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Highly Frustrated Nuclear Magnetism in Monolayer Helium Three on Graphite

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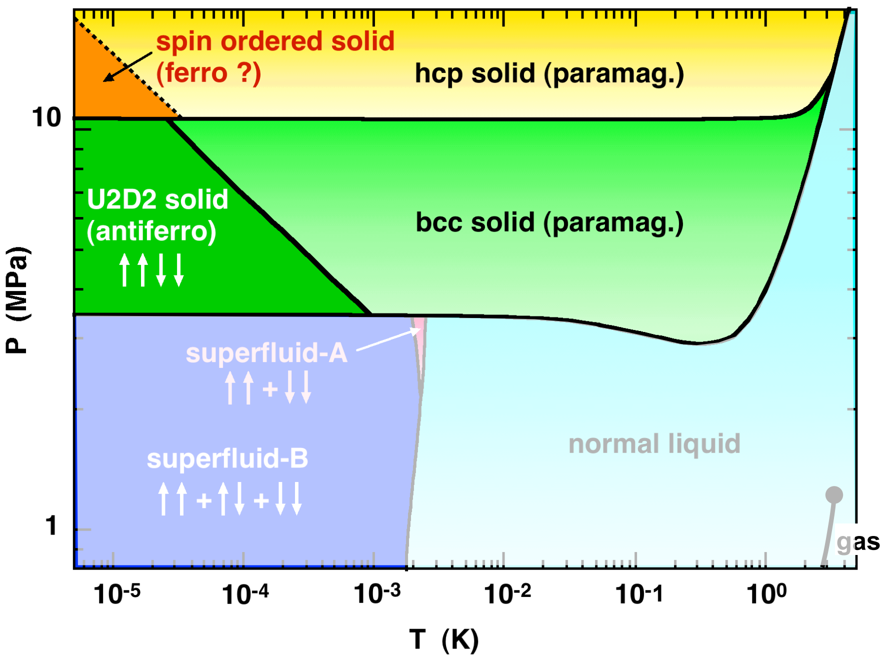
Outline:

1. Bcc solid ^{3}He as a reference 3D AFM with **ring exchanges** (RE)
2. **Frustrated 2D FM** with RE ••• high-density IC solid ^{3}He
3. **2D gapless QSL ••• **4/7 phase

Solid ^{3}He in 3D: ideal quantum spin system (S =1/2)



Ideal, simple and highly tunable

➢No impurities

➢Quantum spin (S = 1/2)

➢No spin-orbit coupling (nuclear-spin system)

➢Isotropic exchange interactions ($J \approx$ 1 mK)

atom-atom exchanges

large zero-point motion

^{1}S closed electronic shell

➢Large energy separation (spin entropy is dominant)

$h\omega_D/J \approx 10^4$, $E_{d\text{-}d}/J \approx 10^{-5}$

➢Highly compressible (large Grüneisen constant)

$K \approx 5 \times 10^{-3}$ bar^{-1} near M. C.

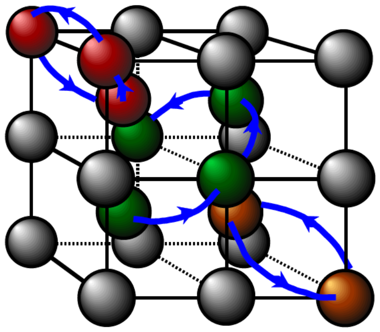
⇐ P (or V) : control parameter

➢Frustration

competing **ring exchanges** (RE) (up to 6-spin)

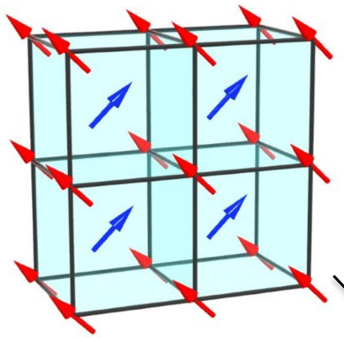
AFM: even number

FM: odd number



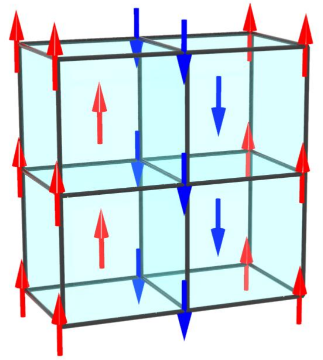
Magnetic phase diagram of bcc solid ^{3}He

H. Fukuyama et al., arXiv:cond-mat/0505177



CNAF phase (HFP)

1sr order transition



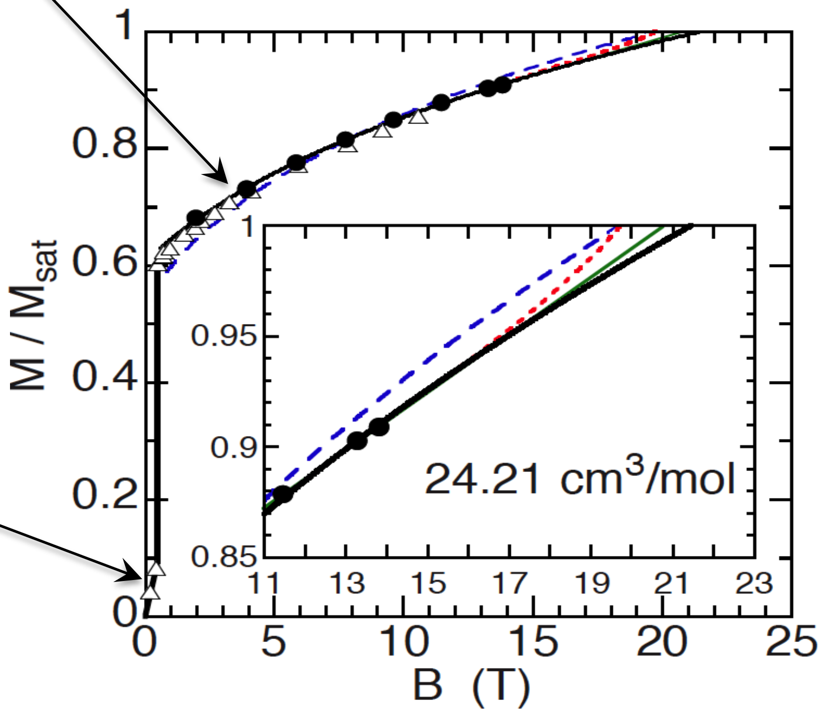
U2D2 phase (LFP)

Frustrated AFM with LRO

$T_N\ (B = 0) = 0.93$ mK

$B_{c2}\ (T = 0) \approx 20$ T

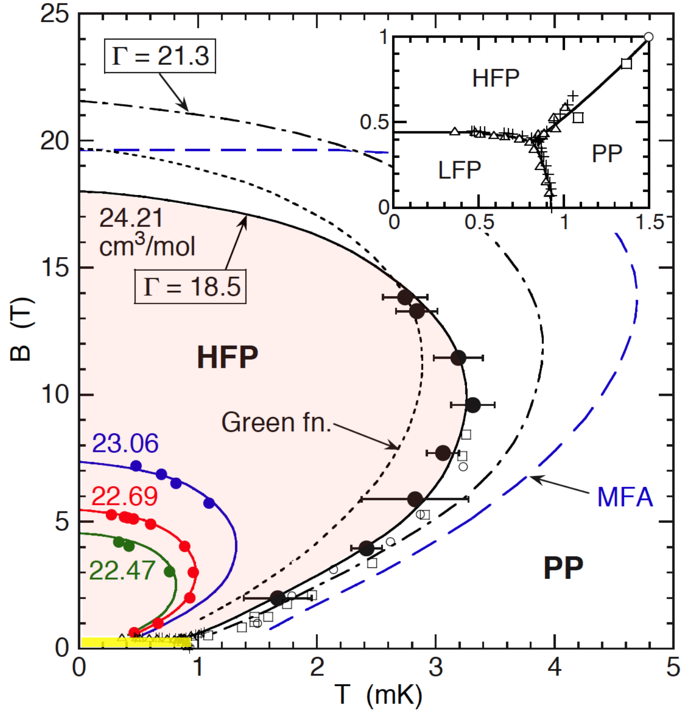
$k_B T_N << \mu B_{c2}$



RE parameters

$J_{1N} = 0.48,\ T_1 = 0.20,\ K_P = 0.28,$
$S_1 = 0.037,\ S_2 = 0.023$ in mK

at $v = 24.21$ cm^3/mol



Ring exchange (RE) model

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

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

P : permutation op. $\boldsymbol{J_P}$: ring exchange interactions

2-spin (AFM) $P_1 = P_{ij} = \frac{1}{2}\left(1 + \sigma_i \bullet \sigma_j\right)$ ← Heisenberg term

3-spin (FM) $P_2 = P_{ijk} + P_{ijk}{}^{-1} = \frac{1}{2}\left(1 + \sigma_i \bullet \sigma_j + \sigma_j \bullet \sigma_k + \sigma_k \bullet \sigma_i\right)$ ← Heisenberg term

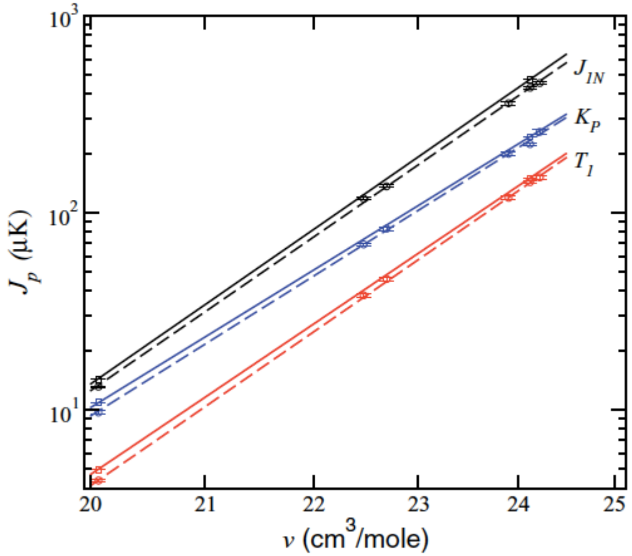
4-spin (AFM) $P_3 = P_{ijkl} + P_{ijkl}{}^{-1} = \frac{1}{4}\left(1 + \sum_{\mu<\nu} \sigma_\mu \bullet \sigma_\nu + G_{ijkl}\right)$

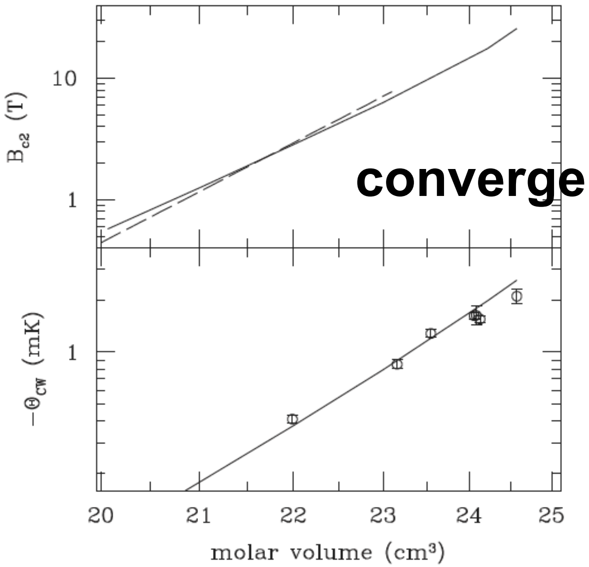
⋮

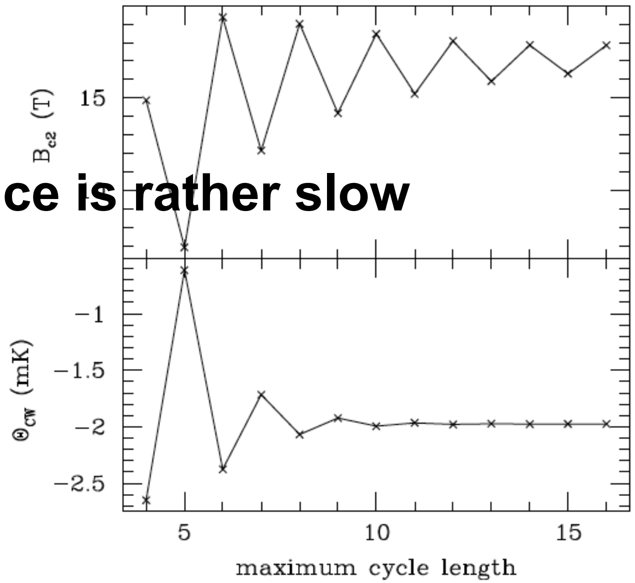
$$G_{ijkl} \equiv \left(\sigma_i \bullet \sigma_j\right)\left(\sigma_k \bullet \sigma_l\right) + \left(\sigma_i \bullet \sigma_l\right)\left(\sigma_j \bullet \sigma_k\right) - \left(\sigma_i \bullet \sigma_k\right)\left(\sigma_j \bullet \sigma_l\right)$$ ← new term

Recent PIMC calculations up to J_8

C^andido, Hai, and Ceperley, PRB. **84**, 064515 (2011)



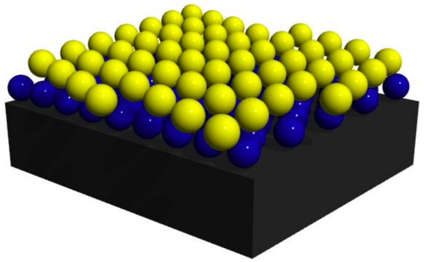




convergence is rather slow

Fabrication of 2D ^{3}He

layer by layer physisorption of ^{3}He on **exfoliated graphite substrate** at low temperatures

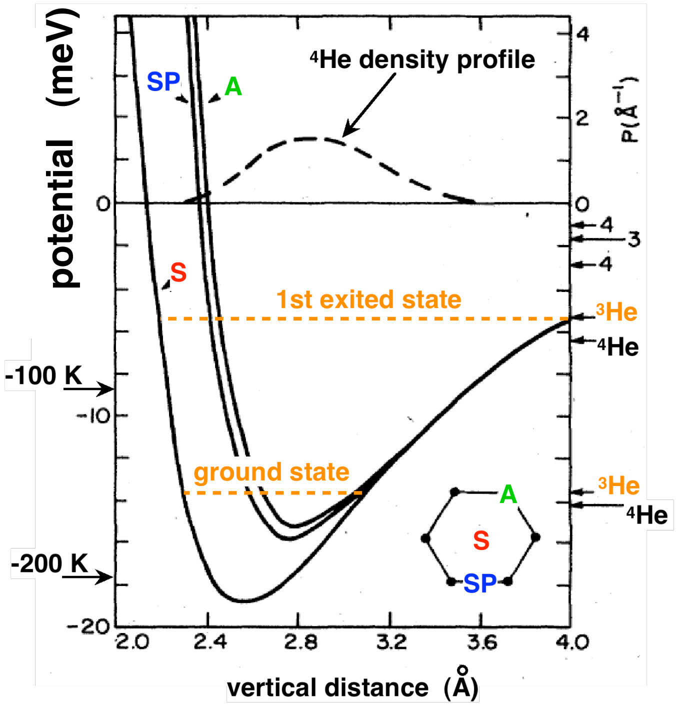


Ideal, simple and highly tunable 2D magnet

➢Much higher frustration
competing ring exchanges + geometrical (triangular)

➢Much larger fluctuation
lower dimension **(2D)** cf. nano cluster (0D)

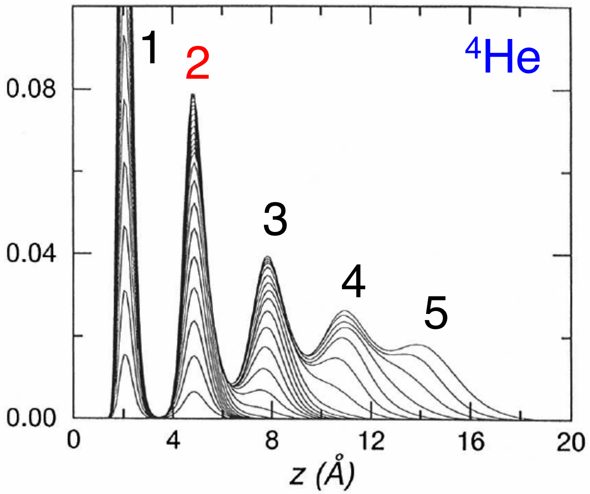
He-graphite potential



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

2nd later ^{3}He has experimentally accessible J (≈ 1-10 mK).

Calculated density profile



➢1st and 2nd layers are well separated.

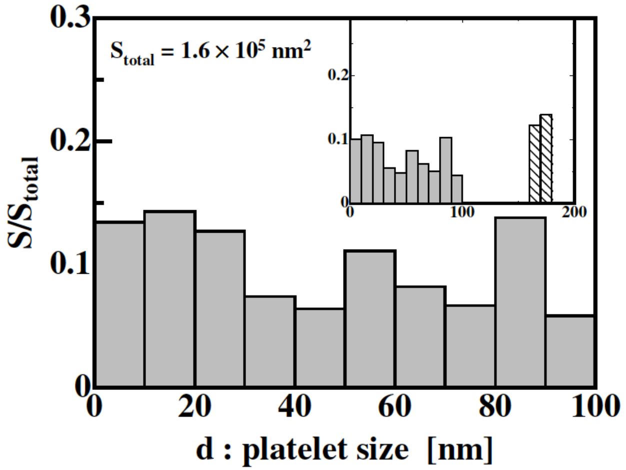
M. Roger et al., JLTP **112**, 45 (1998)

Exfoliated graphite substrate

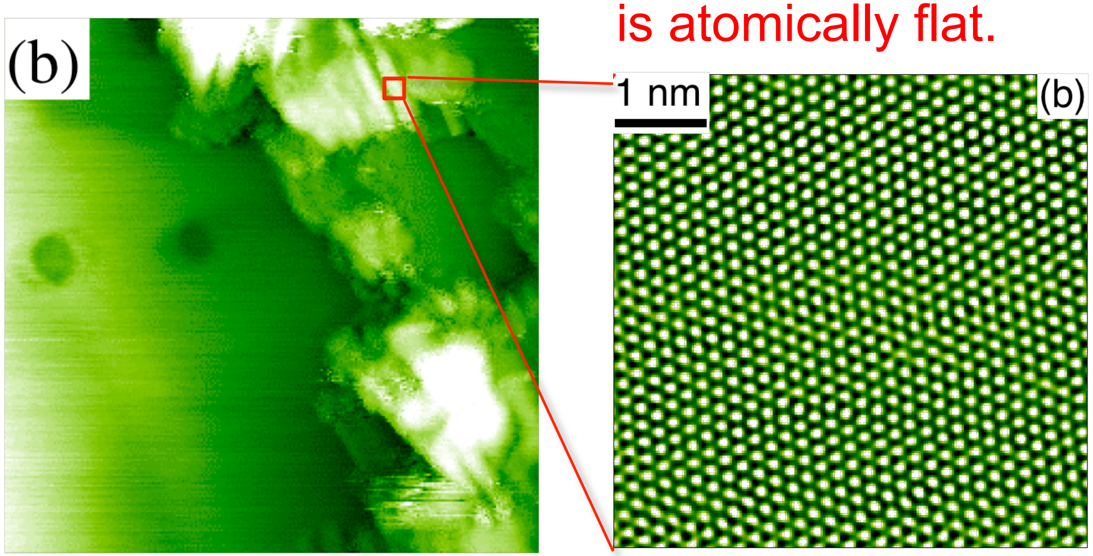
Grafoil

Y. Niimi et al., PRB. **73**, 085421 (2006)
S. Takayoshi and H. Fukuyama, JLTP **158**, 672 (2010)

- exfoliate "graphite foam" by graphite intercalation technique
- surface area: 20 m^2/g
- platelet size: 10-100 nm
- mosaic angle: ±30 deg
- density: 1.0 g/cm^3 (half of graphite)

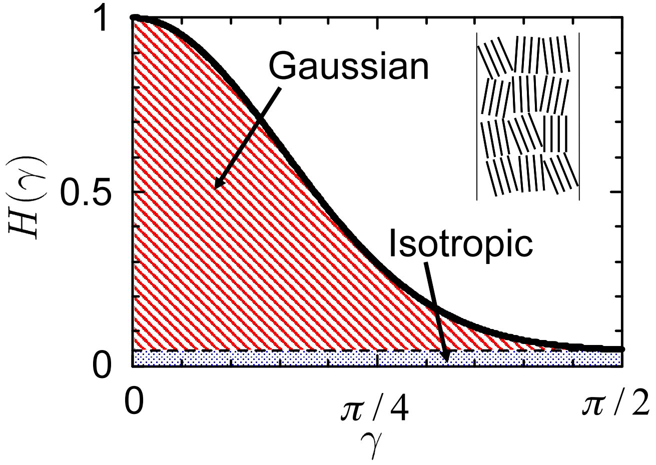


Each platelet surface is atomically flat.



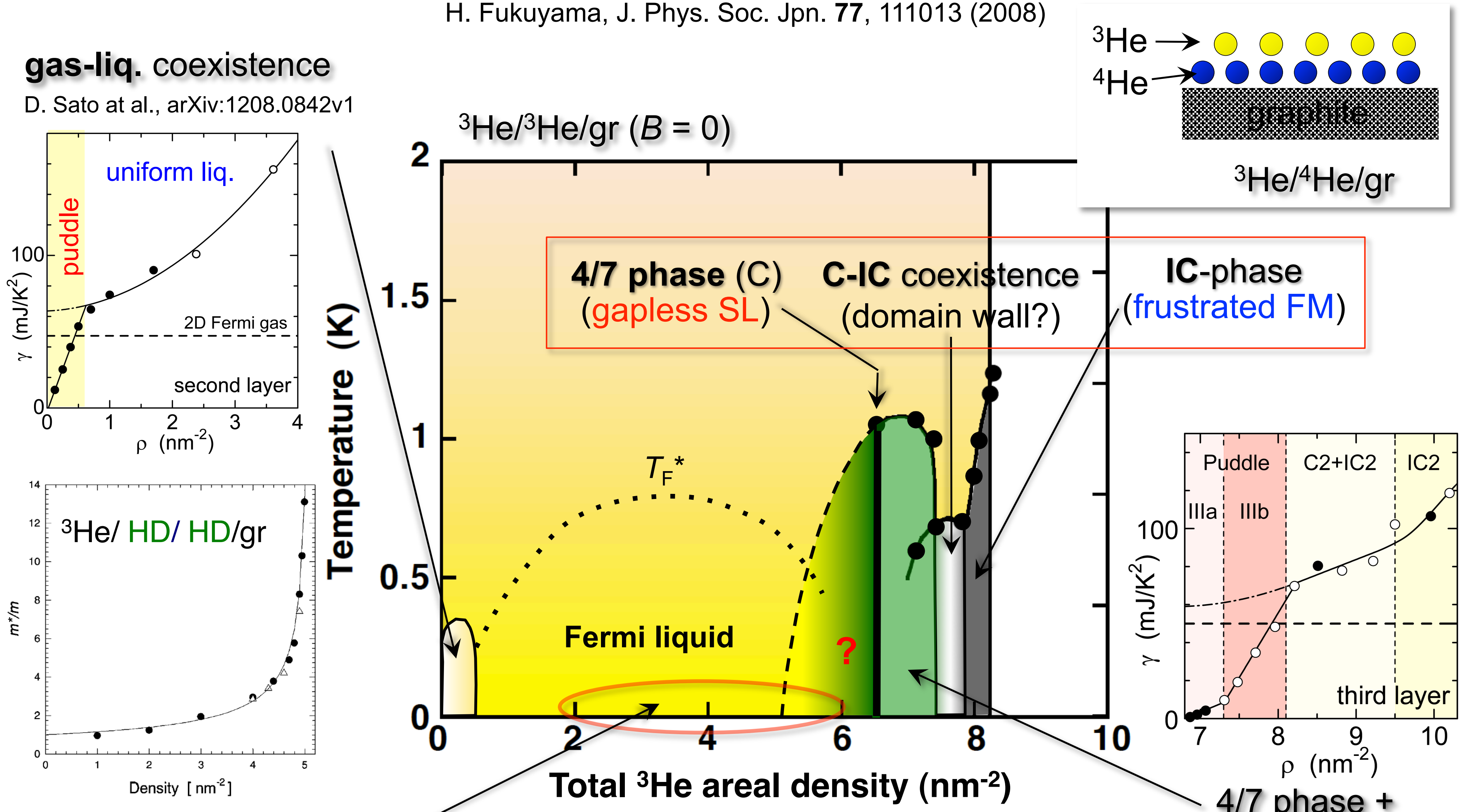
120×120 nm^2

STM images



Phase diagram of 2nd layer ^{3}He on graphite

H. Fukuyama, J. Phys. Soc. Jpn. **77**, 111013 (2008)



gas-liq. coexistence

D. Sato at al., arXiv:1208.0842v1

Mott-Hubbard transition

divergent m^* towards localization

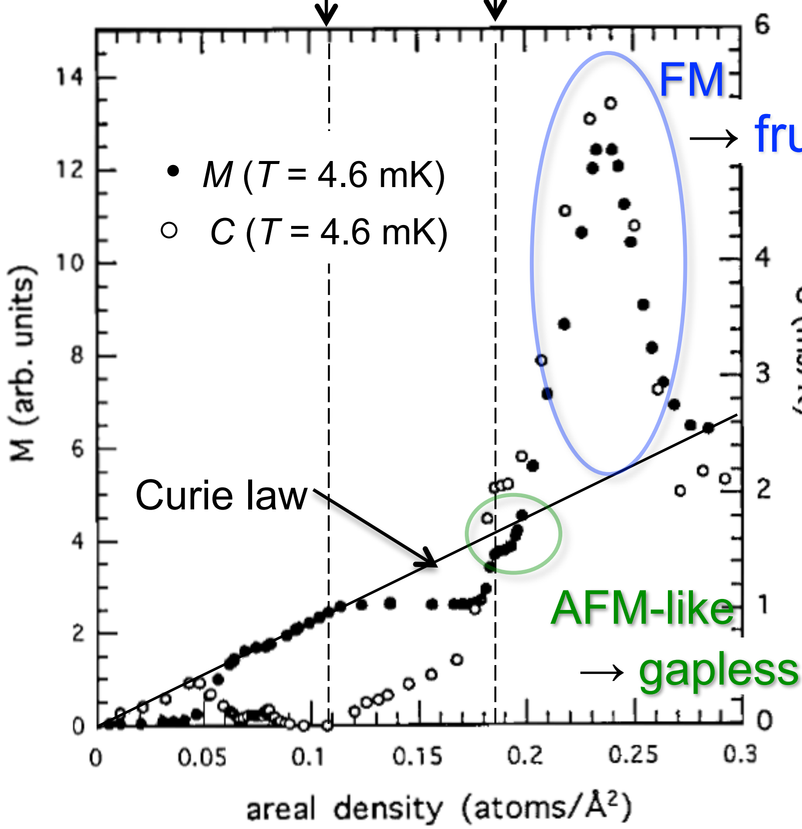
A. Casey et al., PRL **90**, 115301 (2003)

4/7 phase +
3rd layer **puddle**

D. Sato at al., arXiv:1208.0842v1;
JLTP **158**, 201 (2010)

2. Frustrated 2D FM with RE in high-density IC solid ^{3}He

2nd-layer IC slid is a Heisenberg ferromagnet on triangular lattice (HFT)



completion of 2nd layer localization
& 3rd layer promotion

2nd layer promotion

- M (T = 4.6 mK)

○ C (T = 4.6 mK)

FM

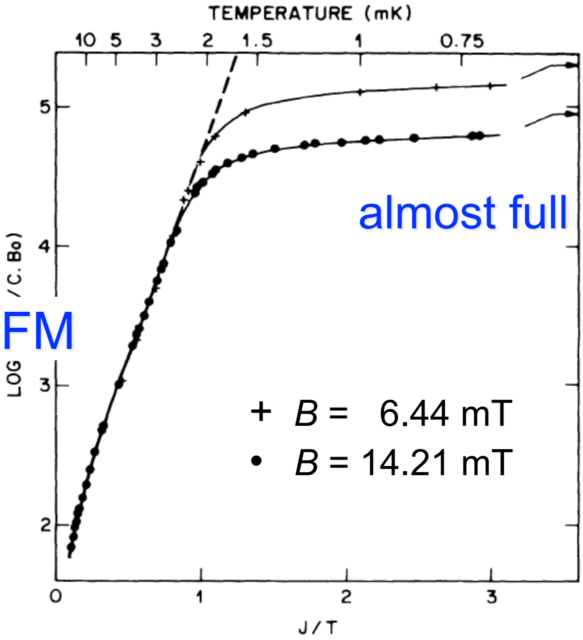
→ frustrated FM

Curie law

AFM-like

→ gapless QSL

C. Bäuerle et al.,Czech. J. Phys .suppl. S1 **46**, 399 (1996)
H. Franco, et al., PRL **57**, 1161 (1986)



M_{sat}

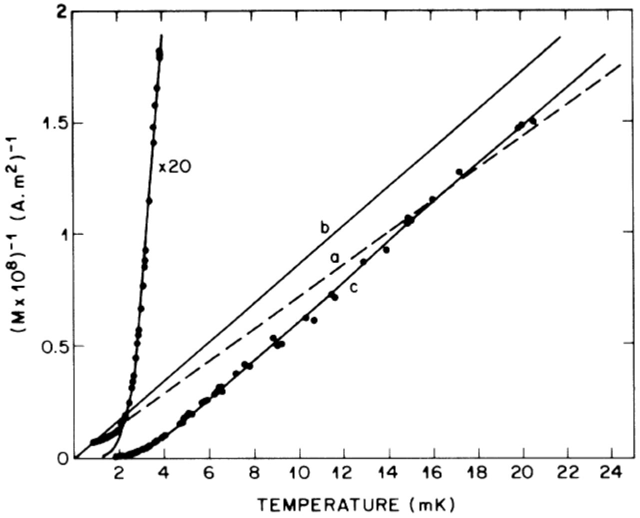
almost full saturation M

+ B = 6.44 mT

• B = 14.21 mT

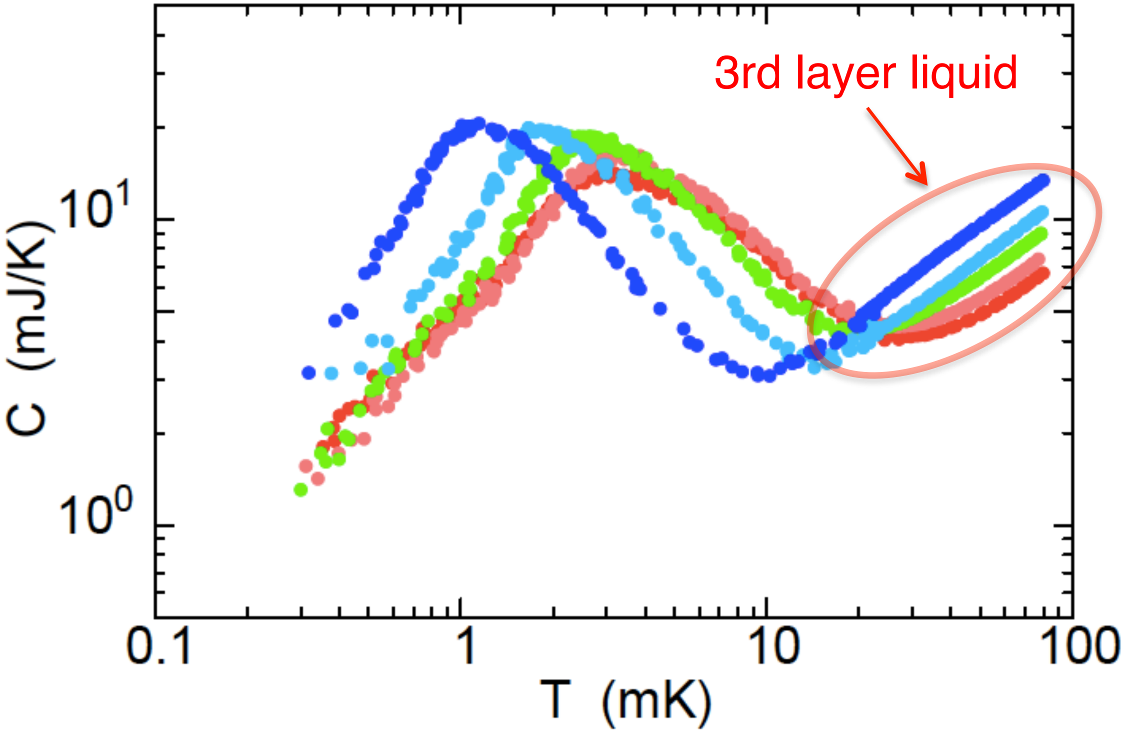
ρ_{tot} = 23.3 nm^{-2}
(ρ_1 = 11.2 nm^{-2},
ρ_2 = 8.1 nm^{-2},
ρ_3 = 4.0 nm^{-2})

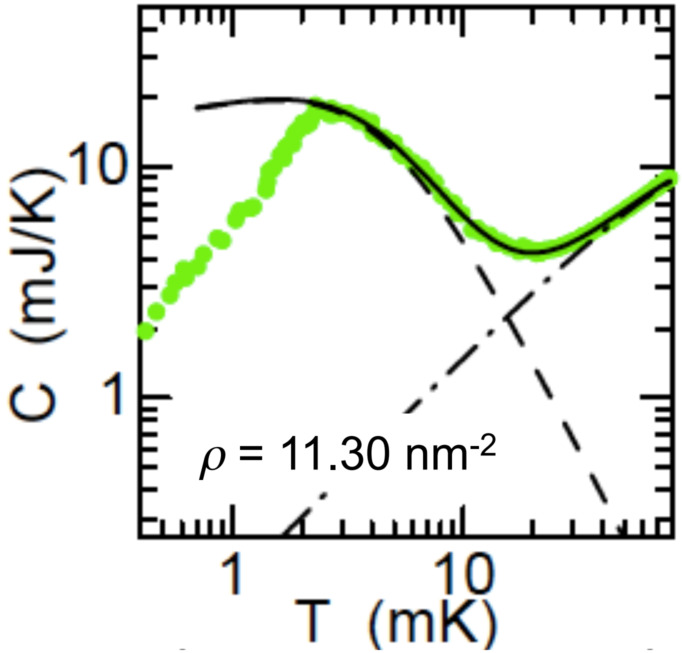
J = 2.1 mK

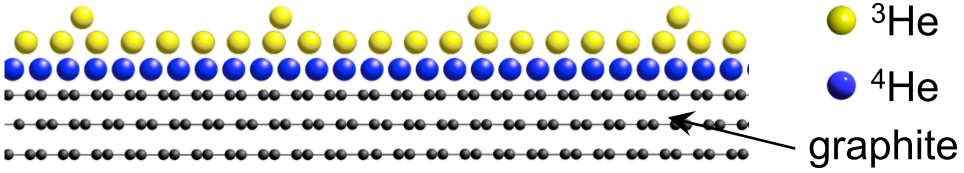


H. Godfrin et al., PRL **60**, 305 (1988)

Heat capacity raw data: 2nd-layer IC(FM) region



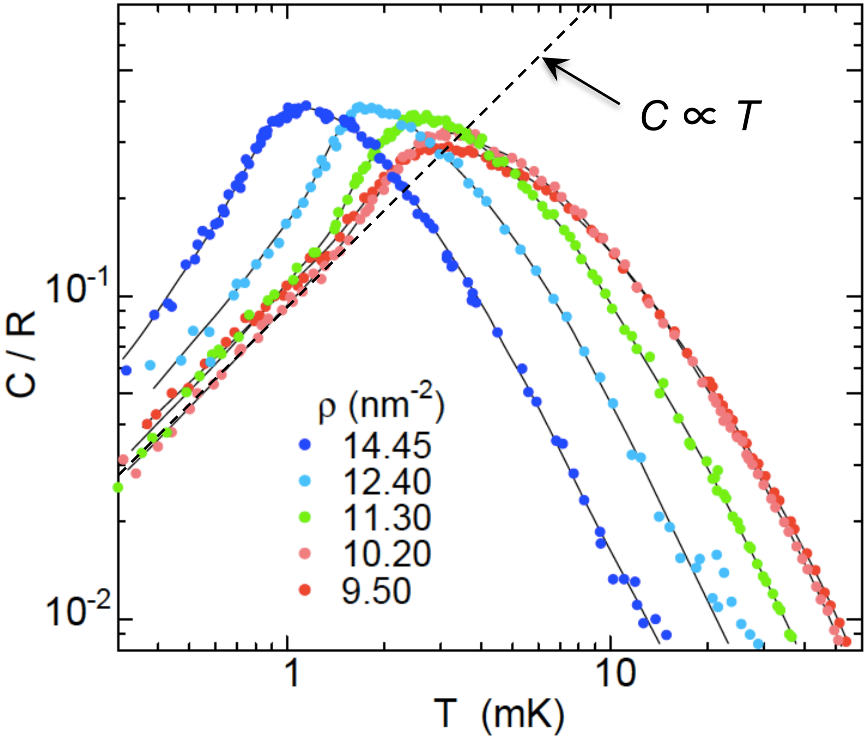


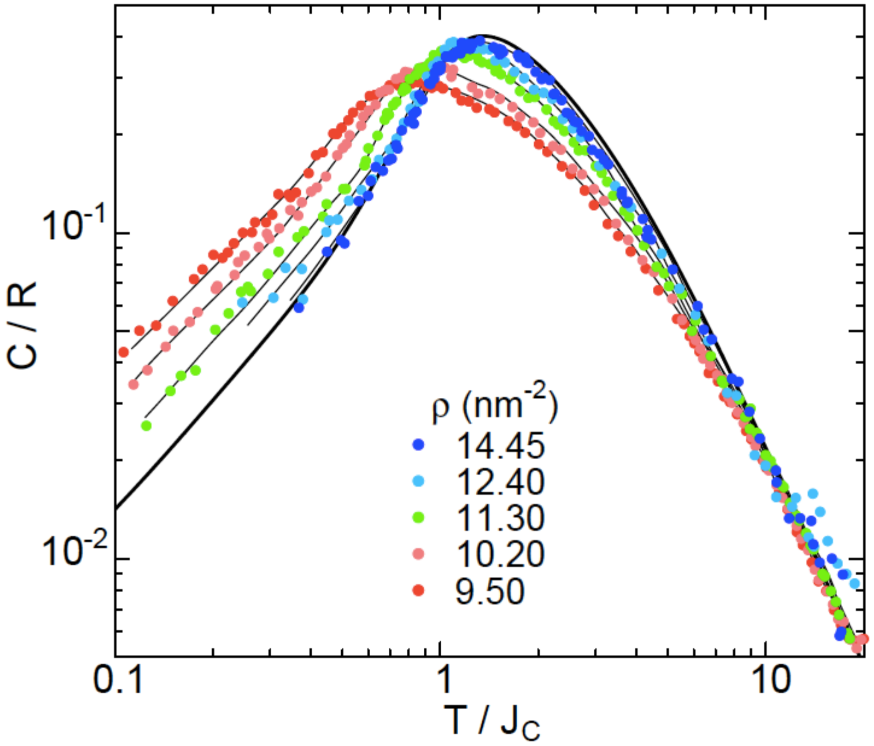


HC contribution from 3rd layer liquid is subtracted to deduce 2nd layer spin HC.

Frustration tunable 2D FM on triangular lattice

Nuclear spin specific heat : 2nd-layer IC (FM) region





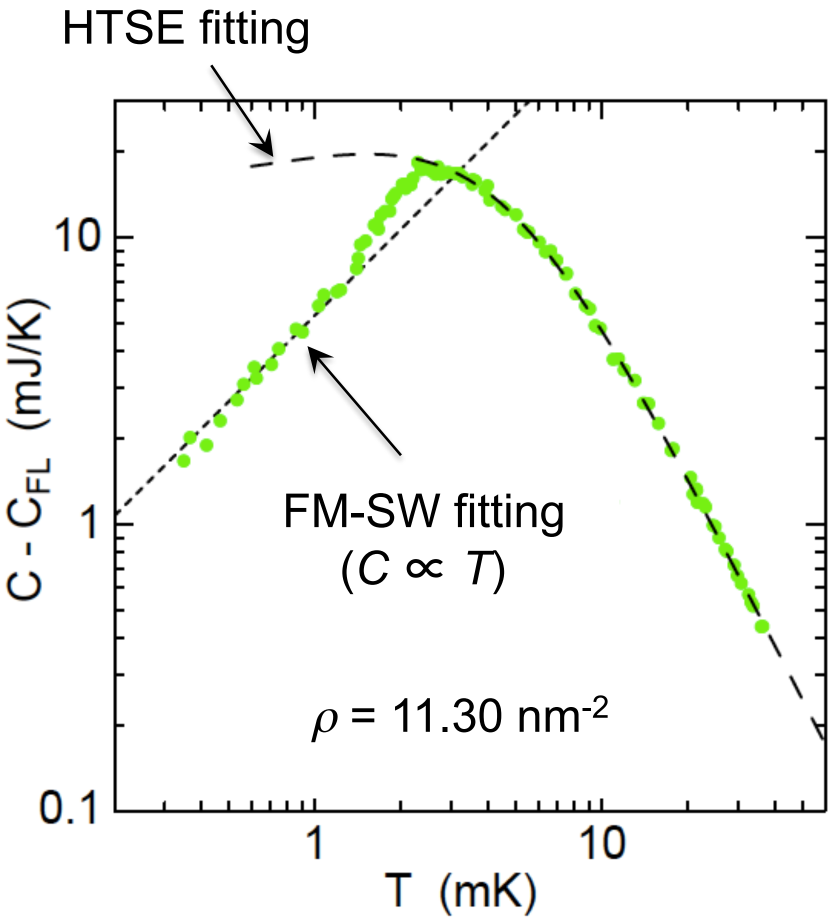
- Single peak in spin heat capacity
- T_{peak} shifts lower-T and peak becomes broader with increasing ρ.
- $C \propto T$ at low-T (FM-SW)

— S = 1/2 Heisenberg ferromagnet on triangular lattice (**HFT**) model

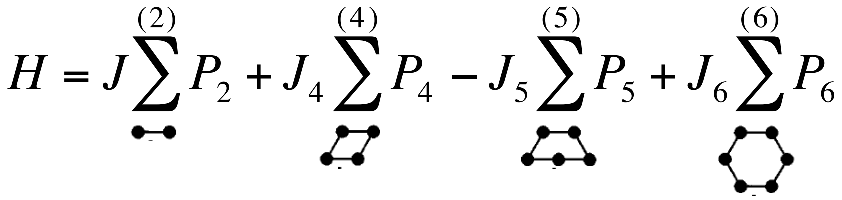
B. Bernu and G. Misguich., PRB **63**, 134409 (2001)

In high-density limit, system behaves almost perfect **HFT**.

Experimental determination of ring exchange (RE) parameters



RE hamiltoniann:



$$H = J\sum^{(2)} P_2 + J_4\sum^{(4)} P_4 - J_5\sum^{(5)} P_5 + J_6\sum^{(6)} P_6$$

High-T series expansion (HTSE} fitting:

high-*T* series expansion of MSE Hamiltonian up to 5th order in β (= 1/*T*)

+

[2,3] Padé approximation after Euler transformation $\beta' = \beta/(1 + \beta)$

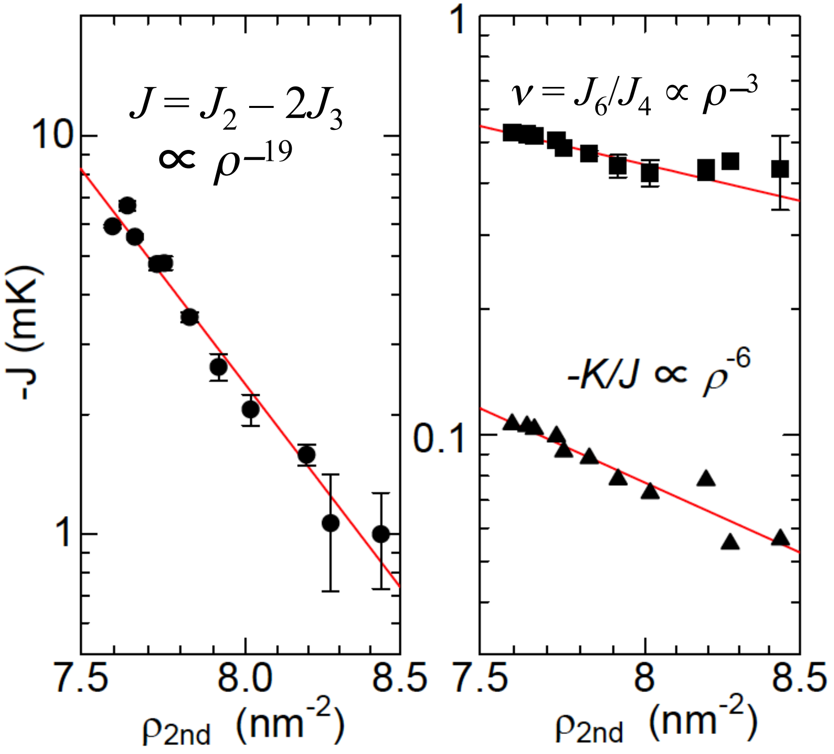
M. Roger, PRB **56**, R2928 (1997)

MSE (fitting) parameters:

$J = J_2 - 2J_3$: effective 2-spin exchange
$K/J = (J_4 - 2J_5)/J$: effective 4-spin exchange
$\mu = J_5/J_4 = 0.3$: 5-spin exchange (fixed)
$\nu = J_6/J_4$: 6-spin exchange

Ring exchange parameters

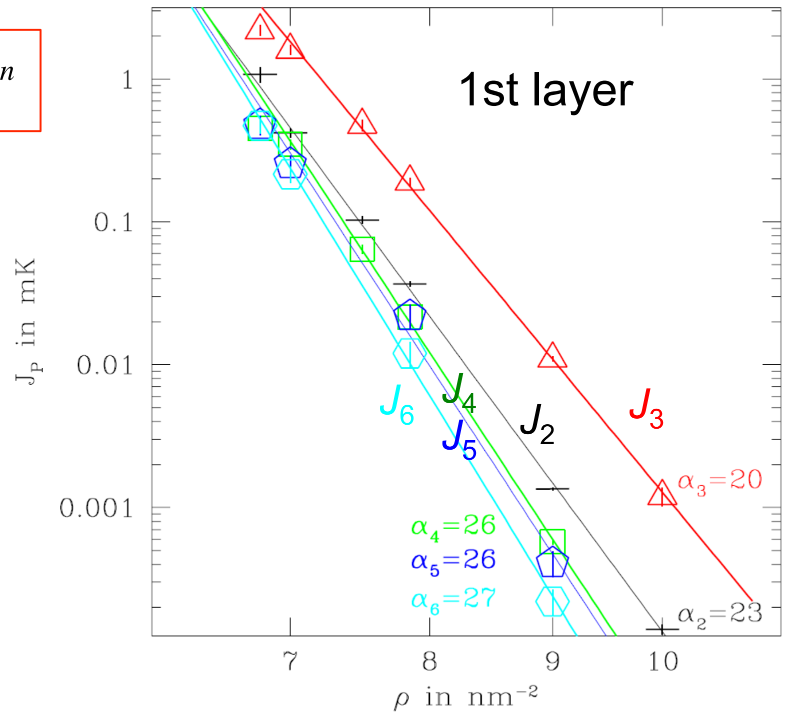
experimental



$$J_n \propto \rho^{-\alpha_n}$$

PIMC calculation

Bernu and Ceperley, JPCM **14**, 9099 (2002)

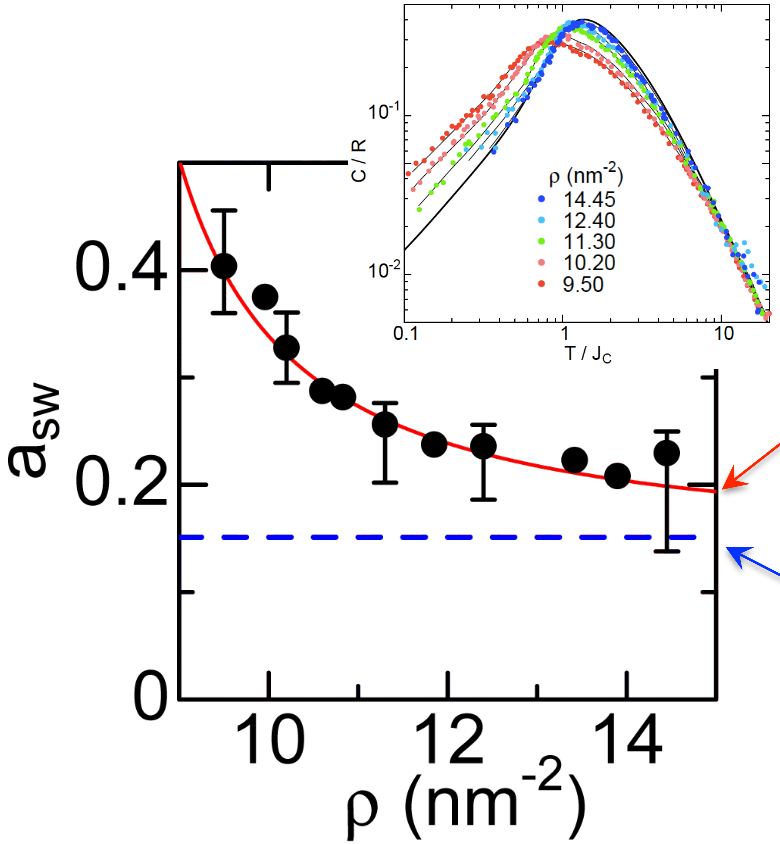


- J (< 0: FM) is dominated by FM J_3 and other J_P decrease much faster than J_3 as density increases.

- Very good agreement between exp. and PIMC cal. for α.

	J	J_4	J_5	J_6
α_n (exp)	19 ± 2	25 ± 2	←	28 ± 3
α_n (PIMC)	20	26	26	27

FM spin wave analysis



Low-*T* heat capacity ($\boldsymbol{C = aT}$) is consistent with FW-SW picture for 2D

RE model M. Roger, PRB **56**, R2928 (1997)

$$a_{\mathrm{RE}} = \pi \left| J + 4J_4 - 10J_5 + 2J_6 \right| / 12\sqrt{3}$$

with RE parameters (J, J_4, J_5, J_6) determined from HT-data

Modified spin-wave theory for HFT

M. Takahashi, Prog. Theor. Phys. Suppl. **87**, 233 (1986)

$$a_{\mathrm{HFT}} = \pi \left| J \right| / 12\sqrt{3}$$

Remarkable agreement between $\boldsymbol{a_{\mathrm{exp}}}$ and $\boldsymbol{a_{\mathrm{RE}}}$.

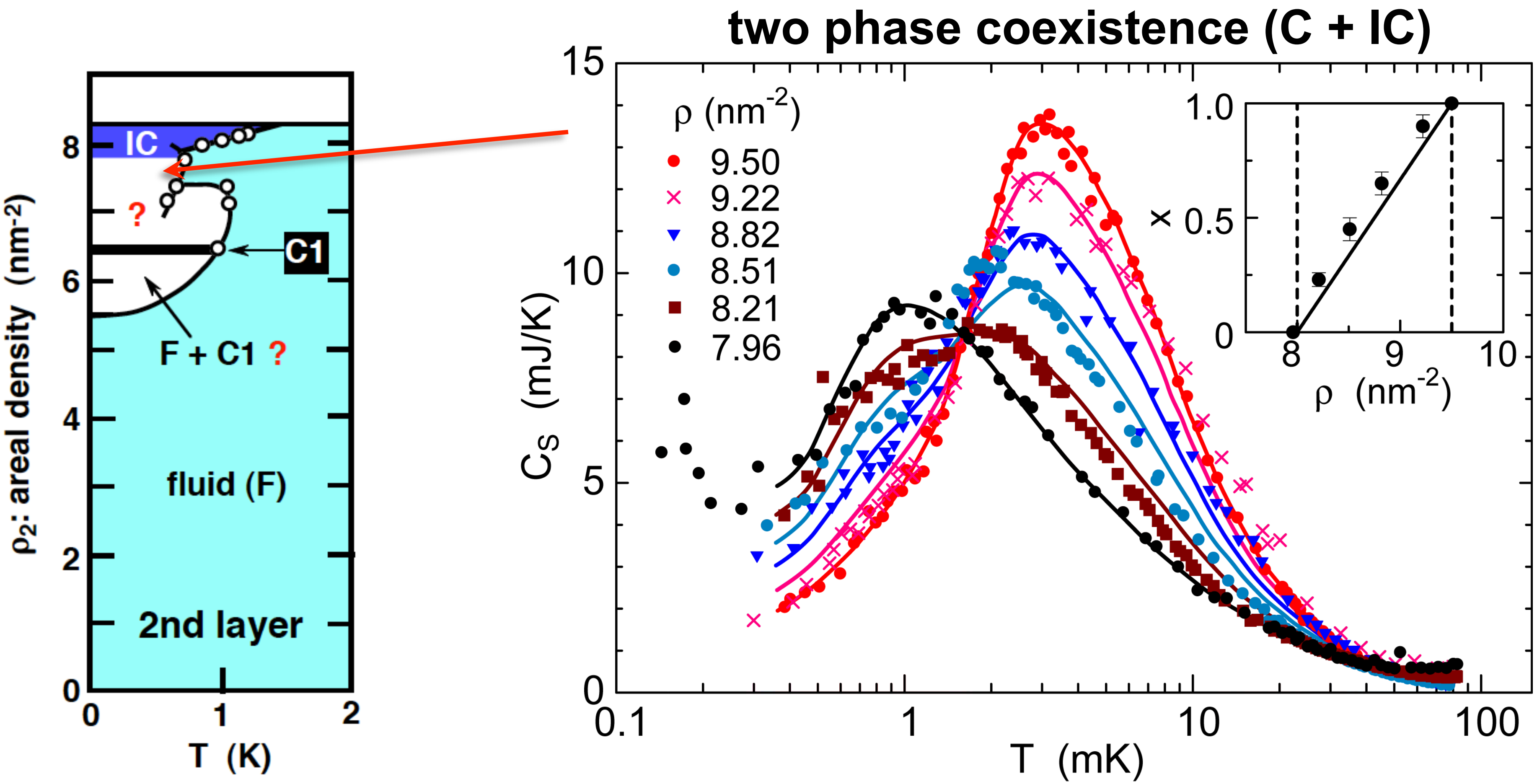
RE model is well applicable to frustrated FM phase in 2nd layer ^{3}He.

C-IC transition: 1st order transition

- Magnetic HCs in C-IC transitional region can be well represented by linear combination of $C_C\,(T)$ and $C_{IC}\,(T)$.

D. Sato et al., JLTP **158**, 201 (2010)
D. Sato et al. to be published

C-IC density change: $\rho_{IC} - \rho_{4/7} = 0.58\ \mathrm{nm}^{-2}$

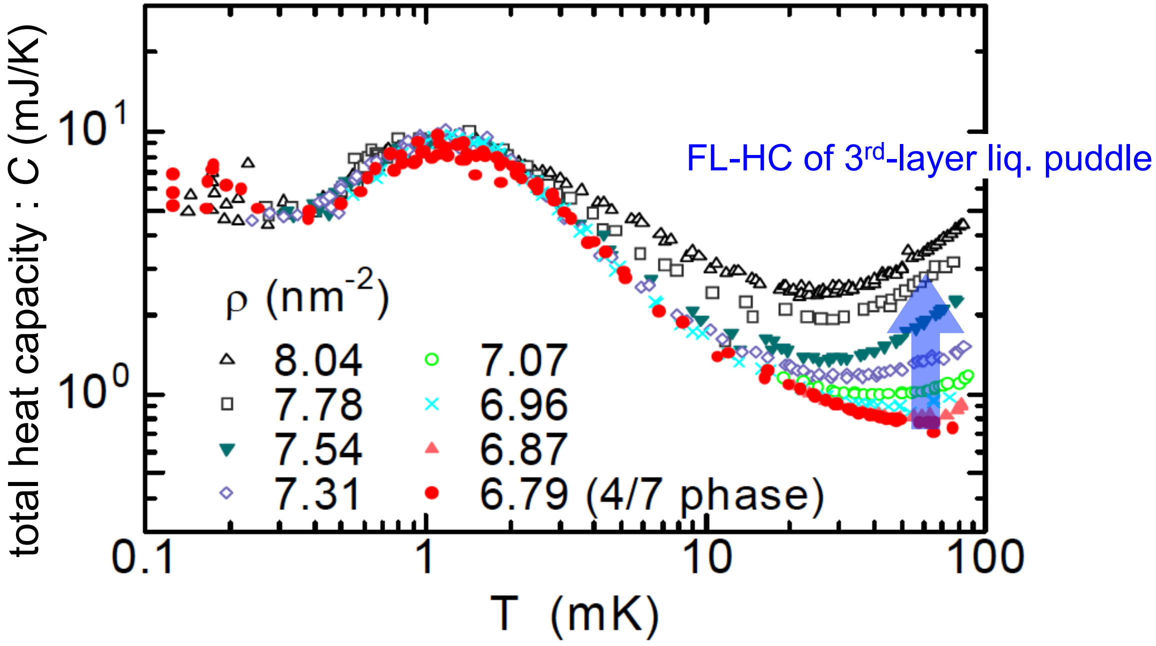


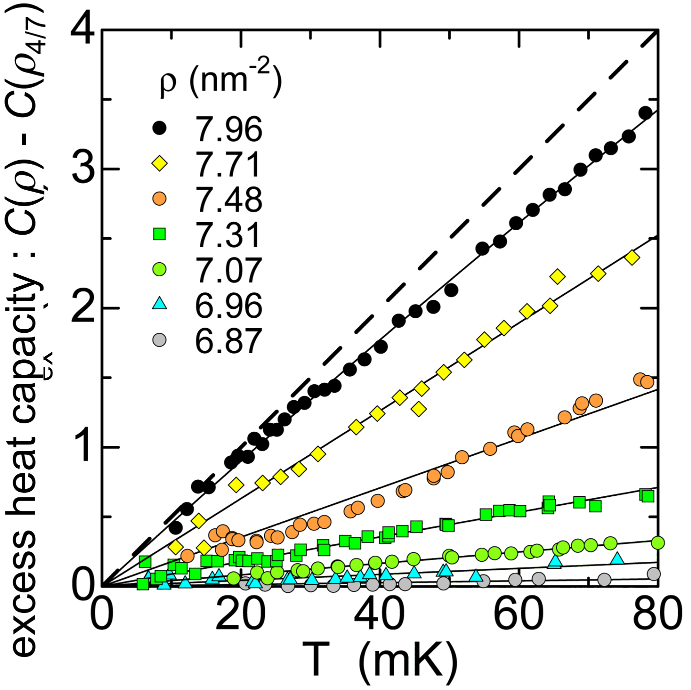
two phase coexistence (C + IC)

4/7 phase with 3rd-layer liquid puddle

D. Sato at al., arXiv:1208.0842v1; JLTP **158**, 201 (2010)

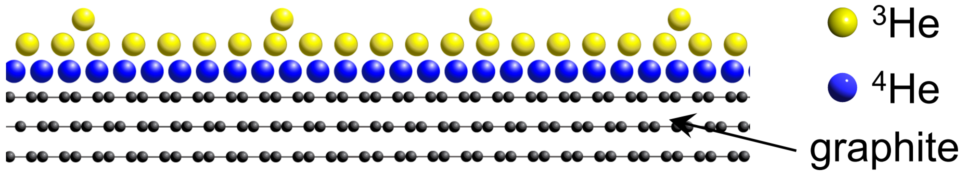
spin HC of 4/7 phase
with double peak





- Only high-T part increases with increasing total ρ by 19%.

- Excess HC varies in proportion to T with γ smaller than γ_0.

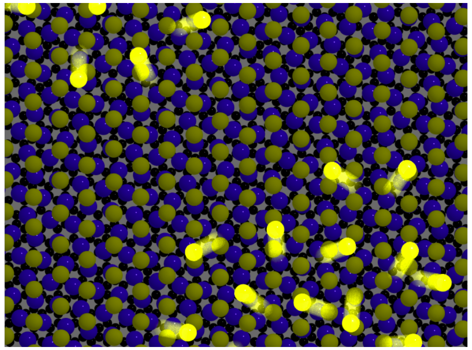


Double peak structure of 4/7 phase does not change with area of 3rd-layer liquid puddle.

Gas-liquid transition in 3rd layer ^{3}He

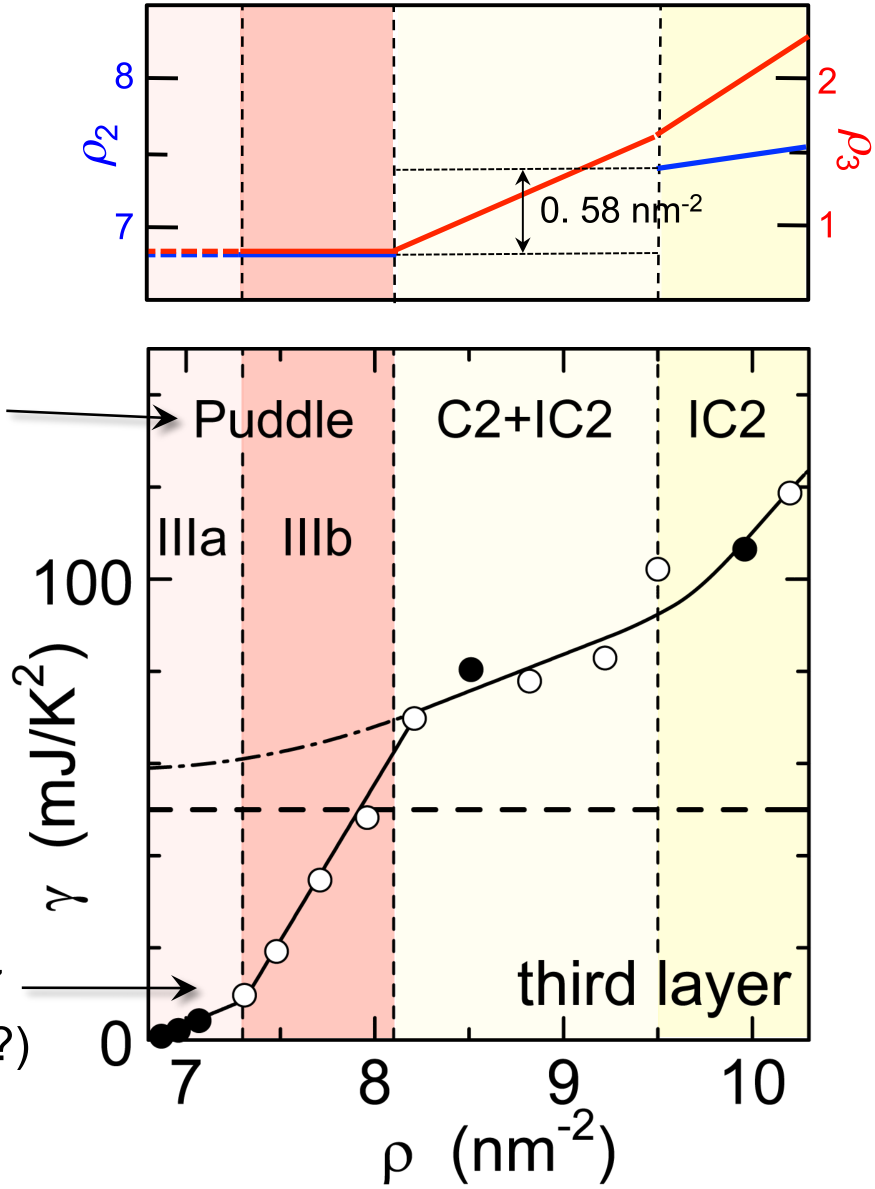
3rd layer puddle phase

$\rho_{puddle} \approx 0.9\ nm^{-2}$



1. 6% compression of 4/7 phase? (or interstitials?)

2. heterogeneity effect?



- ρ-linear increase of γ (two-phase)
- kink in $\gamma(\rho)$ near $m^*/m = 1$

For degenerate 2D Fermi Liquid:

$$C_{FL}(T) = \gamma T + \cdots$$

$$\gamma = (\pi k_B^2/3\hbar^2) A m^*$$

A : surface area

m^* : QP effective mass

● D. Sato at al., arXiv:1208.0842v1

○ D. Sato at al., JLTP **158**, 201 (2010)

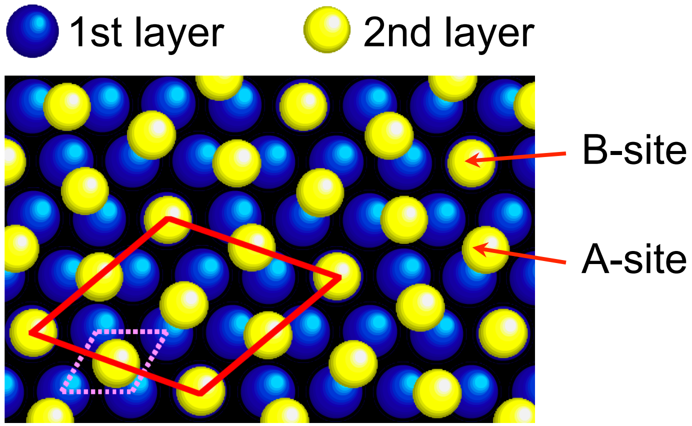
3. 2D gapless QSL state in 4/7 phase

4/7 phase: low-density commensurate (C) solid of He in 2nd layer on graphite

$\sqrt{7}\times\sqrt{7}$ structure

- **Detailed commensurability** differs depending on PIMC calculations.

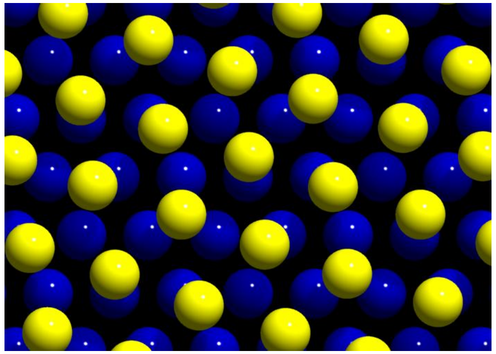
Elser model



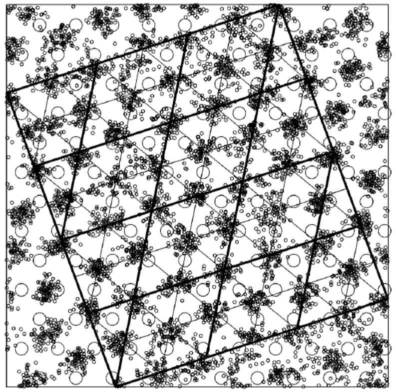
V. Elser, PRL **62**, 2405 (1989)

F.F. Abraham et al., Europhys. Lett. **12**, 107 (1990)

Takagi model



T. Takagi, J. Phys. Conf. Series **150**, 032102 (2009)

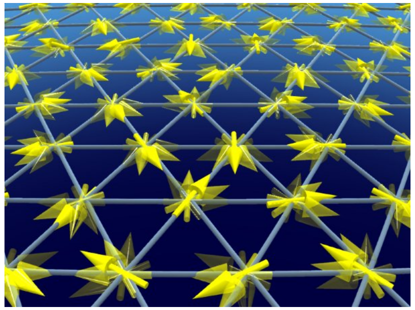


M. Pierce and E. Manousakis, PRL **81**, 156 (1998); PRB **59**, 3802 (1999)

Neutron scattering exp. failed to detect Bragg peaks of 4/7 phase. H. J. Lauter et al., Can. J. Phys. **65**,1435(1987)

- large Debye-Waller factor
- huge B.G. from graphite

Gapless spin-liquid behavior in 4/7 phase



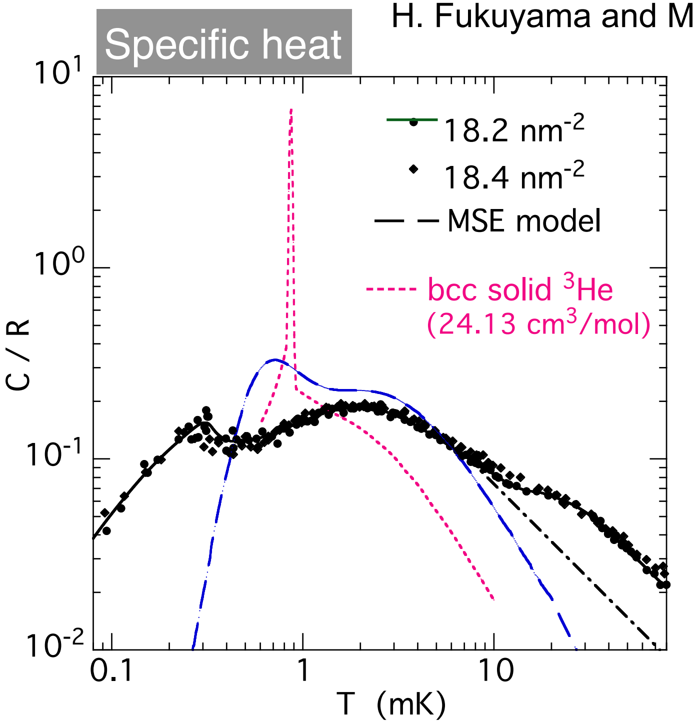
Gapless QSL in 2D

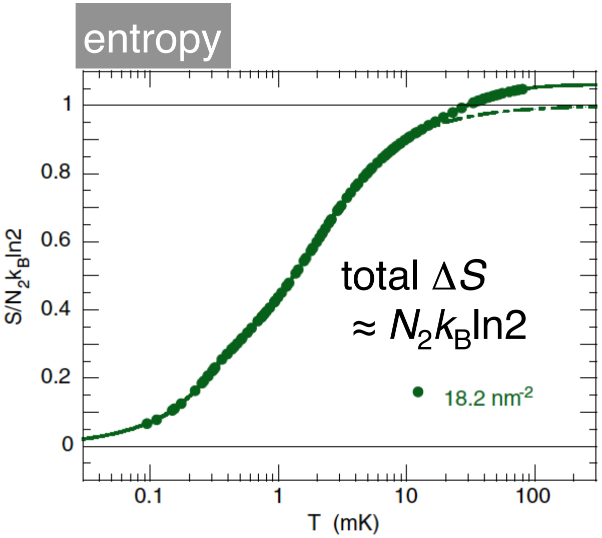
($J \approx 10$ mK)

- No finite-T phase transitions down to $\boldsymbol{T/J \approx 10^{-2}\text{-}3\text{X}10^{-4}}$ … truly 2D
- Double peak in $C(T)$ and $C \propto T$ at low-T … highly frustrated
- No exponential decay ($\chi \rightarrow$ const. as $T \rightarrow 0$) at $T << J$ … gapless spin excitation

First proposed in heat capacity measurements

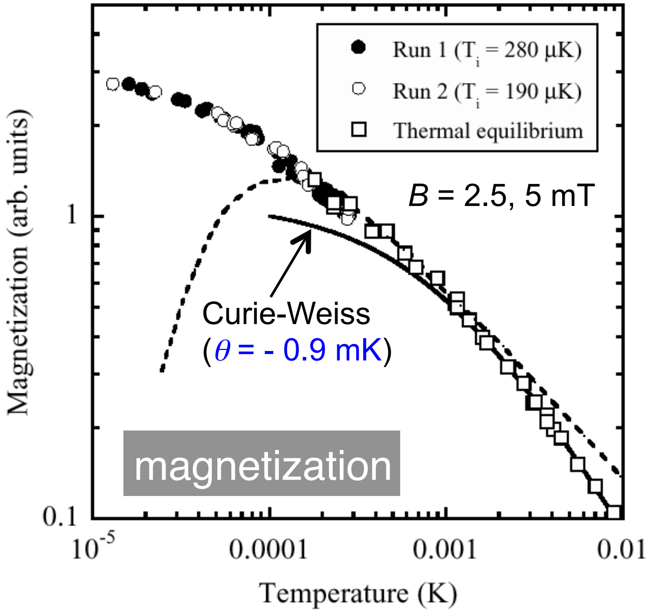
K. Ishida, H. Fukuyama, et al., PRL **79**, 3451 (1997)
H. Fukuyama and M. Morishita, Physica B **280**, 104 (2000)





And, then confirmed by magnetic susceptibility measurements

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



Early history of gapless QSL in 2D

2D ^{3}He

1989 D.S. Greywall
- highly frustrated (missing entropy?)

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

1990 M. Roger
- ring exchange model (4-spin) on triangular lattice

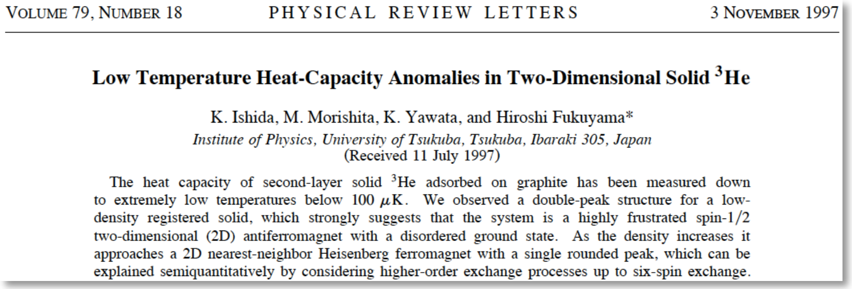
1997 K. Ishida, H. Fukuyama et al.
- double-peak in $C(T)$ and $C \propto T$ down to $T = 10^{-2}\,J$
- "*fully frustrated disordered ground-state*"

1997, 1999 T. Momoi, K. Kubo et al.
- scalar chiral order
- prediction of 1/2 magnetization plateau (RE model with 4-spin)

1998, 1999 G. Misguich, C. Lhuillier et al.
- gapful QSL (exact diagonalization of RE model with 6-spin)

2000 H. Fukuyama
- proposal of gapless QSL

2004 R. Masutomi, H. Ishimoto et al.
- gapless QSL (M measurement down to $T = (10^{-3}\text{-}10^{-4})J$)



VOLUME 79, NUMBER 18 PHYSICAL REVIEW LETTERS 3 NOVEMBER 1997

Low Temperature Heat-Capacity Anomalies in Two-Dimensional Solid ^{3}He

K. Ishida, M. Morishita, K. Yawata, and Hiroshi Fukuyama*

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(Received 11 July 1997)

The heat capacity of second-layer solid ^{3}He adsorbed on graphite has been measured down to extremely low temperatures below 100 μK. We observed a double-peak structure for a low-density registered solid, which strongly suggests that the system is a highly frustrated spin-1/2 two-dimensional (2D) antiferromagnet with a disordered ground state. As the density increases it approaches a 2D nearest-neighbor Heisenberg ferromagnet with a single rounded peak, which can be explained semiquantitatively by considering higher-order exchange processes up to six-spin exchange.

electronic systems

S = ½ 1D HA-chain [Cu($C_4H_4N_2$)(NO_3)$_2$]

1999 P.R. Hammar et al.
- No LRO down to T = $10^{-2}J$
- gapless QSL ($C(T) \propto T$ at low T)

S = ½ 2D HAT [κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$]

2003 Y. Shimizu, K. Kanoda
- No LRO down to $T = 10^{-4}J$
- gapless QSL ($T_1^{-1} \propto T$)

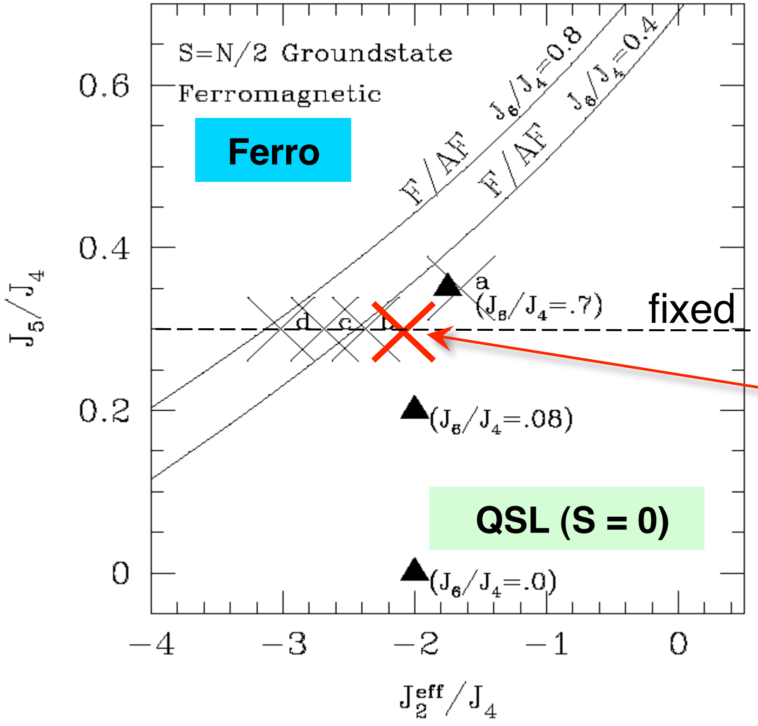
Ring exchange model for 2D ^{3}He

Exact diagonalization of MSE Hamiltonian including J_6

G. Misguich et al., PRL **81**, 1098 (1998); PRB **60**, 1064 (1999)

- gapful QSL ($\Delta \approx J_4$)
- Δ diminishes near the FM-QSL boundary ?
- finite size effect (36 spins)?

G. Misguich, Ph. D. thesis (1999)



RE parameters **extrapolated** to 4/7 phase density from IC phase in ^{3}He/^{4}He/gr

J = -40 mK

J_4 = 19 mK

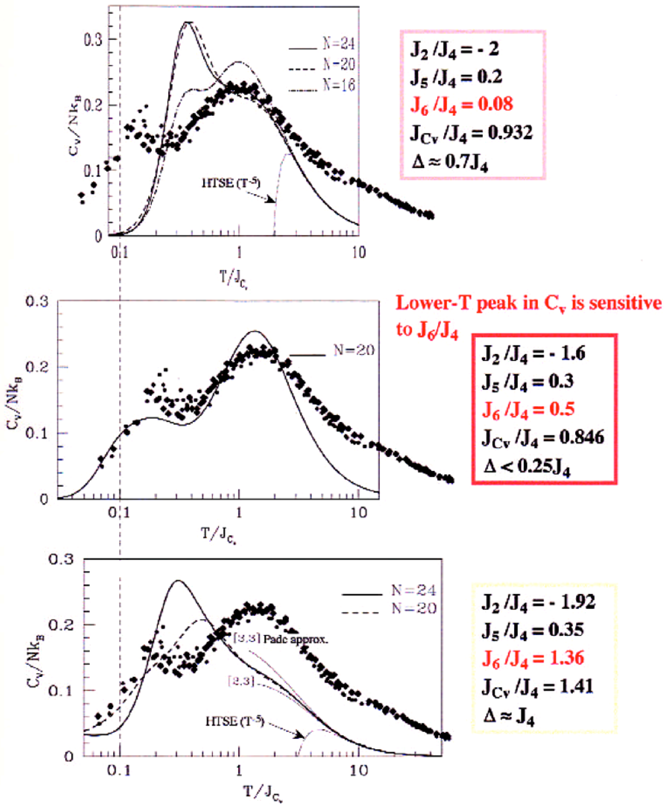
J_5 = 5.7 mK

J_6 = 12 mK

$\nu = J_6/J_4 = 0.63$

$\mu = J_5/J_4 = 0.3$ (fixed)

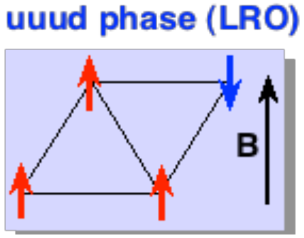
, which are too large presumably because of larger potential barriers in C phase.

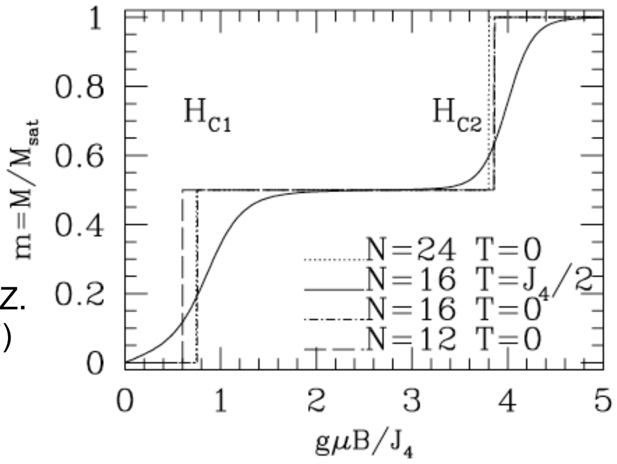


So far, anomalous high-T behavior prevents us to deduce RE parameters of 4/7 phase directly by analyzing HC data with HTSE.

Magnetization curve of 4/7 phase

(Narrow) magnetization plateau at $M = M_{sat}/2$
→ importance of J_4





K. Kubo and T. Momoi, Z. Phys. B **103**, 485 (1997)

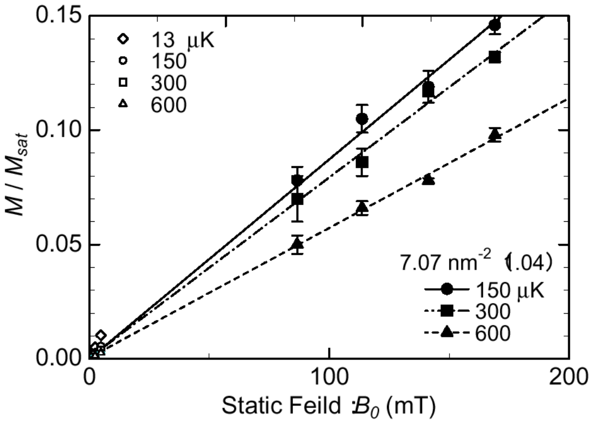
G. Misguich et al., PRL **81**, 1098 (1998)

Truly gapless

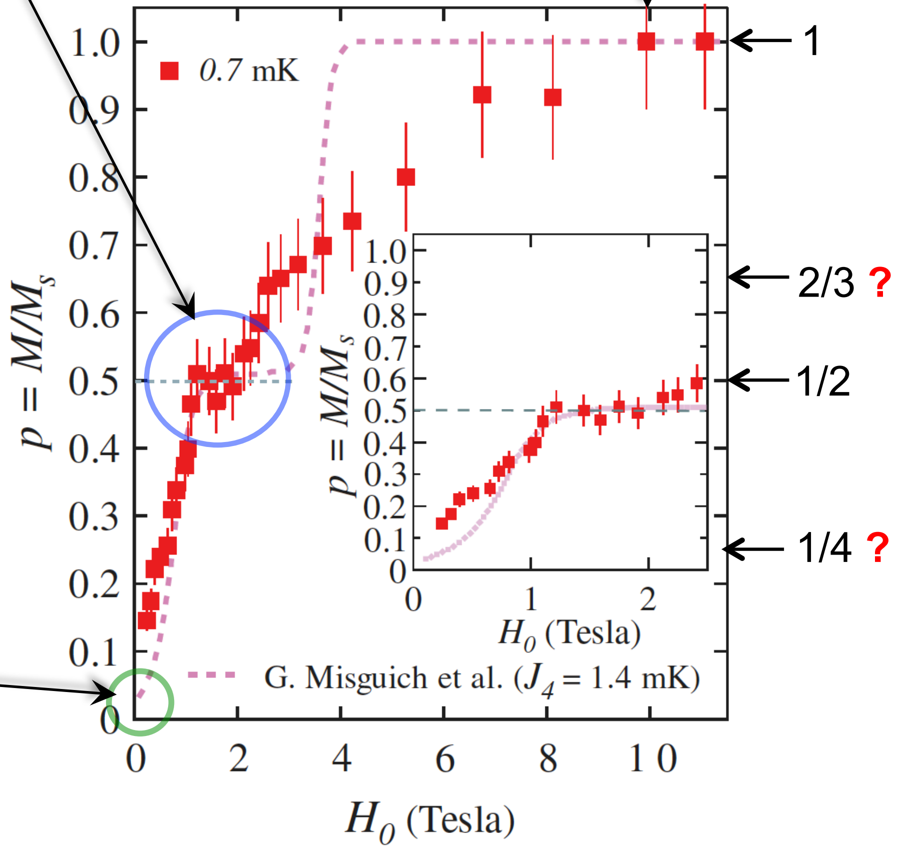
$$\mu B \leq 10^{-4} J$$

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

S. Murakawa, Ph.D. thesis, Univ. of Tokyo (2006, unpublished)



upper critical field ($H_{c2} \approx 10$ T)
$|\theta| << \mu H_{c2} \approx |J|$ ← highly frustrated



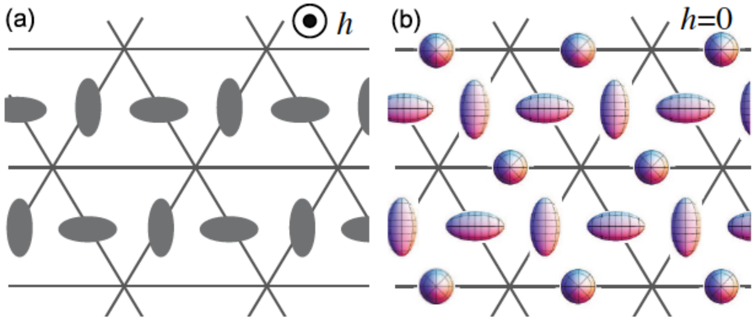
H. Nema, H. Ishimoto et al., Phys. Rev. Lett. **102**, 075301 (2009)

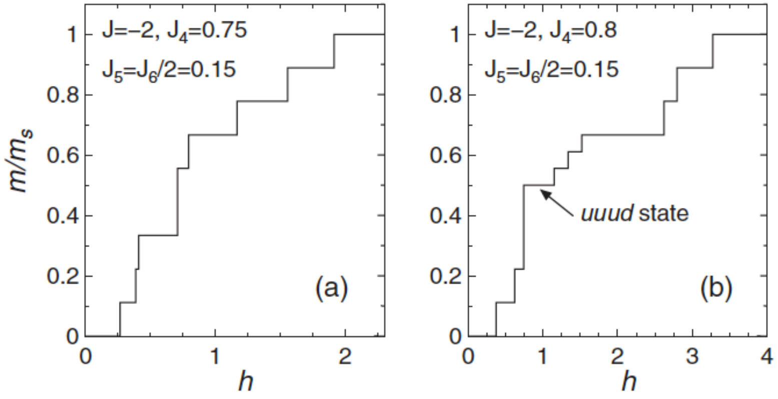
Recent theories

1. Ring exchange (MSE) model

T. Momoi et al., Phys. Rev. Lett. **108**, 057206 (2012)

octahedral spin nematic state
with bond antiferroquadrupolar order

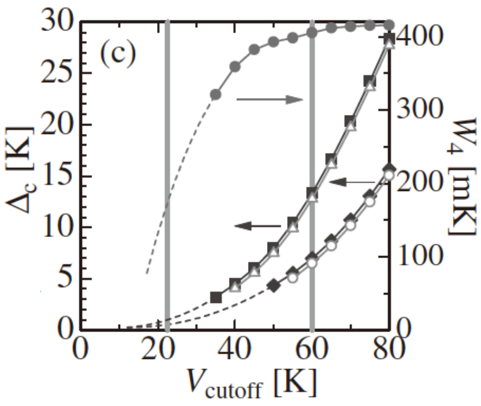




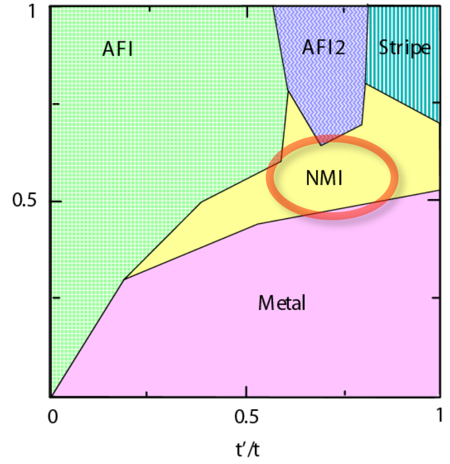
2. Hubbard model

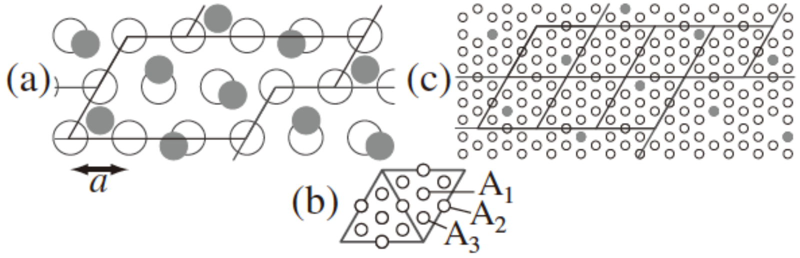
S. Watanabe and M. Imada, J. Phys. Soc. Jpn. **78**, 033603 (2009)

density fluctuations due to virtual promotion
to 3rd layer



T. Mizusaki and M. Imada,
Phys. Rev. B **74**, 0144213 (2006)

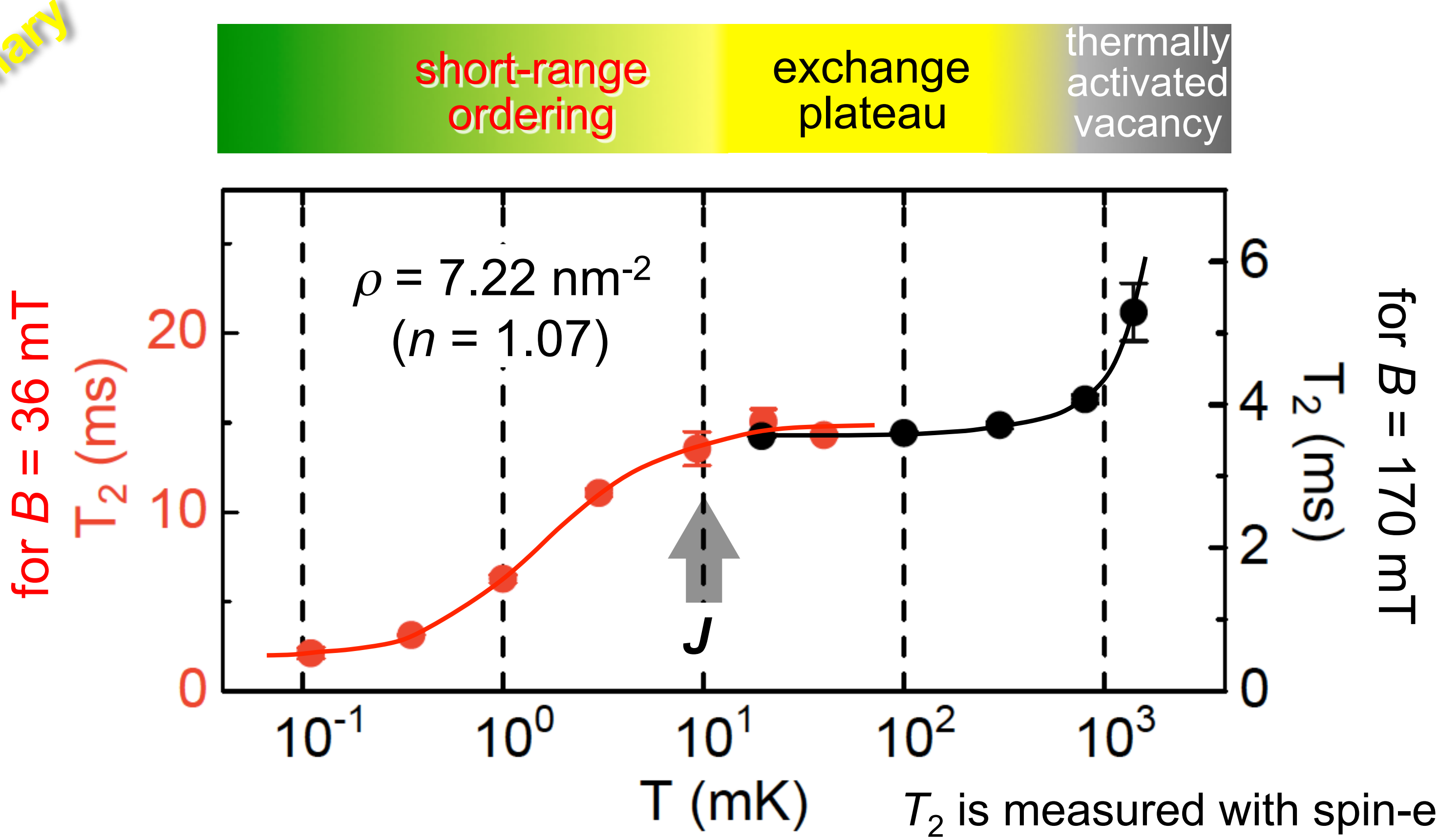




Spin-spin relaxation time (T_2) in 4/7phase: *T*-dependence

D. Sato, Ph.D. thesis, Univ. of Tokyo (2012, unpublished)

preliminary

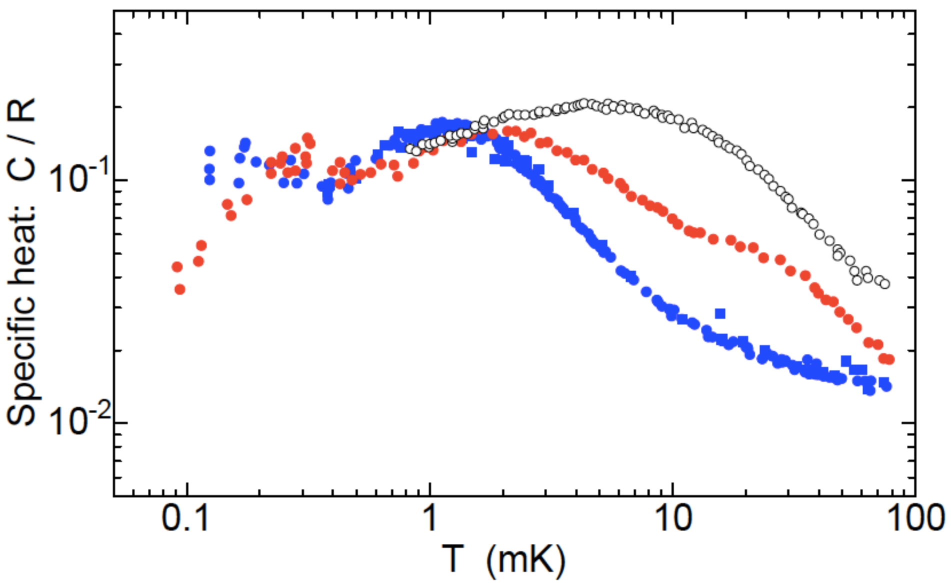


T_2 is measured with spin-echo method

- Gradual transformation from the paramagnetic state at high-*T* to gapless SL state at *T* = 0
- No abrupt change (absence of finite-*T* transition)

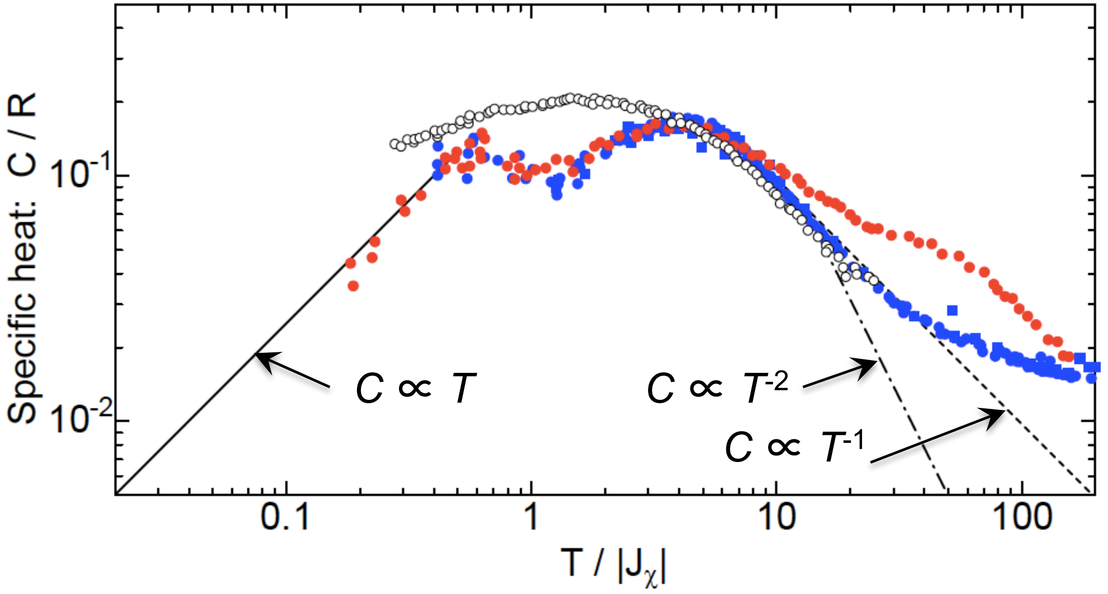
Call for microscopic theory of T_2

4/7 phase on different underlayers



- 4/7 phase exists at least three different underlayers: ^{4}He, ^{3}He and bilayer HD
- ^{3}He/HD/HD/gr system has ten time larger *J* (≈ 30 mK) with single peak.

		J_χ	$\rho_{4/7}$
○	on bilayer HD (Casey et al., 1998)	(3.0 mK,	5.4 nm^{-2})
●	on ^{3}He (Ishida et al., 1997)	(0.5 mK,	6.4 nm^{-2})
●	on ^{4}He (Sato et al., 2010)	(0.3 mK,	6.8 nm^{-2})
■	on ^{4}He (Matsumoto et al., 2005)	(0.3 mK,	6.8 nm^{-2})

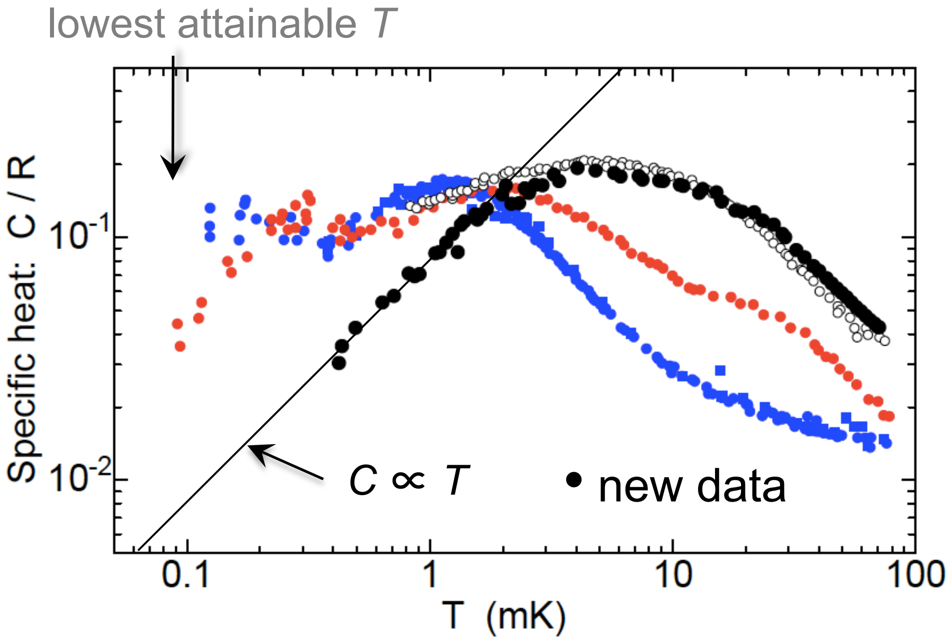


Problems:

- *T*-linear region so far observed is rather narrow.
- anomalous behavior at high-*T*

New HC data of 4/7 phase on bilayer HD

preliminary



- single peak*!*
- $C \propto T$ at low-T as in other systems

	J_χ	$\rho_{4/7}$
● on bilayer HD (This work)	(3.0 mK,	5.4 nm^{-2})
○ on bilayer HD (Casey et al., 1998)	(3.0 mK,	5.4 nm^{-2})
● on ^{3}He (Ishida et al., 1997)	(0.5 mK,	6.4 nm^{-2})
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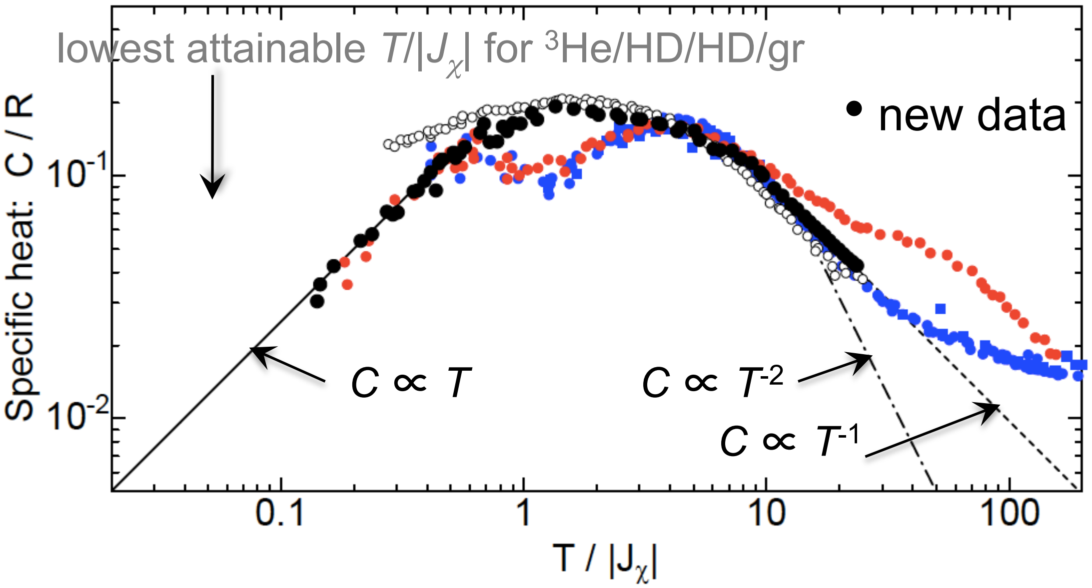
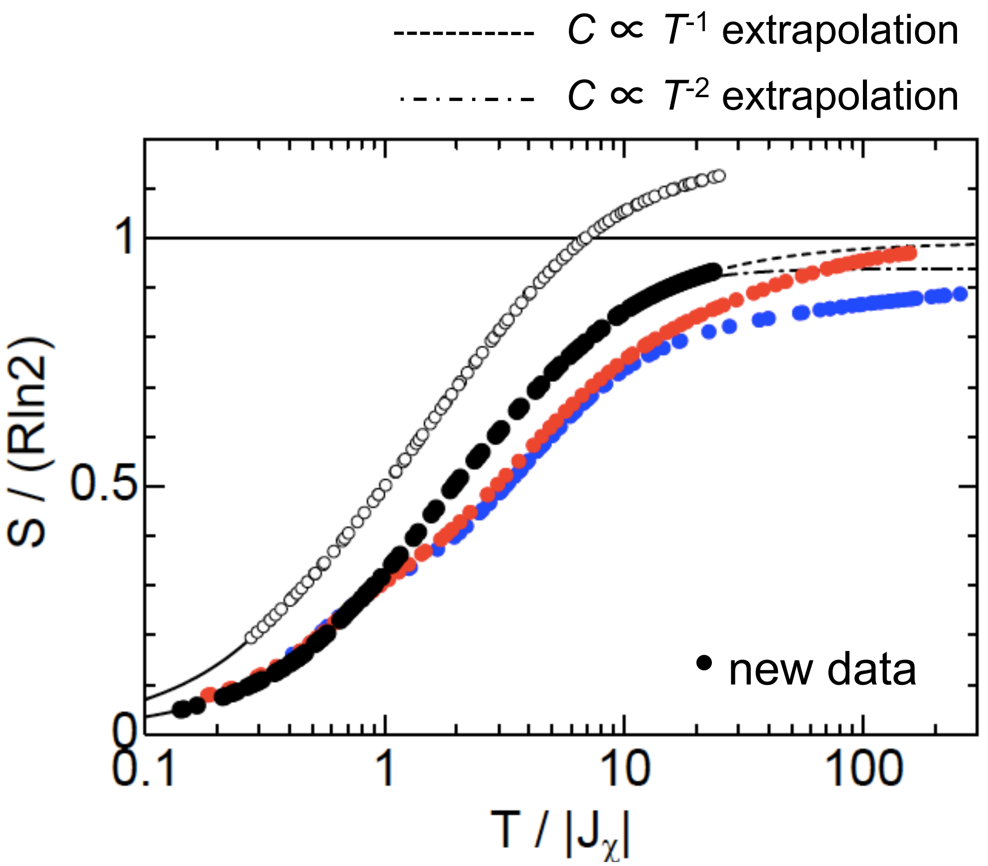


On going and future experiments:

- extending lowest-T data down to 0.1 mK soon and highest-T data up to 500 mK in future.
- study of lattice anisotropy:
  - bilayer H_2: $a/b = 1.116$
  - bilayer D_2: $a/b = 1.078$
  - monolayer hydrogen: $a/b = 1$

Spin entropies in 4/7 phase on different underlayers



		J_χ	$\rho_{4/7}$
●	on bilayer HD (This work)	(3.0 mK,	5.4 nm^{-2})
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■	on ^{4}He (Matsumoto et al., 2005)	(0.3 mK,	6.8 nm^{-2})

- Our new data on ^{3}He/HD/HD/gr seem to show expected total entropy change (= *R*ln2).

- Wilson ratio at $C \propto T$ regime is around 7 (using χ data at T = 0.4 mK by Masutomi et al., 2004).

→ spinon FS ?

Summary

Bcc solid ^{3}He

- Experimental properties are explained semi-quantitatively with RE model considering up to J_6.
- However, recent PIMC calculations show rather slow convergence of B_{c2} and θ against much higher REs.

Frustrated FM in 2D IC solid ^{3}He on graphite

- Determined RE parameters experimentally up to J_6 with good precisions.
- Such RE parameters explain whole T–dependence of specific heat.
- Approach pure HFT model as density increases.

Gapless QSL state in ^{3}He 4/7 phase on graphite

- No LRO and gap down to $10^{-4}J$.
- RE parameters obtained by extrapolation from IC phase seem to be consistent with theoretical QSL or nematic phases.
- Preliminary T_2 data support the gapless QSL picture.
- Preliminary HC data for 4/7 phase on bilayer HD show interesting feature: very broad single peak and $C \propto T$ behavior at low-T with Wilson ratio of 7.