Magnetic Order and Spin Dynamics in $CdCu_2(BO_3)_2$ investigated by ^{11}B NMR and ZF-μSR

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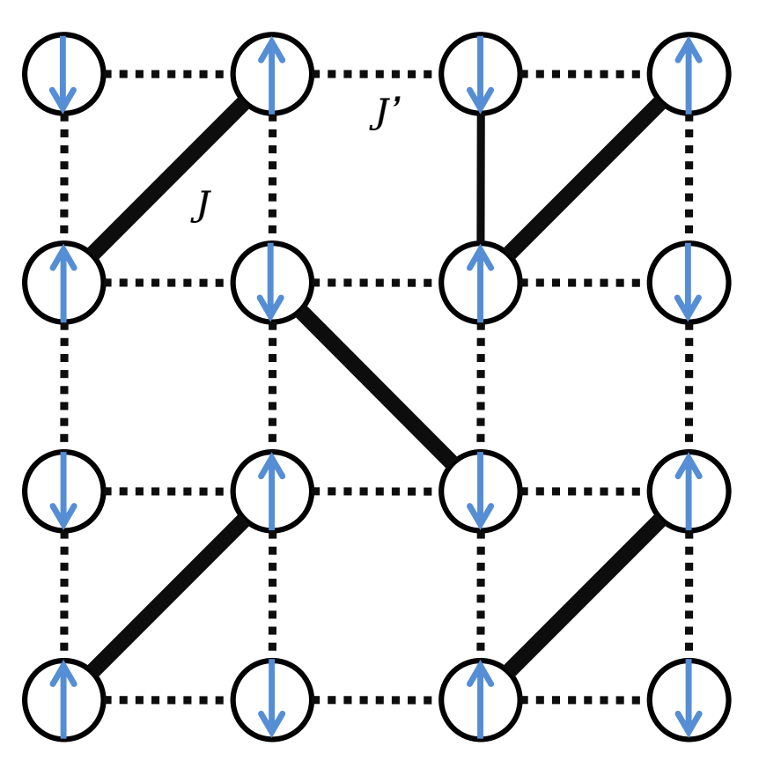
Shastry-Sutherland Lattice

$$\hat{H} = J\sum_{nn} S_i\cdot S_j + J'\sum_{nnn} S_i\cdot S_j$$

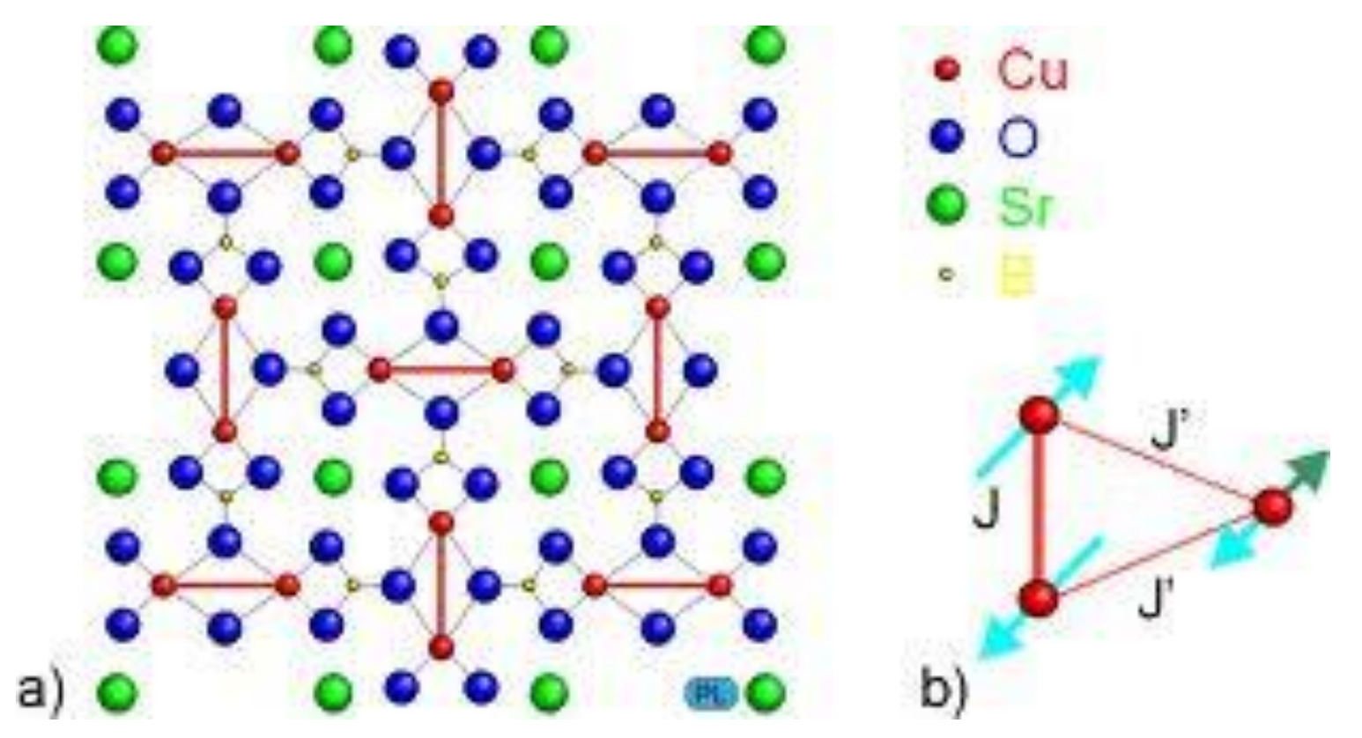


B.S. Shastry and B. Sutherland, Physica B 108, 1069 (81)

topologically equivalent to

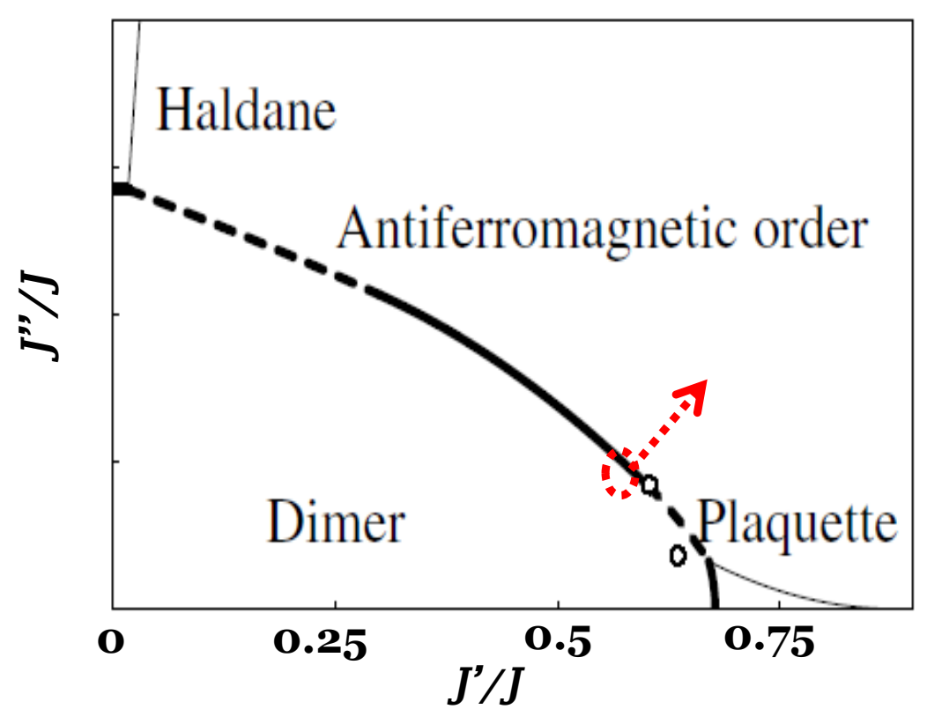
≡

$SrCu_2(BO_3)_2$



H. Kageyama, Phys. Rev. Lett. 82, 3168 (99)

Shastry-Sutherland Lattice (SSL)
→ intradimer coupling interaction J + interdimer coupling constant J'
→ When $J'/J<0.7$, a ground state is an exact product of singlets.



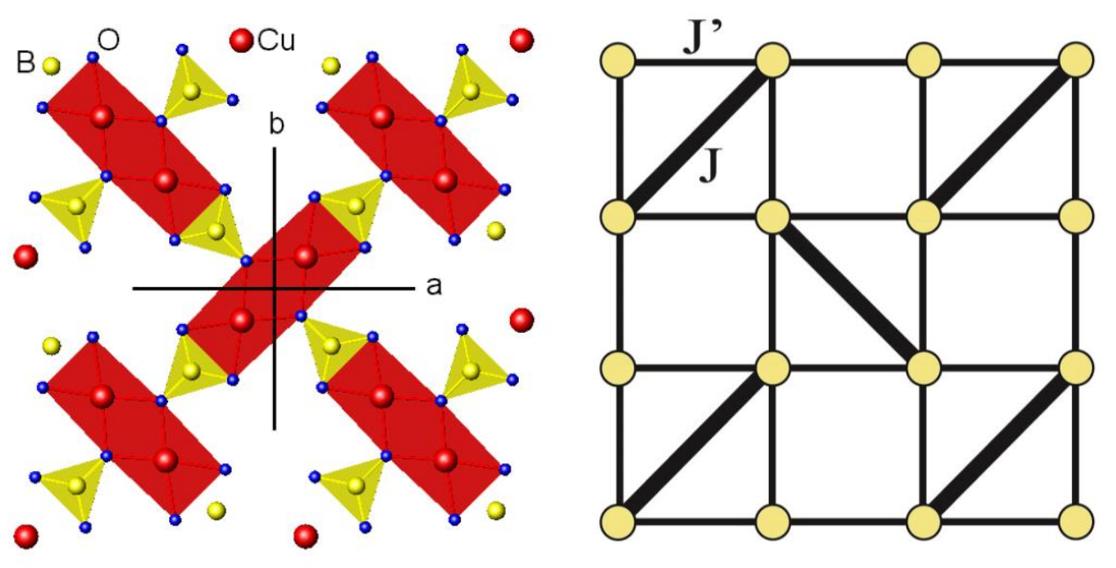
S. Miyahara and K. Ueda, J. Phys.:Condens. Matter 15, R327 (03).

$SrCu_2(BO_3)_2$ -
Experimental Realization of SSL with J'/J=0.63
→ Proximity to a quantum critical point
→ Chemical or hydrostatic pressure might induce a transition to a long-range ordered state.
→ One possible way to tune magnetic couplings is the substitution of Sr by Cd.

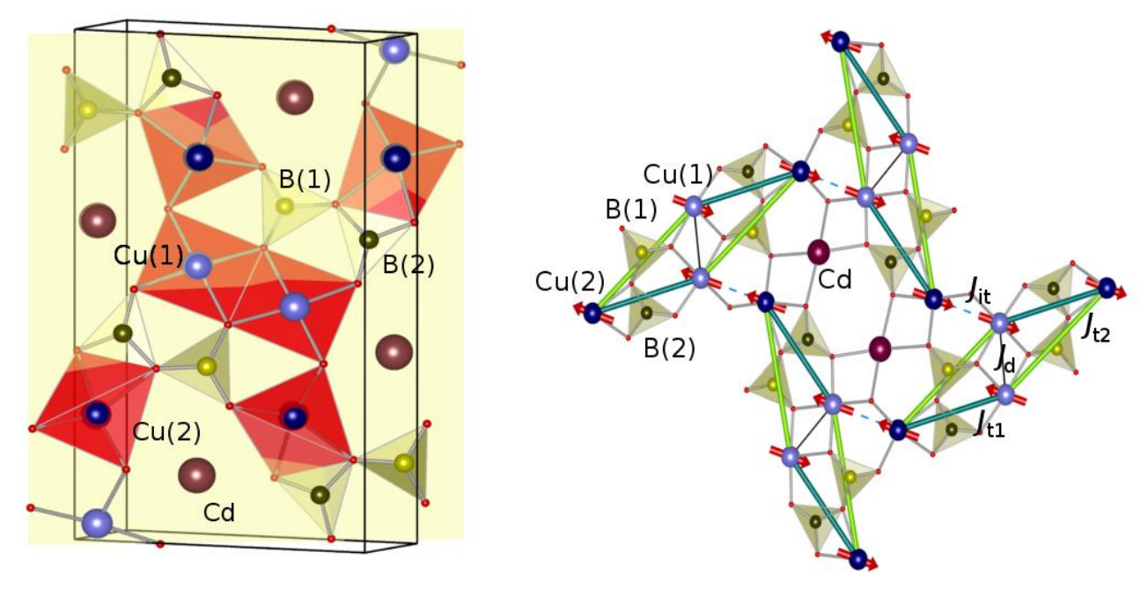
$SrCu_2(BO_3)_2$ vs $CdCu_2(BO_3)_2$

$SrCu_2(BO_3)_2$: tetragonal structure (I-42m)
- Spin dimer with a gap of Δ=33 K

$CdCu_2(BO_3)_2$: monoclinic structure ($P2_1/c$)
- AFLRO T_N=9.8 K

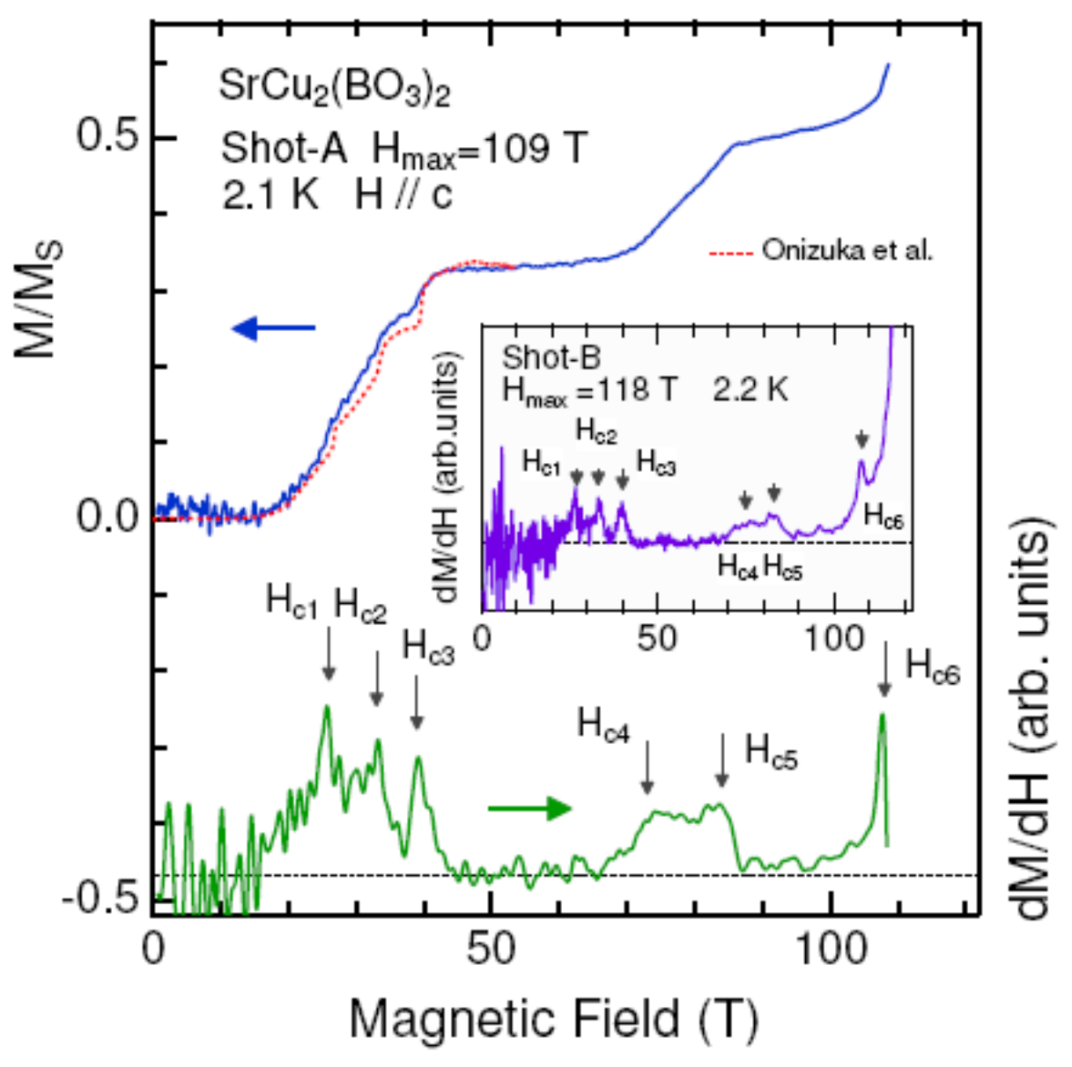


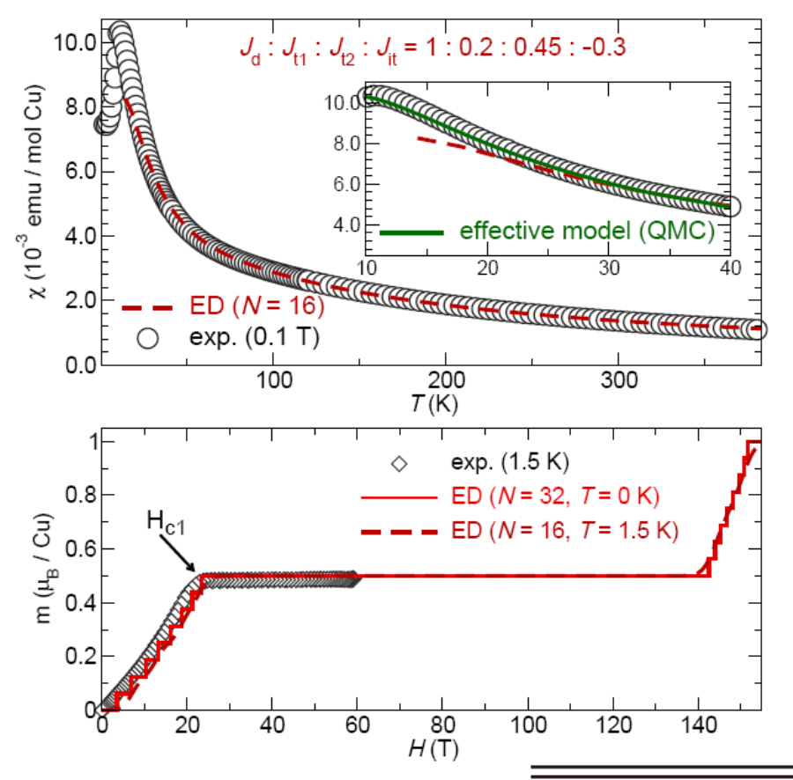
- Structural transformation
- Change of spin topology

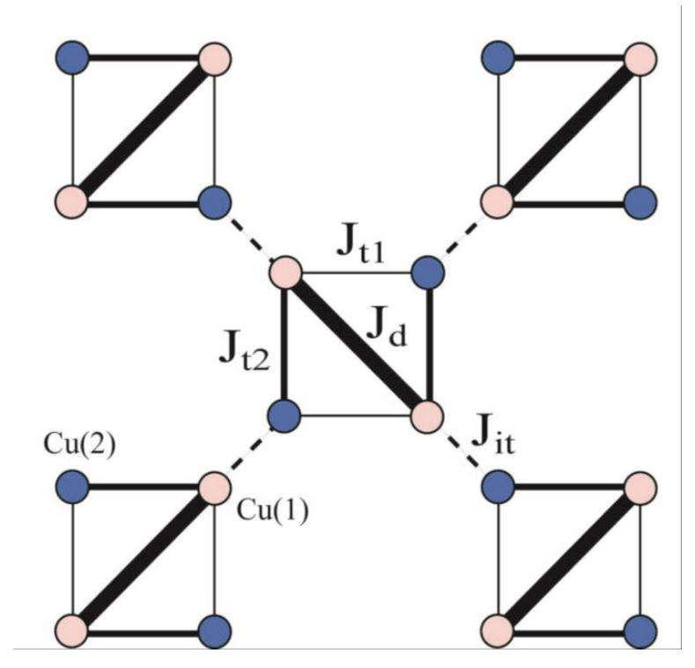


Two inequivalent Cu sites: 0.45 μ_B on Cu(1)
& 0.83 μ_B on Cu(2)

Magnetization



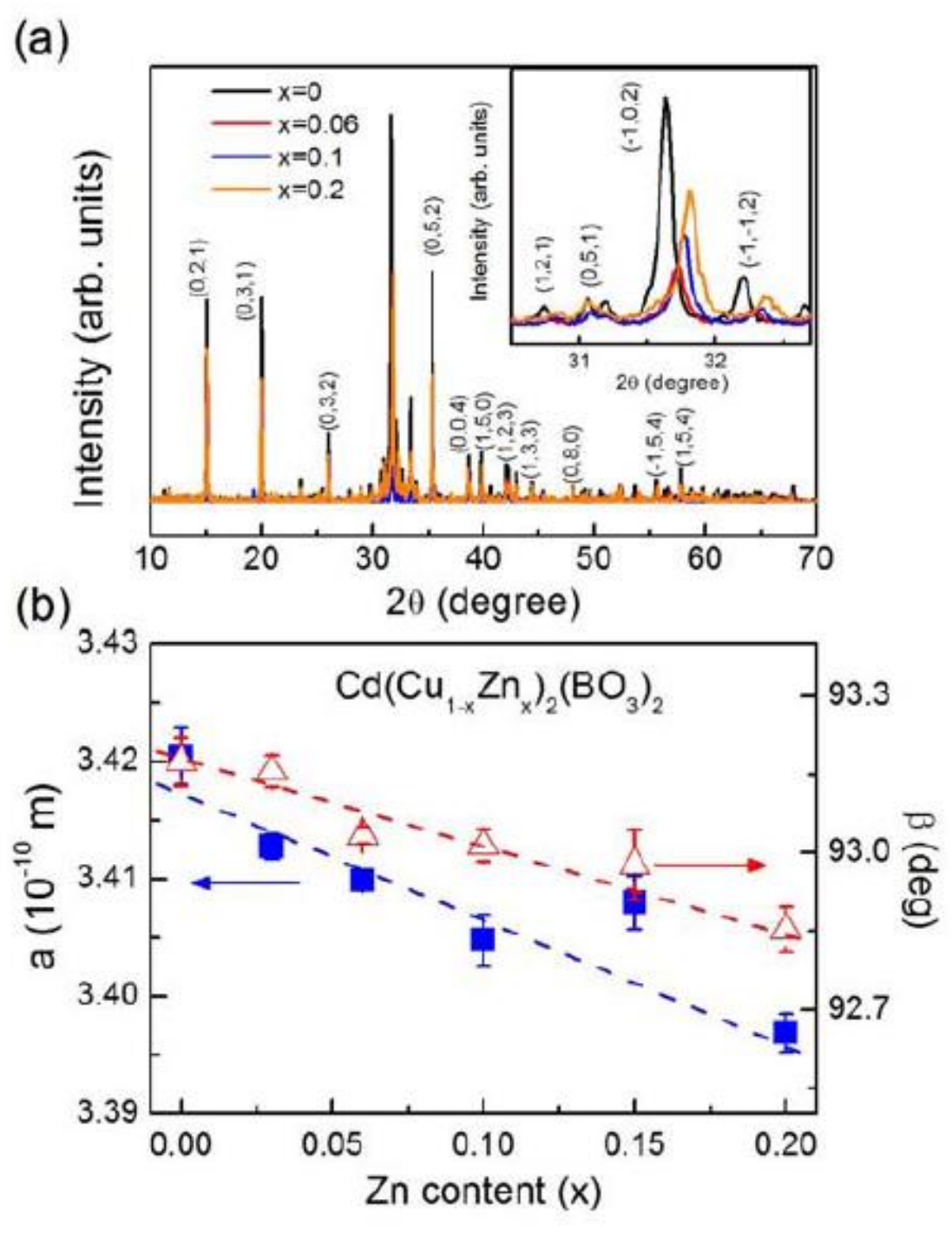


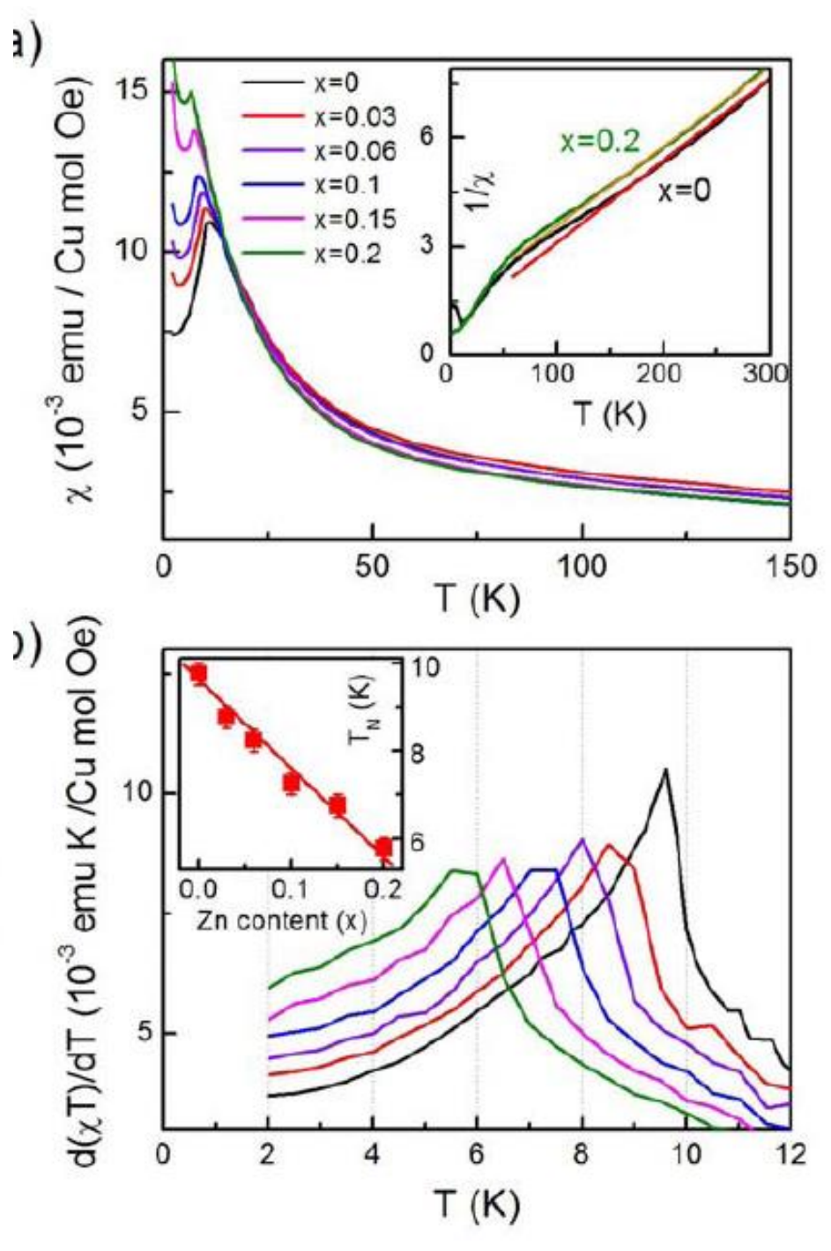


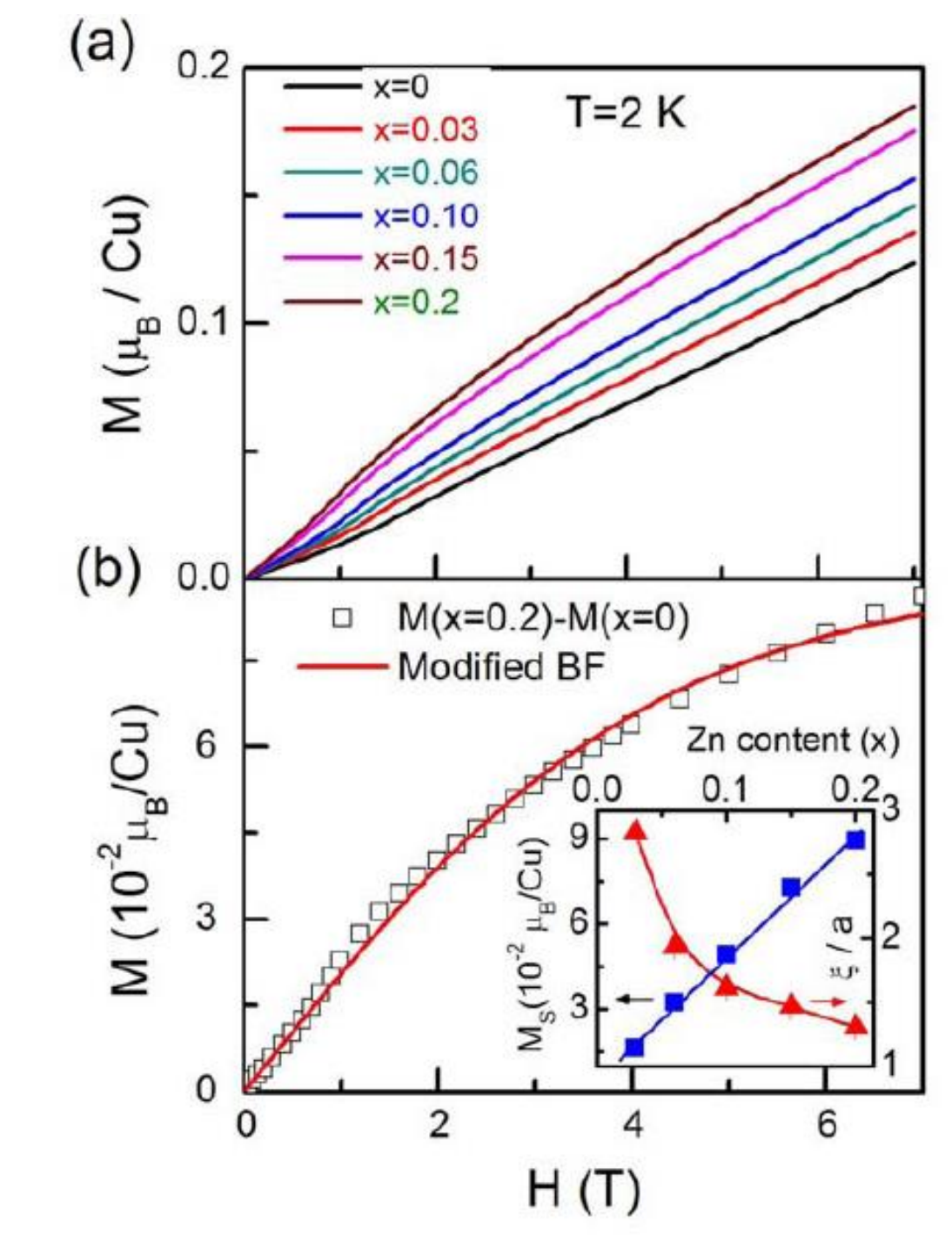
Y. H. Matsuda et al., PRL 111, 137204 (13)
O. Janson, et. al., PRB **85**, 064404 (12)

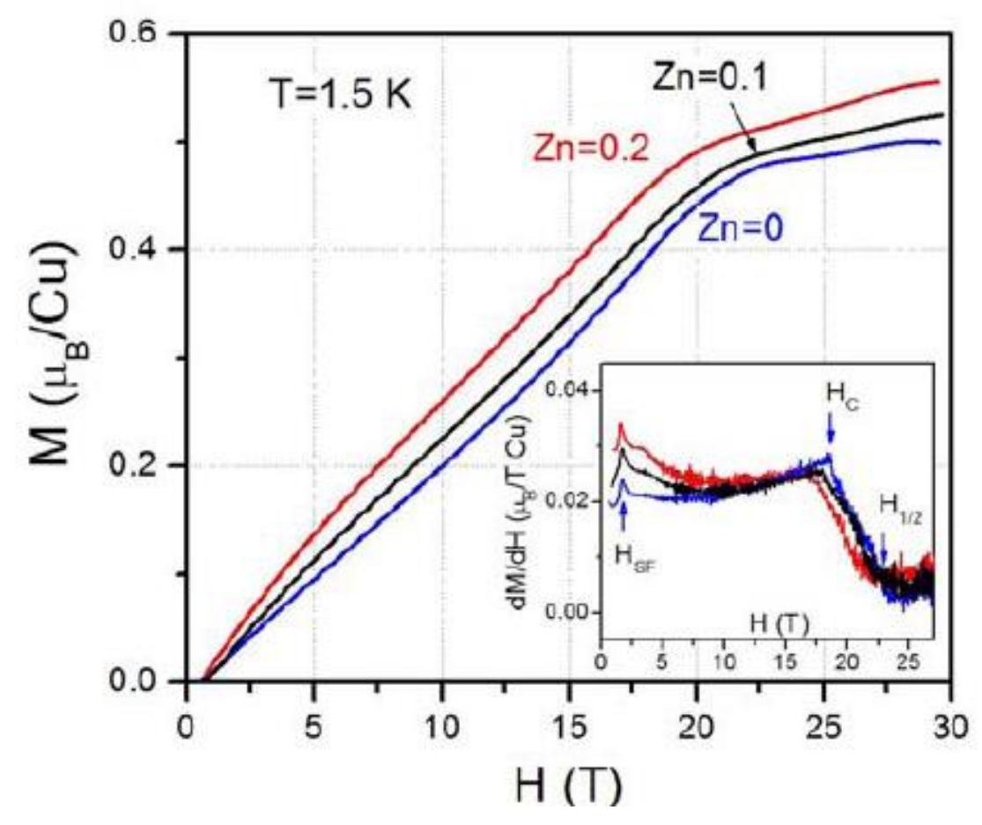
exchange	d	t_i	J_i^{AFM}	J_i (LSDA+U) AMF	 FLL
J_d	2.957	203	425	146	202
J_{t1}	5.268	73	55	30	29
J_{t2}	6.436	85	75	64	55
J_{it}	3.228	47	23	−45	−135
J_{Cd}	6.450	41	17	5	6
$J_{t1} : J_d$ (%)				20	14
$J_{it} : J_d$ (%)				−31	−67
$J_{t1} : J_{t2}$ (%)				47	53

Nonmagnetic impurity effects





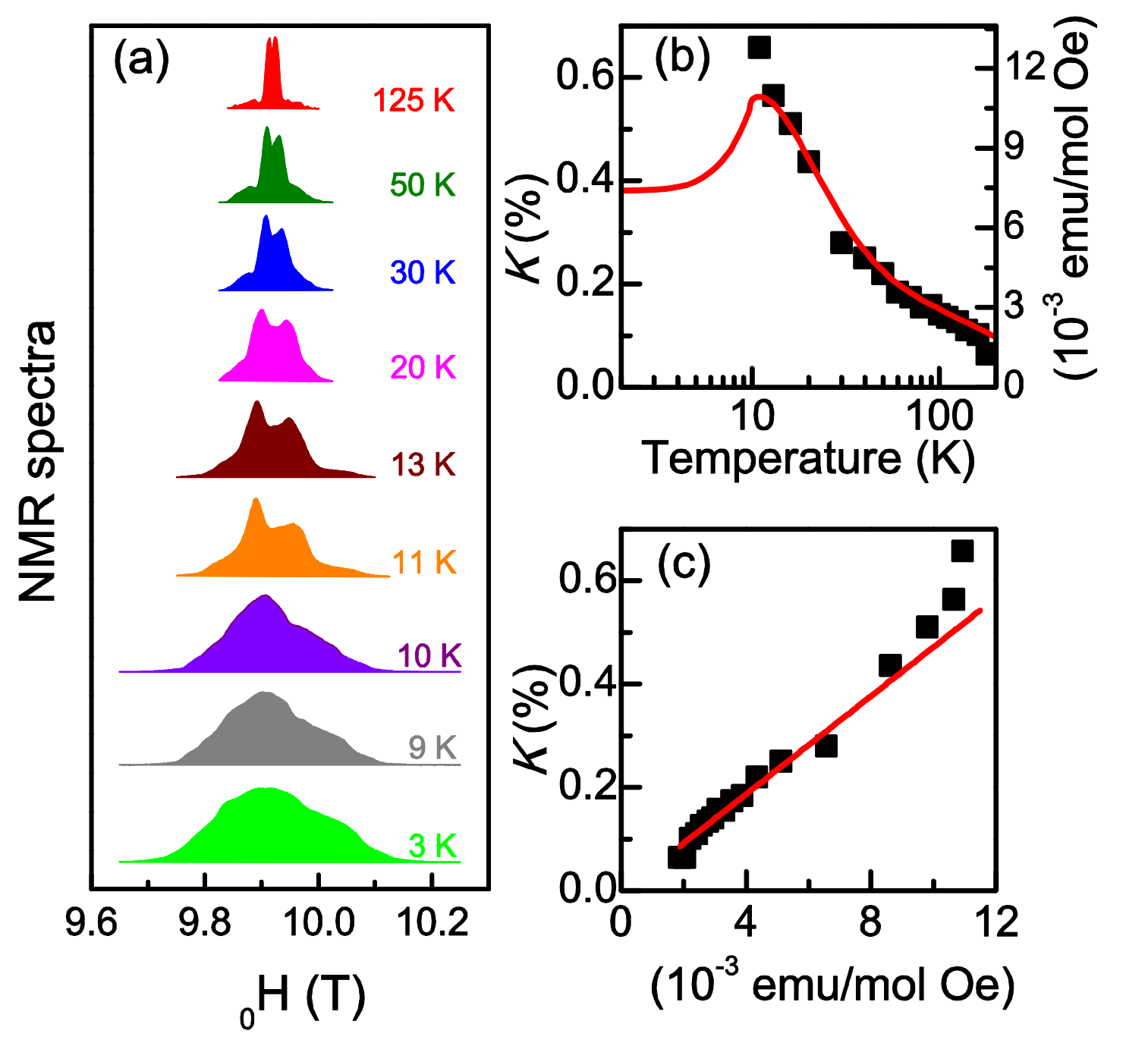




$Cd(Cu_{1-x}Zn_x)_2(BO_3)_2$

With increasing Zn contents,
(i) T_N decreases linearly with increasing x.
(ii) The spin-flop transition field is systematically reduced.
→ a dilution effect
(iii) Θ hardly varies with x.
→ a significant role of inter-tetramer couplings

^{11}B NMR results



◆ Two different Boron sites
(^{11}B : I = 3/2, γ/2π = 13.655MHz)

◆ Magnetic shift K(%)→ Clogston-Jaccarino plot

$$K_S(T) = K_{Chem} + \frac{A_{hf}}{N_A\mu_B}\chi_{spin}(T)$$

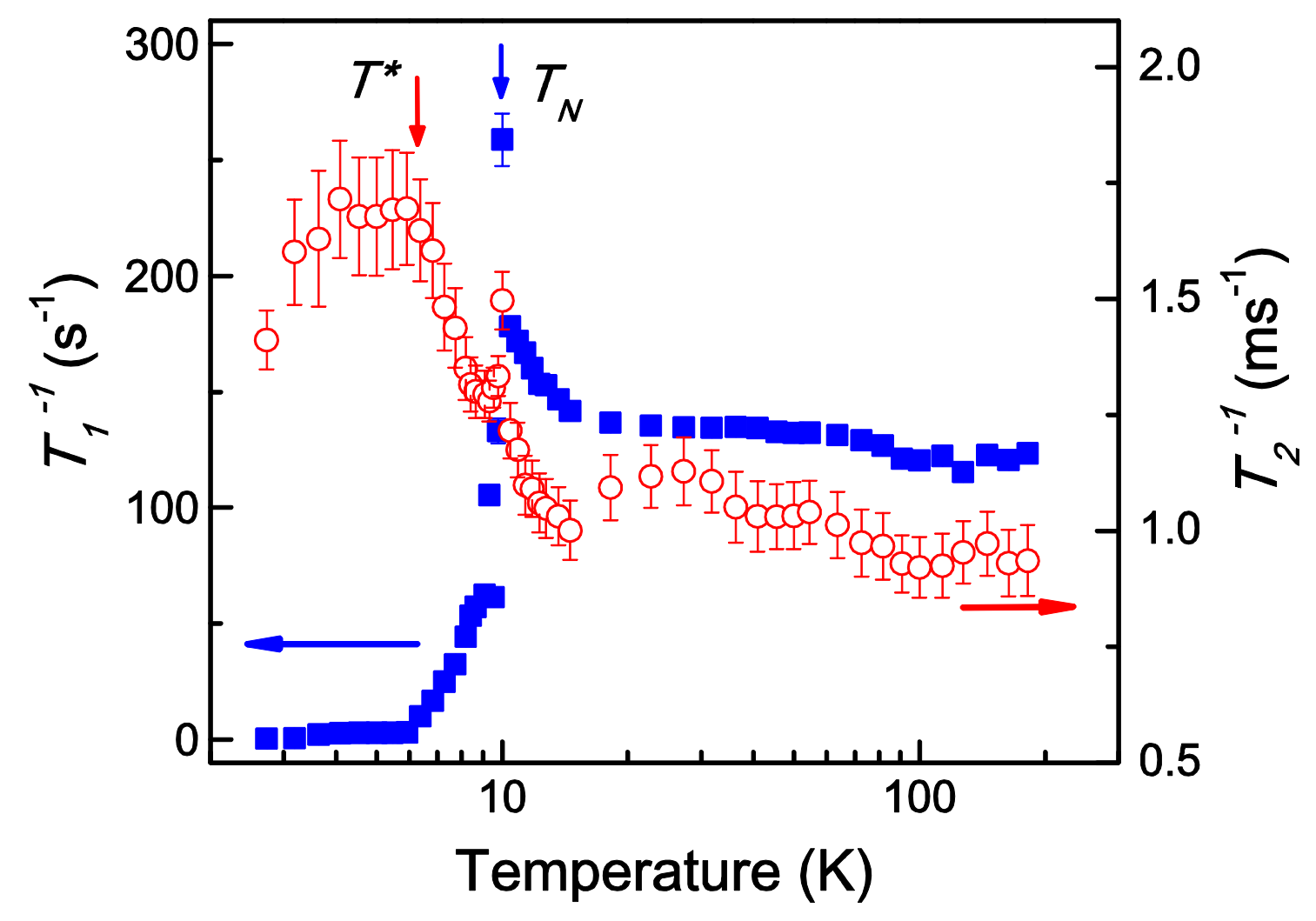
$A_{hf} = 0.047(5)\ \mathrm{T}/\mu_B$ and $K_{Chem} = -0.0037\ \mathrm{T}/\mu_B$

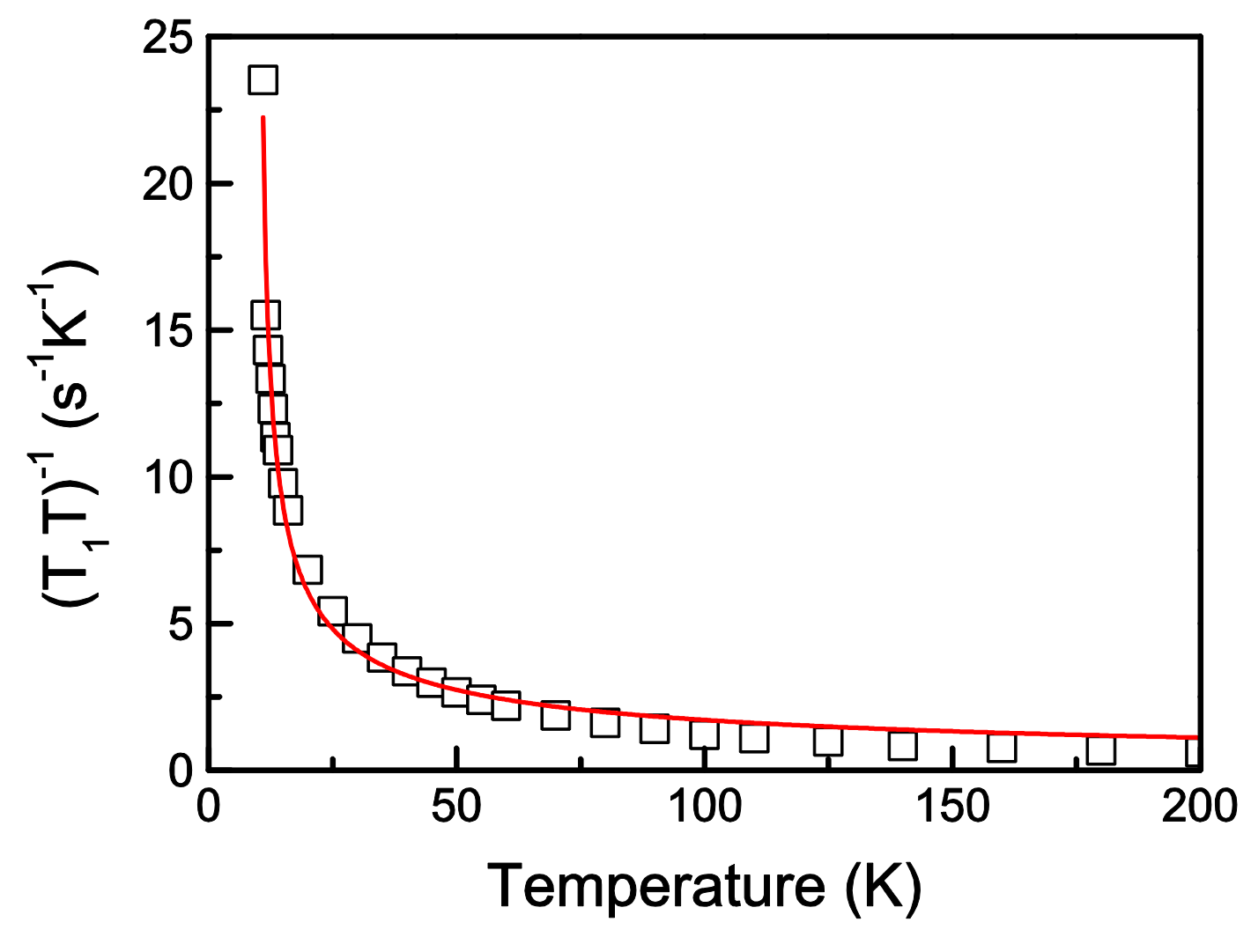
◆ $1/T_1$ diverges at T_N and flattens out below T^*=6.5 K.
$1/T_2$ has a broad maximum around T^* = 6.5 K.
--> Presence of magnetic anomaly in an ordered state

◆ The averaged dynamic susceptibility

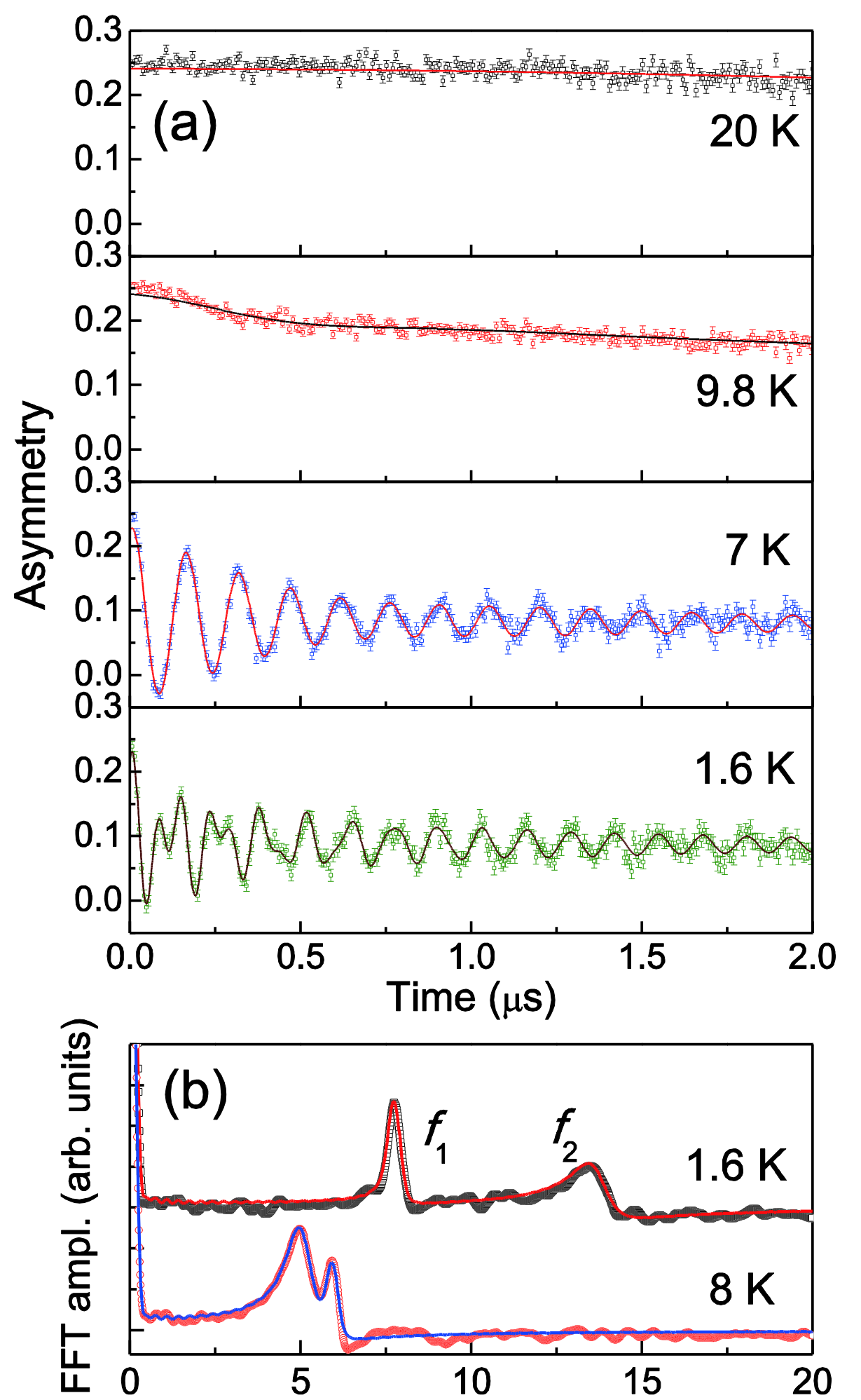
$$\frac{1}{T_1T} \propto (T - T_N)^{-\alpha}$$

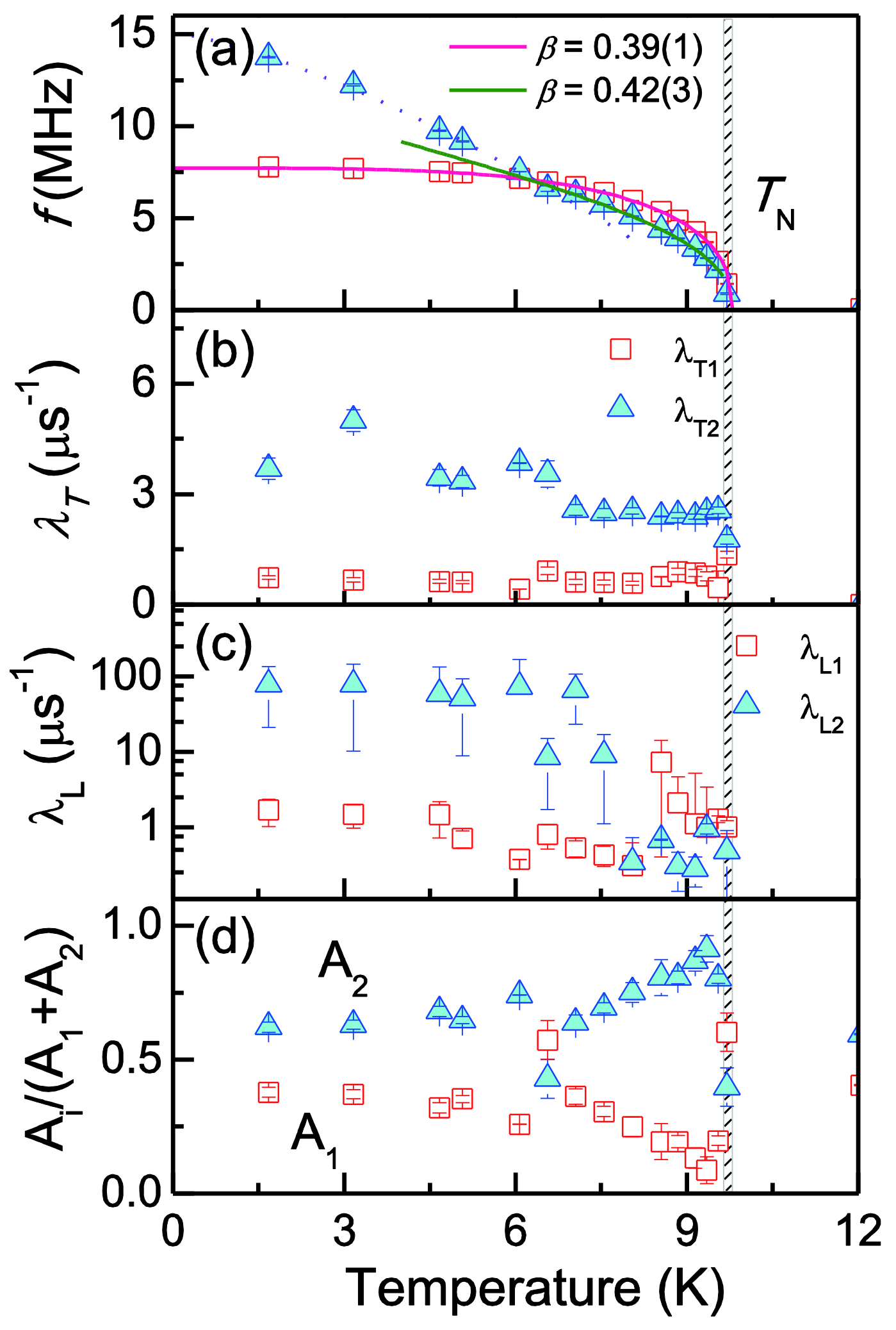
with $T_N = 9.92(9)$ K, $\alpha = 0.57(8)$.





ZF-μSR





◆ symmetry function

$$P(t)= \sum_{j=1}^{2} A_j[\alpha_j cos(2\pi f_j + \phi_j)\exp(-\lambda_{Tj}t)] + (1-\alpha_j)\exp(-\lambda_{Lj}t)]$$

A_j: the volume fraction of the two muon sites
$\lambda_{T,L}$: transverse and longitudinal relaxation rate
ϕ_j : initial phase of the oscillatory signal
f_j: muon Larmor frequency

◆ Two muon frequencies at f_1=7.77 and f_2=13.73 MHz at T=1.6 K.
f_1: f_2=1 : 1.962 by μSR
cf. 0.45 μ_B on Cu(1): 0.83 μ_B on Cu(2)=1 : 1.844 by NS [M. Hase *et al*, PRB **80**, 104405 (09)]
→ Two muon sites near the respective Cu(1) and Cu(2) spins

◆ Order parameter

$$f = f_0 \cdot \left(1-\frac{T}{T_N}\right)^{\beta}$$

- β = 0.42(3) and 0.39(1)
cf) $\beta = 0.367$ for 3D Heisenberg antiferromagnets

◆ The anomalous evolution of the muon frequency below T^* = 6.5 K indicates a reorientation of spins.

◆ A energy hierarchy of frustrated spin topology
- strongly interacting Cu(1) spins with singlets
+ nonfrustrated weakly interacting Cu(2) spins
+ weak AFM intertramer interaction of J_{Cd}
→ Reorientation of the magnetic structure

Conclusion

(1) ^{11}B -NMR and μSR provide a spectroscopic signature for the long-range AFM ordering at T_N.
(2) $1/T_2$ and μSR parameters show magnetic anomaly at T^*= 6.5 K below T_N = 9.8 K.
(3) The ordered moment of Cu(1) and Cu(2) spin evolves in a markedly different manner for temperature below T^*. This indicates site-dependent spin correlations.
(4) The large difference in the degree of frustration and magnetic interactions between Cu(1) and Cu(2) spin, combined with weak intertramer interaction, is ascribed to the reorientation of spins at T^*.

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