SCHOOL OF SCIENCE
THE UNIVERSITY OF TOKYO
Cryogenic Research Center

Grand Challenges in QFS @Buffalo (Aug. 7, 2015)

New Quantum Phases of Monolayers of Helium

Hiroshi FUKUYAMA

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Cryogenic Research Center (CRC),
The University of Tokyo

1. New quantum spin liquid state in 2D ^{3}He
2. Search for supersolidity in 2D ^{4}He
3. Liquefaction of ^{3}He in 2D
4. Search for superfluidity in monolayer liquid ^{3}He

Quantum Liquid Crystal

New quantum state of matter: Quantum Liquid Crystal

fully preserved

quantum liquid

$T = 0$

trans. and rota. symmetries are partially broken

quantum liquid crystal (QSL)

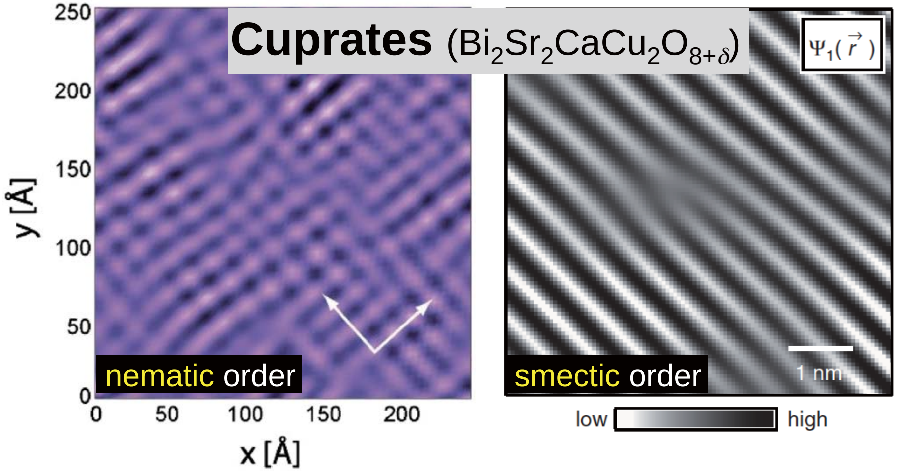
translational and rotational symmetries are fully broken

quantum solid

solid

electronic **Nematic phases**

C. Howald et al., PRB **67**, 014533 (2003)   A. Mesaros et al., Science **333**, 426 (2011)



$Sr_3Ru_2O_7$ R.A. Borzi et al. Science **315**, 214 (2007)

2DES in GaAs/GaAlAs   M.P. Lilly et al., PRL **82**, 394 (1999)

atomic **QLC phases** ?

2D ^{4}He • **supersolidity** ?

2D ^{3}He • **gapless quantum spin liquid**
• **stripe superfluid phase**

Q. interplay between superfluid (or spin) and spatial orders?

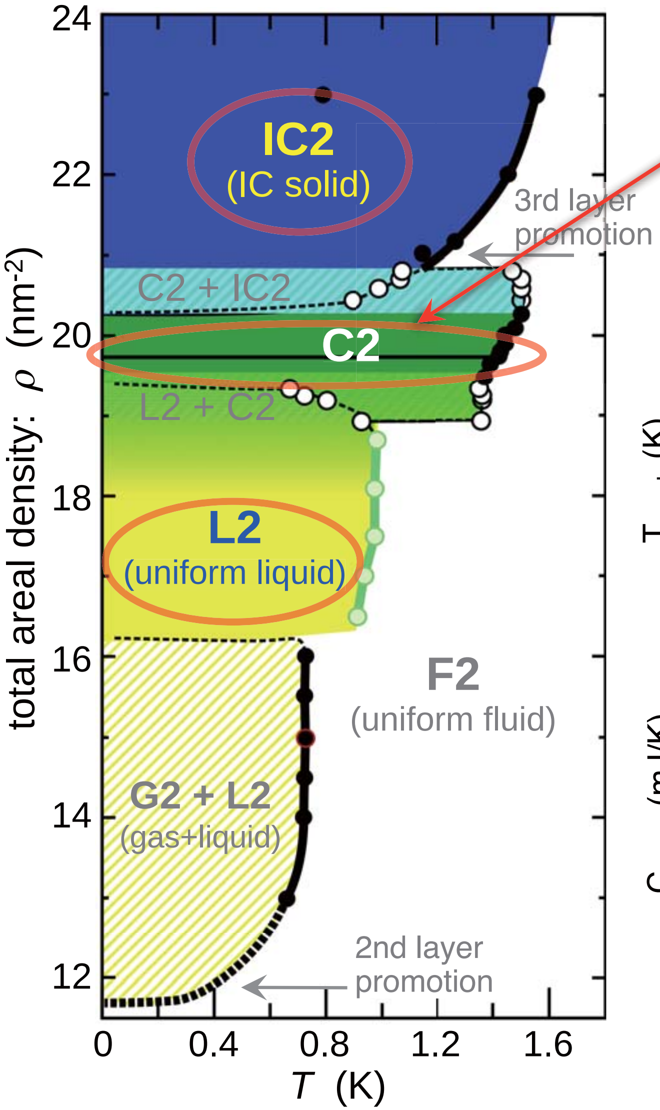
Q. ubiquitous for strongly correlated 2D quantum systems?

Phase diagram of 2nd layer of ^{4}He (2D bosons)

S. Nakamura et al.,arXiv:1406.4388v1

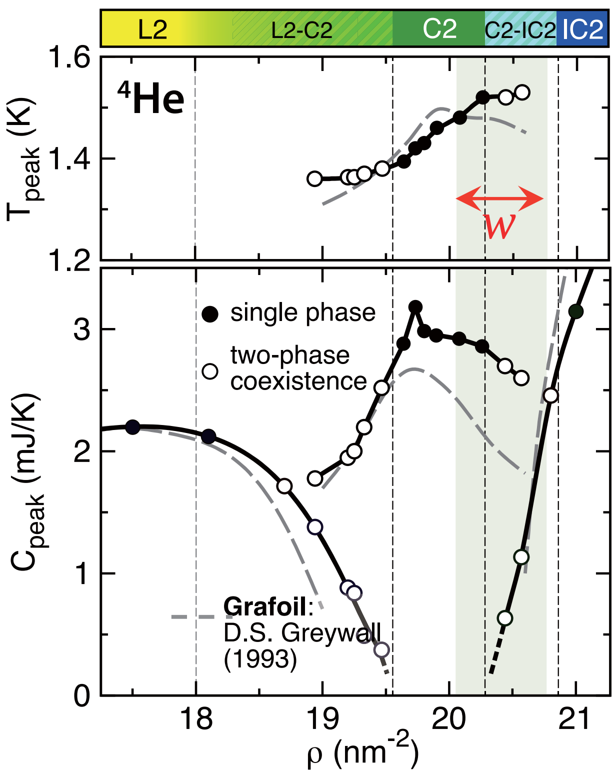
HC data with **ZYX** substrate

S. Nakamura et al., to appear

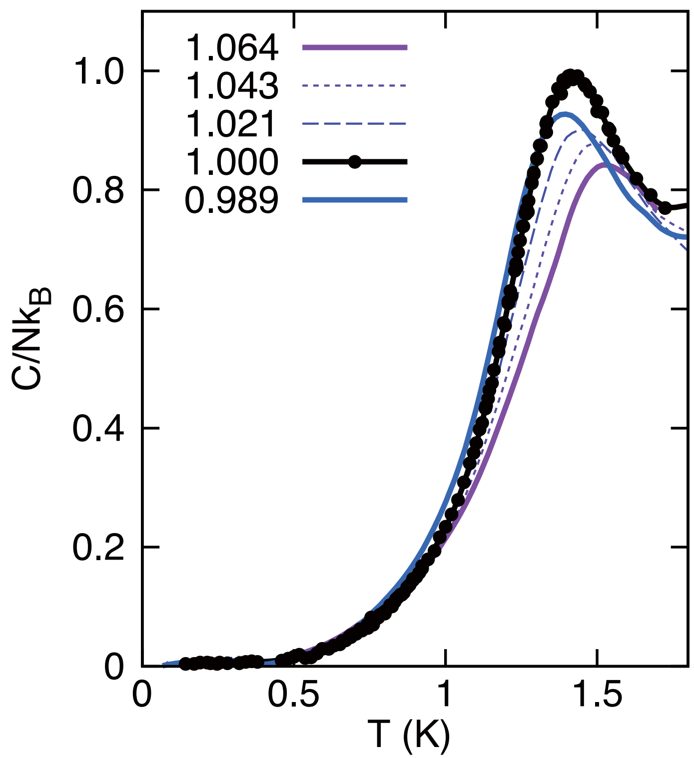


C2: **compressible commensurate phase!?**

$w \equiv \Delta\rho_{C2}/\rho_{C2} \approx 0.10$



pure phase within w



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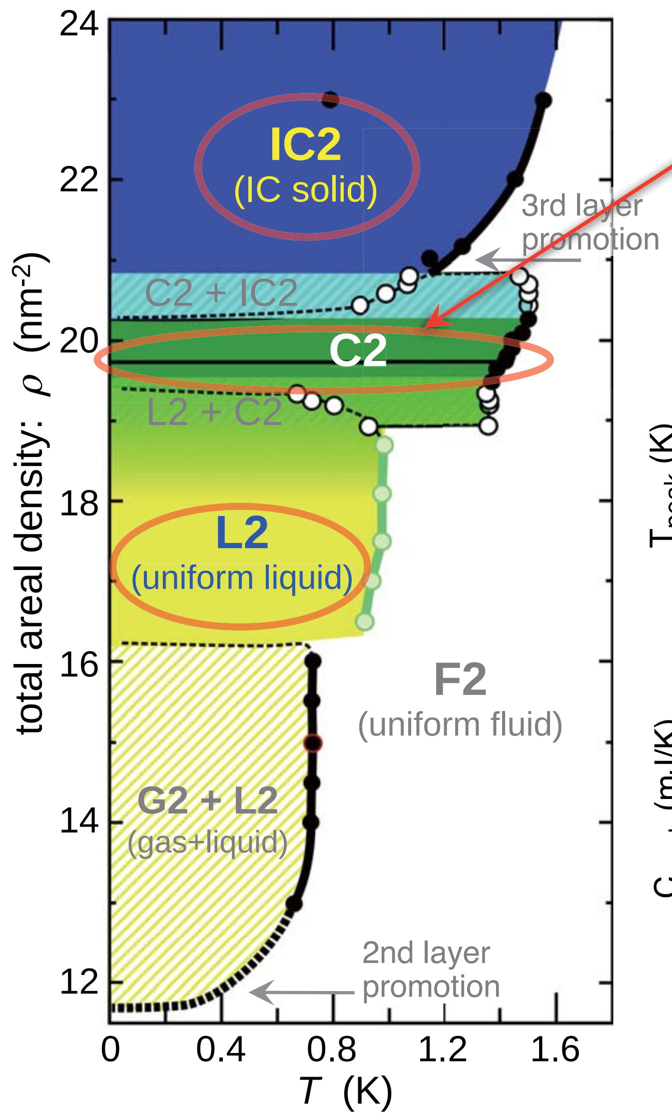

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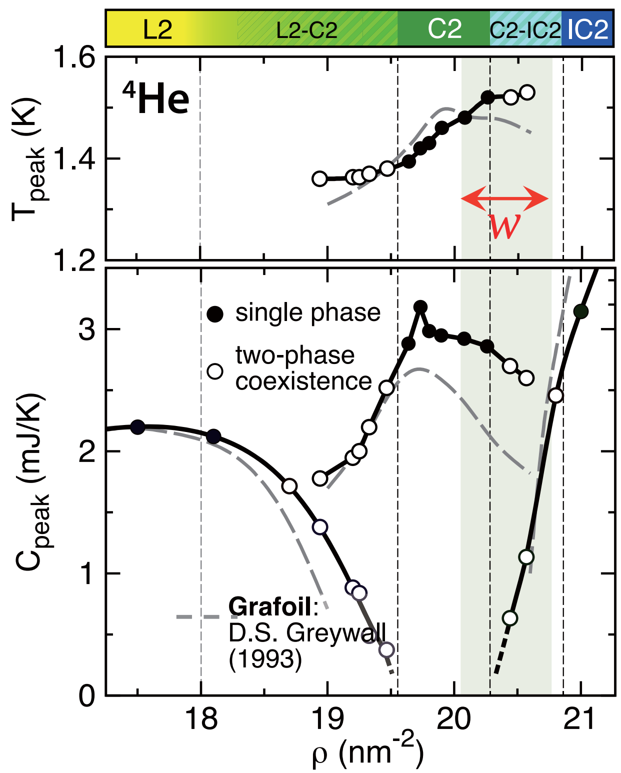
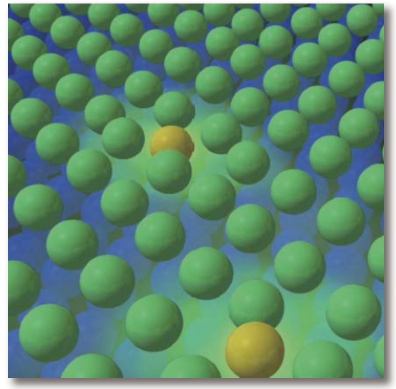
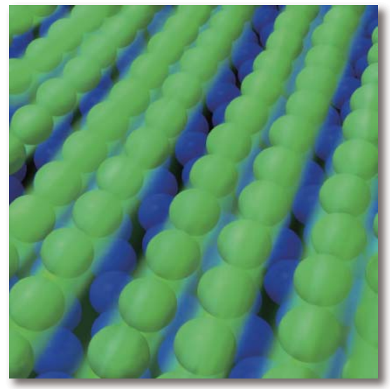


quantum solid with
zero-point defectons



stripe (smectic) phase



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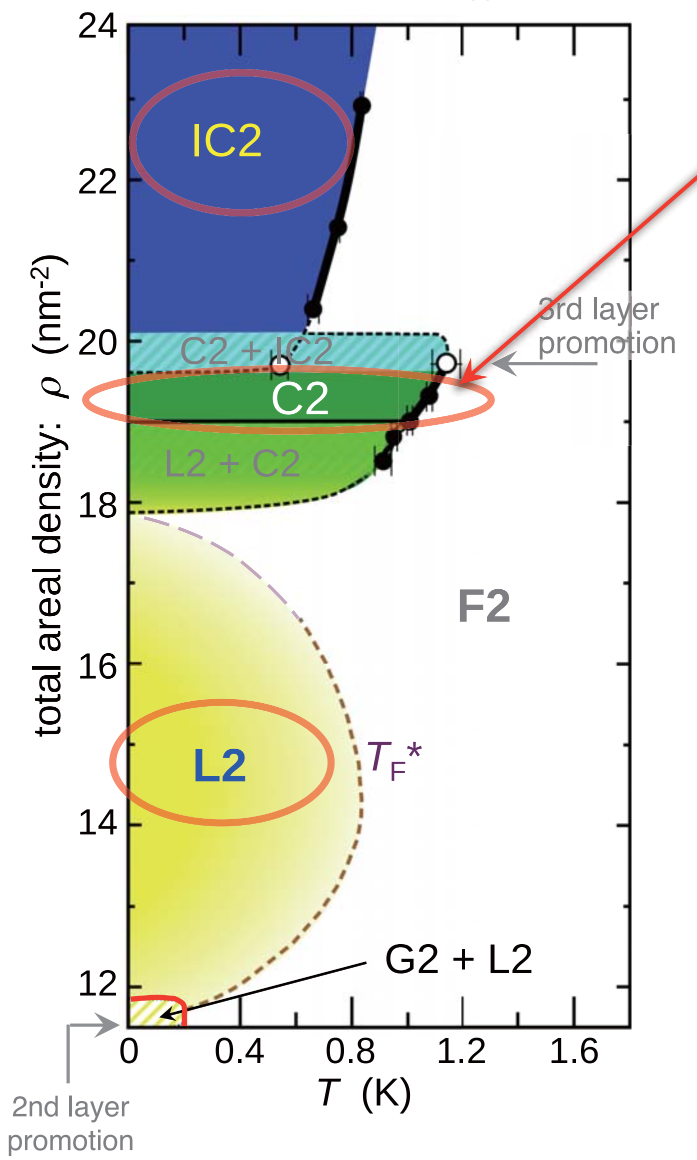

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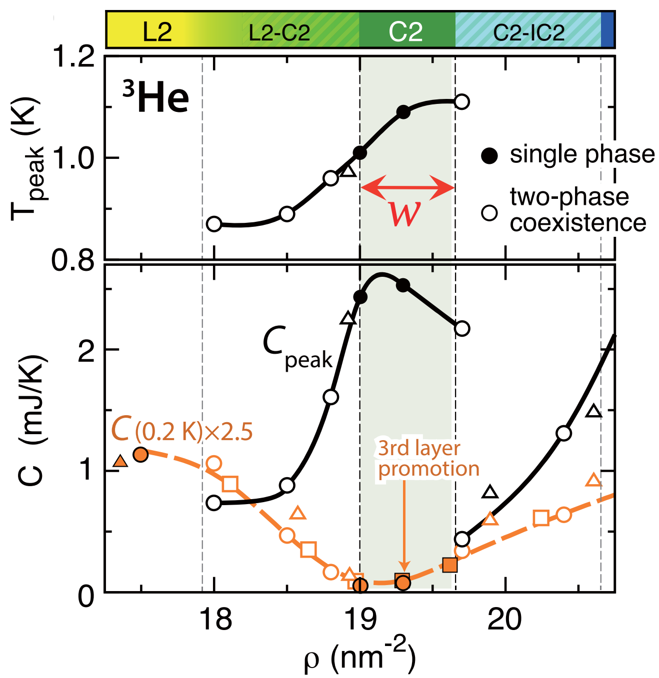

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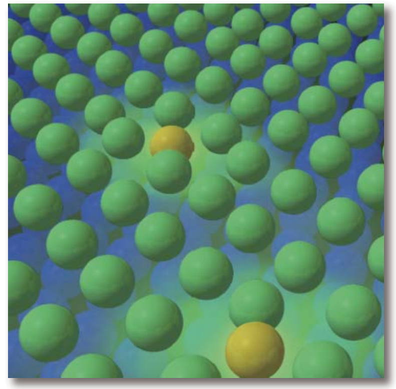

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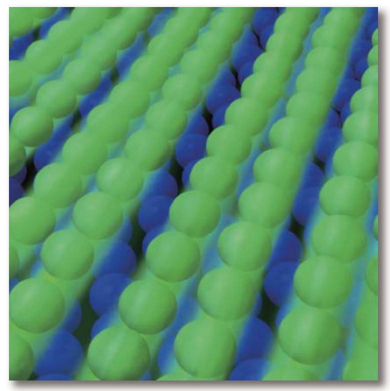


Gapless quantum spin liquid (QSL) in 2D materials

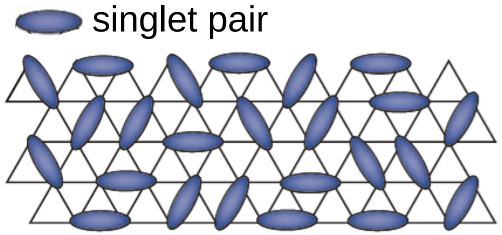
QSL: $\langle S_i \rangle = 0$ without LRO at $T = 0$

theories

Short range RVB (resonating valence bond)

P.W. Anderson (1973, 1987)

S =1/2 Heisenberg N.N. antiferro-magnet on triangular lattice (HAFT)



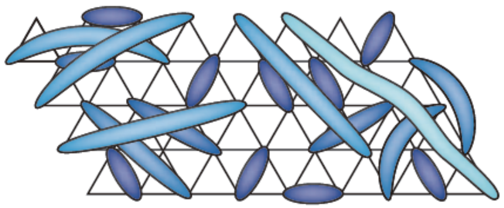
+ • • •

gapful ($\Delta \sim J$)

Long range RVB

L.S. Doucot and P.W. Anderson (1988)

HAFT with longer distance interactions

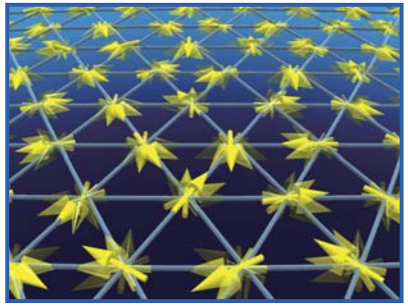


gapless

+ • • •

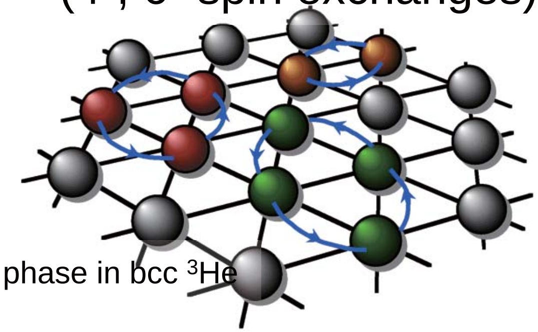
2D ^{3}He

S = 1/2 on triangular lattice



ring exchanges

(4-, 6- spin exchanges)



cf. u2d2 phase in bcc ^{3}He

Exp. claim of **gapless GSL** in 2D ^{3}He

K. Ishida, HF et al. PRL **79**, 3451 (1997)

2D ^{3}He

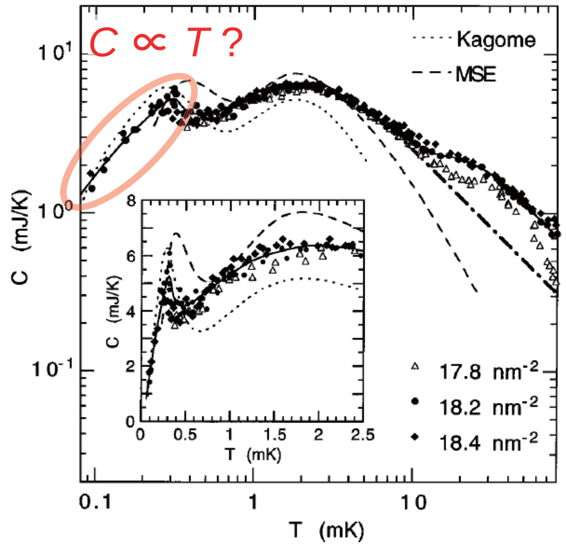
Research history of QSLs in condensed matter

1989 D.S. Greywall ^{3}He/^{3}He/gr
- found highly frustrated magnetism (missing entropy?)

1989 V. Elser
- proposal of 4/7 structure (Heisenberg Kagome)

1993 M. Siqueira et al. ^{3}He/HD/HD/gr
- found 4/7 phase on ^{3}He/HD/HD/gr

1997 K. Ishida et al. ^{3}He/^{3}He/gr
- found double-peak in $C(T)$ and $C \propto T$ at $T < T_{peak}$
- proposal of "gapless QSL"



1998 G. Misguich et al.
- gapful QSL (exact diagonalization of RE model with 6-spin)

2000 H. Ikegami et al. ^{3}He/HD/HD/gr
- M measurement to $T \approx 10^{-2}J$ supporting gapless QSL

2001 E. Collin et al. ^{3}He/^{4}He/gr
- M measurement to $T \approx 10^{-2}J$ suggesting QSL with small gap

2004 R. Masutomi et al. ^{3}He/HD/HD/gr, ^{3}He/^{4}He/gr
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2009 H. Nema et al. ^{3}He/^{4}He/gr
- found $M_{sat}/2$ plateau at $B \approx 1.5$ T suggesting importance of J_4

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- M measurement to $T \approx 10^{-2}J$ … suggesting QSL with small gap

Really $C \propto T$?

2004 R. Masutomi et al. ^{3}He/HD/HD/gr, ^{3}He/^{4}He/gr
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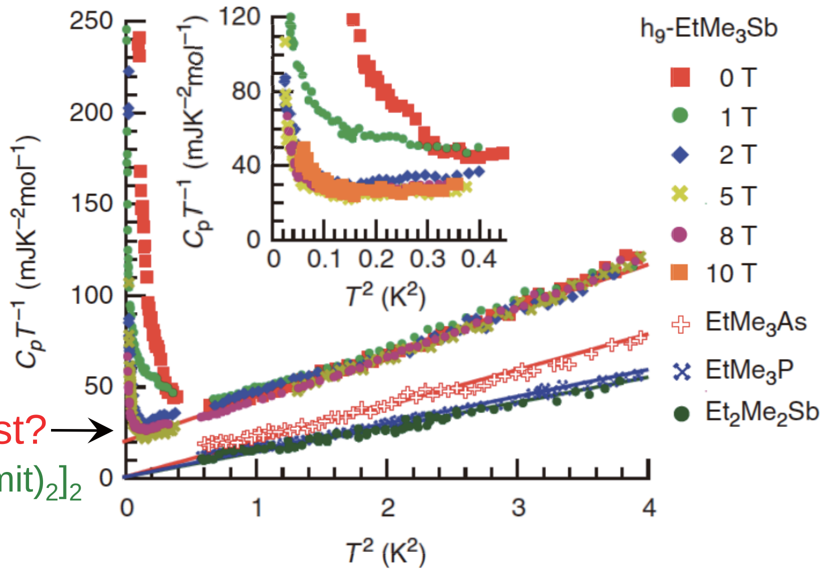
2009 H. Nema et al. ^{3}He/^{4}He/gr
- found $M_{sat}/2$ plateau at $B \approx 1.5$ T suggesting importance of J_4

2015 M. Kamada et al. ^{3}He/HD/HD/gr
- found new gapless QSL

$CT \approx$ const? →

$EtMe_3Sb[Pd(dmit)_2]_2$



electronic systems

[κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$] S=1/2 HAFT

2003 Y. Shimizu et al. NMR-T_2, $\chi \to$ const

2008 S. Yamashita et al. $C \propto T$

2010 M. Yamashita et al. $\kappa/T \to 0$

$EtMe_3Sb[Pd(dmit)_2]_2$ S=1/2 HAFT

2007 T. Itou et al. NMR-T_1&T_2

2010 M. Yamashita et al. $\kappa \propto T$

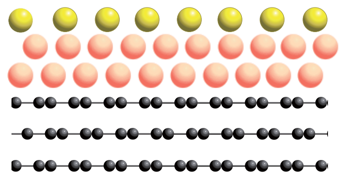
2011 S. Yamashita et al. $C \propto T$

$Na_4Ir_3O_8$ S=1/2 3D hyper-Kagome

2007 Y. Okamoto et al. $C \propto T^{2.5}$, $\chi \to$ const

Found new QSL phase in 2D ^{3}He

- **On bilayer of HD** M. Kamada, QFS2015 (Thursday afternoon)
- **Lowest density** solid ^{3}He ever found (**5.25 nm^{-2}**)



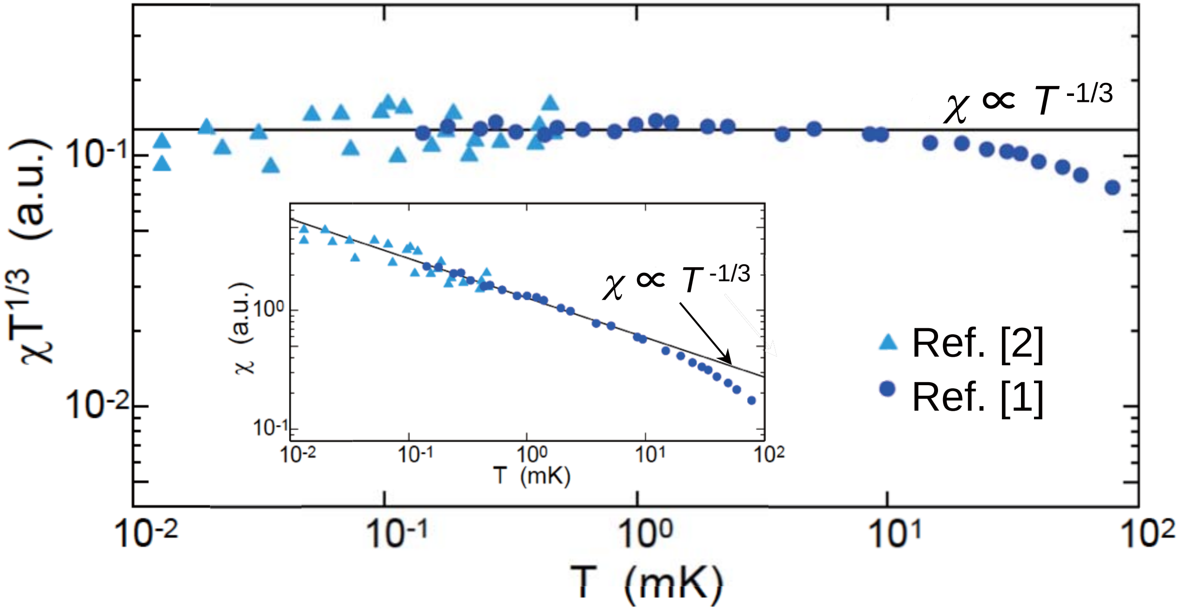
^{3}He/HD/HD/gr

cf. U. London group

$C \propto T^{2/3}$!

TBA

$\chi \propto T^{-1/3}$!



$\chi T/C\ (\propto R_W) = \text{const.}$ ... Can define Wilson ratio ?!

[1] H. Ikegami et al., PRL **85**, 5146 (2000)
[2] R. Masutomi, et al., PRL **92**, 025301 (2004)

Entropy change of new QSL phase

^{3}He/HD/HD/gr

ρ = 5.25 nm^{-2}

TBA

Advantages of 2D ^{3}He:

- phonon contribution to C ($\propto$ T^2) appears only at very high T (>

Spin entropy change deuced from specific heat data:

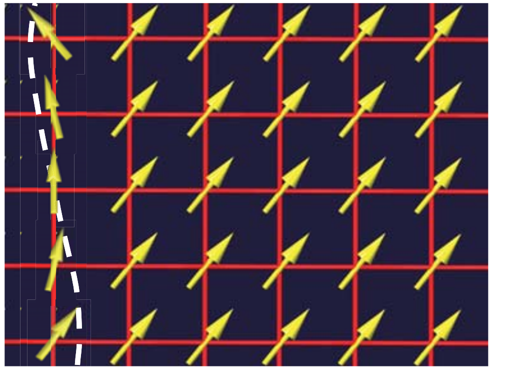
$\Delta S \approx Nk_B \ln 2$ $\rightarrow$ quantum solid (or QLC)

What are elementary excitations in QSLs ?

classical magnets with LRO

- magnon (boson) at T ≠ 0

  e.g. 2D AFM spin wave : $C \propto T^2$, $\chi = \text{const.}$



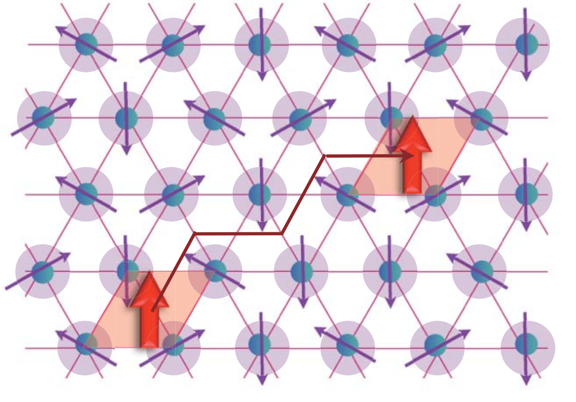
Magnon (spin wave)

gapless QSL

$C \propto T^{2/3}$ has been predicted by several **fermion fractionalization** theories, but has not been experimentally seen before!

→ deconfined **spinon** Fermi surface?

- C.P. Nave and P.A. Lee, PRB **76**, 235124 (2007)
- O. I. Motrunich, PRB, **72**, 045105 (2005):
  Hubbard model with **4-spin exchange**
  $C \propto T^{2/3}$, $\chi = \text{const.}$
- R.R. Biswas, *et al.*, PRB, **83**, 245131 (2011)
  S =1 SU(2) **Majorana fermion**
  $C \propto T^{2/3}$, $\chi \propto T^{-1/3}$, $R_W \approx 4$



Spinon (*S* = 1)
quasi-particle like

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S. Nakamura et al.,arXiv:1406.4388v1

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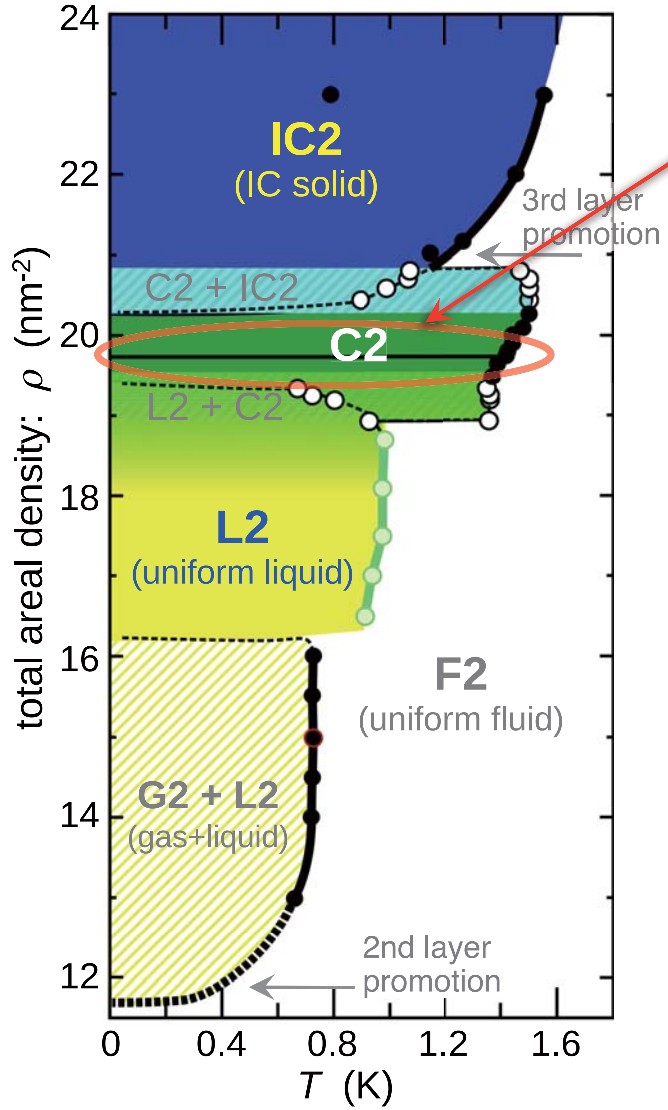
S. Nakamura et al., to appear



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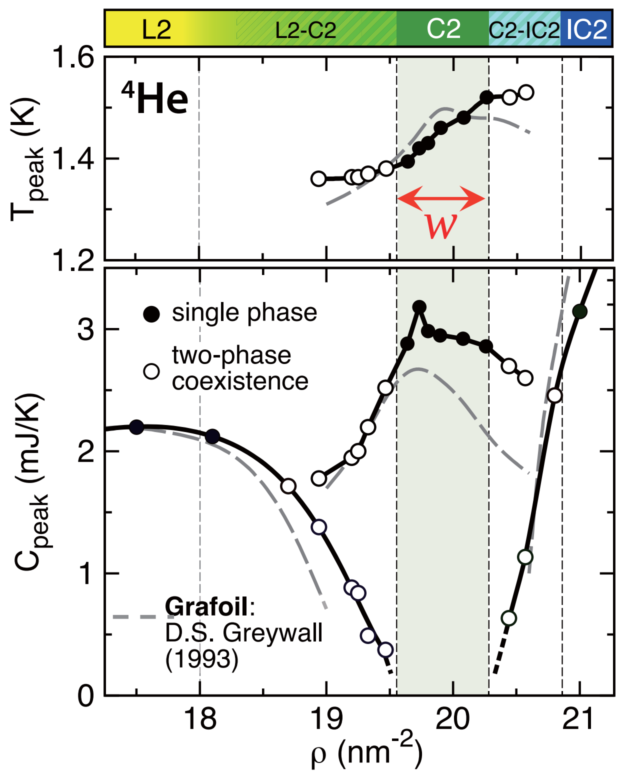

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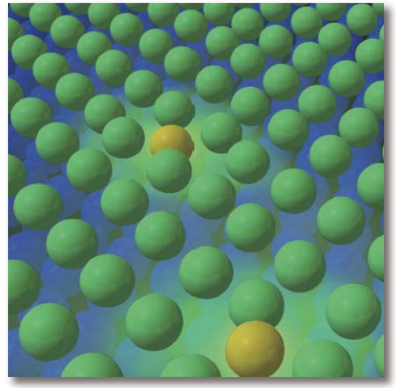

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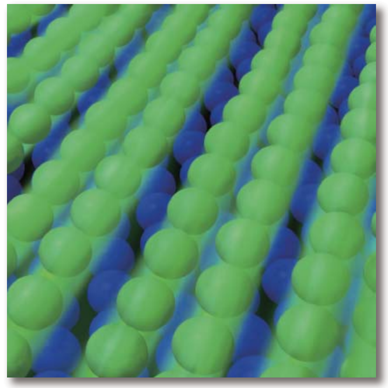
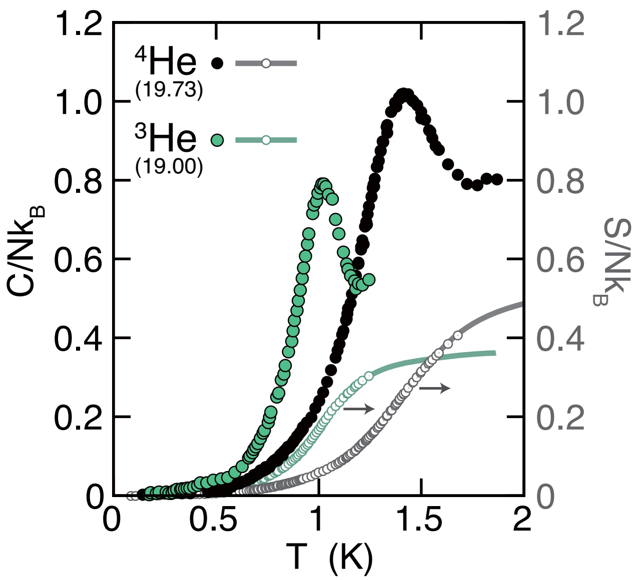


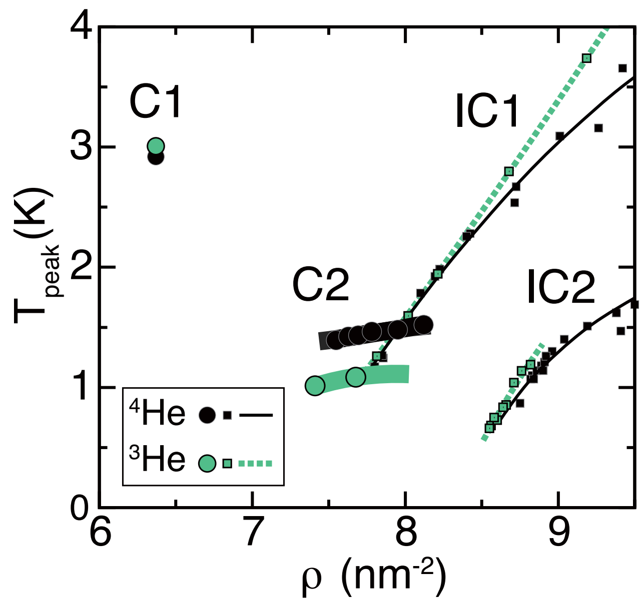
Supersolid transition at $T < 1$ K in ^4He-C2 ?

S. Nakamura et al.,arXiv:1406.4388v1

- Should be of 2D melting
- Anomalous isotope effect:

  $1.4\times T_{peak}(^3He) \approx T_{peak}(^4He)$
- Entropy release for ^4He is larger than ^3He by 40%.





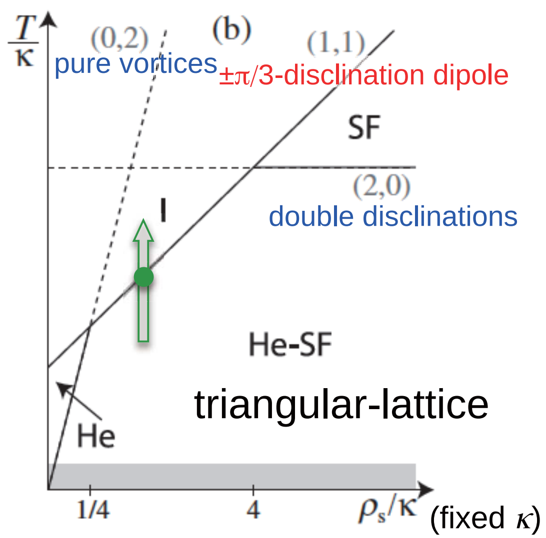
Possible supersolid transition in ^4He-C2 phase

Both **spatial** and **gauge** symmetries are *spontaneously*, ***partially*** and ***simultaneously*** broken below T_{peak}?

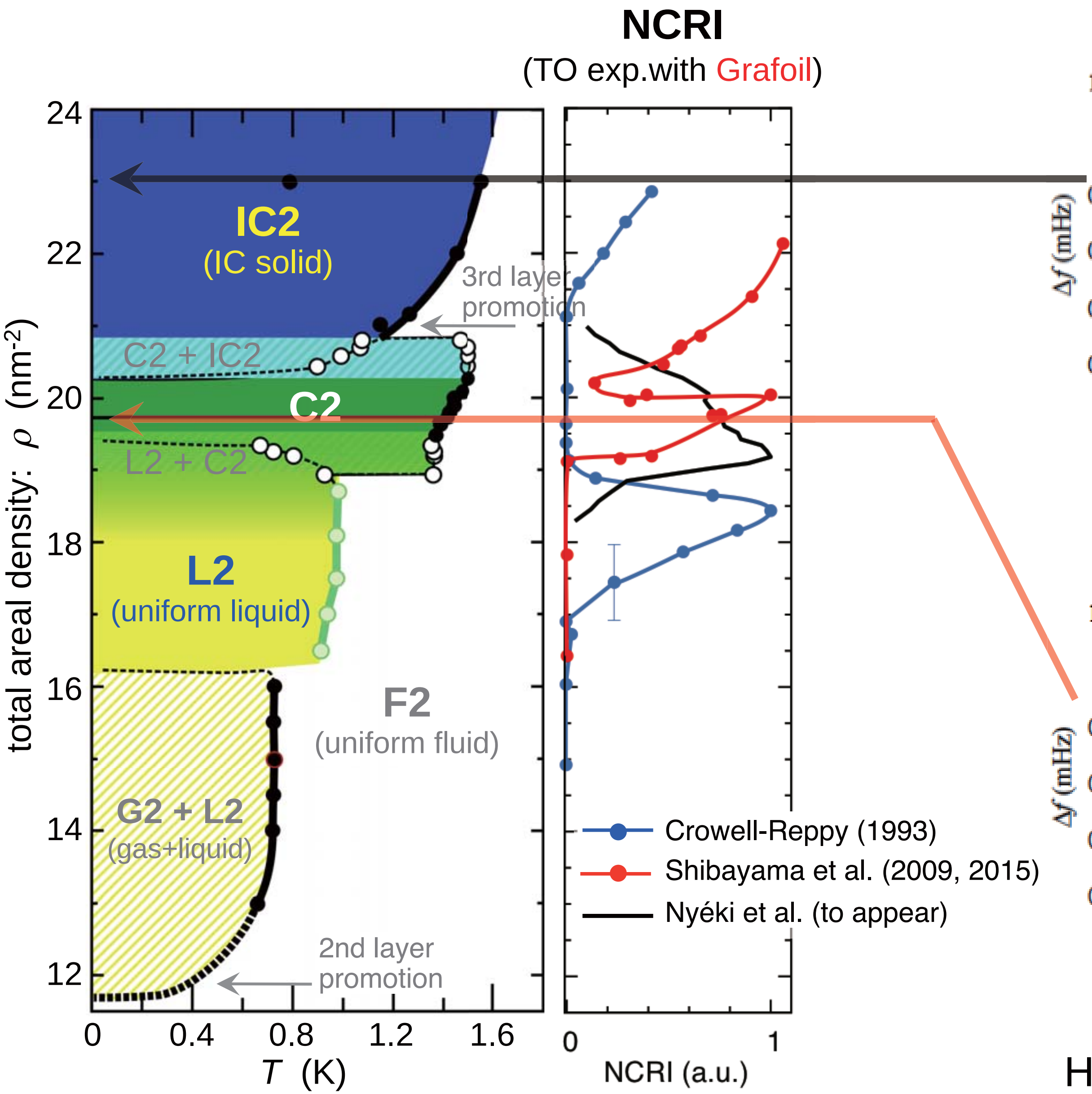
Intertwining of **crystalline** and **superfluid** orders in triangular lattice ?

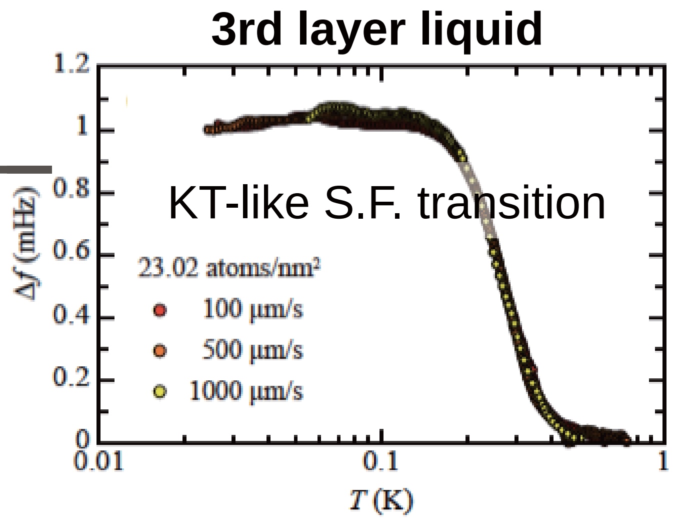
S. Gopalakrishnan, J.C.Y. Teo, and T.L. Hughes, PRL **111**, 025304 (2013)

$$\mathcal{H}_{\rm el.} = \kappa[(\nabla\cdot\mathbf{n})^2 + (\nabla\times\mathbf{n})^2] + \rho_s|\nabla\theta|^2$$

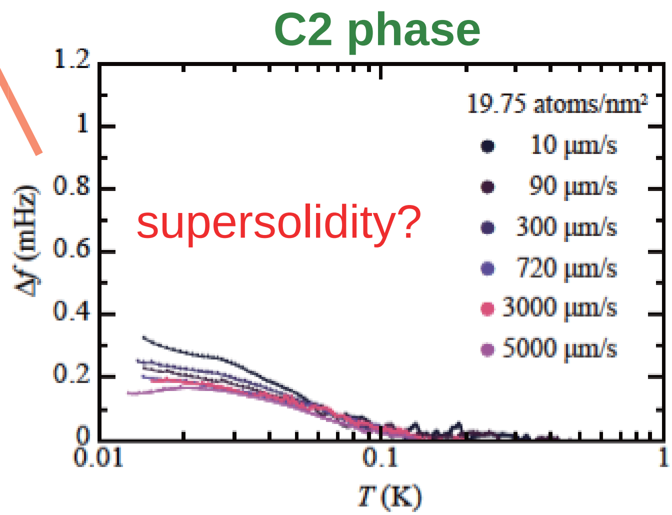


Torsional oscillator measurements with Grafoil substrate





Y. Shibayama et al. (2015)



However, $\Delta\rho_s/\rho \approx$ **0.01** (too small)

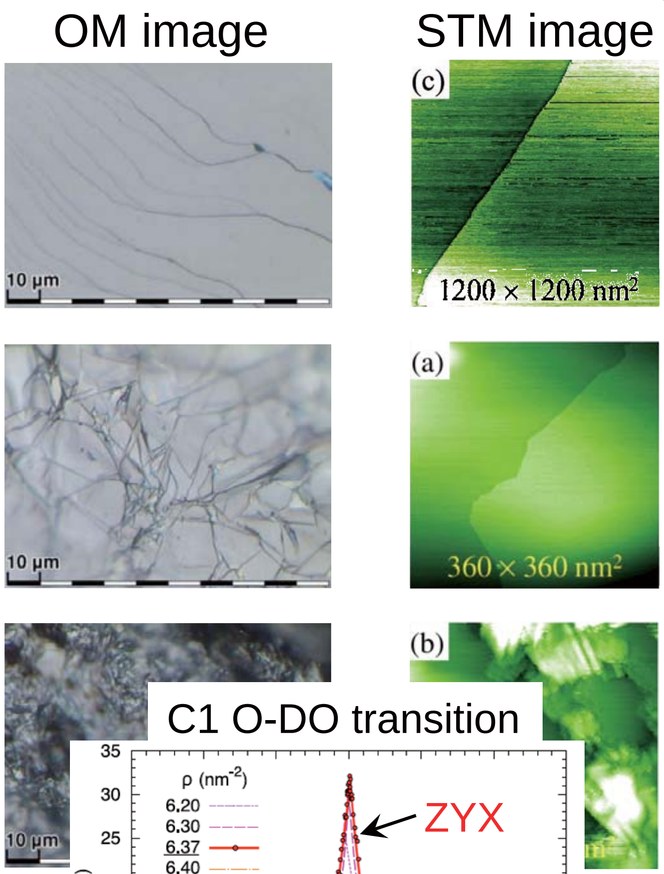
Need for better graphite substrate for T.O. experiments to search for supersolidity

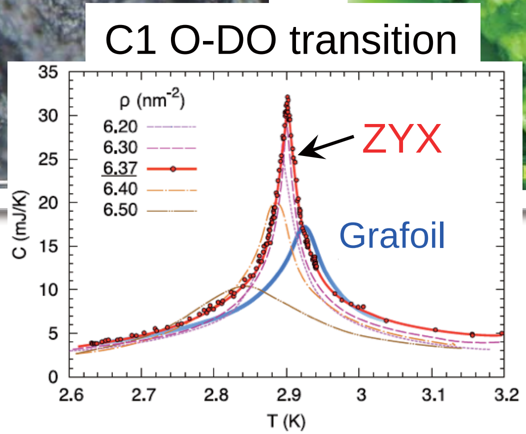
various exfoliated graphite substrate

HOPG
(highly oriented pyrolytic graphite)
only top surface

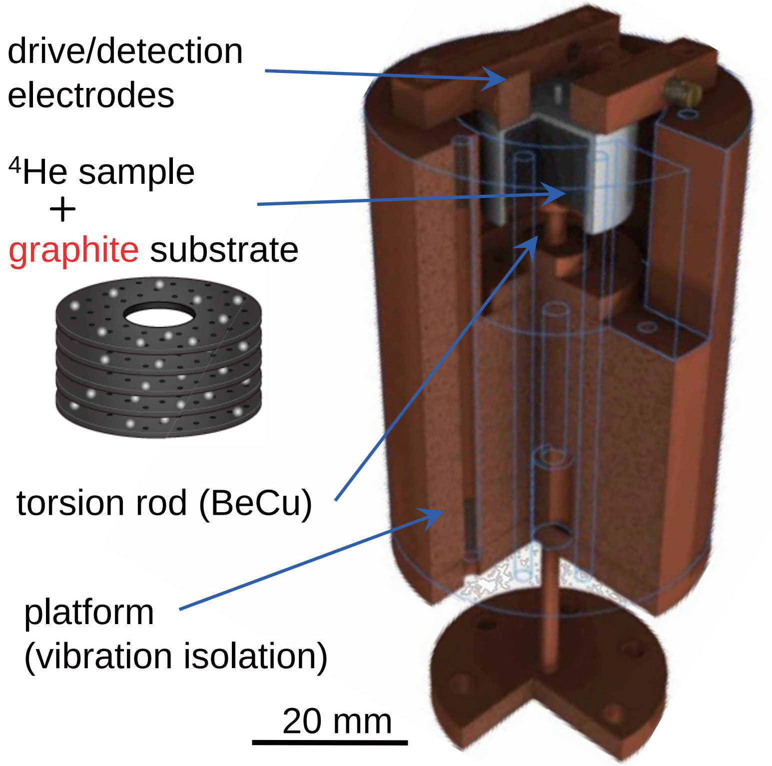
ZYX
(exfoliated ZYH grade HOPG)
2 m²/g, 200 nm

Grafoil
(exfoliated natural graphite flake)
20 m²/g, 10 nm

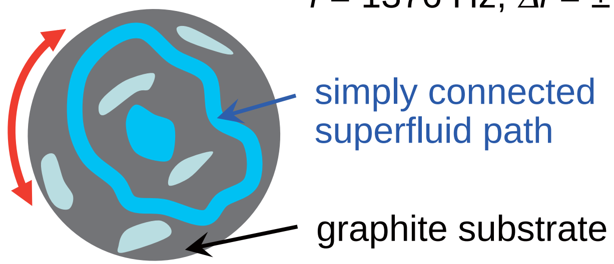




torsional oscillator



- $Q = 3\times10^6$ at $T = 10$ mK
- $f = 1376$ Hz, $\Delta f = \pm 0.1$ mHz

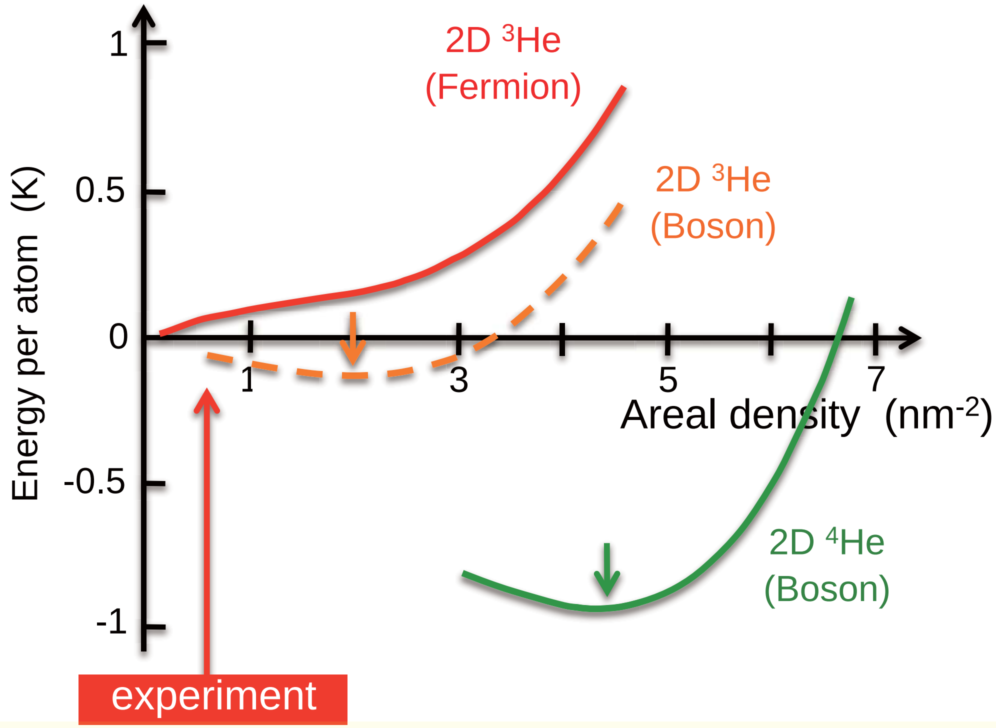


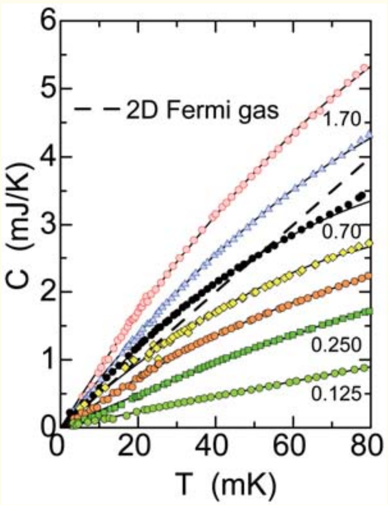
Liquefaction of 2D ^{3}He

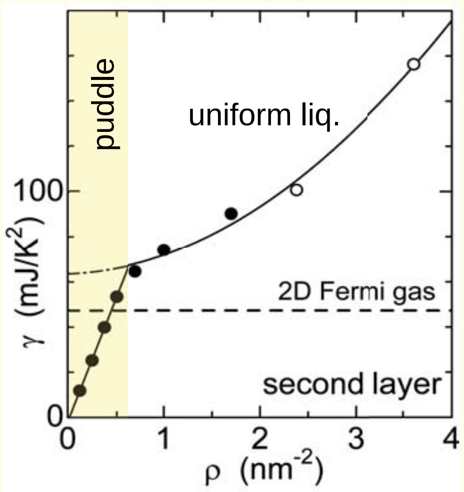
Theoretical predictions

2D ^{3}He is a unique material which **stays quantum gas** at the ground state. (cf. $^\uparrow$H)

VMC	A. D. Novaco and C. E. Campbell, Phys. Rev. B 11, 2525 (1975).
VMC	M. D. Miller and L. H. Nosanow, J. Low Temp. Phys. 32, 145 (1978).
VMC	B. Krishnamachari and G. V. Chester, Phys. Rev. B 59, 8852 (1999).
● DMC	V. Grau, J. Boronat and J. Casulleras, PRL, 89, 045301 (2002)

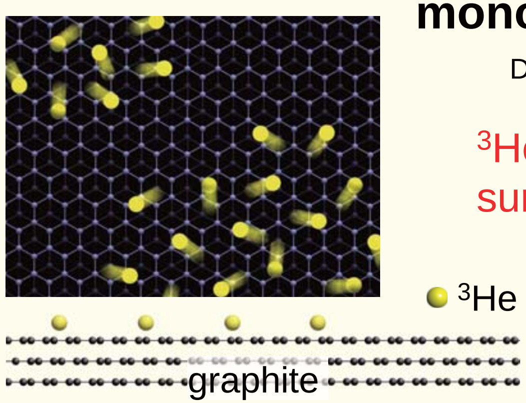






Recent specific heat measurements of monolayer ^{3}He on graphite

D. Sato et al., PRL **109**, 235306 (2012)



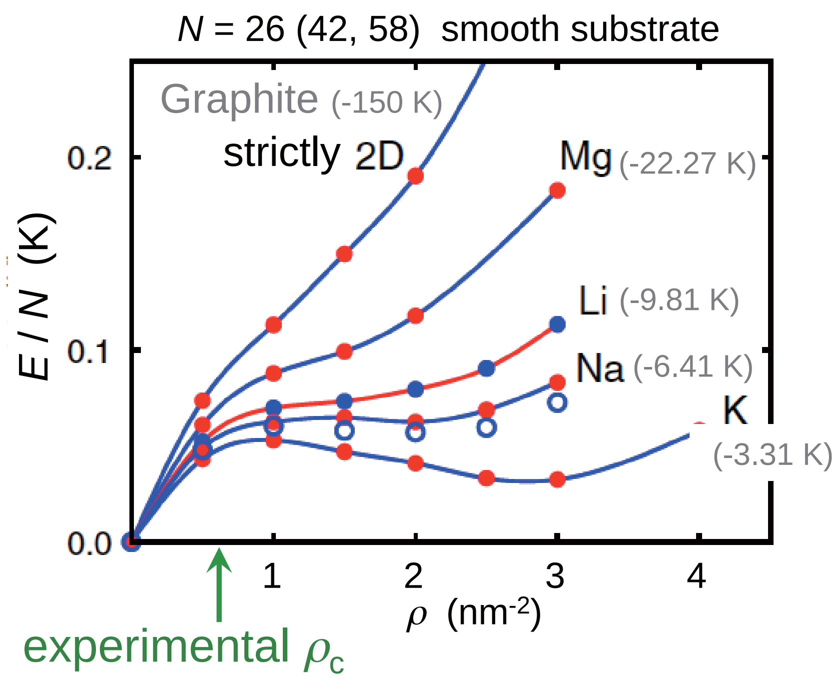
^{3}He atoms liquefy at surprisingly low density:

$\rho_c \approx 0.6$ nm^{-2} ($a \approx 1.4$ nm)

Recent theoretical calculations

New DMC calculation for quasi 2D ^{3}He claimed **no bound state**, again!

M. Ruggeri, S. Moroni, and M. Boninsegni, PRL **111**, 045303 (2013)



- Verified previous results, i.e., no bound state, for strictly 2D.
- Bound states can be **meta-stabilized only on much more weakly attractive substrates** such as Li, Na and K than graphite.

Another approach from clusters:

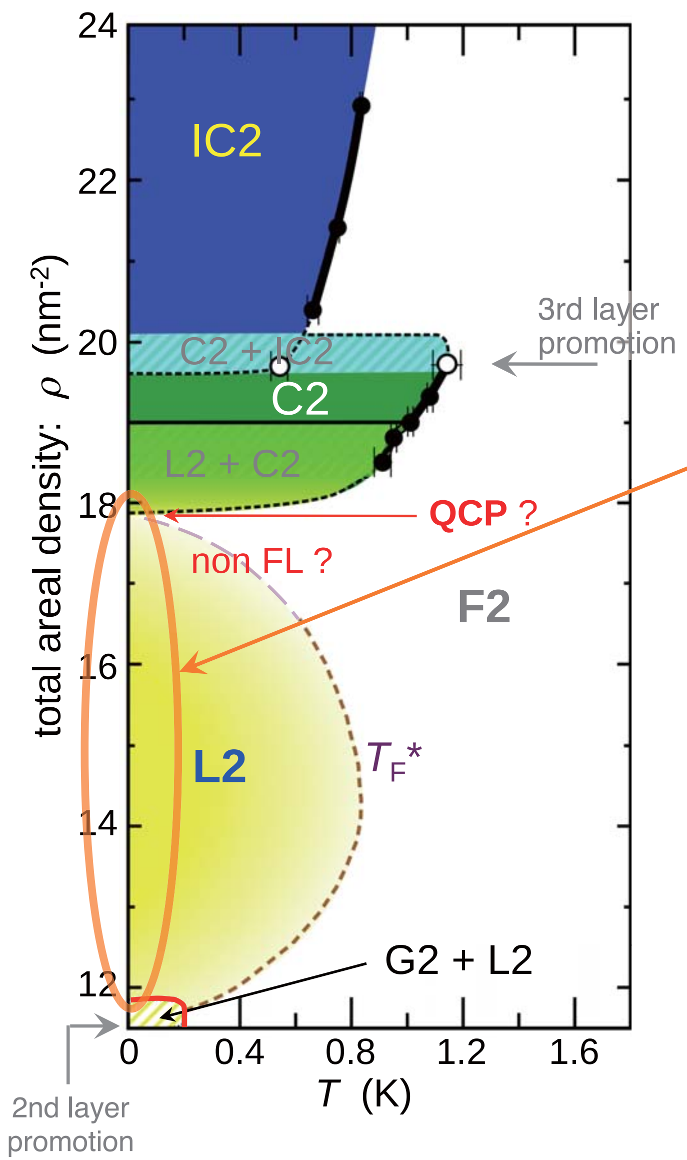
Adiabatic hyperspherical coordinates calculations N. Sakumichi and H. Suno

- Tiny but finite binding energies for N = 2 and 3.
- 2D ^{3}He seems to be self-bound at $N \to \infty$.

Challenge for theorists and experimentalists as well.

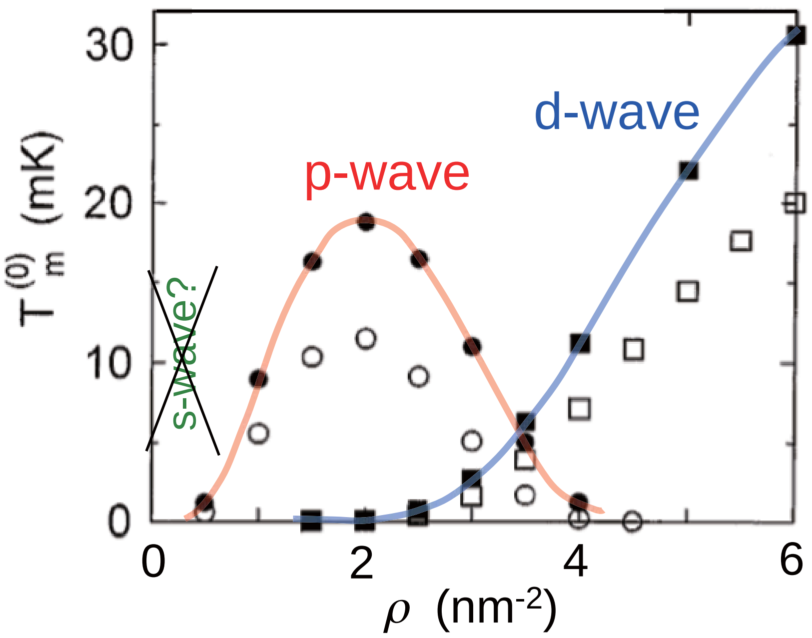
Possible superfluid transitions in monolayer liq. ^{3}He

S. Nakamura et al.,arXiv:1406.4388v1
D. Sato et al., JLTP **158**, 201 (2010); to appear



theoretical predictions

Y. Onishi and K. Miyake, JPSJ **68**, 3927 (1999)
H. Takahashi and D.S. Hirashima, JLTP **121**, 1 (2000)



Wide density range ($0.6 \leq \rho \leq 6$ nm^{-2})

➔ interaction tunable system

Grand challenges in 2D QFS

1. Study and confirmation of quantum liquid crystal
(quantum liquid state, supersolid, etc)

- **Needs direct information on spatial order**
  neutron scattering, synchrotron X-ray scattering, LEED, etc., below 1 K
- **Inconsistency among existing *ab initio* calculations**
  stability of QLC phase (or commensurate phase)
- **Needs better substrate good for transport measurements**
  other graphite than ZYX, ...?

2. Confirmation of liquefaction (many body condensation) **of ^{3}He in 2D**

- **Needs more theoretical efforts**

3. Search for possible superfluidity of monolayer ^{3}He
(mechanism?, *p*-wave → *d*-wave transition?)