

Spin Disorder

in a Bulk Triangular Antiferromagnet

Satoru Nakatsuji,

Y. Nambu (M2), K. Onuma (M1), Y. Maeno

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Department of Physics, Kyoto University



Content

- 2D Triangular AFM
and Geometrically Frustration
- Novel Spin Disordered State
on 2D Triangular Lattice in NiGa_2S_4
- Impurity Effects, Spin Size Effects
- Summary and Future Plans

Search for Novel Quantum Phases in Geometrically Frustrated Lattices

Geometrical Frustration suppresses Magnetic Order.

2D

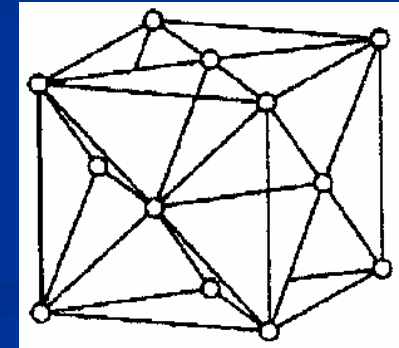
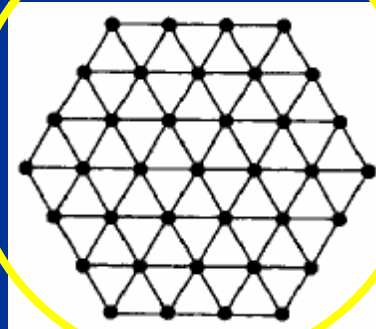
3D

Most basic & studied

Triangle

Face Centered Cubic

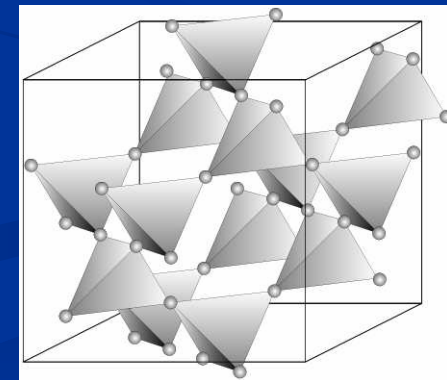
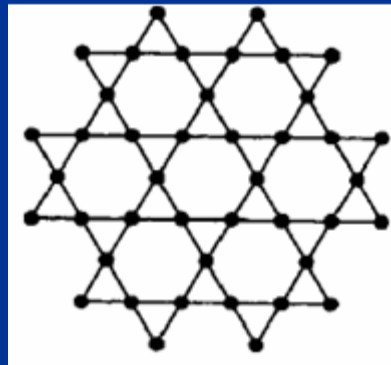
Edge Sharing



Kagomé

Pyrochlore

Corner Sharing

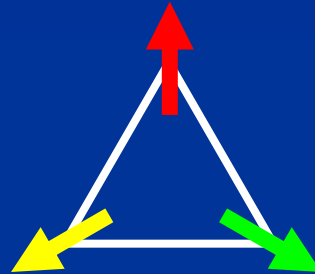


Triangular Lattice AFM

“the only geometrically frustrated 2D Bravais lattice”

- **Ising** (Wannier, Hushimi 1950) Disordered State with Residual Entropy.

- **Classical**



120° spin order, Chirality

- **Quantum** (Anderson 1973) Quantum Spin Liquid

- **XY** (Miyashita and Shiba 1984) Kosteritz-Thouless type transition, (Z_2) Vortex

Heisenberg (Kawamura and Miyashita 1984)

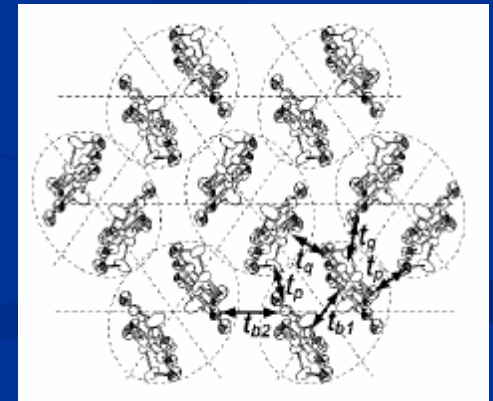
Novel Quantum Phases in 2D Triangular Lattice?

How about Experiments?

Triangular Lattice with Low spin ($S \leq 1$) Experiments

New Candidates from Japan!

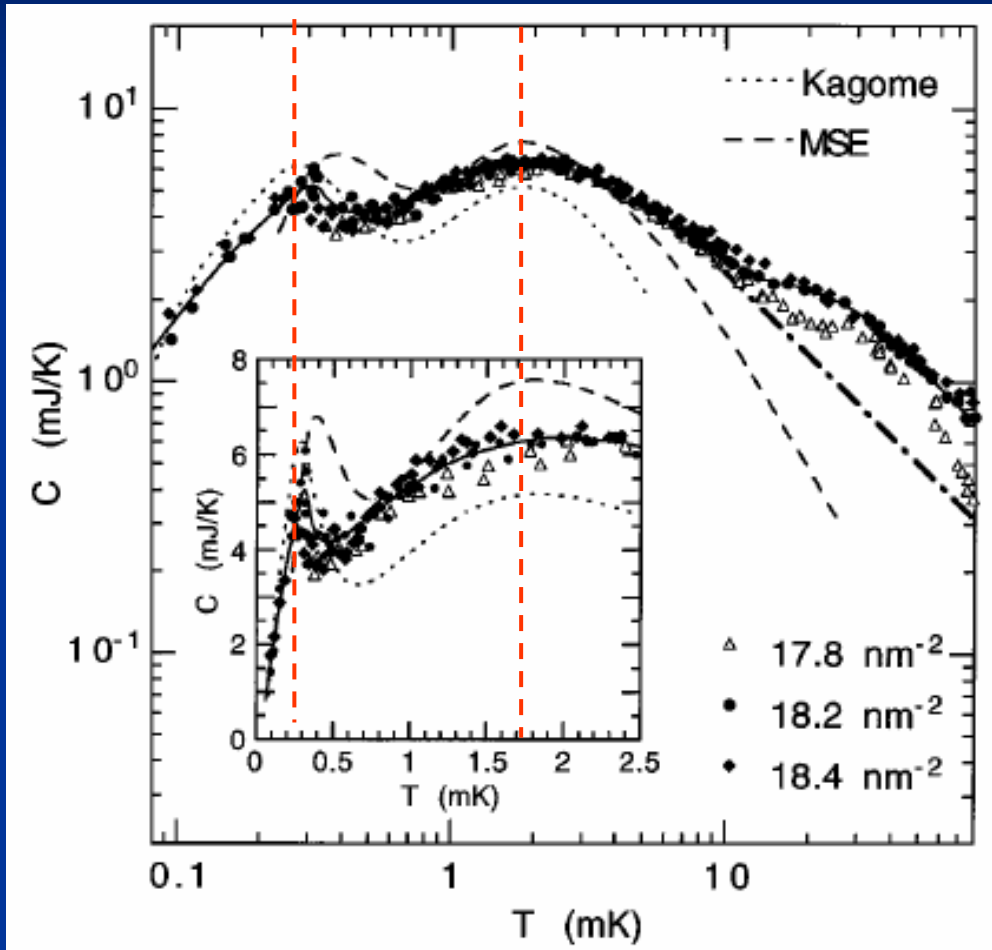
- ^3He thin film absorbed on carbon: O-19 Fukuyama
Gapless Spin Liquid, $S = 1/2$
(K. Ishida et al. PRL 79, 3451 (1997).
R. Matsutomi et al. PRL 92, 025301 (2004).)
- $\kappa\text{-(BETS-TTF)}_2\text{Cu}_2\text{CN}_3$: O-24 Kanoda
Distorted (Anisotropic) triangular lattice
Gapless Spin Liquid, $S = 1/2$
(Y. Shimizu et al. PRL 91, 107001 (2003).)



To our knowledge,
no low spin ($S \leq 1$) bulk system with exact triangular lattice

Signature of Geometrical Frustration

2D Triangular AFM of Solid ^3He



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

Double Peak

in Specific Heat

Higher T peak

$$T \sim \theta_W \sim O(J)$$

Short range order

Lower T peak

$$T \sim \theta_W/10$$

High Degeneracy

at $T \ll J$

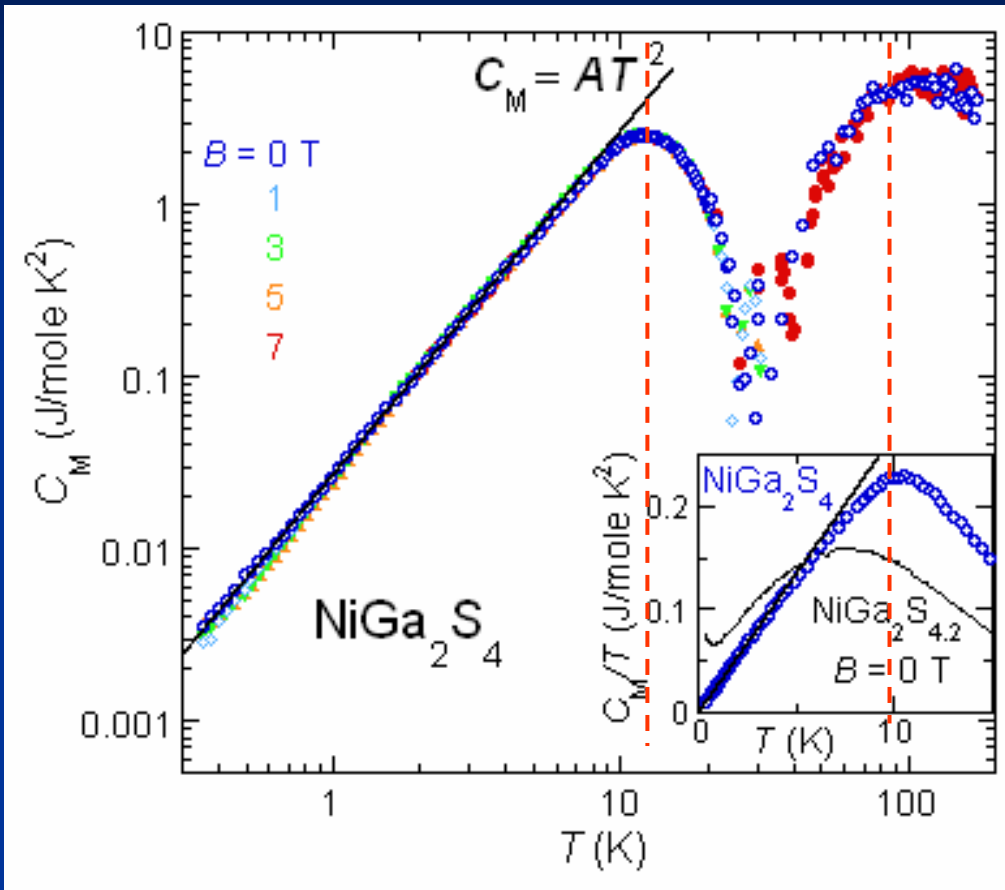
Formation of

Novel Disordered State

Gapless Spin Liquid?

Double Peak in Specific Heat

in NiGa_2S_4



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Formation of
Novel Disordered State?

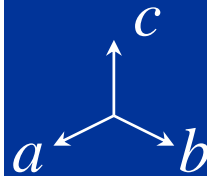
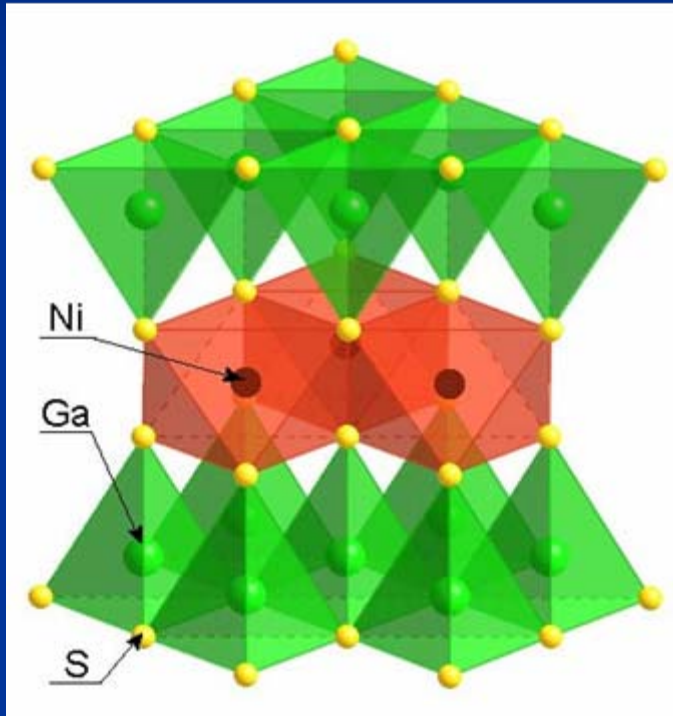
Bulk 2D Triangular AFM: NiGa₂S₄

$\overline{P3m1}$ symmetry above $T > 1.5$ K
No disorder (x-ray, neutron, ICP)

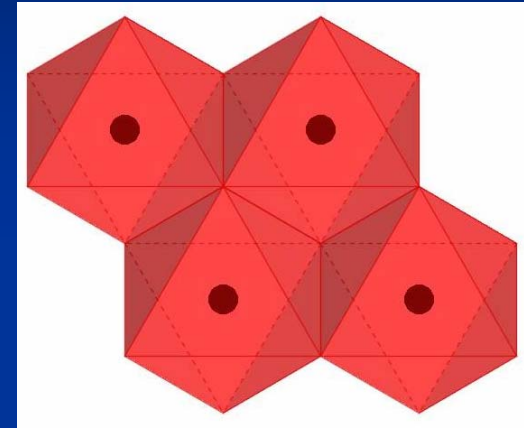
GaS

NiS₂

GaS



Mott Insulator



2D NiS₂ layer

c.f. 1. CoO₂ layer of Na_xCoO₂yH₂O
2. Mott transition in NiS

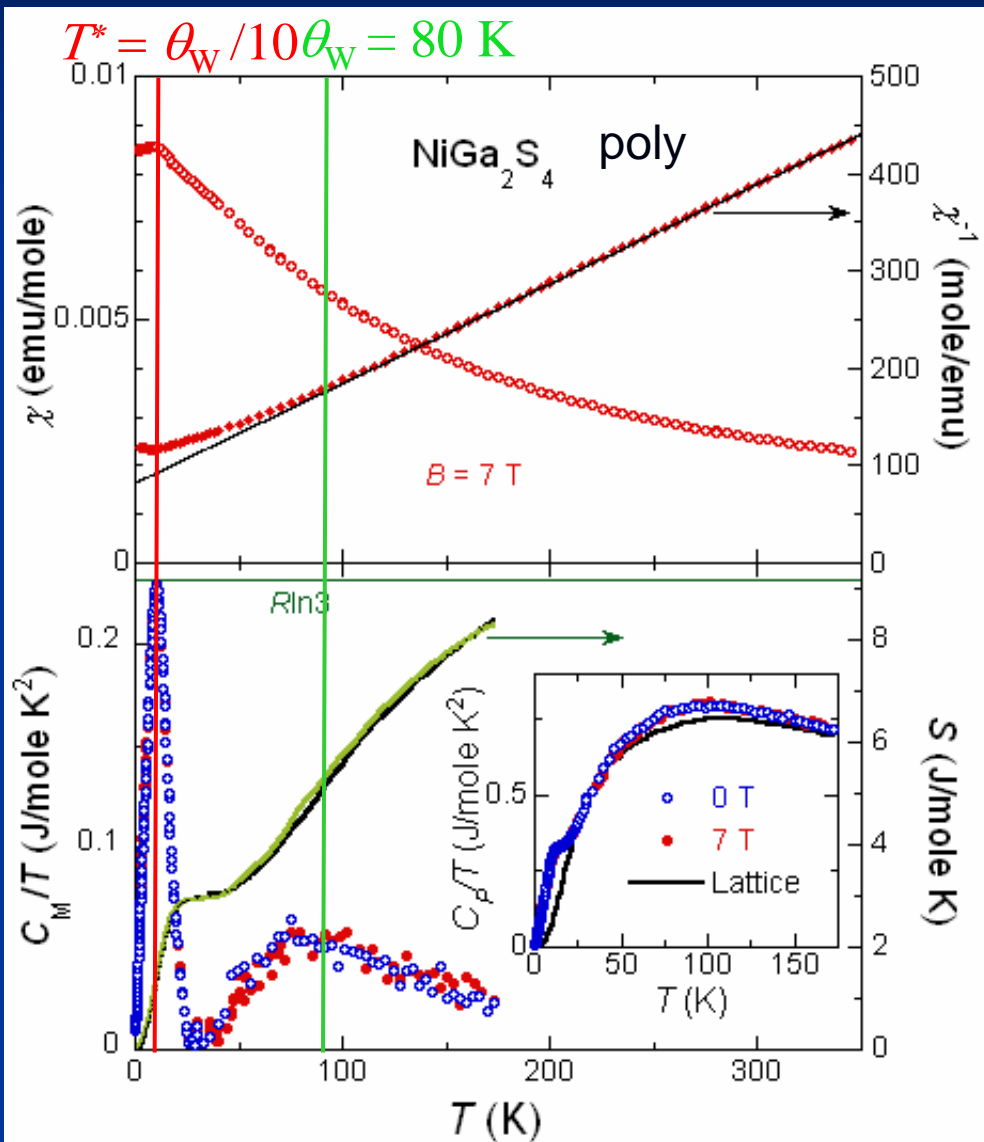


$S = 1$ Heisenberg spin

Ni distance $a = 3.67$ Å intra-layer
 $c = 11.99$ Å inter-layer

S. Nakatsuji, Y. Nambu, H. Tonomura, O. Sakai, S. Jonas, C. Broholm, H. Tsunetsugu, Y. Qiu and Y. Maeno, *Science* **309**, 1697 (2005).

No conventional AF order

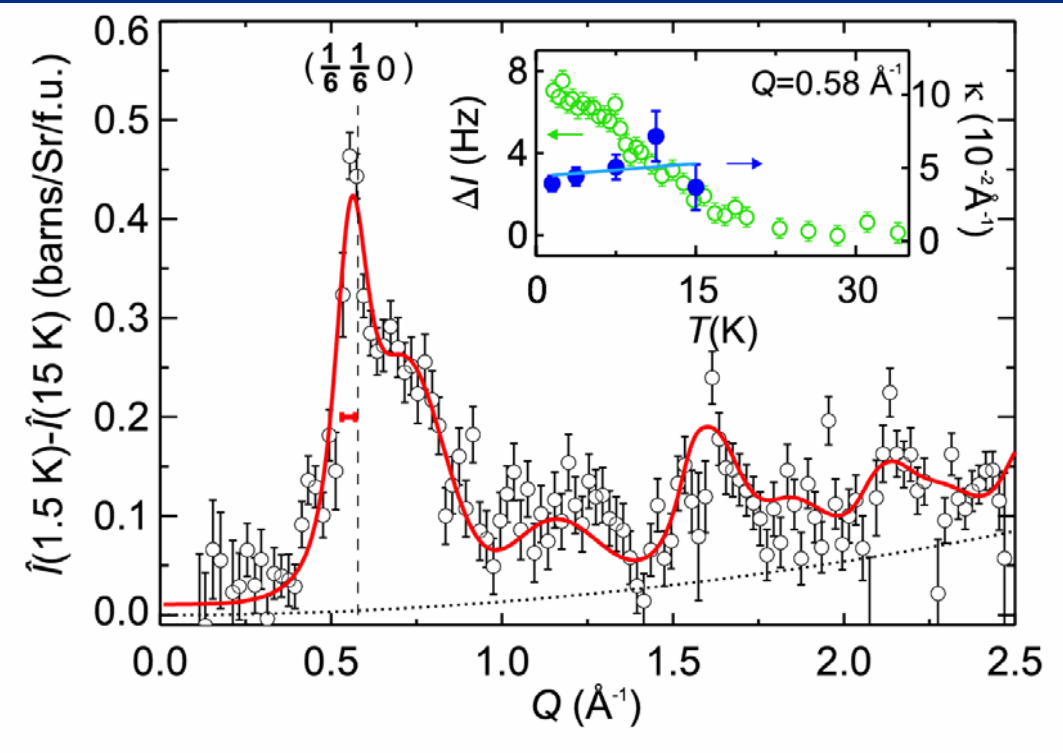


- $p_{\text{eff}} = 2.86 \mu_B$ ($S = 1$, $2.84 \mu_B$)
 $\theta_W = -80 \text{ K}$ (AF)
- No anomaly except a weak peak around 10 K
- No evidence of LRO in C_P
Spins remain disordered.
- Two crossover temperatures:
 1. $T \sim \theta_W$: short range order
 2. $T \sim \theta_W/10$: novel phase formation?

Nano-scale short-range order

Collin Broholm, Seth Jonas @ Johns Hopkins, USA

Magnetic neutron scattering at 1.5 K



No resolution limited peaks

Nearly no correlation between the planes: Highly 2D magnet

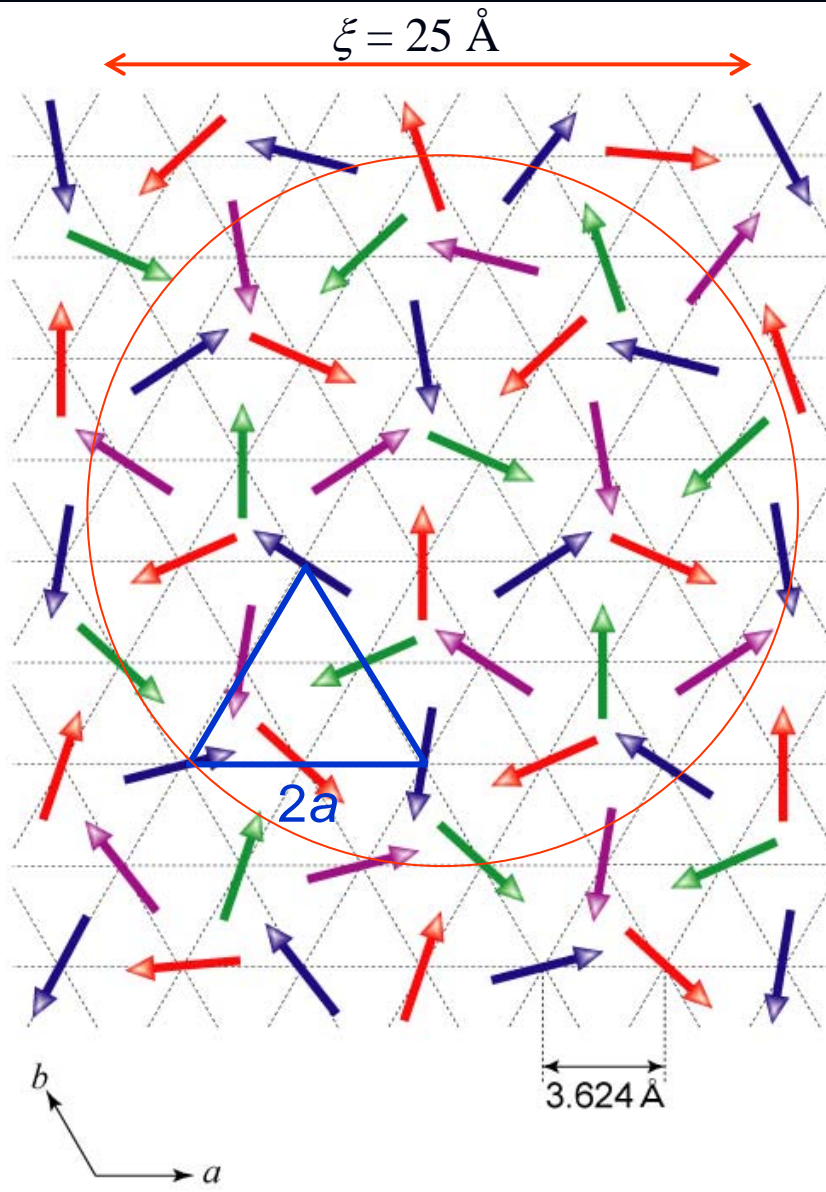
Development of the **incommensurate correlation near $(\frac{1}{6}, \frac{1}{6}, 0)$** below 20 K, corresponding to C_M/T peak.



Normally, 120° structure with $(\frac{1}{3}, \frac{1}{3}, 0)$ correlation.

Nano scale correlation
short: $\xi = 1/\kappa \sim 2.5 \text{ nm}$
slow: $\tau > 10^{-9} \text{ sec}$

$(1/6, 1/6, 0)$ correlation

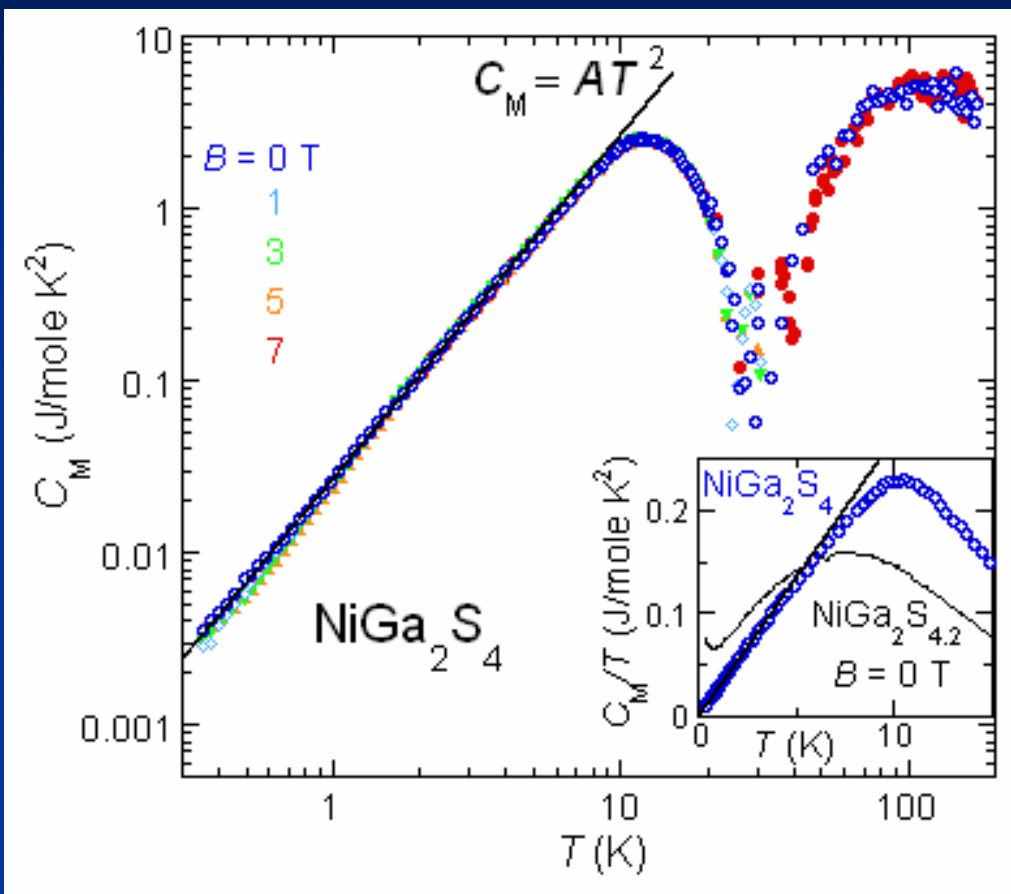


Twice larger unit cell
in comparison with $(1/3, 1/3, 0)$

Possible origins

- Dominant 3^{rd} nearest neighbor interactions
- 4 sublattices of 120° order with $2a$ periodicity
- Multiple spin exchange due to proximity to Metal-Insulator transition

Coherence without magnetic order



• Power Law Behavior

$$C_M \propto T^\alpha \quad \alpha = 2.001(5)$$

at $0.35 \text{ K} < T < 4 \text{ K}$

$$C_M/T \rightarrow 0.0(1)$$

mJ/mole K²

Similar to 2D AFM LRO

Novel Gapless Linearly Dispersive Mode in 2D

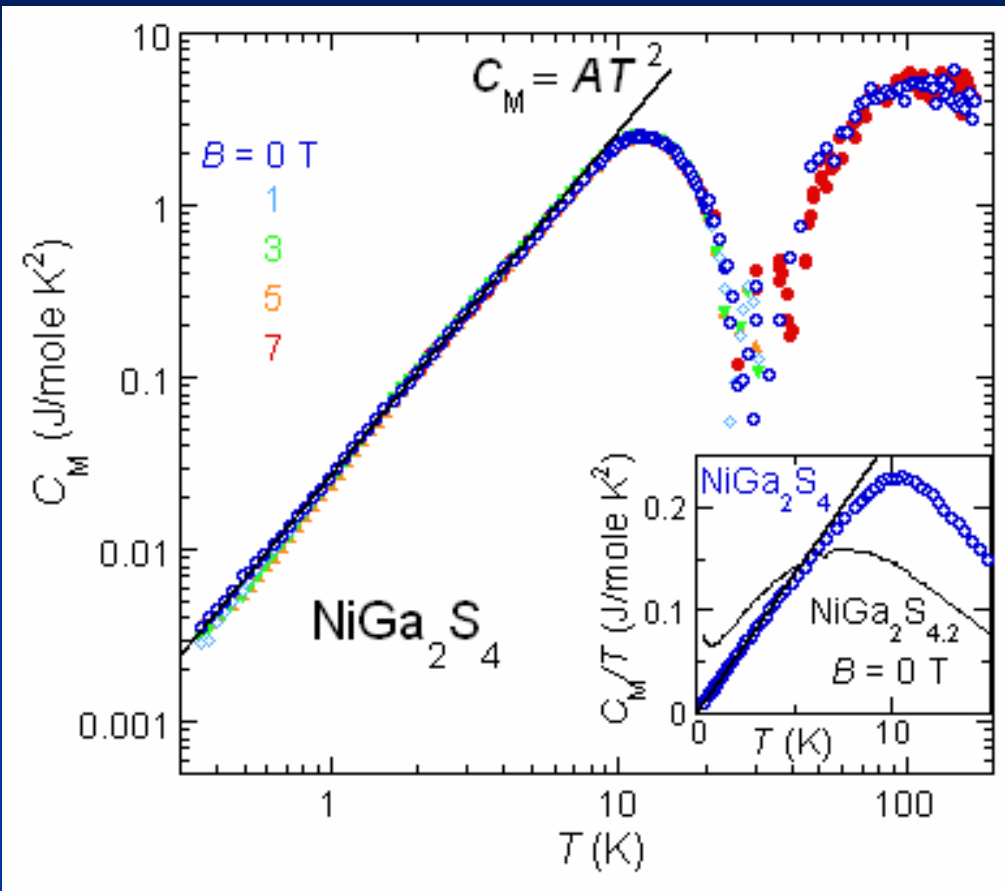
Coherence Length

> 100 nm

• Fragile against Impurities

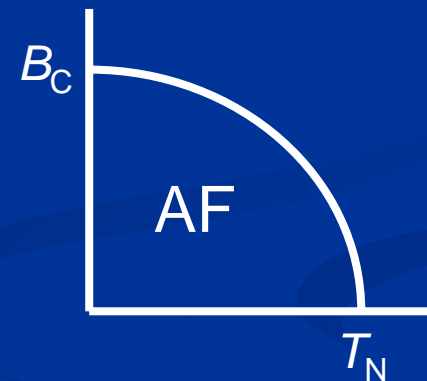
5% extra Sulfur doping \rightarrow Residual C_M/T (0 K)

Moment-Free Excitations



- **Insensitive to Field**

No response even to $B_c = k_B T_{\text{peak}} / \mu_B S \sim 7$ T



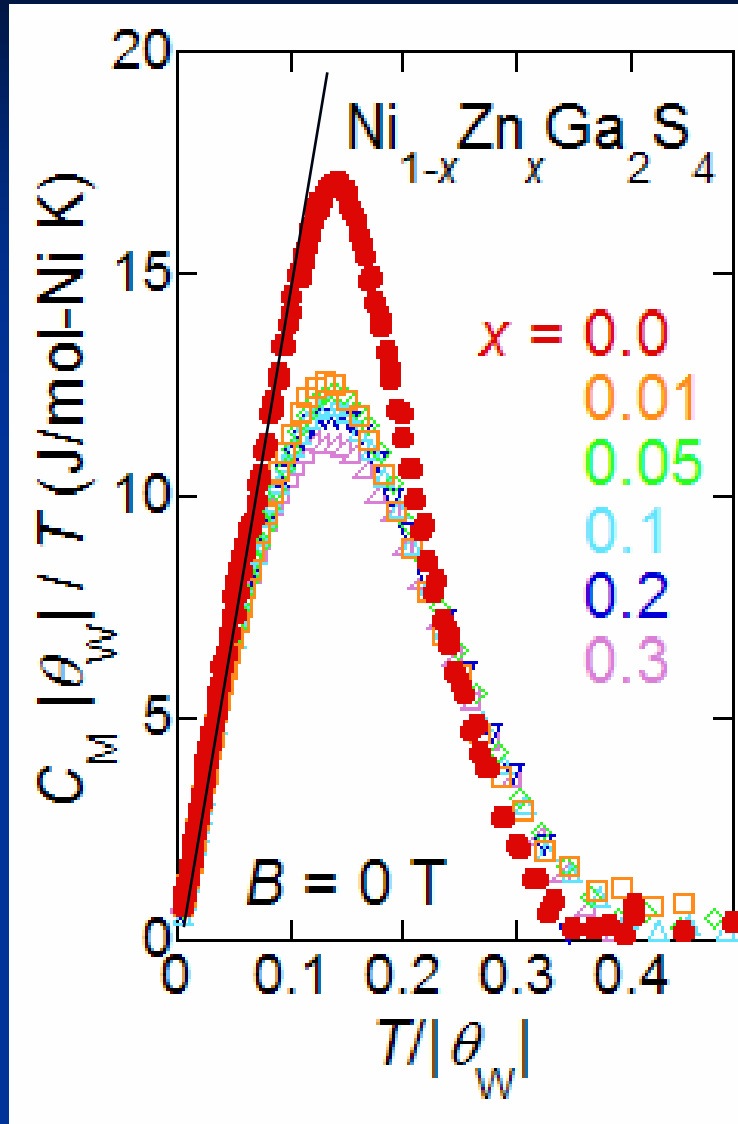
Spin excitation is
Not AF short range order

**But, Moment Free
(Spinless) Clusters**

Impurity Effects

➡ P-68 南部雄亮

Nonmagnetic Impurity (Zn: $S = 0$) Effect



Robust $C_M \sim T^2$ behavior

Scaling with θ_W

$$\rightarrow C_M \propto \frac{T^2}{|\theta_W|^2}$$

Nambu-Goldstone mode
in 2D AFM

↳ Symmetry Breaking

Summary

NiGa₂S₄: the first 2D triangular lattice with $S = 1$

- No AF order despite strong AF coupling
- Long-Range Coherence due to Symmetry Breaking
- Possibility of Novel Type of Magnetic Order
or Spin Liquid State