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Bipartite Elementary Excitations in a Two-dimensional Quantum Spin Liquid

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Elementary excitations in a two-dimensional candidate quantum spin liquid

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Outline

1. Introduction

2. A possible quantum spin liquid state of 2D triangular lattices in organic insulators

   κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ (ET)

   EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ (MIT)

3. Highly unusual elementary excitations in a QSL state

4. Comparison between experiment and theory

5. Summary

M.Yamashita, Y.M. *et al*. Nature Phys. **5**, 44 (2009)

M. Yamashita, Y.M. *et al*. Science **328**, 1246 (2010)

Introduction

Exotic spin state

Liquid, Ice, Nemastic, Chiral, …

Spin Liquids are states which do not break any simple symmetry: neither spin-rotational symmetry nor lattice symmetry.

Quantum spin liquid (QSL)

A novel state of matter where strong quantum fluctuations destroy the long-range magnetic order even at T=0

QSL has attracted much theoretical attention since its proposal in 1973.

The problem of the quantum spin liquid may have close relation to the high-T_c problem

P.W. Anderson, Mater. Res. Bull. 8, 153 (1973)
P.W. Anderson, Science 235, 1196 (1987)

Introduction : Quantum spin liquid (QSL)

One dimension

Notion of QSL is firmly established

Spinon excitation (S=1/2, e=0)

Two and three dimensions

Geometrical frustration

Classical A large ground-state degeneracy

Quantum Quantum fluctuation lifts the degeneracy and a QSL ground state may appear

There are few candidates of the real materials.

2D triangular lattice

^{3}He on graphite
Organic insulators

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$
$EtMe_3Sb[Pd(dmit)_2]_2$

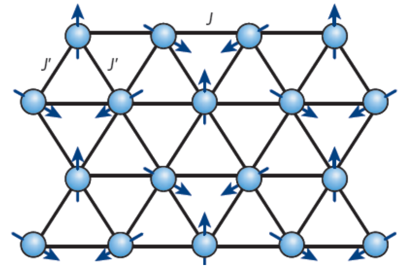
2D kagome lattice

$ZnCu_3(OH)_6Cl_2$ (Herbertsmithite)
$BaCu_3V_2O_8(OH)_2$ (Vesigniete)

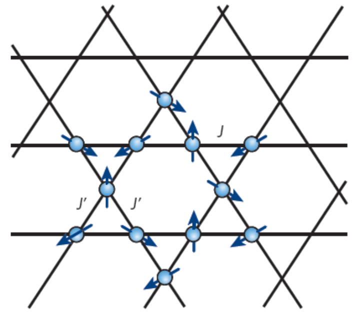
Hyperkagome

$Na_4Ir_3O_8$

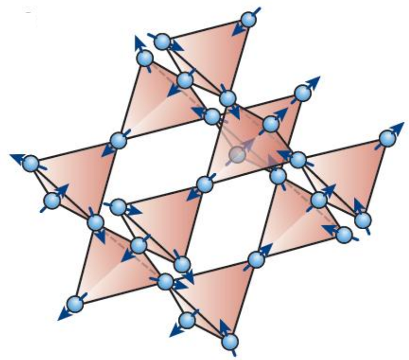
Triangular lattice



kagome lattice



Pyrochlore lattice

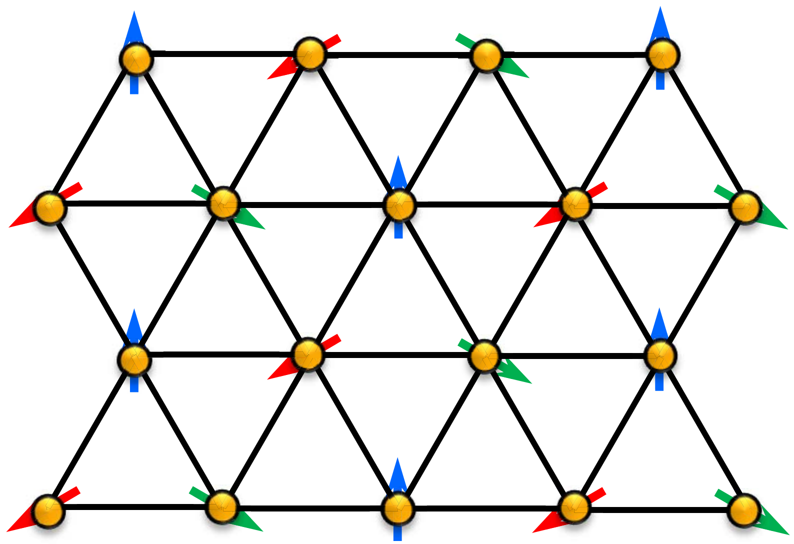


L. Balents, Nature 464, 199 (10)

2D triangular lattice

The 120 degrees antiferromagnetic Néel structure

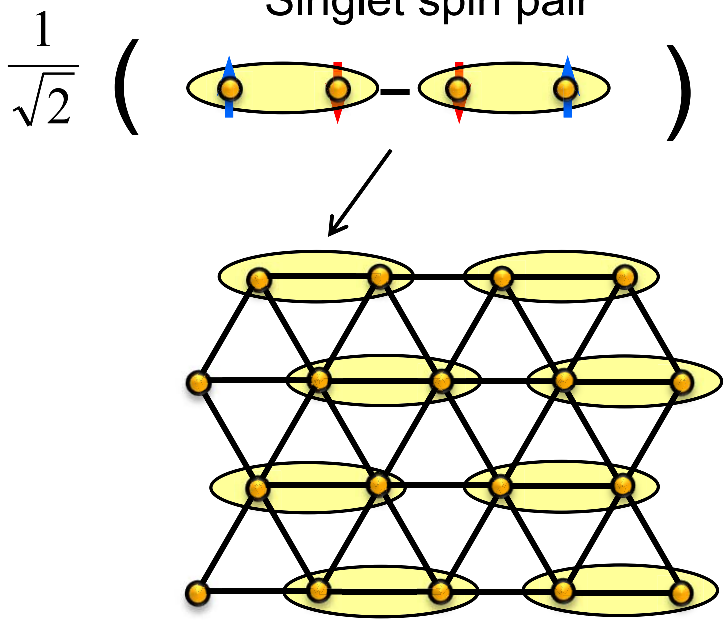
Three sublattice Néel state



2D triangular lattice

Valence Bond Solid

Singlet spin pair



Breaking of the lattice symmetry

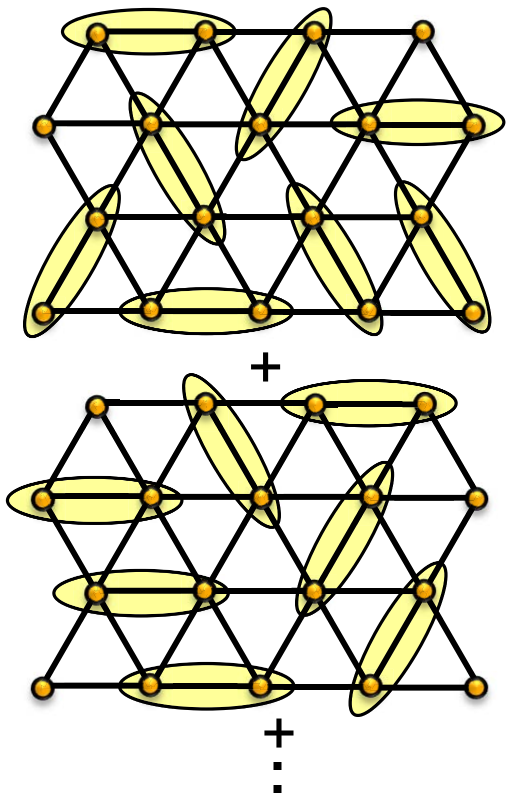
Localized singlets

Long range order of singlet

Resonating Valence Bond (RVB) Liquid

Superposition of different configurations

Resonation between highly degenerated spin configurations leads to a liquid-like wavefunction.

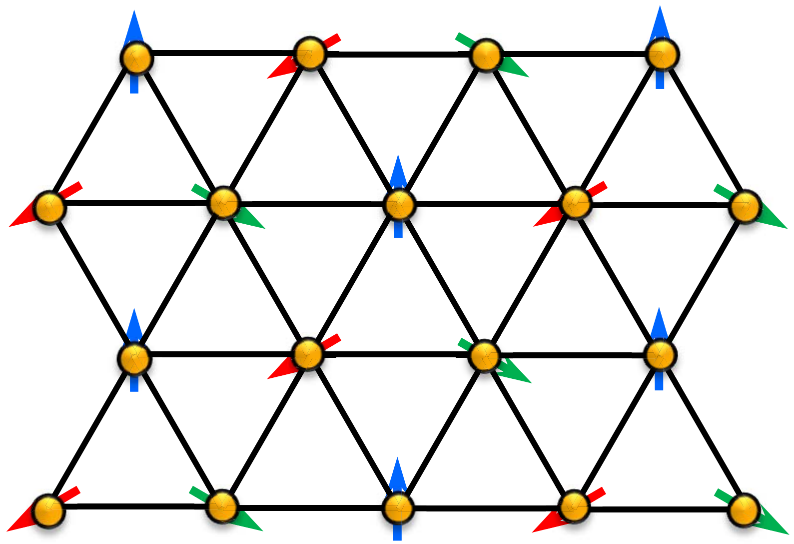


No simple symmetry breaking

P. Fazekas and P.W. Anderson, Philos. Mag. 30, 423 (1974)

2D triangular lattice

Neel order at T=0 even for nearest Heisenberg model with S=1/2



Three sublattice N'eel state

D. Huse and V. Elser, PRL 60, 2531 (88)
B. Bernu, C. Lhuillier and L. Pierre, PRL 69, 2590 (92)
L. Caprioti, A. E.E. Trumper and S. Sorella, PRL 82, 3899 (99)

In real systems, the Neel order occurs at finite temperature due to small but finite interlayer interaction

Additional frustration that enhances the quantum fluctuation is necessary to realize a QSL.

Introduction

How can we identify a QSL in the experiments?

If no symmetry breaking (magnetic order) is observed down to very low temperatures, the system is assumed to be in a QSL state.

What is the elementary excitation of QSLs in the 2D triangular lattice?

Does a QSL host some exotic excitations?

Gapped or gapless?

Magnetic or nonmagnetic?

Localized or itinerant?

If itinerant, what is the statistics, fermion, boson or some exotic?

A wide range of experimental probes are available.

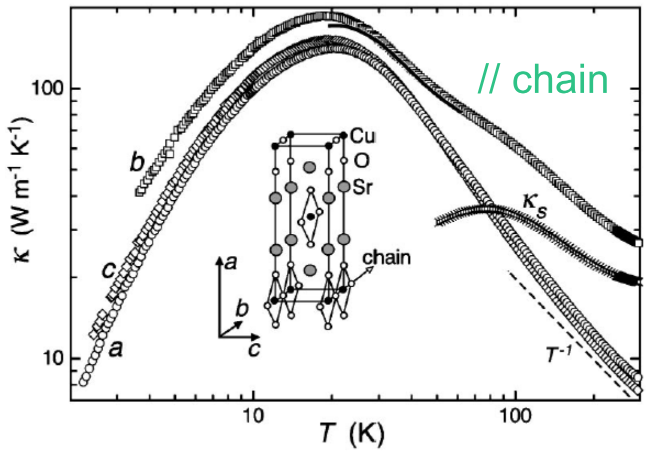
NMR, μSR, Susceptibility, Torque･･･

Specific heat, Thermal conductivity･･･

What the thermal conductivity tells us

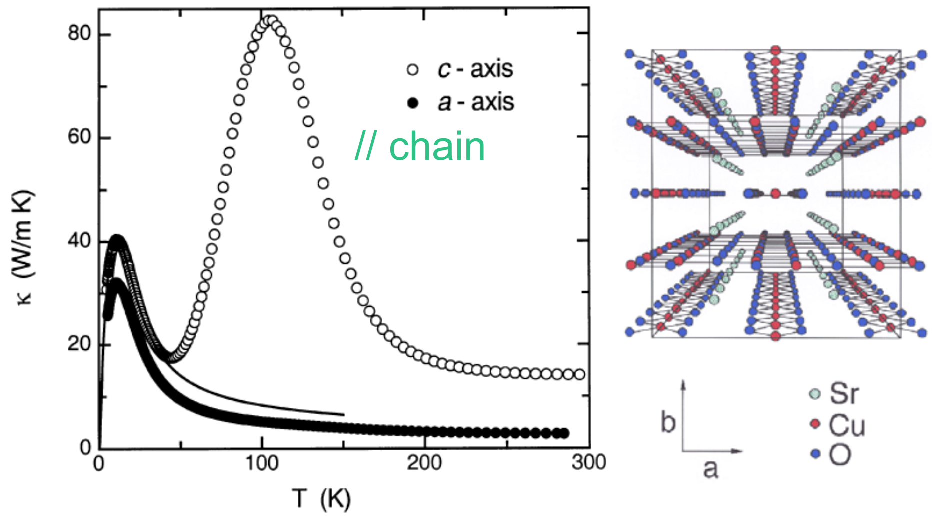
Thermal conductivity provides important information on the elementary excitations, similar to the electronic transport properties in metals.

1D Heisenberg S=1/2 Sr_2CuO_3



A.V. Sologubenko *et al*. PRB 62, R6108 (00)

Spin ladder compound $Sr_{12}Ca_2Cu_{24}O_{41}$



A.V. Sologubenko *et al*. PRL 84, 2714 (00)

Large heat transport by magnetic excitation (spinon) along the 1D chain

What the thermal conductivity tells us

Very powerful probe for *low-energy* and *itinerant* excitations

✓Free from localized excitation coming from impurities
(such as free spins in χ, Schottky anomaly in C)
⇒*Reliable down to mK temperatures*

$$\kappa_s = C_s \upsilon_s \ell_s$$

C_s specific heat
υ_s velocity of excitation
ℓ_s mean free path of excitation

$$\kappa = \kappa_{spin} + \kappa_{phonon}$$

$$\kappa_{phonon} \sim T^3$$

Gapped or gapless?

Gapless spin liquid

$\kappa_{spin} \sim \gamma T$

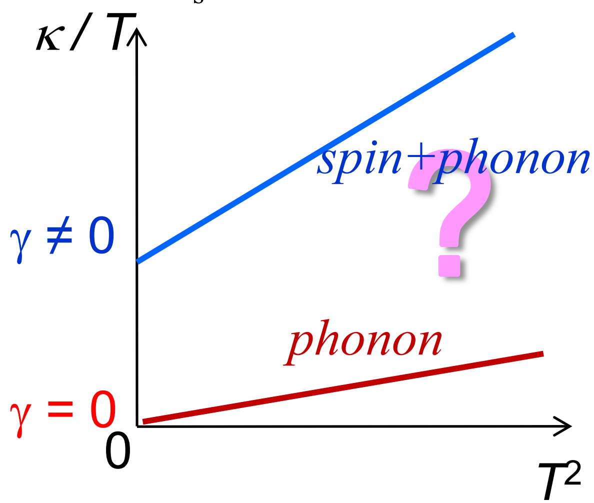
$\kappa_{spin}/T \sim \gamma \neq 0$

Gapped spin liquid

$\gamma = 0$

$\kappa_{spin} \sim \exp(-\Delta/T)$

Residual thermal conductivity
κ_s/T at $T \to 0$ K

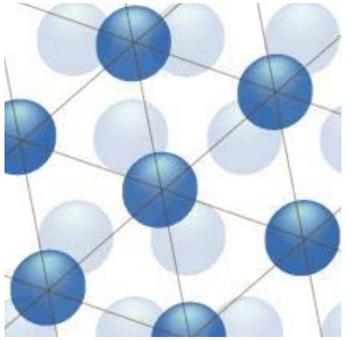


Finite κ/T at $T \to 0$ K immediately indicates the presence of gapless excitation

Magnetic or nonmagnetic? Field dependence of κ_s

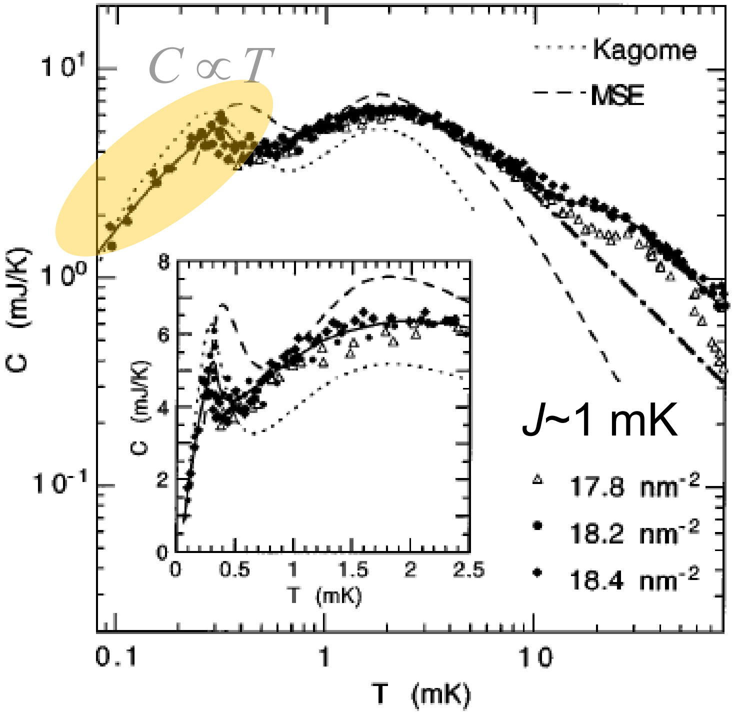
Localized or itinerant? Magnitude of the mean free path l_s

Spin liquid state in 2D ^{3}He on graphite



"4/7 phase"
Triangular lattice formed by ^{3}He atom

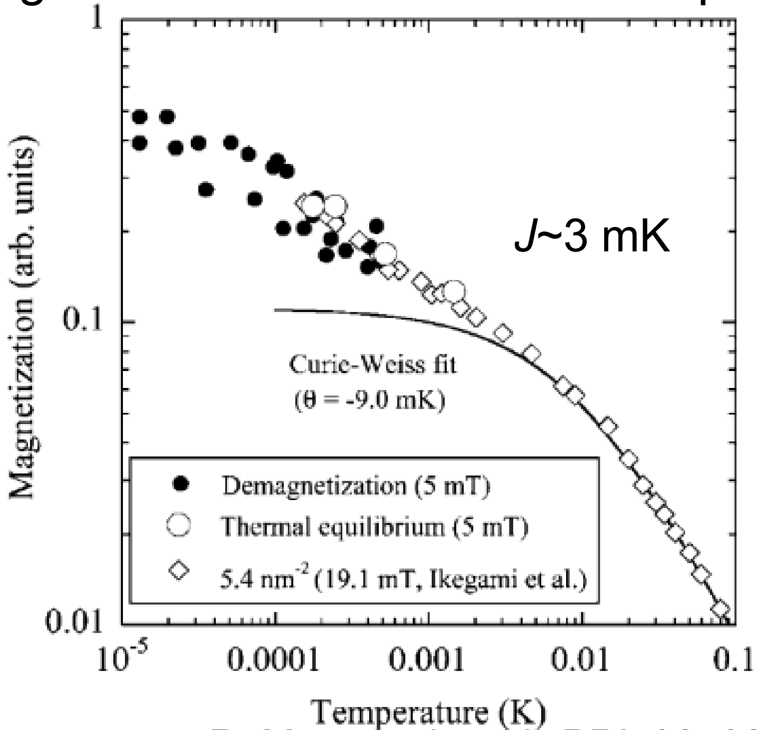
Specific heat of ^{3}He/Graphite



K. Ishida *et al*. PRL 79, 3451 (97)

Gapless down to *T*~*J*/10

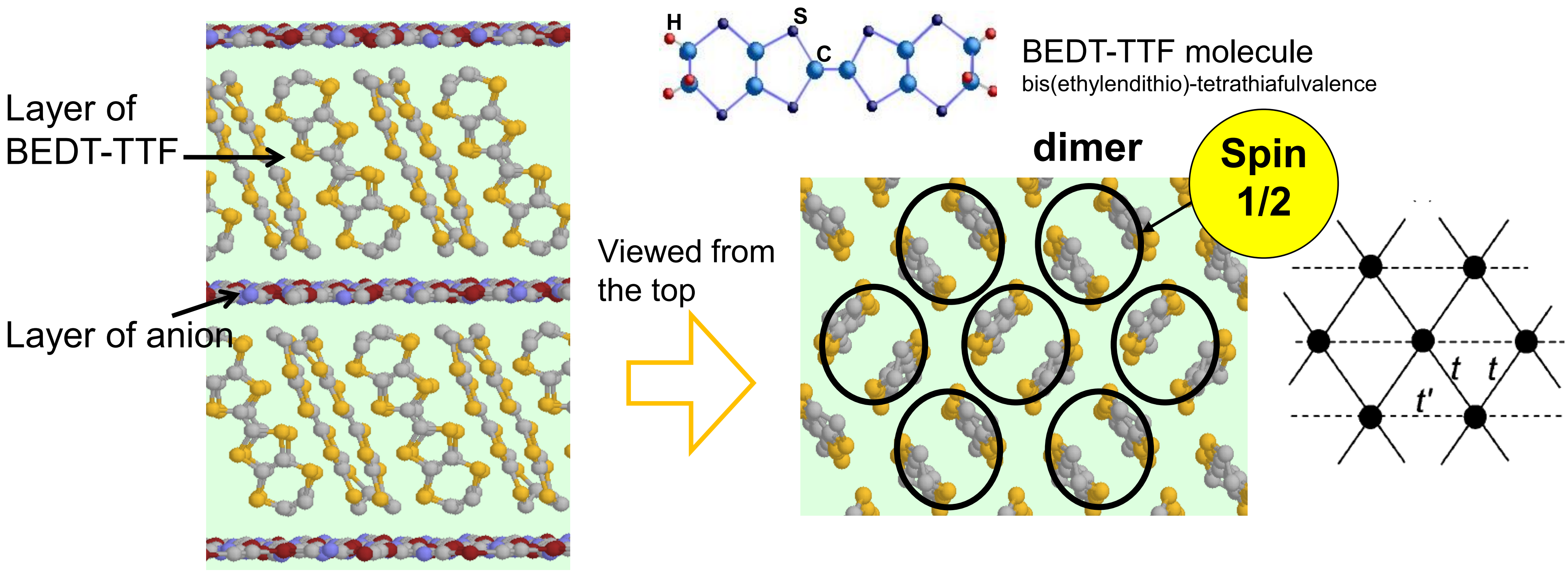
Magnetization of ^{3}He/HD/HD/Graphite



R. Masutomi *et al*., PRL 92, 025301 (04)

No spin gap down to *T*~*J*/300

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$



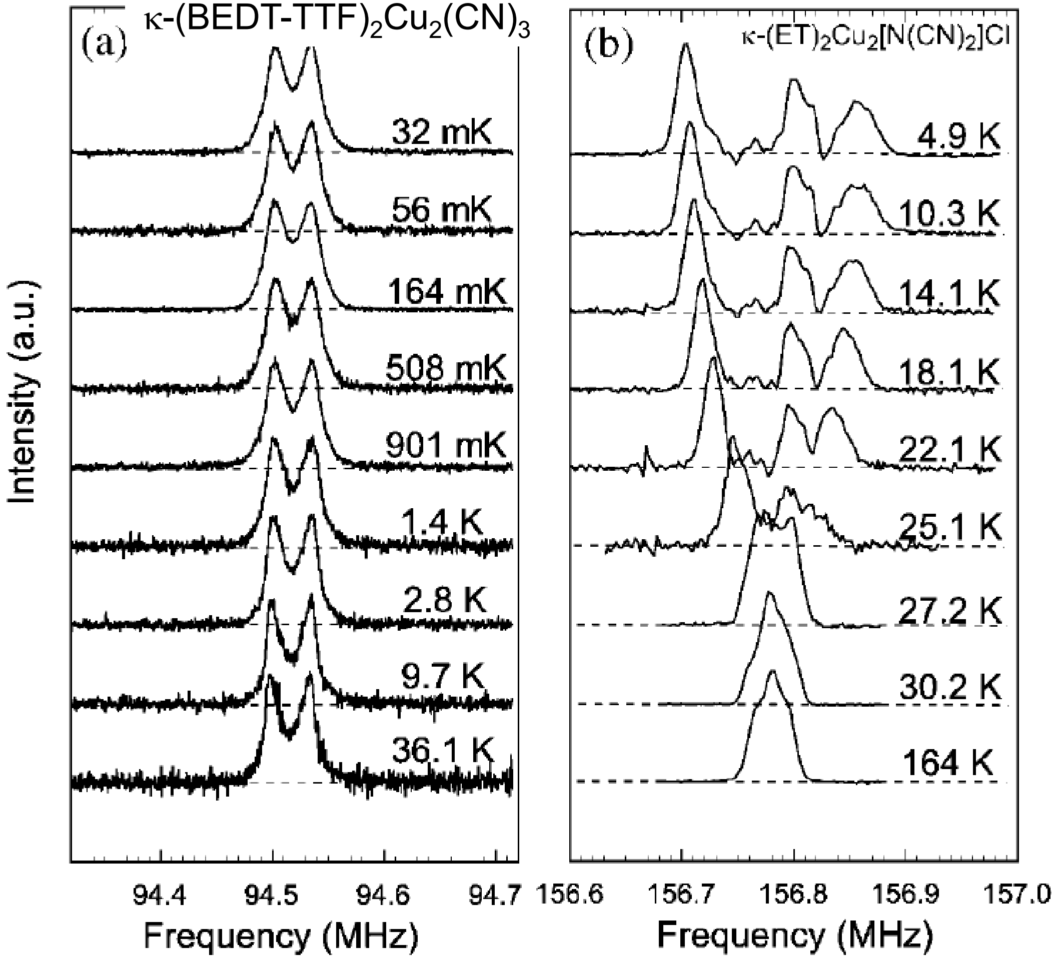
- Mott insulator : t~54.5 meV, t'~57.5 meV and U~448 meV (U/t~8.2)
- Pairs of BEDT-TTF molecule with spin ½ form dimers arranged in a 2D triangular lattice in terms of transfer integrals t and t' between the dimers. (t'/t = 1.06. by Hückel Method)
- Superconductivity under pressure

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

^{1}H NMR

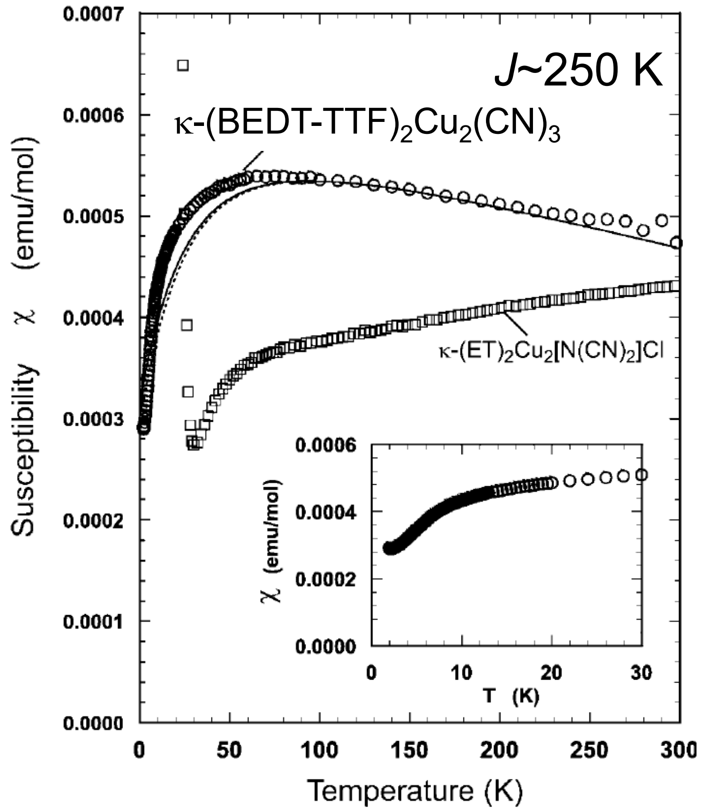
No internal magnetic field

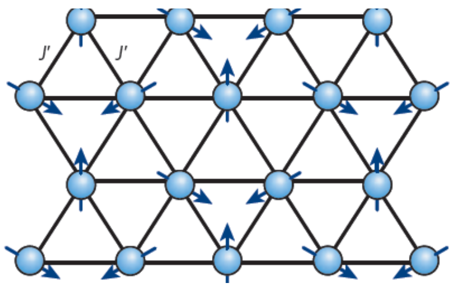
Magnetic order



Y. Shimizu *et al*. PRL 91, 107001 (03)

Magnetic susceptibility

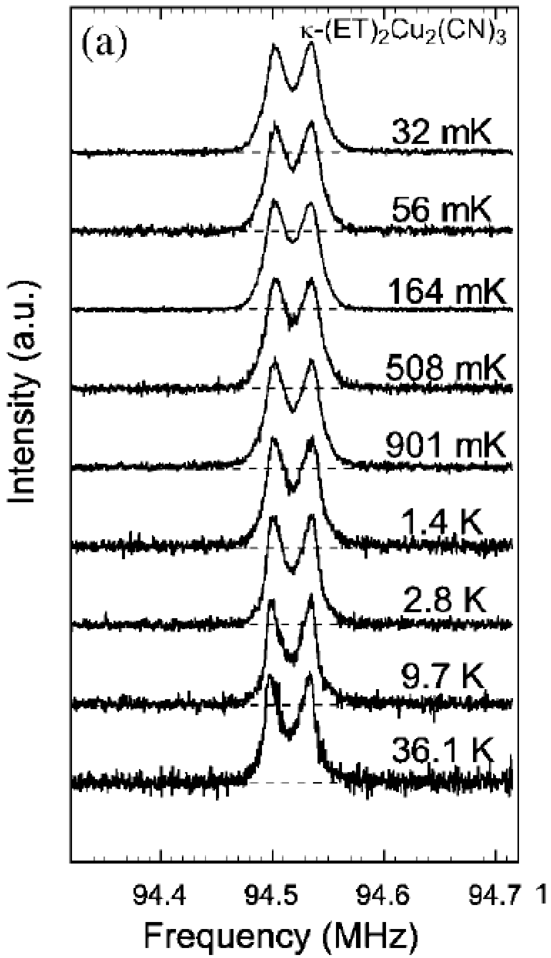




κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

^{1}H NMR

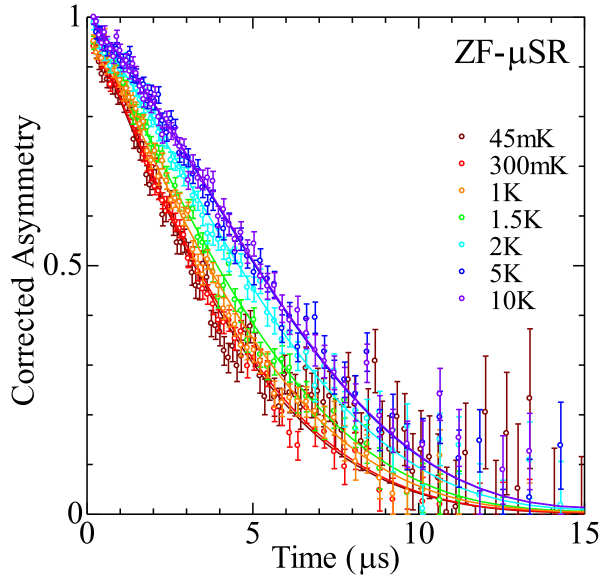
No internal magnetic field



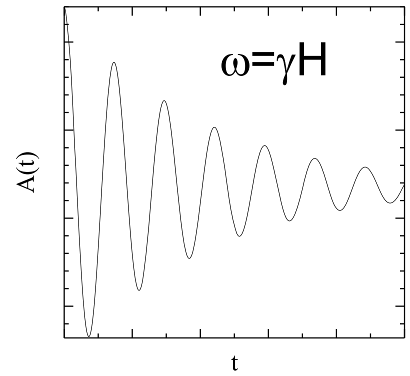
Y. Shimizu *et al*. PRL 91, 107001 (03)

μSR

No muon spin rotation



If magnetic order occurs,



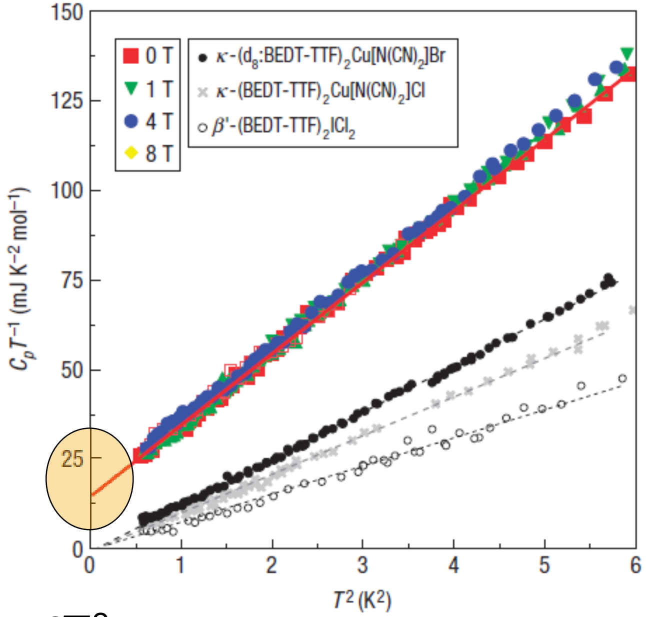
T. Goto *et al*. a preprint (10)

See also S. Oohira, *et al*., JLTP **142** (2007)

No magnetic order down to ~20 mK (~*J*/12,000)

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$: elementary excitation

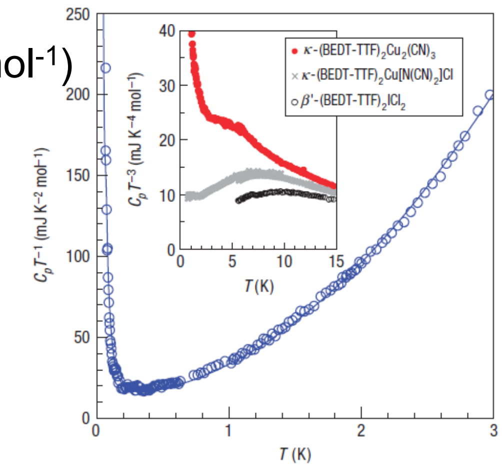
Specific heat



Gapless

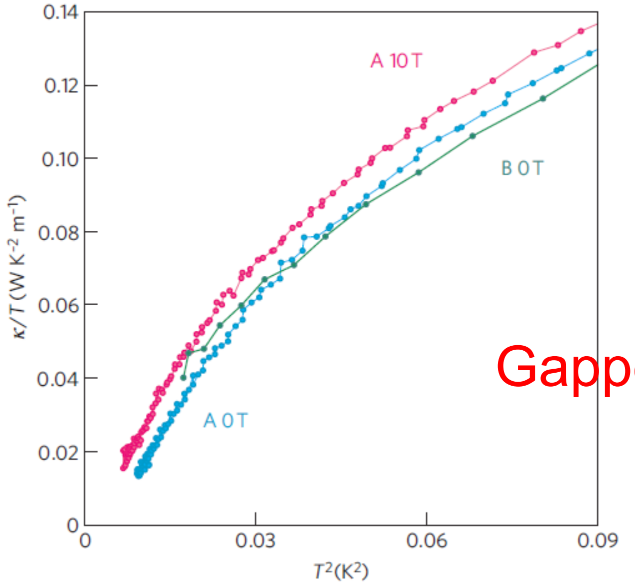
$C_P T^{-1} = \gamma + \beta T^2$

$(\gamma \approx 13\ \mathrm{mJK^{-2}mol^{-1}})$

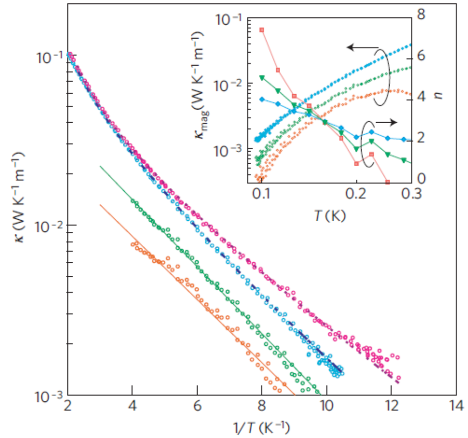


S. Yamashita *et al*. Nature Phys.4, 459 (08)

Thermal conductivity



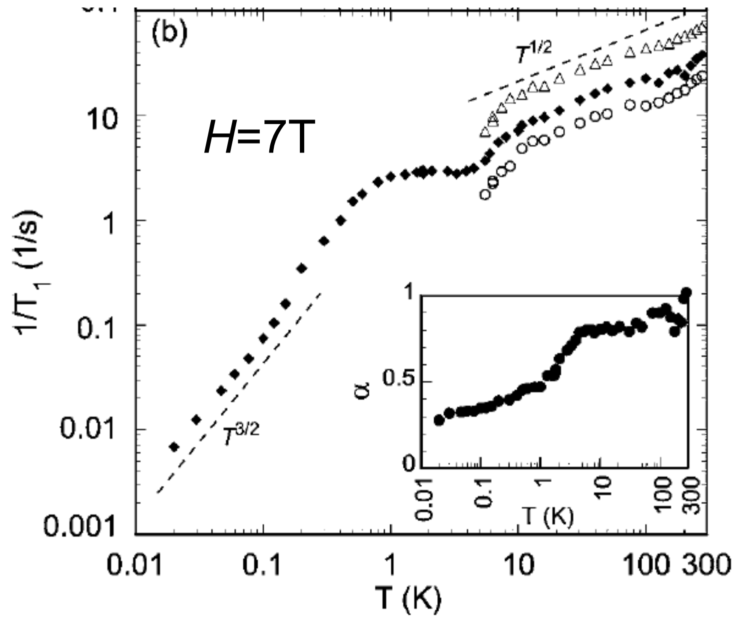
Gapped Δ~0.5K



M.Yamashita *et al*. Nature Phys.5, 44 (09)

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$: inhomogeneity

^{13}C NMR



$$-M(t)/M(\infty) = \exp[-(t/T_1)^{\alpha}]$$

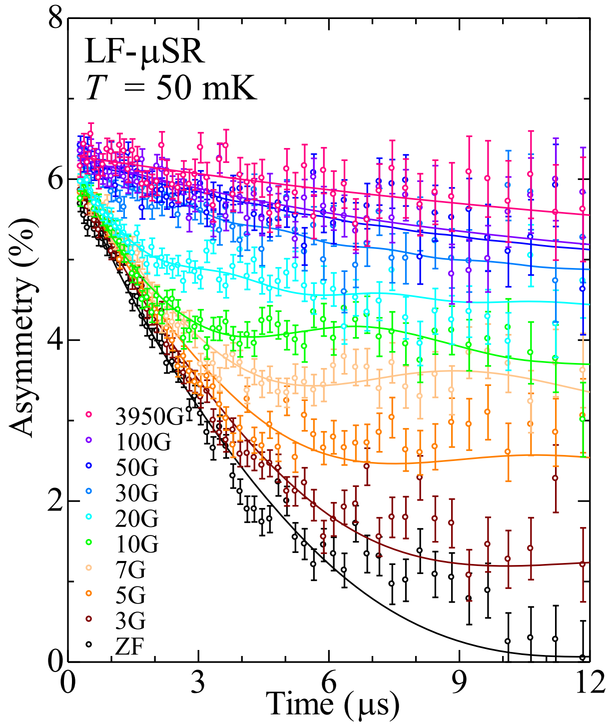
Y. Shimizu *et al*. PRB 73, 140407 (06)

NMR recovery curve shows stretched exponential

α<0.5 below 1K

Distribution of T_1

See also
A. Kawamoto *et al*. PRB 70, 060510 (04)

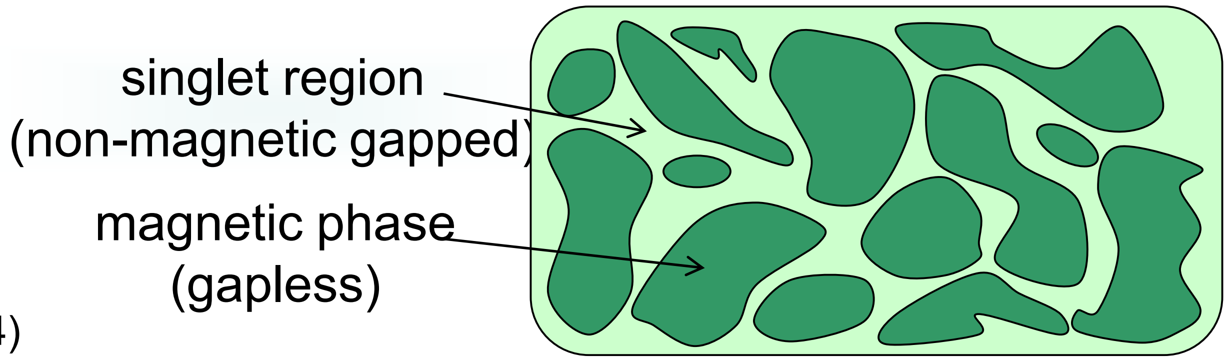


Relaxation curve below 300 mK

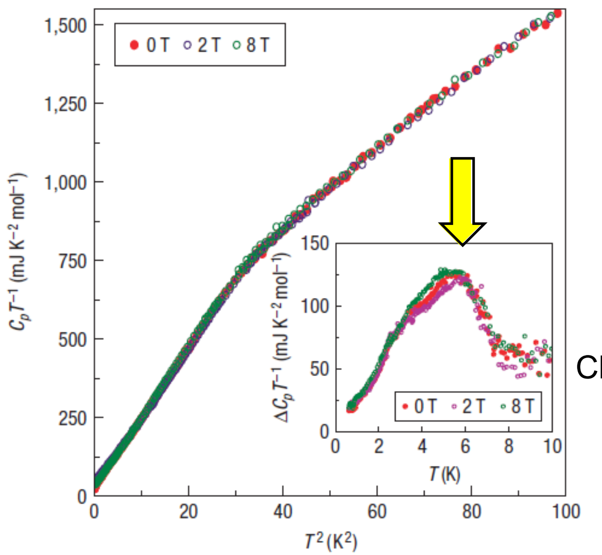
two components

T. Goto *et al.*

Microscopic phase separation between gapped and gapless regions

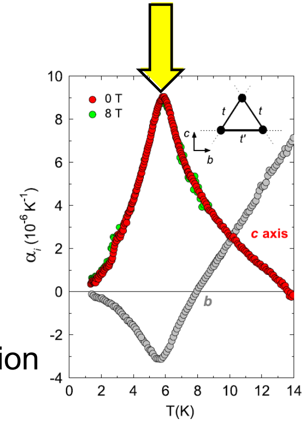


κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$: phase transition at 6 K

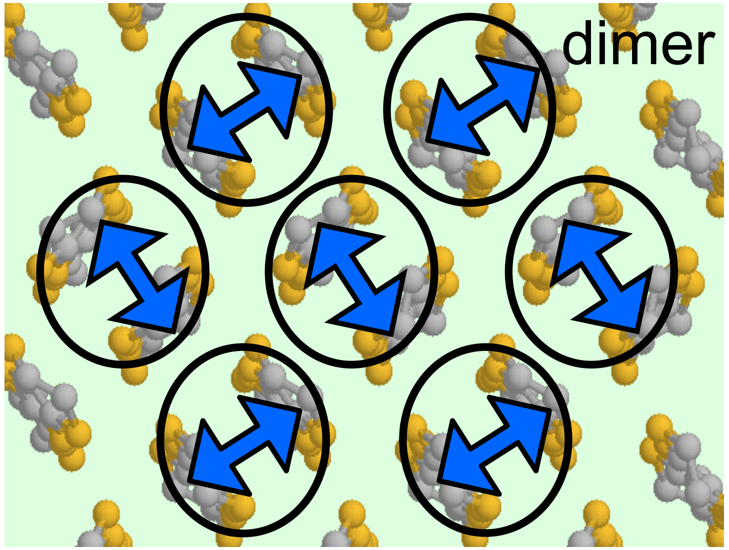


S. Yamashita *et al*. Nature Phys. (08)

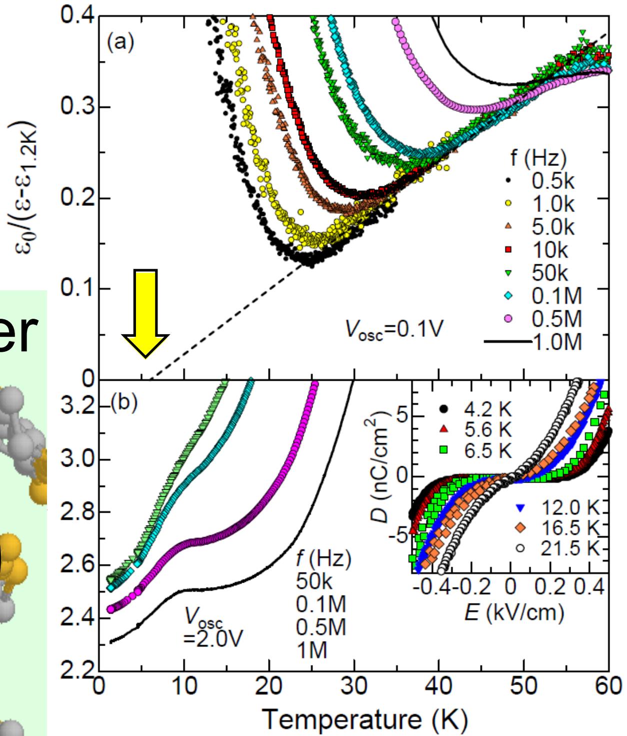
Thermal expansion



Charge disproportionation within the dimer?

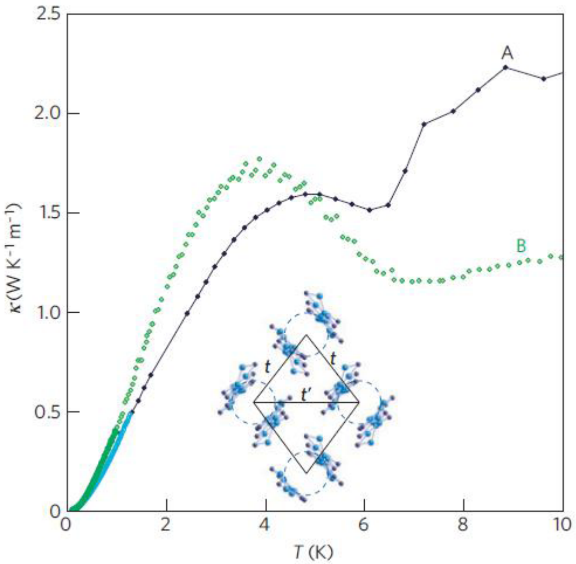


Frequency dependent dielectric constant



M. Abdel-Jawad *et al*. arXiv:1003.3902

Thermal conductivity



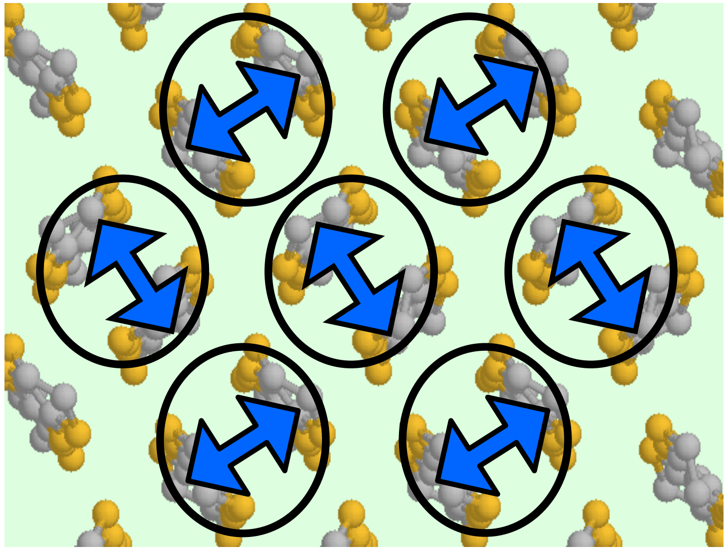
M.Yamashita *et al*. Nature Phys.(09)

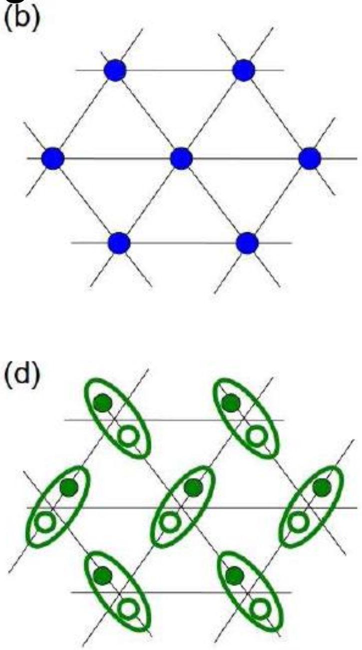
Structural transition associated with the charge degree of freedom?

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$: Anomalous dielectric response

Nontrivial charge degree of freedom within the dimer

dimer





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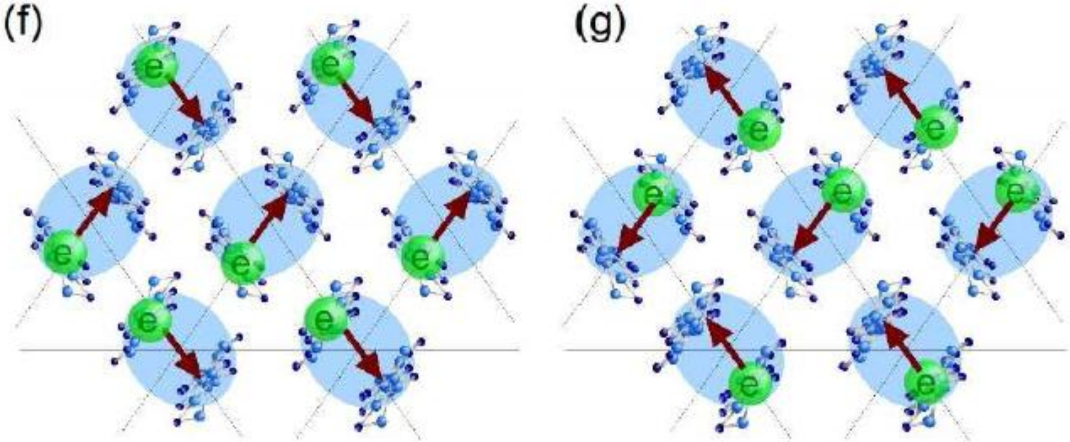


M. Abdel-Jawad *et al*. arXiv:1003.3902

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$: Summary

No magnetic order down to very low temperature $T\sim J/12{,}000$

NMR, μSR, susceptibility

Excitation

Gapless Specific heat

Gapped Thermal conductivity

However,

Strong inhomogeneity and microscopic phase separation

NMR, μSR

Phase transition at 6 K possibly associated with charge degree of freedom within the dimer

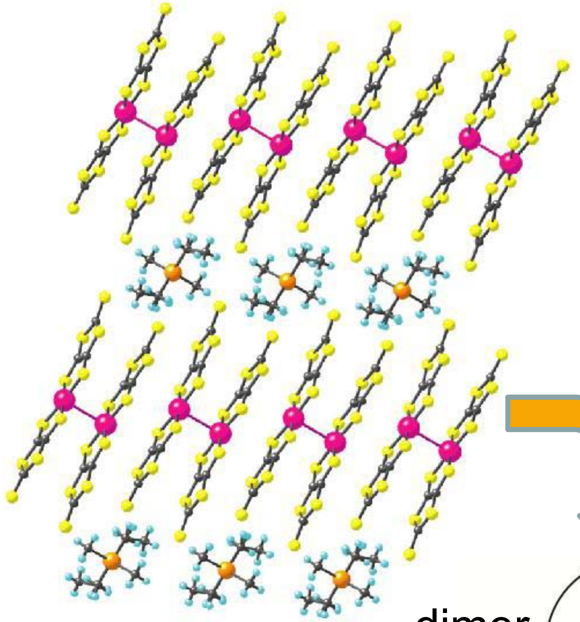
NMR, Heat capacity, Thermal conductivity, Thermal expansion, Dielectric constant

Genuine feature of a QSL appears to be masked by inhomogeneity.
To investigate the QSL, more homogeneous system is required.

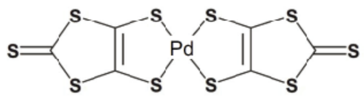
Quantum spin liquid state in β'- $EtMe_3Sb[Pd(dmit)_2]_2$

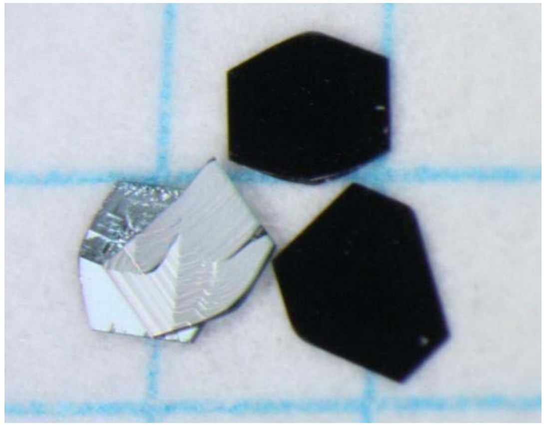
Two-dimensional Mott system with a quasi-triangular lattice

Clean system with small defects



2D layer of
$Pd(dmit)_2$ molecule

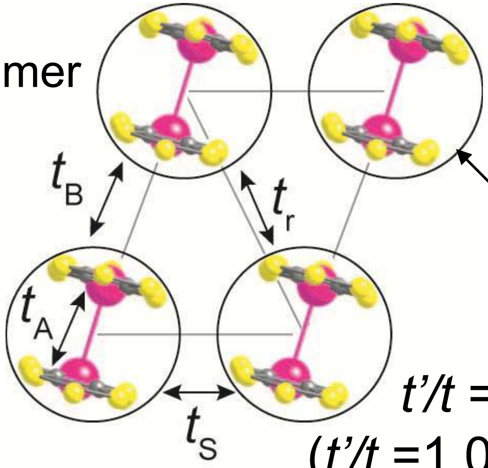




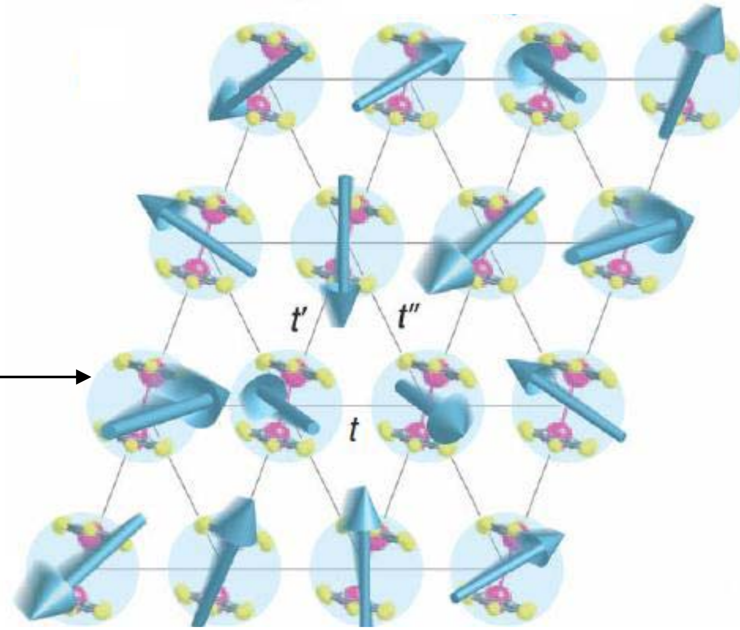
Non-magentic layer
X = $EtMe_3Sb$, Et_2Me_2Sb,,,

EMe_3Sb
$t_S:t_B:t_r$ = 1:1.02:0.93

dimer



Spin 1/2

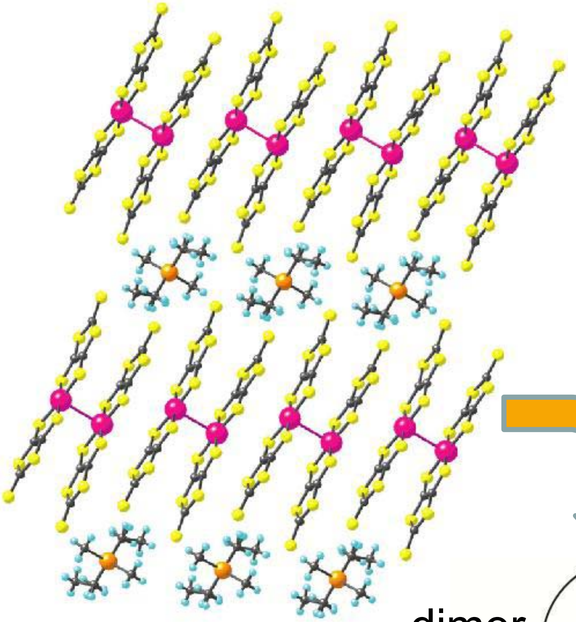


t'/t =0.93
(t'/t =1.06 for ET)

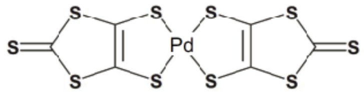
Quantum spin liquid state in β'- $EtMe_3Sb[Pd(d\underline{mit})_2]_2$

Two-dimensional Mott system with a quasi-triangular lattice

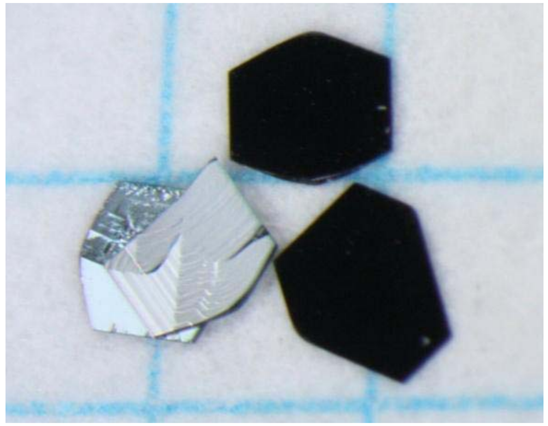
Clean system with small defects



2D layer of $Pd(dmit)_2$ molecule

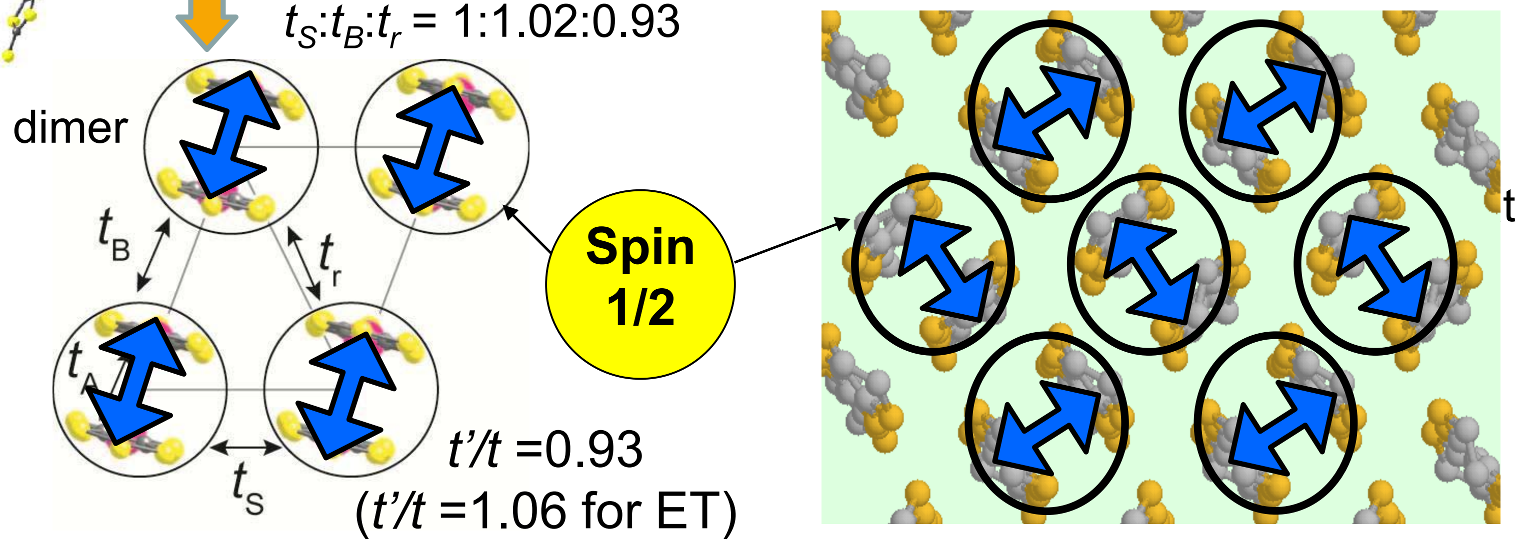


Non-magentic layer
X = $EtMe_3Sb$, Et_2Me_2Sb,,,

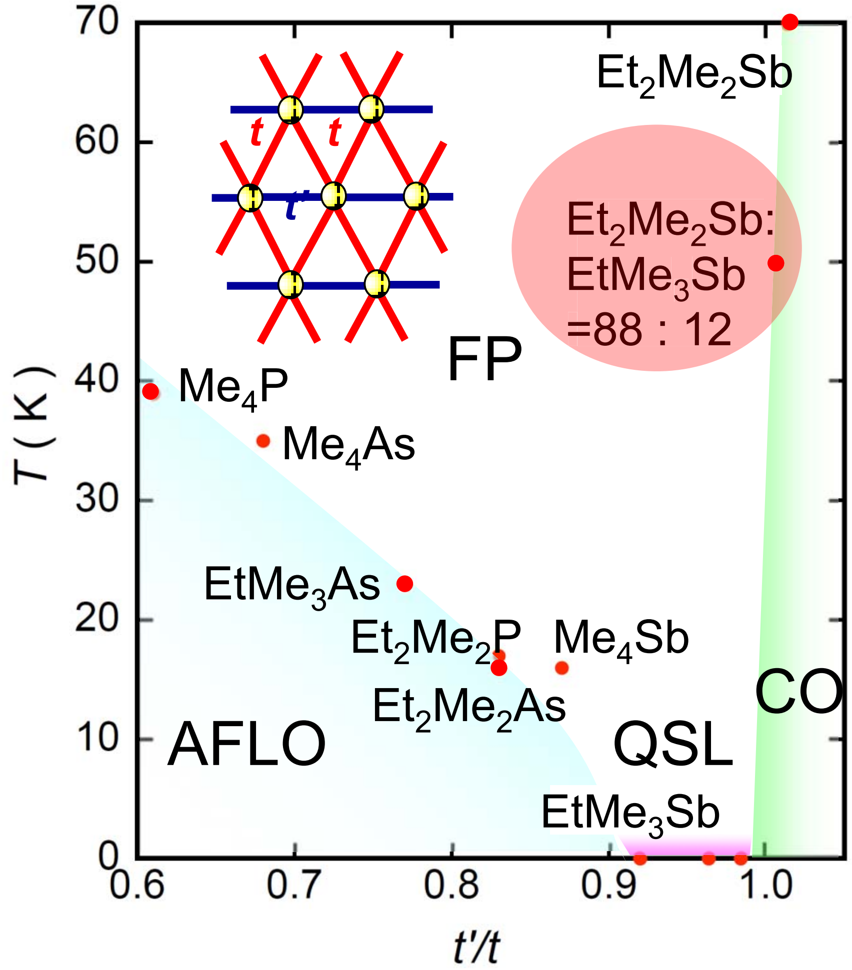


EMe_3Sb
$t_S:t_B:t_r$ = 1:1.02:0.93

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$



β'-(Cation)[Pd(dmit)$_2$]$_2$

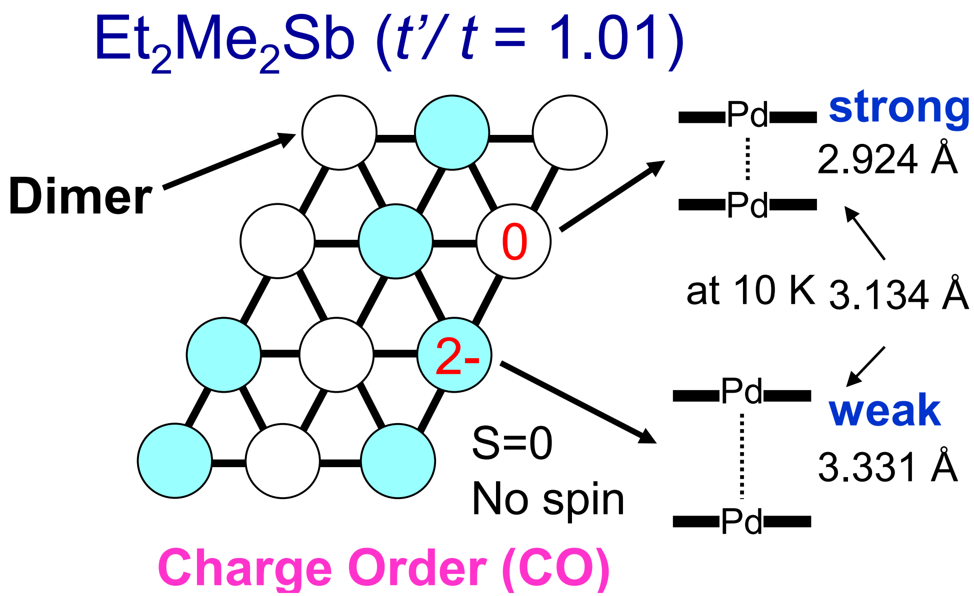


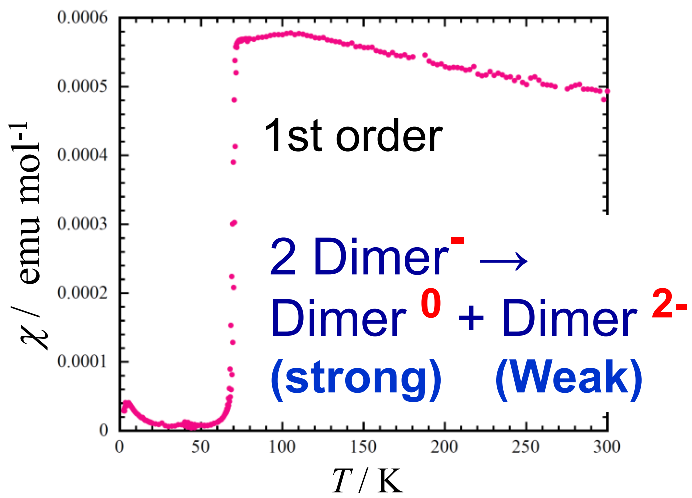
FP: Frustrated paramagnetic state

AFLO: Antiferromagnetic ordered state

CO: Charge ordered state

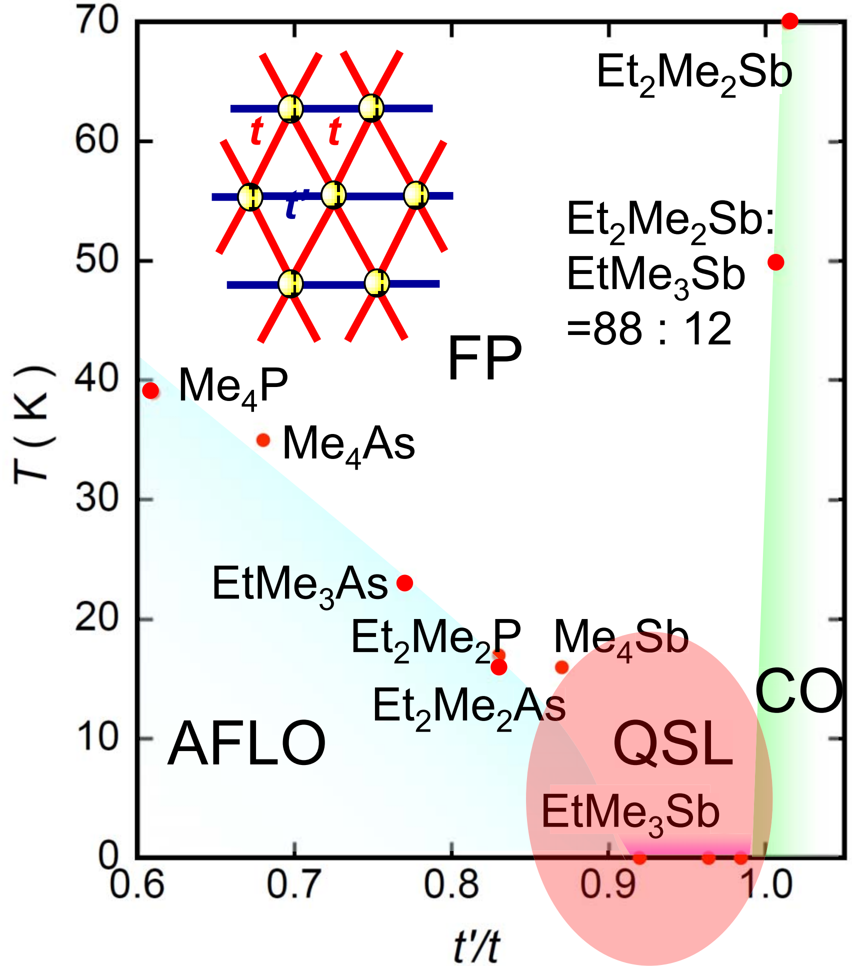
QSL: Quantum spin liquid state

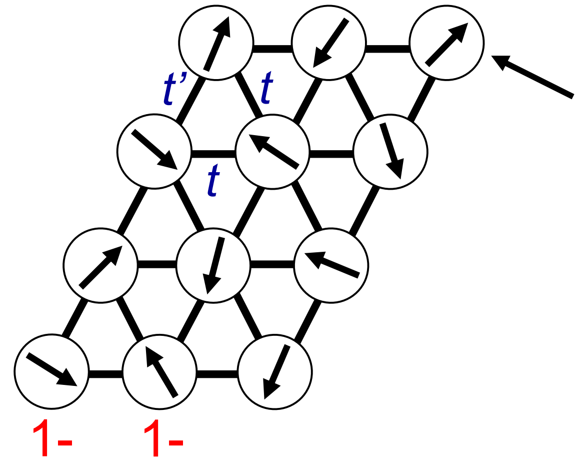




CPL, **411** (2005) 133 / *JPSJ*, **74** (2005) 2754

β'-(Cation)[Pd(dmit)$_2$]$_2$





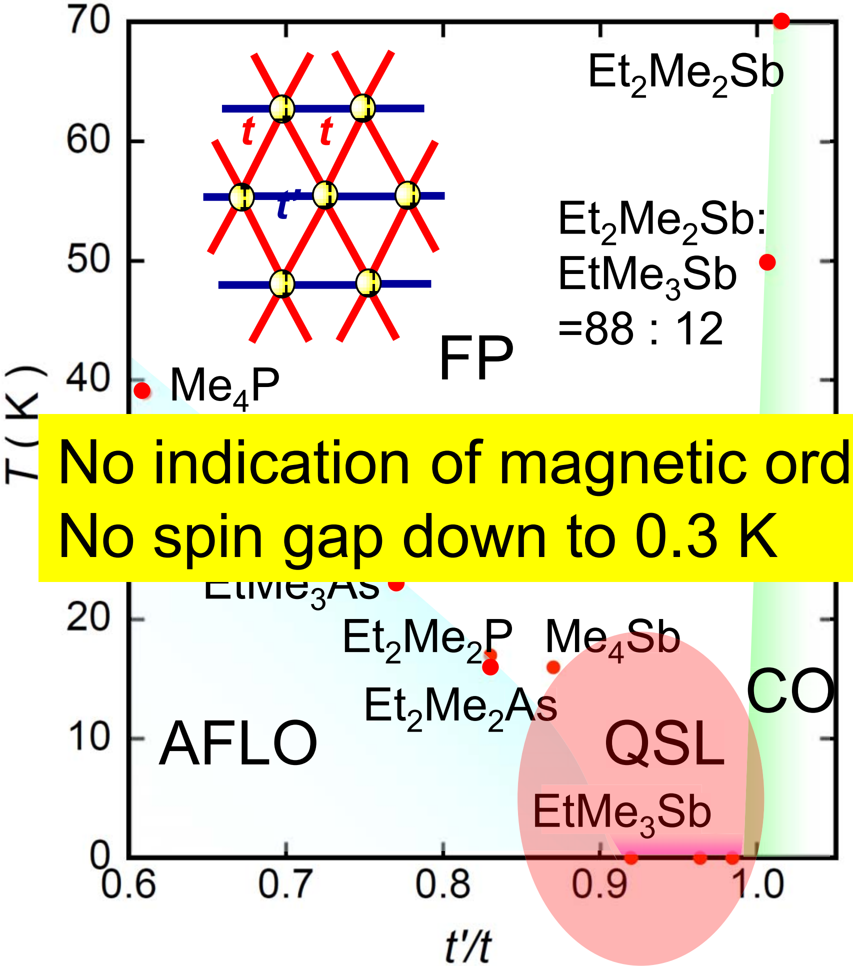
FP: Frustrated paramagnetic state

AFLO: Antiferromagnetic ordered state

CO: Charge ordered state

QSL: Quantum spin liquid state

$EtMe_3Sb[Pd(dmit)_2]_2$ Magnetic susceptibility



No indication of magnetic order
No spin gap down to 0.3 K

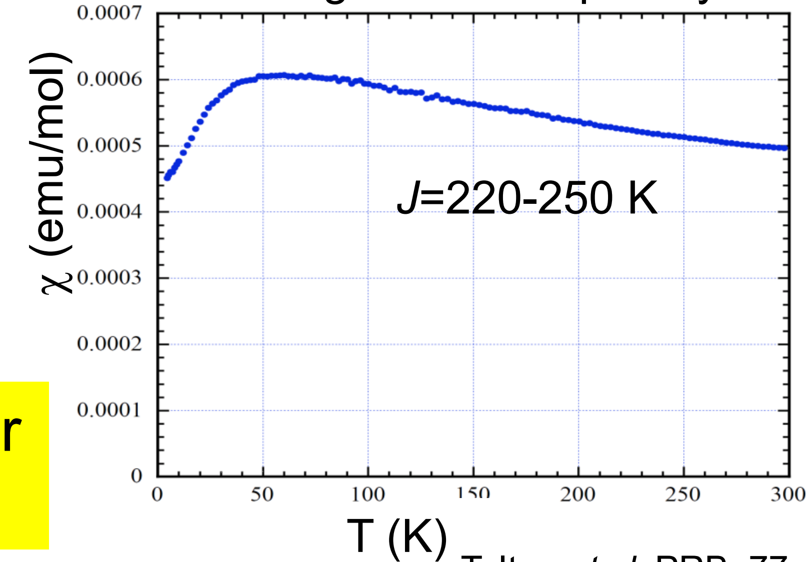
FP: Frustrated paramagnetic state

AFLO: Antiferromagnetic ordered state

CO: Charge ordered state

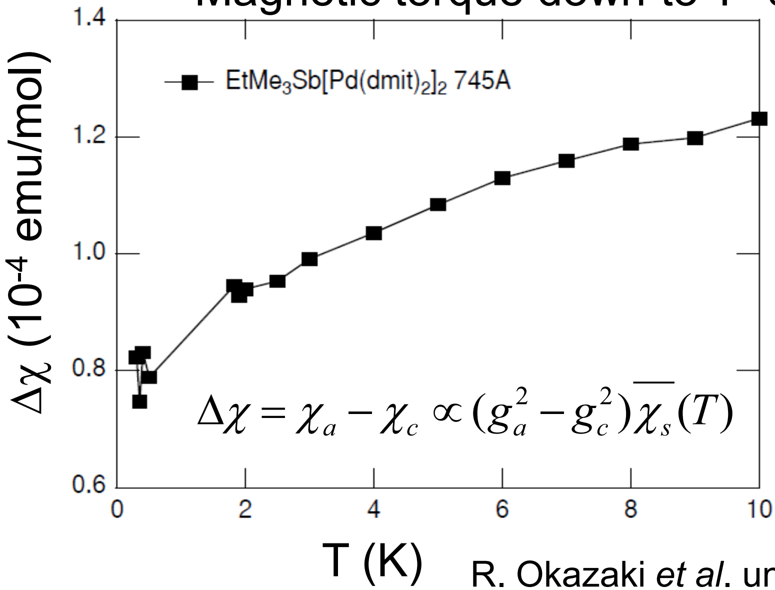
QSL: Quantum spin liquid state

Magnetic susceptibility down to 5 K



T. Itou *et al.* PRB, 77, 104413 (08)

Magnetic torque down to T=0.3 K



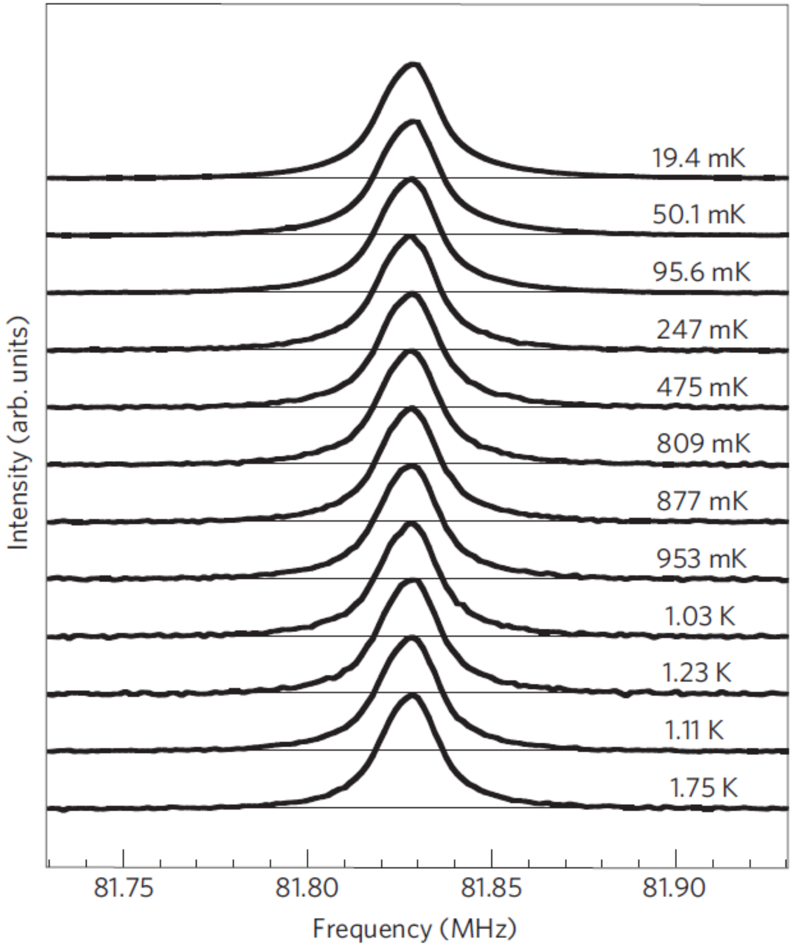
$$\Delta\chi = \chi_a - \chi_c \propto (g_a^2 - g_c^2)\overline{\chi_s}(T)$$

R. Okazaki *et al.* unpublished

$EtMe_3Sb[Pd(dmit)_2]_2$:NMR and μSR

^{13}C NMR *H*=7 T

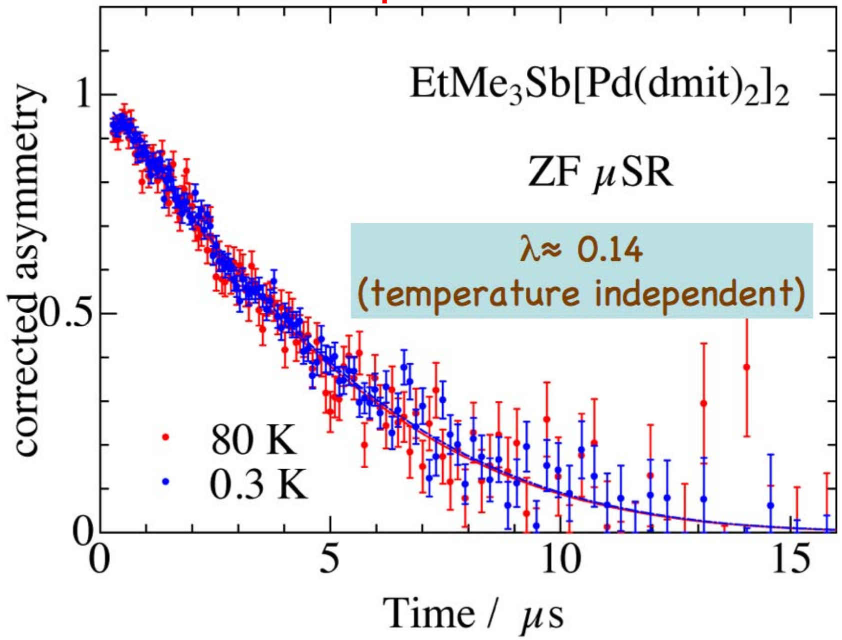
No internal magnetic field



Itou *et al*., Nature Phys. 11, 1 (10)

ZF μSR *H*=0 T

No muon spin rotation



Y. Ishii (RIKEN-RAL) et al.

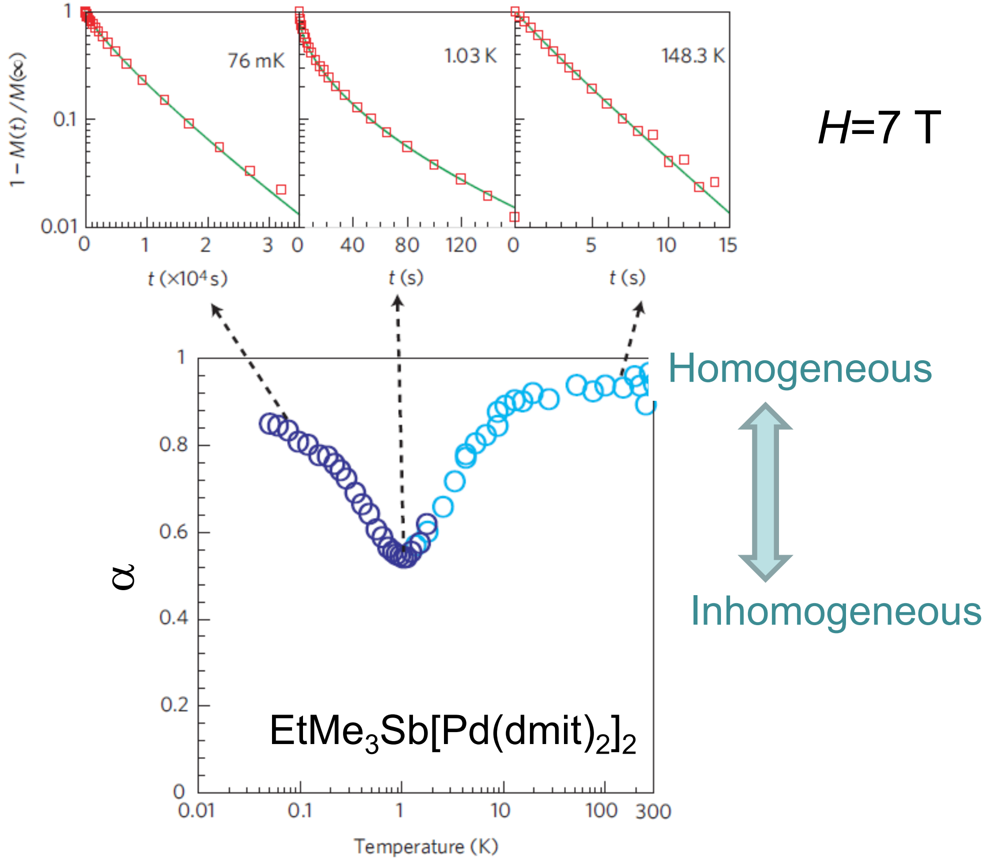
No indication of magnetic order down to ~*J*/12,000

$EtMe_3Sb[Pd(dmit)_2]_2$:NMR

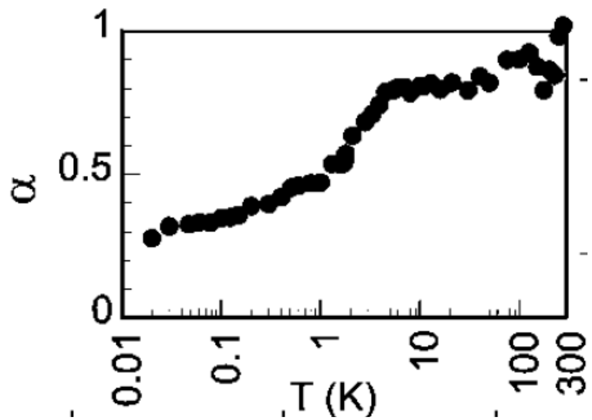
^{13}C NMR

$$-M(t)/M(\infty)=\exp[-(t/T_1)^{\alpha}]$$

H=7 T



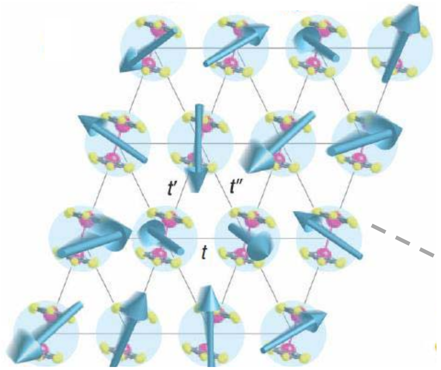
κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$



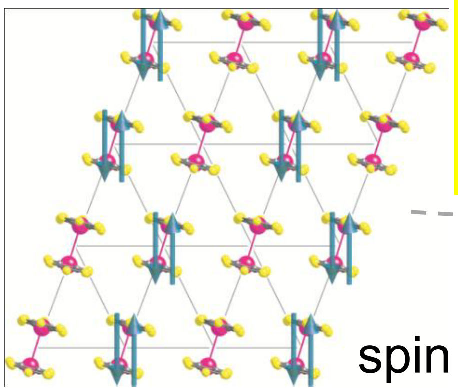
Itou *et al*., Nature Phys. 11, 1 (10)

At low temperatures, homogeneous spin liquid state is realized

$EtMe_3Sb[Pd(dmit)_2]_2$:Low energy excitation



Spin liquid

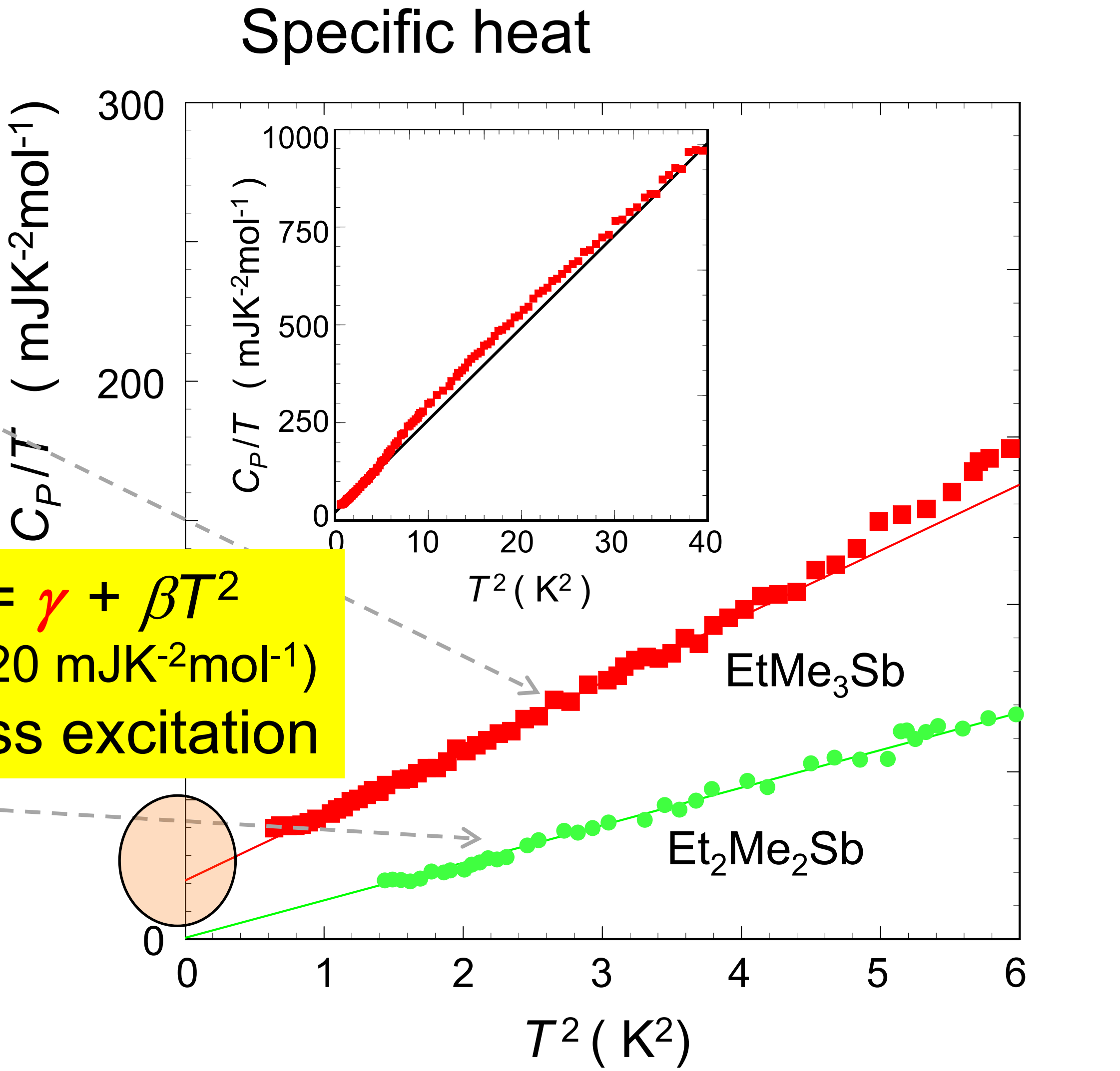


Charge order

$$C_P T^{-1} = \gamma + \beta T^2$$

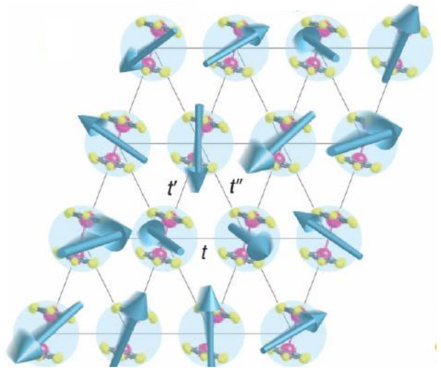
($\gamma \approx 20$ mJK^{-2}mol^{-1})

Gapless excitation

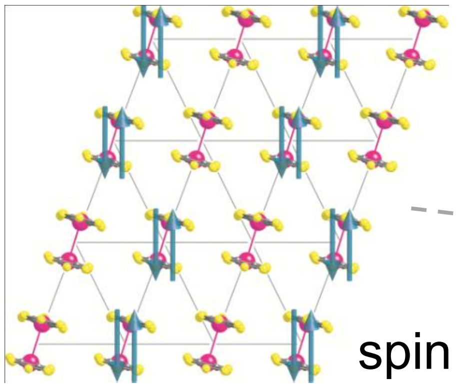


S. Yamashita & Y. Nakazawa (Osaka Univ.)

$EtMe_3Sb[Pd(dmit)_2]_2$:Low energy excitation

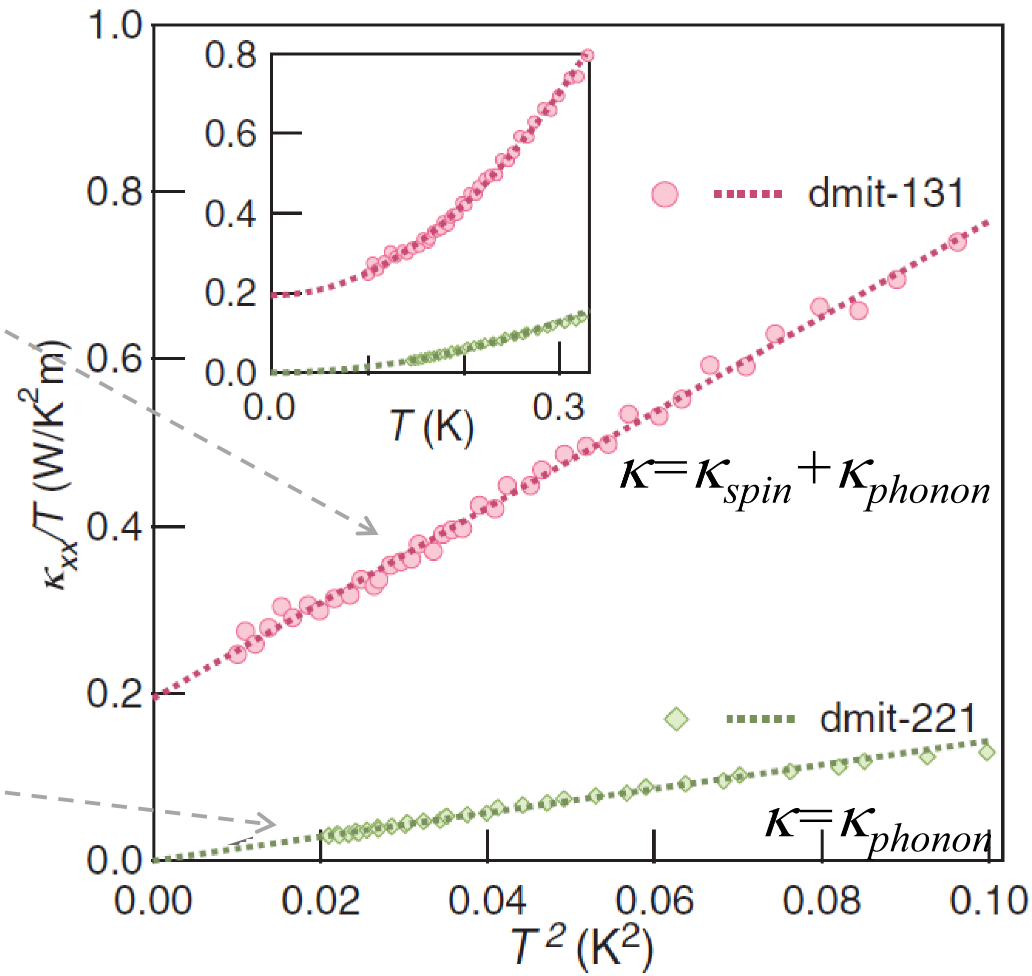


Spin liquid



Charge order

Thermal conductivity



M. Yamashita *et al*. Science 328, 1246 (10)

$EtMe_3Sb[Pd(dmit)_2]_2$:Low energy excitation

Enhancement of κ in spin liquid state
→ $\kappa = \kappa_{spin} + \kappa_{phonon}$

Clear residual of κ/T !
$\kappa/T\ (T \rightarrow 0)$ = 0.19 W/K²m
Evidence for a ***gapless excitation***,
like electrons in normal metals.

Estimation of mean free path

$$\kappa = C \cdot \upsilon_s \cdot \ell$$

$$C/T \sim 20\mathrm{mJ/K^2mol}$$

$$\upsilon_s = Ja\pi / 2\hbar$$

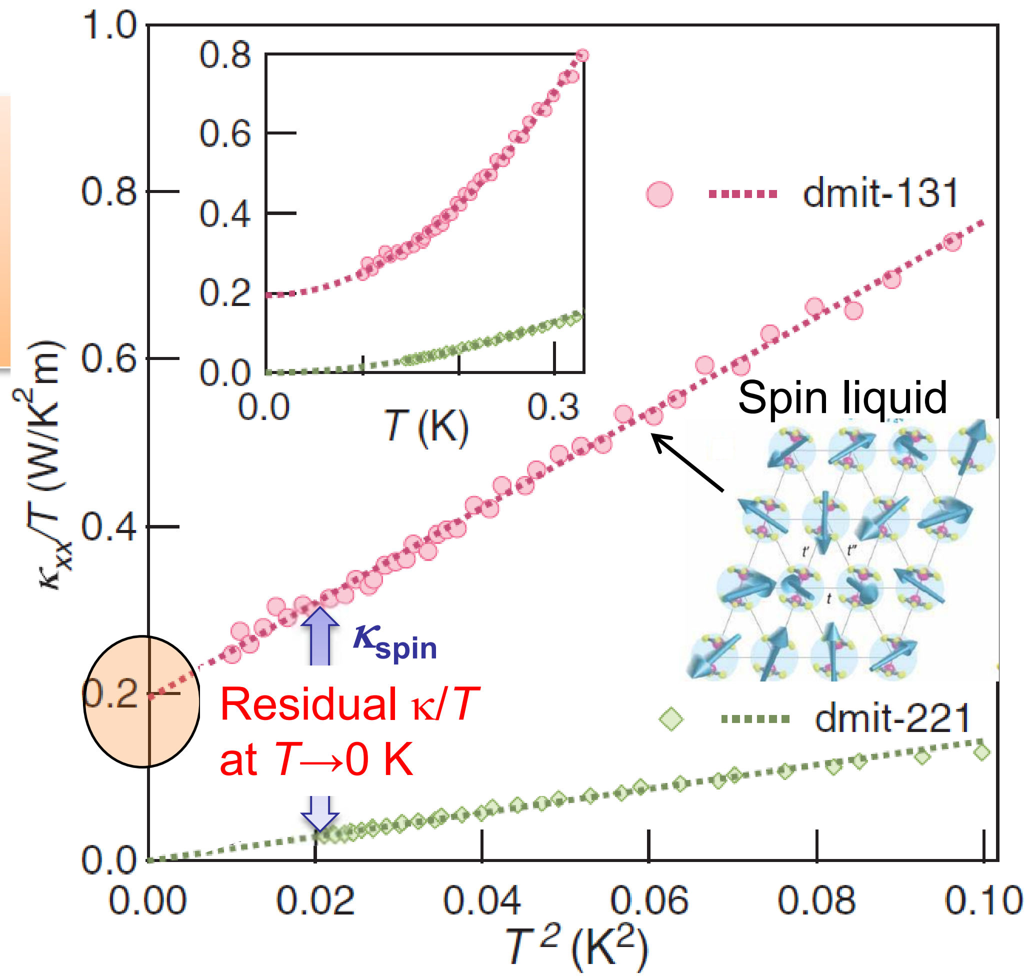
$$\Rightarrow \quad \ell = 1.2\ \mu\mathrm{m} >> a \sim 1\,\mathrm{nm}$$

More than 1000 times longer than the interspin distance!!

Itinerant excitation

Homogeneous
Extremely long correlation length

Thermal conductivity



M. Yamashita *et al.* Science 328, 1246 (10)

Thermal conductivity of 1D Heisenberg chain

In an integrable system such as the Heisenberg spin-1/2 chain, thermal conductivity along the chain is infinite.

X. Zotos *et al*. PRB 55, 11029 (97)
K.Saito, S.Takesue and S. Miyashita, PRE 54, 2404 (96)

Thermal conductivity in a real material is finite due to impurities, phonons or an interchain interaction

1D Heisenberg S=1/2 Sr_2CuO_3

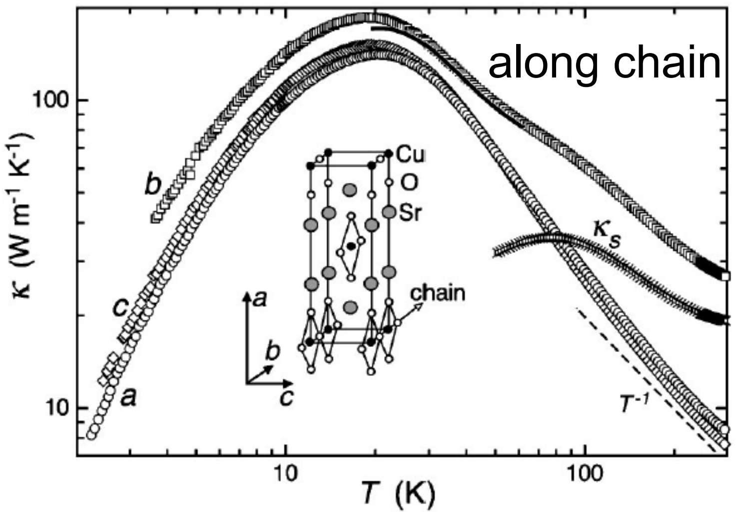
J~2500 K

$$\kappa_s = C_s \upsilon_s \ell_s$$

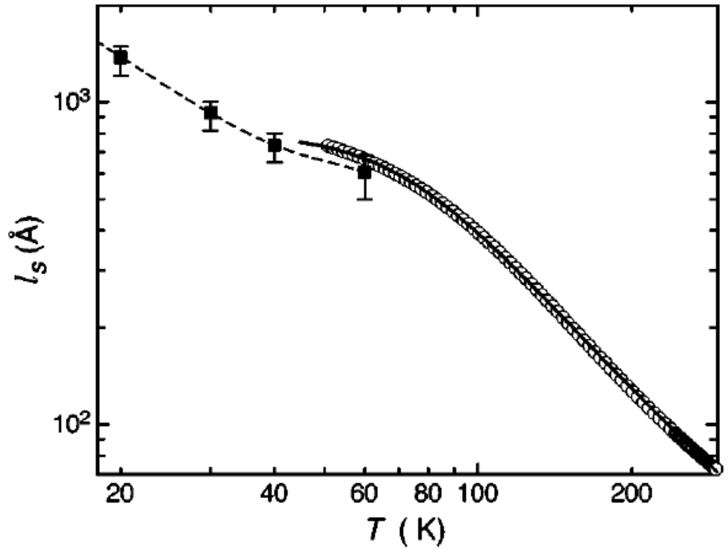
C_s specific heat

$\upsilon_s = Ja\pi / 2\hbar$ velocity of spinon

ℓ_s spinon mean free path



Unusually high quasi-1D thermal conductivity



Very long spinon mean free path

A.V. Sologubenko *et al*. PRB 62, R6108 (00)

$EtMe_3Sb[Pd(dmit)_2]_2$:Low energy excitation

Quantum spin liquid conducts heat very well.

as good as brass

1-yen

Alminium

5-yen (~5 cent)

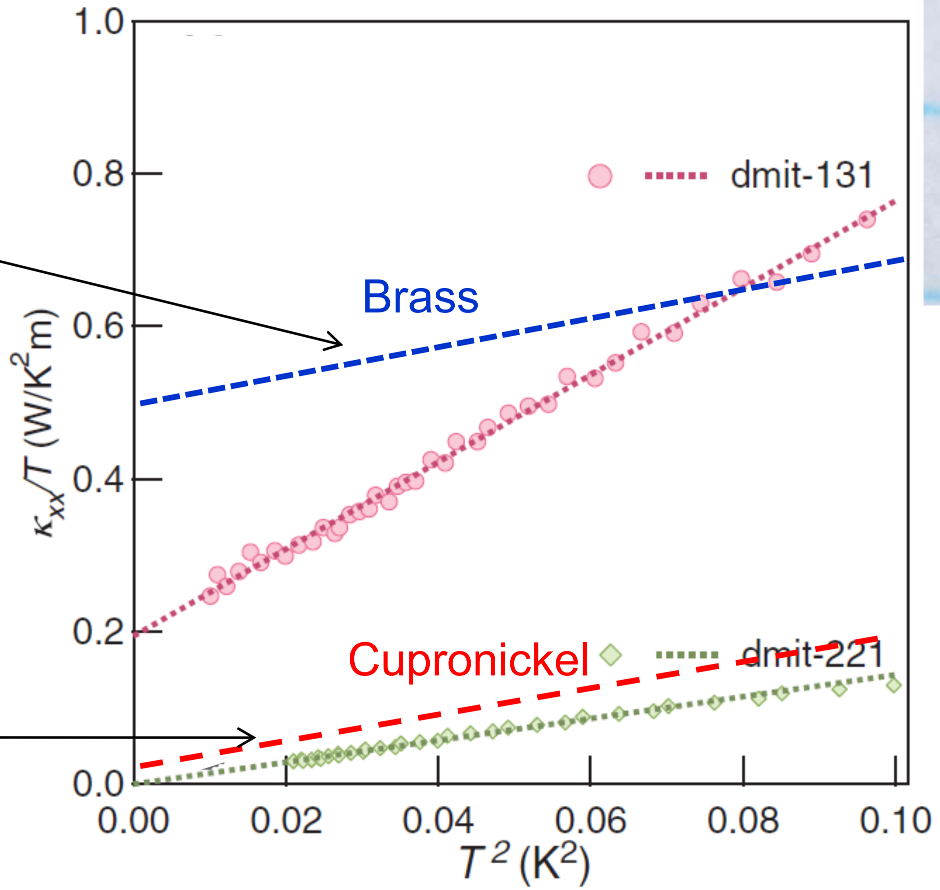
Brass (Cu0.7+Zn0.3)

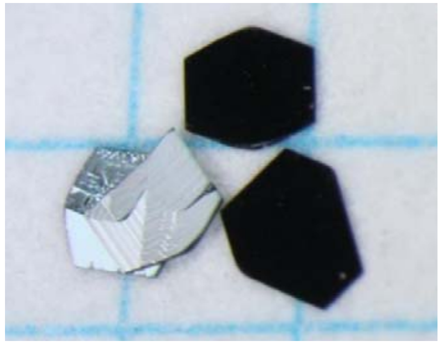
10-yen

Copper

100-yen(~$1~€1)

Cupronickel (Cu0.75+Ni0.25)

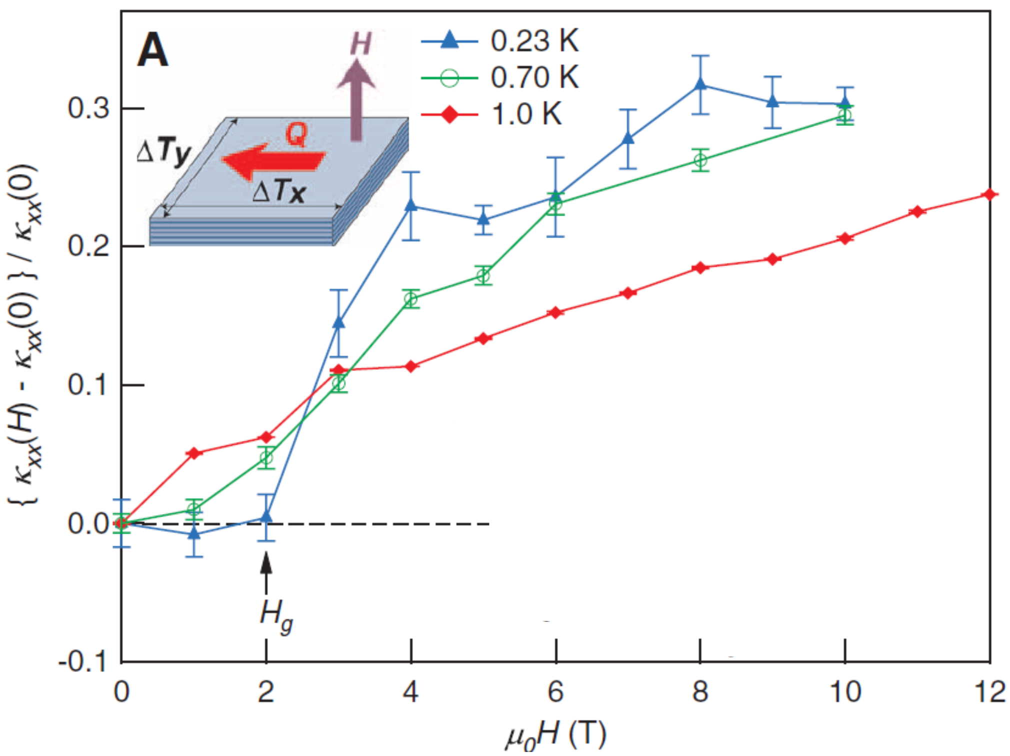




$EtMe_3Sb[Pd(dmit)_2]_2$:Nature of the low energy excitation

Field dependence of the elementary excitations

H-dependence of κ_{xx}



M. Yamashita *et al*. Science 328, 1246 (10)

- H > 1 K

Linear increase

Clear signature of κ_{spin}

- H < 1 K

Spin-gap like behavior

$\mu_0 H_g \sim 2$ T

Bipartite nature of excitations

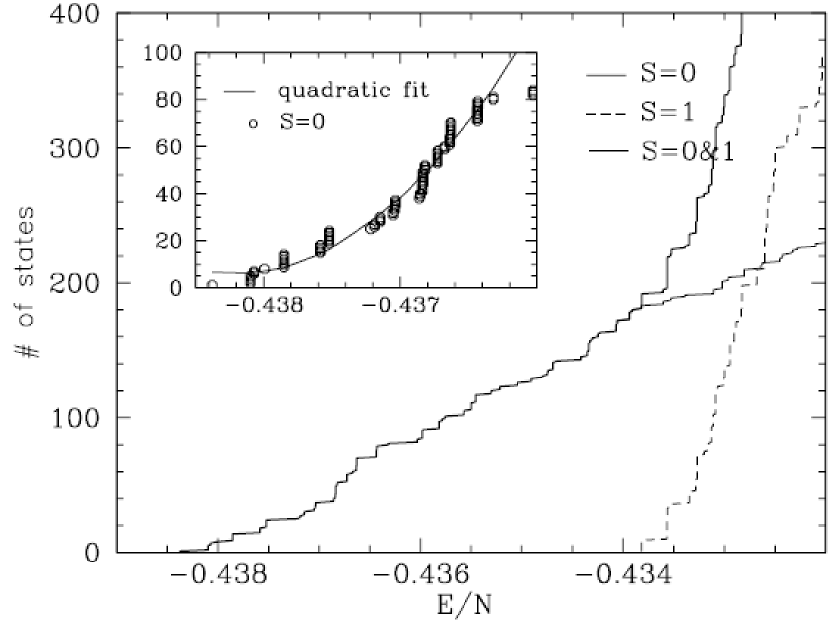
Coexistence of

Nonmagnetic gapless excitation

and

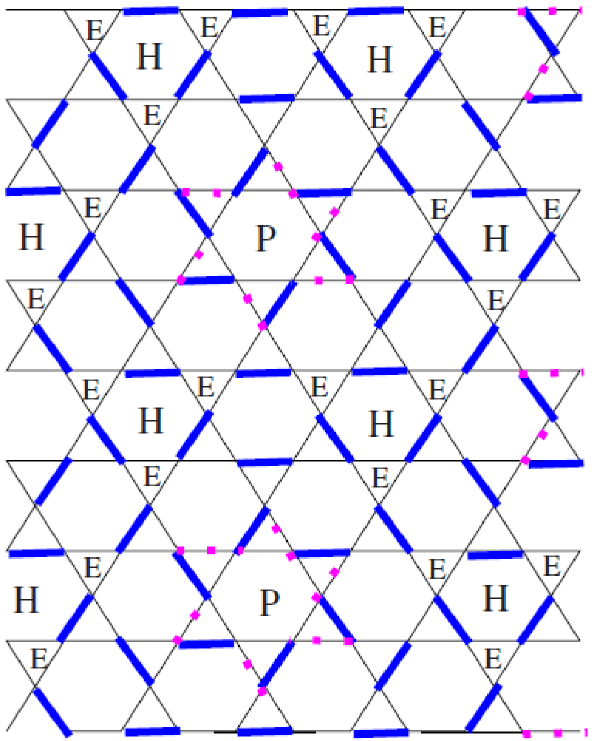
Spin-gap-like excitation that couples to magnetic field

Kagome lattice Heisenberg magnet with *S*=1/2



Gapped magnetic excitation filled with nonmagnetic excitations

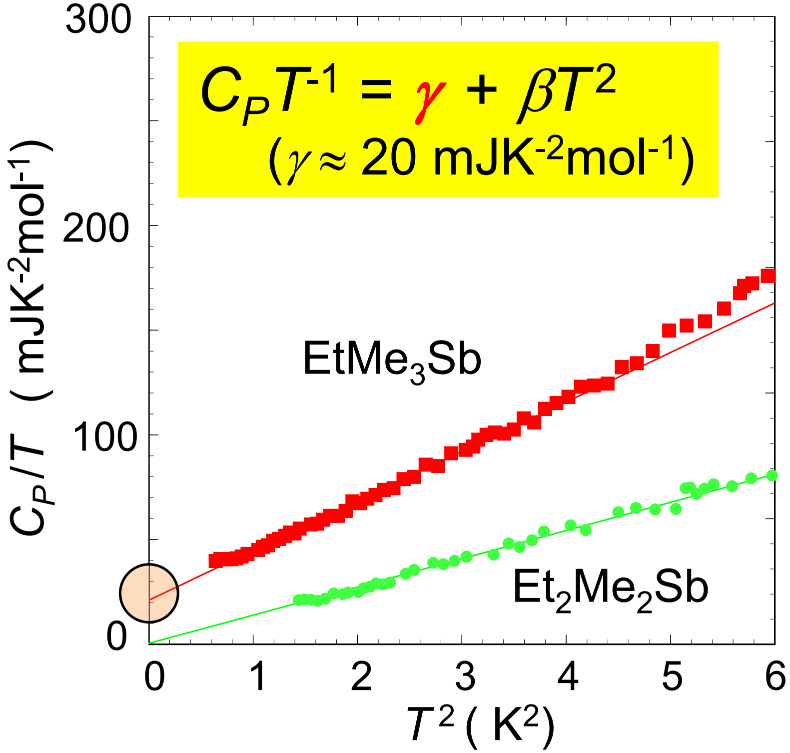
Ch. Waldtmann *et al*. Eur. Phys. J. B 2, 501 (98)



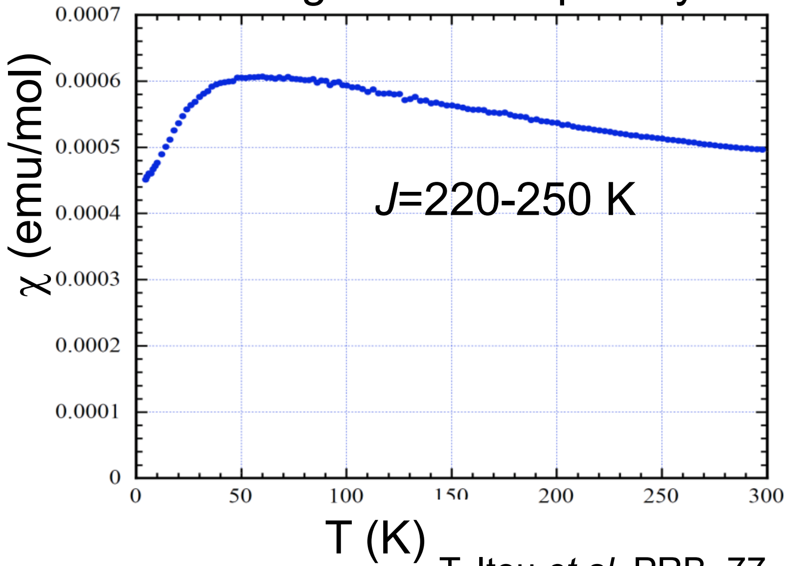
Valence bond solid with a 36 site unit cell

R.P. Singh and D.A. Huse, PRB 76, 180407 (07)

Wilson ratio



Magnetic susceptibility down to 5 K



T. Itou *et al.* PRB, 77, 104413 (08)

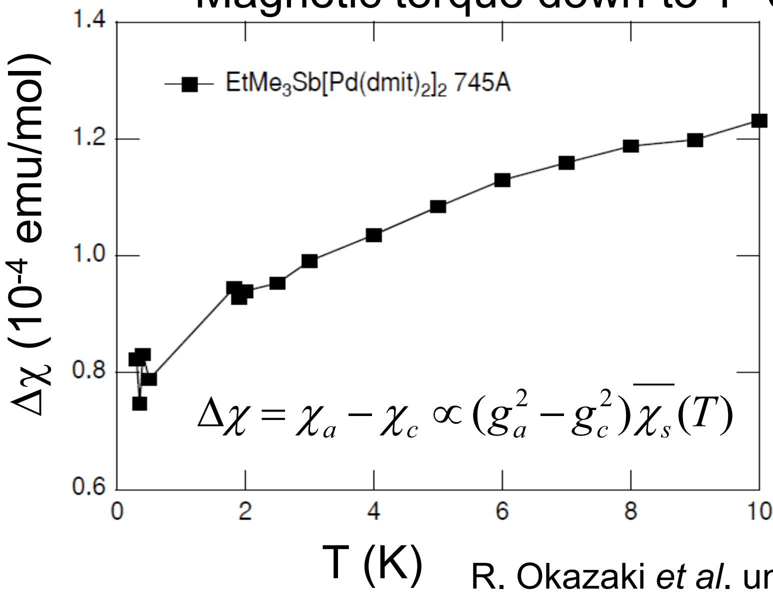
$$R = \frac{4\pi^2 k_B^2 \chi}{3(g\mu_B)^2 \gamma}$$

χ~4.5x10^{-4} emu/mol
γ~20mJ/K^{2}mol

R~1.2 Close to metals

What does this mean??

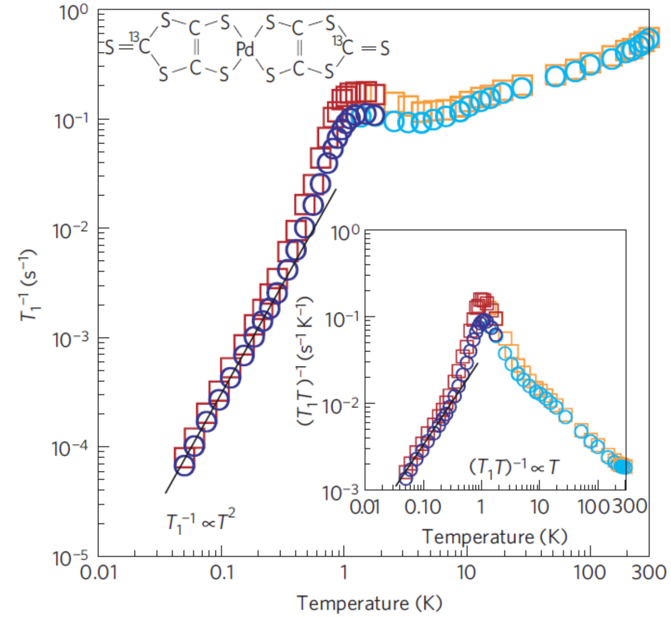
Magnetic torque down to T=0.3 K

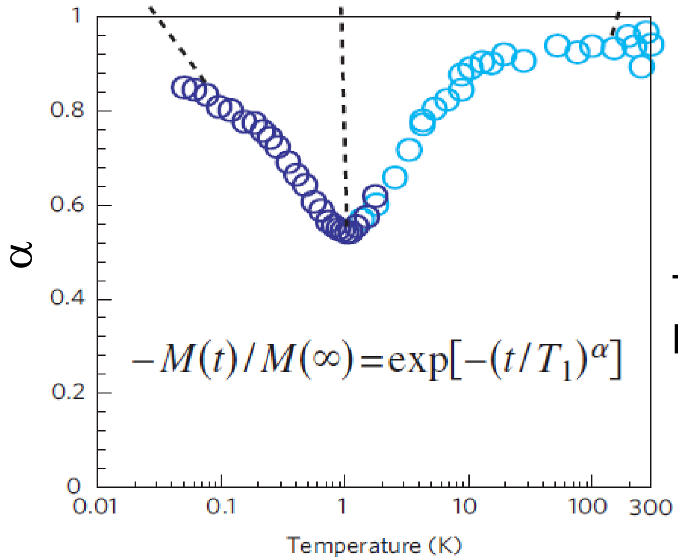


R. Okazaki *et al.* unpublished

$EtMe_3Sb[Pd(dmit)_2]_2$:Symmetry breaking in the QSL?

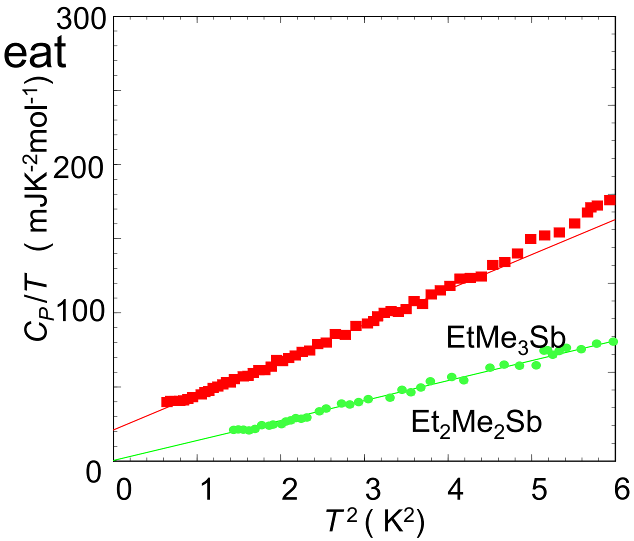
NMR (H=7 T)





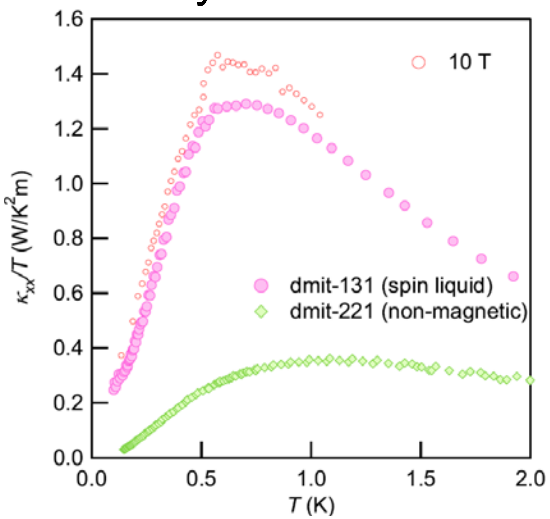
T.Itou *et al.*
Nature phys. (10)

Specific heat



Nakawawa *et al.*

Thermal conductivity



M. Yamashita *et al.*
Science (10)

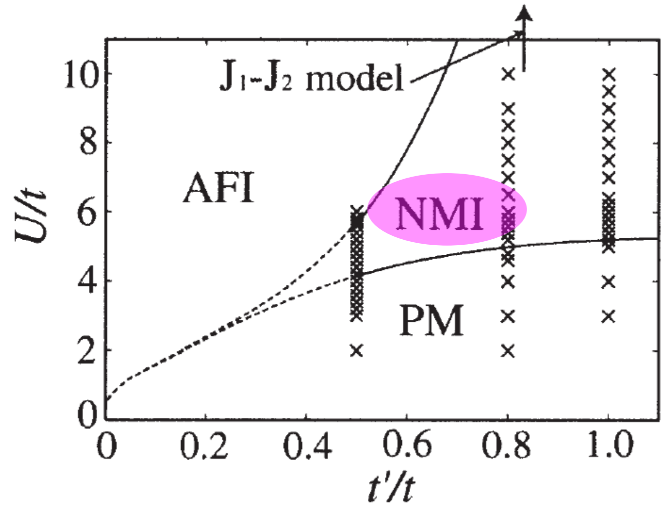
The phase transition suggested by NMR is not confirmed by specific heat, thermal conductivity and magnetic susceptibility

QSL: theory (Hubbard model)

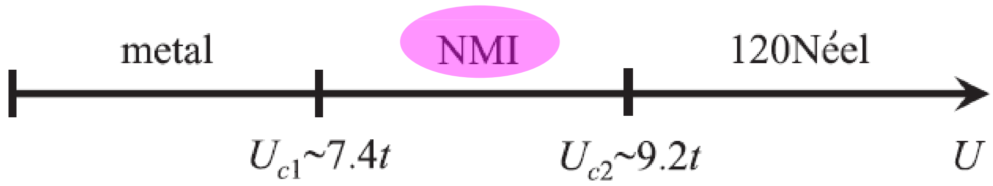
Hubbard model for a triangular lattice

$$\hat{\mathcal{H}} = -t \sum_{\langle i,j \rangle,\sigma} (\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \text{H.c.}) + U \sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

A nonmagnetic insulating state, most likely a QSL ground state, near the Mott transition



H. Morita, S. Watanabe and M. Imada, JPSJ 71, 2109 (02)



T. Yoshioka, A. Koga and N. Kawakami, PRL 103, 036401 (09)

See also

B. Kyung and A.M.S. Tremblay, PRL 97, 046402 (06)
T. Koretsune, Y. Motome and A, Furusaki, JPSJ 76, 07419 (07)

T. Mizusaki and M. Imada, PRB 74, 014421 (06)

Gapless excitation
Localized excitation

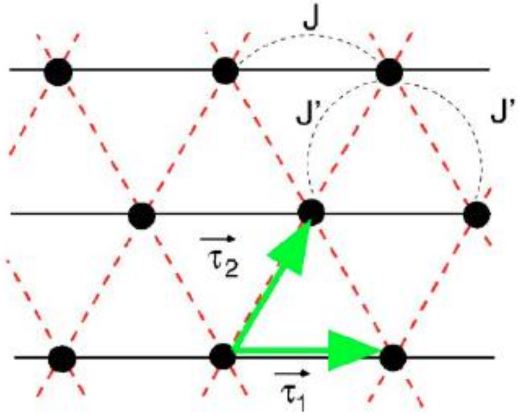
Energy resolution of these calculations is not enough to discuss low energy excitations (E~J/100)

QSL: theory (One dimensionalization)

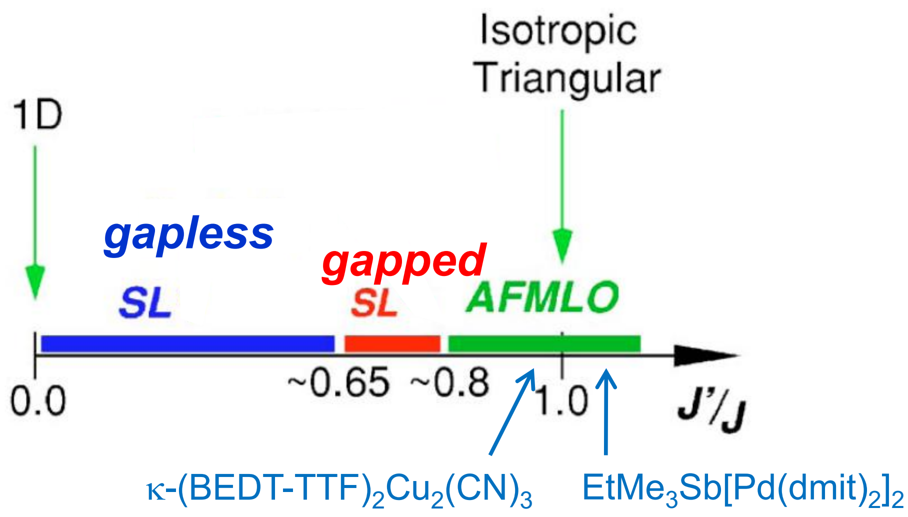
Heisenberg model

One dimensionalization

$$\hat{H} = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j + J' \sum_{\langle\langle i,j \rangle\rangle} \vec{S}_i \cdot \vec{S}_j,$$



S. Yunoki and S. Sorella, PRB 74,014408 (06)



See also Y. Hayashi and M. Ogata, JPSJ 76, 053705 (07)

The present systems are not in the QSL regime in one dimensionalization models.

QSL: theory (Ring exchange)

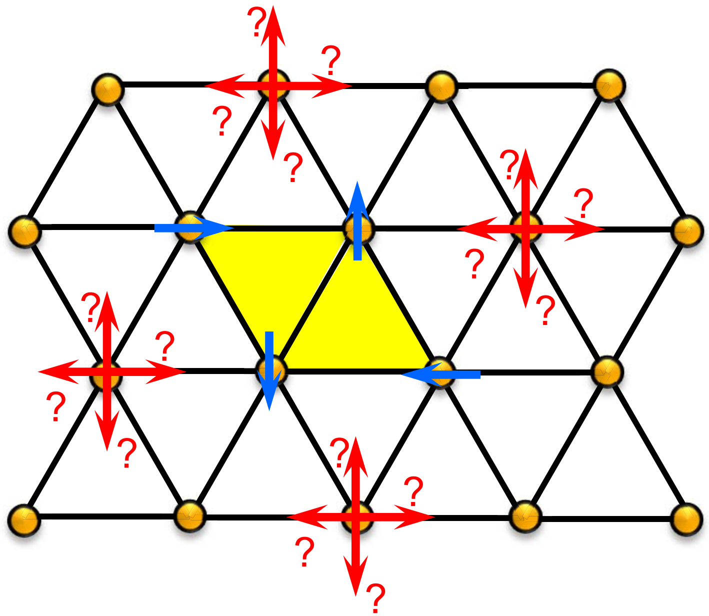
Heisenberg model for a triangular lattice

4 spin ring exchange model

$$\hat{H}_{\mathrm{ring}} = J_2 \sum_{\bullet\!-\!\bullet} P_{12} + J_4 \sum_{\square} \left(P_{1234} + P_{1234}^{\dagger}\right)$$

$$P_{1234} = (\mathbf{s}_1 \cdot \mathbf{s}_2)(\mathbf{s}_3 \cdot \mathbf{s}_4) + (\mathbf{s}_1 \cdot \mathbf{s}_4)(\mathbf{s}_3 \cdot \mathbf{s}_2) - (\mathbf{s}_1 \cdot \mathbf{s}_3)(\mathbf{s}_2 \cdot \mathbf{s}_4)$$

When $J_4>0$ $\theta_1-\theta_2+\theta_3-\theta_4=\pi$



4-spin ring exchange in triangular lattice yields a strong frustration

Quantum Spin Liquid

G. Misguich, C. Lhuillier, B. Bernu and C.Waldtmann, PRB 60, 1064 (99)

Excitation is gapped

W. LiMing, G. Misguish, P. Sindzingre and C. Luhuiller , PRB 62, 6370 (00)

Spin gap filled with a large number of singlets appear in some parameter range

QSL: theory (spinon with Fermi surface)

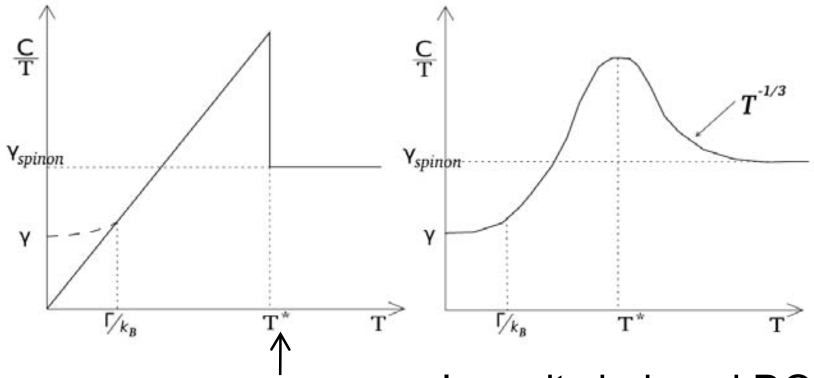
Fermionic excitation
Nodal gap

O.I. Motrunich, PRB 72, 045105 (05).
S.-S.Lee and P.A. Lee, PRL 95, 036403 (05)
S.-S.Lee, P.A. Lee and T. Senthil, PRL 98, 067006 (07)
T. Grover *et al*. PRB 81, 245121 (10)

Gapless excitation

γ-term in specific heat	yes
Finite κ_{xx}/T at $T\rightarrow 0$ K	yes
Wilson ratio $R\sim 1$	yes
NMR T_1 (nodal)	yes?
Thermal Hall angle $\tan\theta_H=\kappa_{xy}/\kappa_{xx}$	no

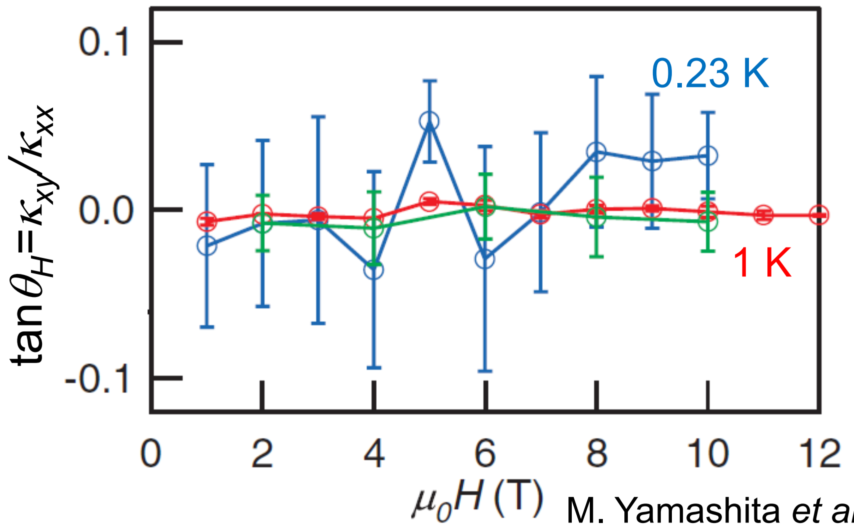
H. Katsura, N. Nagaosa and P.A. Lee, PRL 104, 066403 (10)

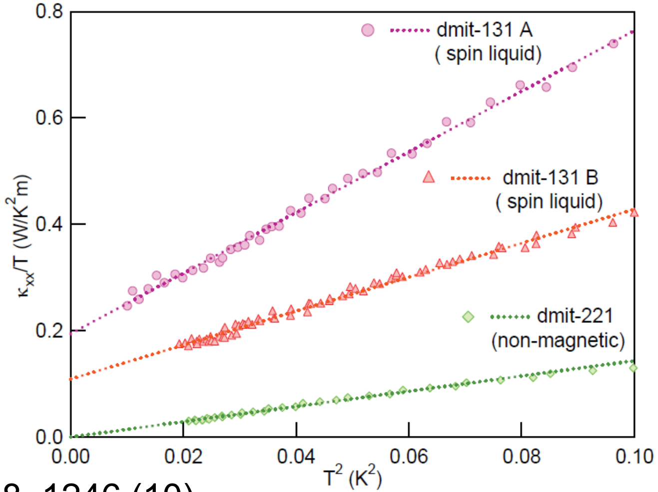


Spinon instability

Impurity induced DOS (*d*-wave superconductor)

Universal thermal conductivity κ_{xx}/T no





M. Yamashita *et al*. Science 328, 1246 (10)

QSL: theory

Quantum dimer model

Gapped

R. Moessner and S. Sondhi
PRL 86, 1881 (01)

Chiral spin liquid

Gapped

V. Kalmeyer and R. B. Laughlin
PRL 59, 2095–2098 (1987)

Z_2 spin liquid

Gapped (Vison)

Y. Qi, C. Xu, and S. Sachdev
Phys. Rev. Lett. 102, 176401 (09)

Algebraic spin liquid

Gapless spinon

X. G. Wen, PRB 65, 165113 (02)

Spin Bose-metal

Gapless boson

D.N. Sheng, O.I.Motrunich and M.P.A.Fisher
PRB 79, 205112 (09)

Summary: Remarkable features in a 2D candidate quantum spin liquid

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ No magnetic order down to T~J/10,000

Gapless or gapped?

Inhomogeneity and microscopic phase separation

EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ No magnetic order down to T~J/10,000

Homogeneous (Suitable for the study of genuine feature in a QSL state)

Highly unusual elementary excitations

Nonmagnetic gapless excitations
Magnetic gapped excitation

Bipartite nature

Excitation is itinerant (highly mobile: mean free path~1 μm)

Extremely long correlation length (Power law correlation function)

No thermal Hall effect up to 12 T

Wilson ration~1.2

Symmetry breaking in the QSL state?

Thermal conductivity

M.Yamashita and Y.M. *et al*. Nature Phys. **5**, 44 (09)

M. Yamashita and Y.M. *et al*. Science **328**, 1246 (10)

Future experiments

Thermal Hall conductivity and torque measurements up to 30 T

In-plane anisotropy (nodal, nematic ･･･)