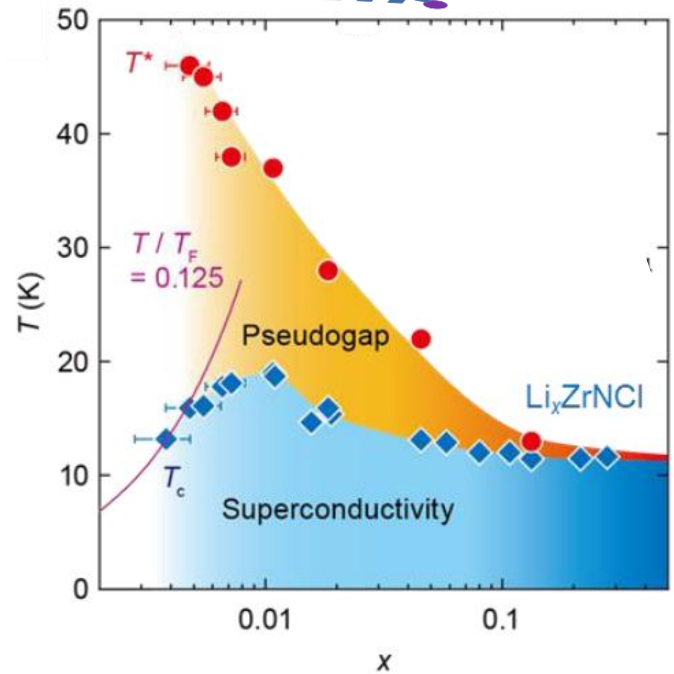


2. 2D BCS-BEC crossover
   - Pesudogap phase
   - Upper limit $T_{BKT}/T_F = 1/8$

   Ideal 2D system with parabolic dispersion
   Without any magnetic or CDW instabilities




3. Vortex Hall effect in the crossover
   - Enhanced Hall angle toward BEC (Signature of superclean region)





Y. Nakagawa *et al.*, *PRB.* **98**, 064512 (2018).
Y. Nakagawa *et al.*, *Science* **372**, 190 (2021).
M. Heyl, K Adachi et al., submitted.