A magnetic analog of the isotope effect in cuprates (and more)

Work of Yuval Lubashevsky, Rinat Ofer, and Amit Kanigel

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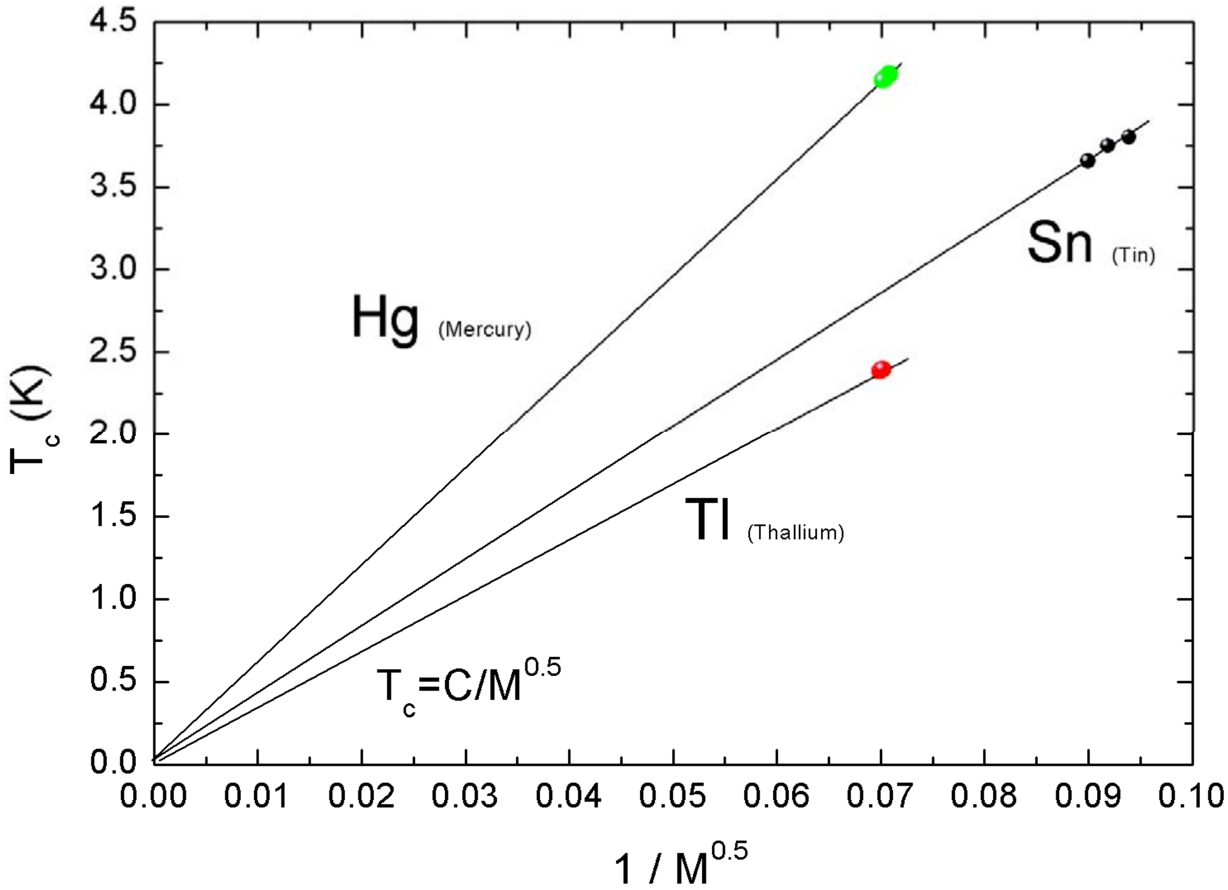
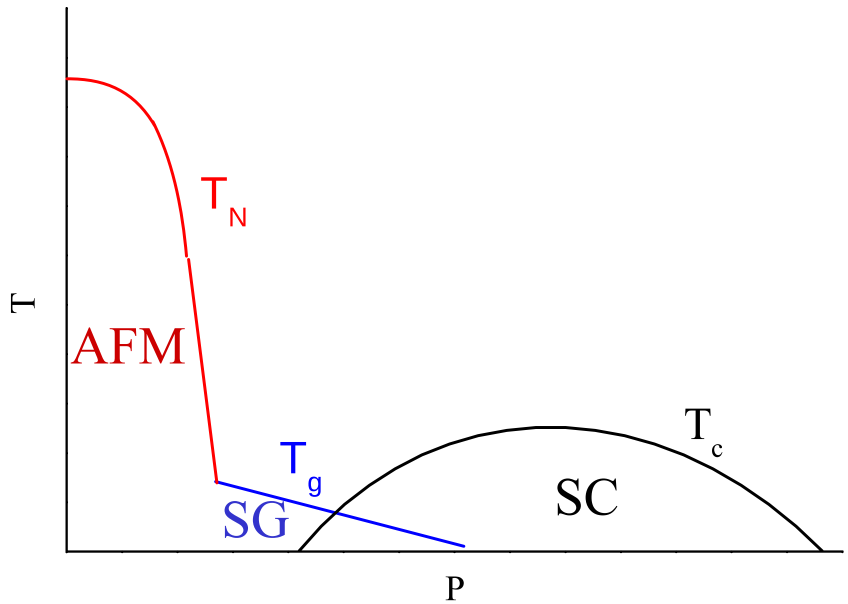
Isotope Effect in metallic superconductor



- C. A. Reynolds et. al., Phys. Rev. 84, 691 (1950).
- B. Serin et al., Phys. Rev. 86, 162 (1952).
- E. Maxwell et al., Phys. Rev. 95, 333 (1954).

- Maximum 4% variation of T_c in Sn.
- The $T_c \propto M^{-1/2}$ is not applicable for different materials.

Our Main Objective:

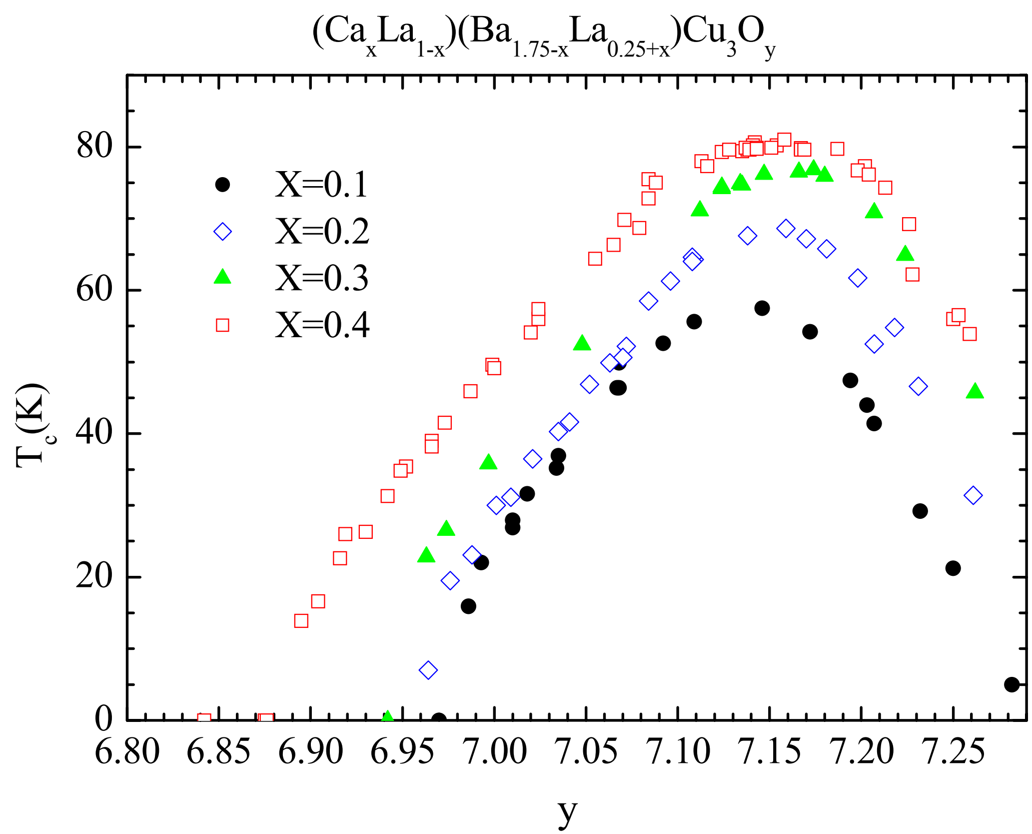


- To strengthen the J **<u>with no other changes</u>**, and see if T_c grows?

- This would be a magnetic equivalent of the isotope effect.
- Experimentally this is difficult but not inconceivable.

CLBLCO; Our Model Compound

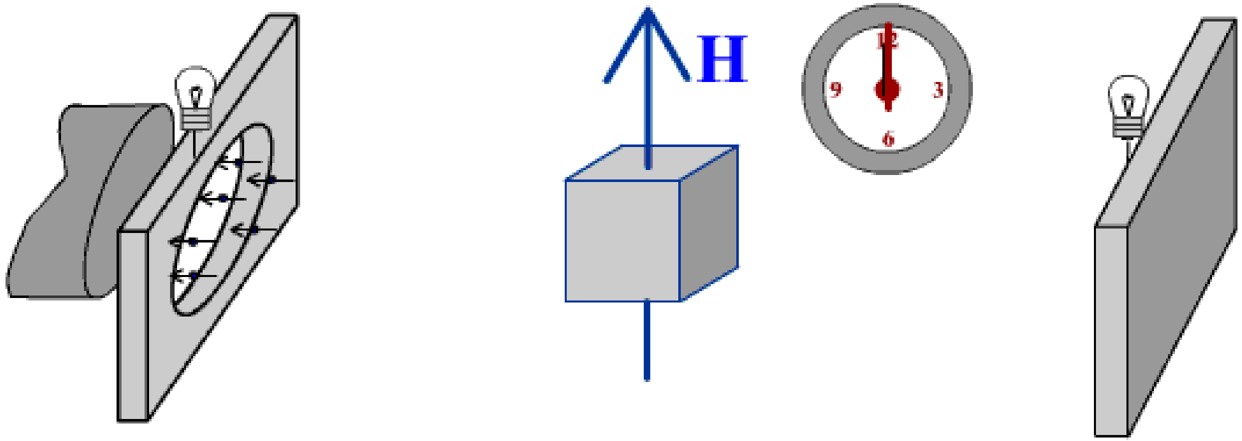
- $YBa_2Cu_3O_y$ structure.
- Tetragonal at all x and y.
- 2 planes per unit cell.
- Over doping is possible.
- T_c variation of 30%.
- Valance Ca=Ba=2, La=3.
- Similar level of disorder (NMR).
- Ionic radii variations are not relevant.



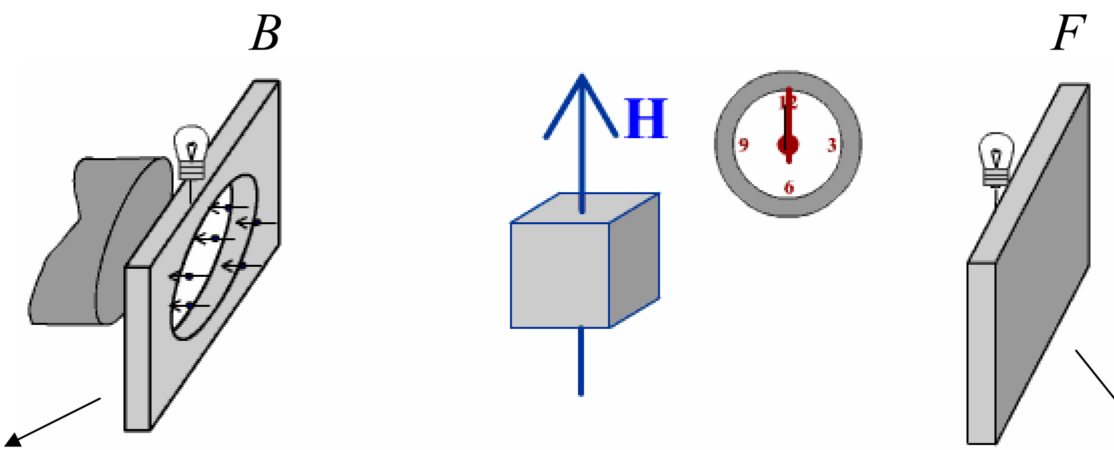
Goldschmidt *et al.*, Phys. Rev. B **48**, 532 1993

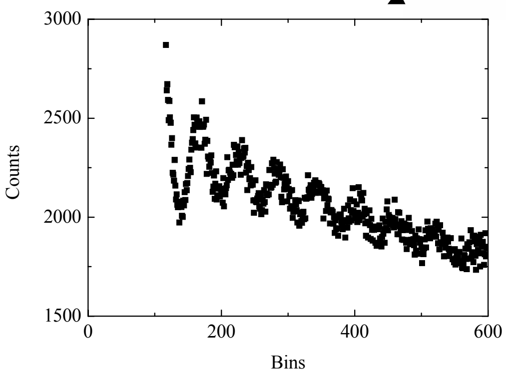
- CLBLCO allows T_c^{max} variations with minimal structural changes.
- What would T_N , T_g, and T^* do?

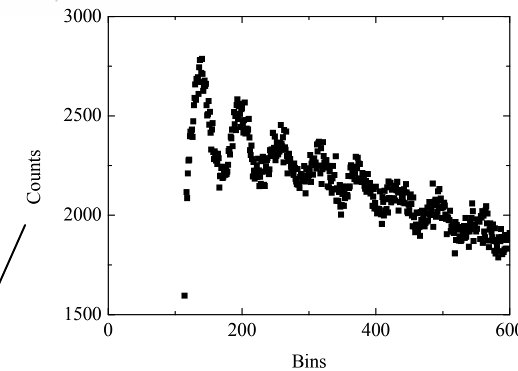
Principals of μSR



Principals of μSR



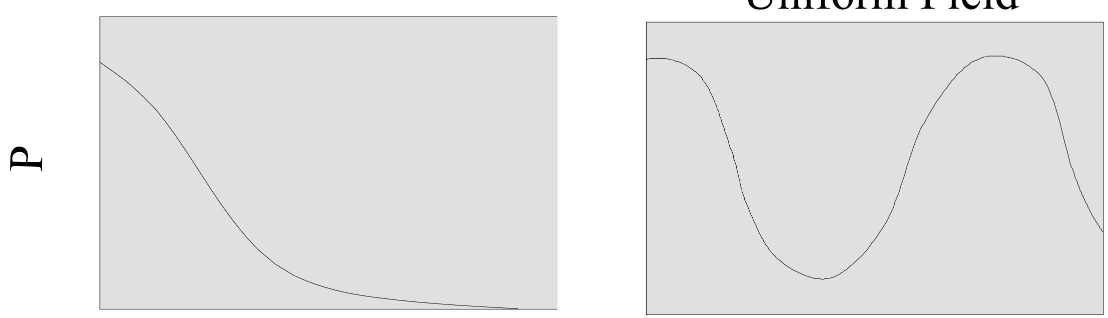




Asymmetry = (F-B)/(F+B)∝ $P_z(t)$.

Random Field

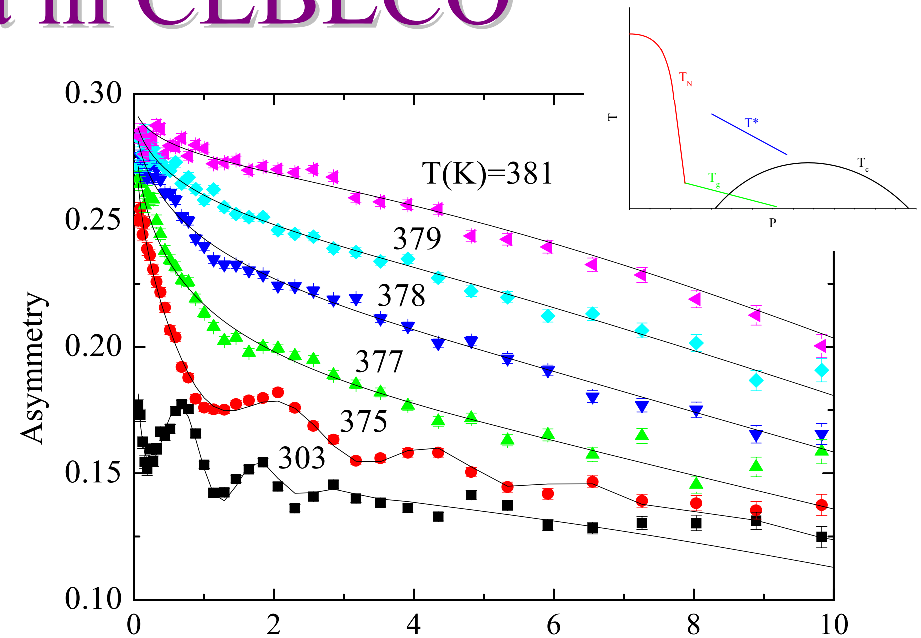
Uniform Field

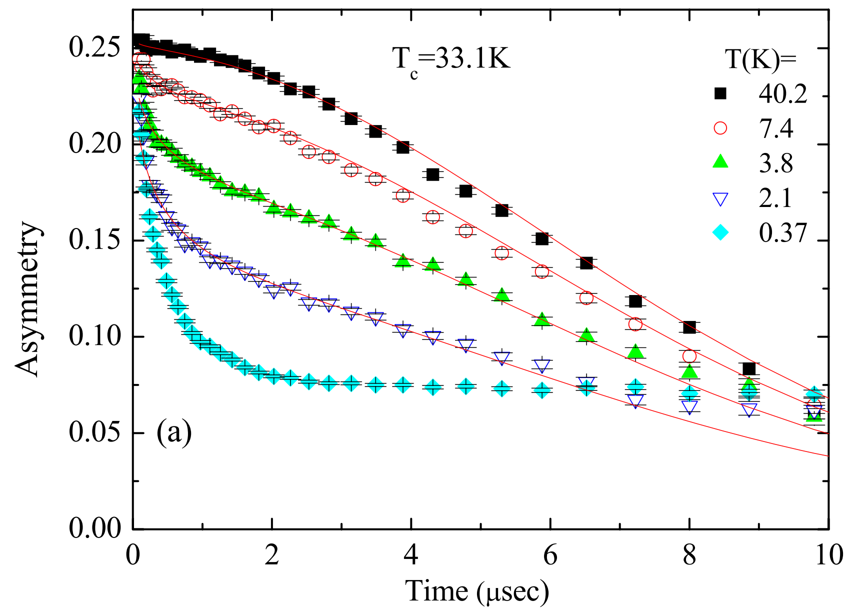


Time

Time

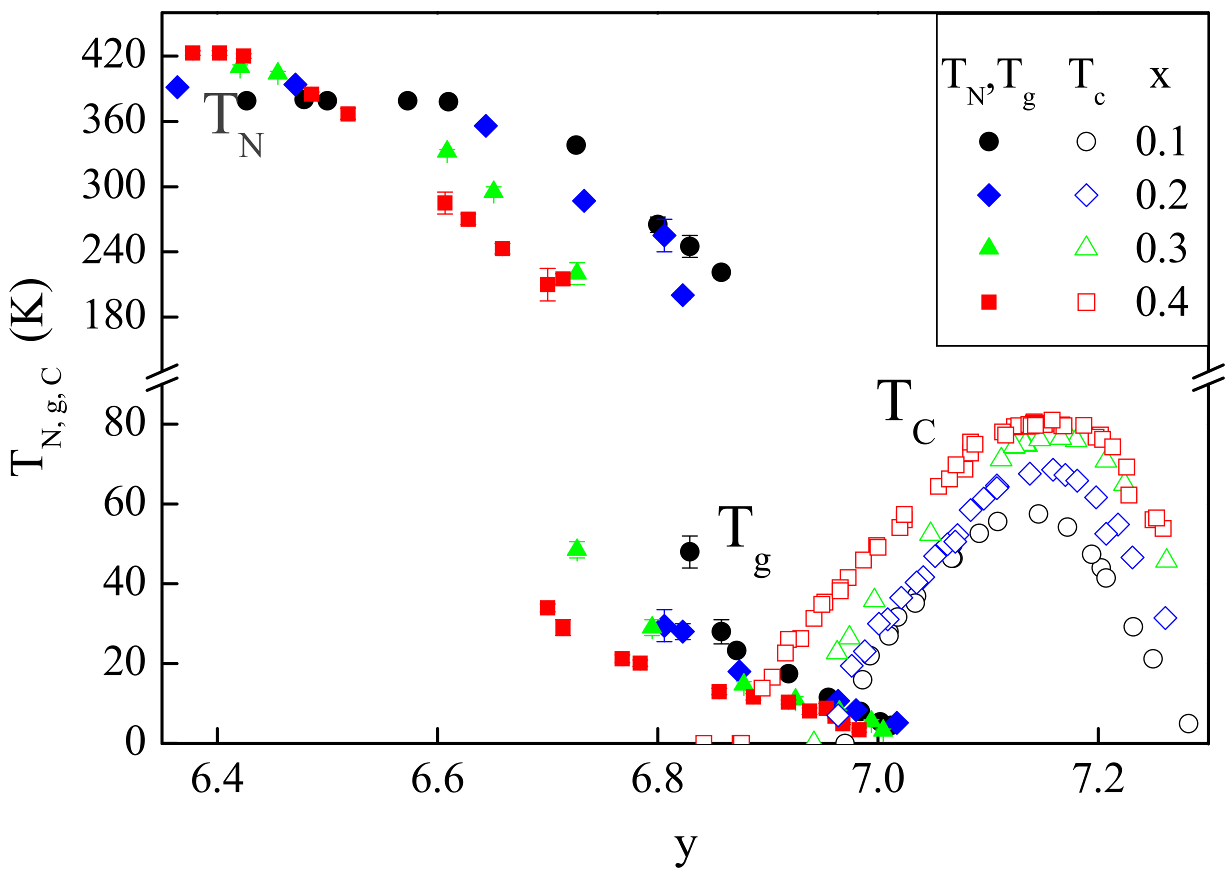
Raw ZF μSR Data in CLBLCO



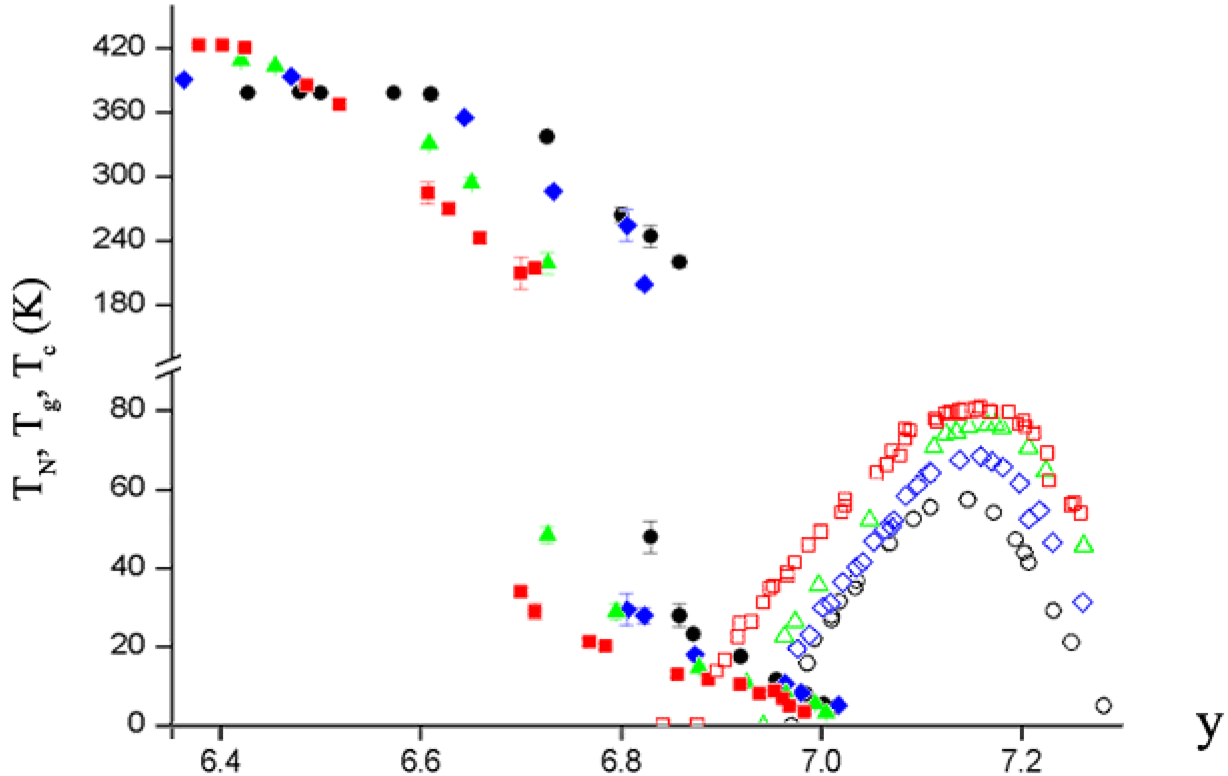


- There is weak relaxation above T_N or T_g.
- There are oscillations in the ordered phase but not in the spin glass phase.
- There are 2 contributors to $P_z(t)$. As one amplitude grows, the other decreases.

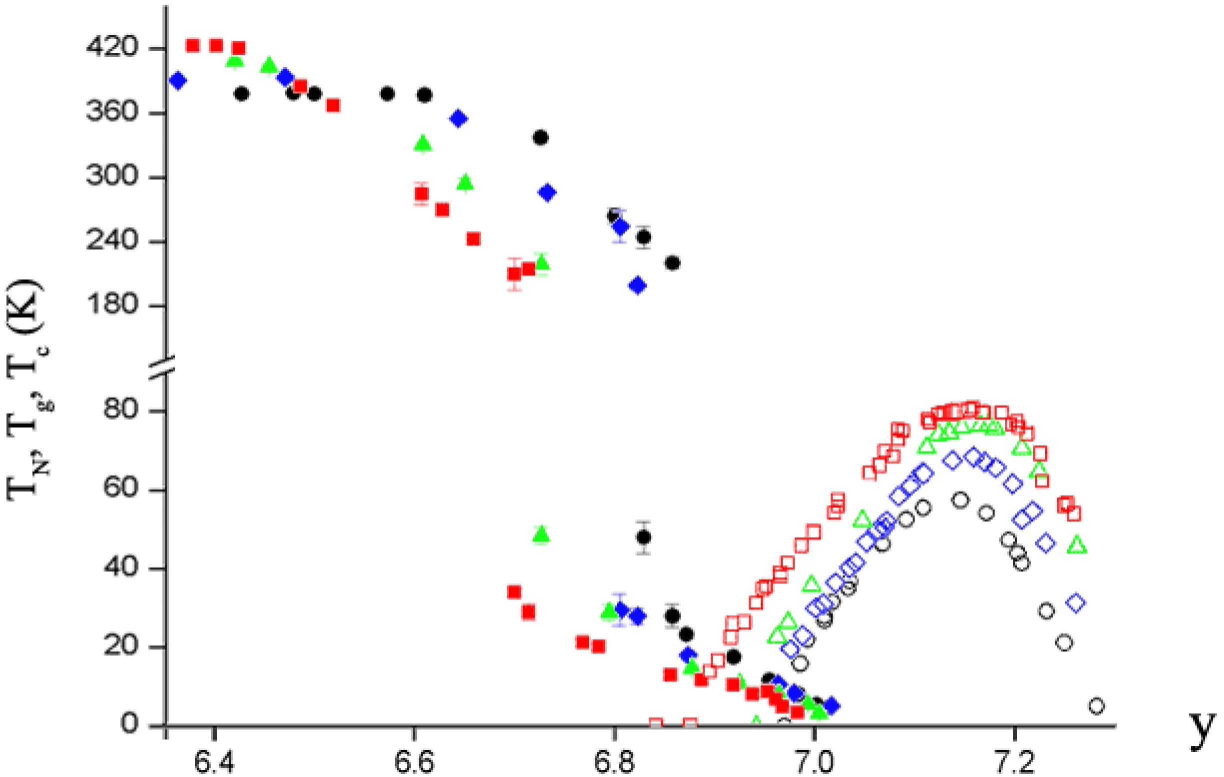
Phase Diagram $(Ca_xLa_{1-x})(Ba_{1.75-x}La_{0.25+x})Cu_3O_y$



- The family with the highest T_c at optimal doping has the highest T_N .
- How do we untangle this phase diagram?



420
360
300
240
180
80
60
40
20
0
6.4
6.6
6.8
7.0
7.2
T_N, T_g, T_c (K)
y



420
360
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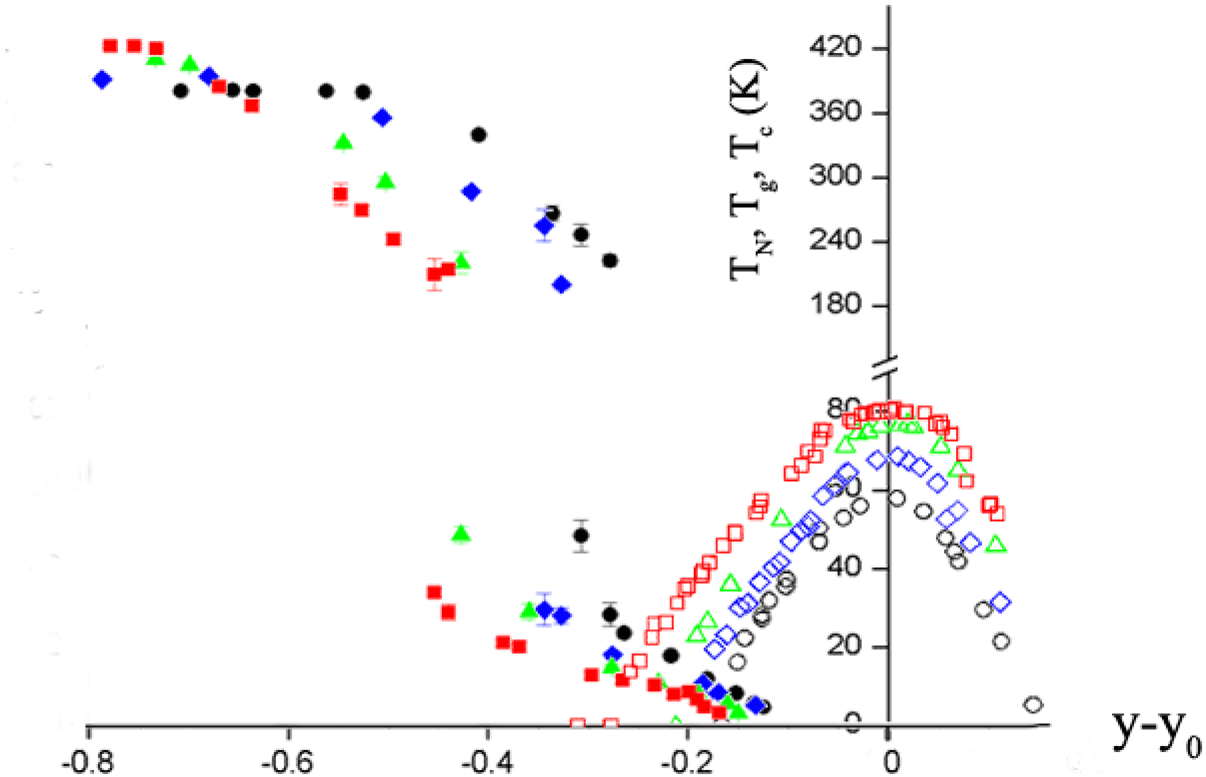
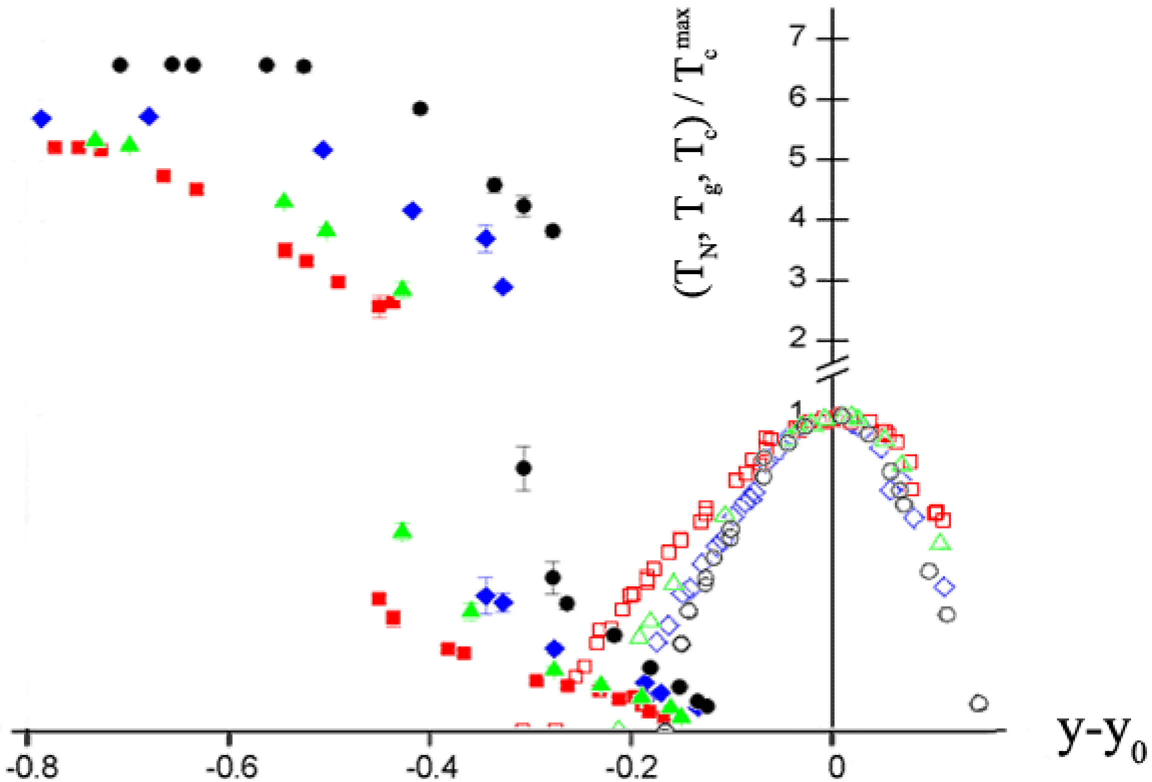
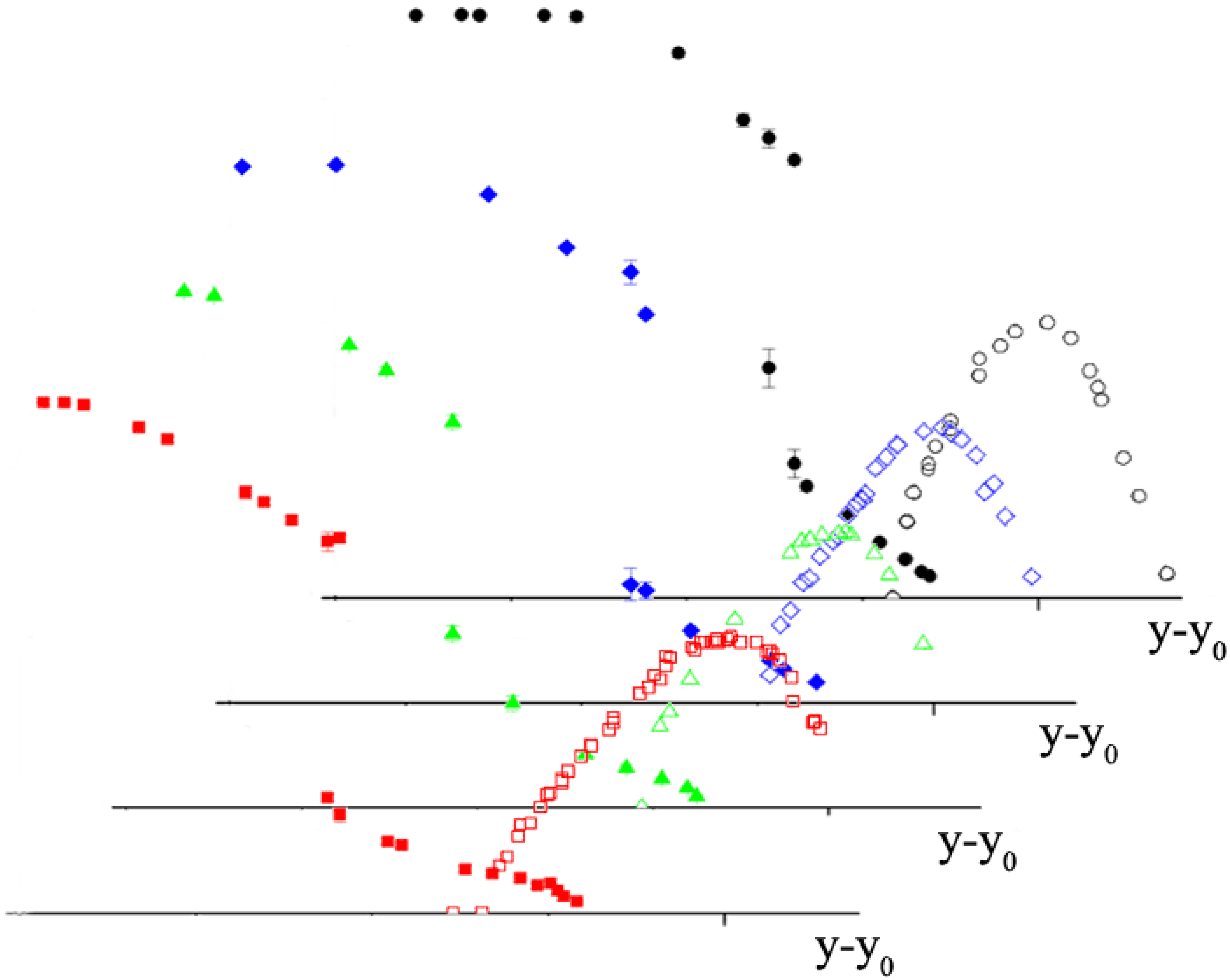
T_N, T_g, T_c (K)
y

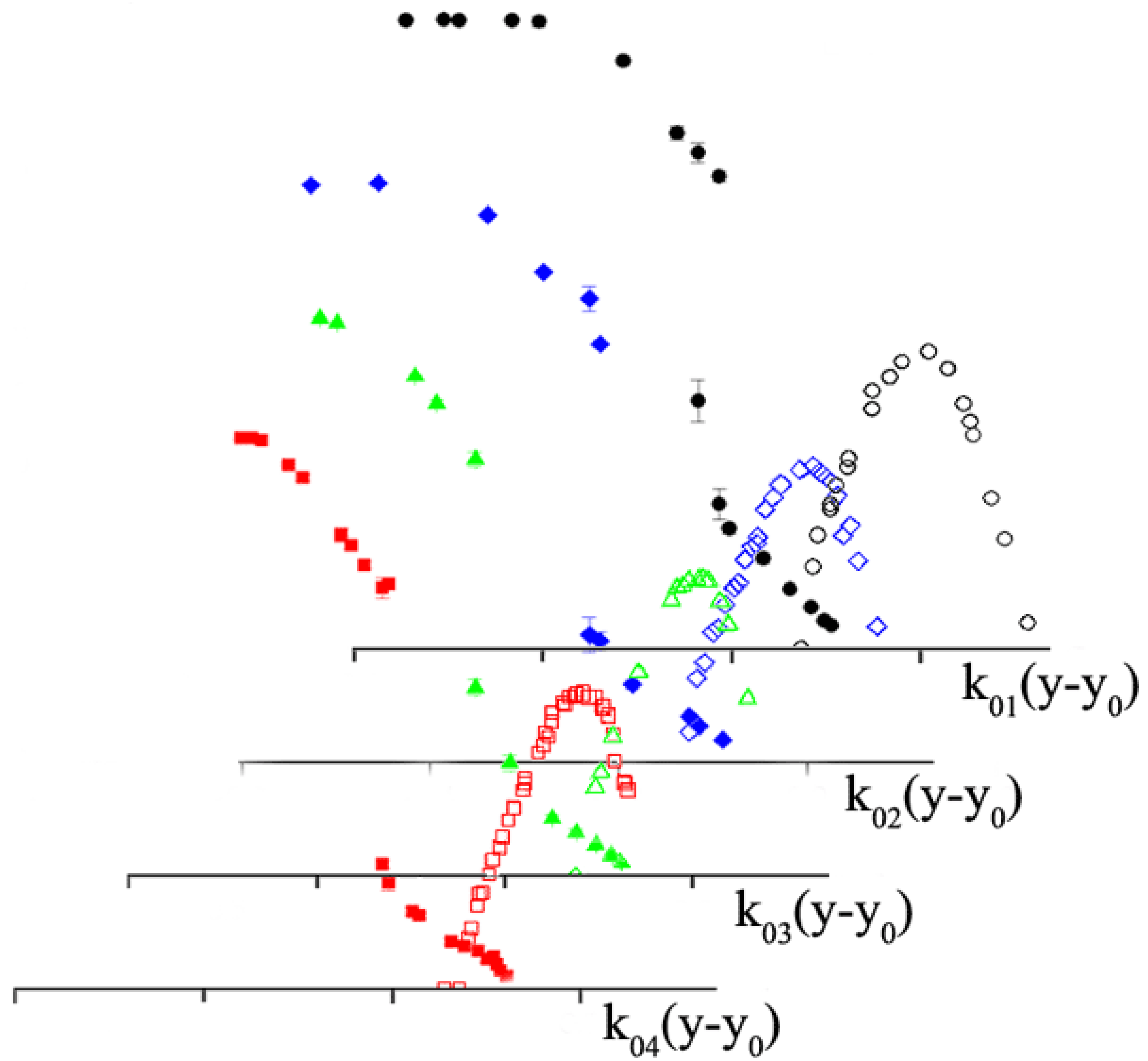


420
360
300
240
180
80
60
40
20
0
T_N, T_g, T_c (K)
-0.8
-0.6
-0.4
-0.2
0
$y-y_0$

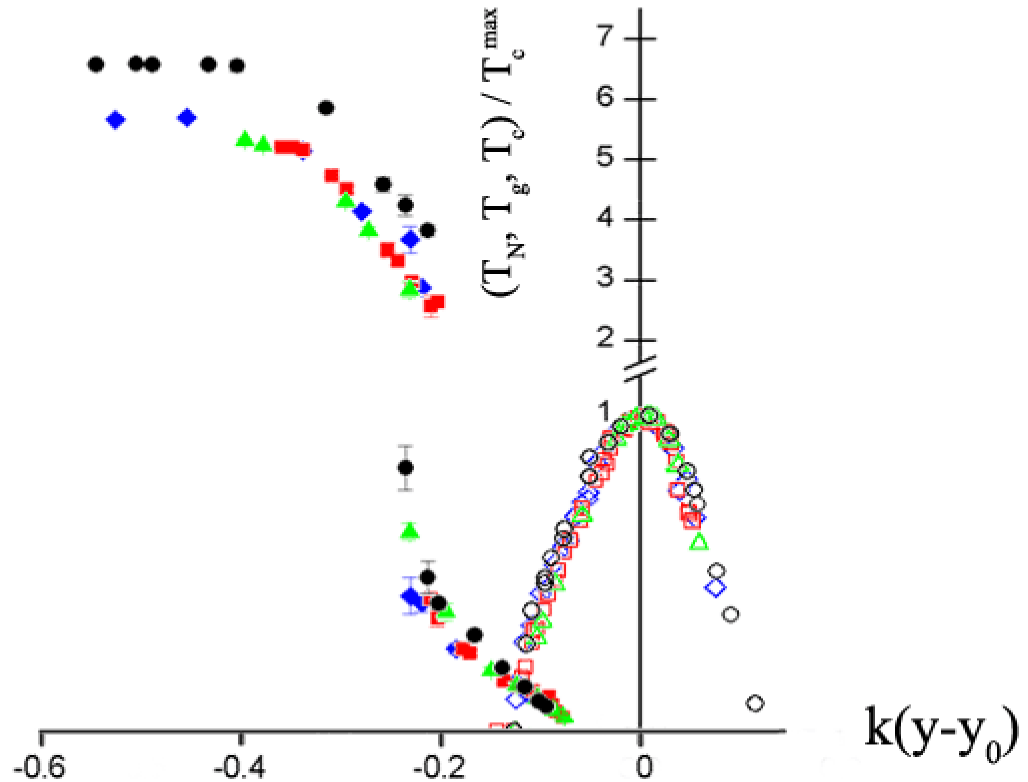


$(T_N, T_g, T_c) / T_c^{max}$
7
6
5
4
3
2
1
-0.8
-0.6
-0.4
-0.2
0
$y-y_0$


y-y0
y-y0
y-y0
y-y0



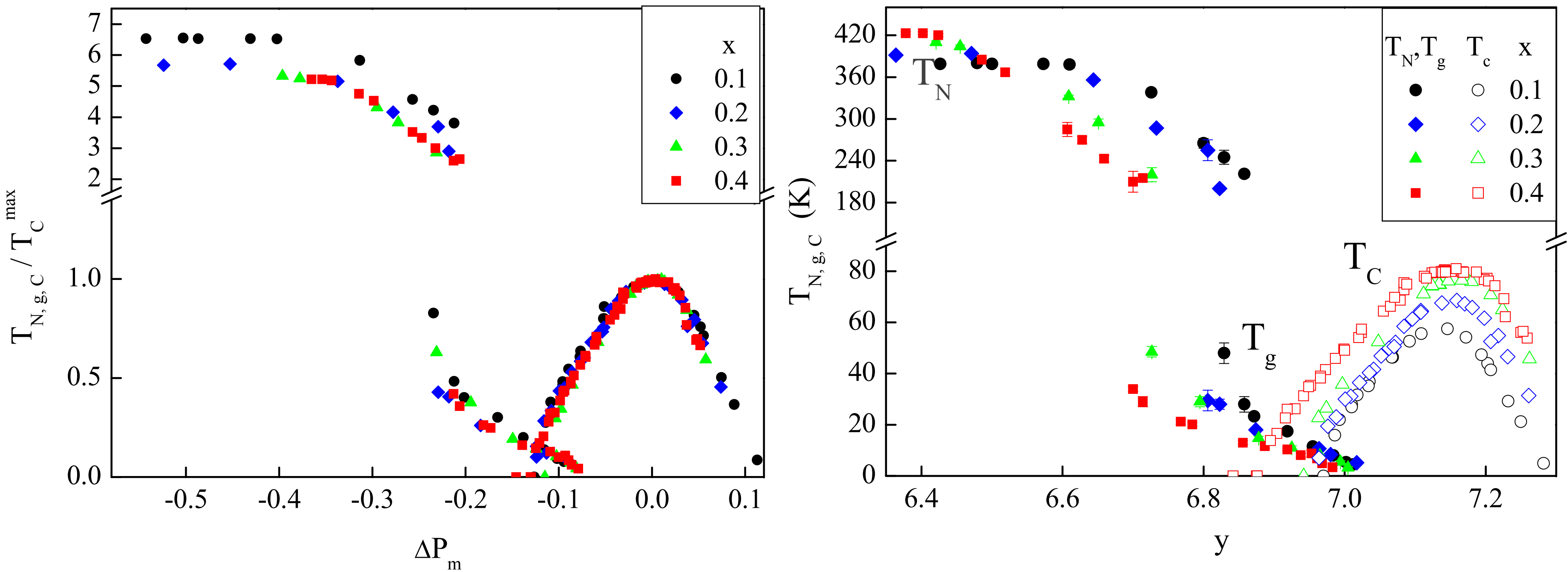
$k_{01}(y-y_0)$
$k_{02}(y-y_0)$
$k_{03}(y-y_0)$
$k_{04}(y-y_0)$



$(T_N, T_g, T_c) / T_c^{max}$
7
6
5
4
3
2
1
-0.6
-0.4
-0.2
0
$k(y-y_0)$

Transformation of the entire doping range.

$(Ca_xLa_{1-x})(Ba_{1.75-x}La_{0.25+x})Cu_3O_y$



$$T_N, T_g, T_c \rightarrow T_N / T_c^{max}, T_g / T_c^{max}, T_c / T_c^{max}$$

$$\Delta p_m = K_x (y - y_x^{opt})$$

- The scaling works for x=0.2 to 0.4 (15% variation in T_c^{max})!
- What about x=0.1?

The role of anisotropies

- T_N is determined by J and anisotropies.
- The spin Hamiltonian is given by

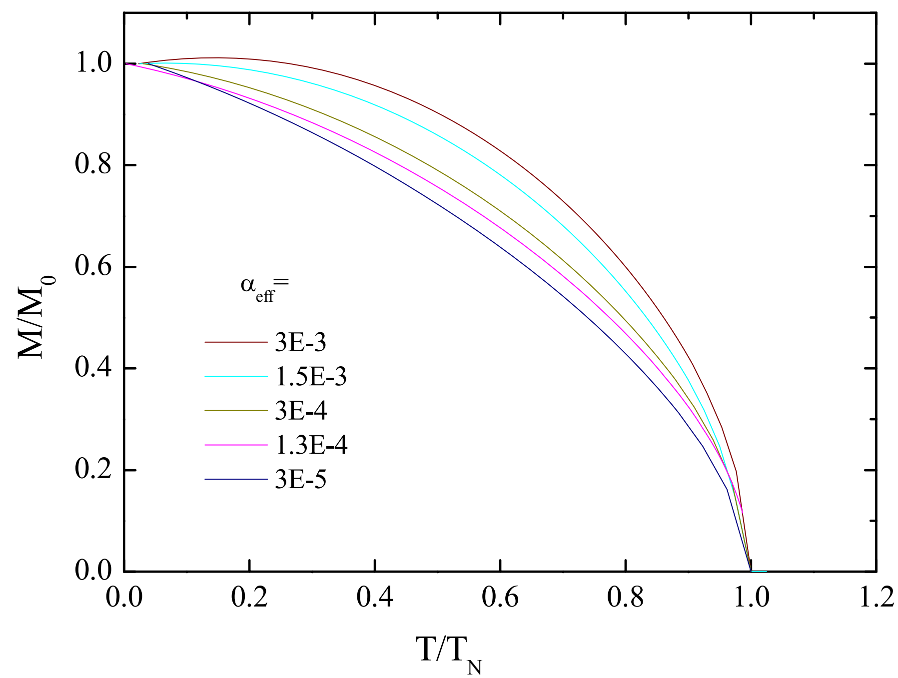
$$H = J\left(\sum_{\text{in plane}} \mathbf{S}_i \mathbf{S}_j + \alpha_{xy} \sum_{\text{in plane}} S_i^z S_j^z + \alpha_{\perp} \sum_{\text{between planes}} \mathbf{S}_i \mathbf{S}_j\right)$$

- Information on anisotropies can be obtained from *M(T)* measurements.

$$\alpha_{eff} = z_{xy}\alpha_{xy} + z_{\perp}\alpha_{\perp}$$

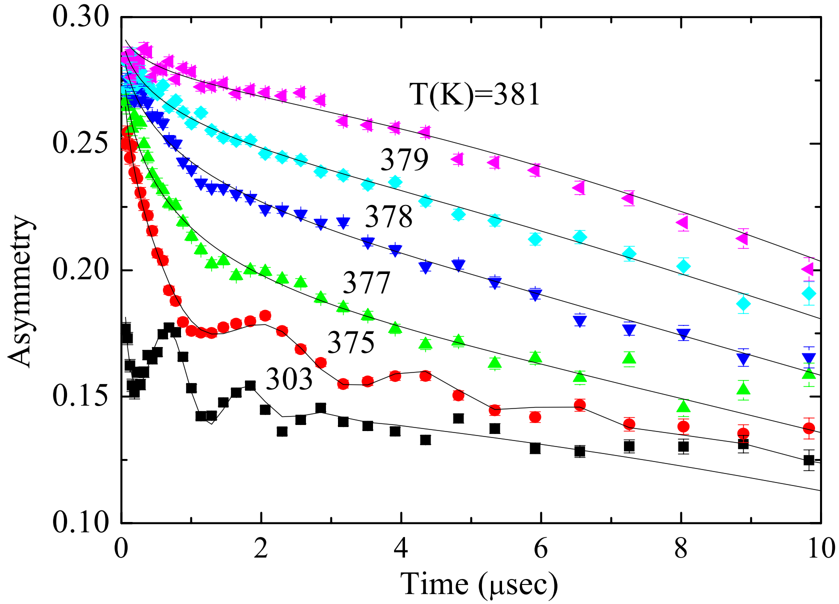
$$T_N = J t_N(\alpha_{eff})$$

$$t_N(\alpha_{eff}) = \pi M_0 \Big/ \left|\ln\left|\frac{4\alpha_{eff}\ln(4\alpha_{eff}/\pi)}{M_0/\pi^2}\right|\right|$$



Arovas and Auerbach, Phys. Rev. B **38**, 316 1998.

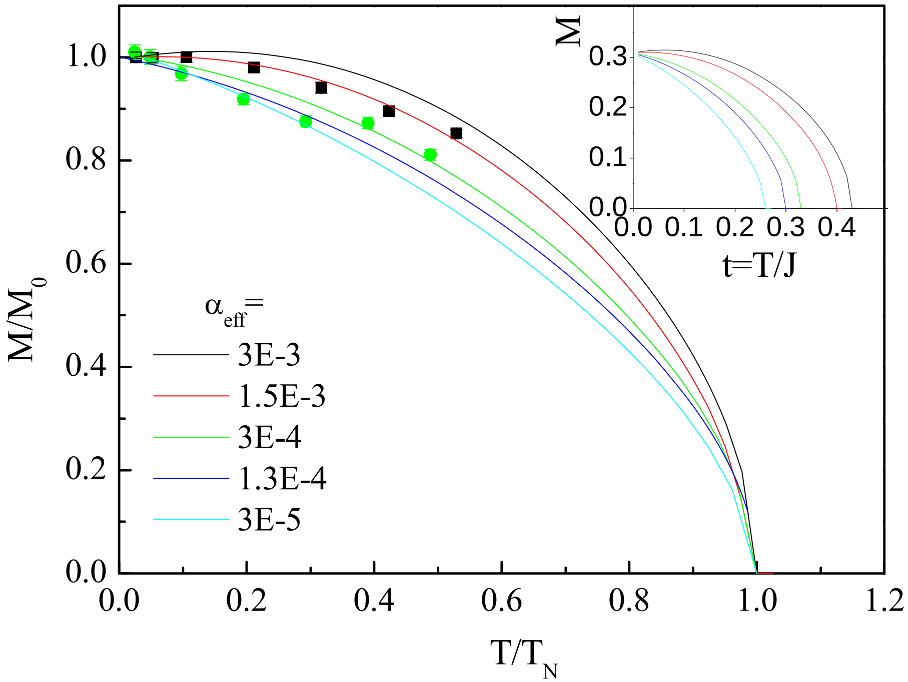
M(T)

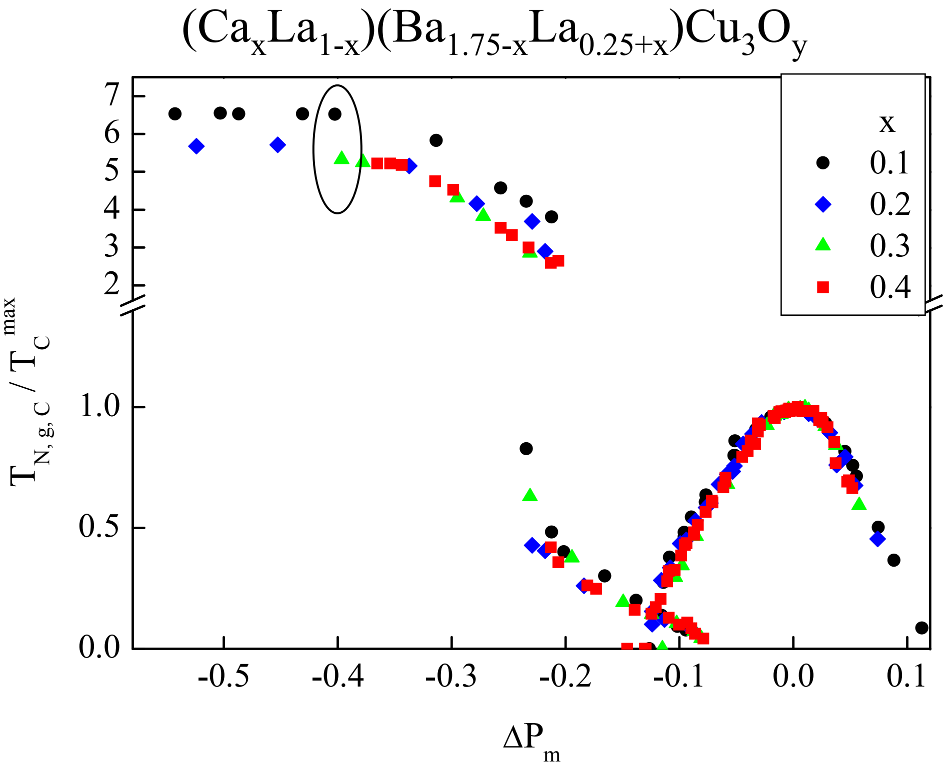


$$\omega \propto M$$

$$\frac{\omega(T)}{\omega(0)} = \frac{M(T)}{M_0}$$

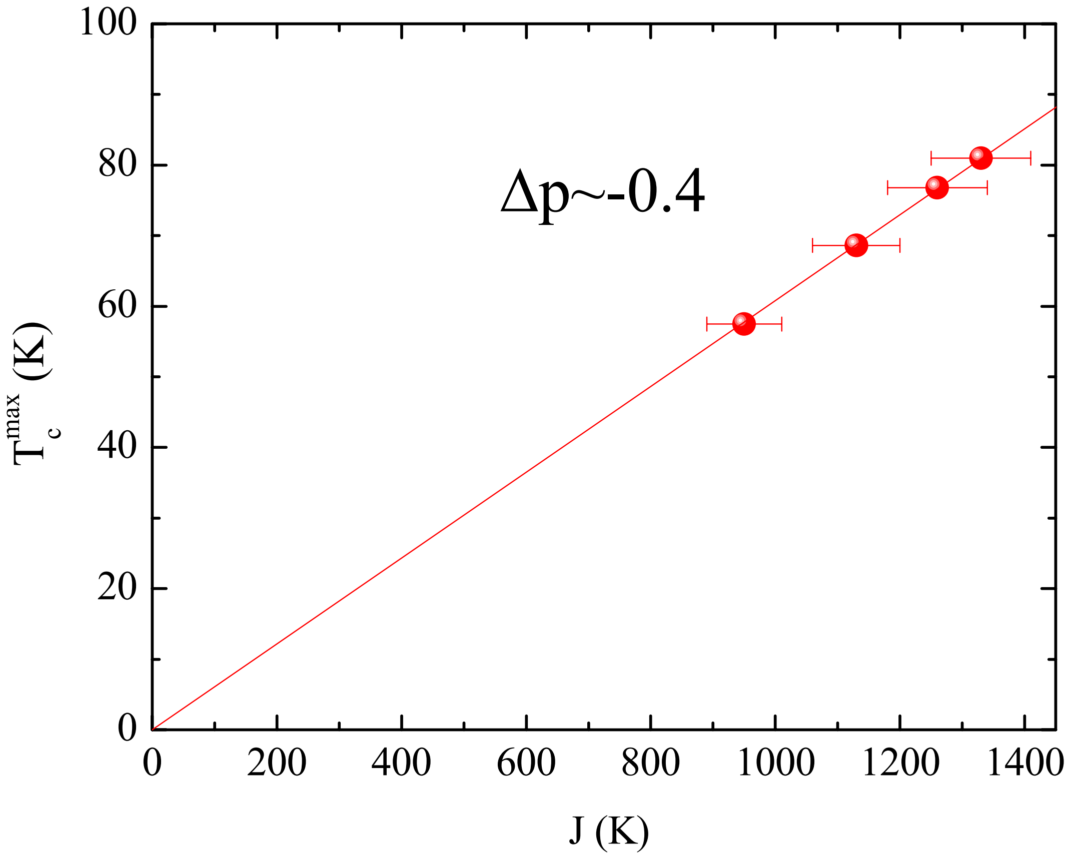
Extracting the anisotropies





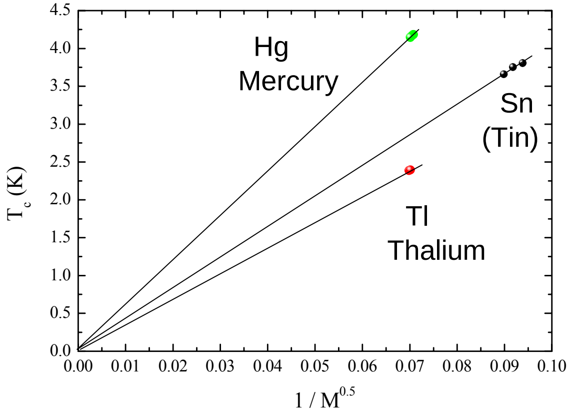
$$J = \frac{T_N}{t_N(\alpha_{eff})}$$

T_c^{max} versus J



} Theory free.

$$T_c^{max} \propto J$$

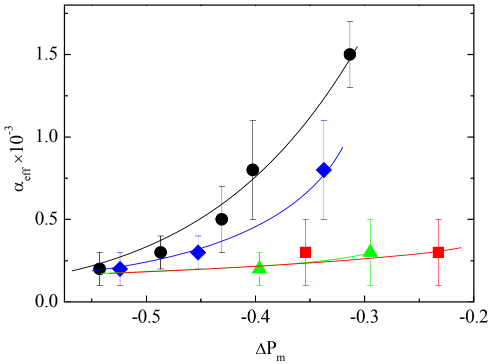


Unified Phase Diagram

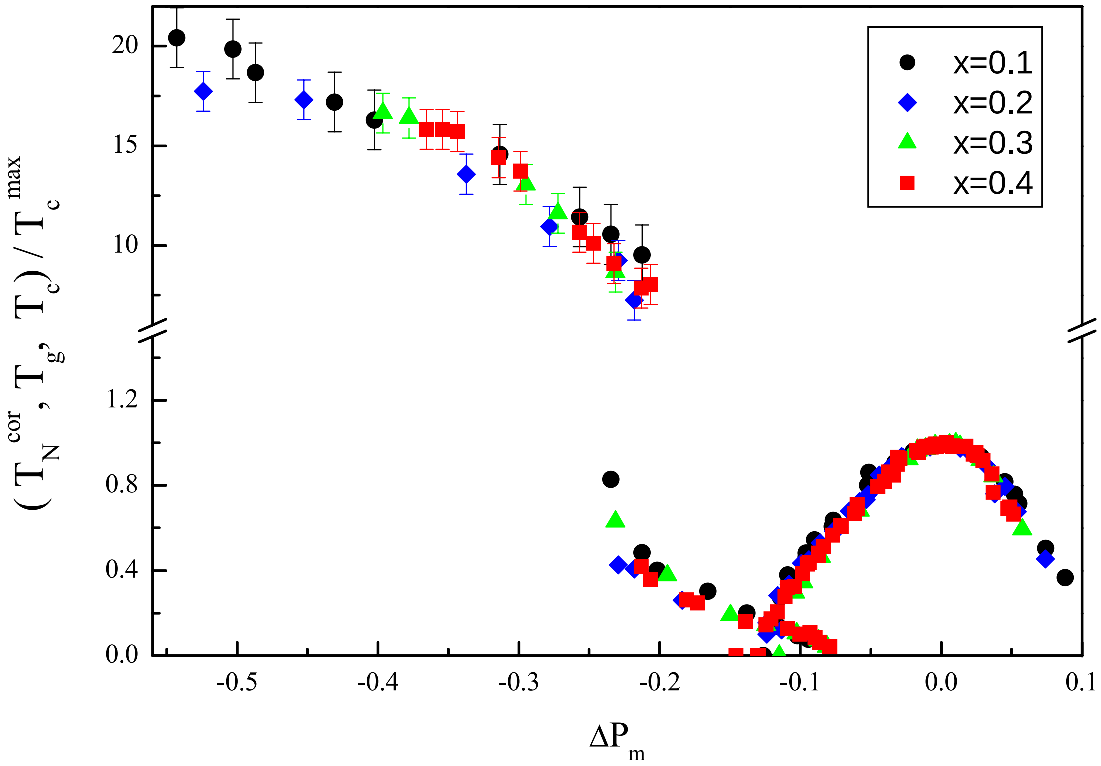
Ofer et al. PRB **74**, 220508(R) 2006

$$T_N^{cor} = \frac{T_N}{t_N(\alpha_{eff})}$$

$$\lim_{y \to 0} T_N^{cor} = J$$

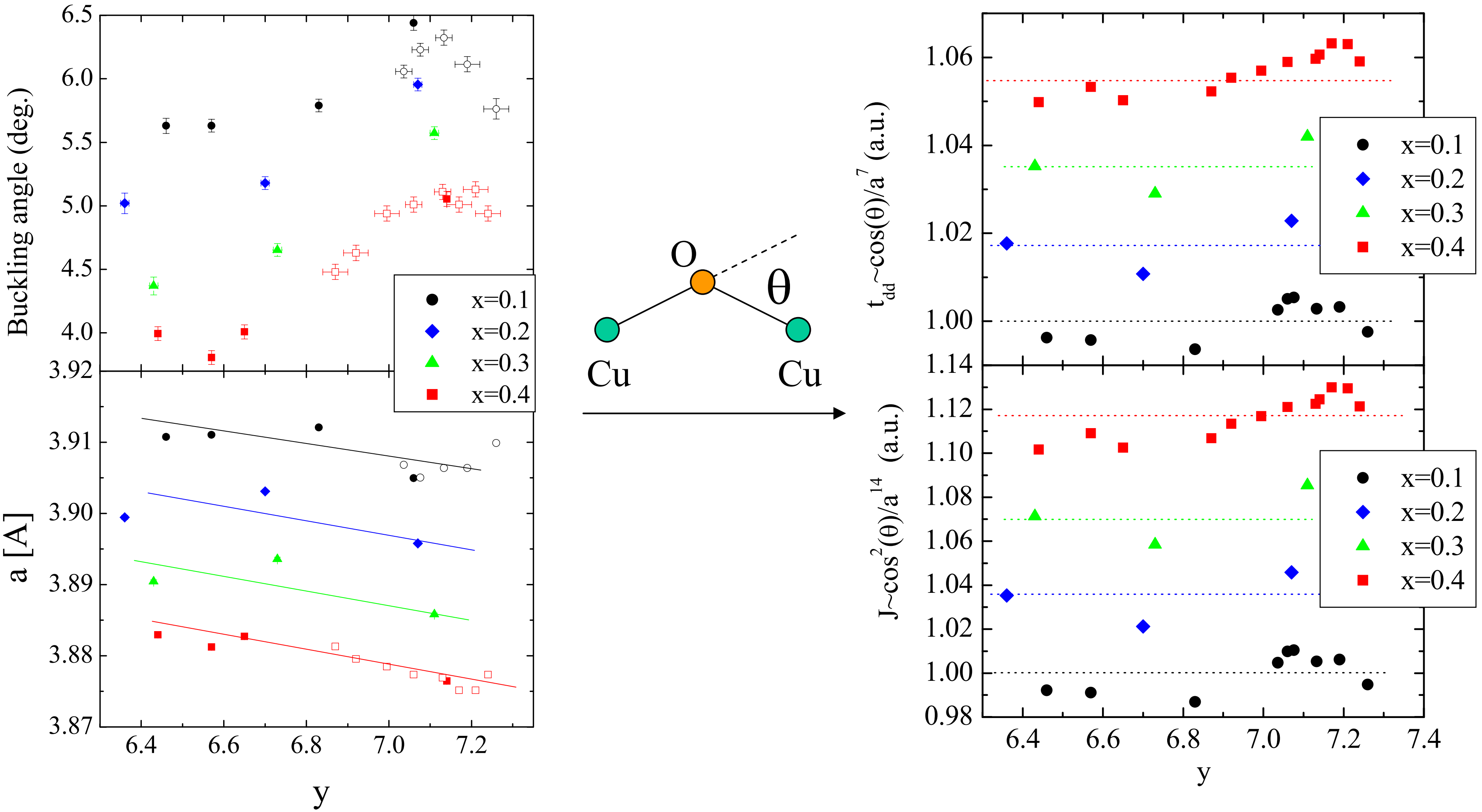


$(Ca_xLa_{1-x})(Ba_{1.75-x}La_{0.25+x})Cu_3O_y$



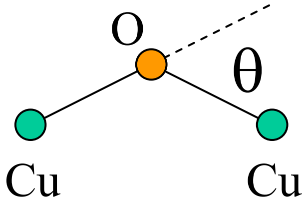
There is a common energy scale *J* for AFM, SG, and SC.

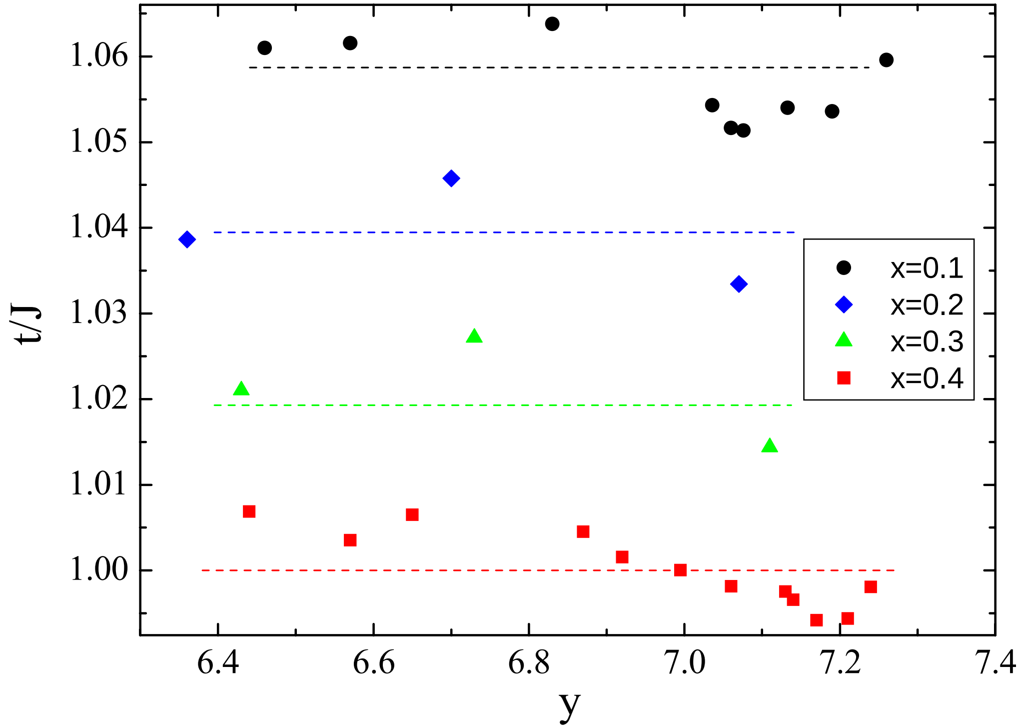
What does vary between families?



•J increases with x mainly as a consequence of decreasing buckling angle.

The implications

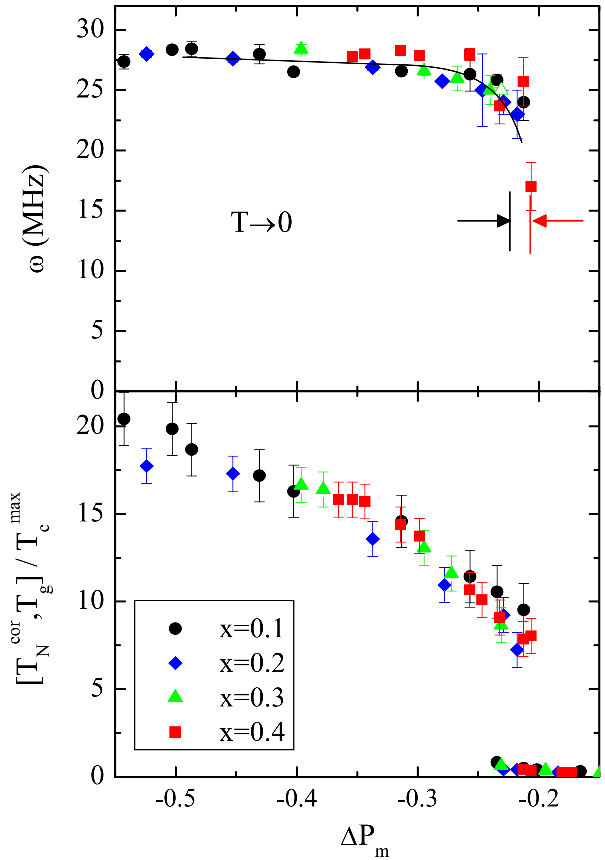


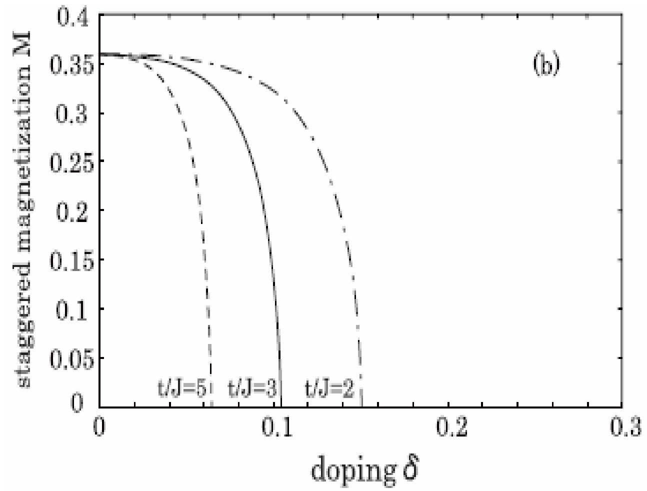


t/J varies by 6% between families.

Alterative estimation of *t/J*

t/J can be extracted from *M(T=0)* (namely ω) versus doping.



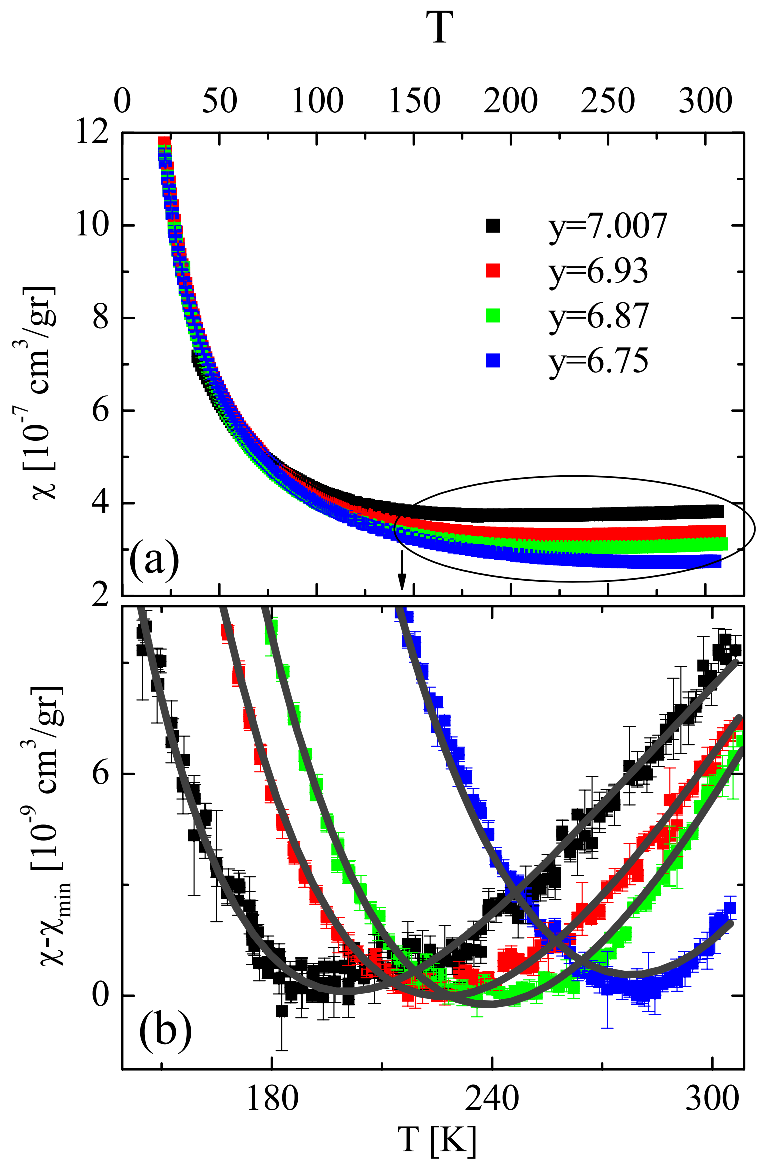


Yamamoto and Kurihara PRB 75, 134520 (2007).
Belkasri and Richard PRB 50, 12869 (1994).
Khaliullin and Horsch PRB 47, 463 (1993).

- Single electron hopping does NOT eliminate *M* upon doping.
- Could electron-pair (bosons) hopping destroy *M*?

What about the PG

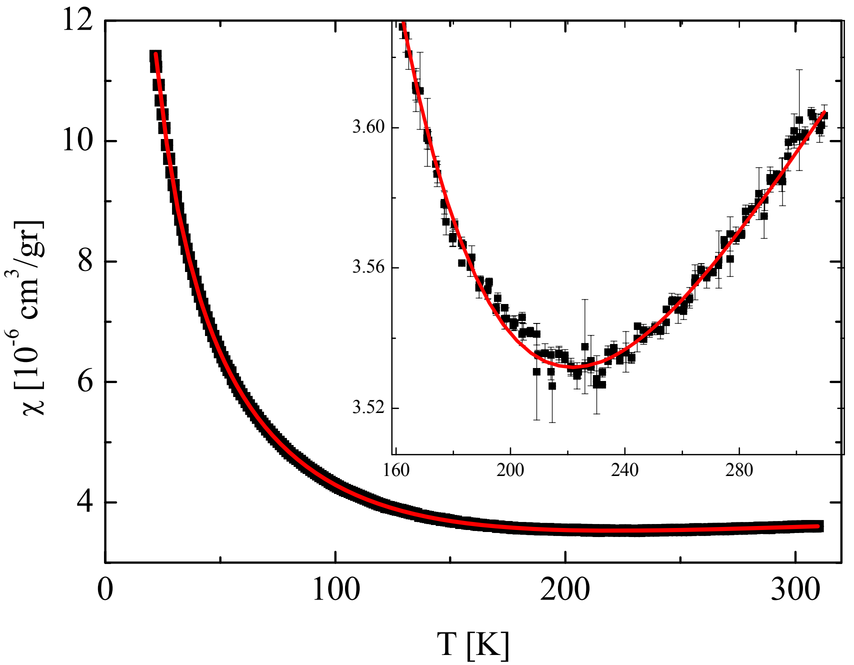
X=0.2

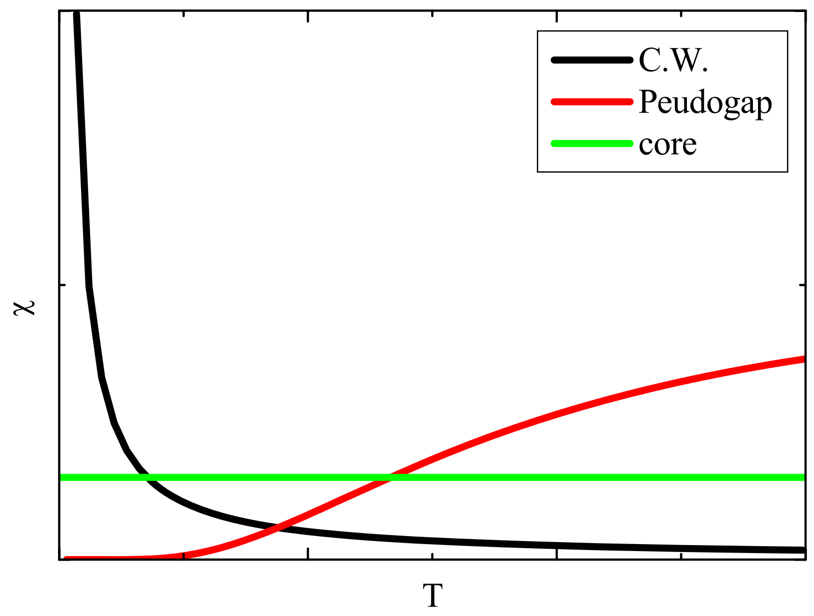


This phenomena was noticed by D. C. Johnston (1988).

The fitting function

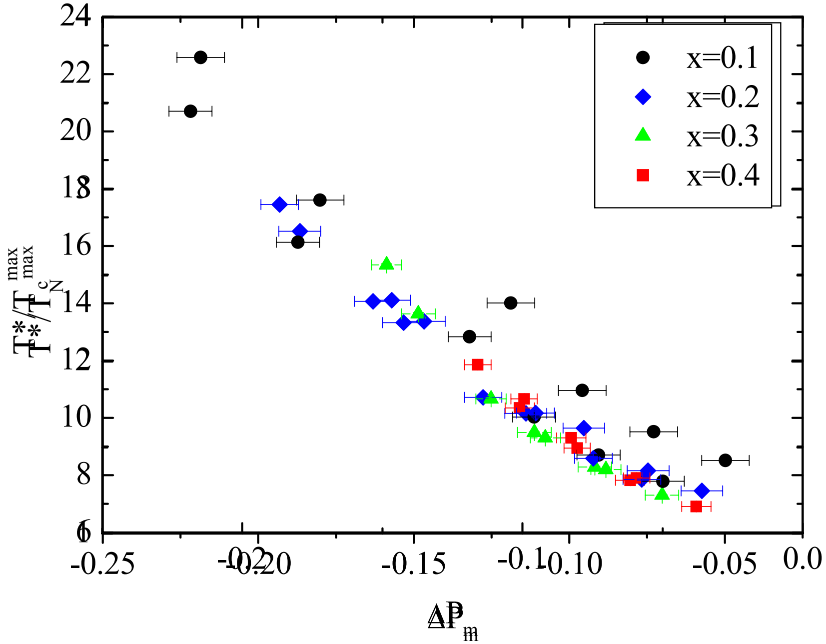
$$\chi_0 = \frac{C_1}{T+\theta} + \frac{C_2}{\cosh\left(\frac{T^*}{T}\right)} + C_3$$



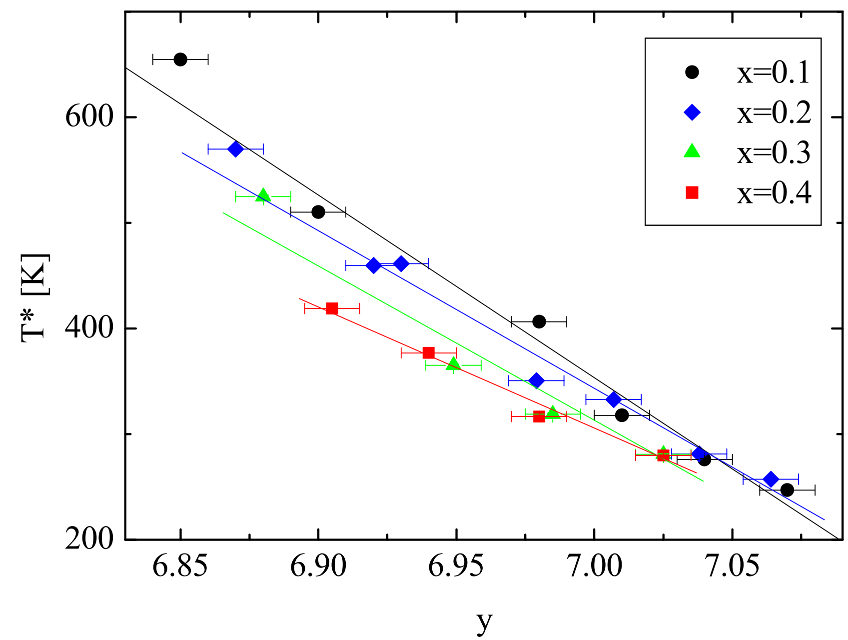


T*

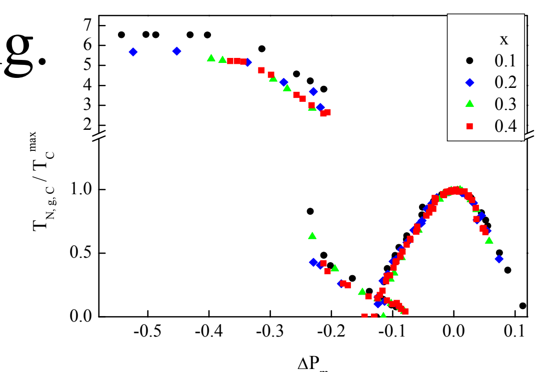
$$T^* \Rightarrow T^*/T_N^{max}$$



$$\Delta p_m = K_x(y - y_x^{opt})$$

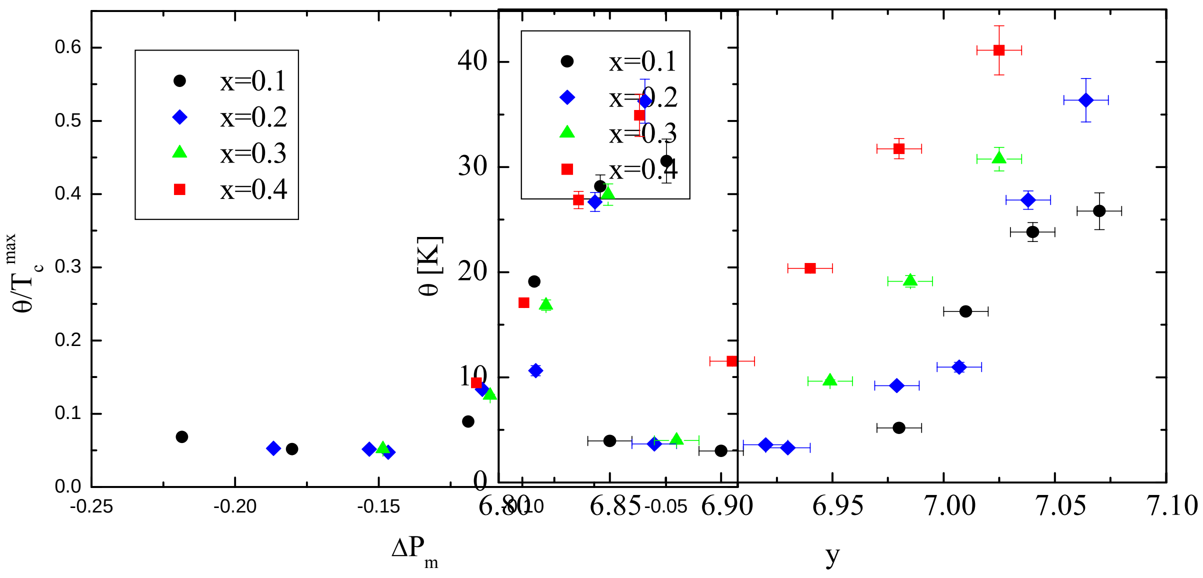


We find a similar problem as in the T_c/T_N scaling.



T^* scales with T_N.

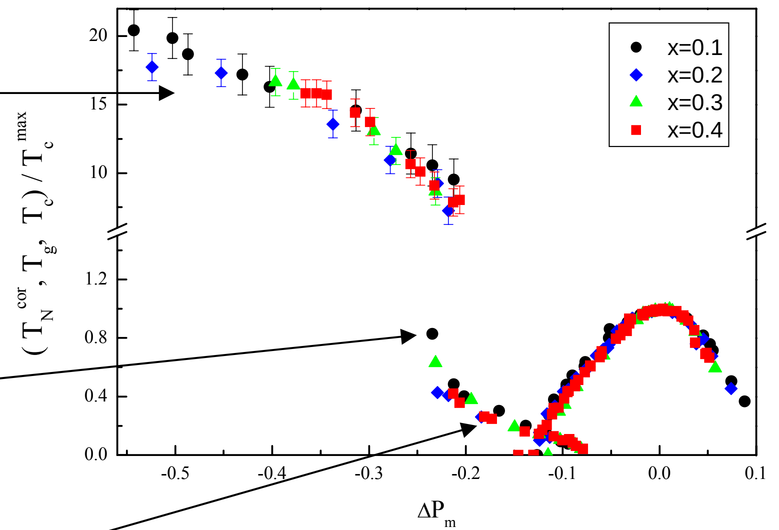
Curie-Weiss temperature



This is another indication that the relevant parameter is Δp_m.

Conclusions

$T_c \propto J$ and is a 2D phenomena.

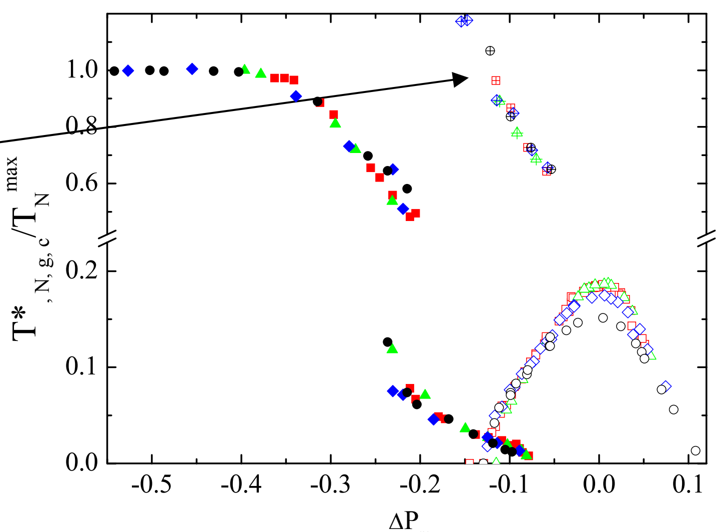


$H_{eff}(T \rightarrow 0)$ is not *t-J*.

Similar levels of disorder.

$$T^* \propto T_N = Jt(\alpha_{eff})$$

and is a 3D phenomena.



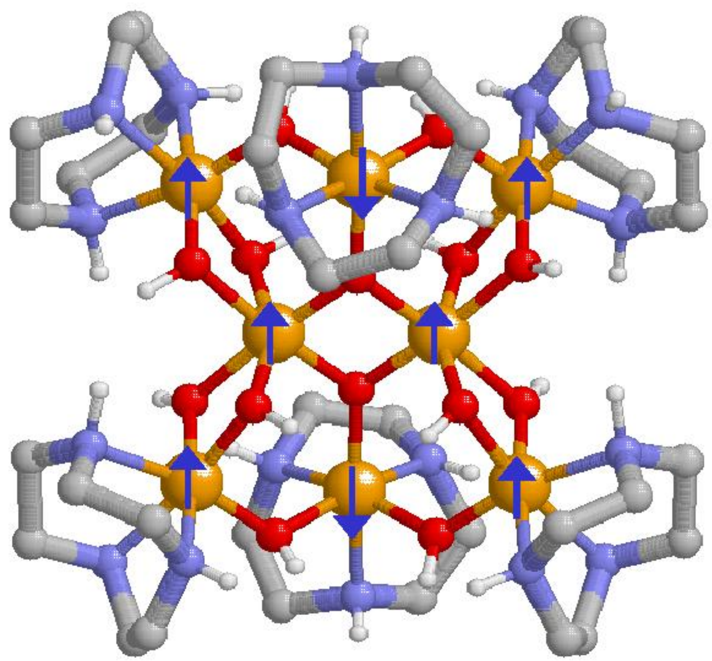
Open question

- What does *K* stands for?
- We know from NQR that it is not a doping efficiency parameter.

Experimental Estimate of Dephasing time in Molecular magnets

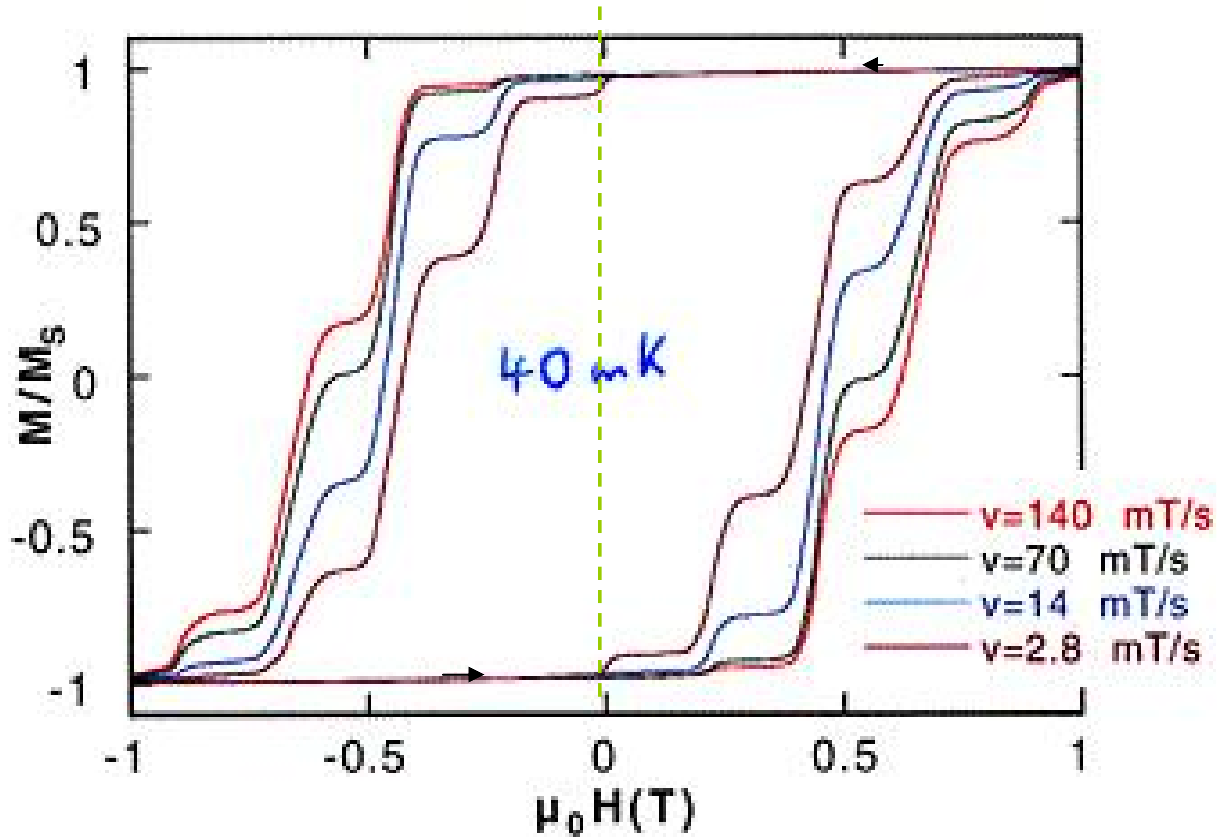
Ph.D. of Oren Shafir.
Collaborators : E. Shimshoni, V. Marvaud.

Fe8



$$S = 6\times\left(5/2\right) + 2\times\left(-5/2\right) = 10$$

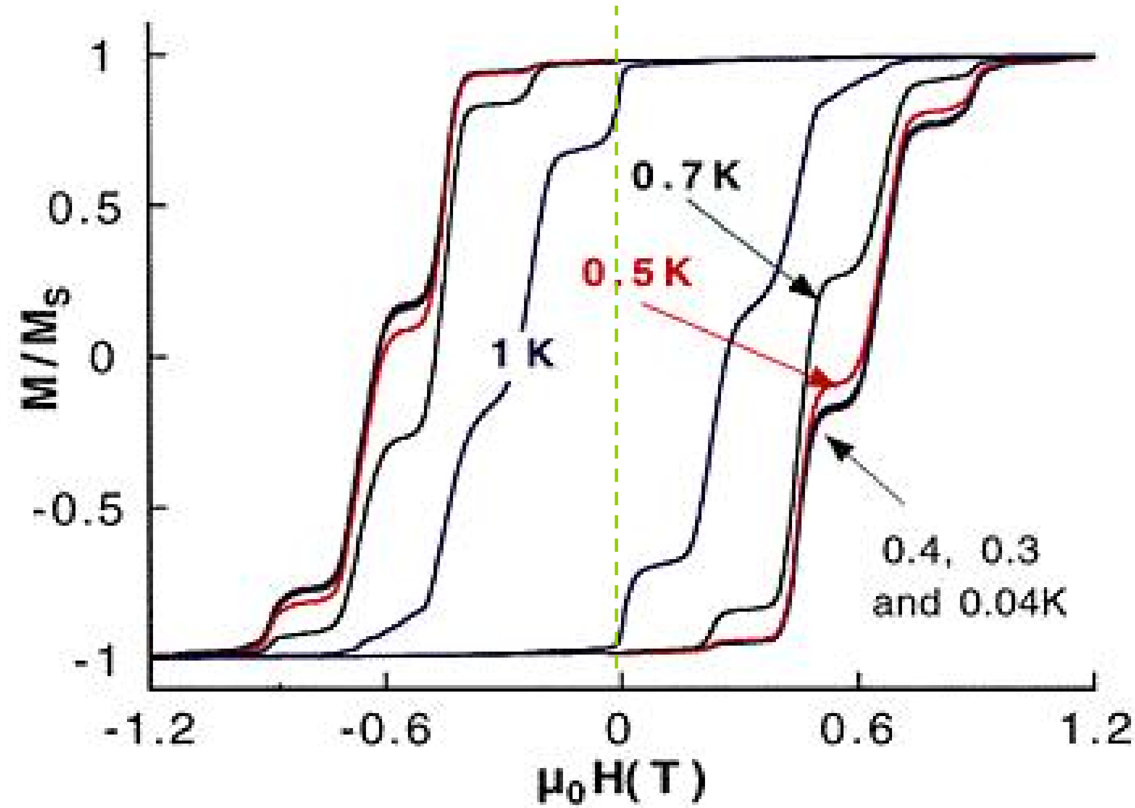
Introduction



A. Caneschi et al. JMMM 200, 182 (1999)

- Steps in the hysteresis loop are found at regular intervals of *H*, indicating magnetic quantum tunneling.

- The magnetization jumps are sweep rate dependent.



A. Caneschi et al. JMMM 200, 182 (1999)

- Below 300mK the step size is *T* independent, meaning pure tunneling.

Theoretical step size calculation

The Landau Zener model

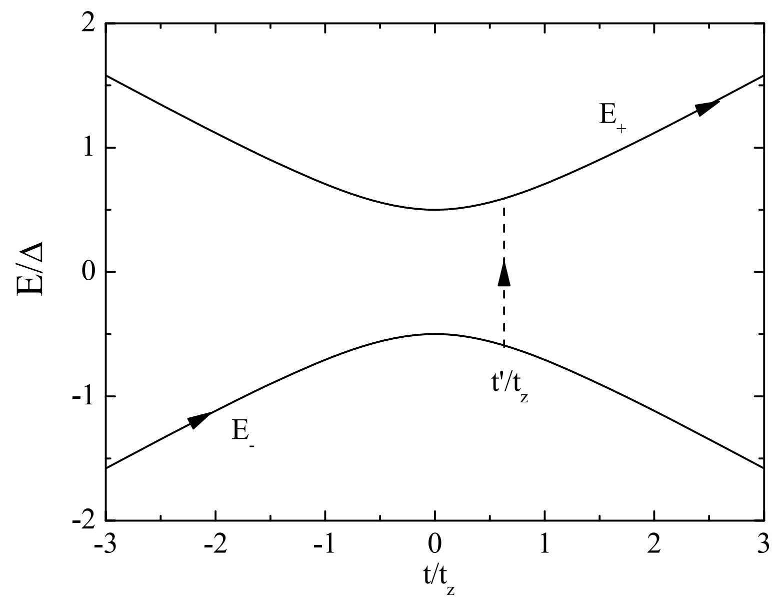
$$H = \alpha t S_z + \Delta S_x$$

where α is sweep rate, Δ is tunnel splitting.

The probability P that a spin flips between energy states is

$$P = \left| \langle + | U | + \rangle \right|^2 = \exp\left(-\pi \frac{\Delta^2}{\hbar \alpha} \right).$$

Where U is the time propagation operator.



Stochastic LZ model*

$$H = \alpha t S_z + \Delta S_x + \mathbf{B}(t)\mathbf{AS}$$

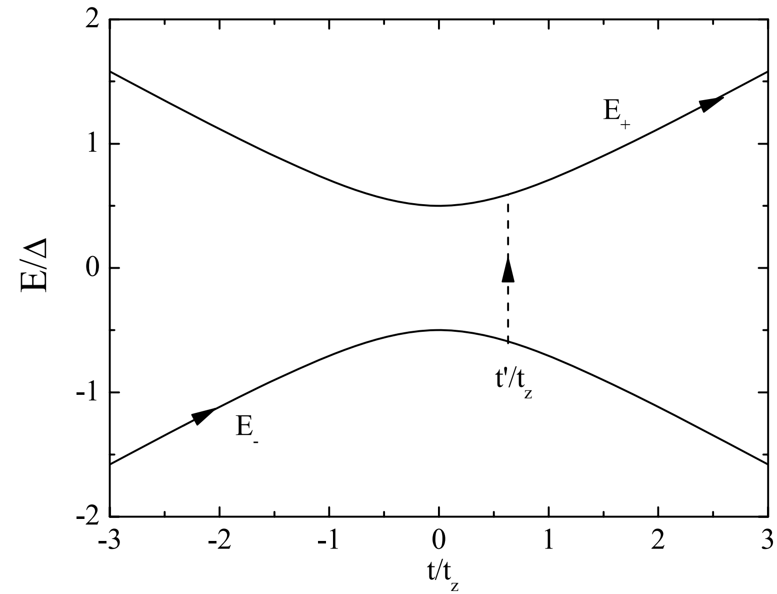
where **A** sets the symmetry of the noise field **B**(t) coupling, and

$$\left\langle \mathbf{B}(t)\mathbf{B}(0) \right\rangle = \left\langle B^2 \right\rangle \exp(-t/\tau_c).$$

There are 4 different time scales:

$$t_T = \frac{\hbar}{\Delta},\ t_z = \frac{\Delta}{\alpha},\ \tau_c,\ \tau_\varphi = \frac{\hbar^2}{\left\langle B^2 \right\rangle \tau_c},$$

and 12 possible theories (not including **A** variations).



For long/short dephasing time τ_φ one should sum up transition amplitude/probability respectively.

Estimating the dephasing time could be very useful.

* Shimshoni, Gefen, and Stern

Our strategy

• Dephasing is a property of the environment, it should be molecule independent.

• In all molecular magnets, the environment is made of a sea of protons.

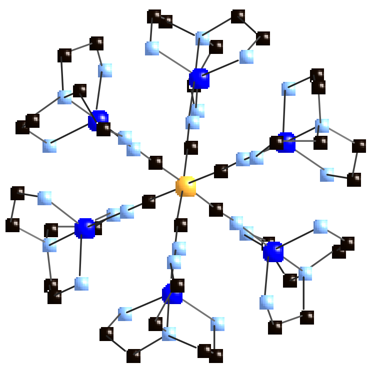
• Lets use isotropic molecule with no tunnel splitting, in zero field, to estimate τ_φ, namely,

$$H = \alpha t S_z + \Delta S_x + \mathbf{B}(t)\mathbf{A}\mathbf{S}$$

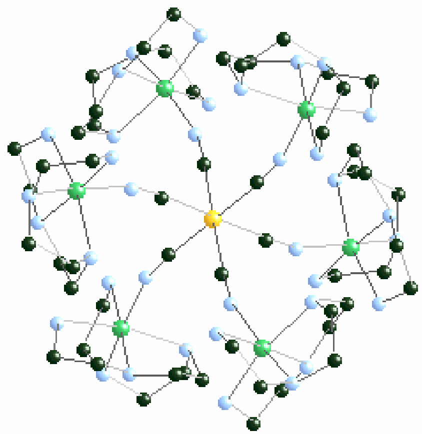
• In our experiment, the only variable is S, and we change it from 7/2 to 27/2.

• This is the first study of this kind.

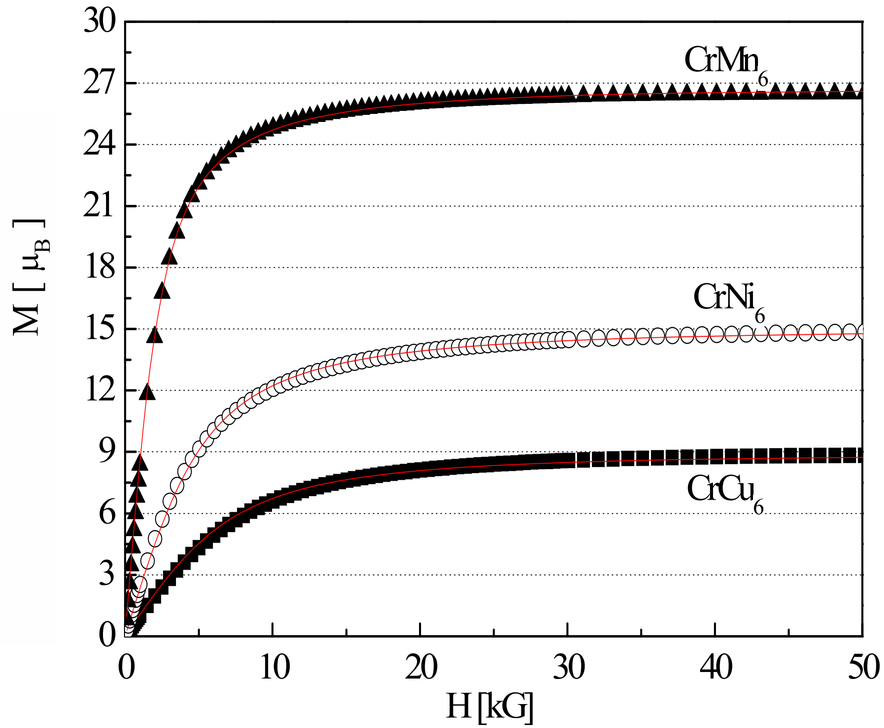
Our molecules



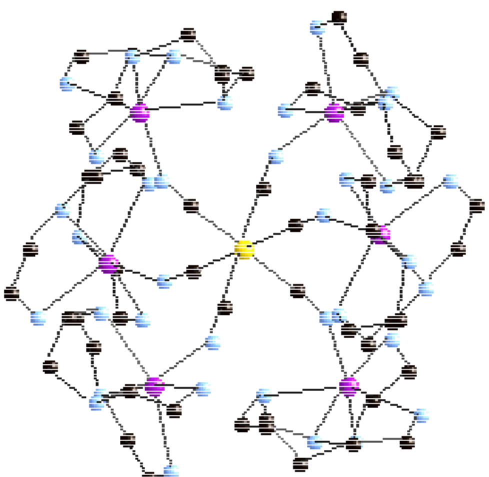
$CrCu_6$
$S=9/2$
$J_{Cr\text{-}Cu}=77K$
$S_{Cu}=1/2$



$CrNi_6$
$S=15/2$
$J_{Cr\text{-}Ni}=24K$
$S_{Ni}=1$



$CrMn_6$
$S=27/2$
$J_{Cr\text{-}Mn}=-11$
$S_{Mn}=5/2$

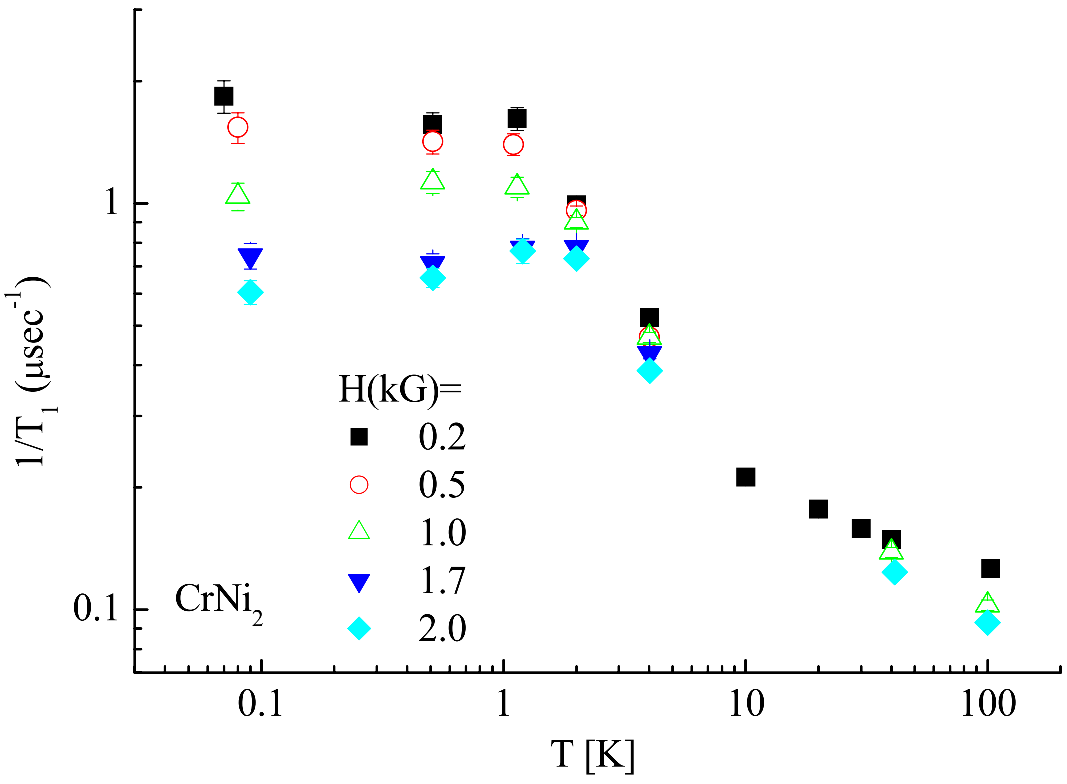


The dynamic measurement

- Spin lattice relaxation of a muon varies with *T* and *H*.

- The muon T_1 is set by τ_φ of the molecules.

- No *T* dependence at low *T*.

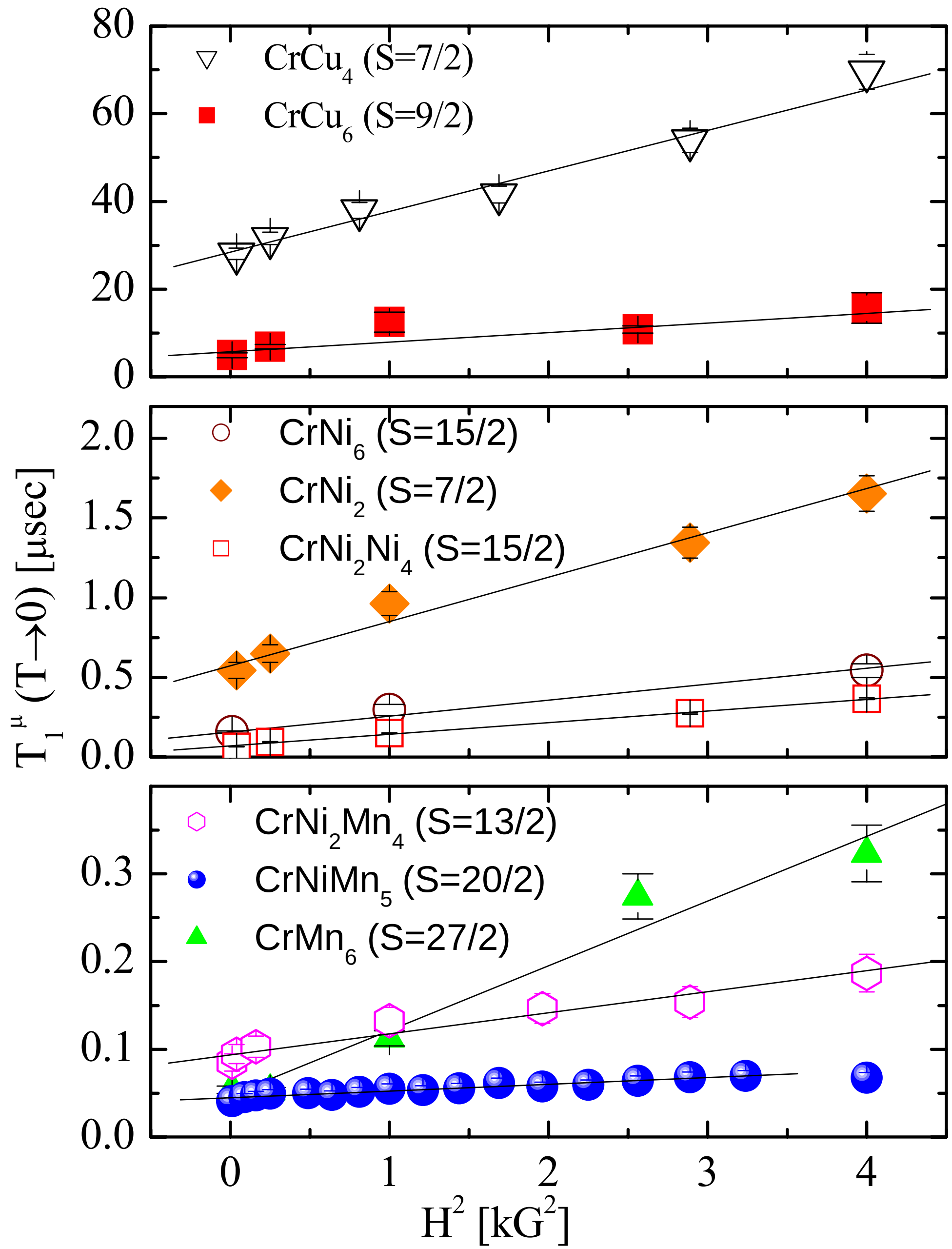


T_1 at 100mK

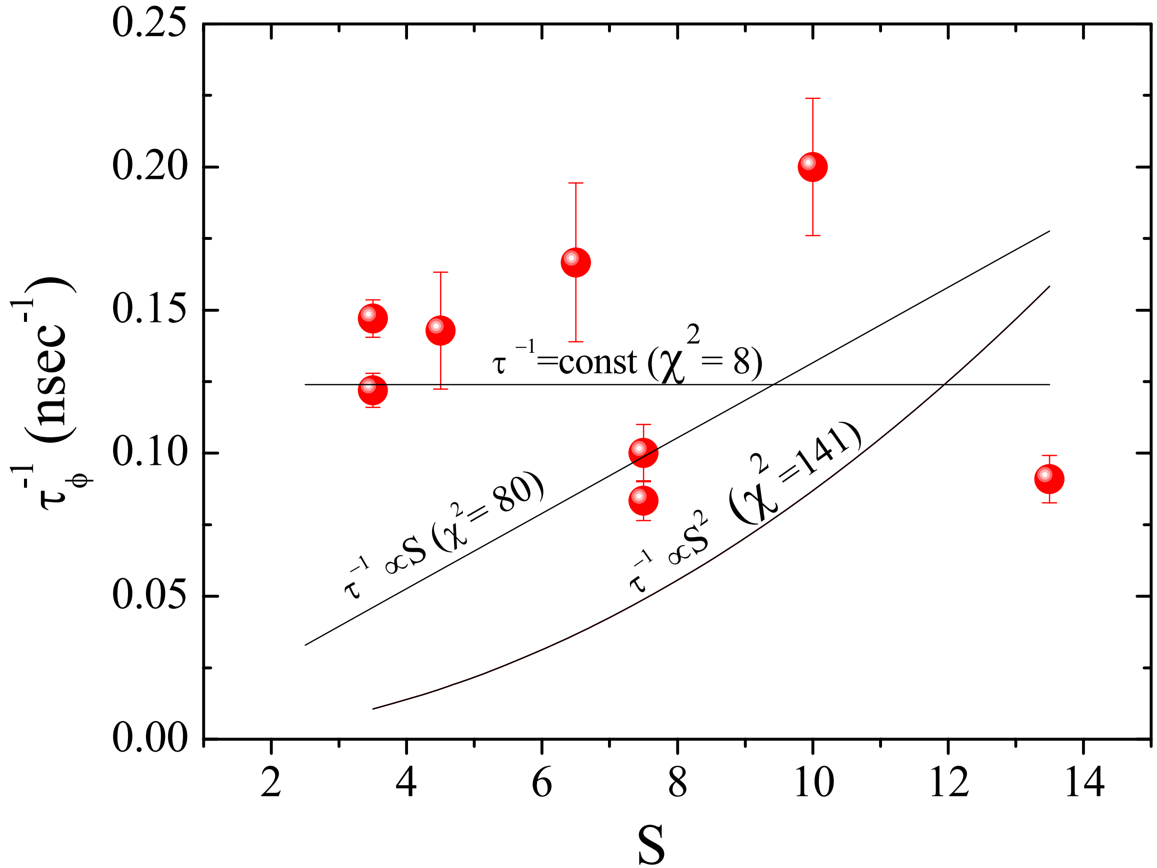
• T_1 varies considerably between samples.

• We can determine τ_φ from

$$\frac{1}{T_1} \propto \frac{\tau_\varphi}{1+(\gamma H \tau_\varphi)^2}.$$



Our major finding



Keren et al. PRL **98,** 257204 (2007)

The data is best explained by τ_φ(S)=const.

Conclusions

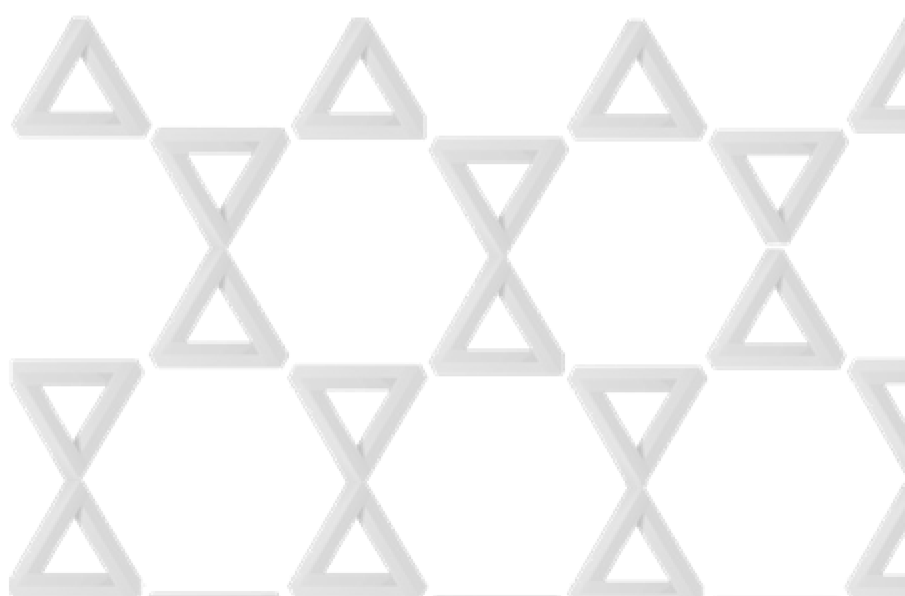
- τ_φ is on the order of 10^{-8} s and is the shortest time scale in the problem of Fe8 where $t_T \sim 10^{-4}$ s, $t_T > t_z$, and $\tau_c \sim 10^{-6}$ s.

- This case was not examined theoretically.

- The S independence of τ_φ is consistent with a nuclear origin for the stochastic field B.

Ground state and excitation properties of Herbertsmithite $ZnCu_3(OH)_6Cl_2$

Ph.D. Oren Ofer
Collaborators: Emily A. Nytko, and Daniel D. Nocera [MIT, USA]

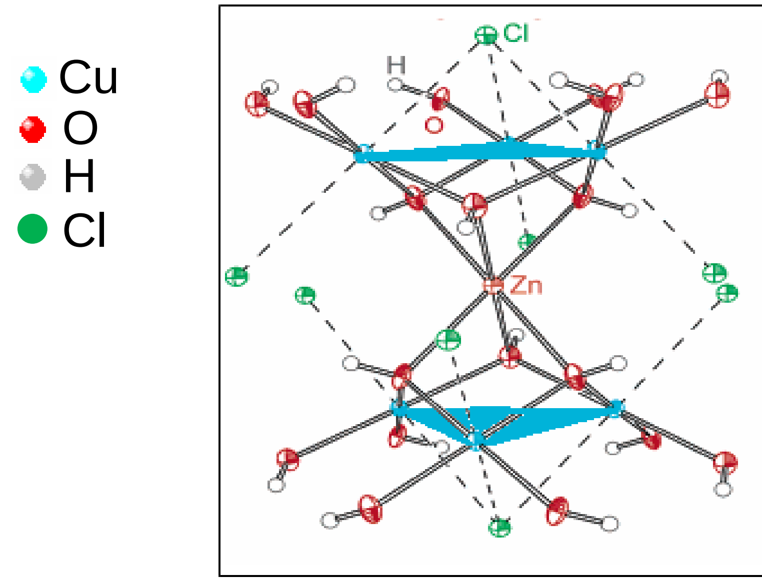


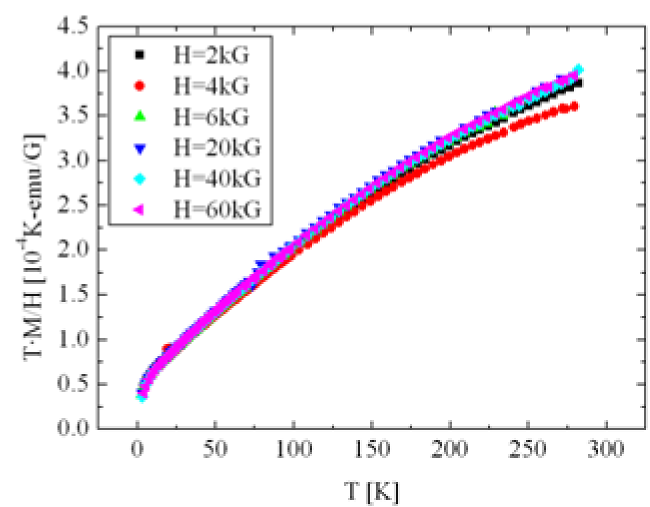
S=½ Kagomé Ground State?

- Do S=½ spins freeze on the kagomé lattice?
- Is the ground state magnetic?
- What can be said about the density of excited states?
- Is there a gap in the spin energy spectra?
- Does the lattice distort to accommodate a spin-Peierls state?

Herbertsmithite $ZnCu_3(OH)_6Cl_2$

- XRD shows perfect S=1/2 kagomé.
- $q_{CW} = -\ 314K$ [1].
- No magnetic order down to 1.2K was found.
- Field-independent M/H at high fields (useful later).
- Growth of χ slower than $1/T$.

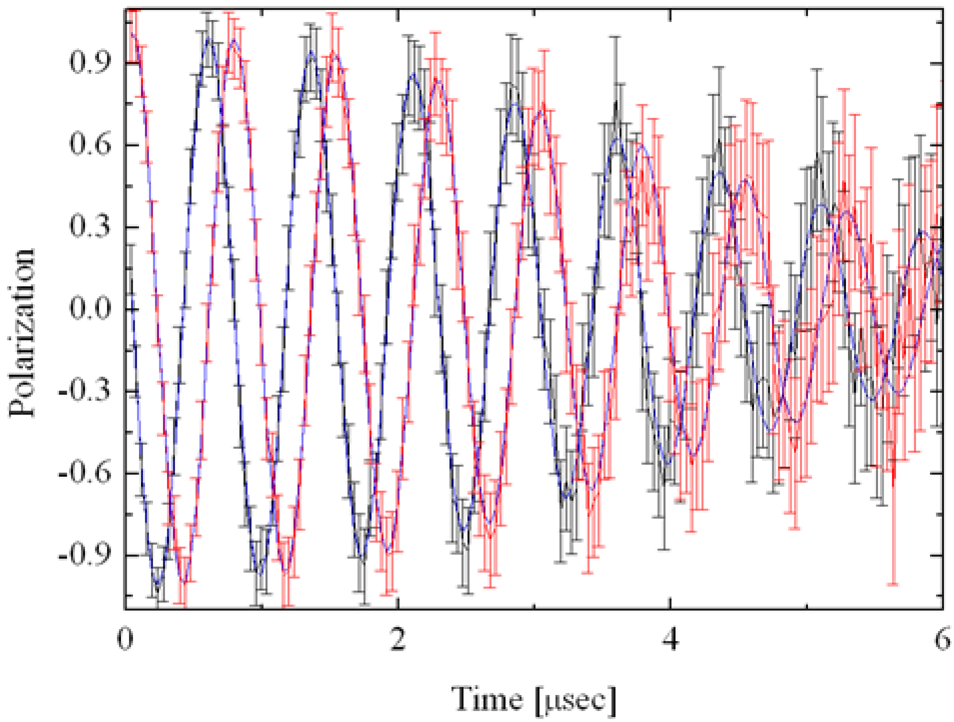




[1] M. P. Shores *et al., J. Am. Chem. Soc.*, ***127***, *13462 (2005*)

Raw μSR Data

- We measure rotation frequency and relaxation of the muon spin.
- The frequency shift is a result of the sample magnetization.
- The transverse-field relaxation is a result of static field inhomogeneities.

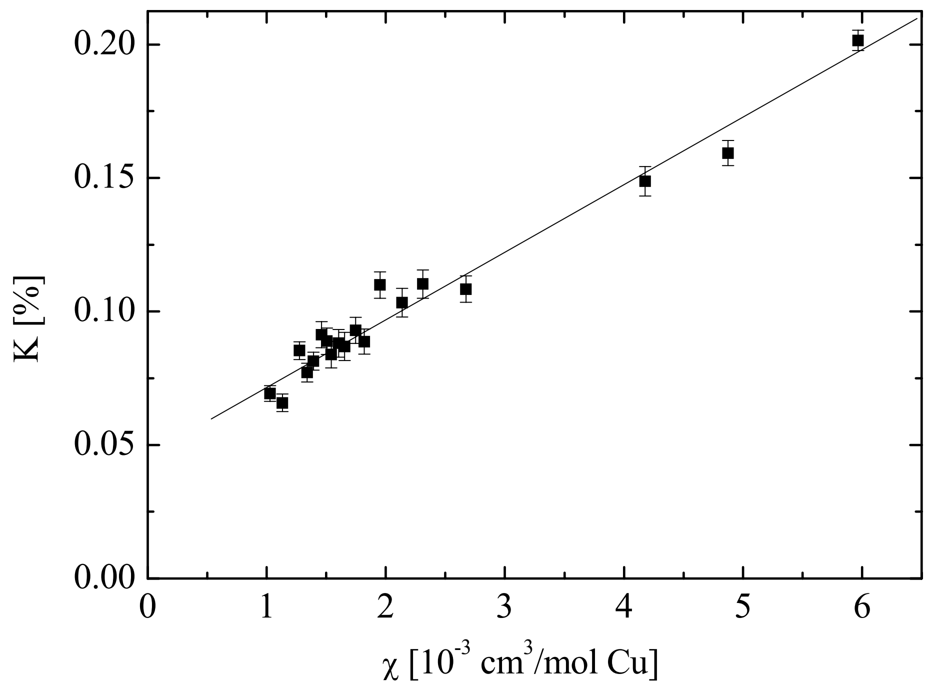


We fit $P_{TF} = A_{TF} \exp\left(-\,(t / T_2^*)^2\right)\cos(wt)$ to the data in the RRF.

The shift, $K = \dfrac{w - w_0}{w_0}$.

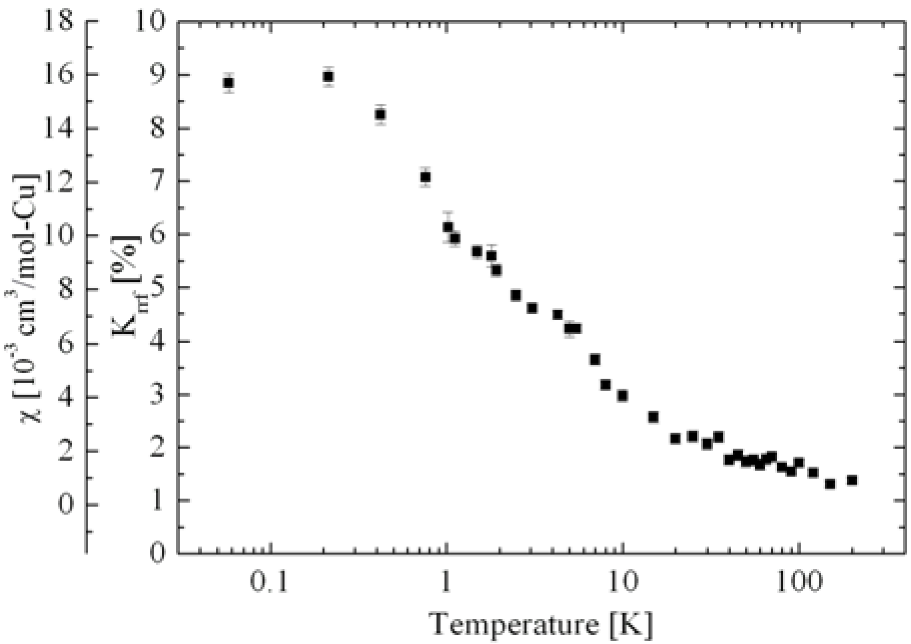
Shift Calibration

- χ and *K* show a linear relation.
- This calibration allows determination of χ from the muon frequency shift, *K*.

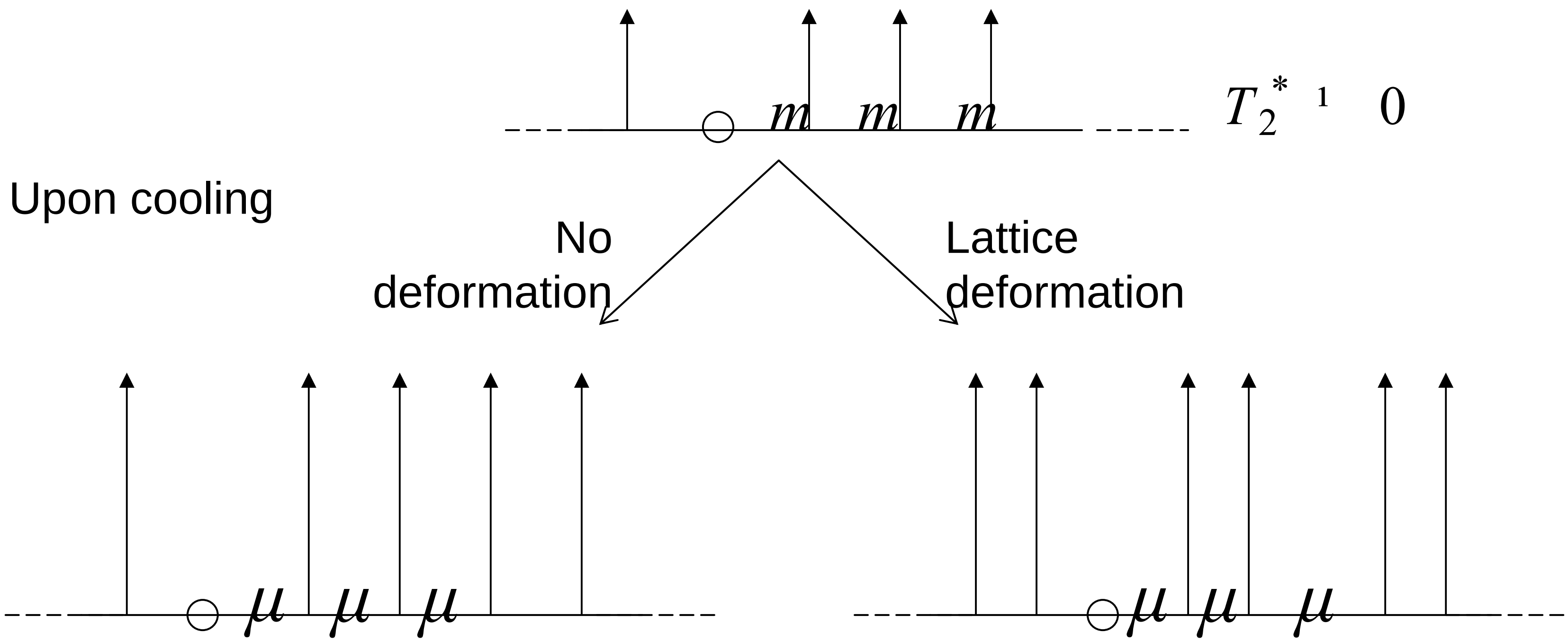


μSR Data

- We measured *K* hence χ down to 60*mK*.
- We found saturation of χ at *T~200mK*.
- As with the susceptibility, μSR shows no spin freezing, or long range order.



$T_2{}^*$ Interpretation



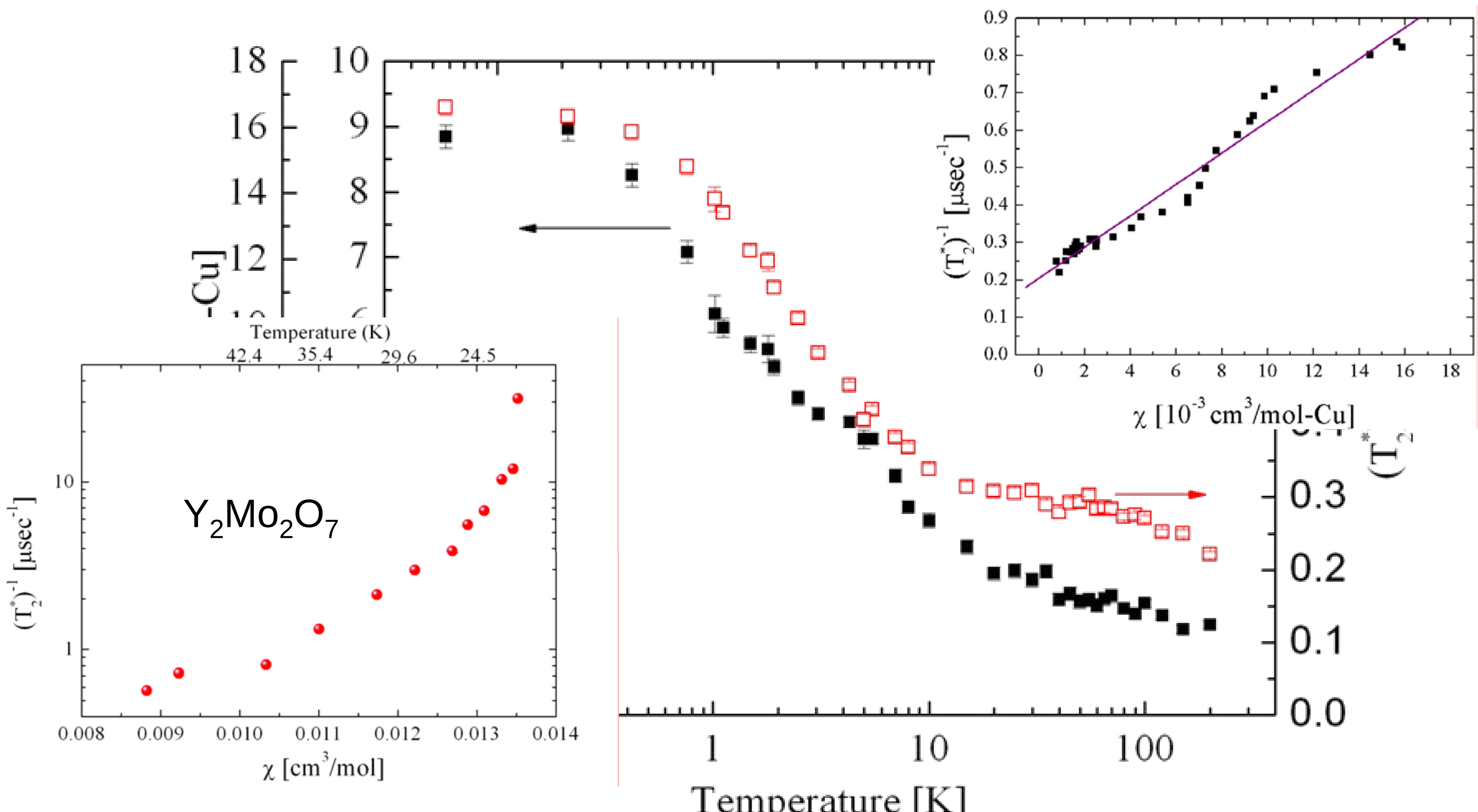
$$\frac{1}{T_2^*}(T) = C \times c(T)$$

$$\frac{1}{T_2^*}(T) \neq C \times c(T)$$

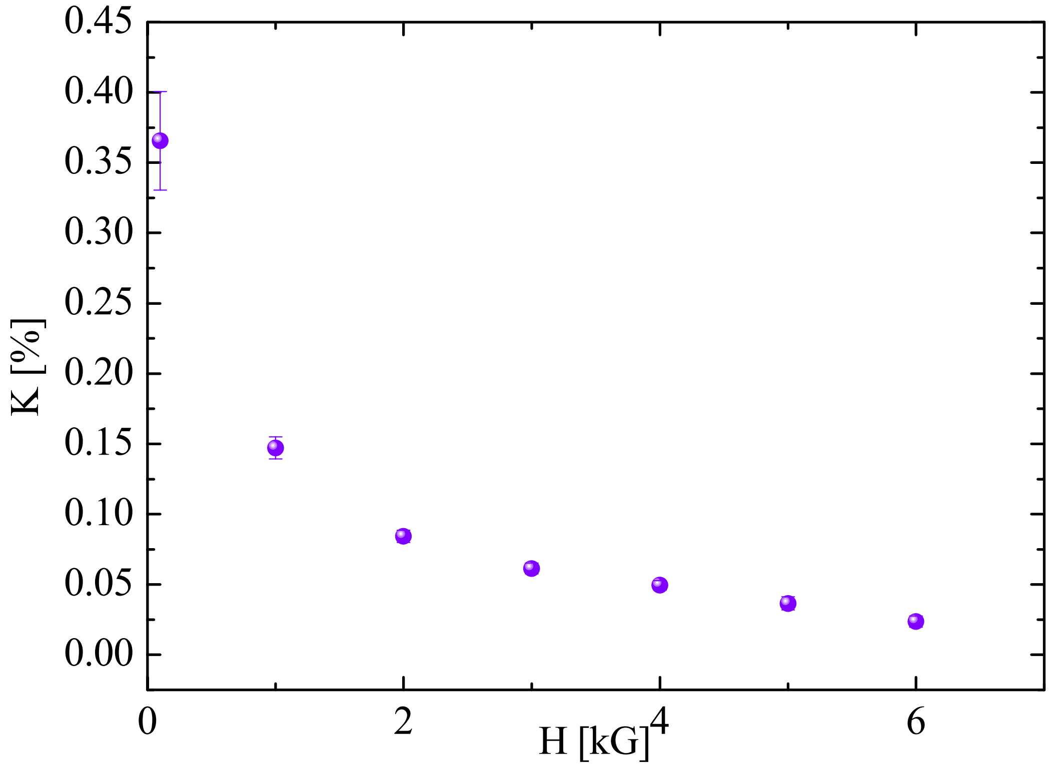
Additional relaxation mechanism.

Absence of Spin-Peierls

- $(T^*_2)^{-1}$ and χ behave similarly.



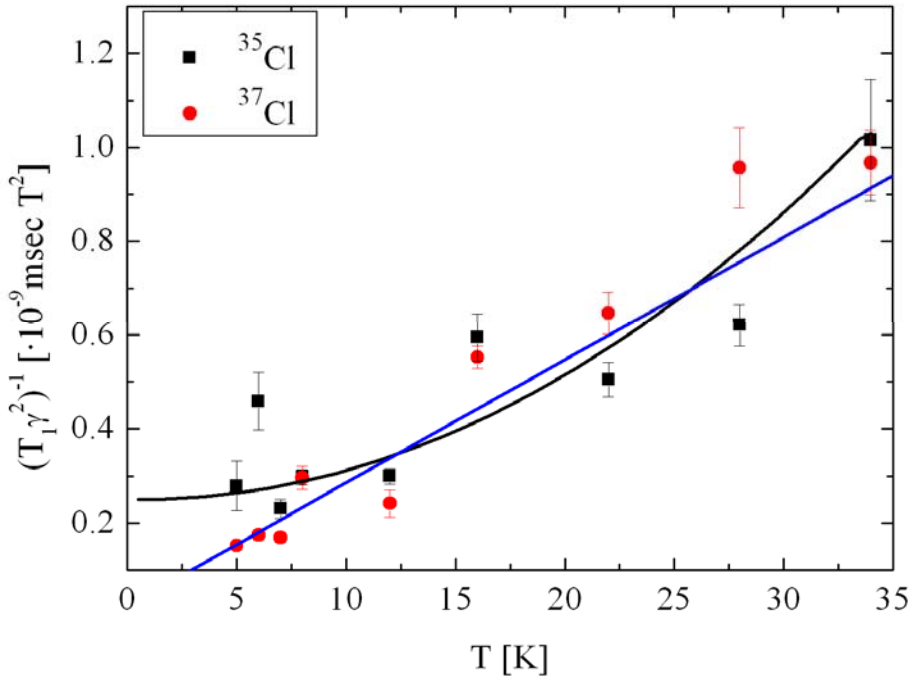
Field dependence of K



The field dependence is not clear!

$^{35,37}Cl\ T_1$ By NMR

- Muon T_1 is longer than muon life-time.
- Powdered Sample.
- Cl T_1 increases down to T~50K and slowly decreases.
- The decrease of T_1^{-1} with decreasing T is slow=gapless.



Spin lattice relaxation can be interpreted by Bosonic excitations: two magnon (or any Boson) Raman scattering,

$$(T_1)^{-1} = A\gamma^2 \int_{\Delta}^{\infty} \rho^2(E) \cdot n(E) \cdot [n(E)+1] dE \Rightarrow \begin{array}{l} \Delta = 0.5(2)K \\ \alpha = 0.23(1) \end{array}.$$

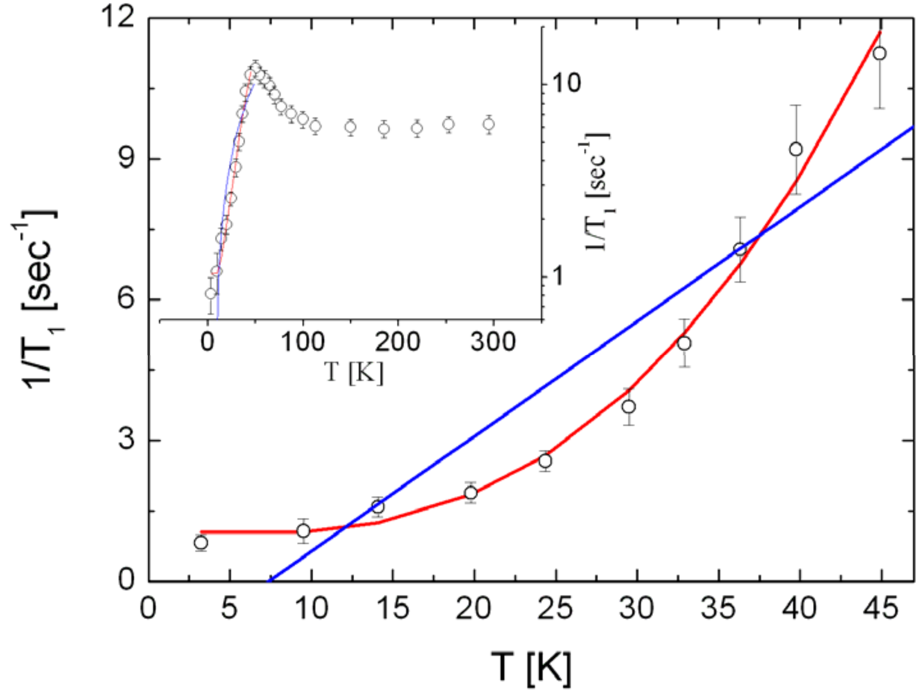
$$\rho(E) \propto E^{\alpha}$$

Imai's data

- Oriented Powder and higher field.

- With this data,

Δ=0.002(3) K
α=0.59(3)



$$E^{a}$$

$$(T_1)^{-1} = A g^2 \int_{\Delta}^{\infty} \rho^2(E) \cdot n(E) \cdot [n(E) + 1] \, dE$$

T. Imai *et al.*, cond-mat/0703141 (2007)

Summary

- Do S=½ spins freeze on the kagomé lattice?
- The spins continue to fluctuate down to ~60*mK*.
- Is the ground state magnetic?
- Saturation of χ meaning no phase transition or singlet formation.
- What can be said about the density of excited states?
- The density of states $r \sim E^{1/2}$.
- Is there a gap in the spin energy spectra?
- Negligible or no gap.
- Does the lattice distort to accommodate a spin-Peierls state?
- $(T_2^*)^{-1}$ scales with χ meaning no evidence of spin-Peierls distortion.