Magnetic Quantum Phase Transitions in Coupled Spin Dimer Systems

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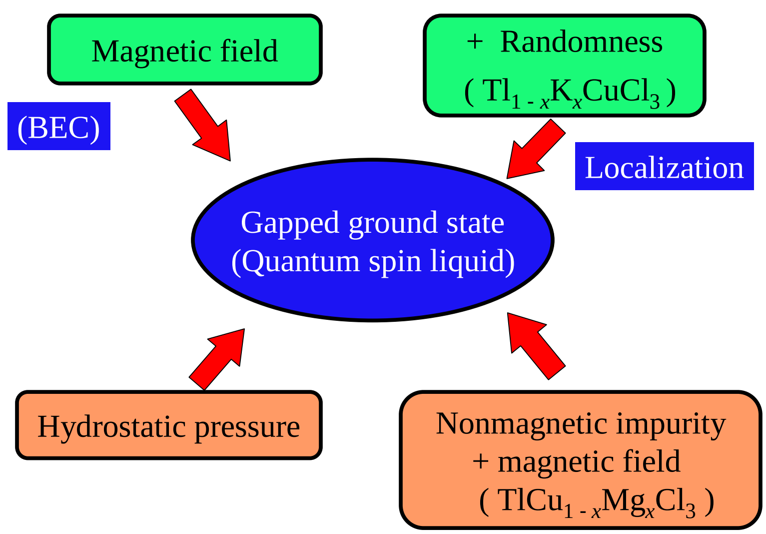
♦ $TlCuCl_3$, $KCuCl_3$, (NH_4CuCl_3)

- Magnetic insulator.
- S=1/2 Heisenberg system.
- 3D coupled spin dimer system.
- Singlet ground state with excitation gap.

♣ Quantum Phase Transitions



♣ Collaborators

- Sample preparation, magnetization, specific heat and ESR measurements:
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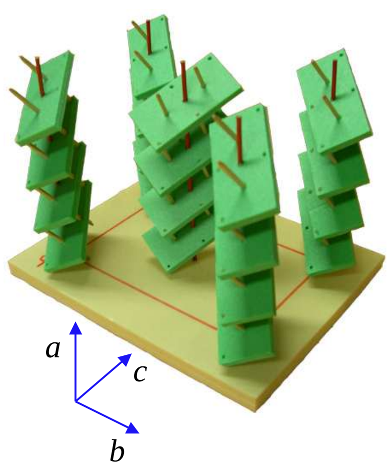
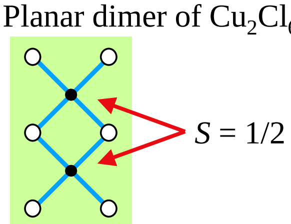
- Neutron scattering: K. Kakurai, A. Oosawa (JAERI), T. Kato (Chiba Univ.), A. Hoser, (HMI), Ch. Rüegg, M. Oettli, Ch. Niedermayer, A. Furrer (PSI)
- High field magnetization and ESR measurements: T. Goto, H. Mitamura (ISSP), K. Kindo, S. Kimura (Osaka), H. Nojiri, M. Motokawa (IMR), H. Ohta, S. Obubo (Kobe), V. N. Glazkov, A. I. Smirnov (Kapitza Inst.)
- NMR: M. Takigawa, O. Vyaselev (ISSP), Y. Shimaoka, T. Goto (Kyoto), T. Goto (Sophia)
- Light scattering and ultrasonic measurements: P. Lemmens (MPI), K. -Y. Choi (Aachen), B. Busse (Braunschweig), S. Schmidt, S. Zherlitsyn, B. Wolf, B. Lüthi (Frankfurt)
- Heat conductivity: K. Kudo, Y. Koike (Tohoku)
- Theory: M. Oshikawa, T. Nikuni (Tokyo Tech), E. Ya. Sherman (Graz), M. Müller, H. -J. Mikeska, A. K. Kolezhuk (Hannover)

♣ Crystal Structure of $ACuCl_3$ (A=Tl, K, NH_4)



Planar dimer of Cu_2Cl_6



AF dimer exchange

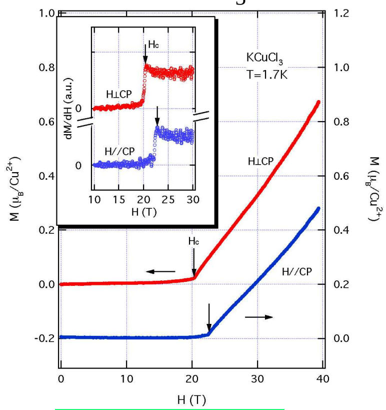
J = 5.68 meV for $TlCuCl_3$
4.34 meV for $KCuCl_3$

$J_A \approx$ 0.3 meV, $J_B \approx$ 1.8 meV,
$J_C \approx$ 3.0 meV for NH_4CuCl_3

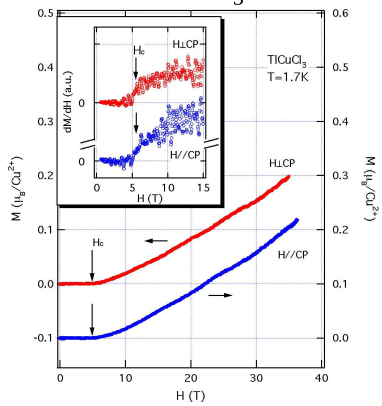
Willett *et al*.: J. Chem. Phys. **38** (1963) 2429
Takatsu *et al*.: JPSJ **66** (1997) 1611

♣ Magnetization Curves of $KCuCl_3$ and $TlCuCl_3$

$KCuCl_3$



$TlCuCl_3$

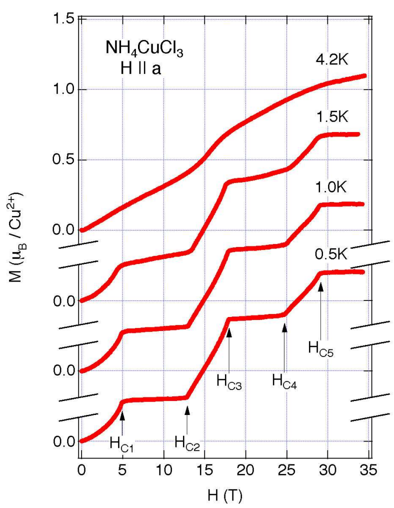


$(g/2)H_c = 23.1$ T

$(g/2)H_c = 5.6$ T

Shiramura *et al*.: JPSJ **66** (1997) 1900

♣ Magnetization Curve of NH_4CuCl_3

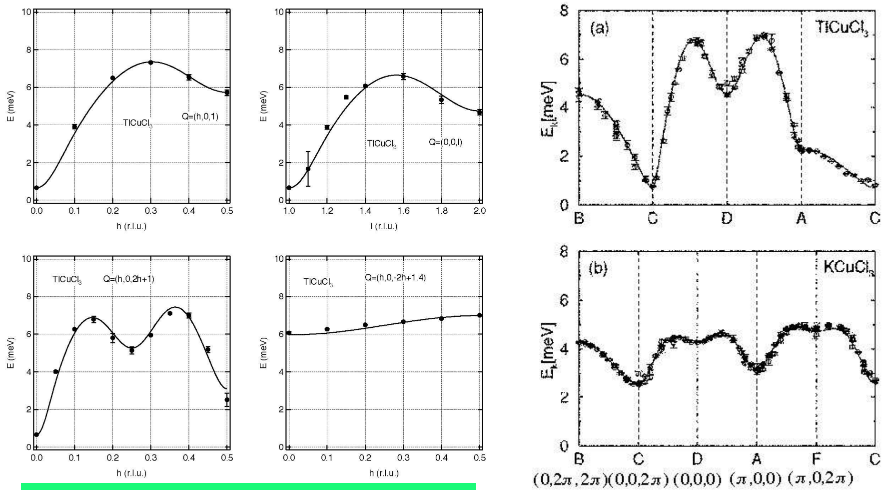


- Magnetization plateaus at $M_s/4$ and $3M_s/4$.
- Chemical unit cell is enlarged twice along the *b*-axis below 70 K.

Shiramura *et al*.: JPSJ **67** (1998) 1548

Rüegg *et al*. PRL in production

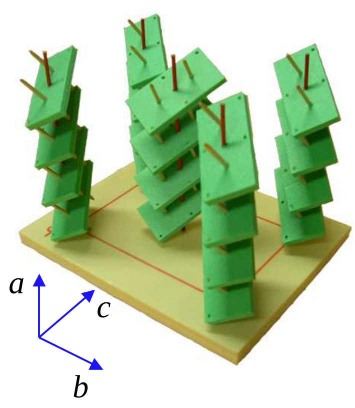
♣ Magnetic Excitations in $TlCuCl_3$ ($KCuCl_3$)

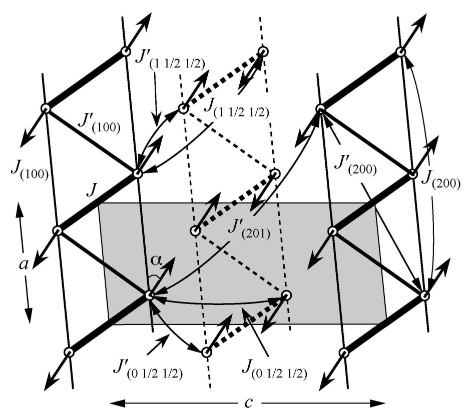


• Lowest excitation at $\boldsymbol{Q}$ = (0,0,1).

T. Kato *et al.*:JPSJ **67** (1998) 752, N. Cavadini *et al.*: Eur. Phys. J. B **7** (1999) 519
N. Cavadini *et al.*: PRB **63** (2001) 172414, Oosawa *et al.*: PRB. **65** (2002) 094426

♣ Exchange Network in $TlCuCl_3$



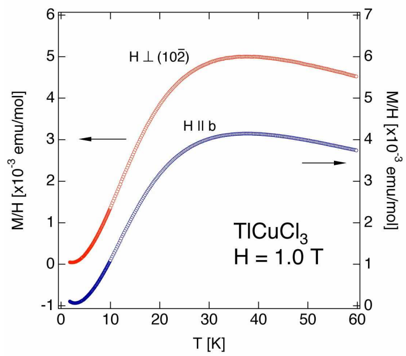


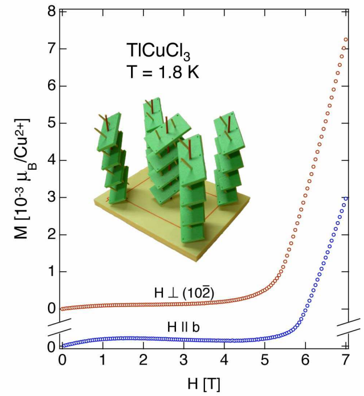
• 3D coupled Heisenberg spin dimer system.

$J = 5.68$ meV $J_{(100)} = 0.34$ meV $J'_{(100)} = 1.70$ meV

$J_{(1\frac{1}{2}\frac{1}{2})} = 0.91$ meV $J'_{(1\frac{1}{2}\frac{1}{2})} = -0.57$ meV $J'_{(201)} = 2.56$ meV

♣ Magnetic Properties of $TlCuCl_3$



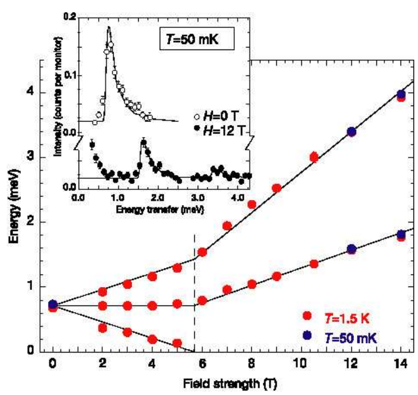


- Singlet ground state with an excitation gap Δ = 0.65 meV.

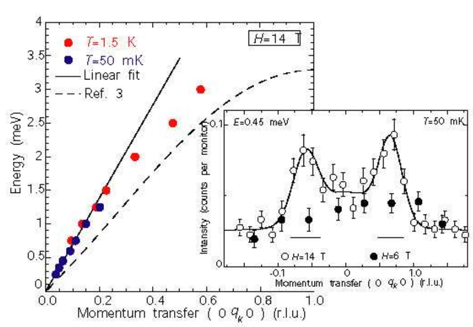
Oosawa *et al.*:J. Phys.: Condens. Matter **11** (1999) 265

♣ Field-Induced Quantum Phase Transition

♦ Field dependence of magnetic excitations



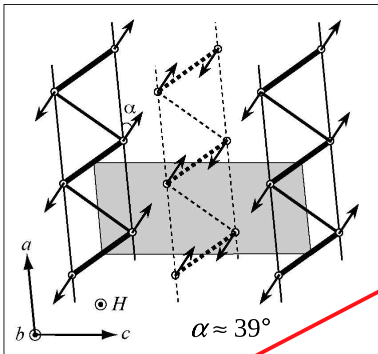
♦ Low-lying magnetic excitations

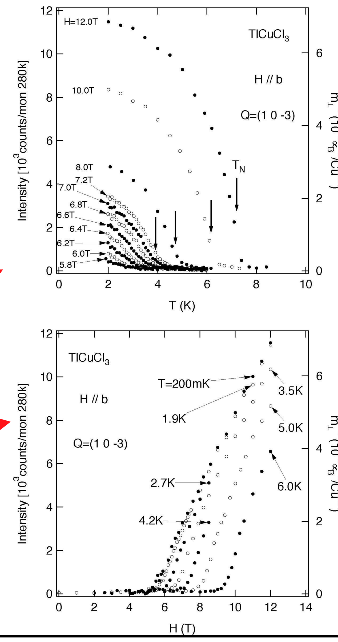


Rüegg *et al.*: Nature **423** (2003) 62
Matsumoto *et al.*: PRL **89** (2002) 077203, PRB **69** (2004) 054423

♣ Neutron Scattering in High Magnetic Field

♦ Transverse Néel ordering

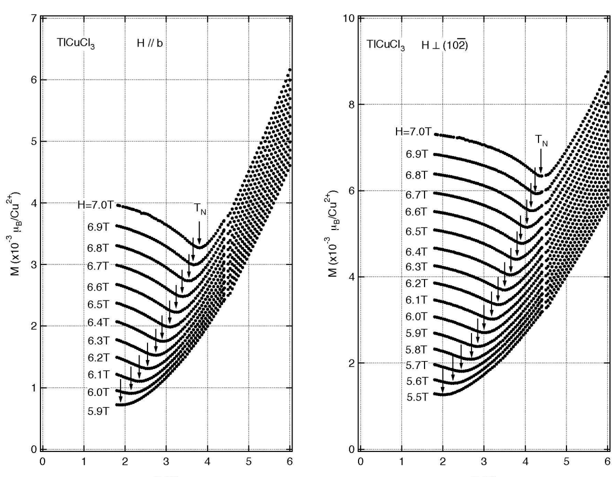




♦ Temerature and field dependence of Bragg peak intensities.

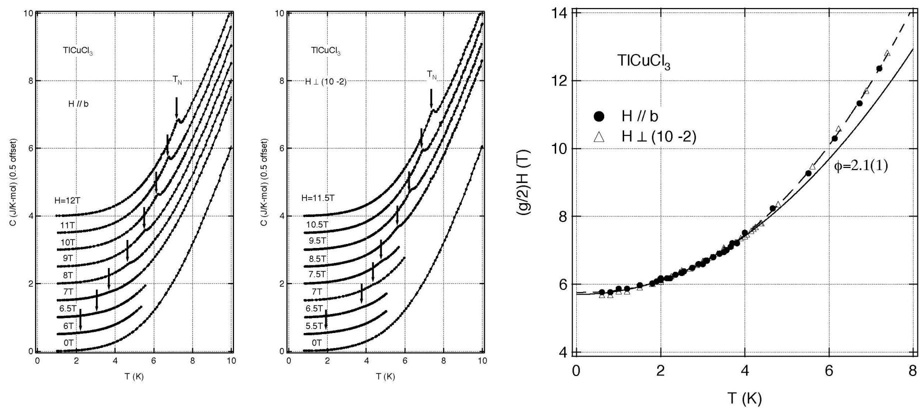
Tanaka *et al.*: JPSJ **70** (2001) 939

♣ Low Temperature Magnetizations in $TlCuCl_3$



• Cusplike minimum at T_N.

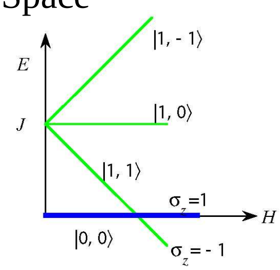
♣ Phase Boundary

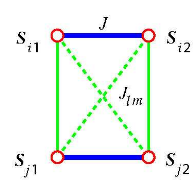


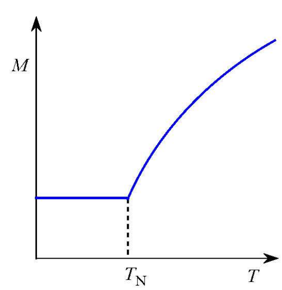
• Power law: [$H_N(T) - H_N(0)$] ∝ T^{ϕ} . $\phi \approx 2.0$)

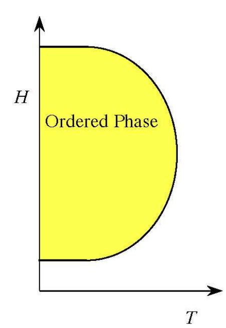
♣ Molecular Field Theory in the Real Space

Tachiki & Yamada: JPSJ **28** (1970) 1413

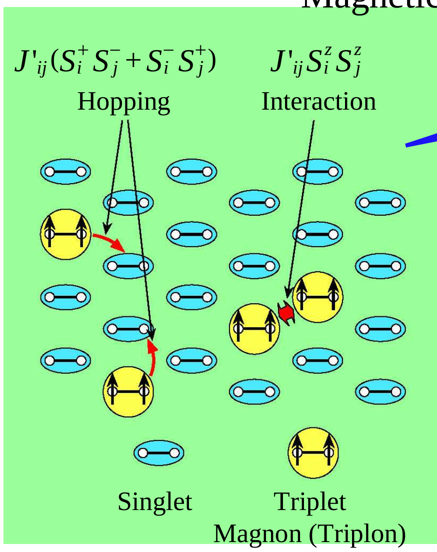








♣ Field-Induced Magnetic Ordering



System of interacting bosons

When hopping is dominant, phase transition is Bose-Einstein condensation.

Transverse magnetic ordering.

When interaction is dominant, Wigner crystal of spin triplets.

→ $SrCu_2(BO_3)_2$

Giamarchi & Tsvelik: PRB **59** (1999) 11398,
Nikuni *et al.*: PRL **84** (2000) 5868, Rice: Science **298** (2002) 760.

♣ Model

Nikuni *et al.*: PRL **84** (2000) 5868

$$\mathcal{H} = \sum_i (J - H) a_i^\dagger a_i + \sum_{i,j} t_{ij} a_i^\dagger a_j + \frac{1}{2}\sum_{i,j} U_{ij} a_i^\dagger a_j^\dagger a_j a_i$$

$$n << 1 \quad (H \sim H_g)$$

$$\mathcal{H} = \sum_{k} \left(\varepsilon_k - \mu\right) a_k^\dagger a_k + \frac{U}{2N}\sum_{k,k',q} a_{k+q}^\dagger a_{k'-q}^\dagger a_k a_{k'}$$

with $\varepsilon_k = \dfrac{\hbar^2 k^2}{2m}$ (quadratic dispersion)

- ♦ Relations between magnetic and particle quantities

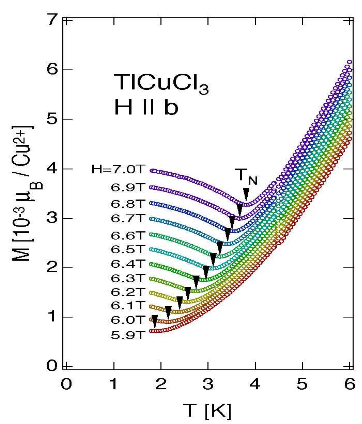
  $\mu \Leftrightarrow H \qquad \mu = g\mu_B (H - H_g)$

  $N \Leftrightarrow M \qquad M = g\mu_B N$

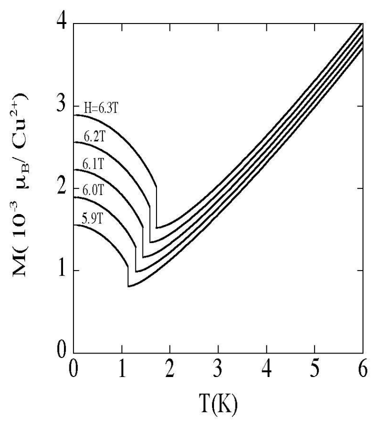
  Compressibility: $\dfrac{\partial n}{\partial \mu}$ $\Leftrightarrow$ Susceptibility: $\dfrac{\partial M}{\partial H}$

- ♦ Hartree-Fock-Popov approximation

♣ Low Temperature Magnetization



• Experiment



• Calculation

cf. BEC theory with Dzyaloshinsky-Moriya term $\boldsymbol{D}\cdot[\boldsymbol{S}_i \times \boldsymbol{S}_j]$

Sirker *et al.*: cond-mat/0403311

♣ Phase boundary

- $\mu_{\mathrm{c}} = 2U\left(\dfrac{mk_{\mathrm{B}}T}{2\pi\hbar^2}\right)^{3/2}\zeta(3/2) \;\Longrightarrow\; H_{\mathrm{N}}(T) - H_{\mathrm{N}}(0) \propto T^{3/2}$

- Experiment: [$H_{\mathrm{N}}(T)$ - $H_{\mathrm{N}}(0)$] $\propto T^{\phi}$. ϕ = 1.7~ 2.2)

. The value of .ϕ becomes smaller with decreasing fitting window.

□□□□□ theory

- Relativistic dispersion of the form $\varepsilon_{\boldsymbol{k}} = \sqrt{\Delta^2 + Ak^2}$

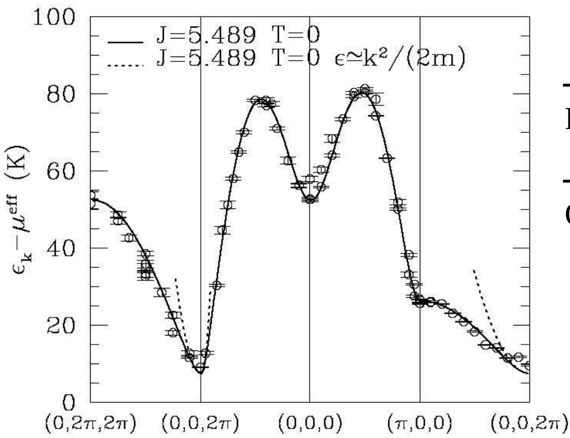
  Sherman *et al.*: PRL **91** (2003) 057201

- Quantum Monte Carlo on cubic dimer lattice

  Nohadani *et al.*: PRB **69** (2004) 220402R

♣ Hartree-Fock Calculation with Realistic Dispersion

Misguich & Oshikawa: cond-mat/0405422

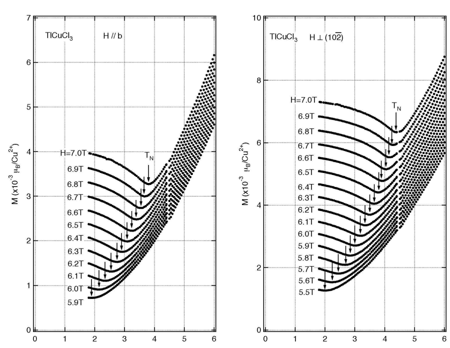


Realistic dispersion (solid line)

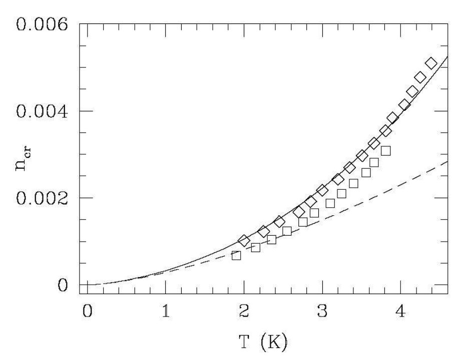
Quadratic dispersion (dashed line)

♣ Critical density n_{cr} as a function of temperature

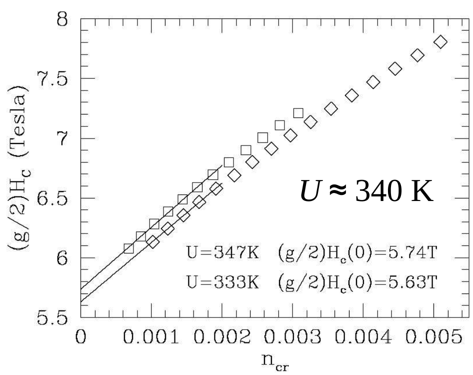
- Experiment

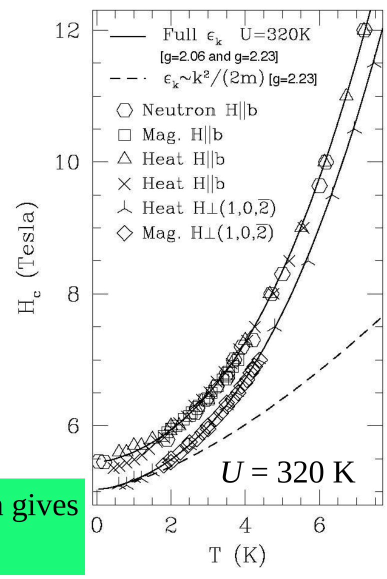


- Calculation with realistic dispersion (solid line)



♣ Critical field $H_N(T)$ versus n_{cr} and phase boundary

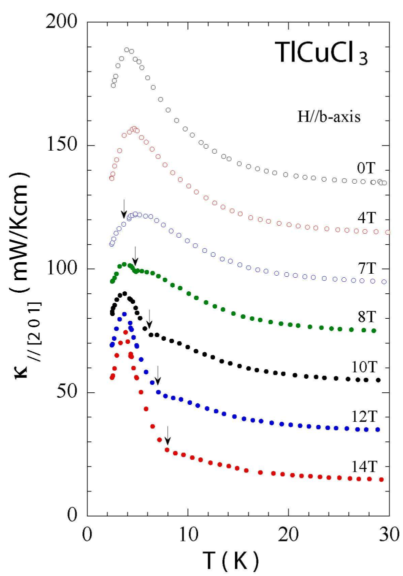




- $(g/2)[\ H_N(T) - H_N(0)\ ] = 2Un_{cr}(T)$

- BEC theory with realistic dispersion gives very good description *!*

♣ Thermal Conductivity κ

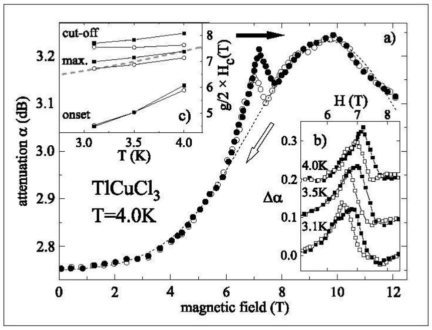


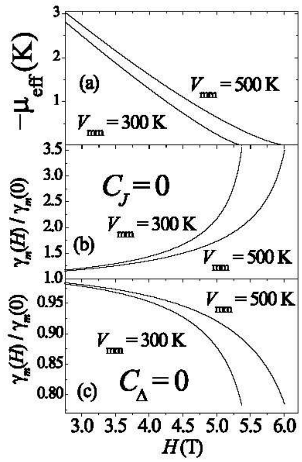
- Drastic enhancement of κ in the ordered state.

- Drastic enhancement of the mean free path of magnons in the ordered state.

Kudo *et al*.: cond-mat/0312063,
J. Magn. & Magn. Mater. In press

♣ Sound Attenuation





- Sound attenuation increases with H and has a sharp peak at $H_N(T)$.

➡ Softening of the lowest excitation.

Sherman *et al.*: PRL **91** (2003) 057201

♣ Localization of Spin Triplets in $Tl_{1-x}K_xCuCl_3$ ($x < 0.3$)

Partial K^+ substitution may produces
(1) Chemical pressure and (2) exchange randomness.

$$\mathscr{H} = \sum_i (J_i - H) a_i^\dagger a_i + \sum_{i,j} t_{ij} a_i^\dagger a_j + \frac{1}{2}\sum_{i,j} U_{ij} a_i^\dagger a_j^\dagger a_j a_i$$

Randomness in intradimer coupling J ➡ Randomness in local potential.

J = 5.68 meV for $TlCuCl_3$
4.34 meV for $KCuCl_3$.

Randomness in interdimer coupling. ➡ Randomness in hopping amplitude and interaction.

♣ Bose Lattice Gas in the Presence of Random Potential

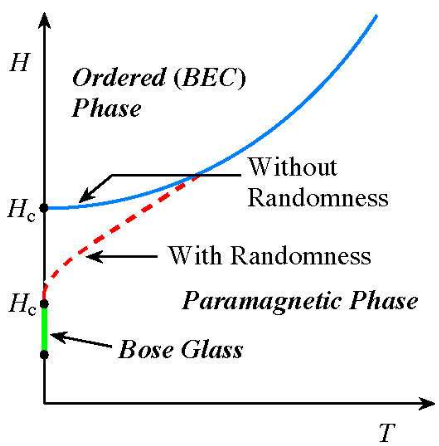
Fisher *et al.*: PRB **40** (1989) 546

• Soft core and on-site interaction.

• Bose glass phase at $T = 0$.
Bosons localize (insulator), no gap, finite compressibility.
• New critical behavior.
Bose glass-superfluid transition.

$T_c \sim [n_s(0)]^x$ and $n_s(0) \sim (n - n_c)^\zeta$
with $x = 2/3$, $\zeta = 1$ for the pure system,
$x = 3/4$, $\zeta \geq 8/3$ for the disordered system.

♣ Phase Diagram Expected for $Tl_{1-x}K_xCuCl_3$



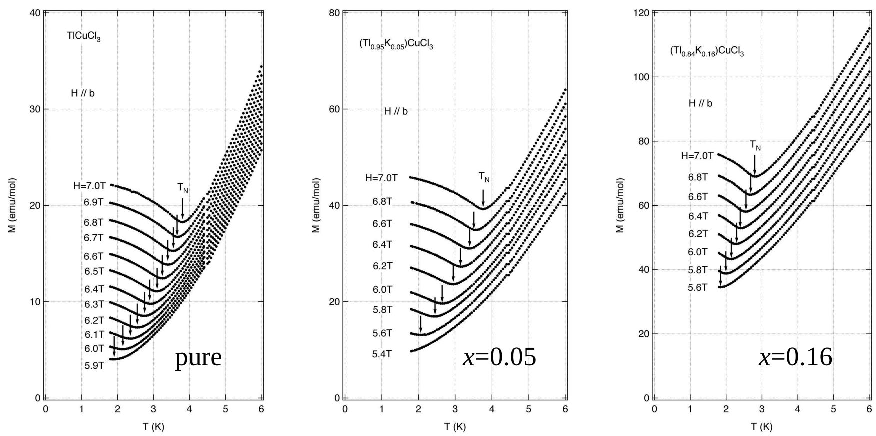
Superfluid phase
➡ Ordered phase.

Mott insulating phase
➡ Gapped singlet phase.

Bose glass phase of spin triplets.

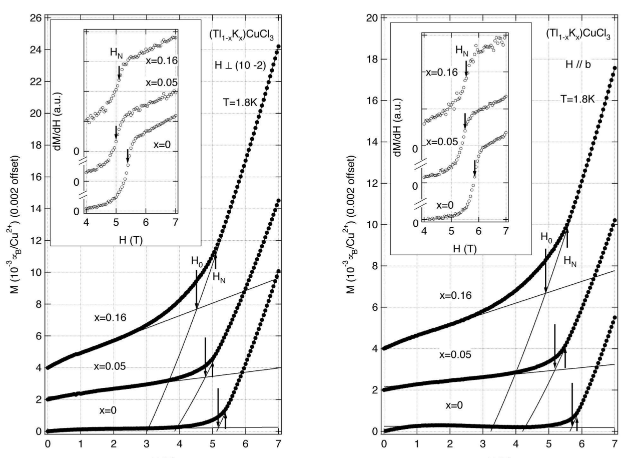
♣ □□□□□□□□□□□□□□□ Magnetization in $Tl_{1-x}K_xCuCl_3$

Oosawa & Tanaka: PRB **65** (2002) 184437



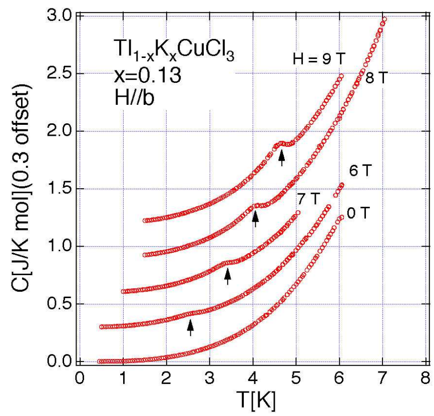
- Cusp like minimum at T_N as observed in $TlCuCl_3$.

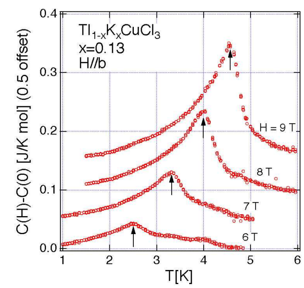
♣ □□□□□□□□□□□□□□□ Magnetization Curves in $Tl_{1-x}K$

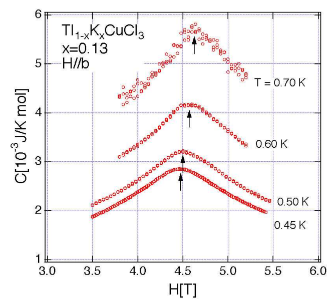


- Finite slope in magnetization curves for $x \neq 0$.
  ➡ No gap and finite compressibility.

♣ Specific Heat in $Tl_{1-x}K_xCuCl_3$



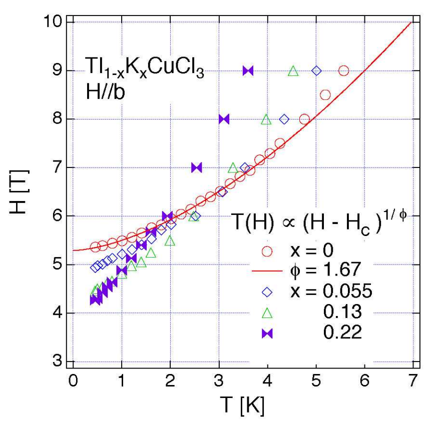


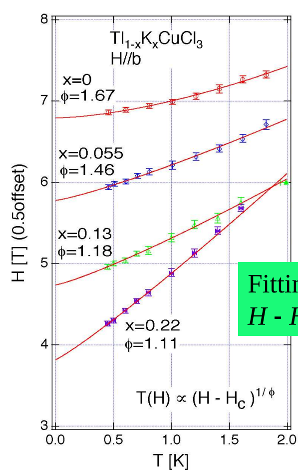


- Sharp anomaly
  $\Rightarrow$ good homogeneity.

Shindo & Tanaka: cond-mat/0310691

♣ Phase Boundaries





Fitting field range:
H - $H_N(0) < 1$ T

- Low temperature phase boundaries for $x\neq0$ are close to linear function of T, □□□ concave form was not observed down to 0.45 K.

♣ Summaries on $Tl_{1-x}K_xCuCl_3$

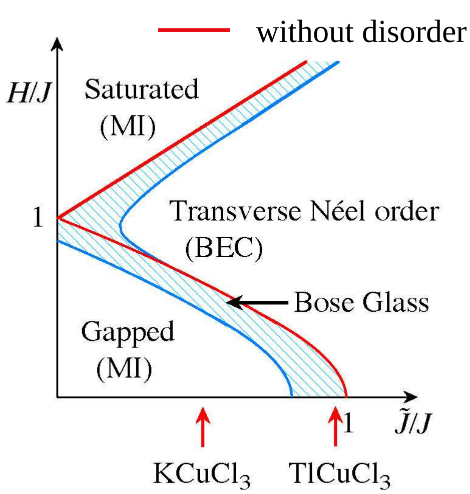
For $H > H_N(0)$
- $\phi \Rightarrow 1$ ➡ Different critical behavior and crossover from convex form to concave form.

For $H < H_N(0)$
- Finite susceptibility ➡ No gap and finite compressibility.
- Absence of long range order ➡ Localization of spin tri

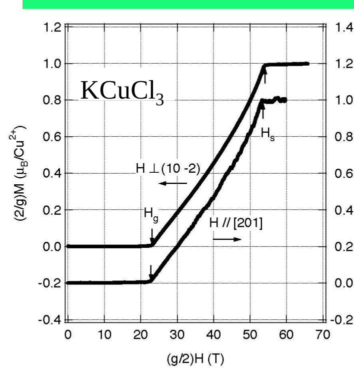
⬇

- Ground state is Bose glass phase of spin triplets.
- Gapped singlet (Mott insulating) phase disappears.

♣ Phase Diagram for $Tl_{1-x}K_xCuCl_3$ ($x < 0.3$)



- $TlCuCl_3$ is close to QCP.
- Small amount of disorder wipes out MI phase.



Oosawa *et al.*: PRB **66** (2002) 104405