KITP Conference 8th Oct. 2012

Exotic Phases of Frustrated Magnets

# Fermi Liquid Character of Organic Spin Liquids in $X[Pd(dmit)_2]_2$ System

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Satoshi YAMASHITA

# Outline

- Background : organic spin liquid compounds

  Gap-less characters of organic spin liquids in specific heat

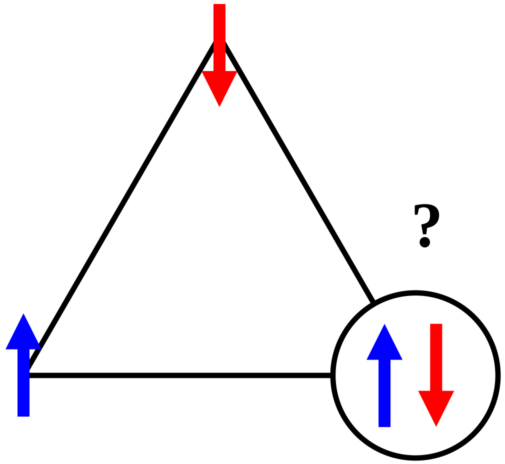
- Motivation : Fine tuning of degree of frustration in $XY[Pd(dmit)_2]_2$

- Result I : Fermi liquid like character

- Result II : The possibility of critical behavior on the phase boundary

- Summary

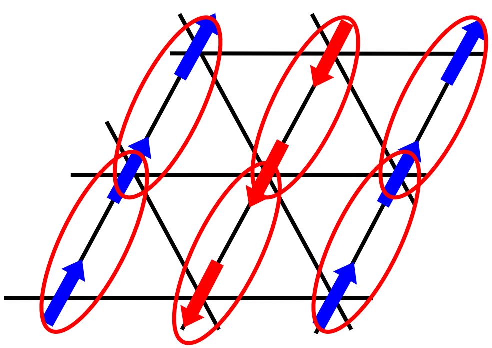
# Quantum spin liquid on 2D triangular system



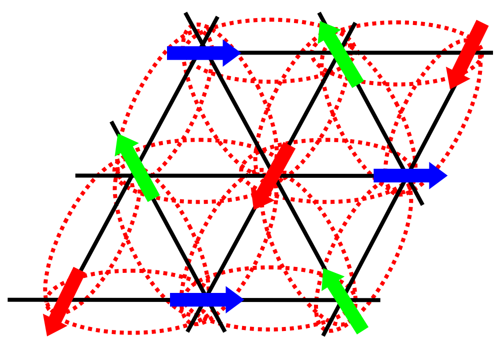

**Geometrical frustration problem**

Neél order is unstable

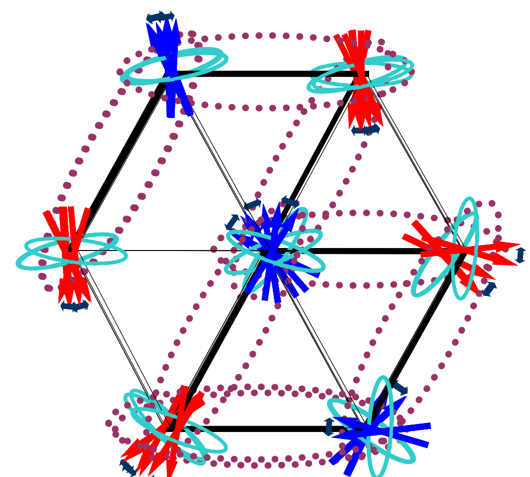
Ising type



Heisenberg



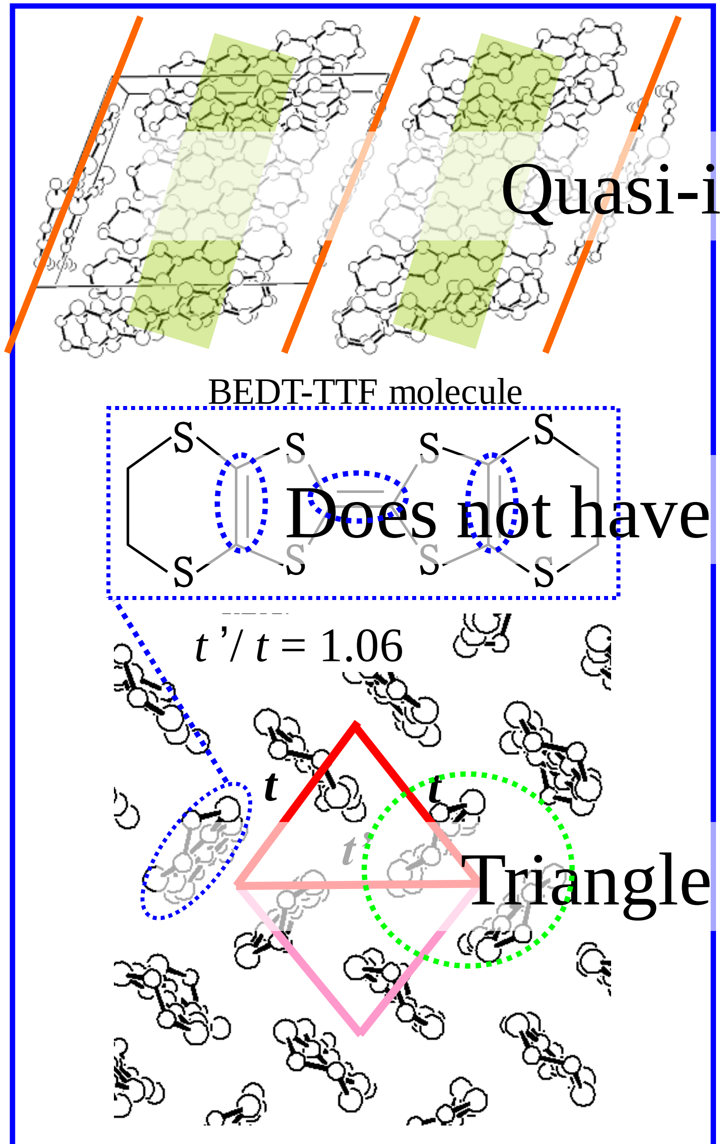
Possibility of spin liquid

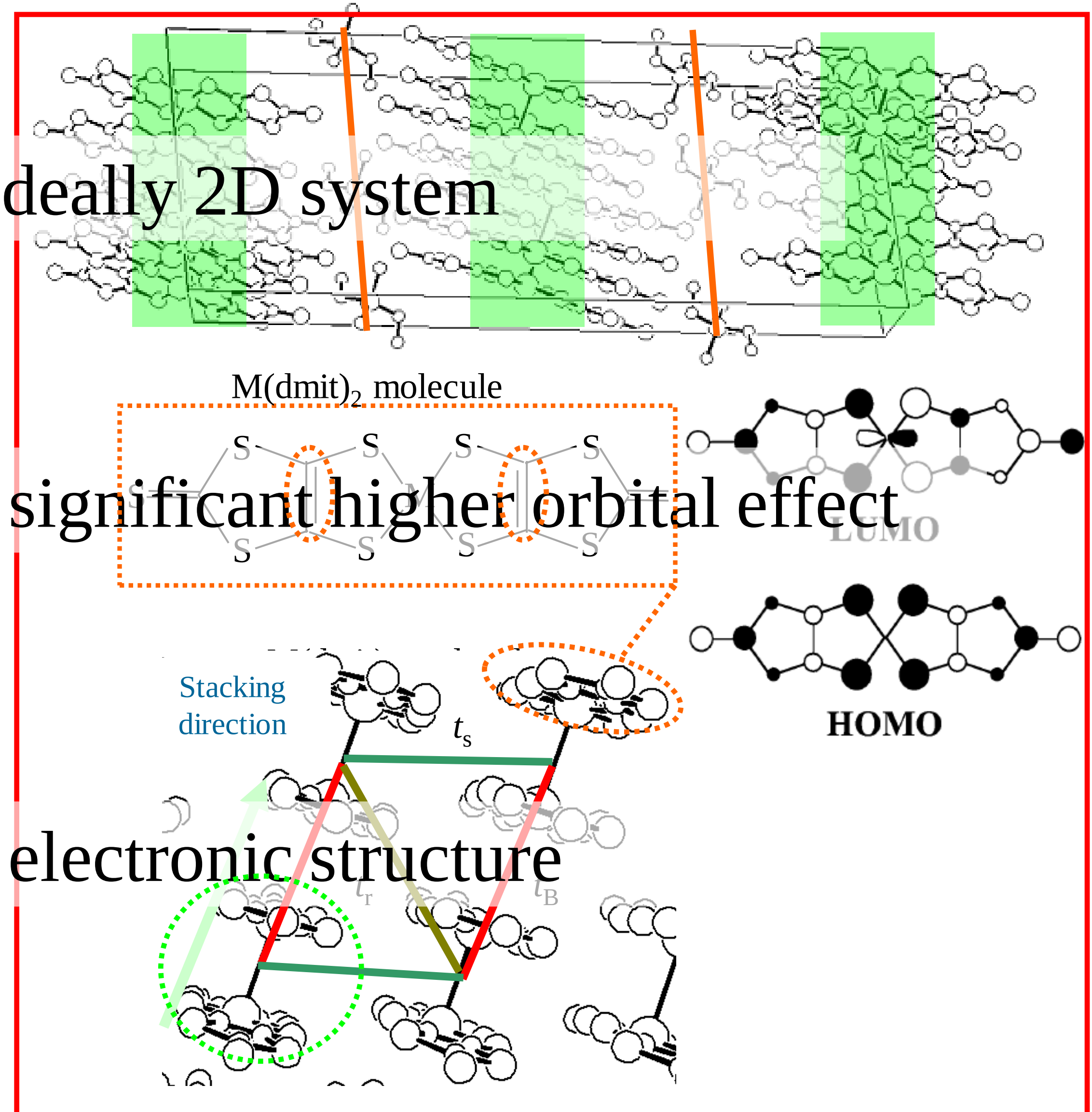


# Organic spin liquid compounds

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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




Quasi-ideally 2D system

Does not have significant higher orbital effect

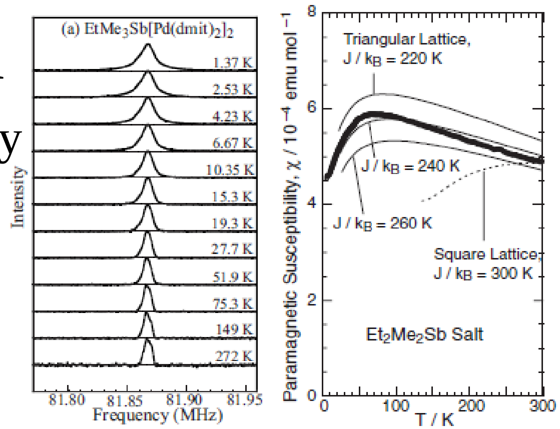
Triangle electronic structure

# Realization of quantum spin liquid

## No long range ordering

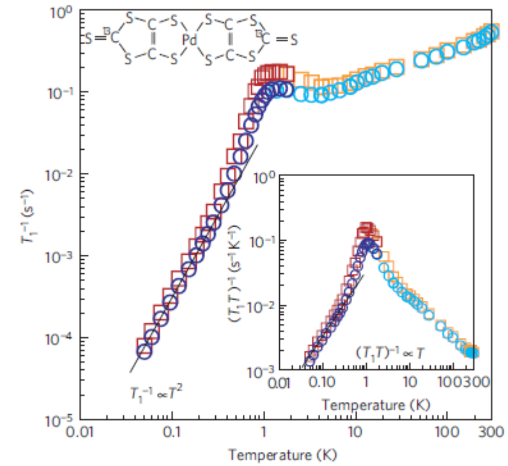
$^{13}$C NMR

Susceptibility




T. Itou *et. al.*, *Phys. Rev. B* **77**(2008)104413

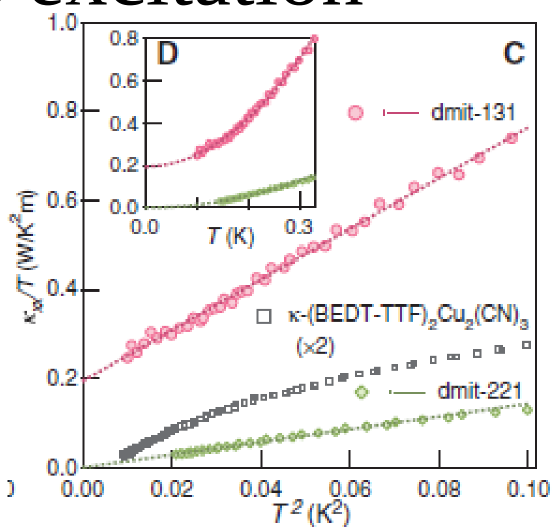
## Gap?




T. Itou, S. Maegawa *et al.*, *Nature Phys* 6, *673-676.* (2010)

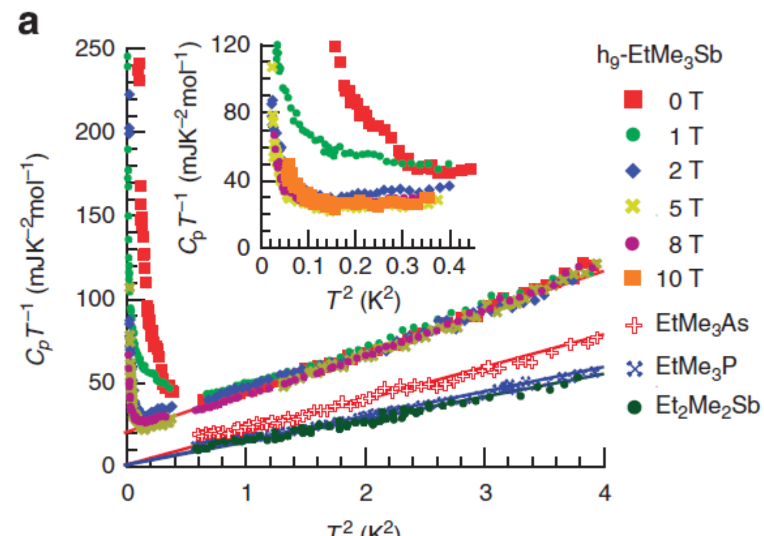
## Gap-less excitation

Thermal Conductivity




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

## Specific Heat

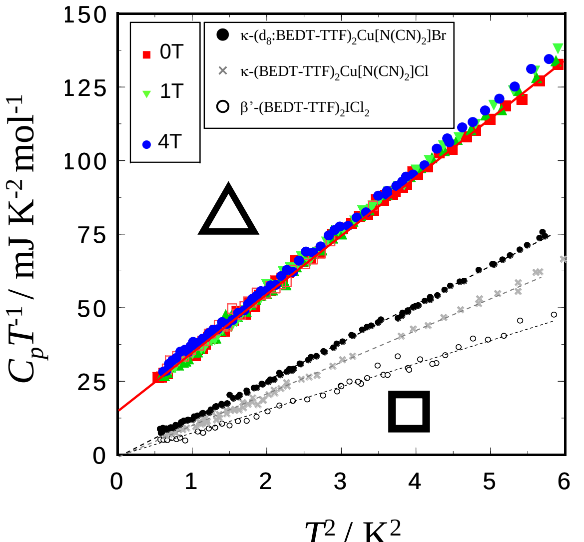



S. Yamashita *et al.*, *Nature Commun.*, 2, 275 (2011).

# Specific heat measurements

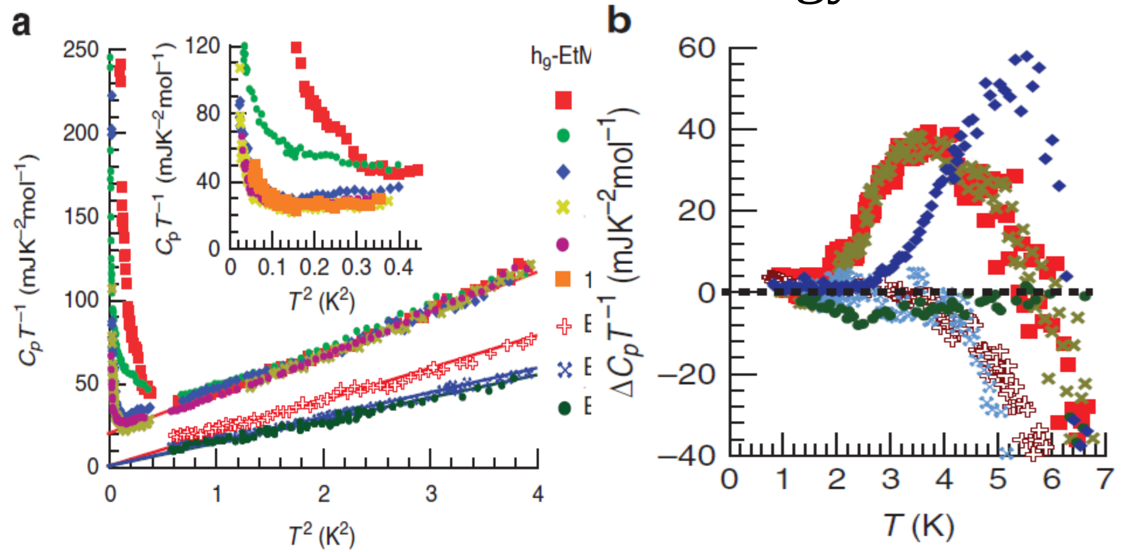
## Gap-less behavior

## Low-energy excitation




$\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

S. Yamashita *et al.*, *Nature Phys.* **4**, 459(2008)




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

S. Yamashita *et al.*, *Nature Commun.*, 2, 275 (2011).

$$C_p \approx \frac{\partial U}{\partial T} (at \quad low \quad temp.)$$

Effective method to detect low-energy excitation

Quantitative discussion to clarify the excitation structure

We can determine absolute value of $C_p$

$$\gamma = \frac{\pi^2}{3} k_B N D(E_F)$$

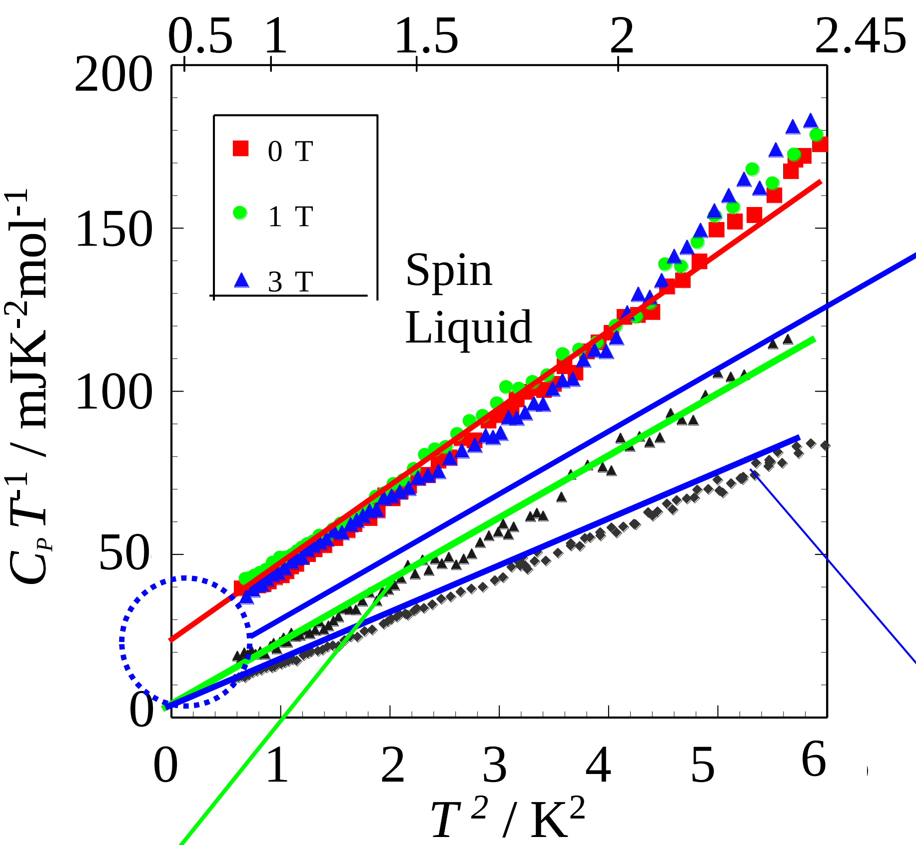
# The Fermi liquid character of organic spin liquid

Specific heat data of $EtMe_3Sb[Pd(dmit)_2]_2$

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

*T*-linear term

Lattice contribution




$EtMe_3Sb[Pd(dmit)_2]_2$

$\gamma = 19.9\ mJK^{-2}mol^{-1}$

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ ×1.5

$\gamma = 12.9\ mJK^{-2}mol^{-1}$

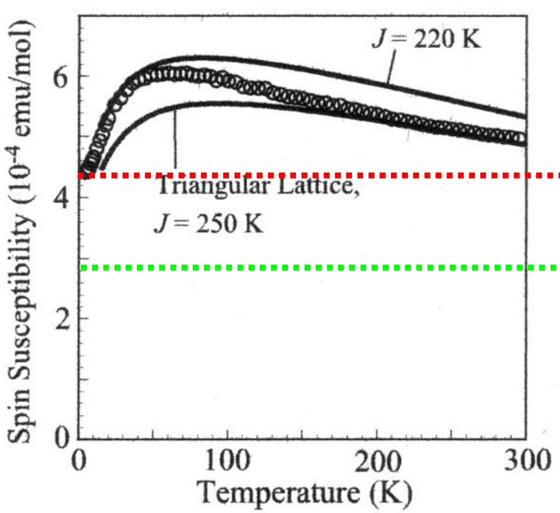
$EtMe_3As[Pd(dmit)_2]_2$ $T_N \approx 23$ K AFI

$EtMe_3P[Pd(dmit)_2]_2$ (monoclinic) $T_{trs} \approx 25$ K Non magnetic (VBS)

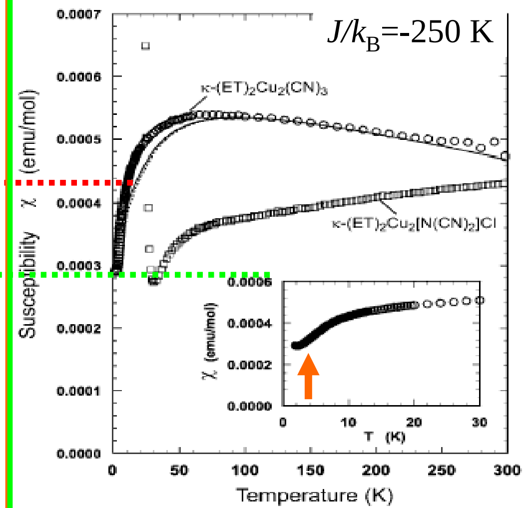
# The Fermi liquid character of organic spin liquid

## Magnetic susceptibility

$EtMt_3Sb[Pd(dmit)_2]_2$




κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$




T. Itou *et. al.*, *Phys. Rev. B* **77**(2008)104413 Y. Shimizu *et. al.*, *Phys. Rev. Lett.* **91** 107001(2003)

Wilson Ratio

$R_w \approx 1.5\text{-}1.6$

γ is same order to organic metals.

( γ of κ-$(BEDT\text{-}TTF)_2Cu(NCS)_2$ is 30 $mJK^{-2}mol^{-1}$)

Fermi Liquid character

$EtMe_3Sb[Pd(dmit)_2]_2$

$\chi_0 = 4.4\times10^{-4}$ emu / mol

$\gamma = 19.9\ mJK^{-2}mol^{-1}$

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$

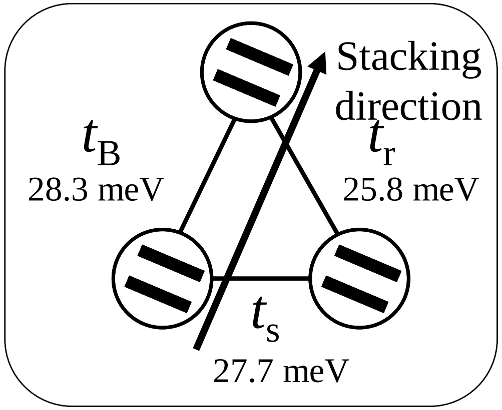
$\chi_0 = 2.9\times10^{-4}$ emu / mol

×1.5

$\gamma = 12.9\ mJK^{-2}mol^{-1}$

# Motivation : Fine tuning of anisotropy of triangle structure

**$EtMe_3Sb[Pd(dmit)_2]_2$**



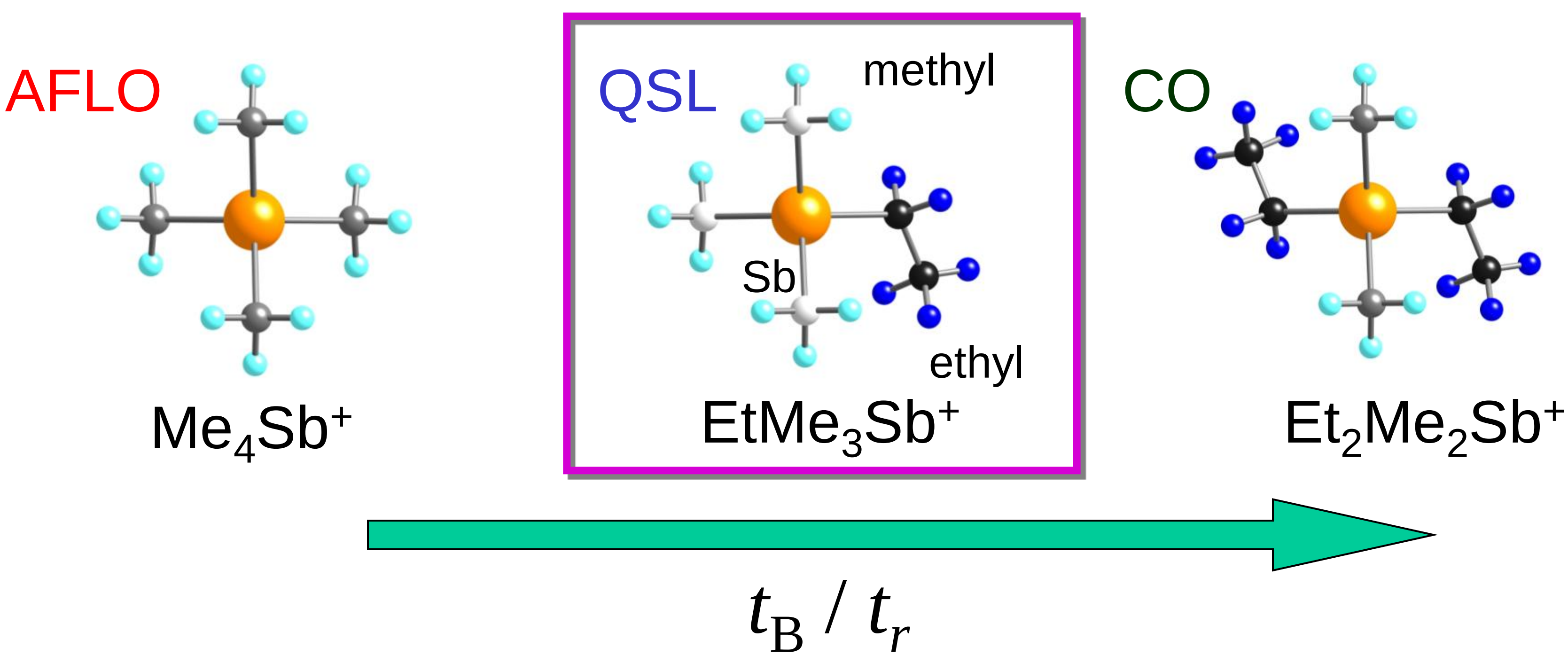

T. Itou *et. al.*, *Phys. Rev. B* **77**(2008)104413

**Difference of transfer integrals ($t_B$, $t_s$, $t_r$)**

$$t_B : t_s : t_r = 1.09 : 1.07 : 1$$

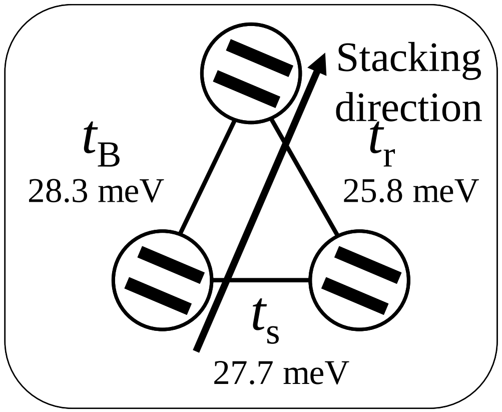
Anisotropic triangle structure

## Cation



# Motivation : Fine tuning of anisotropy of triangle structure

**$EtMe_3Sb[Pd(dmit)_2]_2$**

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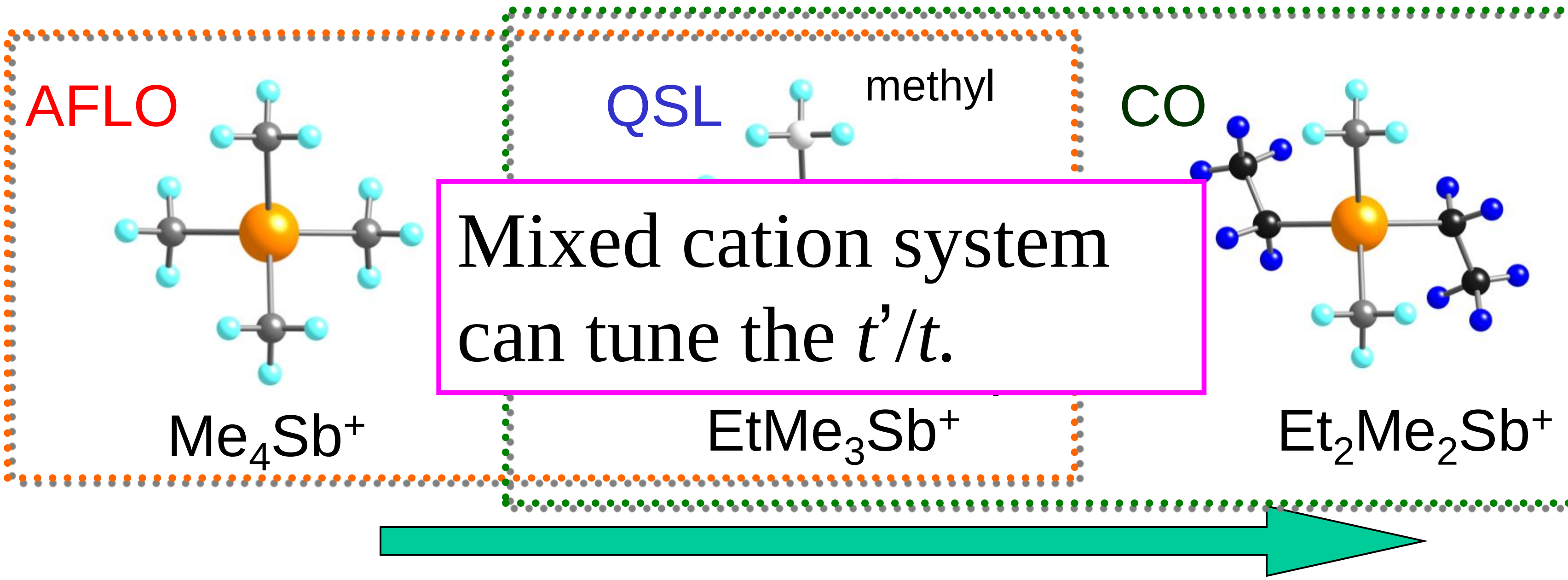
Anisotropic triangle structure

T. Itou *et. al.*, *Phys. Rev. B* **77**(2008)104413

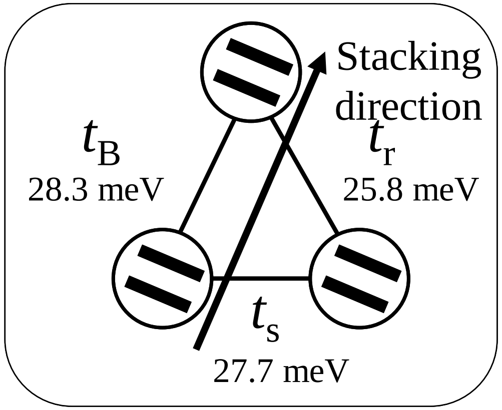
Cation




$t_B$ / $t_r$

# Motivation : Fine tuning of anisotropy of triangle structure

**$EtMe_3Sb[Pd(dmit)_2]_2$**

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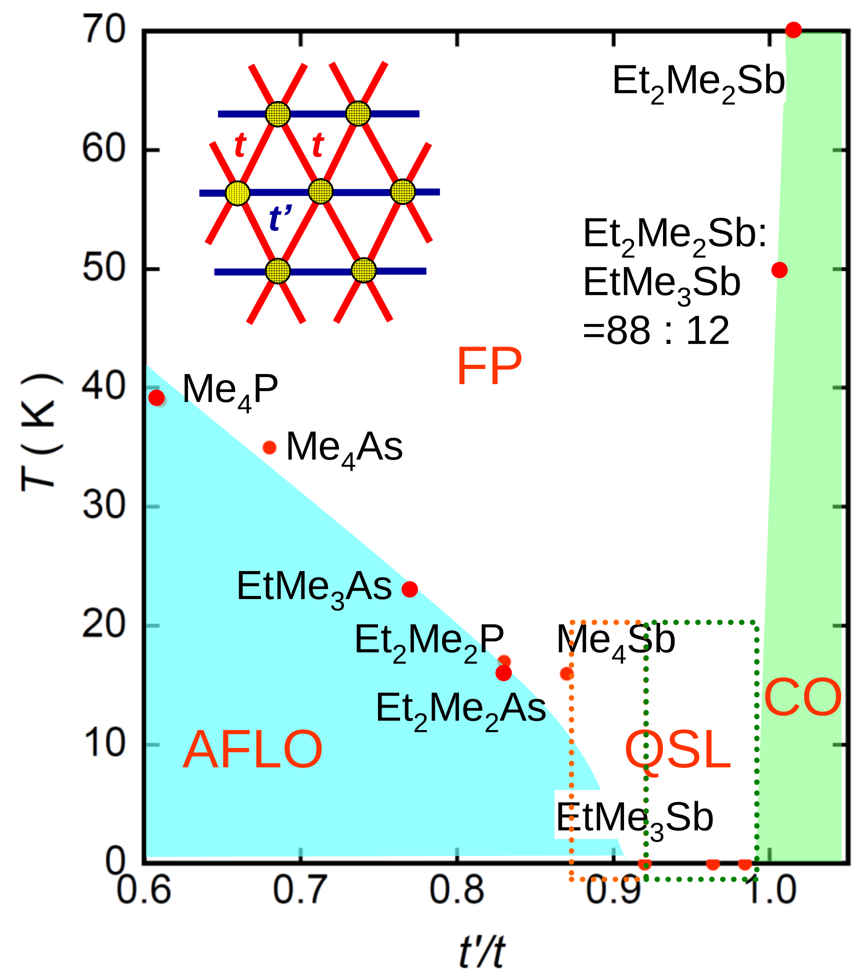
**Difference of transfer integrals ($t_B$, $t_s$, $t_r$)**

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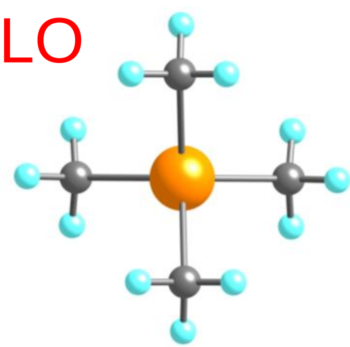
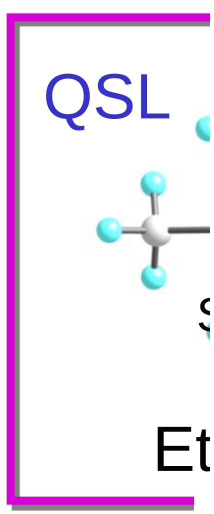


$Me_4Sb^+$

C




Fine tuning by mixing cation

$t_B$ / K. Kanoda and R. Kato. Annu. Rev. Condens. Matter Phys., **2**, (2011).167-188

# Motivation

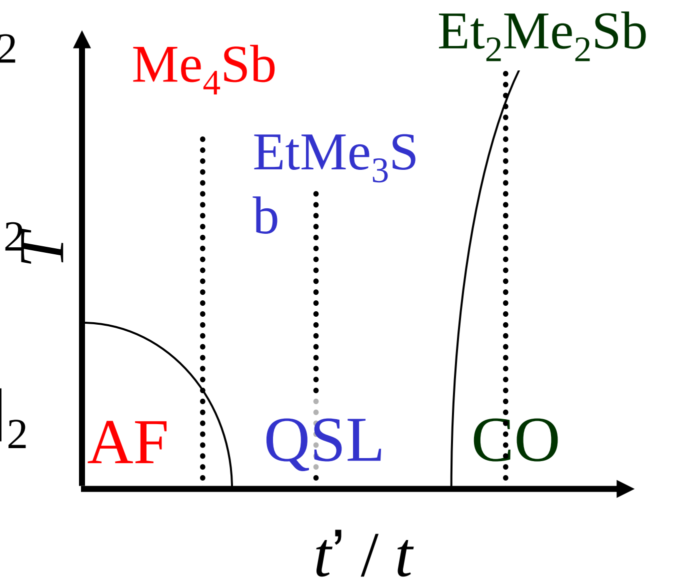
To clarify the excitation structure of organic spin liquids from the stand point of quantitative discussion by specific heat measurement.

- Relation between anisotropy of triangle and gap-less character.

AF + QSL $(Me_4Sb)_x(EtMe_3Sb)_{1-x}[Pd(dmit)_2]_2$

QSL+CO $(EtMe_3Sb)_{1-y}(Et_2Me_2Sb)_y[Pd(dmit)_2]_2$

AF +CO $(Et_2Me_2As)_{1-z}(Et_2Me_2Sb)_z[Pd(dmit)_2]_2$



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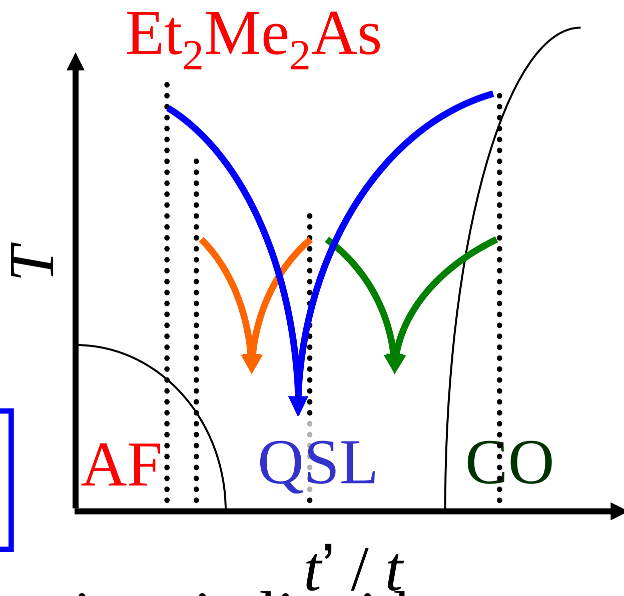
AF +CO $(Et_2Me_2As)_{1-z}(Et_2Me_2Sb)_z[Pd(dmit)_2]_2$



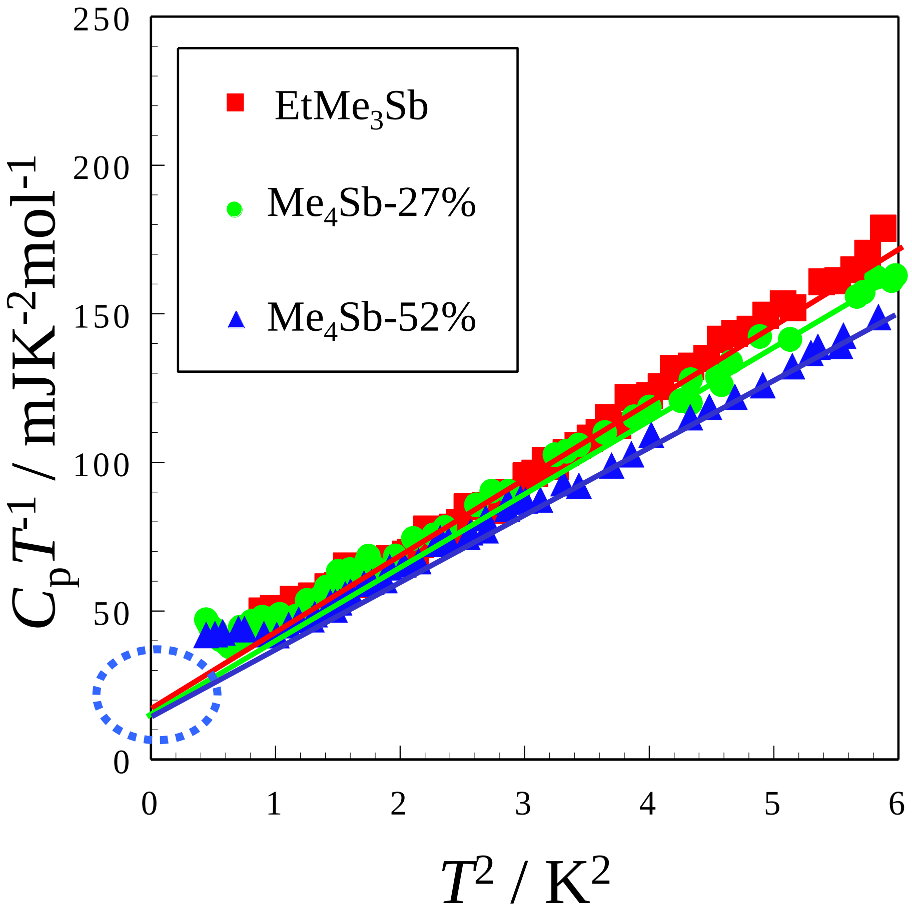

Three types of approach to organic spin liquid.

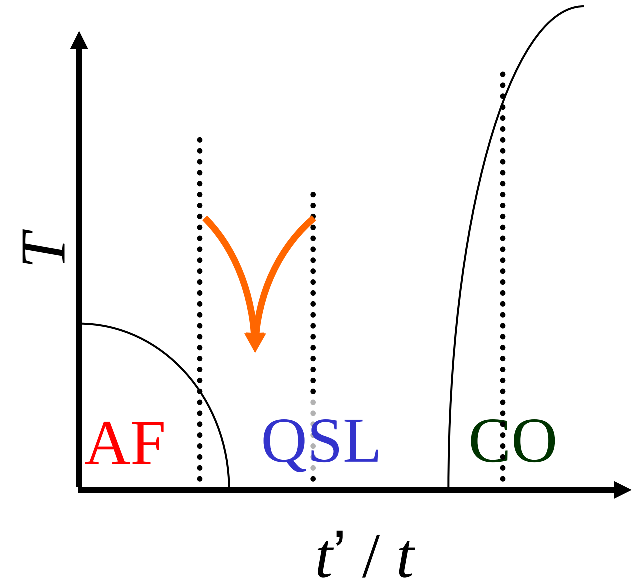
Systematic study

**In this work, we have performed specific heat measurement for these three types of mixed compounds.**

# Quantum spin liquid in mixing cation system $XY[Pd(dmit)_2]_2$

Specific heat data of $(Me_4Sb)_x(EtMe_3Sb)_{1-x}[Pd(dmit)_2]_2$



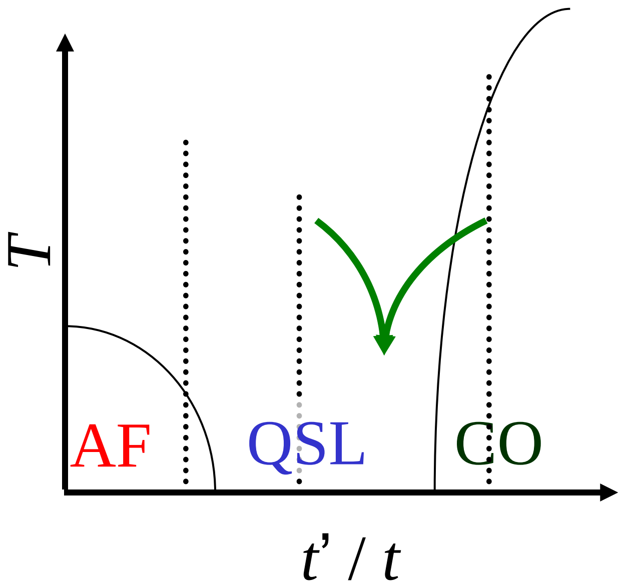

In this region, the bulk spin liquid is realized.

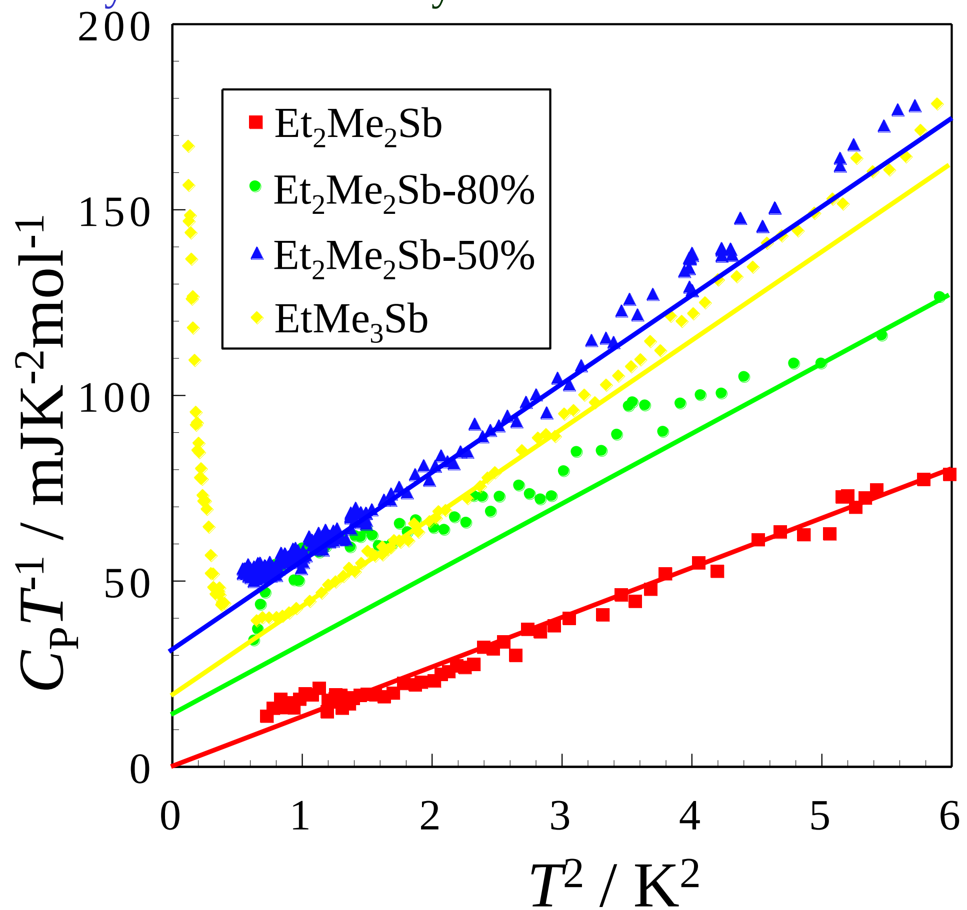
$\gamma$ : 14-20 mJK$^{-2}$mol$^{-1}$

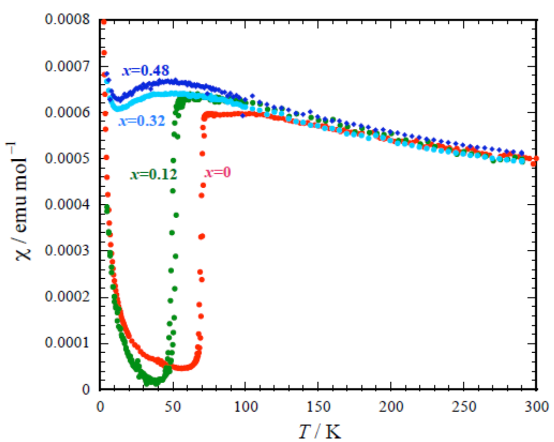
$R_W$ : 1.2-1.6

# Quantum spin liquid in mixing cation system $XY[Pd(dmit)_2]_2$

## Specific heat data of $(EtMe_3Sb)_{1-y}(Et_2Me_2Sb)_y[Pd(dmit)_2]_2$



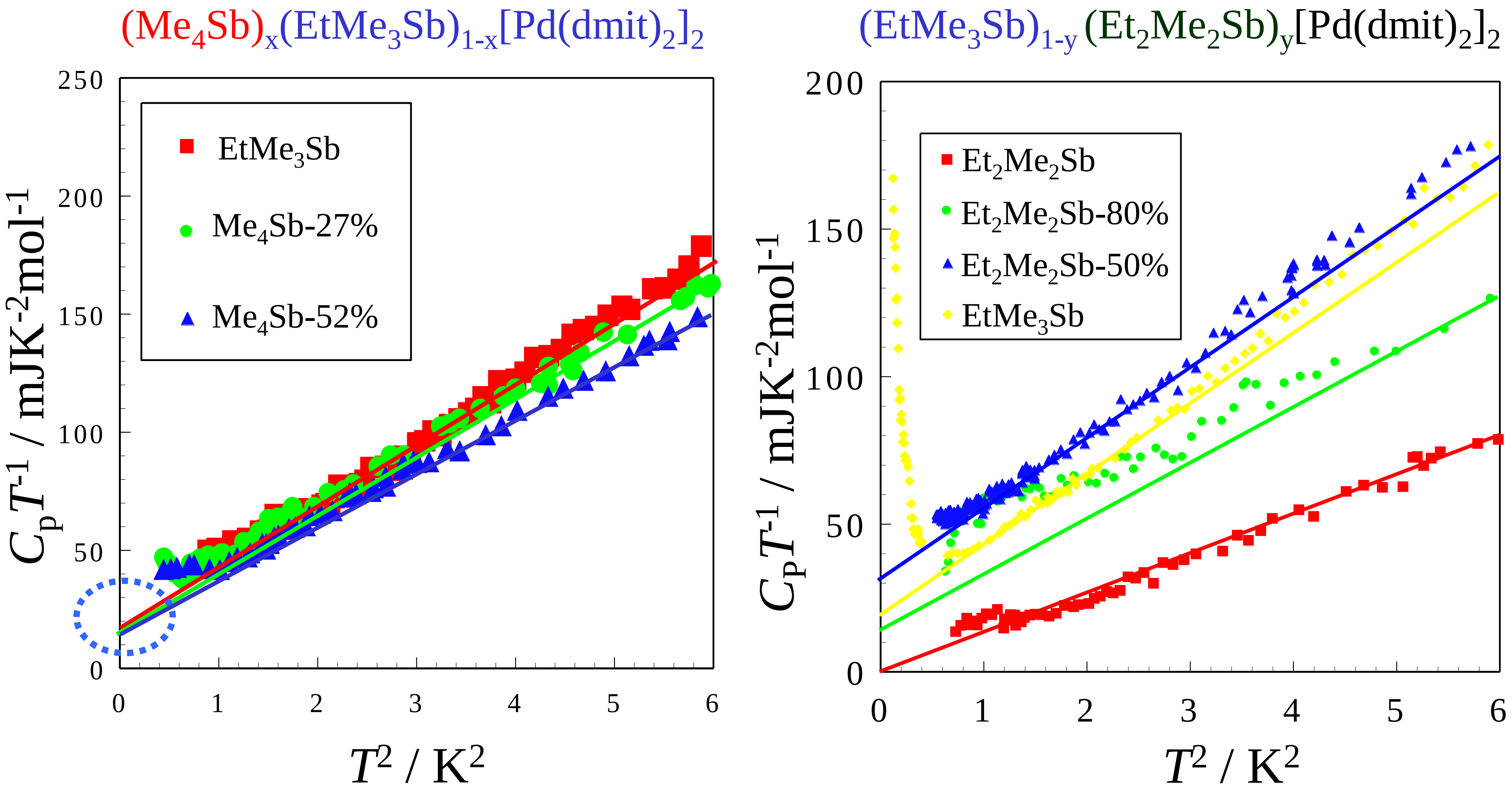




$(EtMe_3Sb)_{0.5}(Et_2Me_2Sb)_{0.5}[Pd(dmit)_2]_2$

$\gamma$=30-35 mJ K$^{-2}$mol$^{-1}$

$\chi_0 = 6.2 \times 10^{-4}$ emu/mol $R_w$ = 1.3-1.4

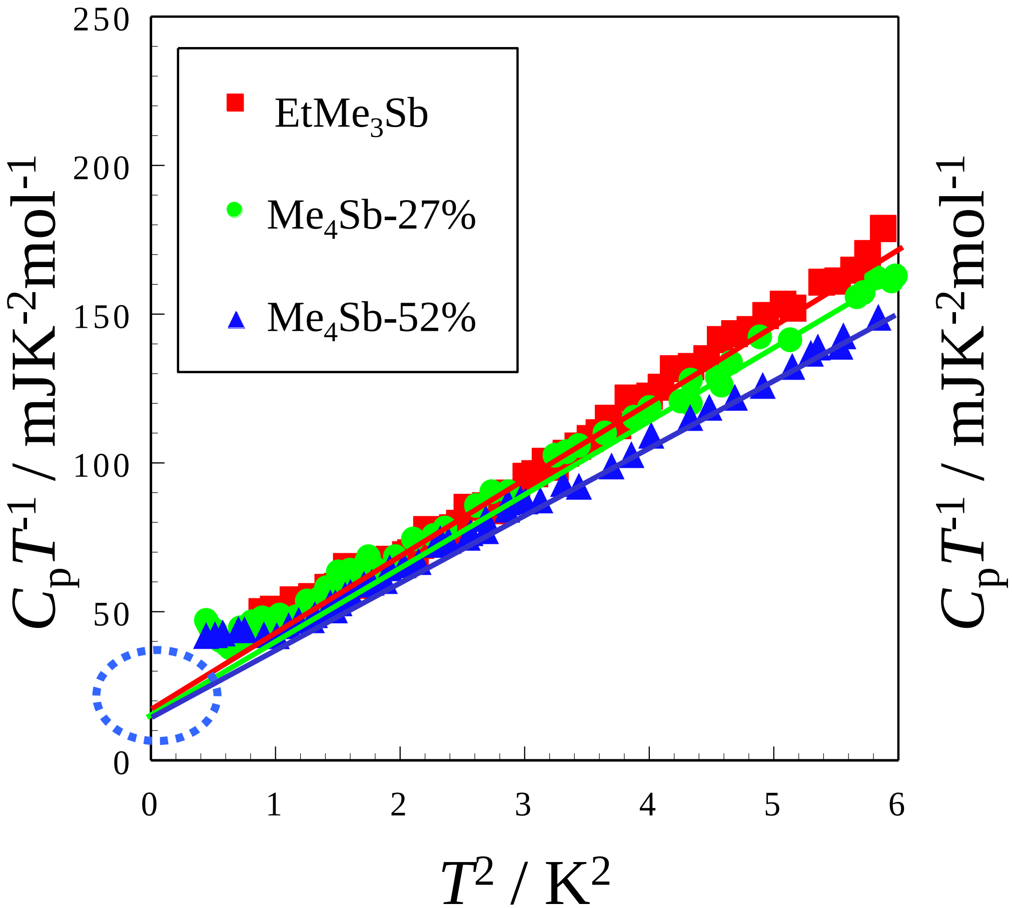
# Quantum spin liquid in mixing cation system $XY[Pd(dmit)_2]_2$

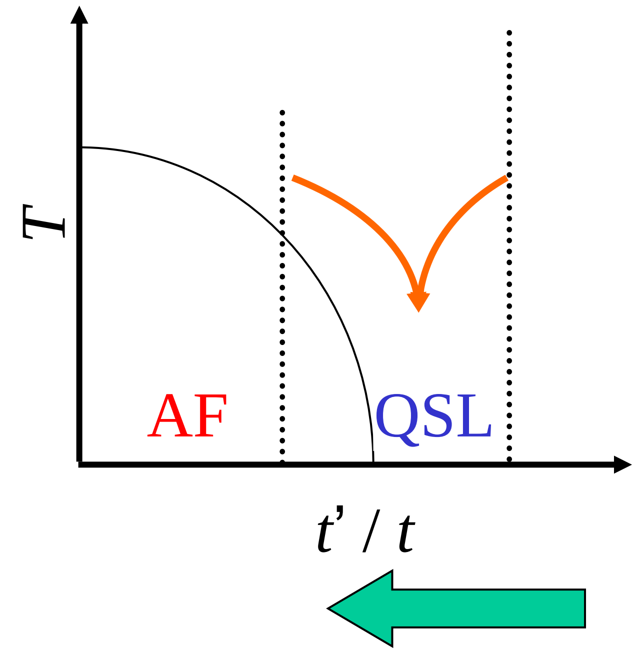



AF+QSL and QSL+CO system show **bulk quantum spin liquid** behavior with Fermi liquid character ($R_w$ is close to 1).

# Quantum spin liquid in mixing cation system $XY[Pd(dmit)_2]_2$

## Specific heat data of $(Me_4Sb)_x(EtMe_3Sb)_{1-x}[Pd(dmit)_2]_2$




We also approach to the phase boundary between AF and QSL.

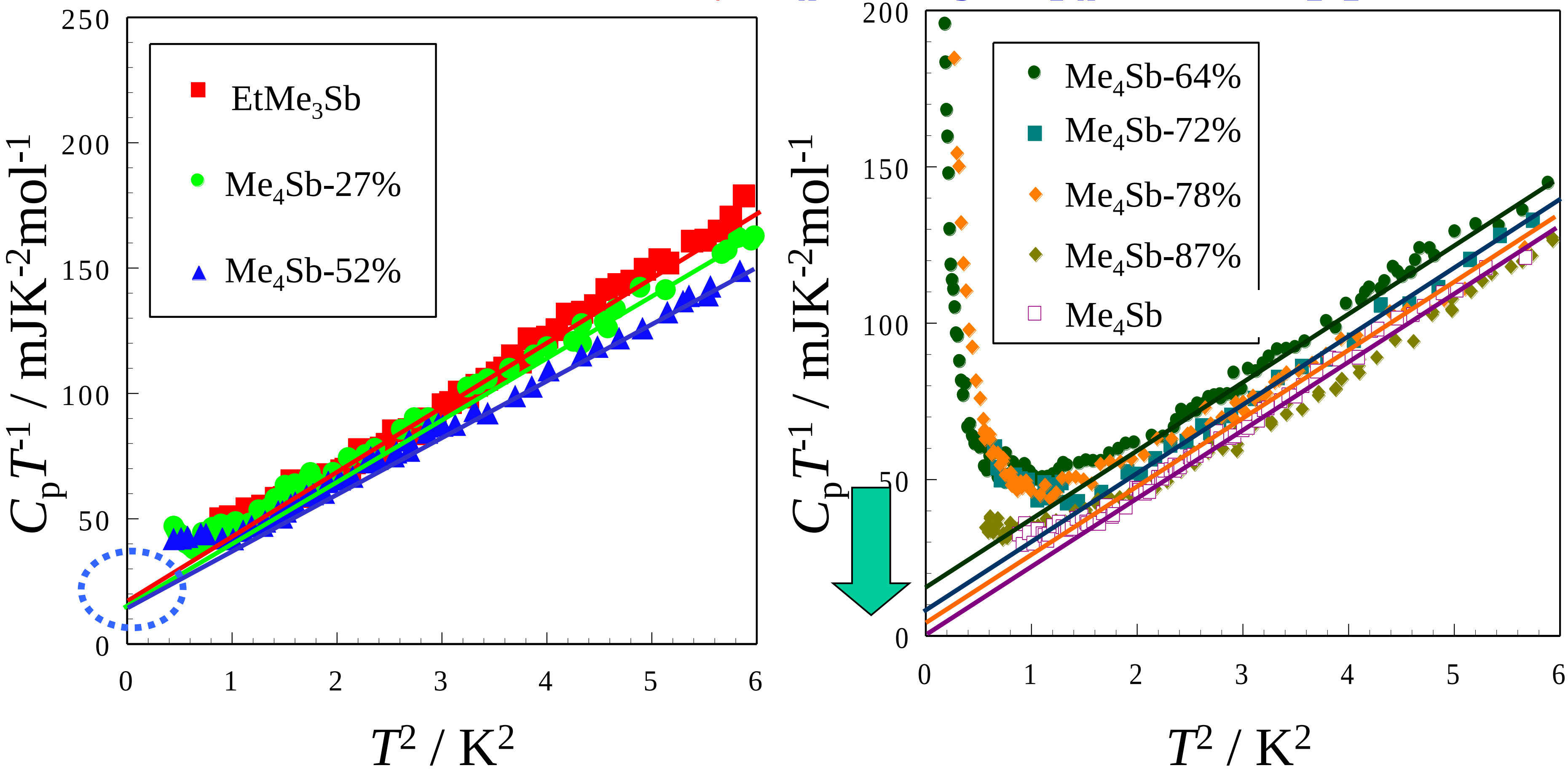
$$C_p T^{-1} = A T^{-3} + \gamma + \beta T^2$$

**γ=0 Gapped state**

**γ=finite Gap-less Spin liquid**

# Gap-less behavior on the phase boundary in $XY[Pd(dmit)_2]_2$

Specific heat data of $(Me_4Sb)_x(EtMe_3Sb)_{1-x}[Pd(dmit)_2]_2$

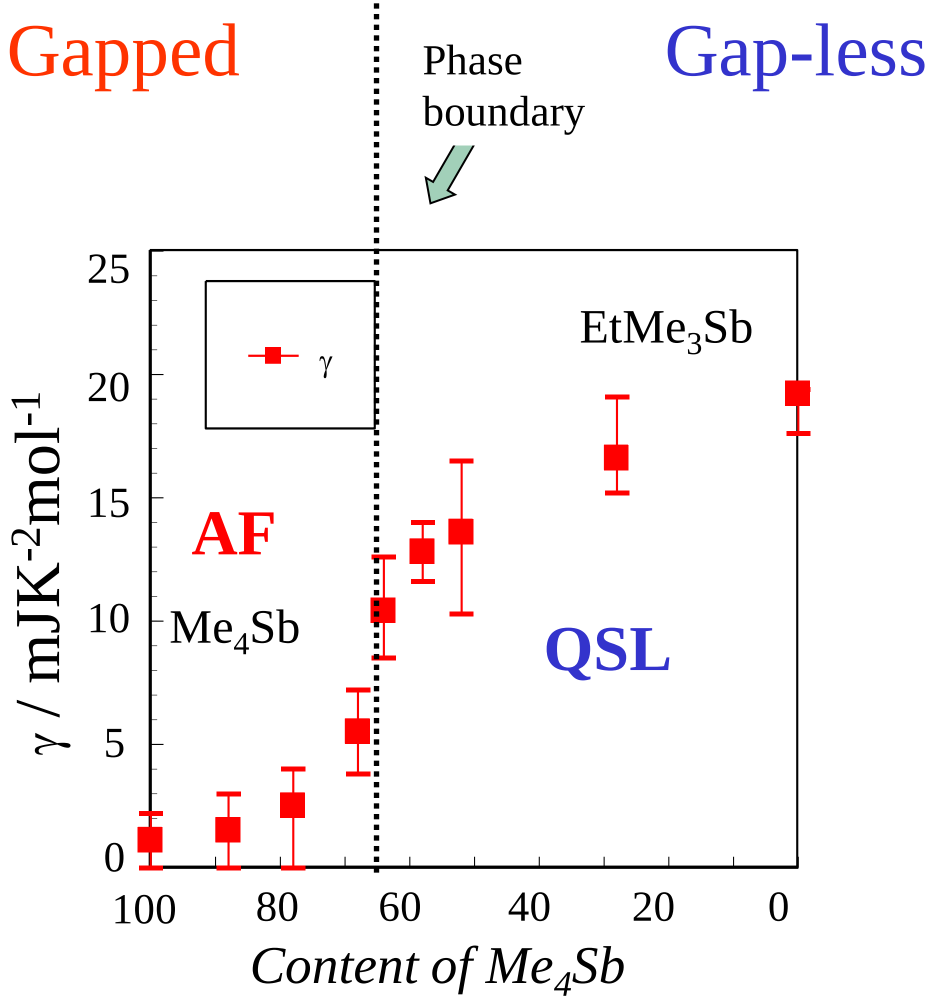


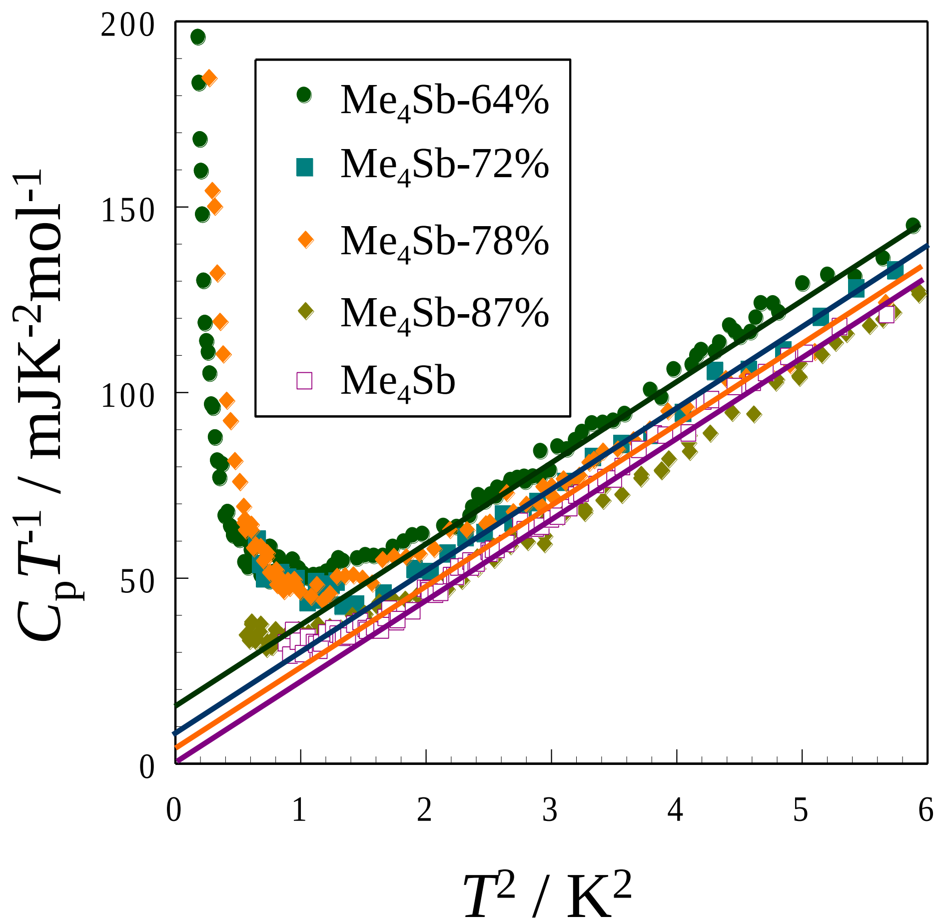

$$C_p T^{-1} = AT^{-3} + \gamma + \beta T^2$$

**γ=0 Gapped state**

**γ=finite Gap-less Spin liquid**

# The vanishing of gap-less character at the phase boundary

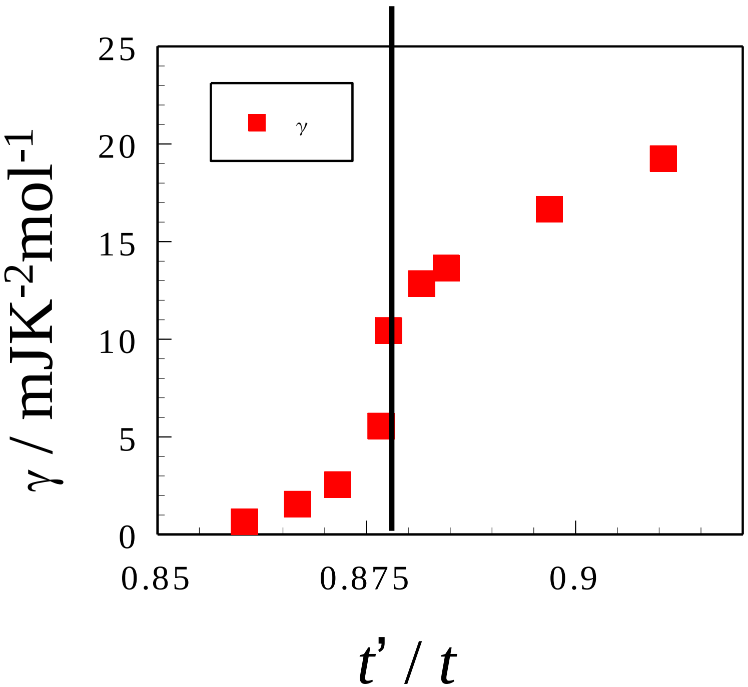


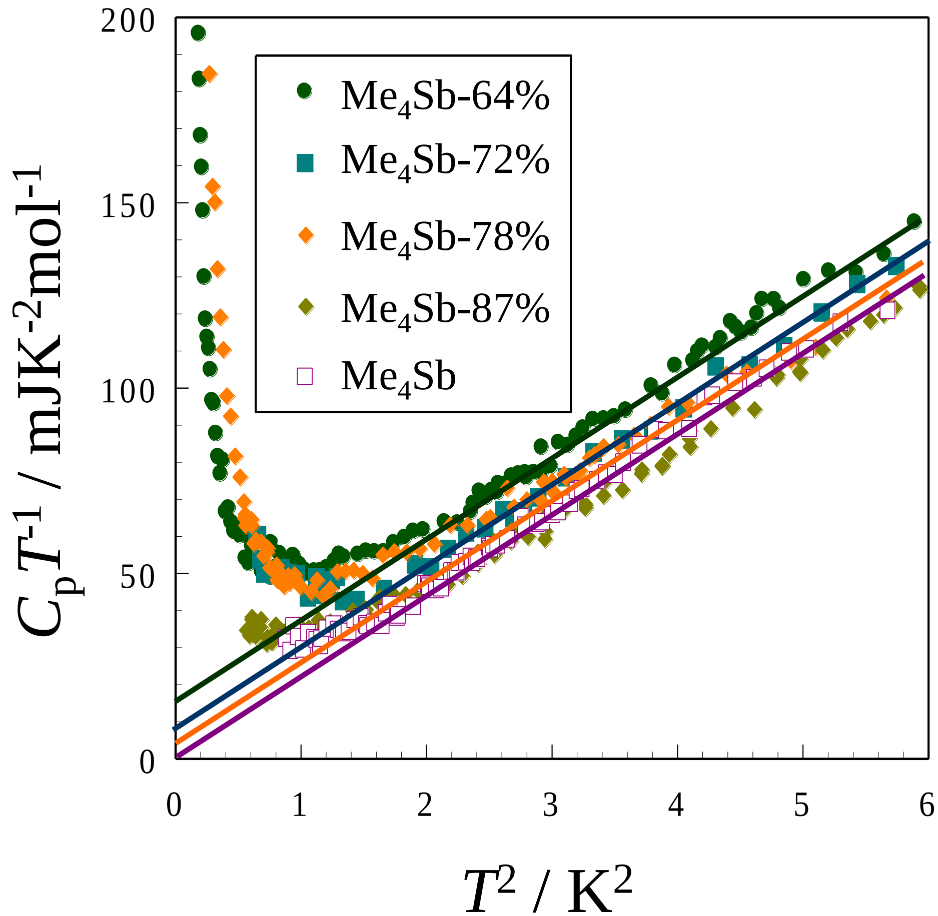




**The phase boundary**

$(Me_4Sb)_{0.64}(EtMe_3Sb)_{0.36}[Pd(dmit)_2]_2$

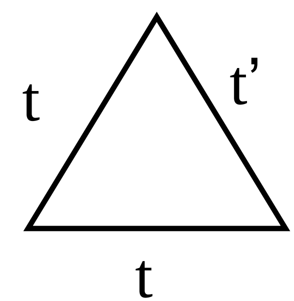
# The vanishing of gap-less character at the phase boundary




**The phase boundary**

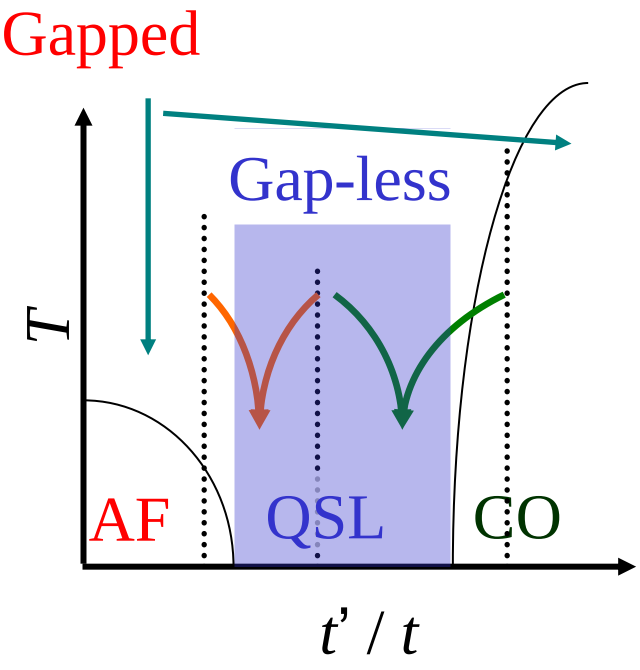
$t' / t = 0.87_8$




$(Me_4Sb)_{0.64}(EtMe_3Sb)_{0.36}[Pd(dmit)_2]_2$

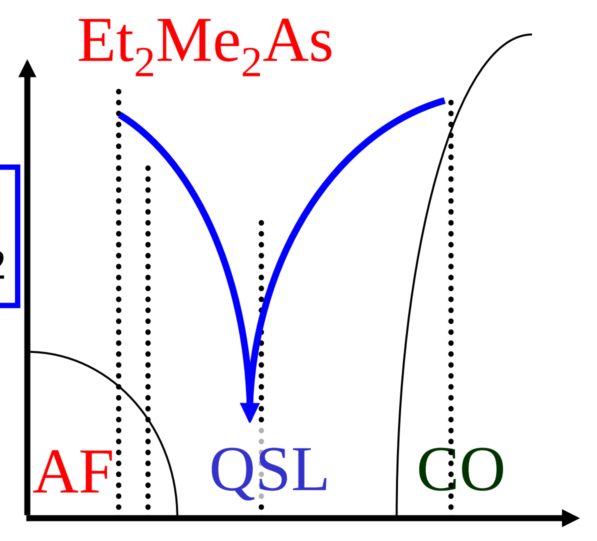
AF + QSL $(Me_4Sb)_x(EtMe_3Sb)_{1-x}[Pd(dmit)_2]_2$

QSL+CO $(EtMe_3Sb)_{1-y}(Et_2Me_2Sb)_y[Pd(dmit)_2]_2$

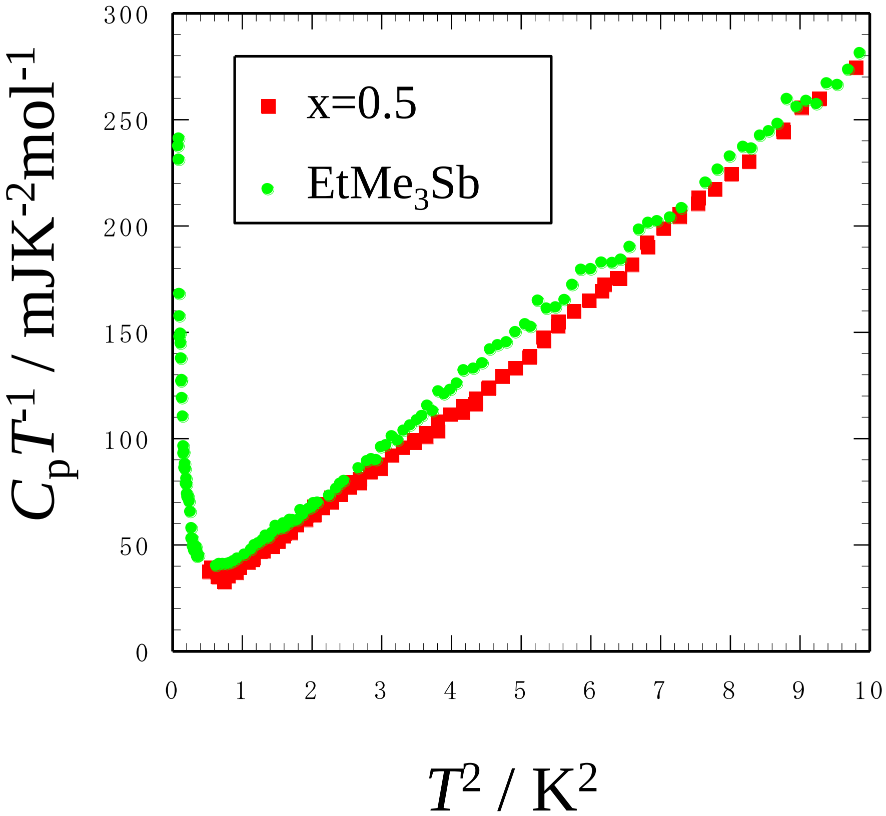


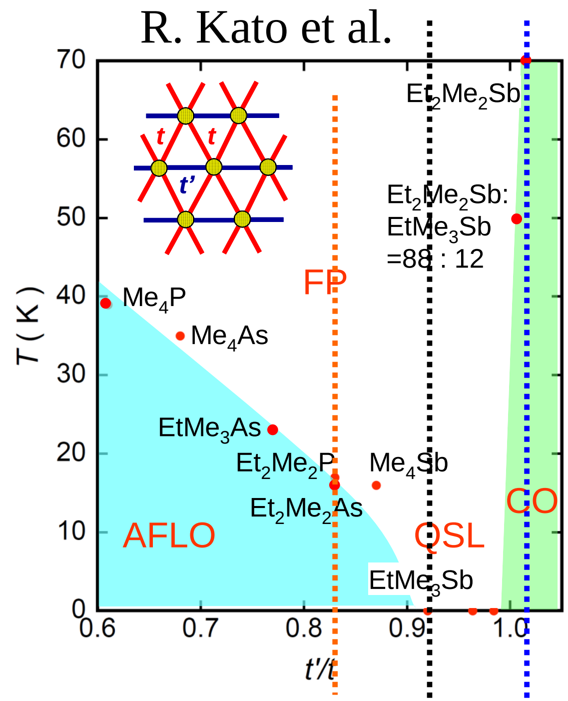

Bulk spin liquids with gap-less (Fermi liquid) character are confirmed !!

AF +CO $(Et_2Me_2As)_{1-z}(Et_2Me_2Sb)_z[Pd(dmit)_2]_2$



# Spin liquid state in $Et_2Me_2As_xSb_{1-x}[Pd(dmit)_2]_2$

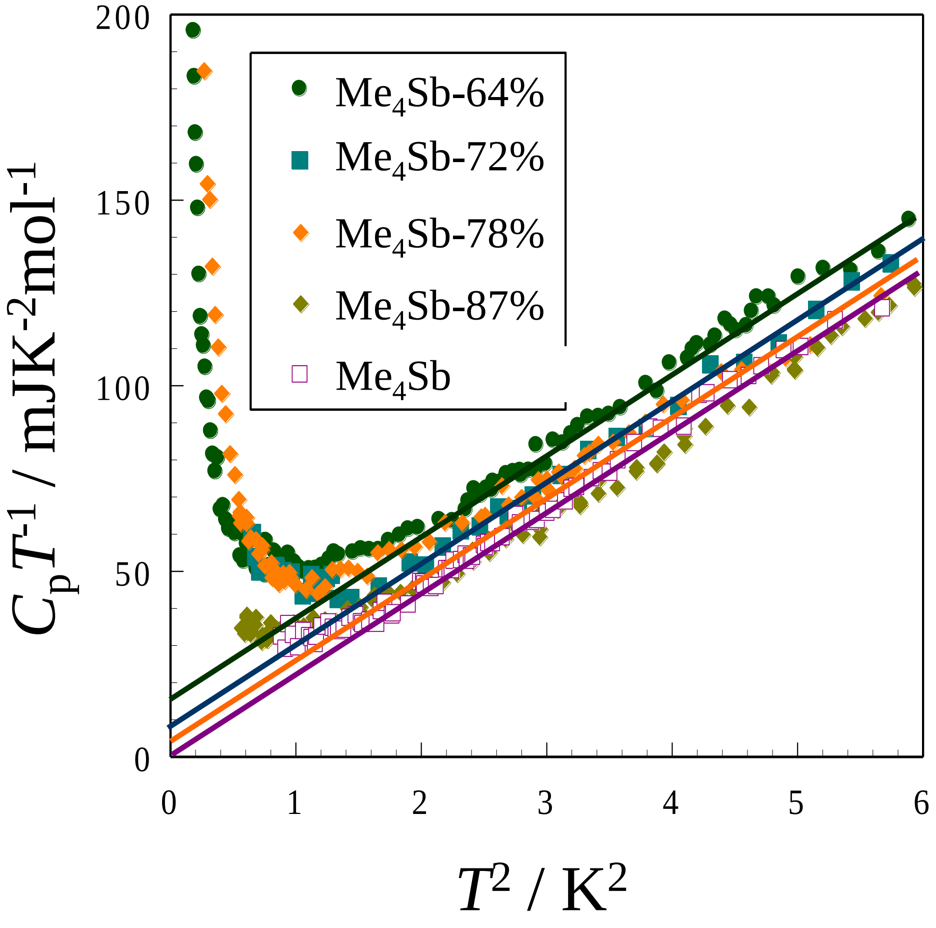


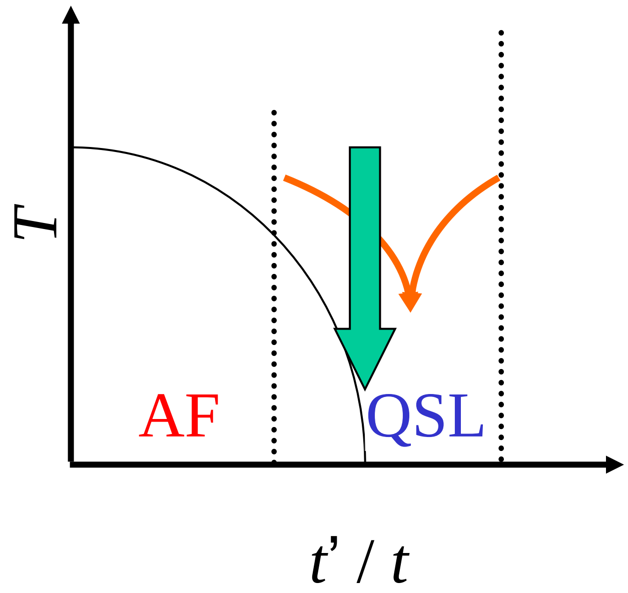




The behavior of $EtMe_3Sb[Pd(dmit)_2]_2$ is almost reproduced in $Et_2Me_2AsSb[Pd(dmit)_2]_2$ system.

- The *t*'/*t* is fine tuned by mixing cation without disorder.
- The Fermi liquid behavior with gap-less excitation is intrinsic character of QSL phase.

# Possibility of critical behavior



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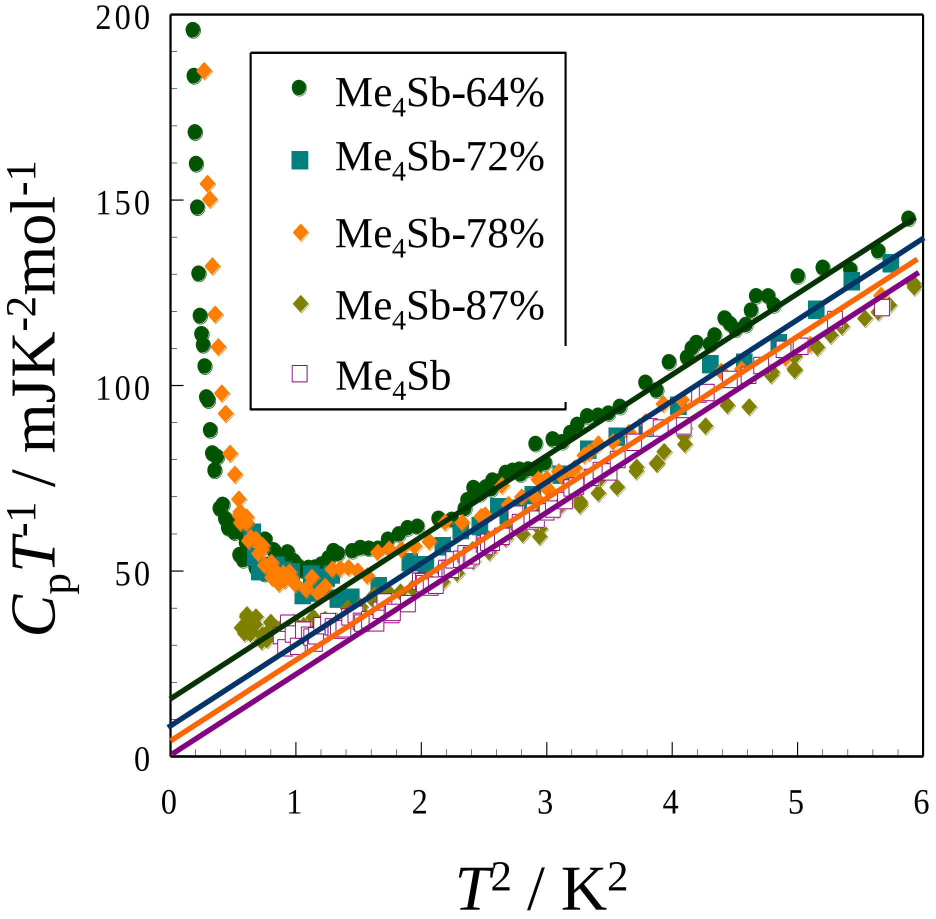


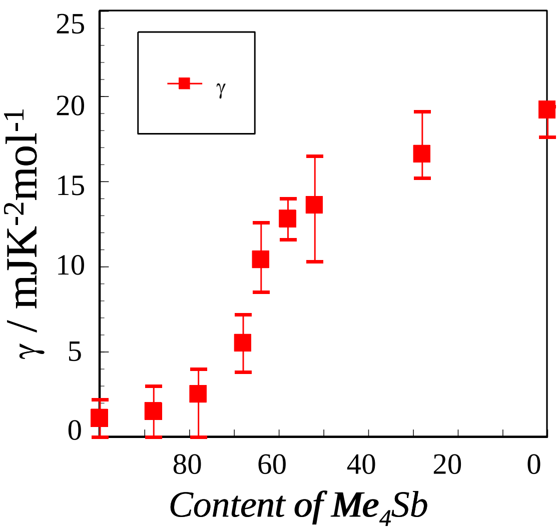

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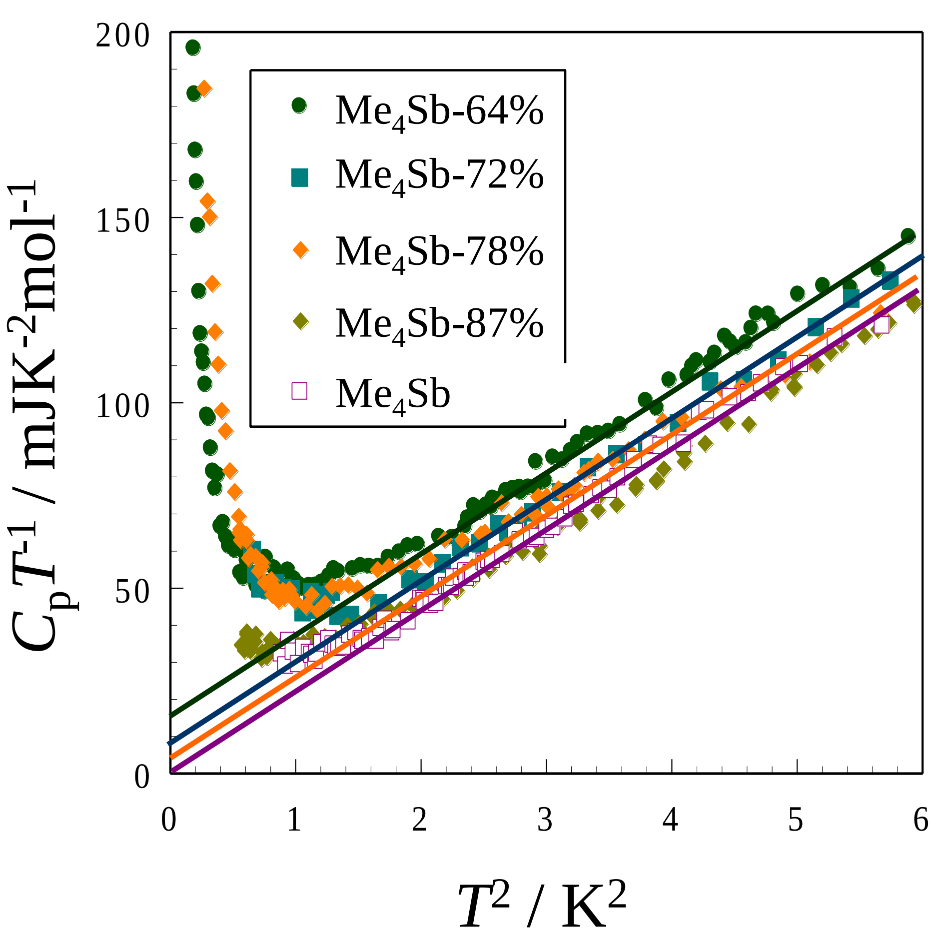
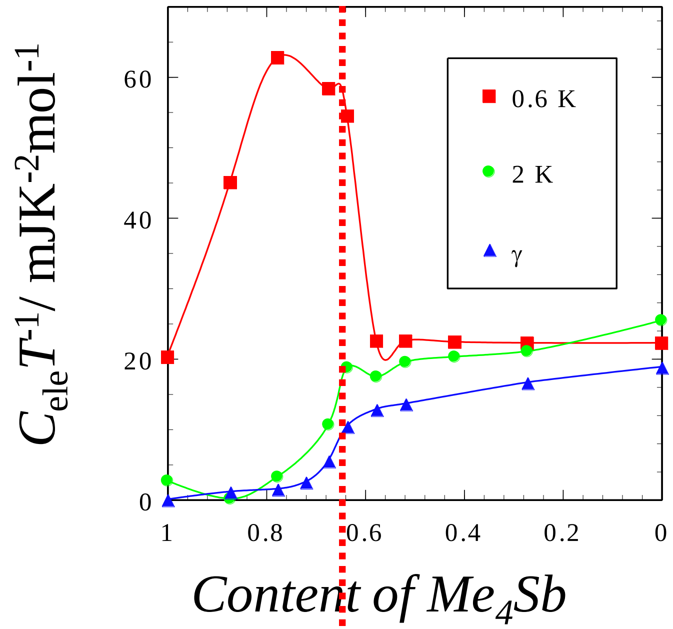
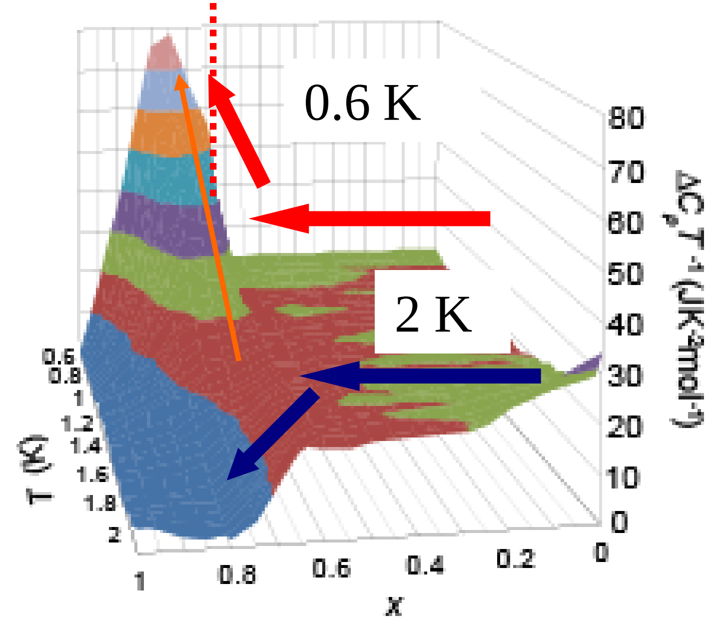
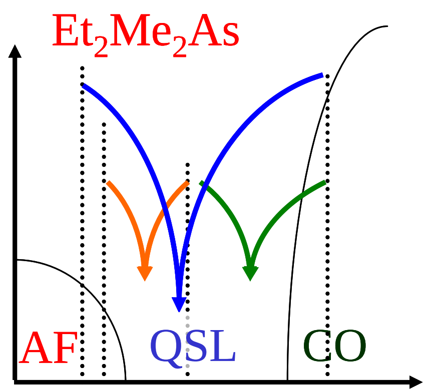



Possibility of critical behavior on phase boundary between AF and QSL.

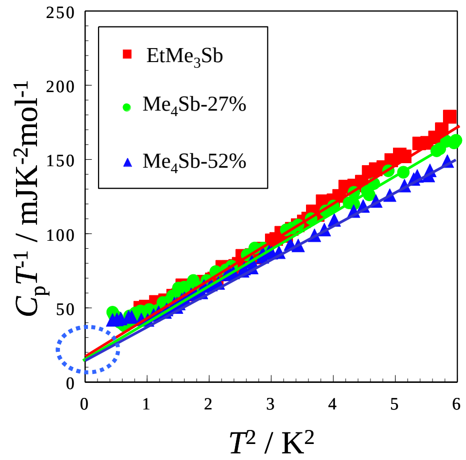
# Summary




We have measured three types of mixing cation systems $XY[Pd(dmit)_2]_2$.

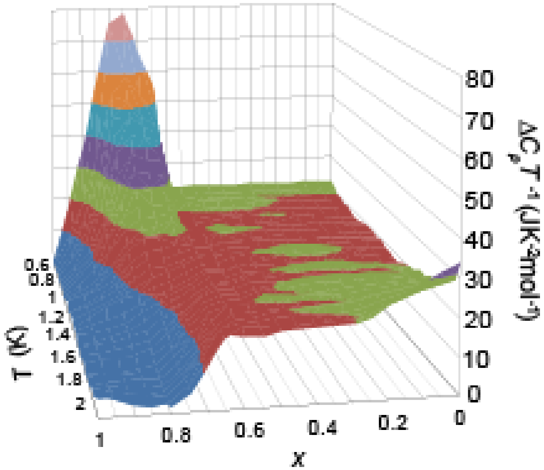
These three system show bulk spin liquid behavior with Fermi liquid character.

The t'/t is fine tuned without significant disorder.




The Fermi liquid character is intrinsic.

We also detected phase boundary between AF and QSL state.




The possibility of critical behavior on the phase boundary is also suggested.