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Miniworkshop on Strong Correlations in Materials and Atom Traps

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Frustrated magnetism and strong correlation effects in 2D He-3

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Frustrated Magnetism and Strong Correlation Effects in 2D ^{3}He

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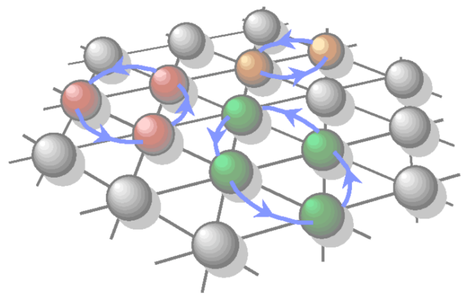
1. Introduction (2D ^{3}He systems)
2. Gapless spin-liquid state in the 4/7 phase
3. Possible vacancy (hole) doping into the 4/7 phase
4. Particle doping into the 4/7 phase
5. Summary



Interaction ranges in He and electron systems

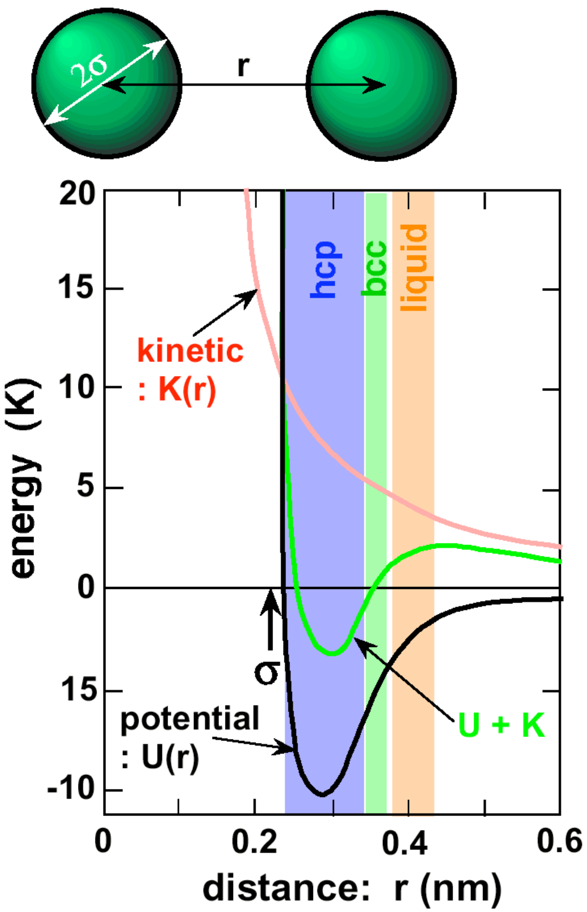
He systems

short-range repulsion: $U_{\mathrm{L-J}}(r) = 4\varepsilon\left\{\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right\}$

Kinetic energy:

- large zero-point energy $K(r) = \frac{\hbar^2}{2mr^2}$

stronger correlations at **higher** densities: $\frac{U(r)}{K(r)} \propto r^{-10}$



Electron systems

long-range repulsion: $U_{\mathrm{coulomb}}(r) = \left(\frac{1}{4\pi\varepsilon_0}\right)\frac{e^2}{r}$

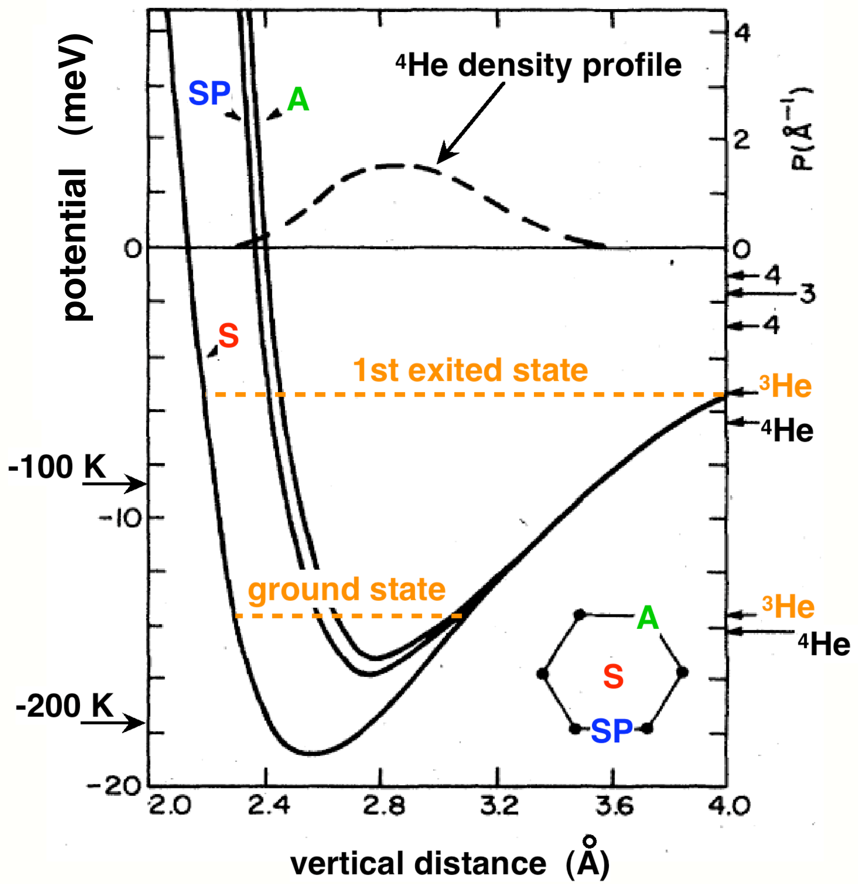
stronger correlations at **lower** densities: $\frac{U(r)}{K(r)} \propto r$

§1. 2D ^{3}He systems
(1st and 2nd layers on graphite)

Nearly ideal 2D systems adsorbed on atomically flat graphite surface

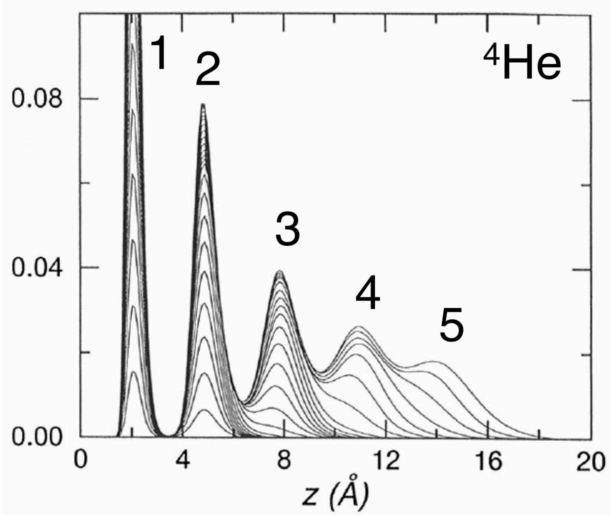
2D Helium adsorbed on graphite

He-graphite potential



M.W. Cole et al., Rev. Mod. Phys. **53**, 199 (1981)

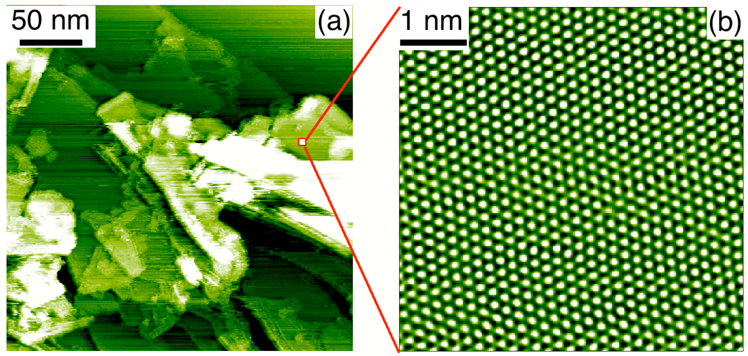
Layer-by-layer growth of He monolayers on atomically flat graphite surfaces



Calculated density profile

STM images of **Grafoil** substrate (exfoliated graphite)

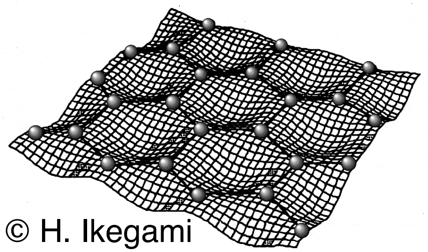
Y. Niimi et al., PRB. **73**, 085421 (2006)



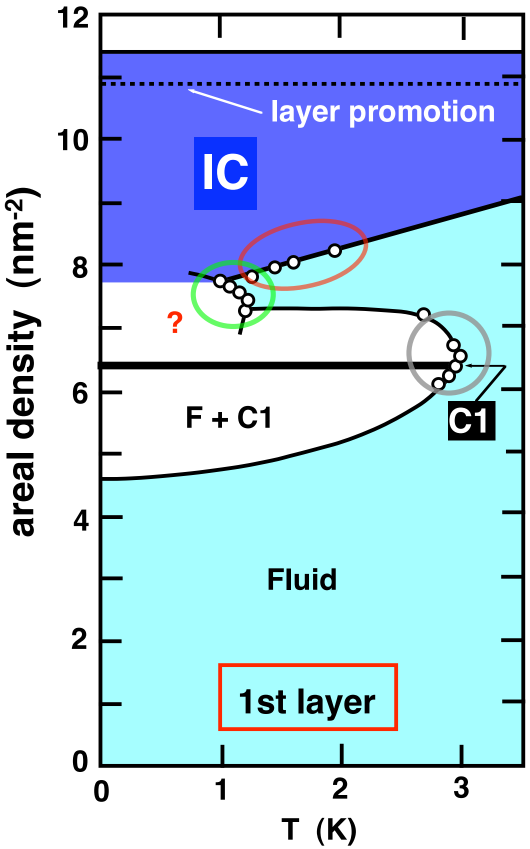
A = 18 m^2/g

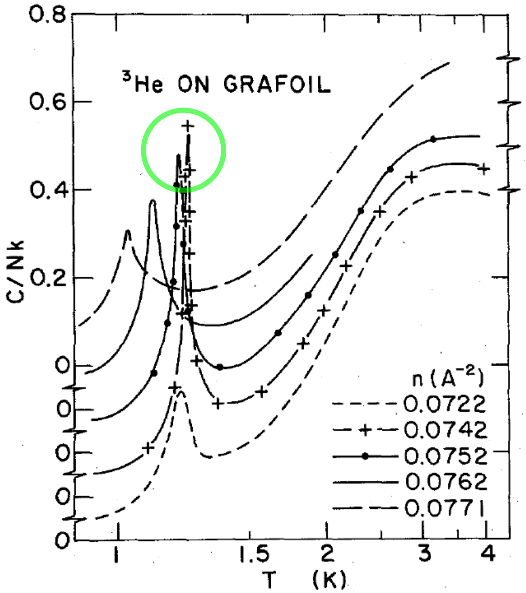
single crystallite size = 10-40 nm

Finite-*T* phase diagram of 1st layer ^{3}He on graphite

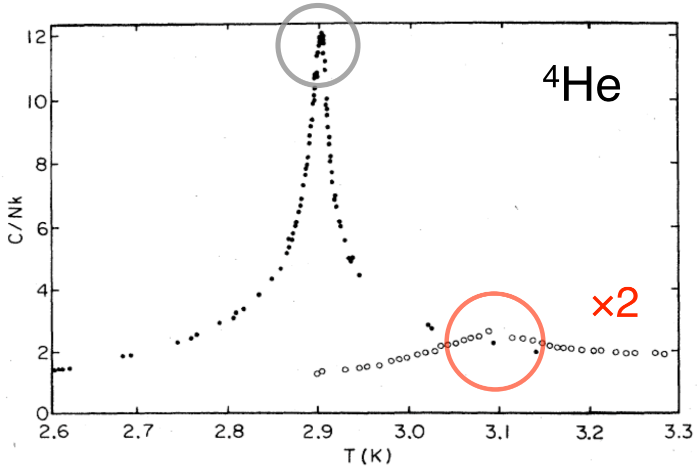


potential corrugation





S.V. Hering et al., JLTP **25**, 793 (1976)



J.H. Campbell and Bretz, PRB **32**, 2861 (1985)

2D melting

- IC solid ⇆ fluid
- KT-like transition

C-IC transition

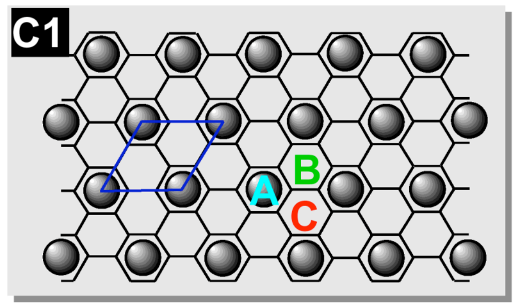
- striped domain wall melting?

Order-disorder transition

- $\sqrt{3}\times\sqrt{3}$ ⇆ fluid
- 3-states Potts model

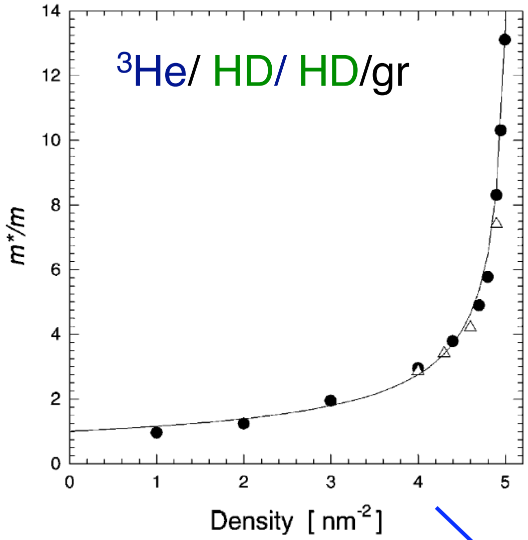
$$C = A\left|(T - T_c)/T_c\right|^{-1/3} + B$$

$\sqrt{3}\times\sqrt{3}$ commensurate phase



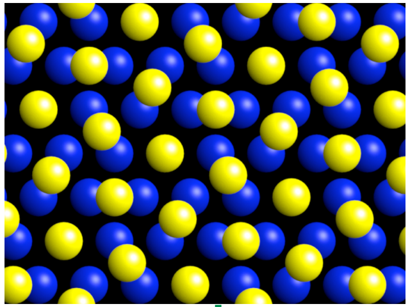
Finite-*T* phase diagram of 2nd layer ^{3}He on graphite

Mott-Hubbard transition



Diverging m^* towards localization

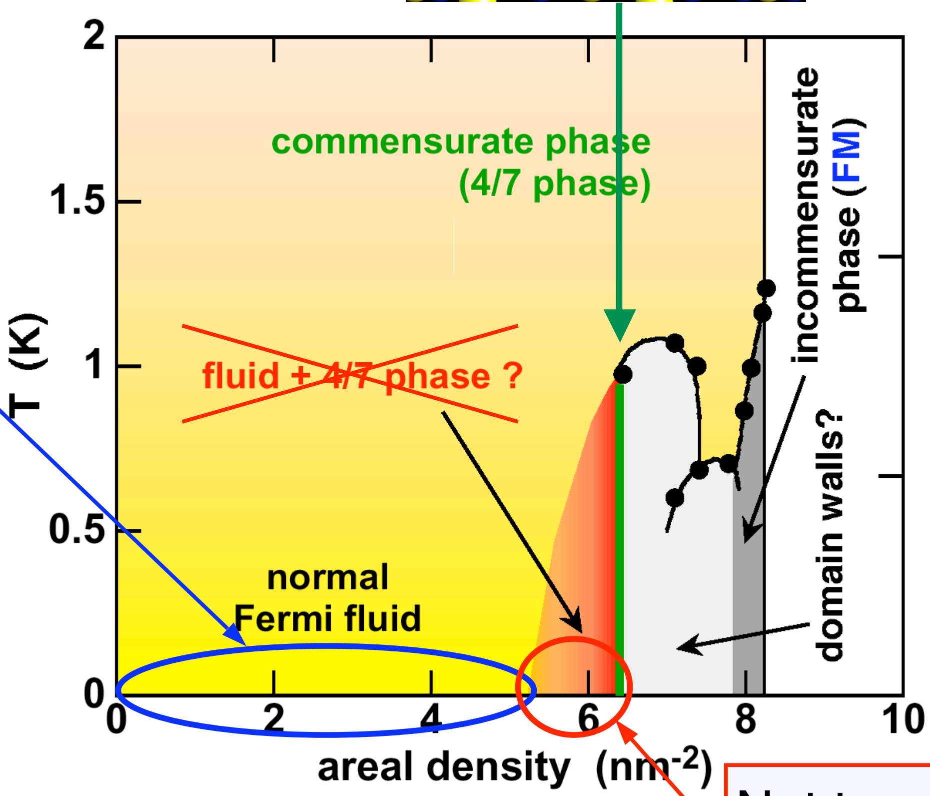
A. Casey et al., PRL **90**, 115301 (2003)



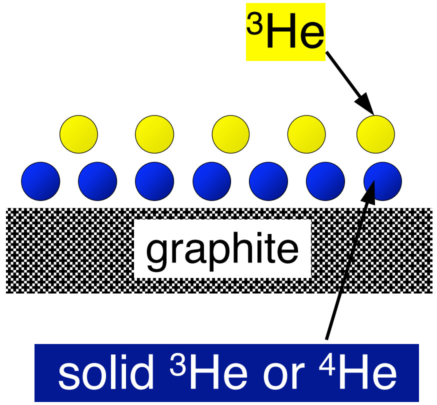
4/7 phase

low-density quantum solid with a triangular lattice

Elser, PRL **62**, 2405 (1989)
Piece&Manousakis, PRB **59**, 3802 (1999)



D.S. Greywall, PRB **41**, 1842

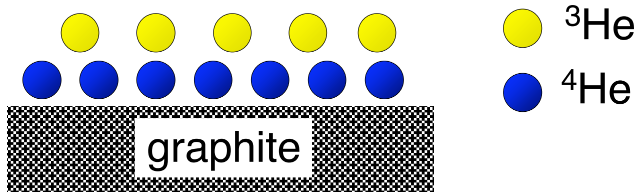


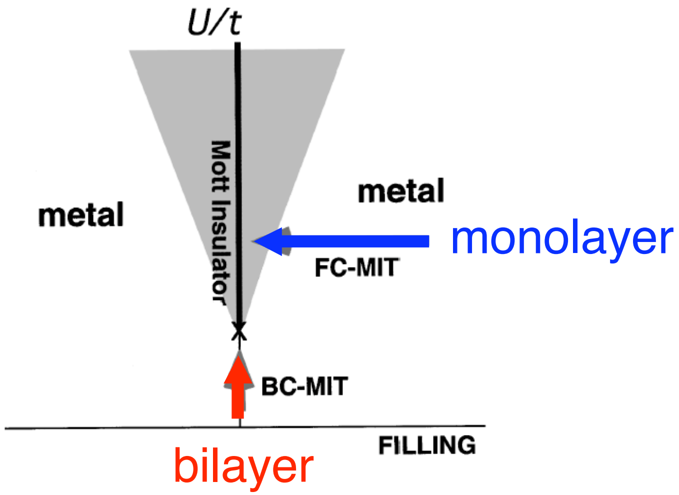
Not two-phase coexistence, but 4/7 phase doped with ZPV

Variety of 2D ^{3}He systems

^{3}He monolayer on ^{4}He monolayer (our system)

^{3}He monolayer localizes without adding overlayer (filling control)

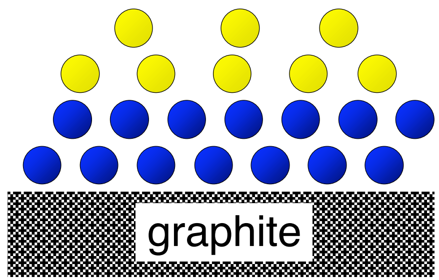


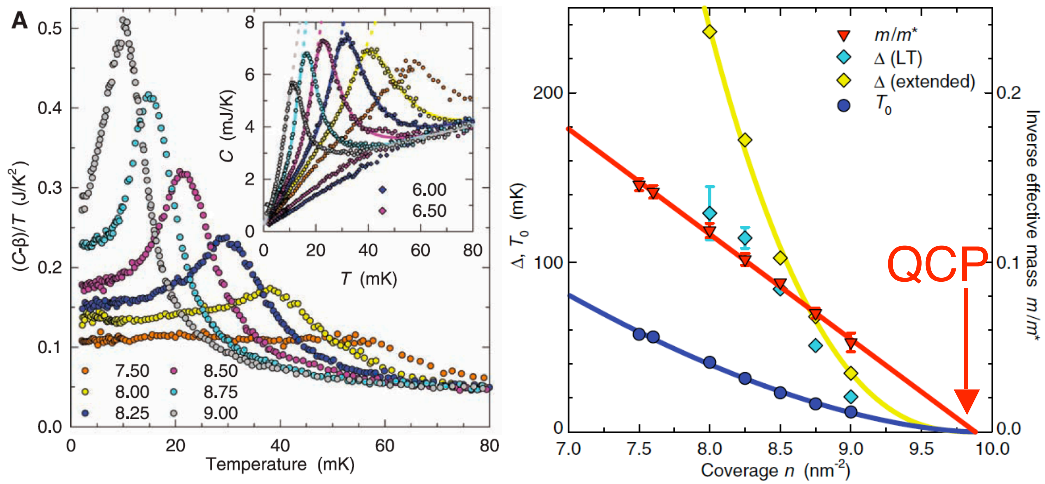


^{3}He bilayer on ^{4}He bilayer

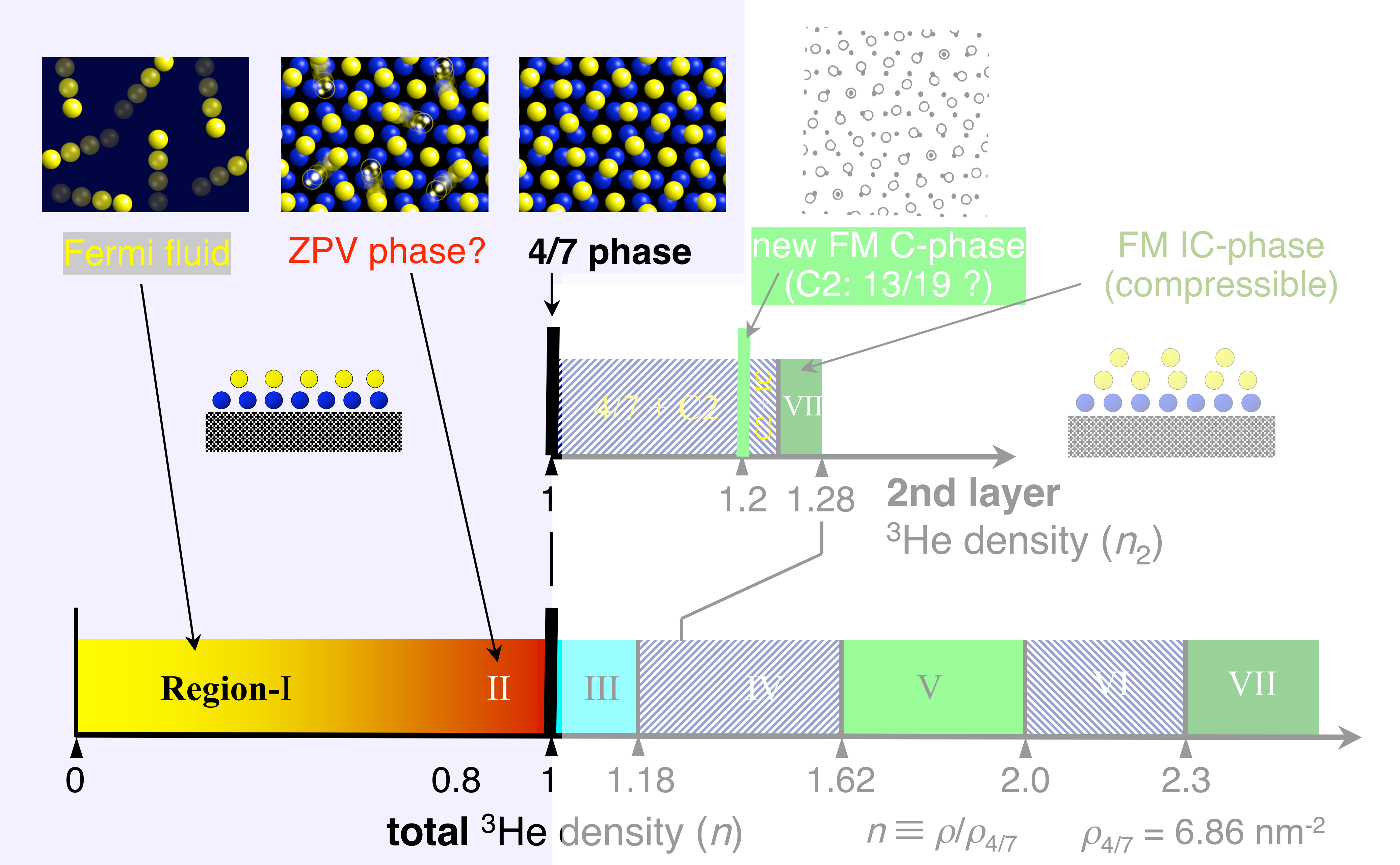
M. Neumann et al., Science **317**, 1356 (2007)

^{3}He underlayer localizes only by adding ^{3}He overlayer (band width control)





$T = 0$ phase diagram of 2D ^{3}He (2nd layer)

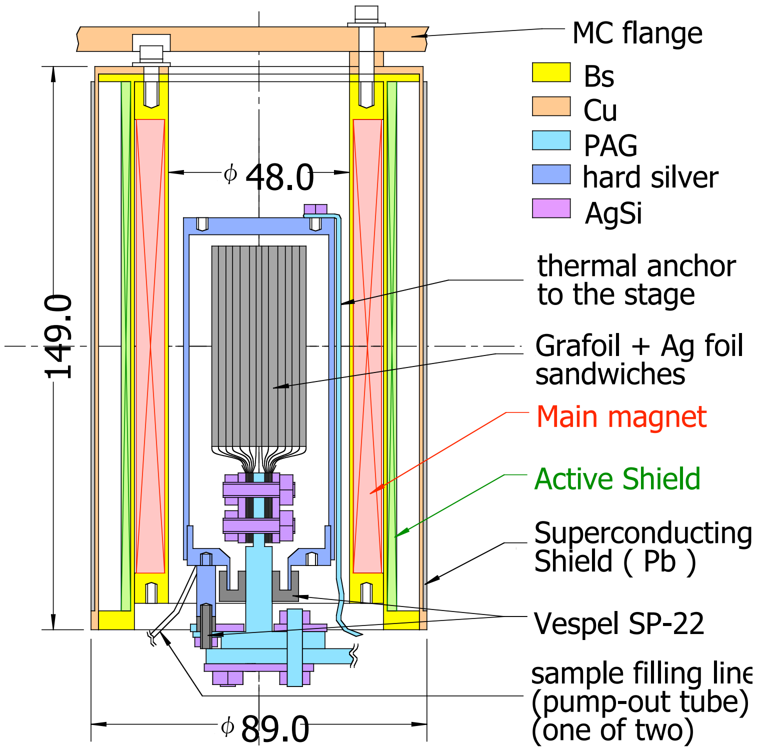


Experimental setup

Heat capacity cell

Y. Matsumoto et al.,
Physica B **329-333**, 146 (2003)

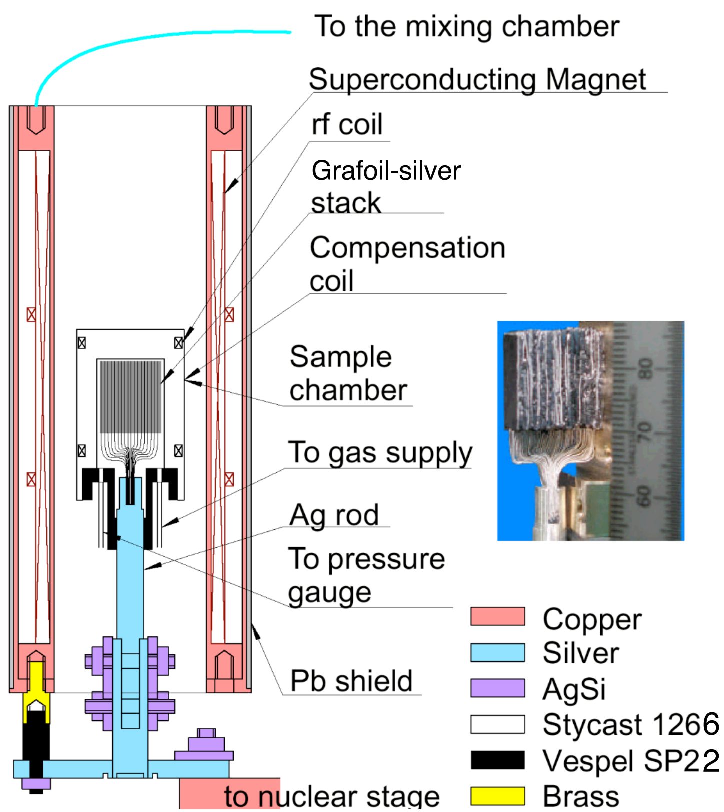
$100\ \mu K \le T \le 80\ mK$
$0 \le B \le 1.2\ T$



NMR cell

S. Murakawa et al.,
Physica B **329-333**, 144 (2003)

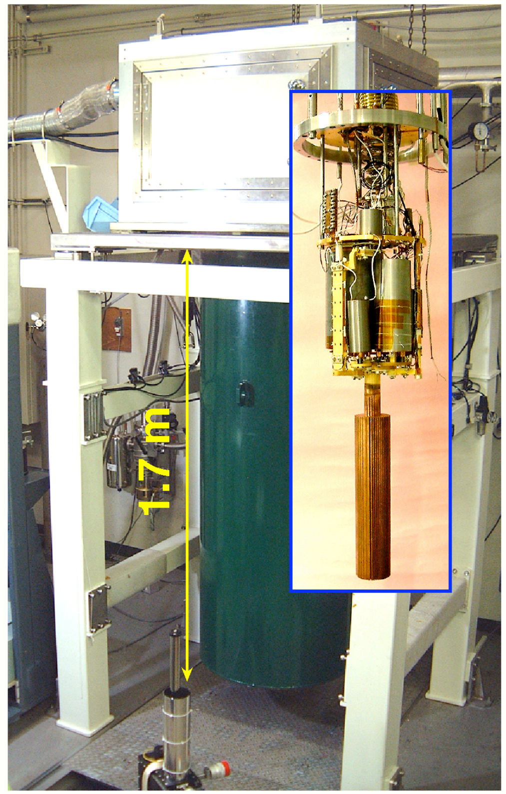
$60\ \mu K \le T \le 2\ K$
$0 \le B \le 1.17\ T$



Nuclear demag. fridge

Y. Matsumoto et al.,
JLTP **134**, 61 (2004)

- T_{min} = 51 μK
- holding $T \le 200\ \mu K$ for one week



§2. Gapless spin-liquid state in the 4/7 phase

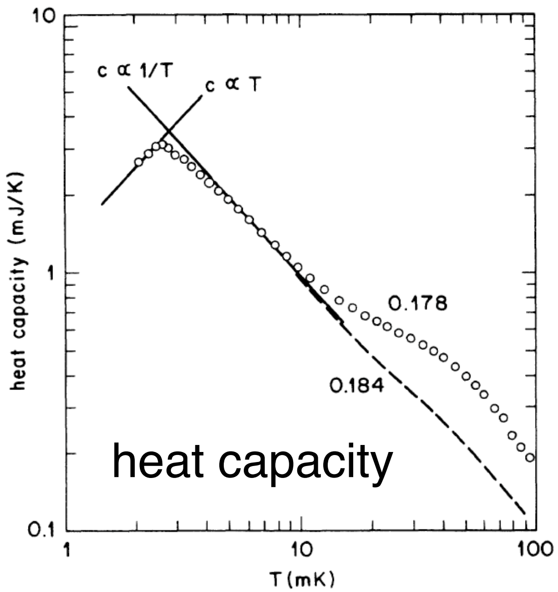
Highly frustrated quantum spin system (S =1/2) on a triangular lattice

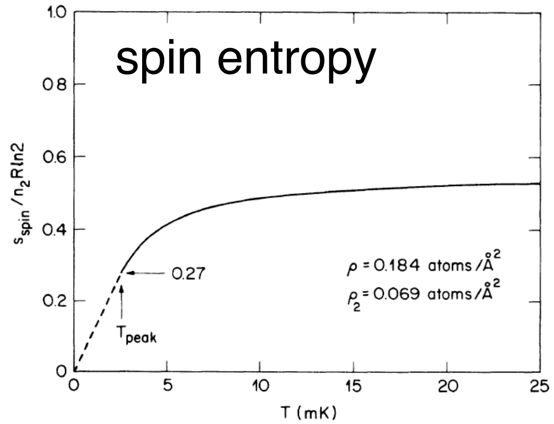
Frustrated magnetism of the 4/7 phase (earlier works)

Heat capacity measurements

D.S. Greywall, PRL **62**, 1868 (1989)

- A round maximum at $T \approx 2$ mK.
- Missing entropy ($\approx 0.5\ln 2$)?
- Highly frustrated antiferromagnet



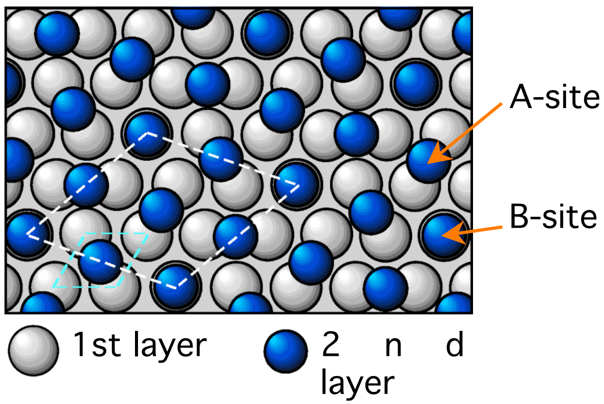


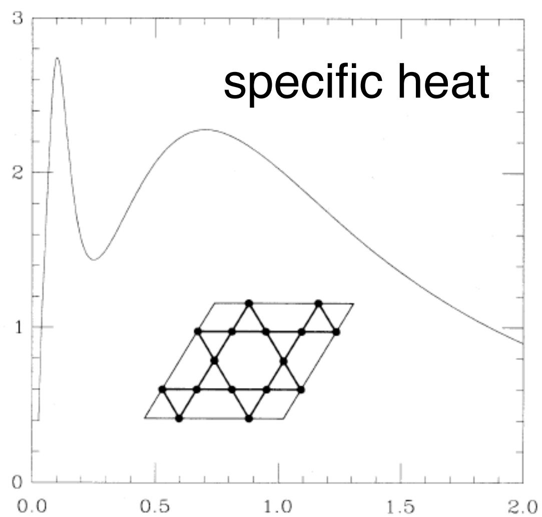
Exact diagonalization and PIMC calculations

V. Elser, PRL **62**, 2405 (1989)

- $\sqrt{7}\times\sqrt{7}$ structure (**4/7 phase**)
- S = 1/2 Heisenberg AF on a **Kagome** lattice (HAFK) model

$$H = J \sum_{i<j}^{A(\mathrm{N.N.})} S_{\mathrm{i}} \bullet S_{\mathrm{j}} \qquad |J_{\mathrm{AB}}| << |J_{\mathrm{AA}}|$$

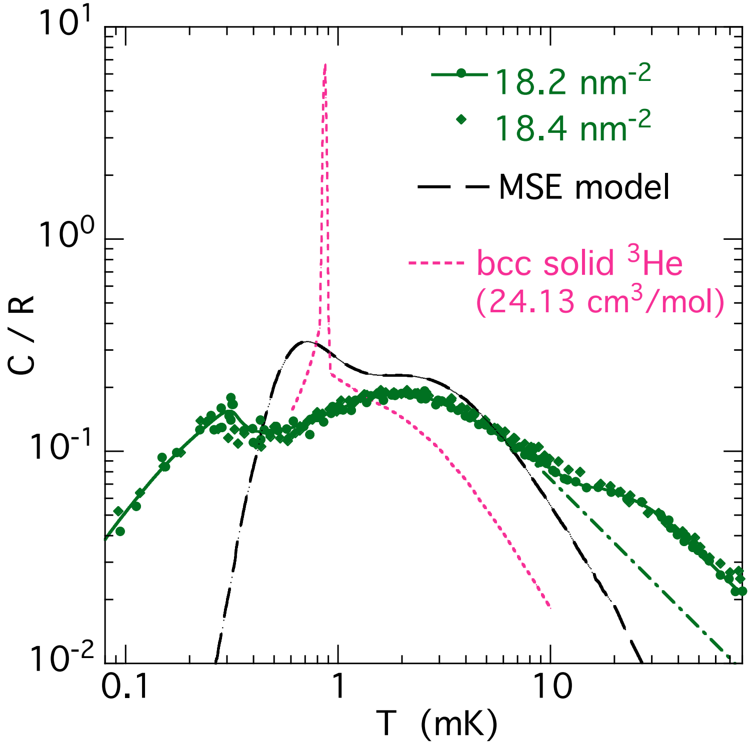




Gapless spin-liquid behaviour in 4/7 phase

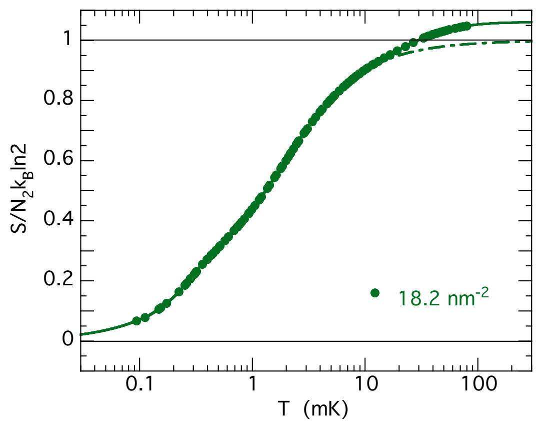
- Absence of finite-*T* phase transitions … truly 2D
  down to $T/J \approx 10^{-2}$-10^{-3} (J = 1-10 mK)
- Double peak in $C(T)$ … highly frustrated
- No exponential behaviour at $T \ll J$ … gapless excitation

Specific heat



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

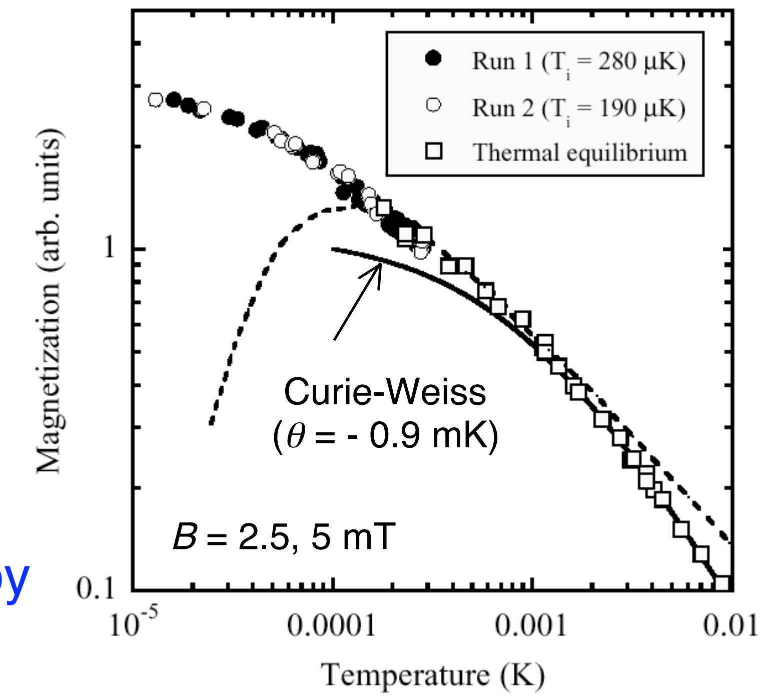
entropy



- $\Delta C(T) \approx N_2 k_B \ln 2$

… no missing entropy

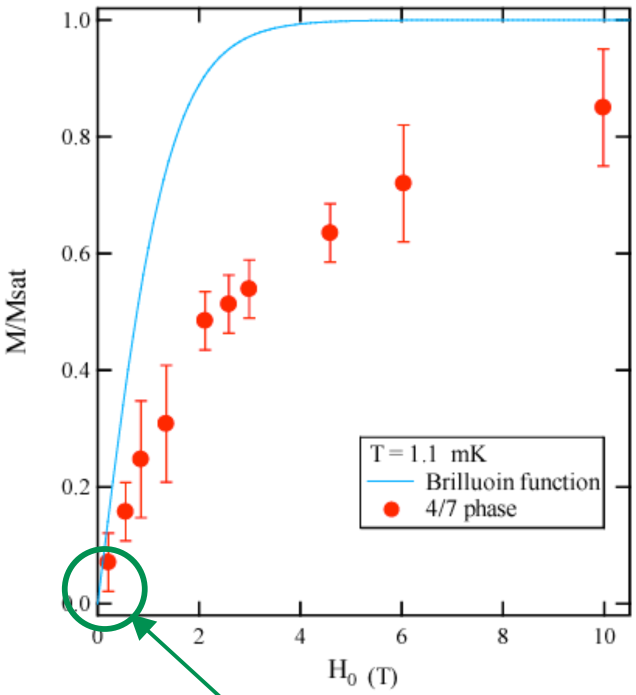
magnetization



R. Masutomi, et al., PRL **92**, 025301 (2004)

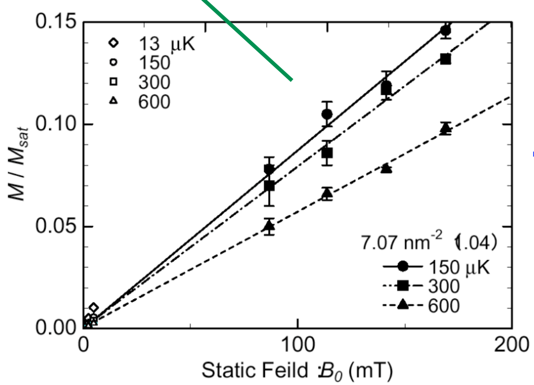
Magnetization curve of the 4/7 phase

Magnetization does not saturate even at $B = 10$ T (= $10\,T_{C\text{-peak}}$).



A. Yamaguchi et al., JLTP **148**, 755 (2007)

Highly frustrated 2D AFM



Truly gapless

S. Murakawa et al., PhD thesis (Univ. of Tokyo, 2006)

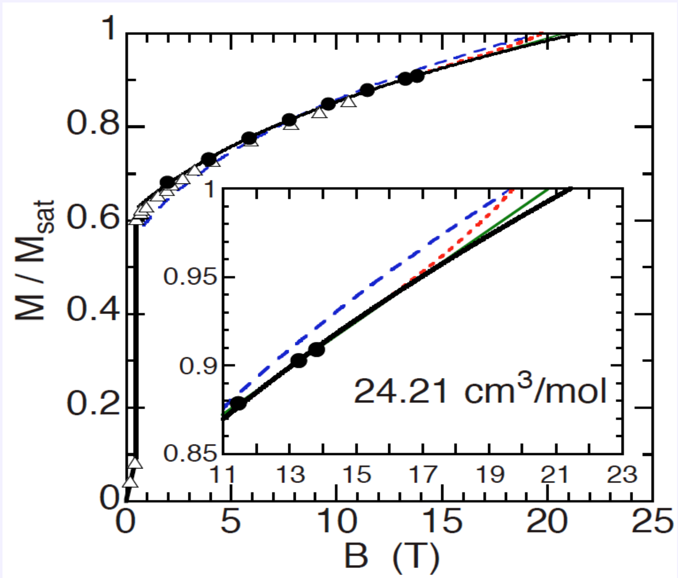
3D solid ^{3}He (bcc)

Highly frustrated 3D AFM

$k_B T_N << \mu B_{c2}$

T_N ($B = 0$) = 0.93 mK

B_{c2} ($T = 0$) ≈ 20 T



△ D.D. Osheroff et al., PRL **58**, 2458 (1987)

● H. Fukuyama et al., to be published

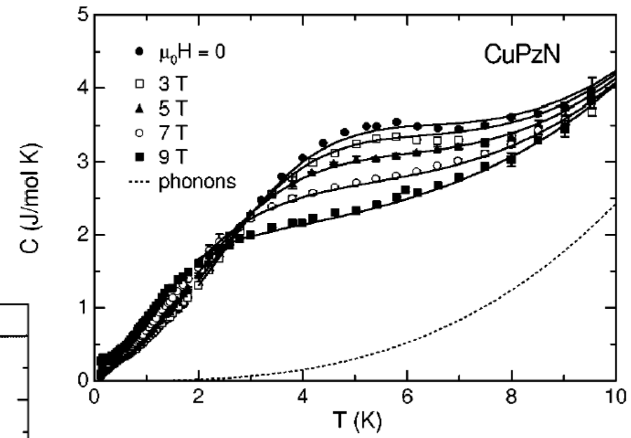
Gapless QSLs found in other materials

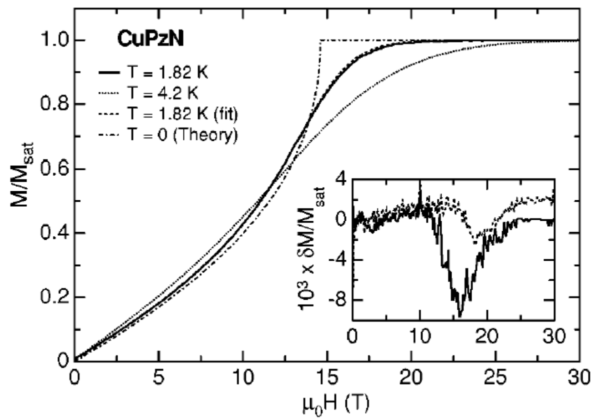
1D AFM Heisenberg chain (S = 1/2)

Copper pyrazine dinitrate: $[Cu(C_4H_4N_2)(NO_3)_2]$

P.R. Hammar et al. (PRB **59**, 1008 (1999))

- No LRO down to 0.1 K: $J = 11$ K, $J'/J < 10^{-4}$
- $C(T) \propto T$ at low T (spinons?)



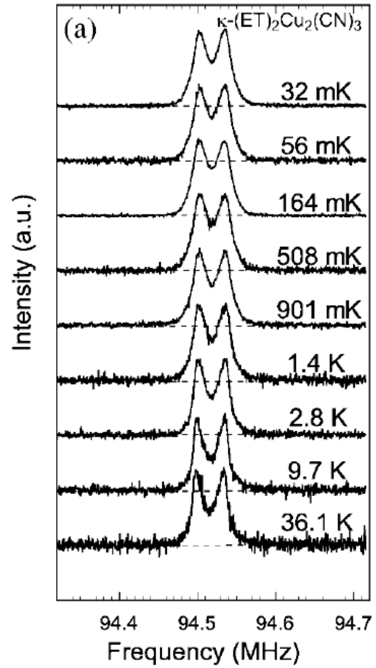


2D Heisenberg spins on triangular lattice (S = 1/2)

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$: organic Mott insulator

Y. Shimizu, K. Kanoda et al. (PRL **91**, 107001 (2003))

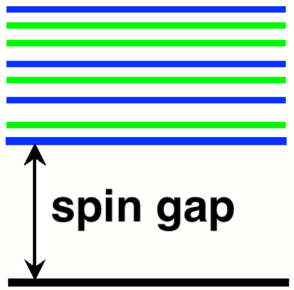
- No LRO down to 32 mK: $J = 250$ K, $J'/J < 10^{-4}$
- $\tau_1^{-1} \propto T, T^2$ (two components)



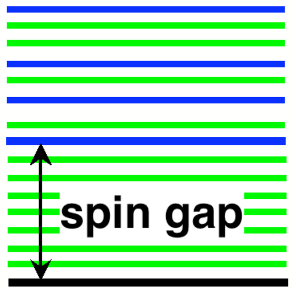
A variation of spin liquids

Gapped spin liquids

class1



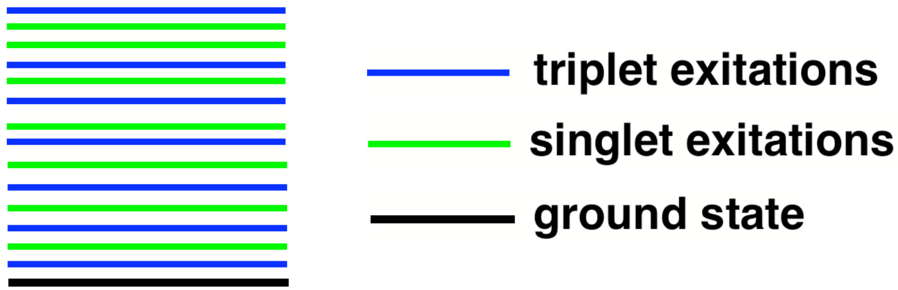
class2



- RVB

- kagome lattice
- Δ-chain

Gapless spin liquids



- **2D ^{3}He**
- $Cu(C_4H_4N_2)(NO_3)_2$
- κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

Multiple-spin exchange model -- general

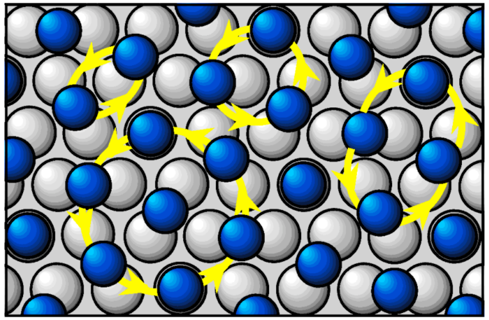
Many-body interactions are essentially important in hardcore quantum solids due to the steric hindrance.

D.J. Thouless, Proc. Phys. Soc. **86**, 893 (1965), *ibid*. **86**, 905 (1965)
M. Roger, J.H. Hetherington and J.M. Delrieu, Rev. Mod. Phys. **55**, 1 (1983)

$$H = \sum_n (-1)^n J_P \left(P_n + P_n^{-1} \right)$$

P_n, P_n^{-1}: cyclic permutation operator of n particles

All $J_P > 0$



2-spin exchange: $P_2 = P_{i,j} = 2S_i \bullet S_j + \frac{1}{2}$ (AFM)

3-spin exchange: $P_3 = P_{1,2,3} + P_{1,2,3}^{-1} = P_{1,2} + P_{2,3} + P_{3,1} - 1$ (FM)

4-spin exchange: $P_4 = P_{1,2,3,4} + P_{1,2,3,4}^{-1} = P_{1,2}P_{3,4} + P_{1,4}P_{2,3} - P_{1,3}P_{2,4} + P_{1,3} + P_{2,4} - 1$ (AFM)

5-spin exchange: $P_{1,2,3,4,5} + P_{1,2,3,4,5}^{-1} = P_{1,2}P_{3,4} + P_{1,4}P_{2,3} - P_{1,3}P_{2,4} + P_{1,3} + P_{2,4} - 1$ (FM)

$$H = J\sum_{i<j} \left(S_i \bullet S_j \right) + K \sum_{i<j<k<l} \left\{ \left(S_i \bullet S_j\right)\left(S_k \bullet S_l\right) + \left(S_i \bullet S_l\right)\left(S_j \bullet S_k\right) - \left(S_i \bullet S_k\right)\left(S_j \bullet S_l\right) \right\}$$
$$+ J_6 \sum_{i<j<k<l<m<n} \left\{ \left(S_i \bullet S_j\right)\left(S_k \bullet S_l\right)\left(S_m \bullet S_n\right) + \left(S_j \bullet S_k\right)\left(S_l \bullet S_m\right)\left(S_n \bullet S_i\right) + \cdots \right\}$$

$J = J_2 - 2J_3$: effective 2-spin exchange (< 0)

$K = J_4 - 2J_5$: effective 4-spin exchange (> 0)

WKB calculations of MSE frequencies

M. Roger, PRB **30**, 6432 (1984)

2D systems with r^{-12} potential

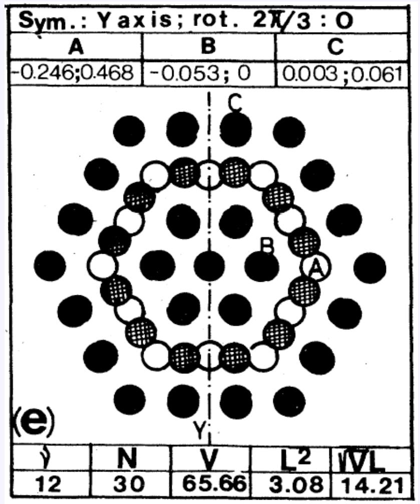
$$J_P = C_P s_P \exp(-A_P/g), \quad A_P \approx L\sqrt{V_{\max}}$$

$$\Gamma(J_P) \equiv \frac{\partial \ln J_P}{\partial \ln V} \approx \frac{5A_P}{3g}$$

$$J_3 > J_4 \approx J_6 > J_2$$

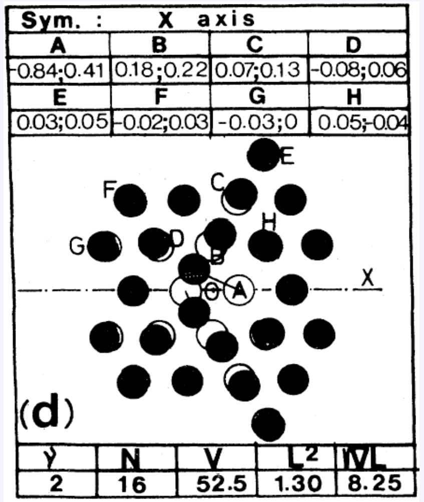
Twelve-spin

$V_{\max} = 65.7$
$L^2 = 3.08$
$\boldsymbol{A_P = 14.21}$
$N = 30$



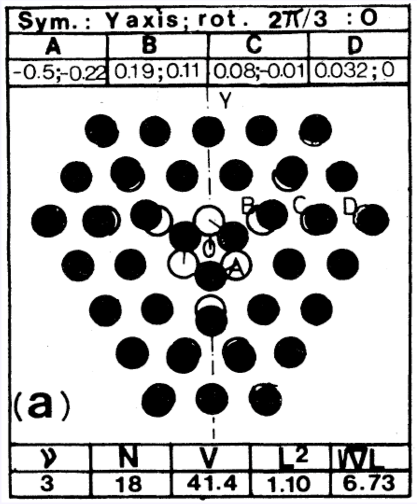
Two-spin

$V_{\max} = 52.5$
$L^2 = 1.30$
$\boldsymbol{A_P = 8.25}$
$N = 16$



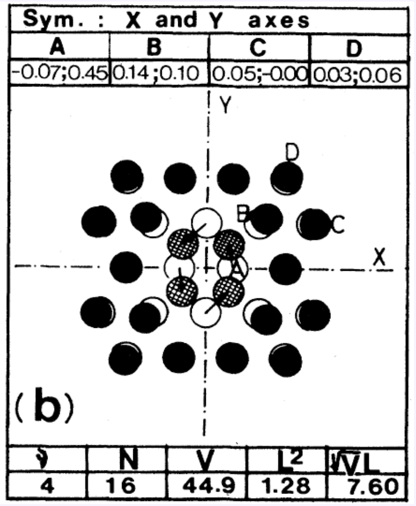
Three-spin

$V_{\max} = 41.4$
$L^2 = 1.10$
$\boldsymbol{A_P = 6.73}$
$N = 18$



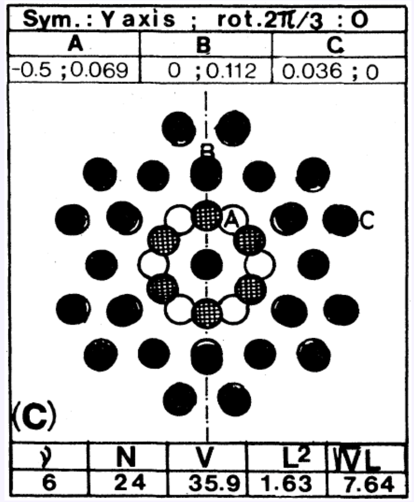
Four-spin

$V_{\max} = 44.9$
$L^2 = 1.28$
$\boldsymbol{A_P = 7.60}$
$N = 16$



Six-spin

$V_{\max} = 35.9$
$L^2 = 1.63$
$\boldsymbol{A_P = 7.64}$
$N = 24$

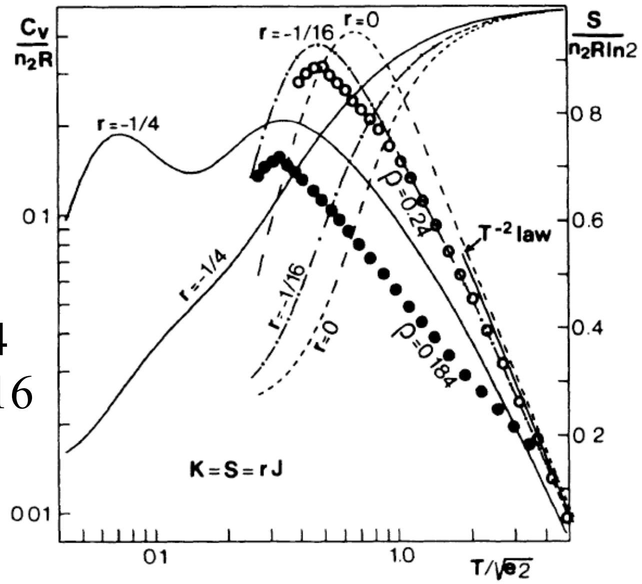


Multiple-spin exchange model for 2D ^{3}He

M. Roger, PRL **64**, 297 (1990)

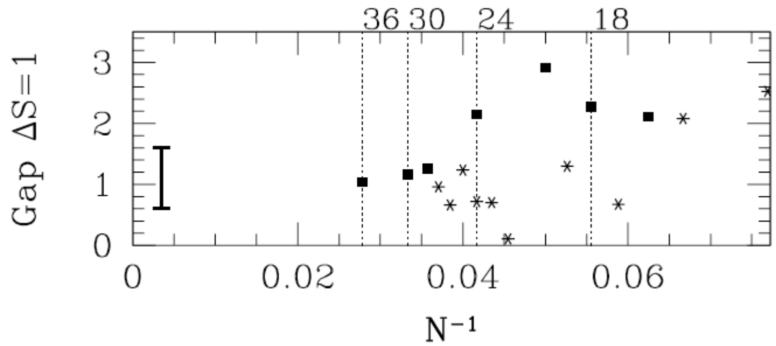
- Exact diagonalization of 16 spins
- frustration caused by competing interactions, i.e., J (< 0) and K (> 0), and geometrical frustration (triangular).
- can explain density dependence
- double peak in specific heat

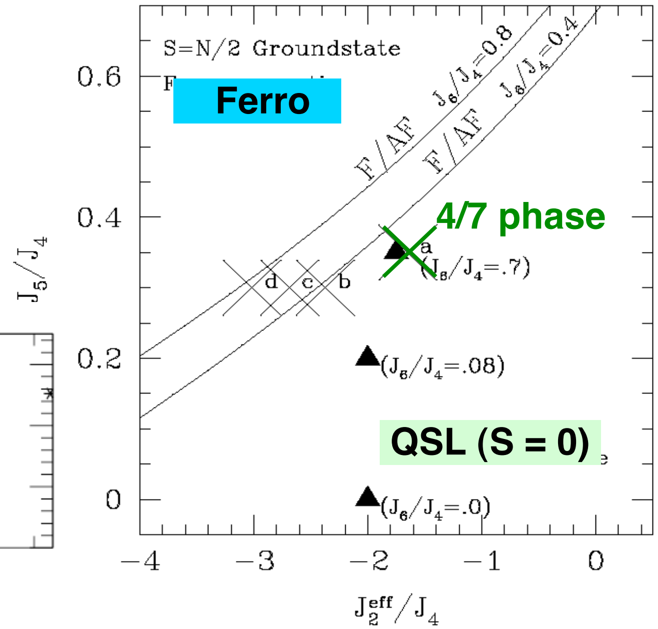
$r \equiv K/J = S/J = -1/4$ (solid line)
$= -1/16$ (dash-dot line)
$= 0$ (dashed line)



G. Misguich et al., PRL **81**, 1098 (1998)

- Exact diagonalization of 36 spins
- taking account of up to J_6
- gapped spin liquid next to FM phase
- double peak in specific heat

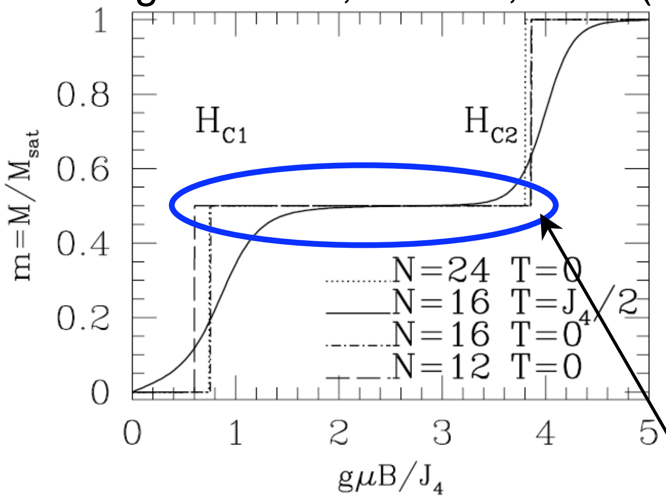




Theoretical magnetic phase diagram of 2D ^3He

Predictions of a **plateau at $M = (1/2)M_{sat}$**

G. Misguich et al., PRL **81**, 1098 (1998))

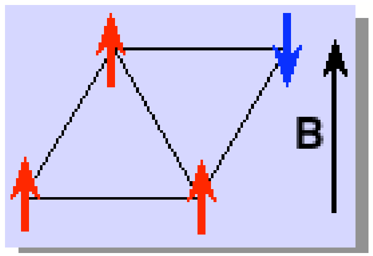


$J/J_4 = -2$

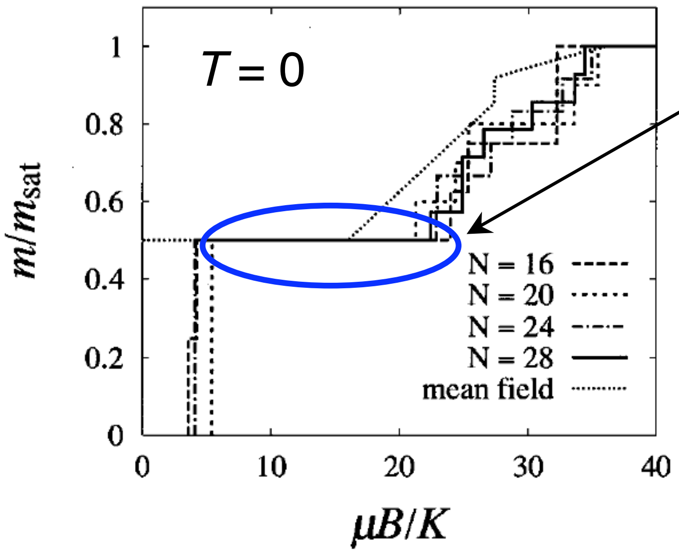
$J_5/J_4 = 0.2$

$J_6/J_4 = 0.08$

LRO to the **uuud** phase in magnetic fields



T. Momoi et al., PRB **59**, 9491 (1999)



$K = J_4$

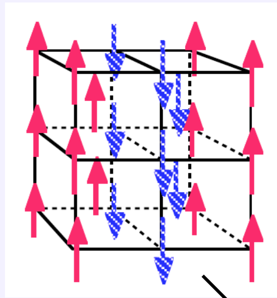
$J/J_4 = 0$

$J_5/J_4 = 0$

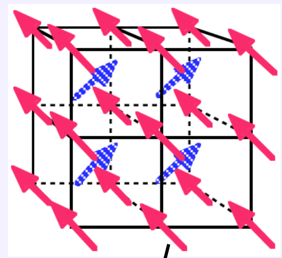
$J_6/J_4 = 0$

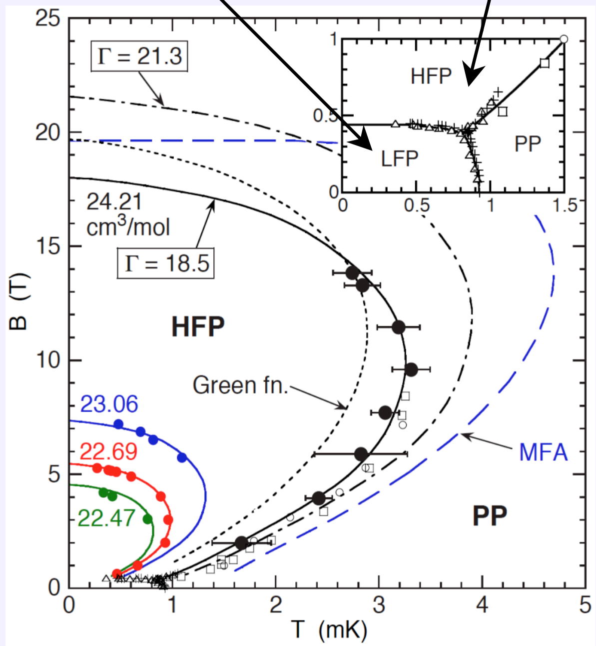
3D solid ^3He (bcc)

UUDD phase (LFP)



CNAF phase (HFP)





H. Fukuyama et al., Physica B **169**, 197 (1991); to be published

Theoretical approaches to gapless QSL in 4/7 phase

Elementary excitation?

spinon   (e.g., magnon in systems with LRO)

Effective Hamiltonian?

Ring exchange

$$H_{\mathrm{eff}} = \sum_P (-1)^P J_{\mathrm{P}} P$$

Momoi, Kubo

↓

hidden orders

Hubbard

$$H = -t \sum_{\langle i,j \rangle, \sigma} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

Imada, Watanabe

↓

density fluctuations

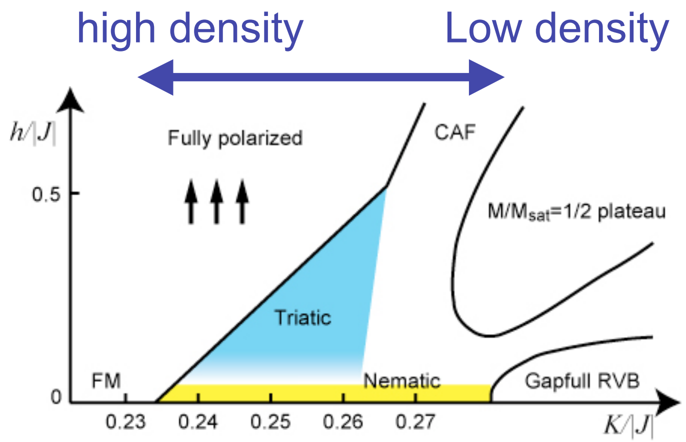
t-J

$$H = -t \sum_{\langle i,j \rangle, \sigma} P\left(c^\dagger_{i\sigma} c_{j\sigma} + h.c.\right)P + J \sum_{\langle i,j \rangle} S_i \bullet S_j$$

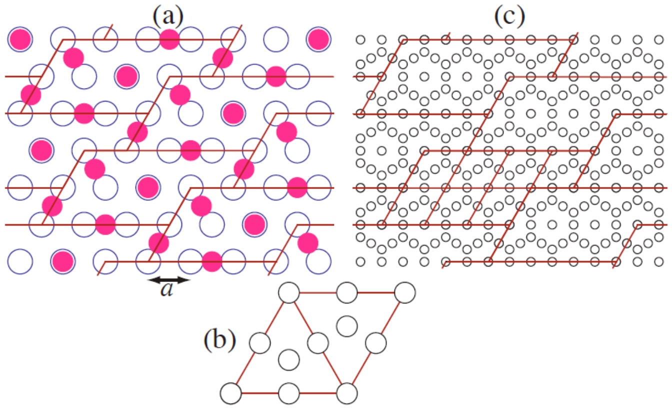
Ogata, Koretsune

↓

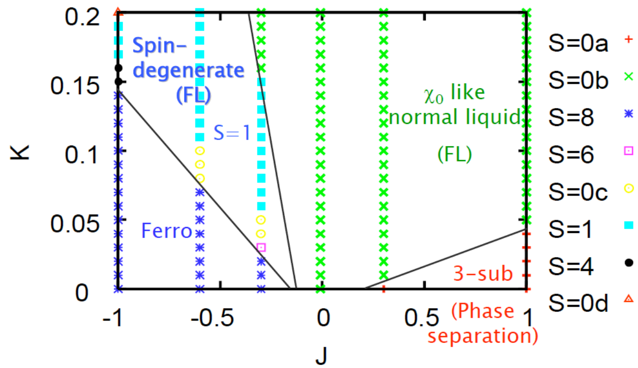
***t-J-K* model**



T. Momoi et al., PRL **97**, 257204 (2007)



S. Watanabe and M. Imada, JPSJ **76**, 113603(2007)



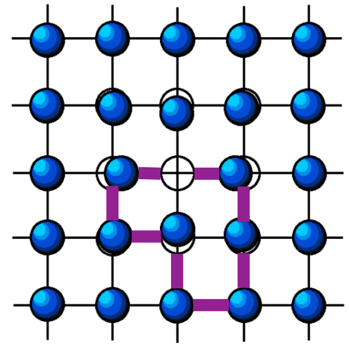
Y. Fuseya and M. Ogata, To bepublished

§3. Possible vacancy (hole) doping into the 4/7 phase

Hypothesis of zero-point vacancy (ZPV) phase

ZPVs in solid He -- holes in quantum solids

1. In 3D solid He



So far, no experimental evidence

M.W. Meisel, Physica B **178**, 121 (1990)

Theories (PIMC, SWF) contradict each other.

Prediction of supersolidity due to BEC of ZPVs.

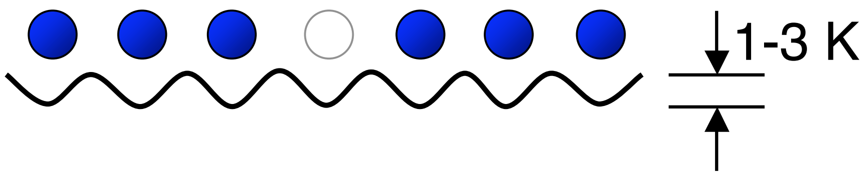
A.F. Andreev and I.M. Lifshitz, Sov. Phys. JETP **29**, 1107 (1969)

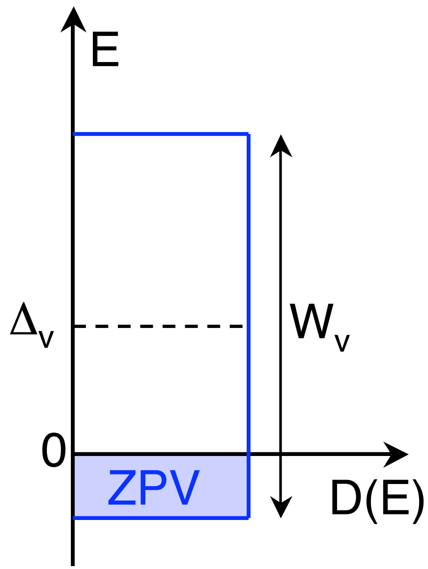
^{3}He *impuriton* … experimentally observed

2. ZPV may be more favorable in 2D

Substrate potential corrugation may decrease Δ_v and increase t due to much lower density.

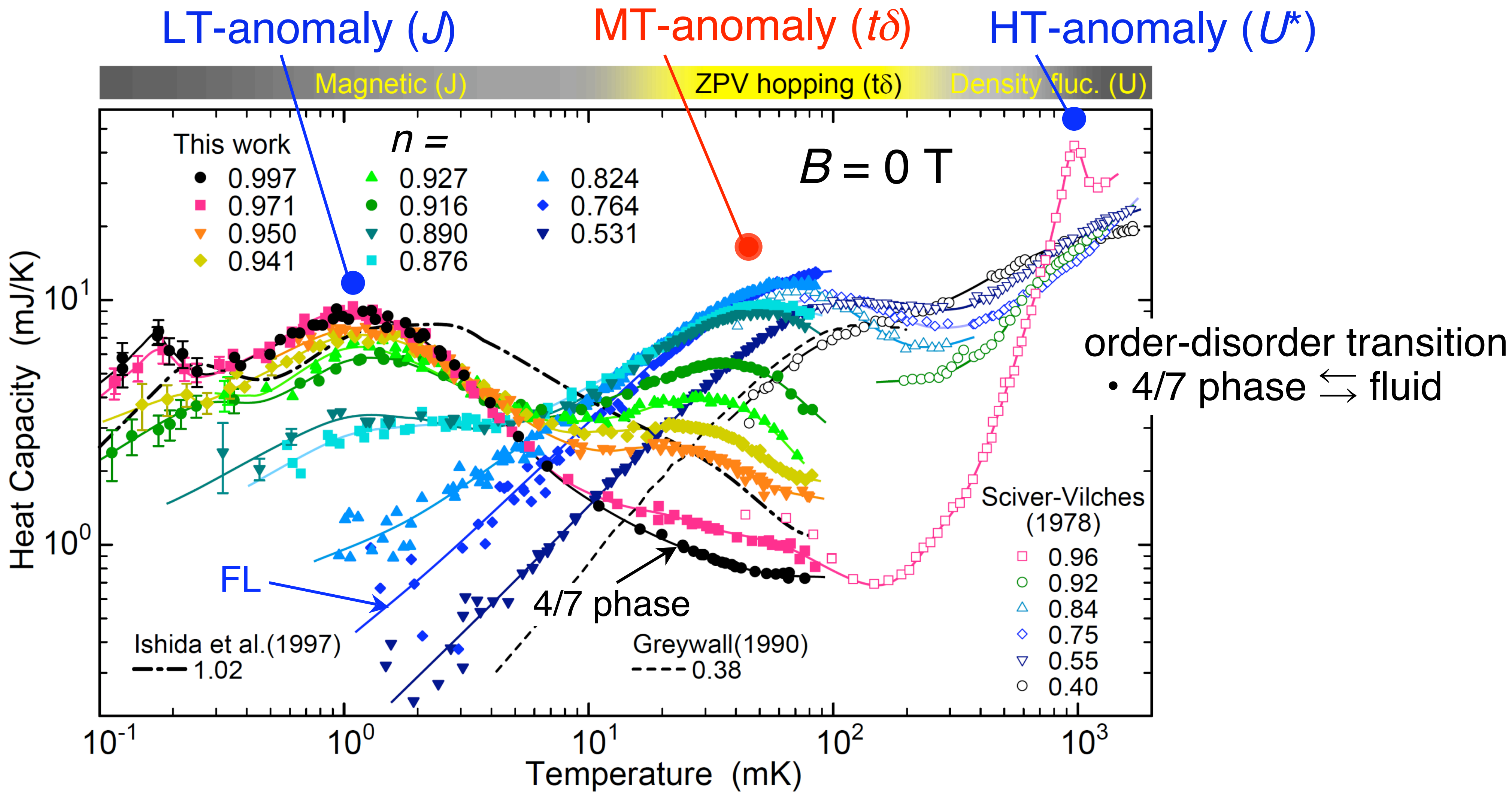
H. Matsuda and T. Tsuneto, Suppl. Prog. Theor. Phys. **46**, 411 (1970)





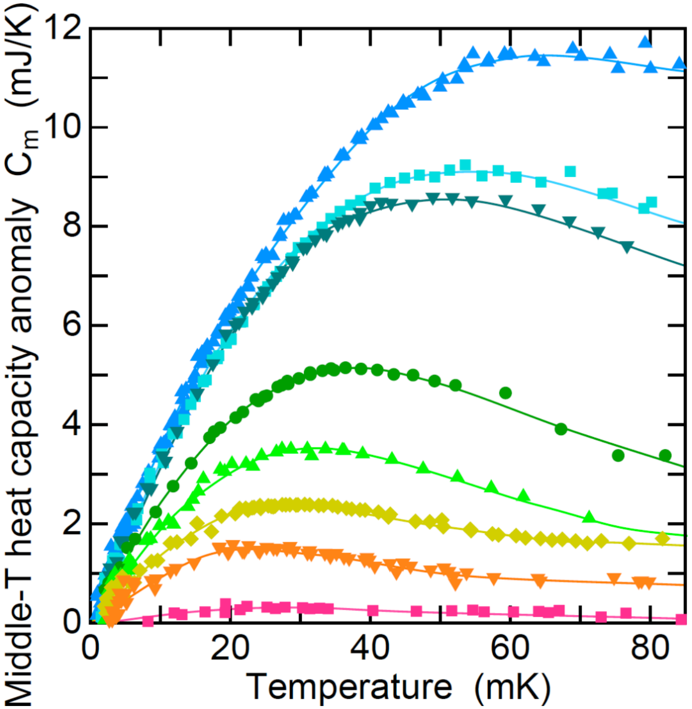
Heat capacities of Region-II

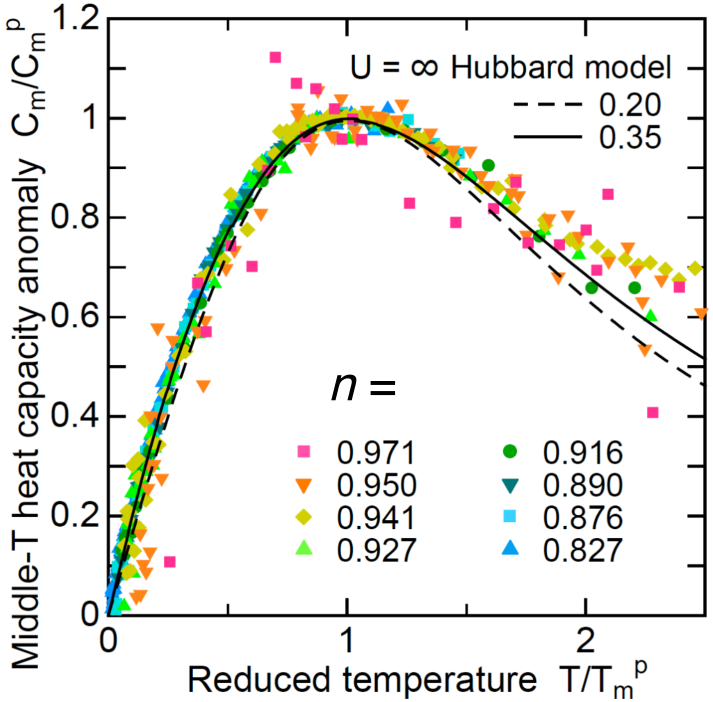
Three distinct energy scales over three orders of magnitude



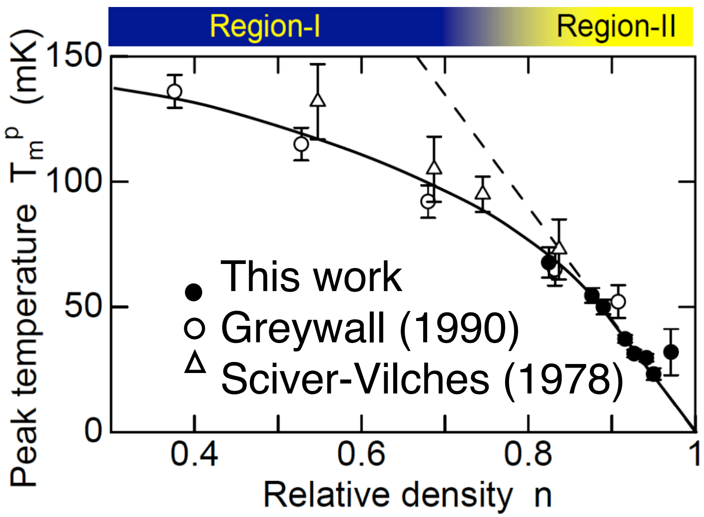
relative density: $n \equiv \rho/\rho_{4/7}$ $\rho_{4/7} = 6.86\ \mathrm{nm}^{-2}$ $A = 556\ \mathrm{m}^2$

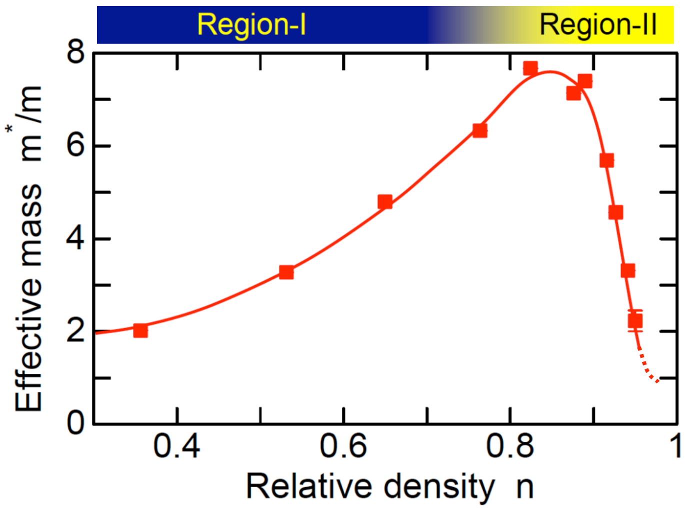
MT-anomalies of heat capacity in Region-II





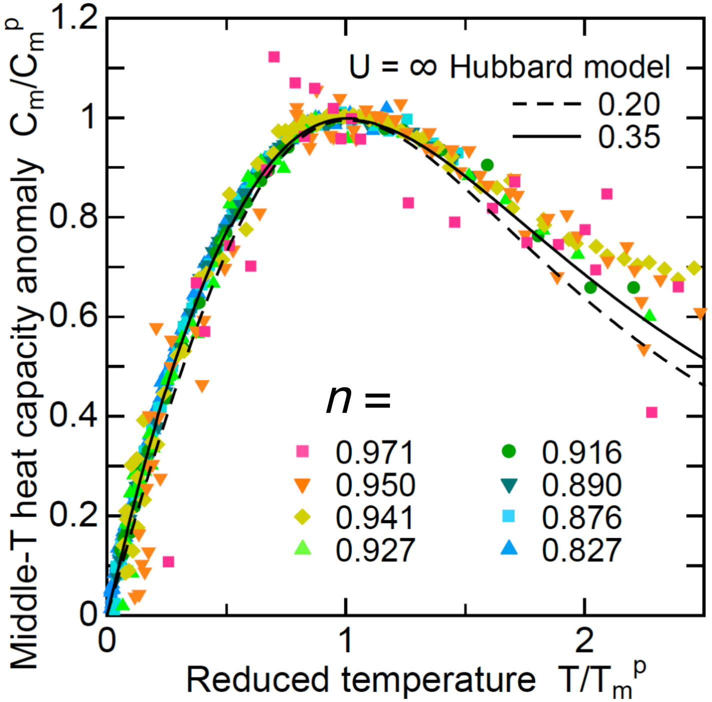
- T_m^p shifts to higher-T with decreasing n.
- C/C_m^p scales with T/T_m^p.





- $T_m^p \propto \delta \equiv (1 - n)$
- γ value turns around at $n \approx 0.8$.

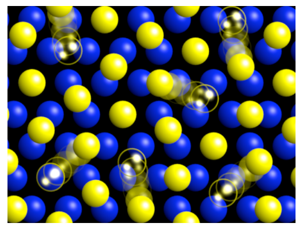
2D hole (ZPV) band picture for Region-II

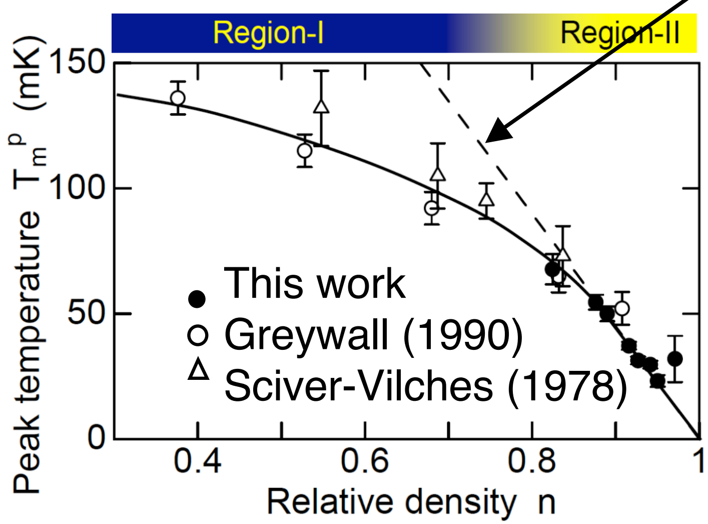


$U = \infty$ Hubbard model* for low particle densities gives good description for measured $C_m(T)$ at high particle densities.

*Koretsune-Ogata (2006)

- 2D hole (ZPV) band is created as a result of strong correlation effects near localization.
- Lattice models are applicable.





- $T_m^p \approx t\delta$ → $t = 430$ mK
- tight binding cal. (Koretsune-Ogata) → $t = 320$ mK
- $J = 4t^2/U^*$ → $t = 200$ mK

Consistent each other.

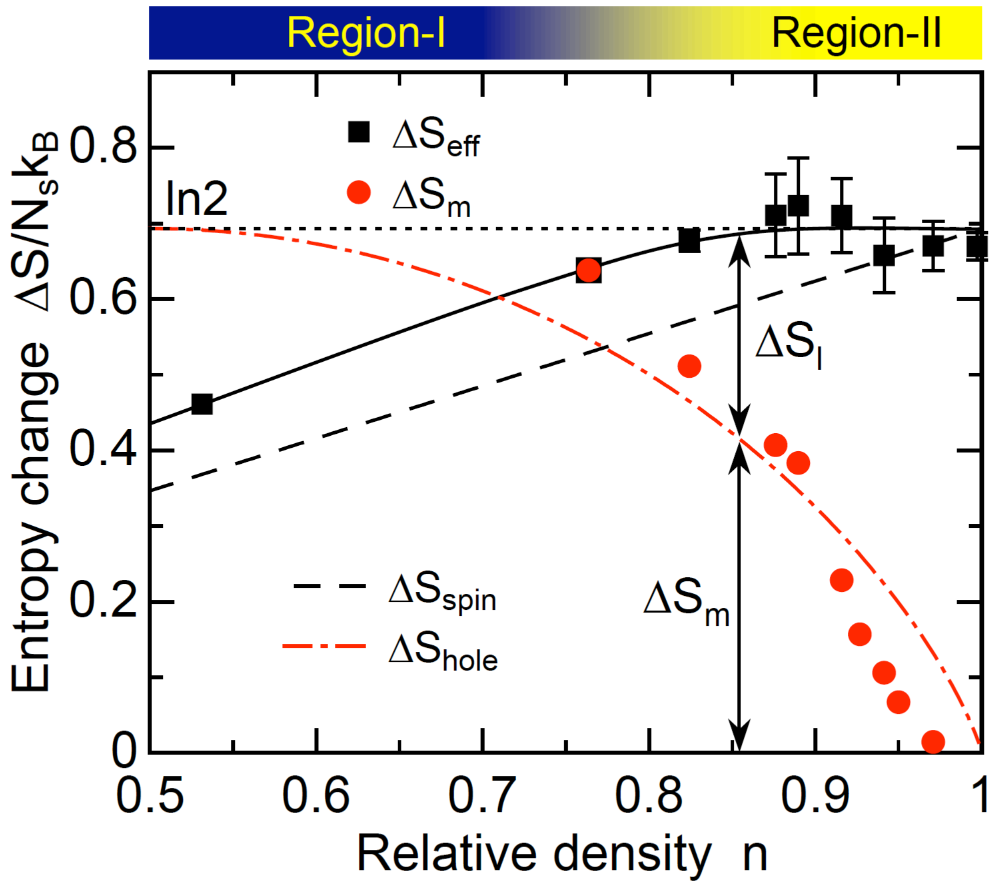
$U^* \approx$ 1-10 K (short-range repulsion)
:density fluctuations such as layer promotion or interstitial creation

$U^*/t \approx 50$

J = 10-20 mK > T_1^p : due to frustration

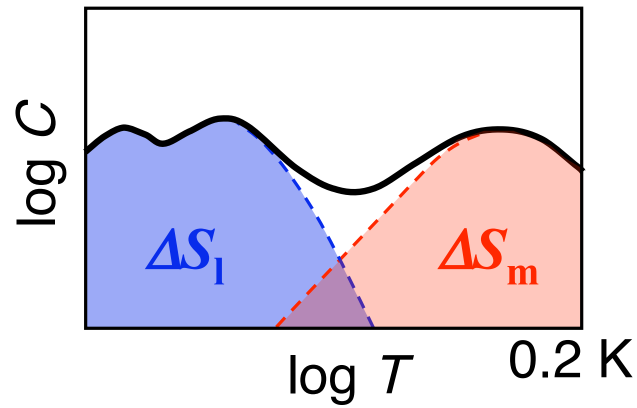
Entropies of Region-II

ΔS_{eff} (= ΔS_m + ΔS_l): deduced from $C(T)$ below 0.2 K



ΔS_m: deduced from $C_m(T)$

ΔS_l: deduced from $C_l(T)$



$\Delta S_{eff} \approx$ ln2 : **almost localized model**

$\Delta S_m \approx \Delta S_{hole}$: entropy to distribute $N_s\delta$ vacancies on N_s lattice-sites

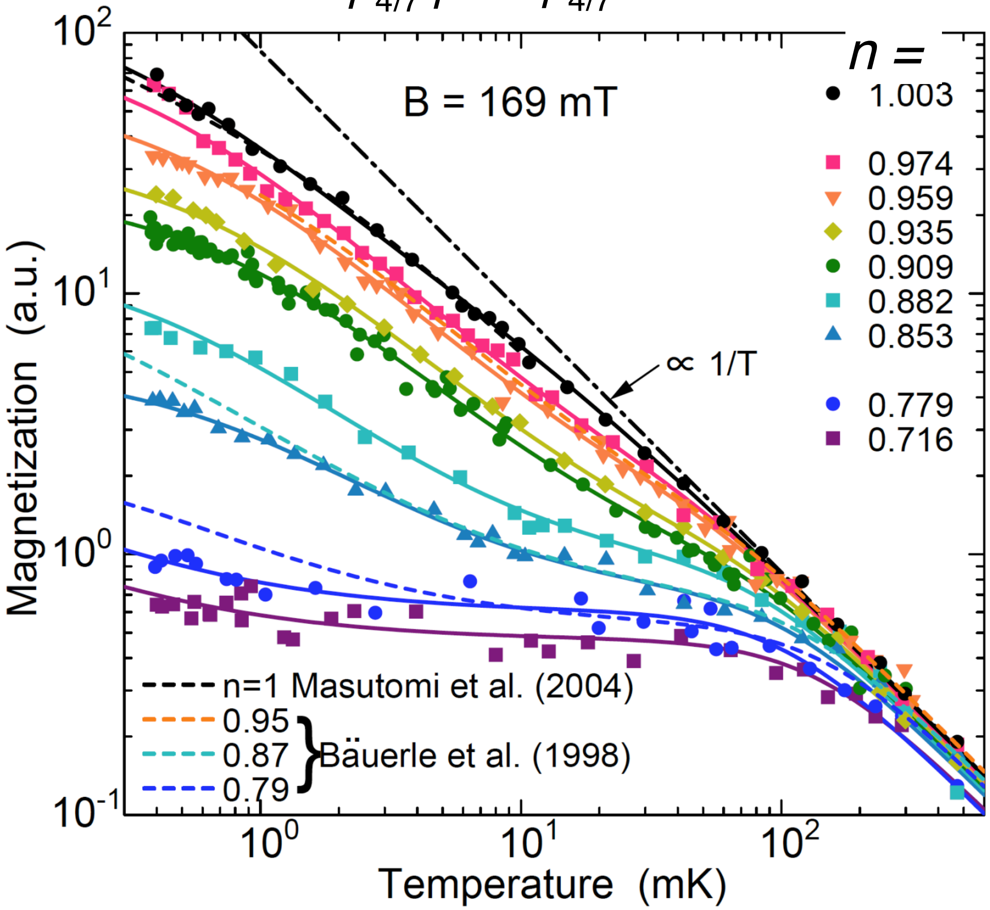
$$\Delta S_{hole} = N_s k_B\{-(1-n)\ln(1-n) - n\ln n\}$$

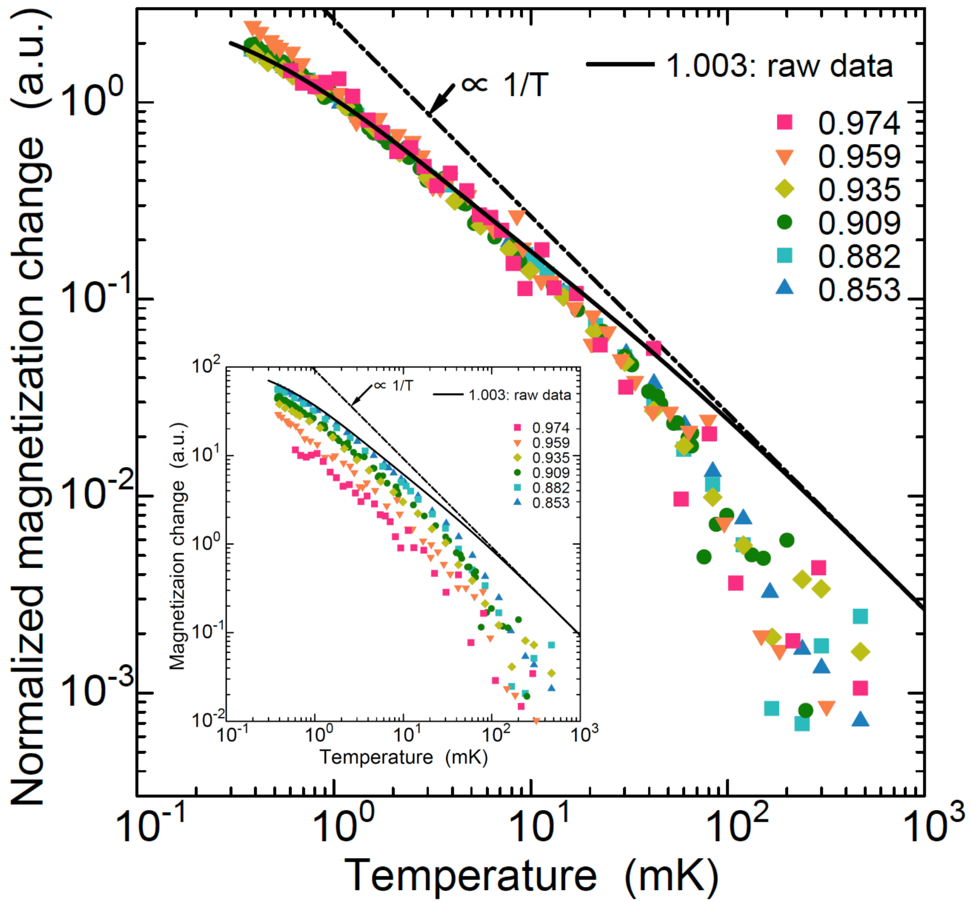
- spin mass separation in 2D? Y, Fuseya and M. Ogata, submitted to JPSP(2008)
- fermion differentiation? T. Misawa and M. Imada, PRB **75**, 115121 (2007)

Magnetization of Region-II

$n \equiv \rho_{4/7}/\rho \qquad \rho_{4/7} = 6.86\ \mathrm{nm}^{-2}$

S. Murakawa et al., to be published





$\Delta M(\rho, T) \equiv M(\rho_{4/7}, T) - M(\rho, T)$

All $\Delta M(\rho, T)$ have the same T-dependence.

→ phase separation?! •••• contradicts heat capacity data

High-*T* series expansions for *t-J* models

Exp. Magnetization data are consistent with *t-J* model for triangular lattice.

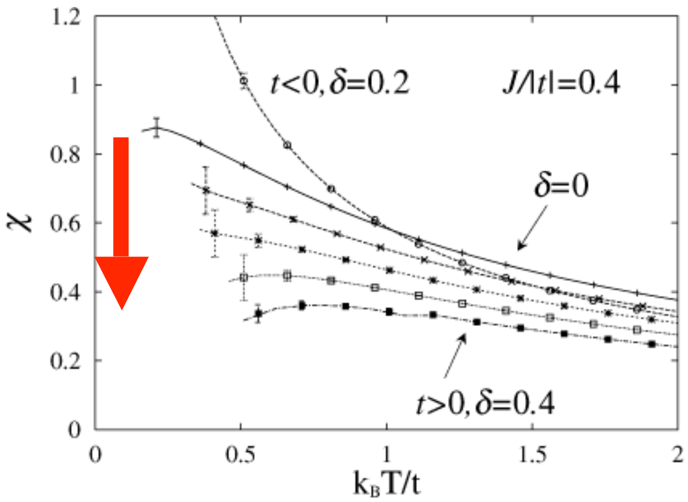
$$H = -t \sum_{\langle i,j \rangle,\sigma} P\left(c_{i\sigma}^{\dagger} c_{j\sigma} + h.c.\right)P + J \sum_{\langle i,j \rangle} S_i \bullet S_j \qquad P : \text{operator to prohibit doublon}$$

Triangular lattice

T. Koretsune and M. Ogata,PRL **89**,11640 (2002)

Hole doping decreases χ

- release of spin frustration
- giving gaped nature
- nontrivial AFM

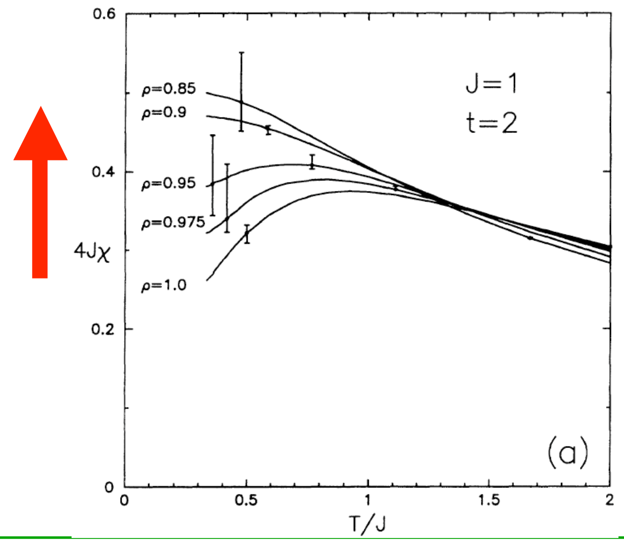


Square lattice

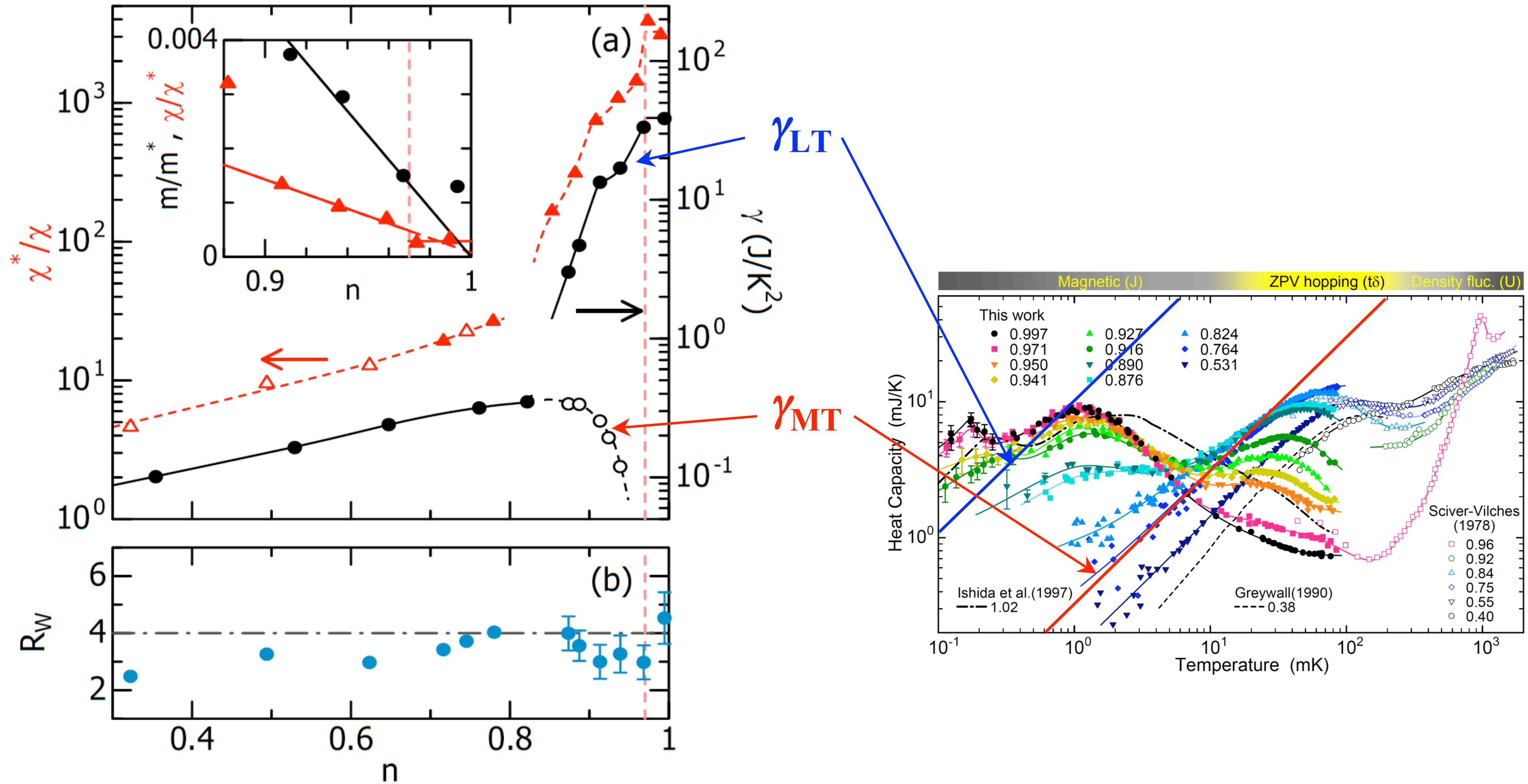
R.R.P. Singh and R.L. Glenister,PRB **46**, 11871 (1992)

Hole doping increases χ

- destructing Neel coupling
- Nagaoka FM



Wilson ratio in Region-II

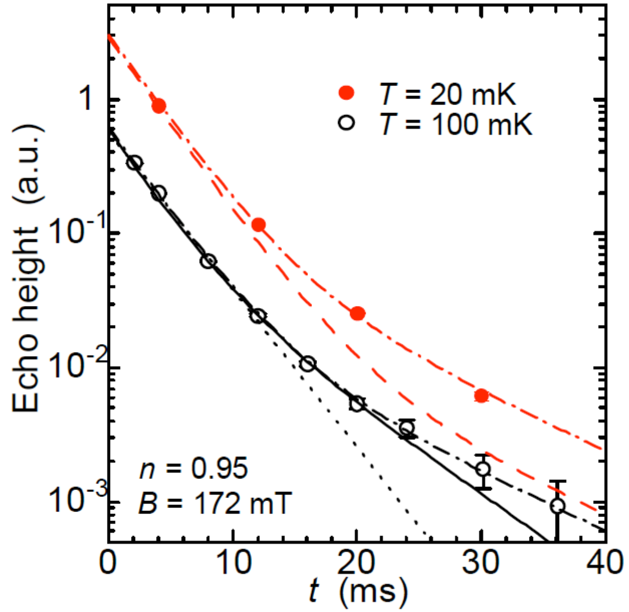


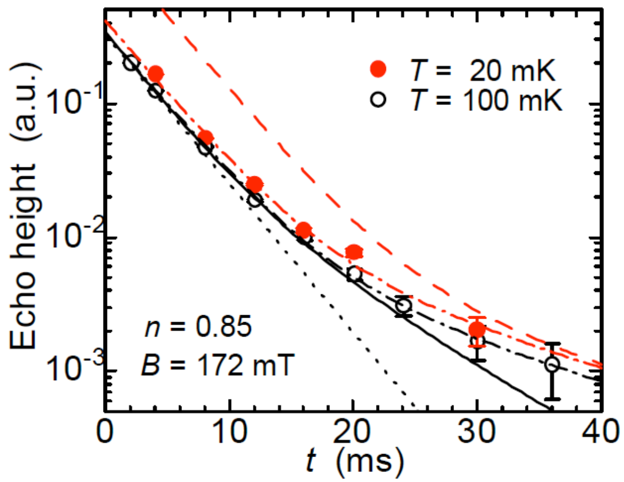
Wilson ratio (R_W) = 3 - 4 •••• Quite heavy fermions set in to create at low-T.

➡ **Spinon Fermi surface**?

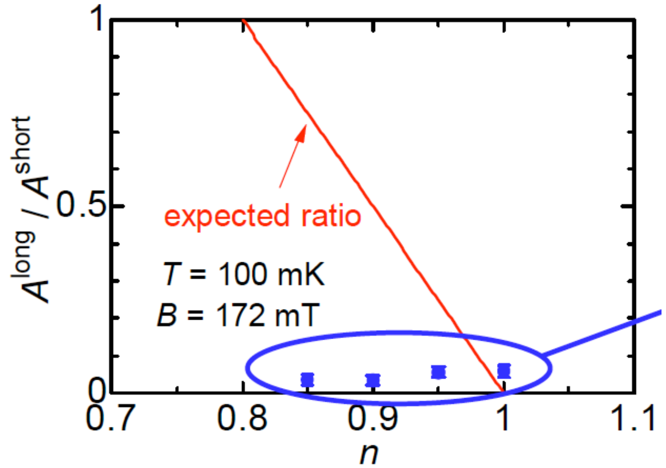
Spin-spin relaxation time (T_2) measurements

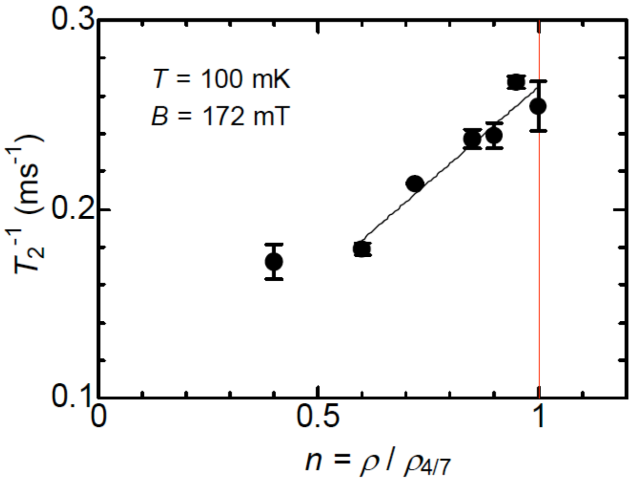
S. Takayoshi et al., *to be published*





T_2 is sensitive to local spin dynamics:
exchange and motional narrowing



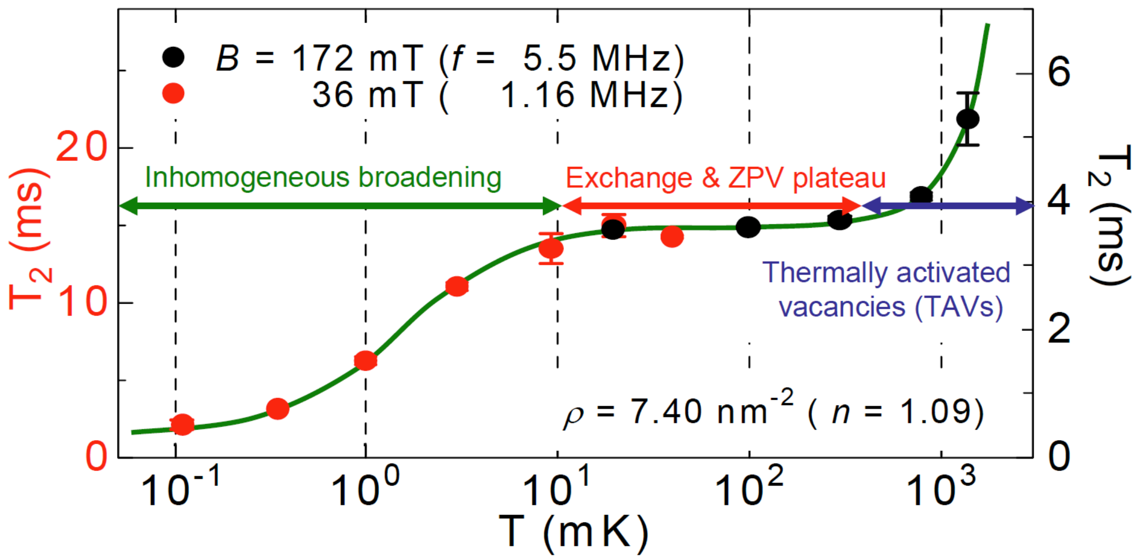


Shorter T_2 component (95%): *intrinsic*
single exponential → single phase
: supporting the ZPV scenario

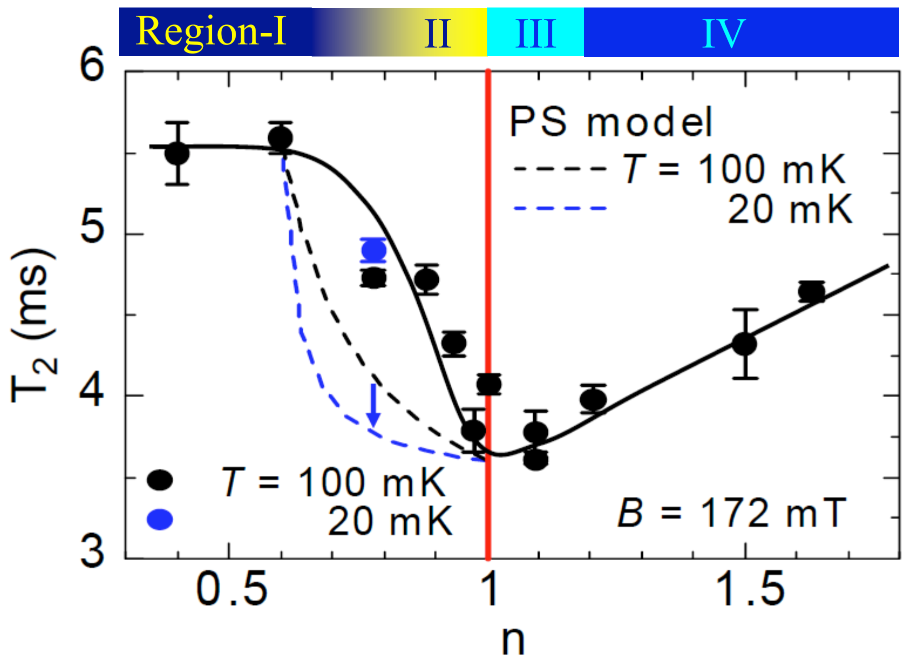
Longer T_2 component (5%): *extrinsic*
inisotropic T_2 for 2D systems and mosaic angle spread of Grafoil

T_2 measurements near the 4/7 phase

temperature dependence



density dependence



There are three *T*-regimes:

(1) $T \leq 10$ mK: short-range ordering?

(2) $10 \leq T \leq 300$ mK: *T*-independent regime determined by quantum motions

(3) $T \geq 300$ mK: thermally activated density fluctuations

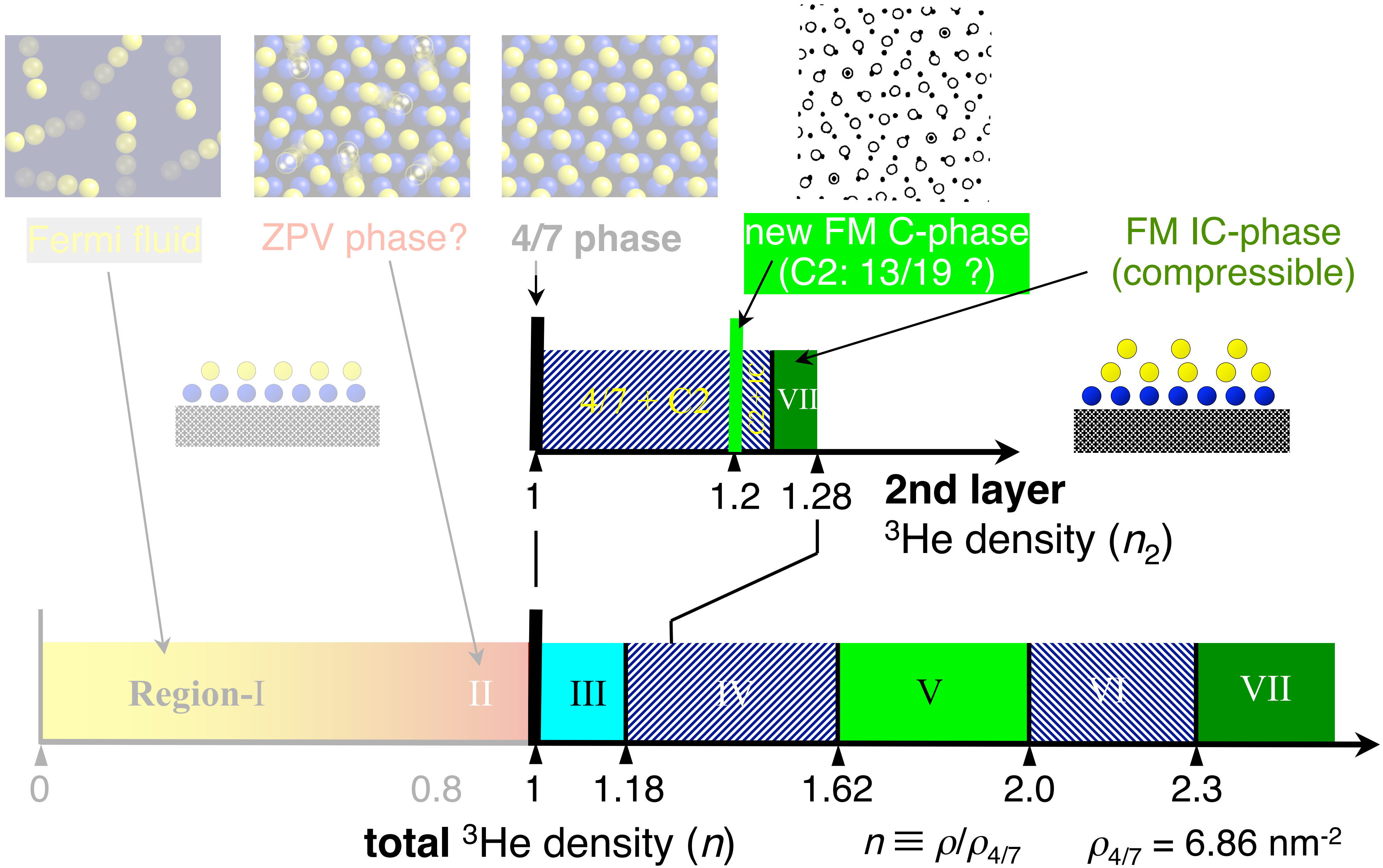
Region-II :
motional-narrowing presumably due to ZPV

Region-III, IV :
exchange-narrowing due to interlayer exchanges

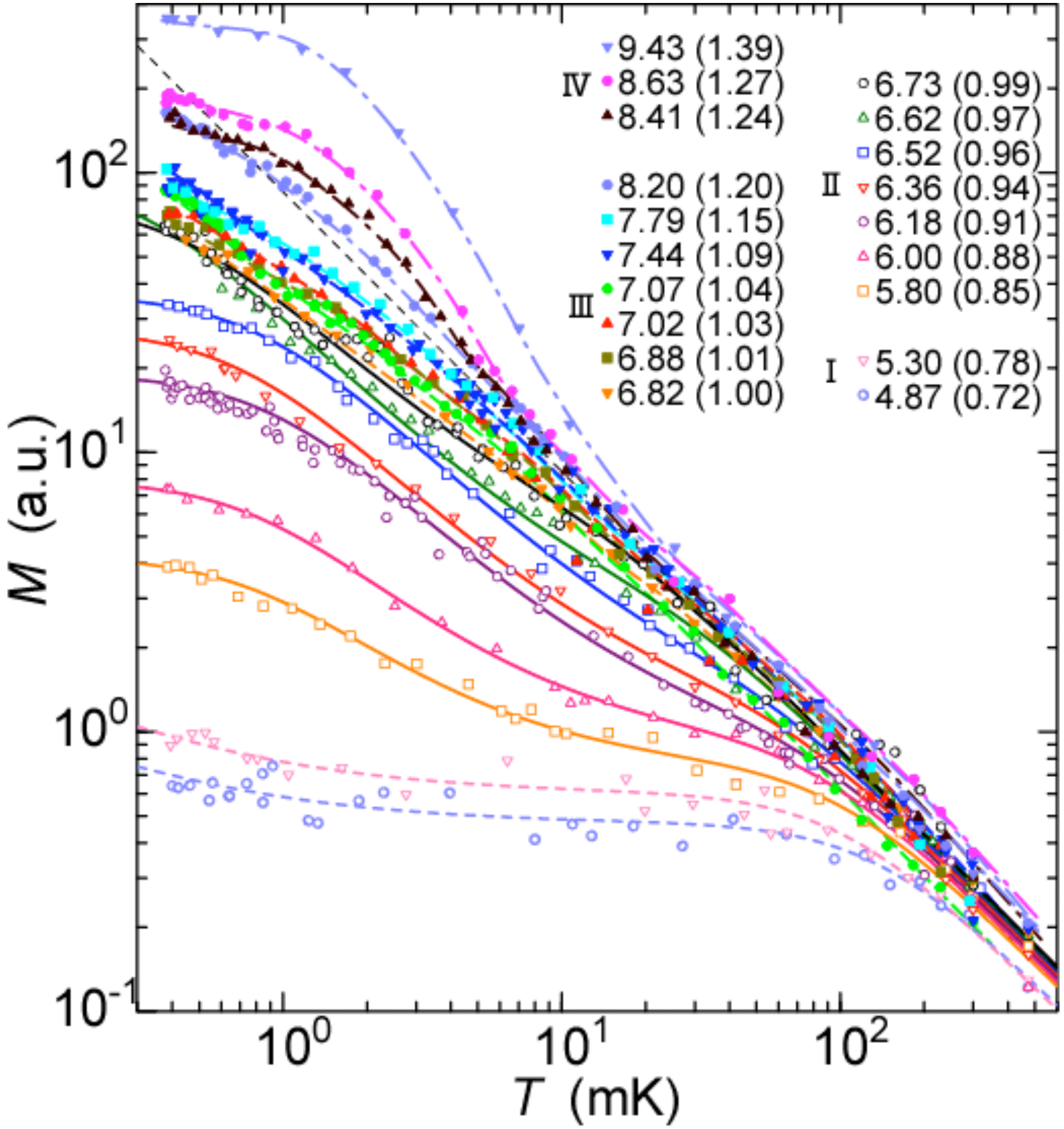
§4. Particle doping into the 4/7 phase

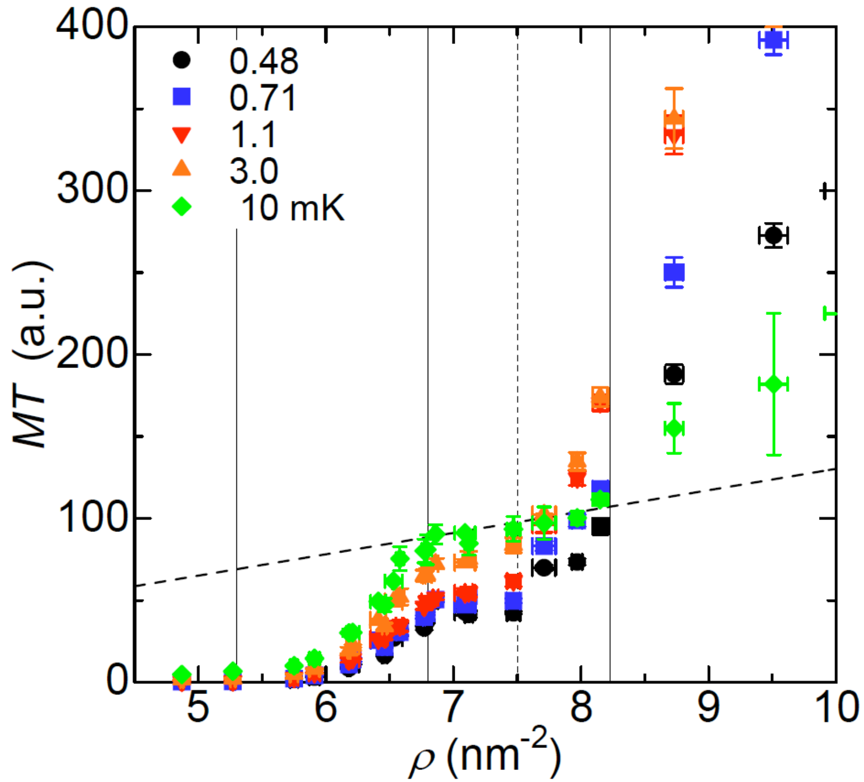
FM phases appear due to reduced frustration.

$T = 0$ phase diagram of 2D ^{3}He (2nd layer)

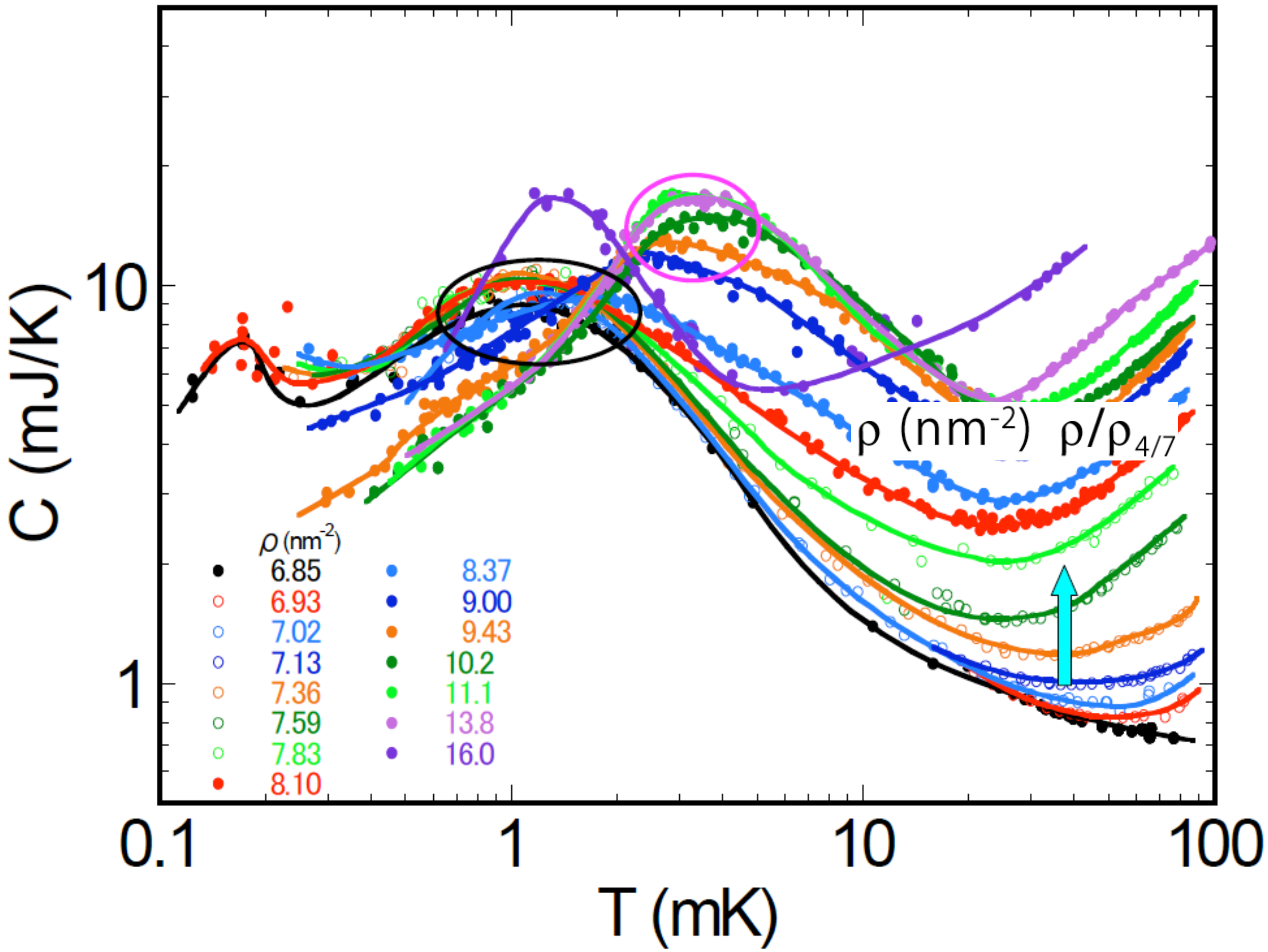


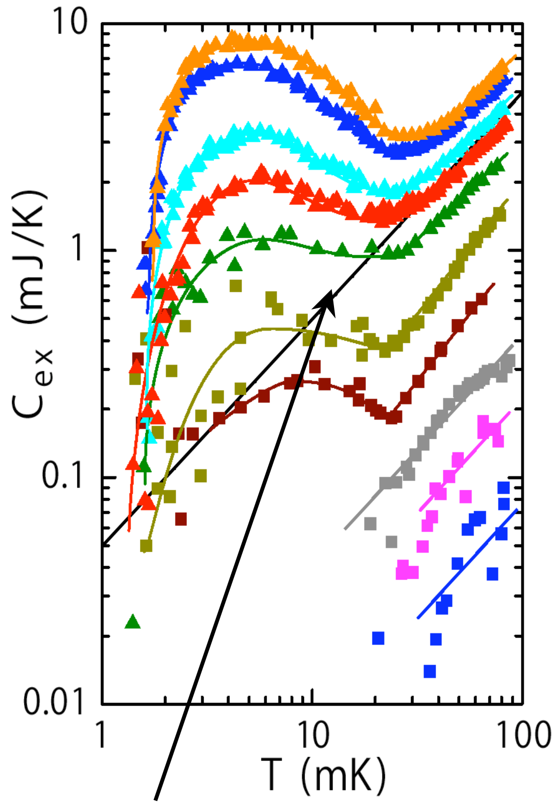
Magnetization of 2D ^{3}He





Heat capacities in Regions III-VII ($\rho \geq \rho_{4/7}$)

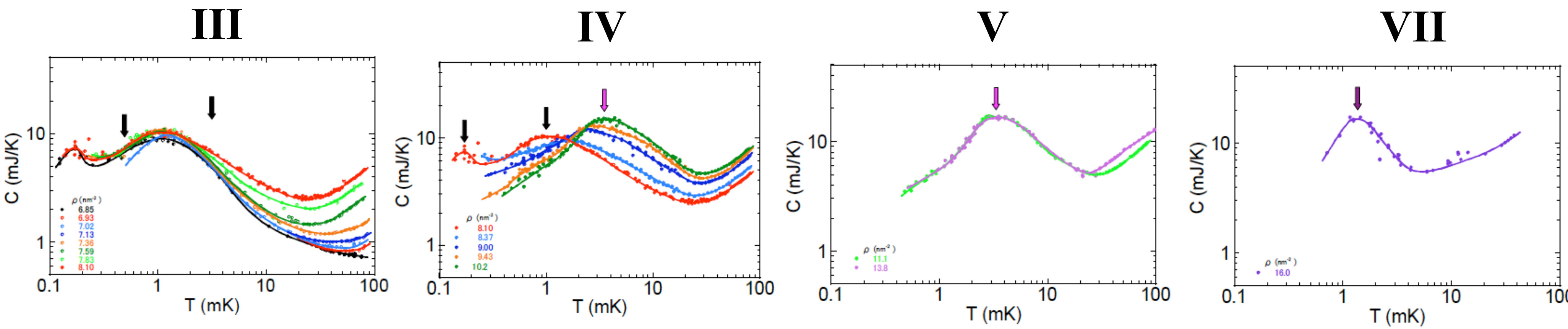




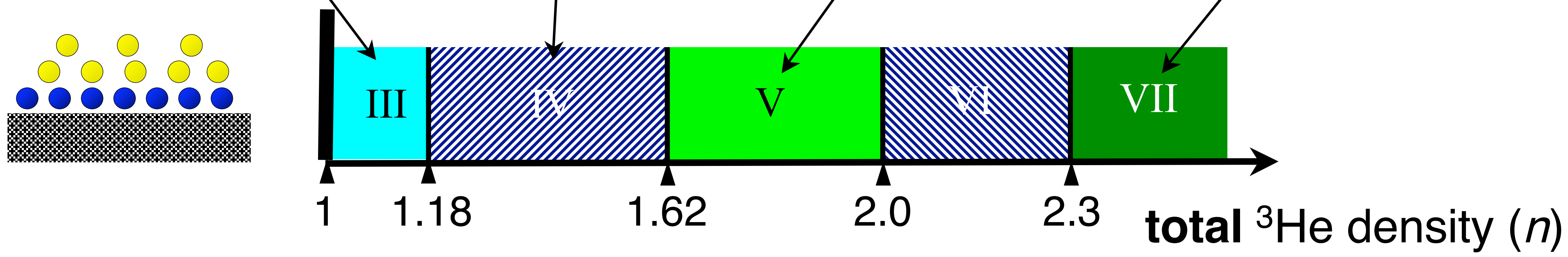
Ideal 2D Fermi gas

- Excess particles are promoted into 3rd layer ($1 \leq n \leq 1.2$) and behave as a degenerate Fermi liquid (puddle).

Heat capacities in Regions III-VII ($\rho \geq \rho_{4/7}$)

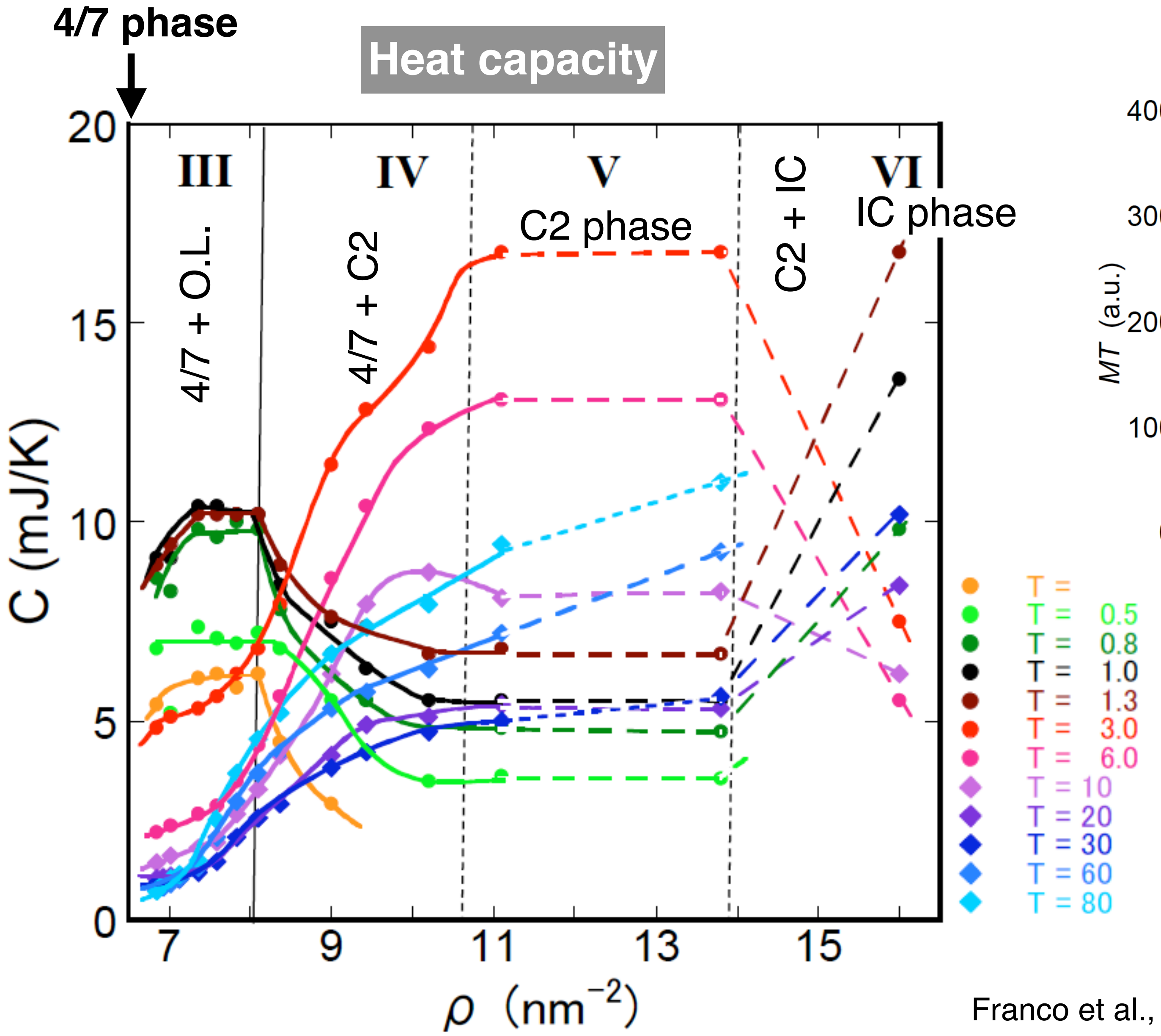


3rd layer	over layers	over layers	over layers
+	+	+	+
4/7 phase	(**4/7 phase** + C2 phase)	C2 phase	IC phase

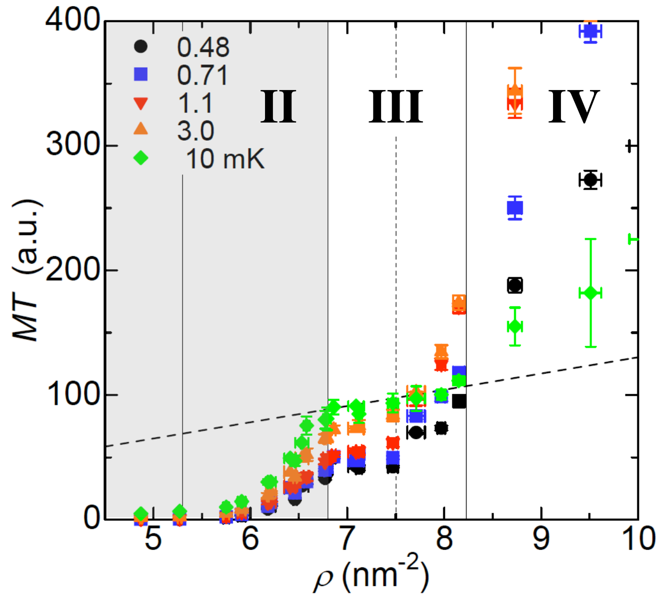


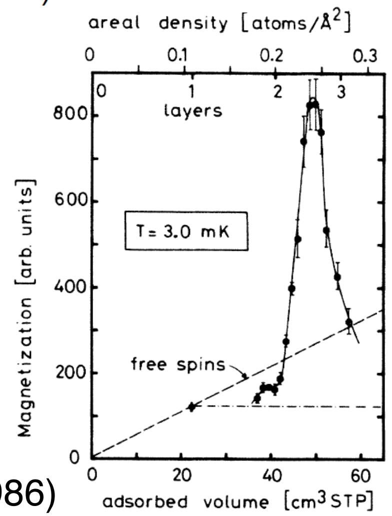
Isotherms of heat capacity and magnetization in excess particle regions

Heat capacity



Magnetization

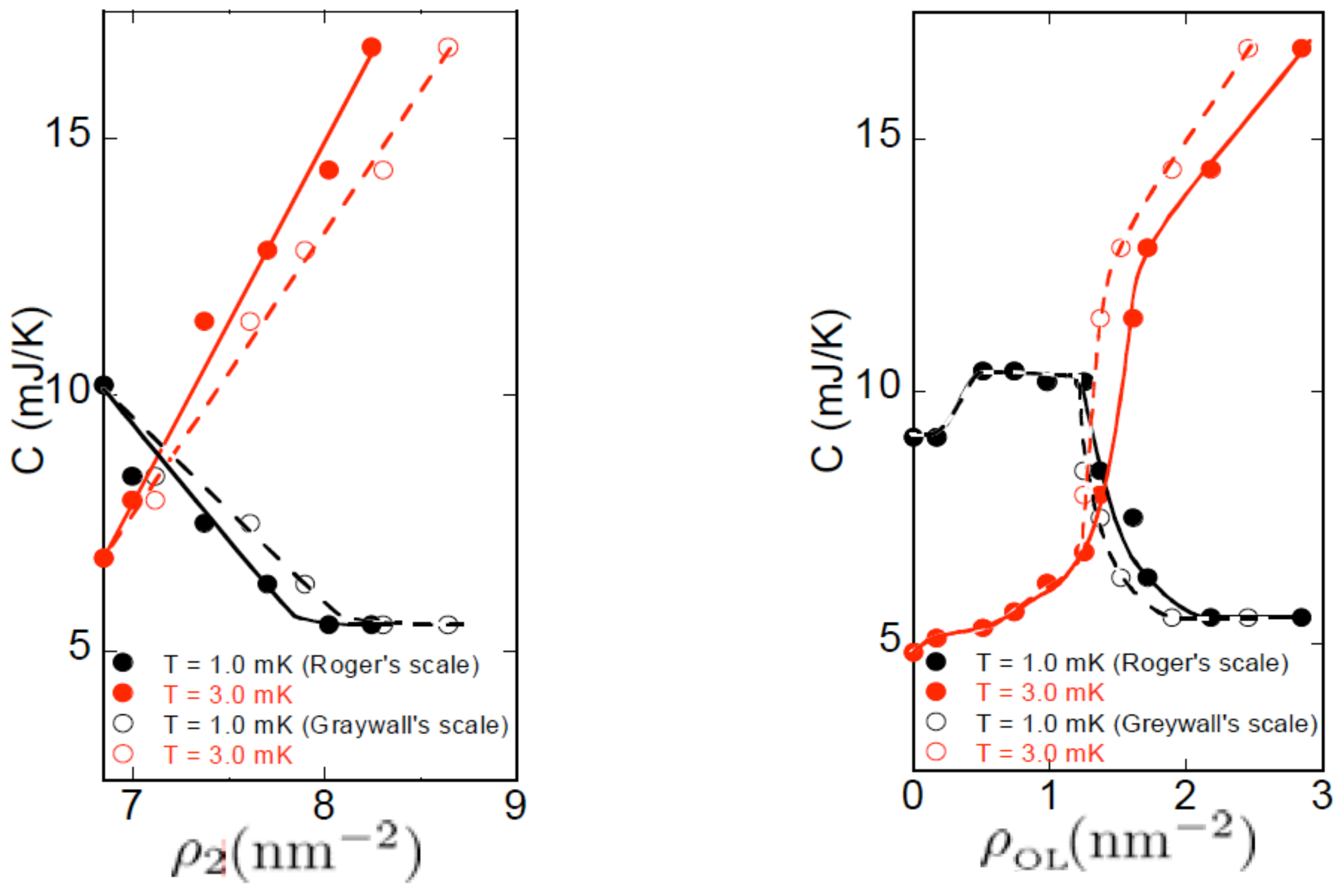




Franco et al., PRL **57**, 1161 (1986)

Coexistence within 2nd layer (Region-IV)

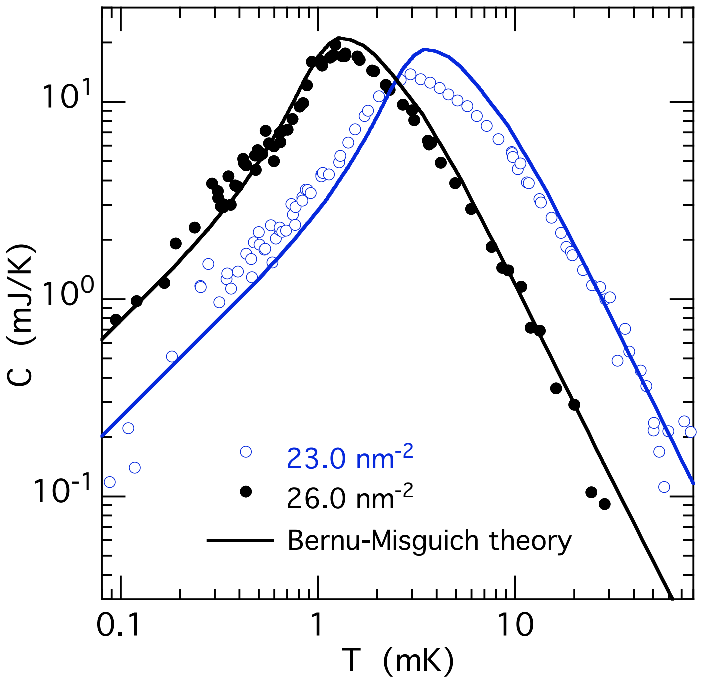
Heat capacity isotherms vary linearly with ρ_2 not ρ_3.

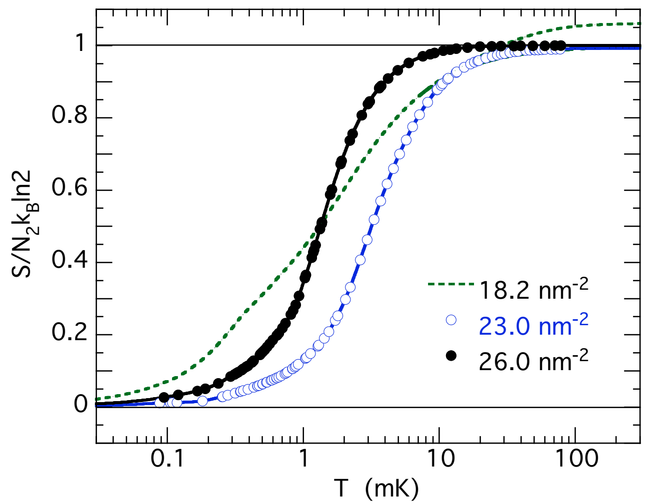


The 4/7 phase coexists with the other commensurate phase (C2-phase).

Heat capacities of C2 and IC phases

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





C2 phase (FM ground state)

- slightly frustrated 2D FM on a triangular lattice

J (magnetization) = -(1.7 - 2.1) mK

J (heat capacity) = -2.7 mK

$\eta \equiv \left(K + J_6\right)/\left|J\right| = 0.10\text{-}0.12$

IC phase (FM ground state)

- nearly ideal 2D FM on a triangular lattice

J (heat capacity) = -1.0 mK

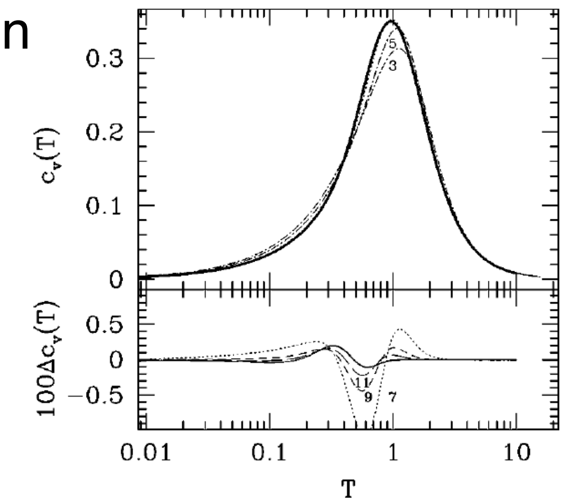
$\eta \approx 0$

S = 1/2 Heisenberg model on a triangular lattice:
HAFT model

B. Bernu and G. Misguich., PRB **63**, 134409 (2001)

Interpolation scheme between

- HT series expansion (with Pade approx.) regime
- LT spin-wave regime



Future prospect

Region-II : $(0.8 \leq n \leq 1)$ ··· hole doping

1. Verification of the ZPV model

- more complete spin-echo measurements
- vapor pressure measurement at fixed T → compressibility

2. Experimental determination of the 4/7 structure

- LEED or neutron scattering

3. Search for supersolid phase in 2D ^{4}He

- torsional oscillator (with Shirahama group)
- 2nd sound

Region-III, IV : $(1 \leq n)$ ··· particle doping

1. Full survey of the phase diagram

- heat capacity and pulsed-NMR

2. Experimental determination of structural change

- LEED or neutron scattering

§5. Summary

1. 4/7 phase

- First gapless spin-liquid state experimentally observed.
- MSE model explain exp. data but not the gapless nature.

2. Region-II (hole doping)

- Possible ZPV phase suggested from heat capacity data.
- T_2 data support single phase (ZPV phase) at least at high-T ($\geq$ 20 mK).
- But magnetization data suggest phase separation being inconsistence with heat capacity data.
- spin-mass separation?, fermion differentiation?, gradual phase separation?

3. Region-III-VII (particle doping)

- The 4/7 phase is stable until 20% excess particles. The newly added particles are promoted into 3rd layer forming FL paddles.
- A new commensurate phase (C2) was found. It is a slightly frustrated 2D FM on a triangular lattice.