NMR studies of cuprates
pseudogap, correlations, phase diagram: past and future?

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NMR
LPS, Orsay

Transport,
SPEC (CEA, Saclay)
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,J.F. Marucco , V. Viallet

Single Crystal Samples:
P. Lejay and D. Colson

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Observation = Amazing Phenomena

H. Alloul , Introduction to the Physics of Electrons in Solids
Editions de l'Ecole polytechnique
English edition , Springer (to appear , december 2010)

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NMR studies of cuprates :
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- ***Magnetic spin susceptibilities in NMR :***
  - **Usual metals and superconductors**
  - **The case of cuprates:**
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  - **Single spin fluid in the normal state**
- *Dynamic susceptibilities and spin lattice relaxation :*
  - Magnetic correlations in the phase diagram
  - d- wave SC
- *The pseudogap and questions on the phase diagram*
  - NMR as a local magnetic probe
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  - Some answers about the phase diagram

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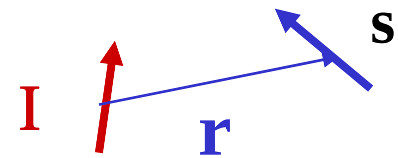
Hyperfine Intercations - NMR Frequency Shifts

Interactions between nuclear moments *I* and electronic moments *s* et *l*



Dipolar

$$H_{dd} = -\frac{\hbar^2 \gamma_n \gamma_e}{r^3}\left\{\vec{I}.\vec{s} - 3\frac{(\vec{I}.\vec{r})(\vec{s}.\vec{r})}{r^2}\right\}$$

Orbital

$$H_{orb} = -\frac{\hbar^2 \gamma_n \gamma_e}{r^3}\ \vec{I}.\vec{l}$$

Contact

$$H_c = \frac{8\pi}{3}\hbar^2 \gamma_n \gamma_e\ \vec{I}.\vec{s}\ \delta(\vec{r})$$

- **Filled atomic shells :**

$$H_{orb} \equiv 0\ ;\ H_{dd} \equiv 0$$

- **Paramagnetic or diamagnetic compounds:**

$$H_T = H_Z + H_{dd} + H_{orb} + H_c = -\hbar\gamma_n\ \vec{I}.(\vec{B}_0 + \vec{B}_L)$$

$$\vec{B}_L = <\vec{B}_L> + \left[\vec{B}_L - <\vec{B}_L>\right]$$

Relaxation time

Mean field Linear response

$$<\vec{B}_L> \propto \chi B_0$$

Frequency shift

Local measurement of the electronic susceptibility

Insulators H_{orb}
Chemical shift
(orbital currents)

metals χ_{Pauli}
Knight shift
(unpaired electrons)

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Magnetic susceptibilities

SQUID Measures

always

Ion cores **orbital** **Spin**

$$\chi_\alpha^m(T) = \chi^{dia} + \chi_\alpha^{orb} + \chi_\alpha^s + (\chi^{imp})$$

$$= \chi^{dia} + \sum_i \left[\chi_{i,\alpha}^{orb} + \chi_{i,\alpha}^s(T)\right]$$

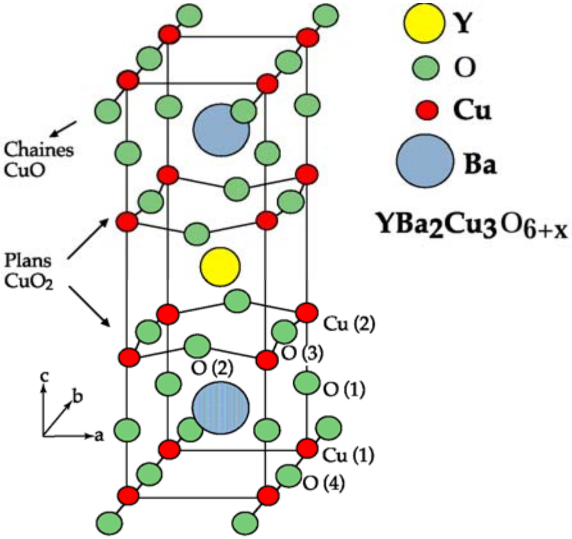
$\alpha = (x,y,z)$

i = atomic sites

NMR shift measures on each nuclear site i

$$K_{i,\alpha}(T) = K_i^{dia} + K_{i,\alpha}^{orb} + K_{i,\alpha}^s(T)$$

$$= K_i^{dia} + A_{i,\alpha}^{orb}\chi_{i,\alpha}^{orb} + A_{i,\alpha}^s\chi_{i,\alpha}^s(T)$$



Local magnetic measurement on each nuclear site i

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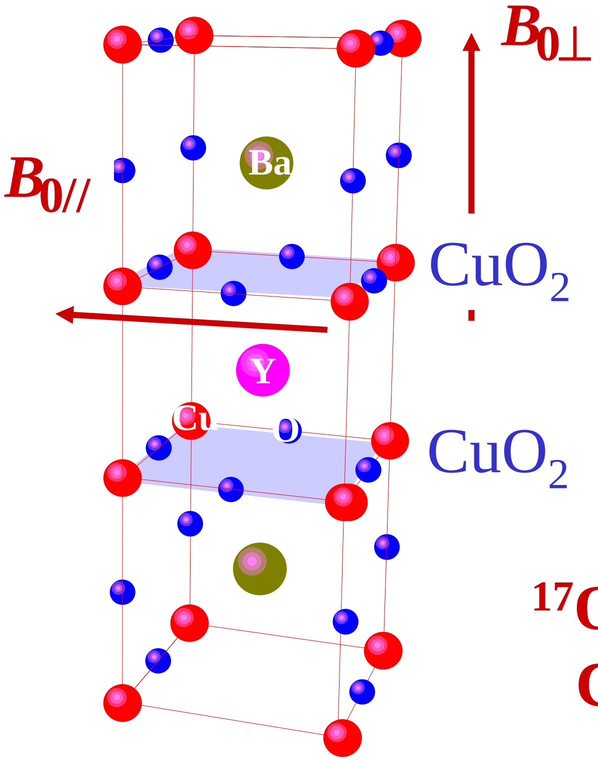
Knight Shift in Superconductors

High T_c cuprates

$YBCO_7$

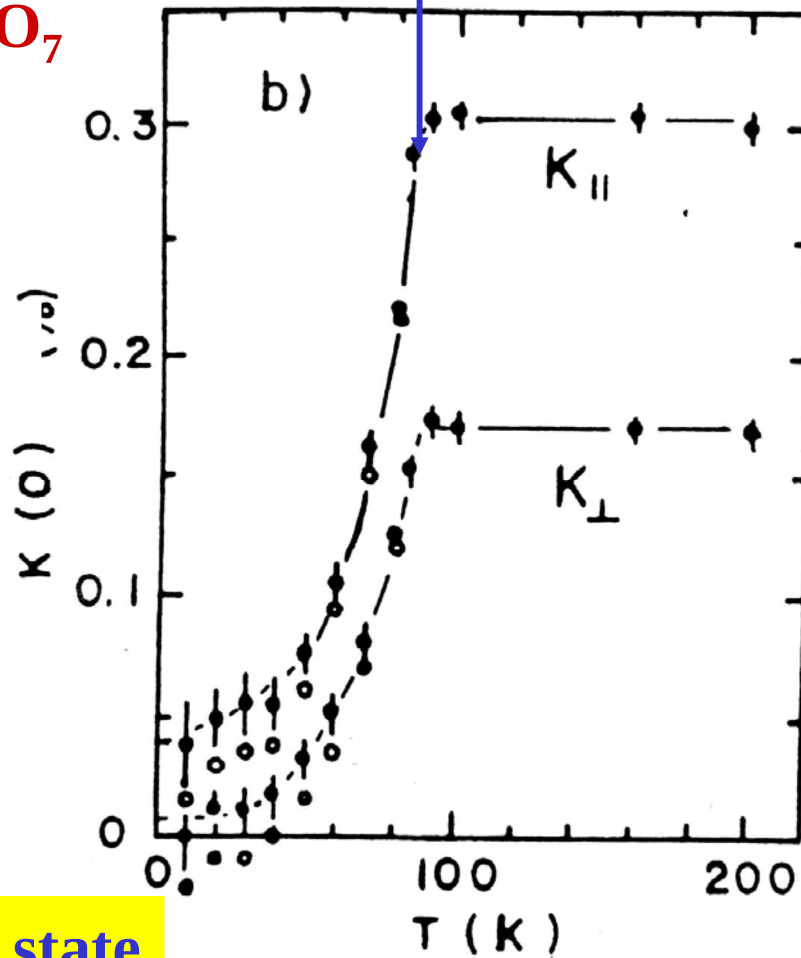


CuO_2 conducting planes

CuO_2

^{17}O NMR of the CuO_2 planes

T_c



M. Takigawa et al PRL 1989

Vanishing of χ_P in the superconducting state
Cooper pairs are in a singlet state

^{89}Y NMR shift in the metallic state

H.A , T. Ohno and P. Mendels, PRL 1989

- ΔK

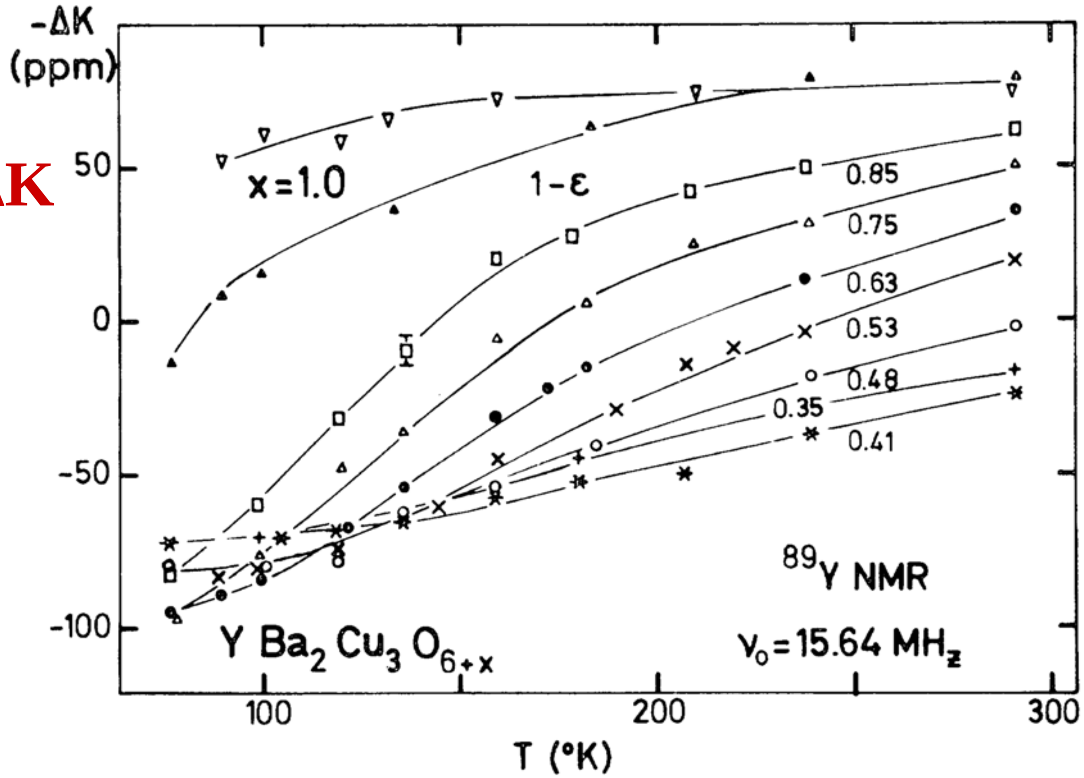
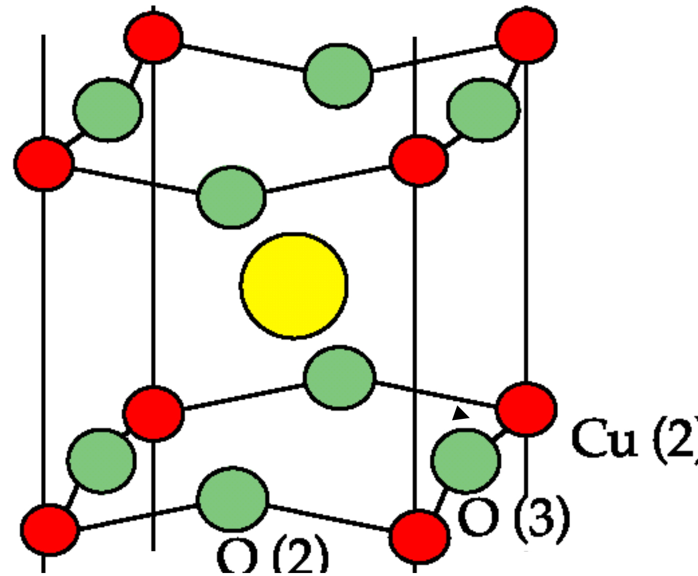


FIG. 1. The shift ΔK of the ^{89}Y line, referenced to YCl_3 plotted vs *T*, from 77 to 300 K. The lines are guides to the eye.

CuO$_2$ plane



CuO$_2$ plane

$$K_{i,\alpha}(T) = K_i^{dia} + A_{i,\alpha}^{orb}\chi_{i,\alpha}^{orb} + A_{i,\alpha}^{s}\chi_{i,\alpha}^{s}(T)$$

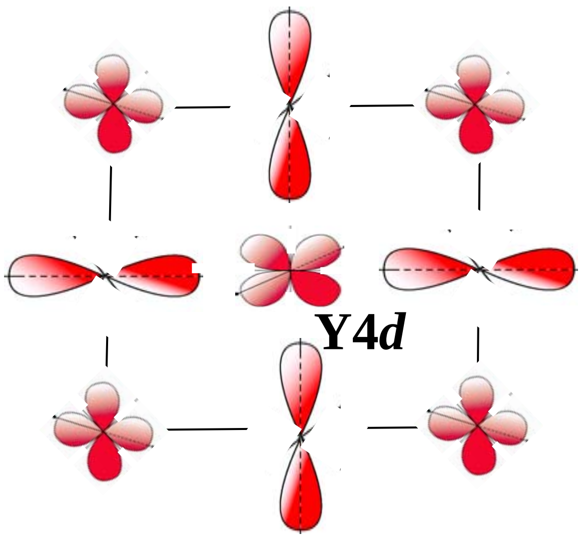
Local magnetic measurement

But transferred hyperfine couplings

Sign of ^{89}Y NMR shift

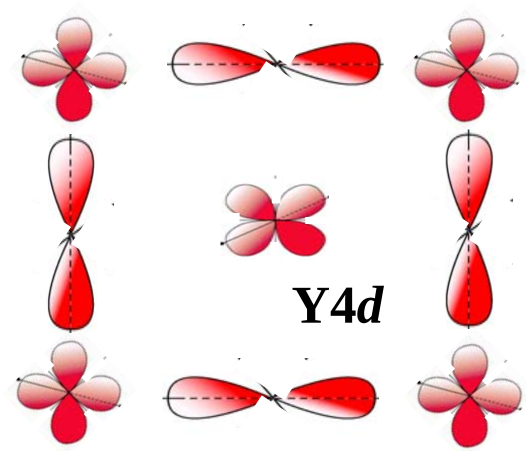
Negative sign comes from Y4*d* orbitals: core polarization

pπ Y4*d*



Very weak

pσ Y4*d*

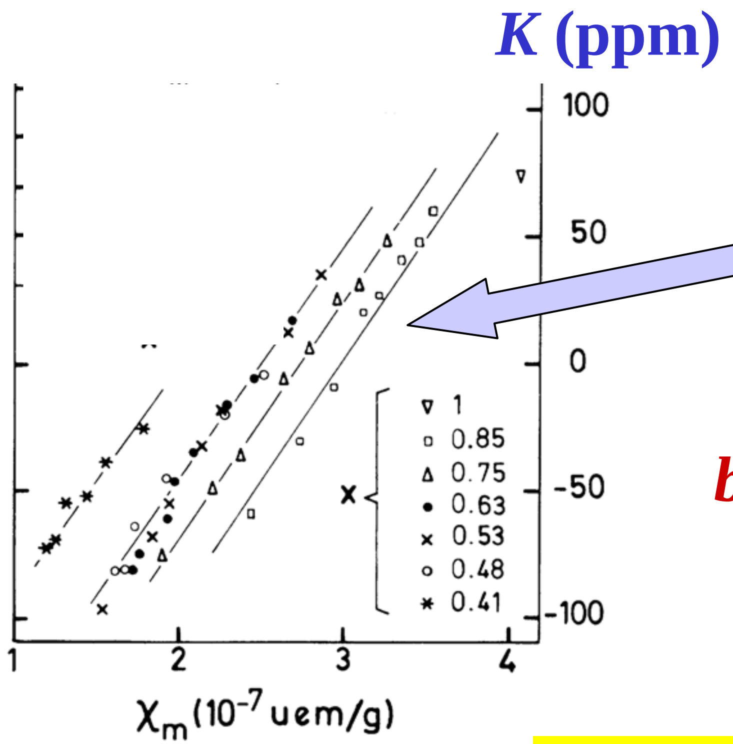


OK

So negative sign comes from pσ -Y4*d* hybridization

Is there an independent oxygen band at the Fermi level?

H.A. , T. Ohno and P. Mendels, PRL 1989



$K^s(T) = A\ \chi^s(T)$

does not change with hole doping

A is driven
by (Y4d-O2pσ)–Cu(3d) covalency

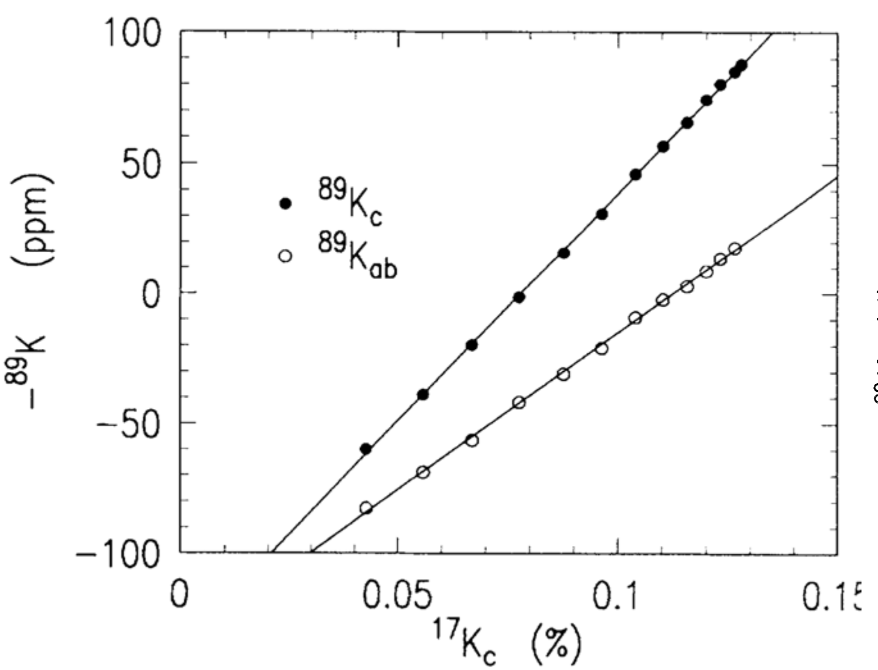
So there is no independent oxygen spin degree of freedom at E_F

Single spin fluid behaviour

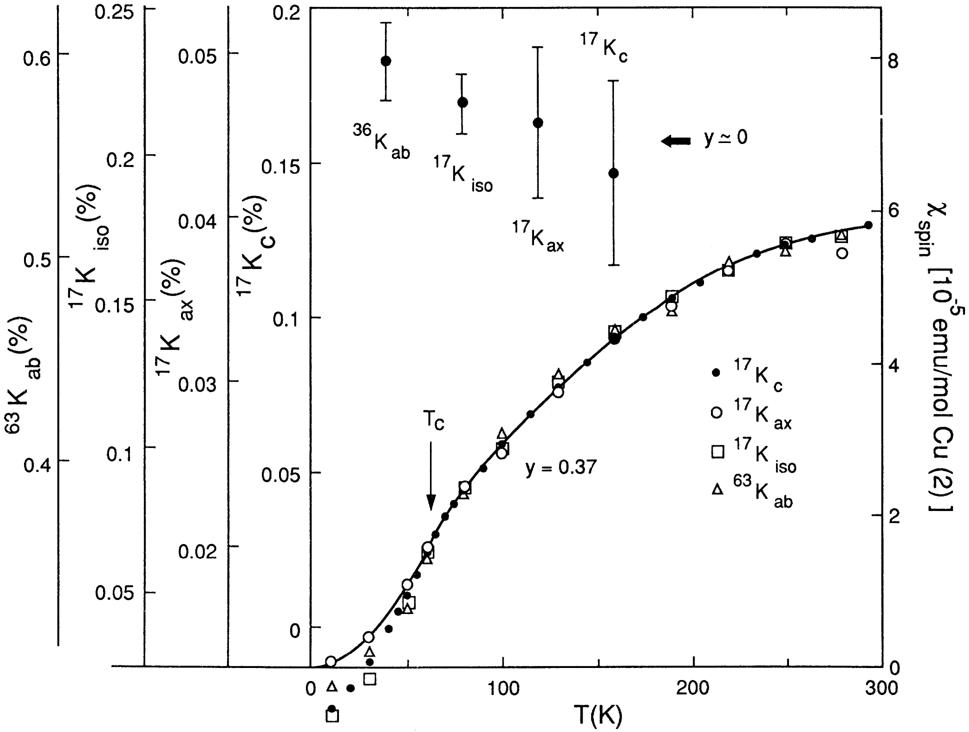
YBCO$_{6.63}$

$$K_{i,\alpha}(T) = K_i^{dia} + A_{i,\alpha}^{orb}\chi_{i,\alpha}^{orb} + A_{i,\alpha}^{s}\chi_{i,\alpha}^{s}(T)$$

M. Takigawa et al 1991, 1993



^{89}Y versus ^{17}O



^{63}Cu versus ^{17}O

A single T dependence for $K_{i,a}(T)$: due to $\chi_{Cu}(T)$

Notice: this allows determinations of the shift references for all nuclei

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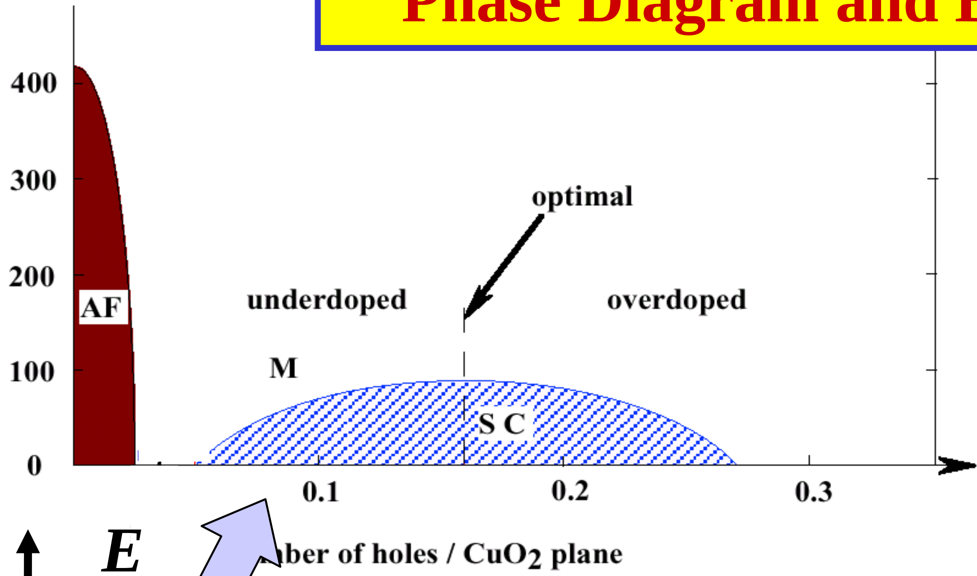
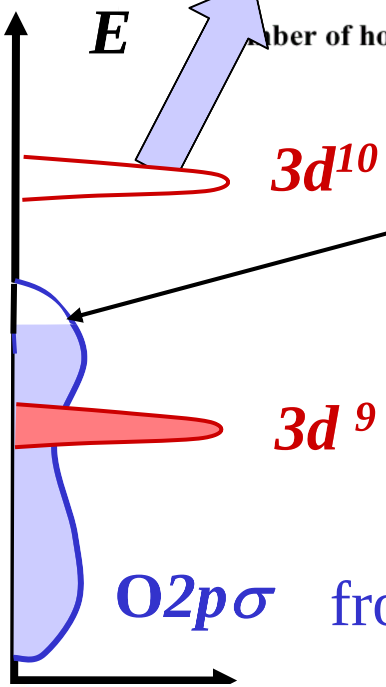
Phase Diagram and Band Structure

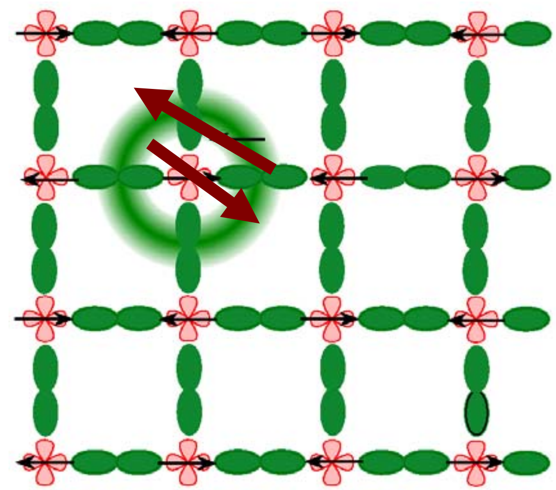




NO!!
Strong O2*p* - Cu *3d* hybridization
Magnetic correlations

There is a single spin fluid

Zhang Rice spin singlets
Cu3*d* - O2*p*σ



The holes steal spins
from the copper hole background

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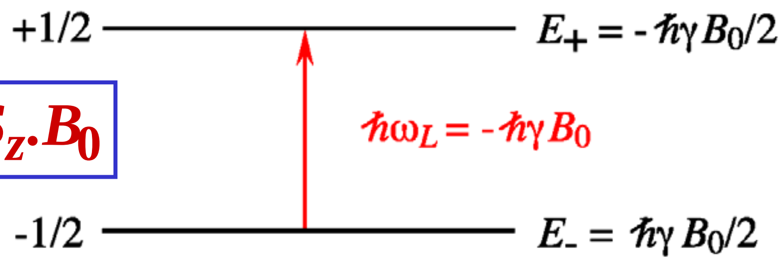
Physical Origin of the Spin Lattice Relaxation

$$H_Z = -\hbar\gamma S_z . B_0$$



B_0 // z

transitions $|-1/2\rangle \rightarrow |1/2\rangle$

if $\langle 1/2 | H_{rf} | -1/2 \rangle \neq 0$

$B_1 \perp z$

rf excitting field
perturbation for H_Z

$$H_{rf} = -\hbar\gamma \, \mathrm{S.B_1} \cos\omega_L t$$

$$\vec{B}_L = \langle \vec{B}_L \rangle + \left[\vec{B}_L - \langle \vec{B}_L \rangle \right]$$

Relaxation: transverse components of the fluctuating field at the Larmor frequency

Transition probability

$$\frac{1}{T_1} = \gamma_n^2 \int_{-\infty}^{\infty} \langle B_L^+(t) \, B_L^-(0) \rangle \exp(-i\omega_n t) \, dt$$

Correlation function of the local field

T_1 results from the coupling with the equilibrium fluctuations of the electron spins degrees of of freedom

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Spin lattice relaxation in a free electron metal

$$\frac{1}{T_1} = \frac{2A^2}{\hbar^2 \gamma_e^2} k_B T \frac{\chi_T''(\omega_n)}{\omega_n}$$

$$\chi_T''(\omega_n) = \sum_q \chi_T''(q,\omega_n)$$

Al metal

For a free electron gas χ ''(q, ω_n) is q independent

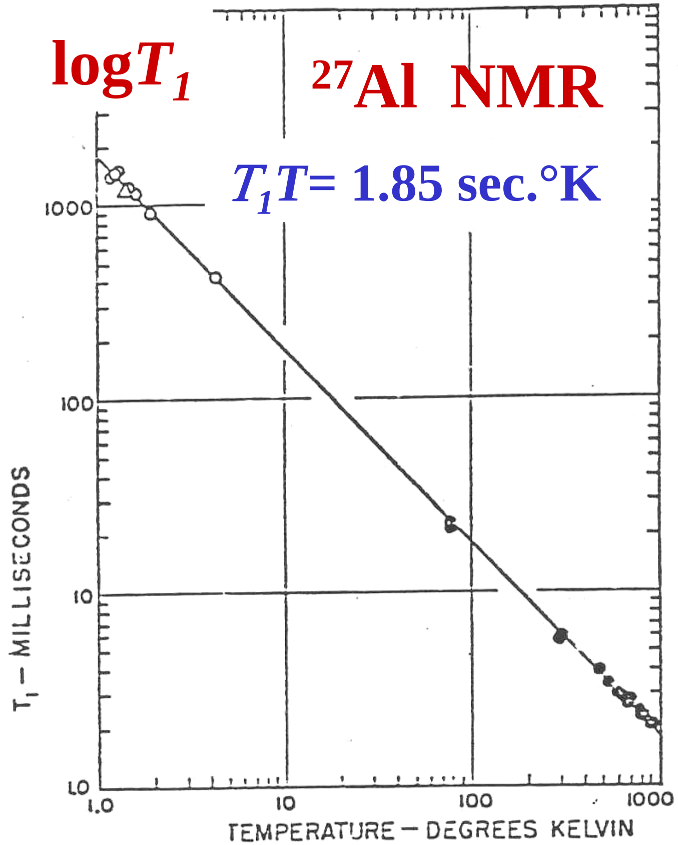
$$\chi_T(\omega) = \frac{1}{2} \hbar^2 \gamma_e^2 \left\{ n(E_F) + i\pi\hbar\omega n^2(E_F) \right\}$$

$$\frac{1}{T_1} = \frac{\pi}{\hbar} A^2 n^2(E_F) k_B T$$

$$K = \frac{A}{\hbar^2 \gamma_e \gamma_n} \chi_P = \frac{A\gamma_e}{2\gamma_n} n(E_F)$$

$$T_1 T K^2 = \frac{\hbar}{4\pi k_B} \left(\frac{\gamma_e}{\gamma_n} \right)^2 = S_0$$

logT_1



log(1/T)

Korringa law for a metal

Thermometry

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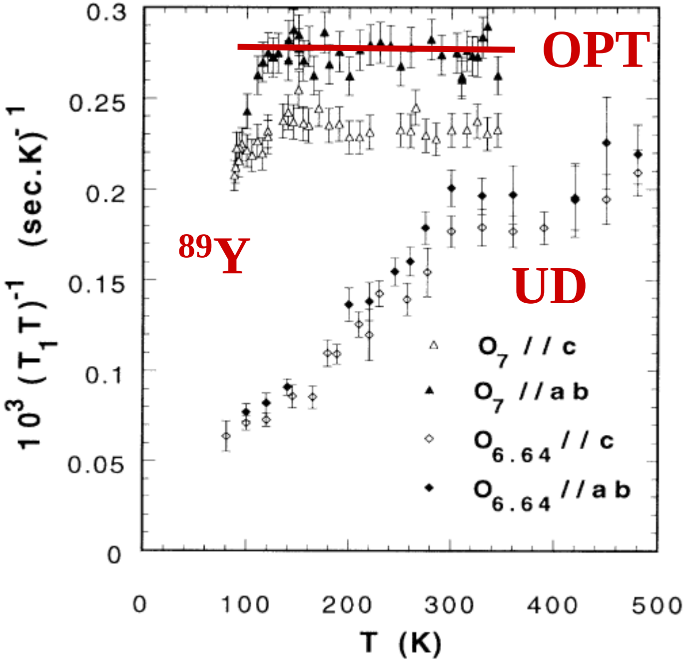
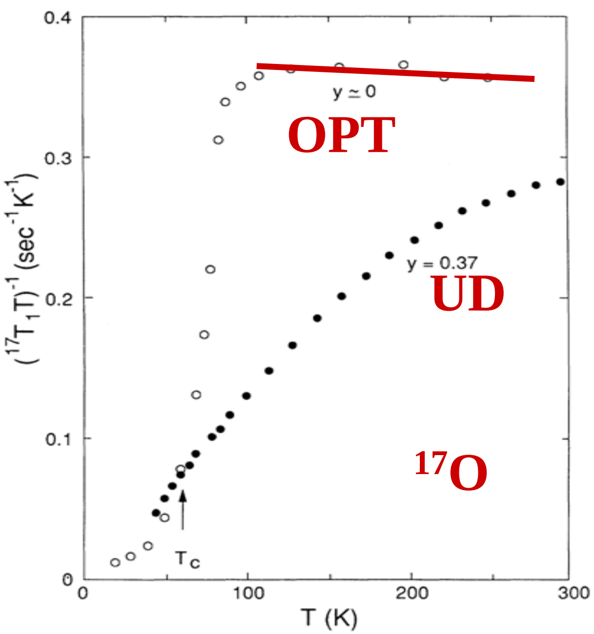
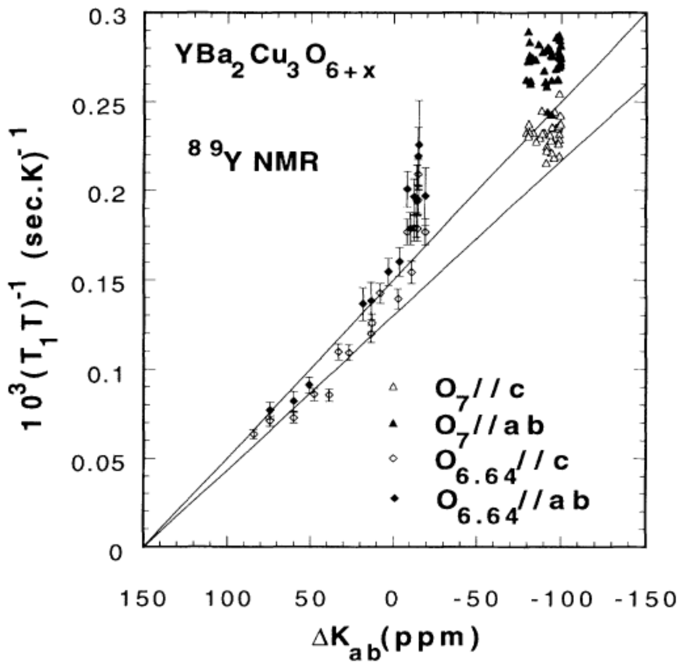
$(T_1T)^{-1}$

Comparison of $(T_1\,T)^{-1}$ on ^{89}Y and ^{17}O





In $YBCO_7$ T_1T is nearly constant on ^{17}O and ^{89}Y
Like in a free electron metal



In $YBCO_{6.6}$

$$\frac{1}{T_1T} \propto K$$

Metallic like component

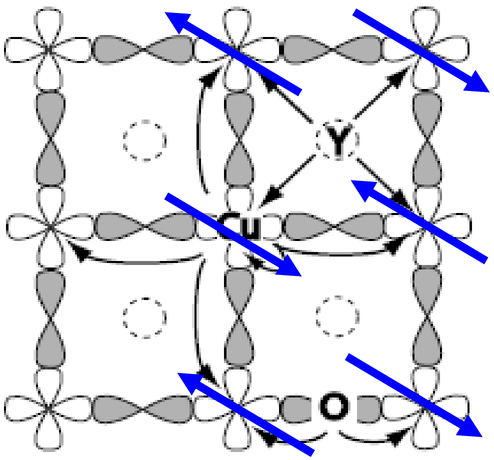
^{89}Y NMR Evidence for a Fermi-Liquid Behavior in $YBa_2Cu_3O_{6+x}$

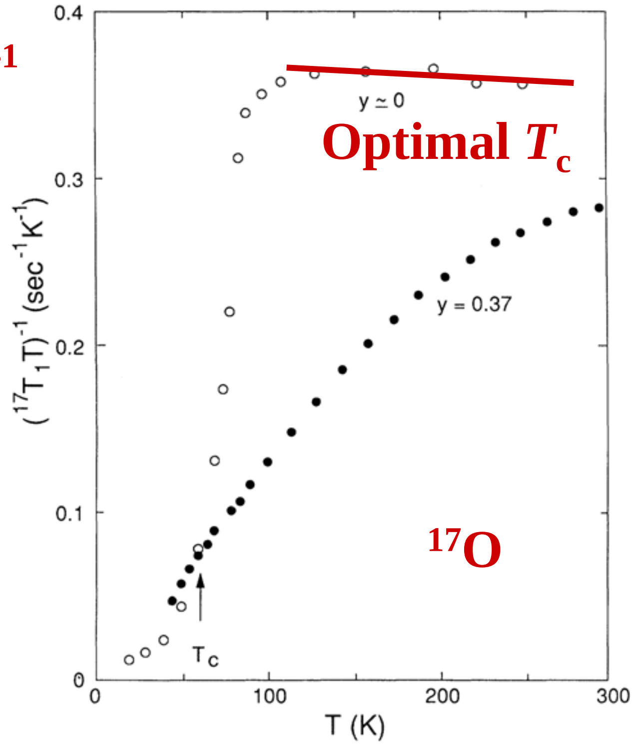
H. Alloul, T. Ohno,[a] and P. Mendels
Physique des Solides, Université de Paris-Sud, 91405 Orsay, France

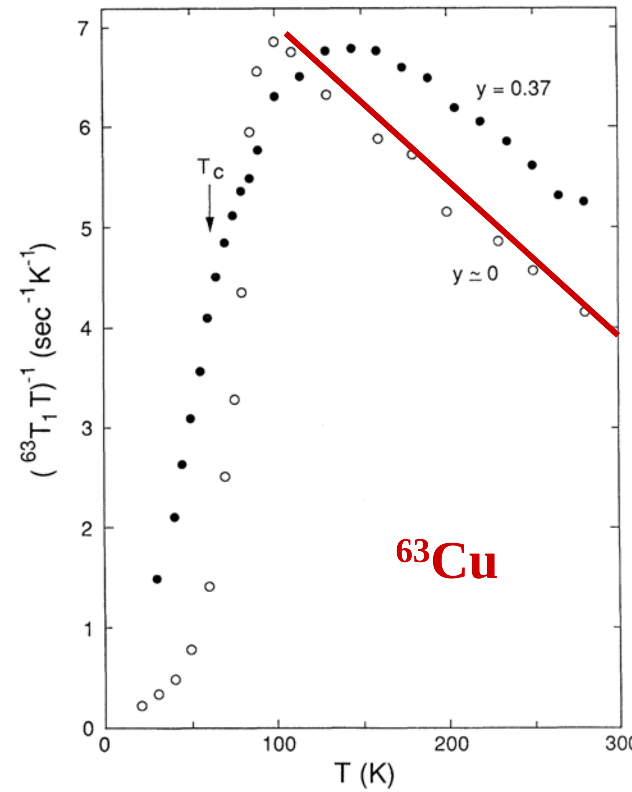
(Received 15 May 1989)

Distinct behaviour of $(T_1T)^{-1}$ on the Cu site: AF correlations

$(T_1T)^{-1}$







In $YBCO_7$ T_1T is nearly constant on ^{17}O but increases at low T for ^{63}Cu

O and Y are insensitive to AF correlations while Cu probes them fully

Increase of AF correlations at low T
Even more for the underdoped case

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H. Alloul, Cours A. Georges CDF, 9/11/2010

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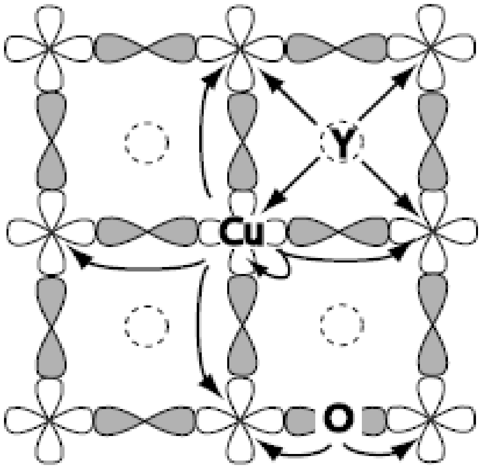
T_1 for nuclei coupled to neighbouring sites

Non local hyperfine coupling
q dependence of the HF coupling

$$A^s_{O,\alpha}(\mathbf{q}) = A^s_{O,\alpha} \sum_{\mathbf{r}_i} \exp(i\ \mathbf{q}.\mathbf{r}_i)$$



^{89}Y $$A^s_{Y,\alpha}(q) = 8D_\alpha \left(\cos q_x a/2\ \cos q_y a/2\right)$$

^{17}O $$A^s_{O,\alpha}(q) = 2C_\alpha \cos q_x a/2$$

^{63}Cu $$A^s_{Cu,\alpha}(q) = A_\alpha + 2B_\alpha \left(\cos q_x a + \cos q_y a\right)$$

For Y and O, A(q) vanishes for q_{AF}= (π/a,π/a)
The AF fluctuations are filtered out by A(q)

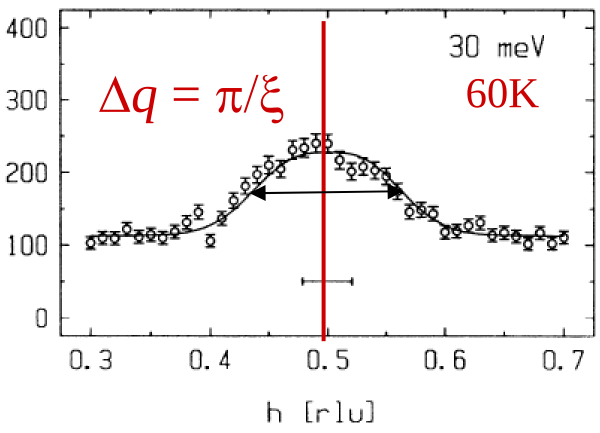
$$\frac{1}{T_1} = \frac{2k_B T}{\hbar^2 \gamma_e^2} \sum_{\mathbf{q}} A^2(\mathbf{q})\ \chi_\perp''(\mathbf{q},\omega_n)/\omega_n$$

$\xi(T)$ **is the AF correlation length probed by ^{63}Cu NMR**

$\omega \rightarrow 0$

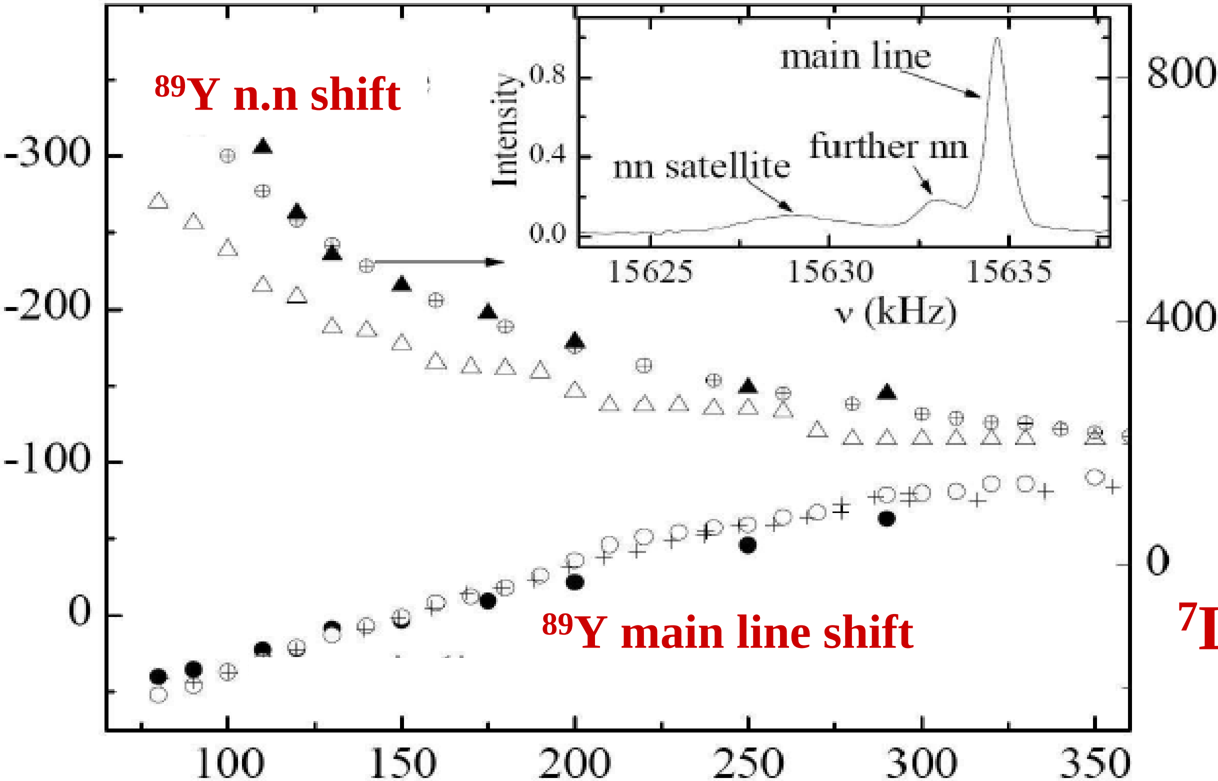
Neutron scattering

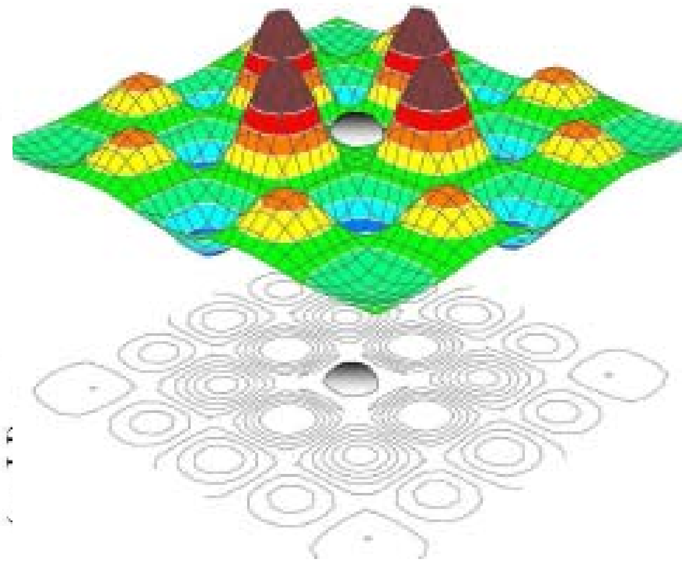
$\chi_\perp''(q,\omega_n)$ **YBCO$_{6.6}$**



Zn^{2+} or Li^{+} (no spin) substituted to Cu^{2+} (spin 1/2)

^{89}Y NMR shift





^{7}Li NMR shift

Empty symbols: Zn^{2+} — *A. Mahajan, H.A …PRL 1994;*

Full symbols : Li^{+} — *J. Bobroff, H. A … . PRL, 1999*

The spinless character of the impurity dominates the magnetic response

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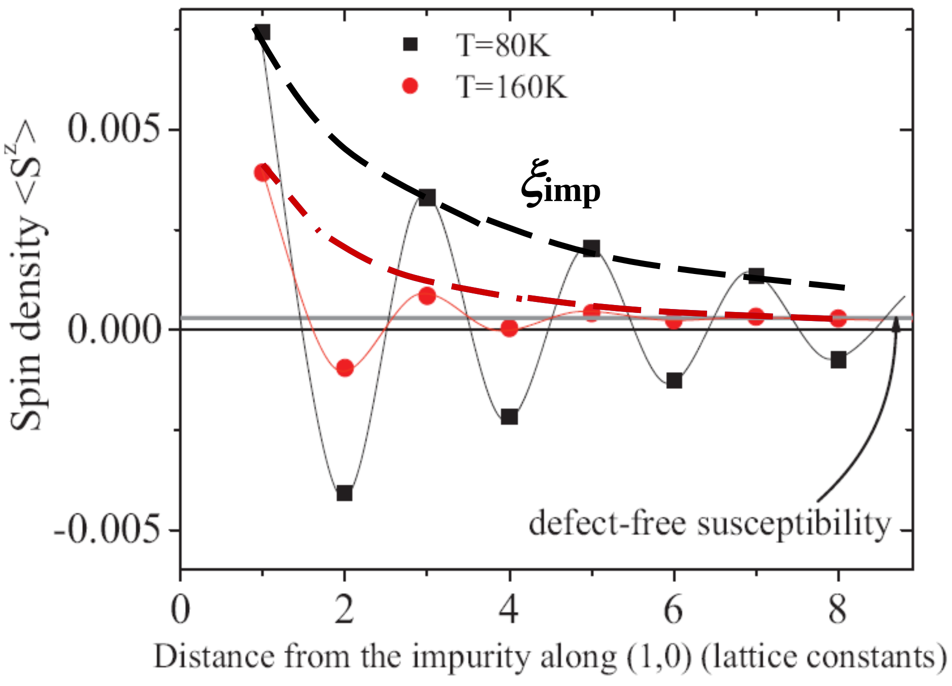
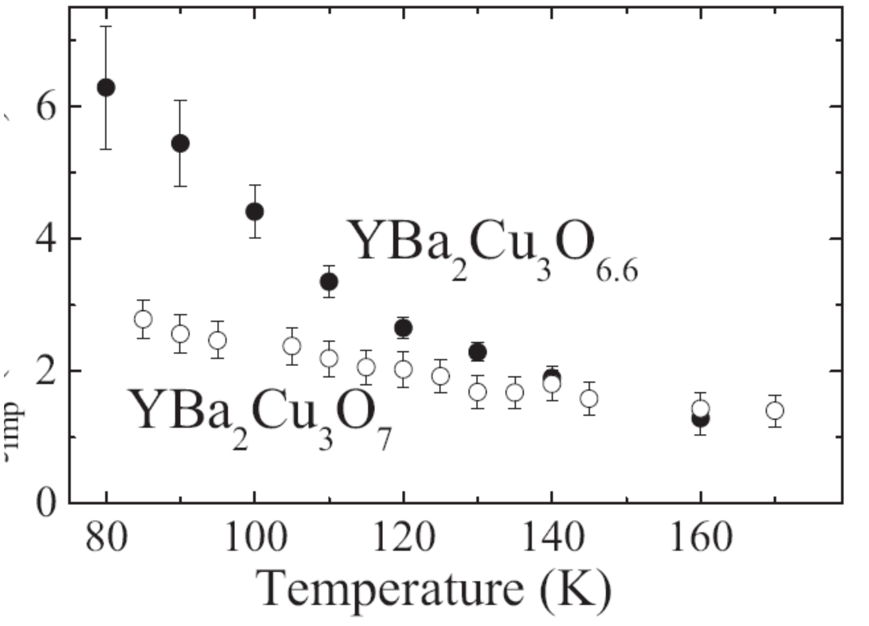
Spatial extent of the staggered moment

S. Ouazi , J. Bobroff, H. A , PRB 2004



$\xi_{imp}(T)$



ξ_{imp}(T) varies smoothly with *T* and doping

Review article: H.A, J. Bobroff, P. Hirschfeld and M. Gabay, RMP 2009

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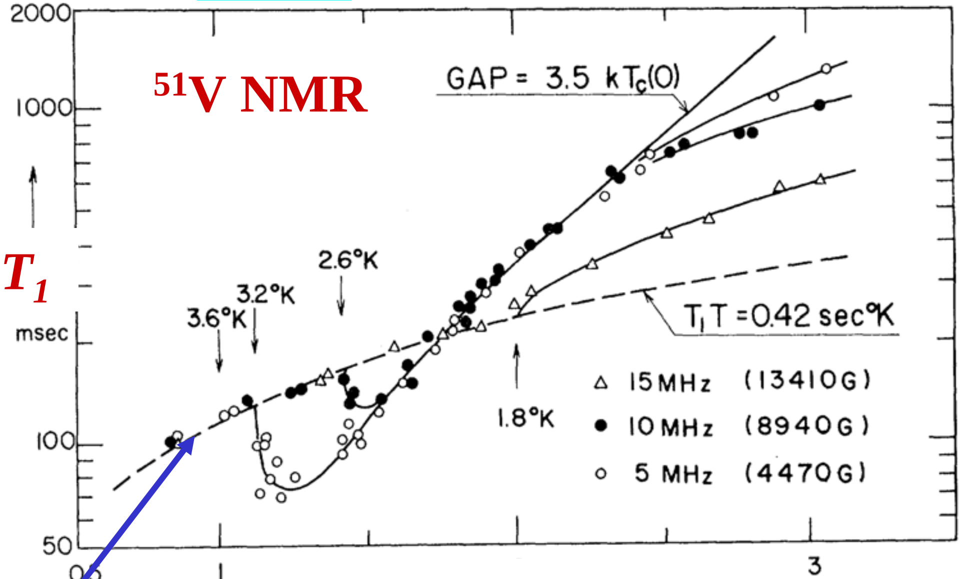
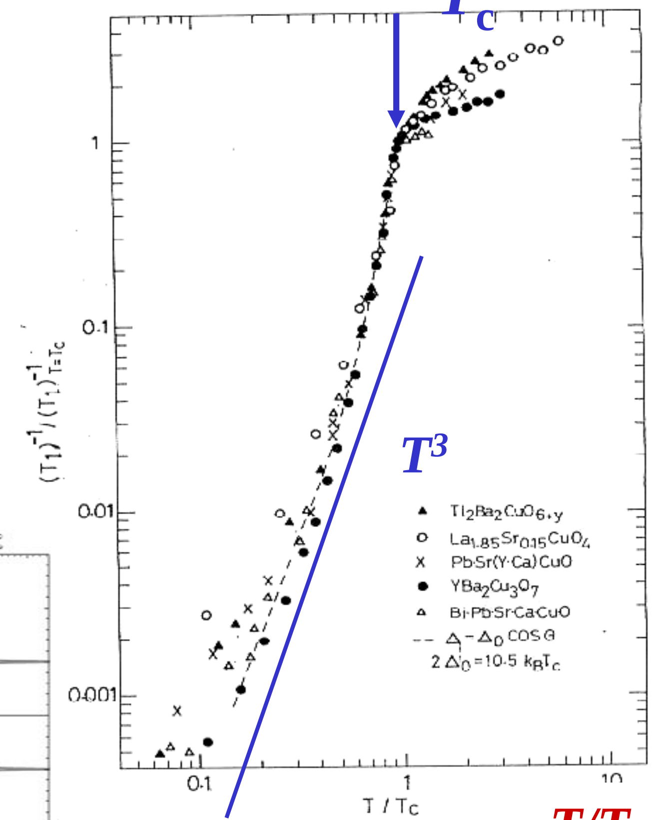
T_1 in the superconducting state

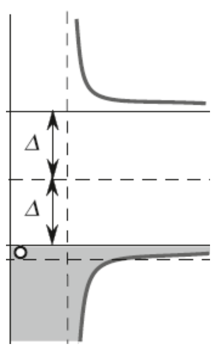
V_3Sn



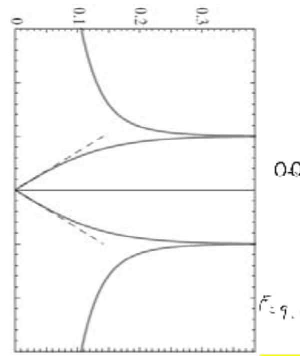
Cuprates

$(T_1T)^{-1}/(T_1T)^{-1}_{T=Tc}$





s wave superconductor



$(T_1T)^{-1} \sim \; exp\,(-\Delta/k_BT)$

for $T<<T_c$

T_1 minimum below T_c

(Hebel-Slichter peak in 1/ T_1)

d wave superconductivity

T^3 variation for $T<<Tc$

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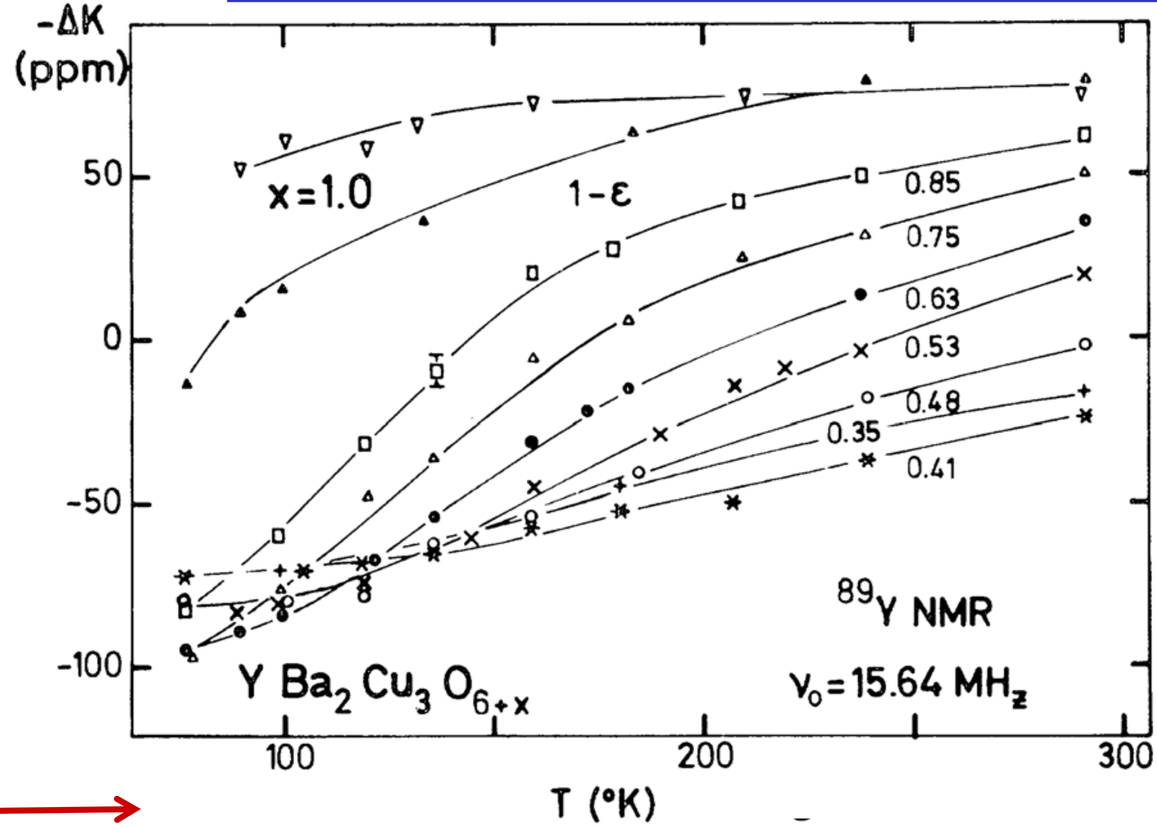
What about the origin of this *T* dependence ?



Large decrease (nearly full loss) of $\chi^s(T)$ above T_c

Pseudogap in the electronic excitations

Origin for K^s

H.A, T. Ohno and P. Mendels, PRL 1989

tuations on the Cu than on the Y or O, which are symmetric sites for the AF lattice of the O_6 compound.[7] In the band picture, AF correlations might induce a pseudogap, as suggested by Friedel,[24] which could explain the reduction of χ_s at low T. However, it is less clear whether this approach is compatible with the smooth variation of χ_s and K_s from the metal to the insulat

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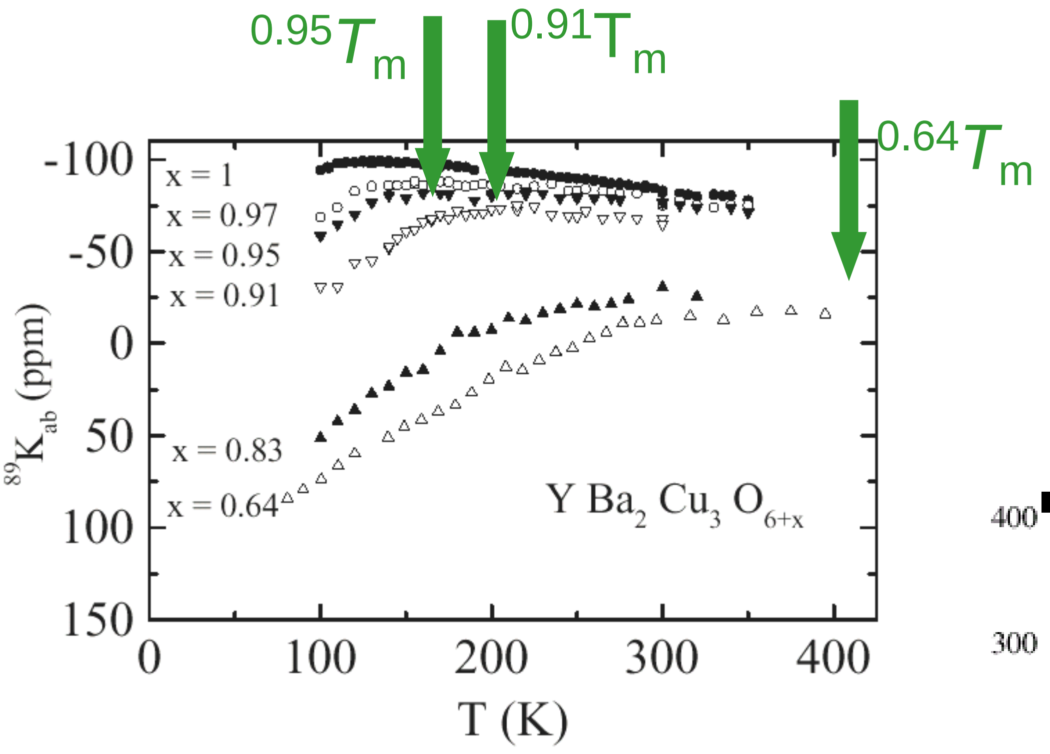
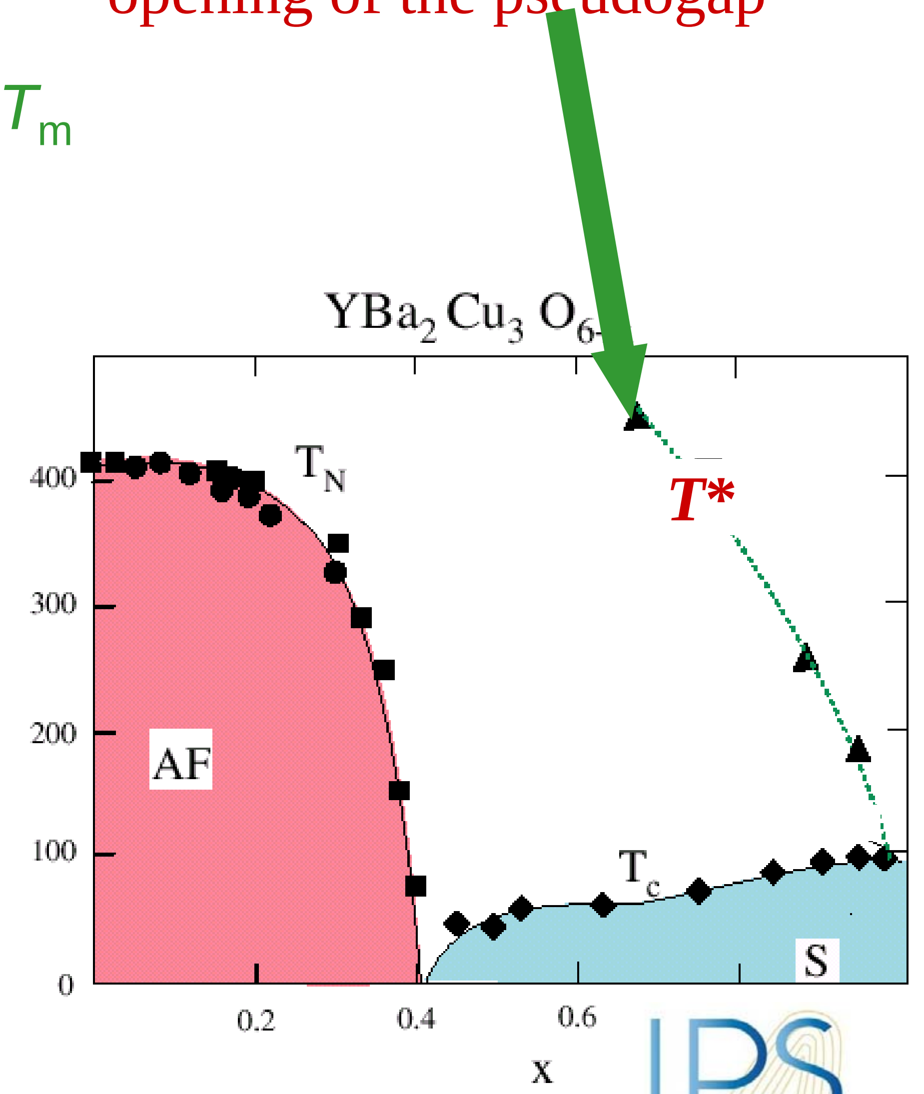
Phase Diagram and Pseudogap

^{89}Y NMR shift

Low *T* decrease of the susceptibility: opening of the pseudogap

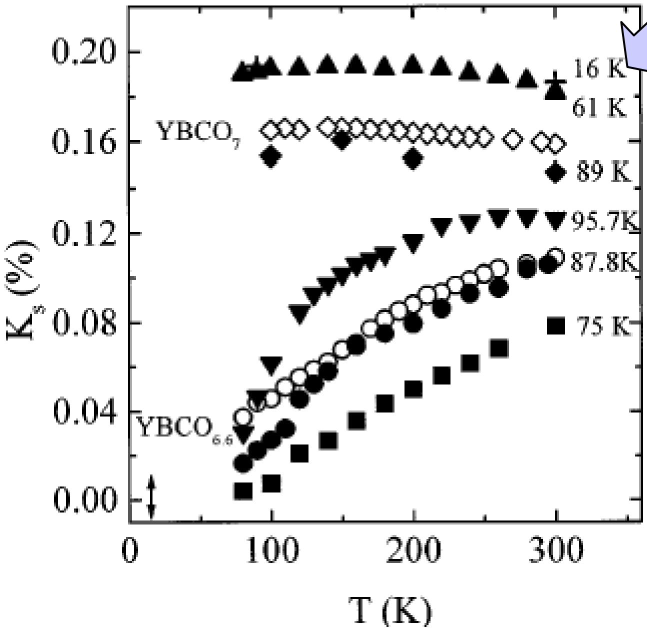




Alloul et al, ????

The drop of χ(T) is generic of underdoped cuprates

[17]O NMR **Hg1201 : one CuO_2 layer,** *J. Bobroff, H.A,… PRL 1997*

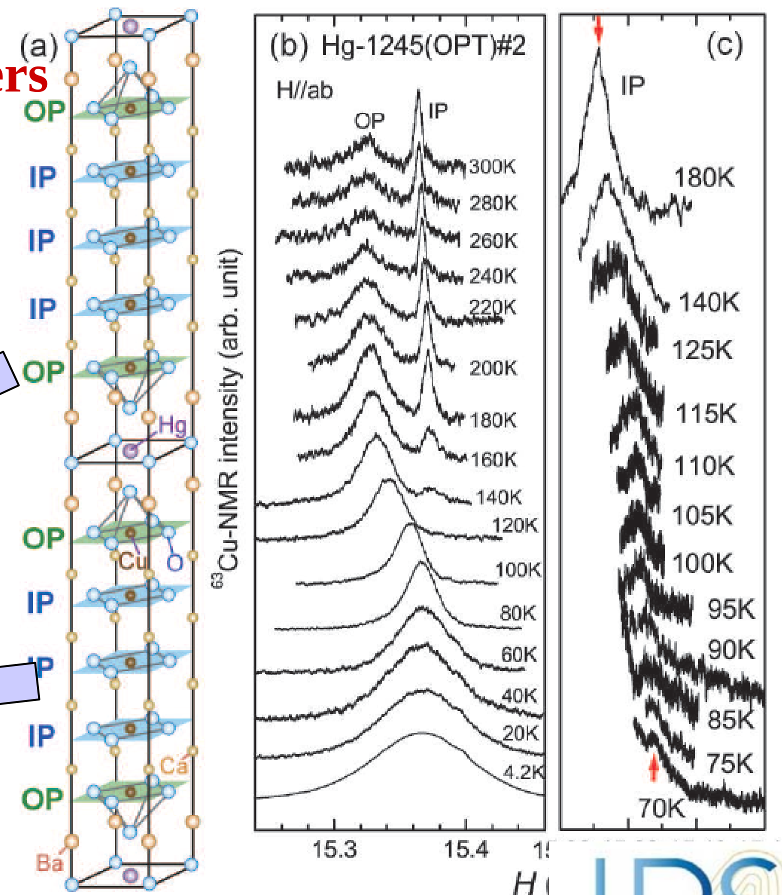


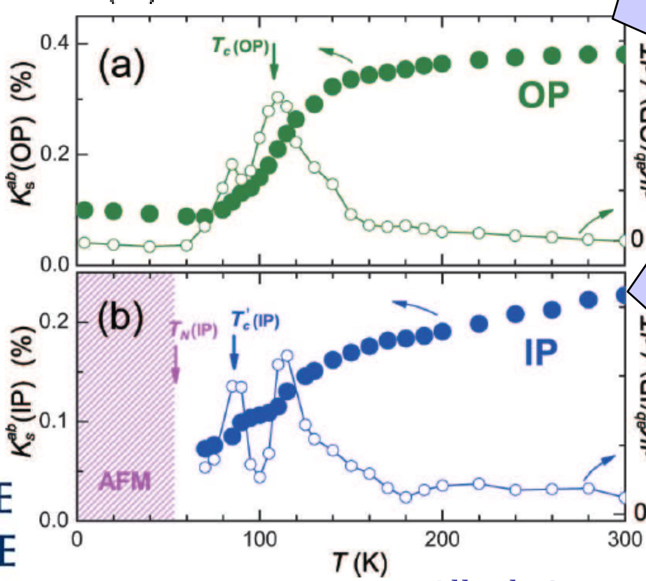
Can be used to estimate the hole doping!

Bi2223 : three CuO_2 layers, *A Trokiner et al ,…*

five CuO_2 layers
Hg1245

Mukuda et al, JPSJ 2008





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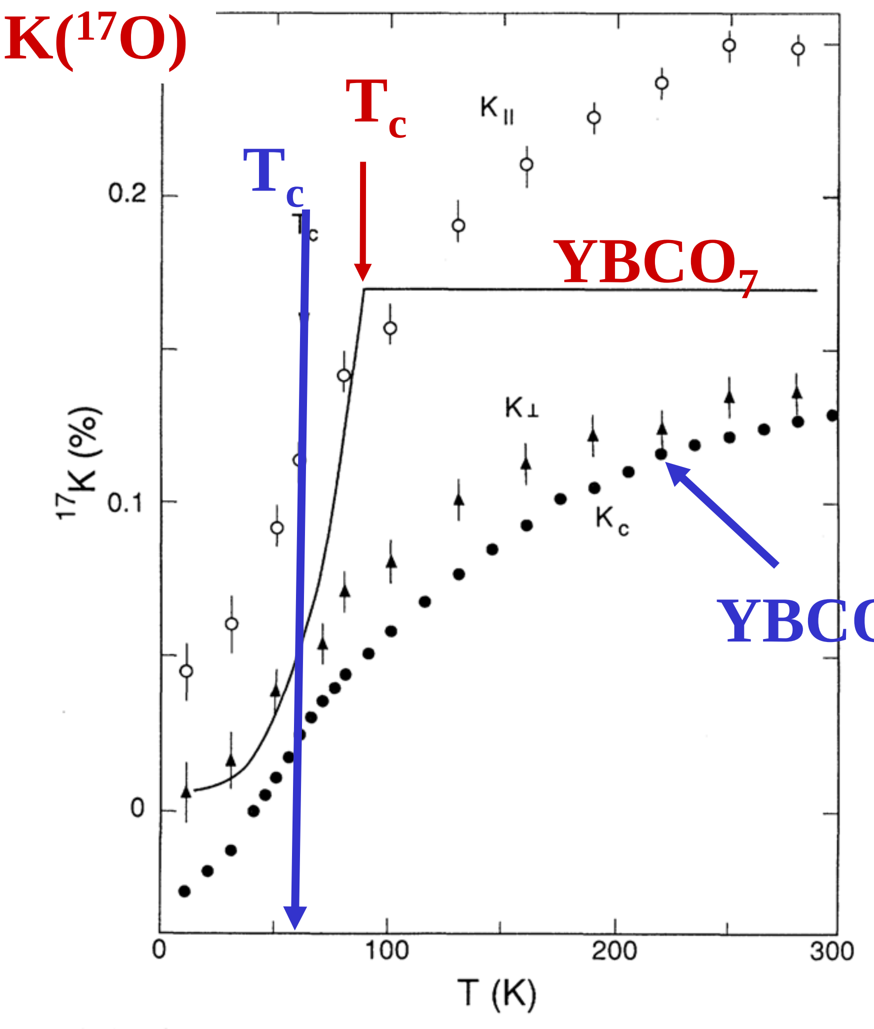
Knight Shift in underdoped YBCO



M. Takigawa et al PRB

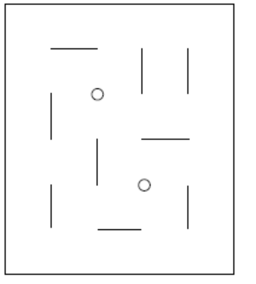
The large drop of $\chi_s(T)$ above T_c could be due to Cooper pair formation

T_c being then due to phase coherence of these preformed pairs
Bose Einstein condensation ?

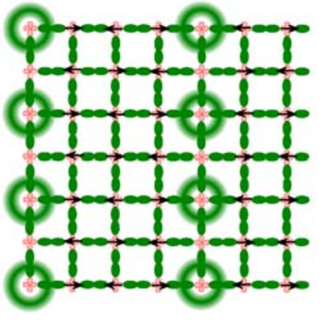
Other scenarios

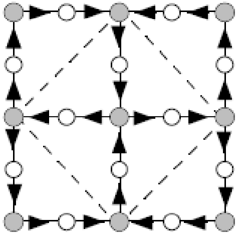
competing magnetic orders

RVB spin liquid

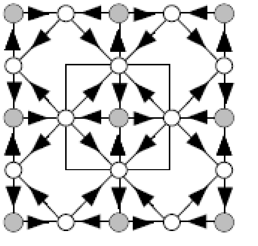


Stripes





ddw order

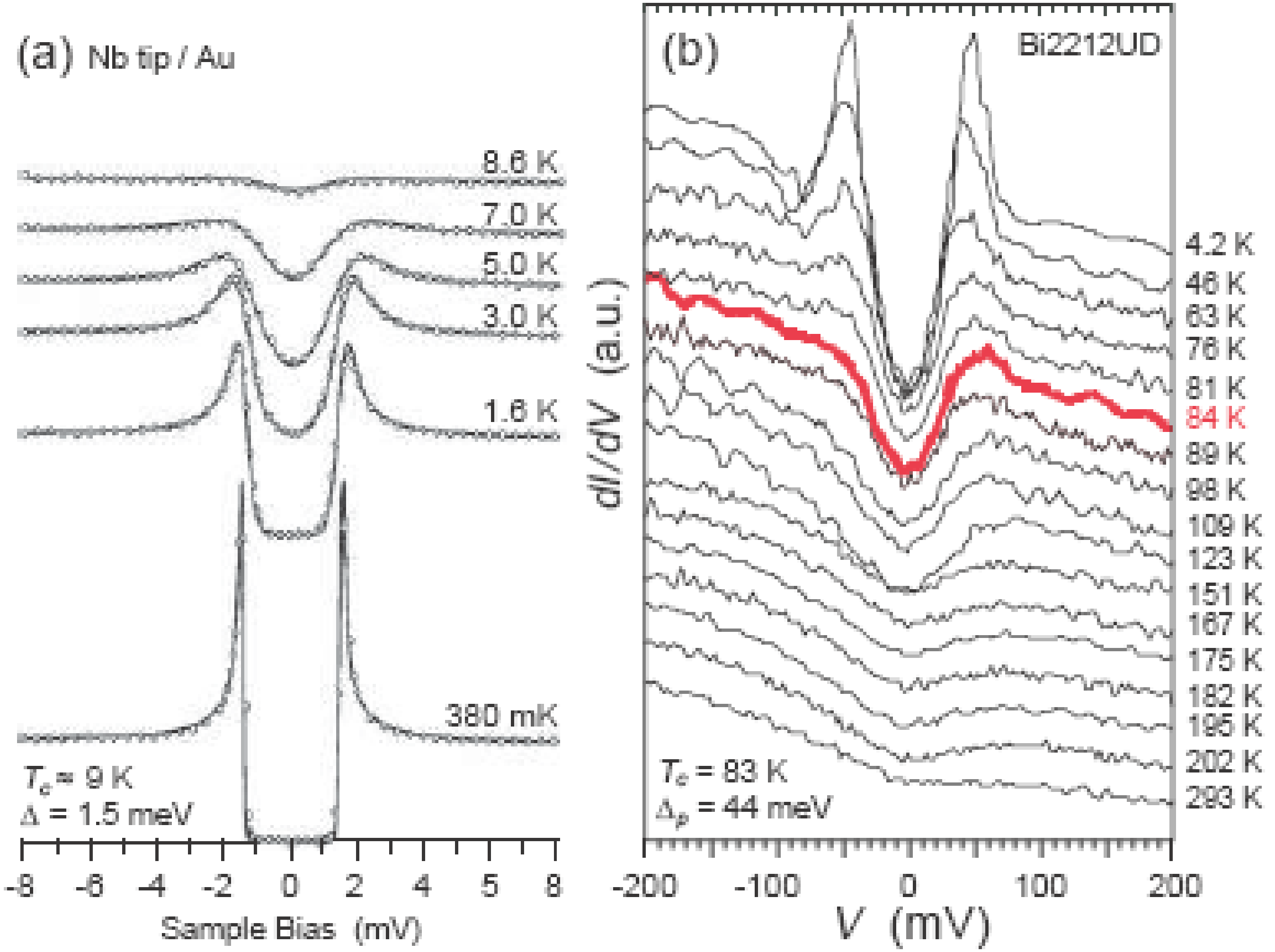


Orbital currents

TRIANGLE DE LA PHYSIQUE

LPS ORSAY

Superconductivity in Bis$_2$r$_2$CaCu$_2$O$_{8+x}$ T$_c$=95K



PSEUDOGAP

C. Renner et al, PRL 1998

Niobium BCS

Underdoped cuprate

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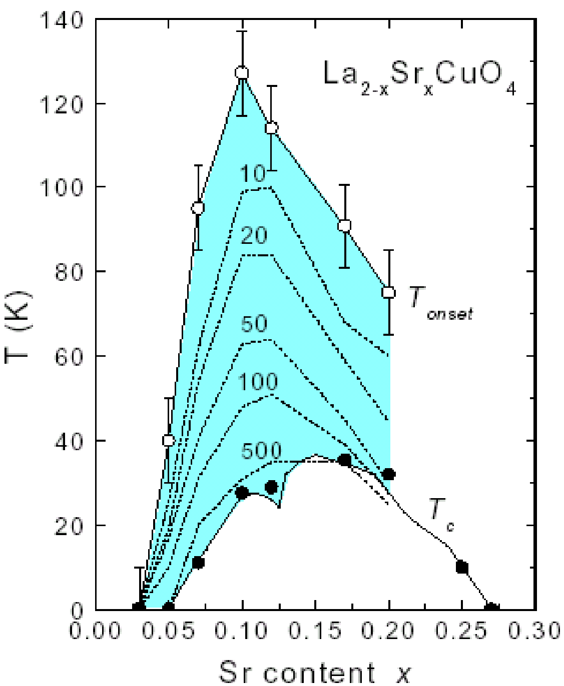
Superconducting fluctations in the normal state of cuprates

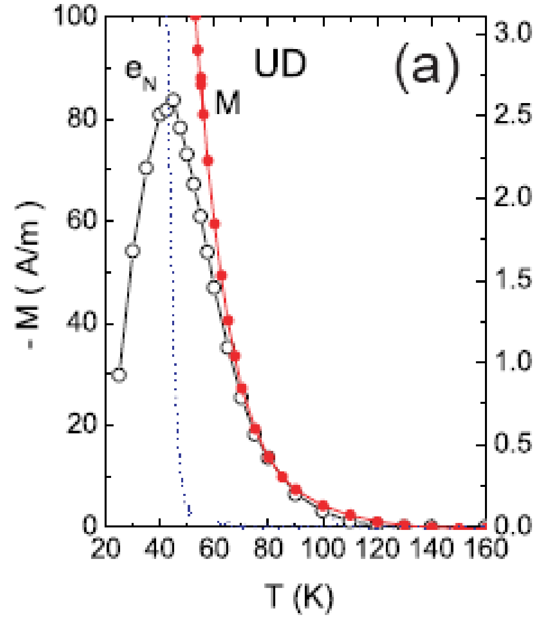
Bi 2212

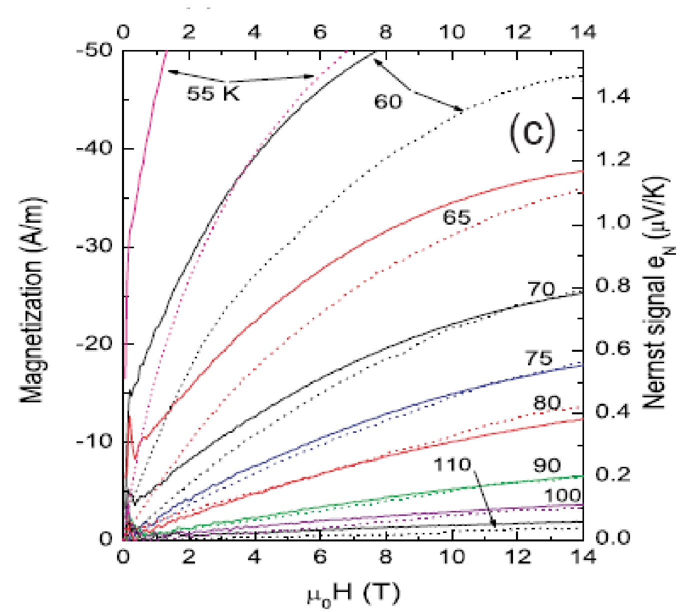
Nernst effect

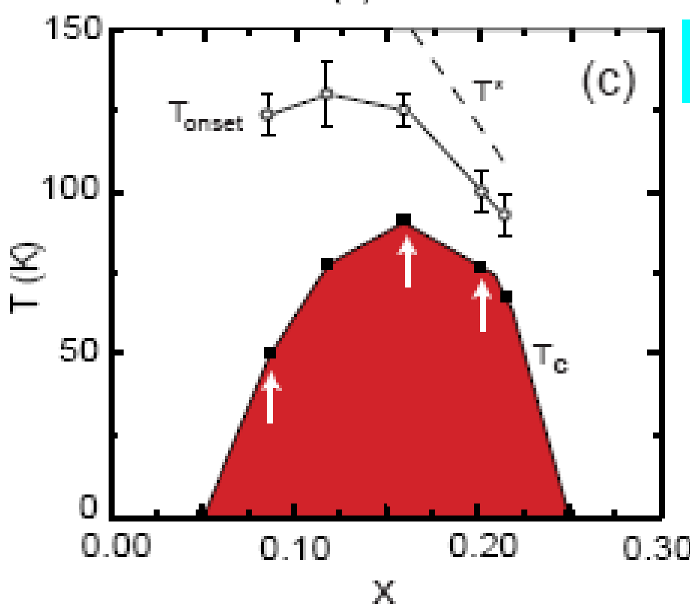
$La_{2-x}Sr_xCuO_4$



Wang et al, PRB 64 (2001)







High field diamagnetism

Wang et al, PRB 64 (2005)

Precursor pairing ?

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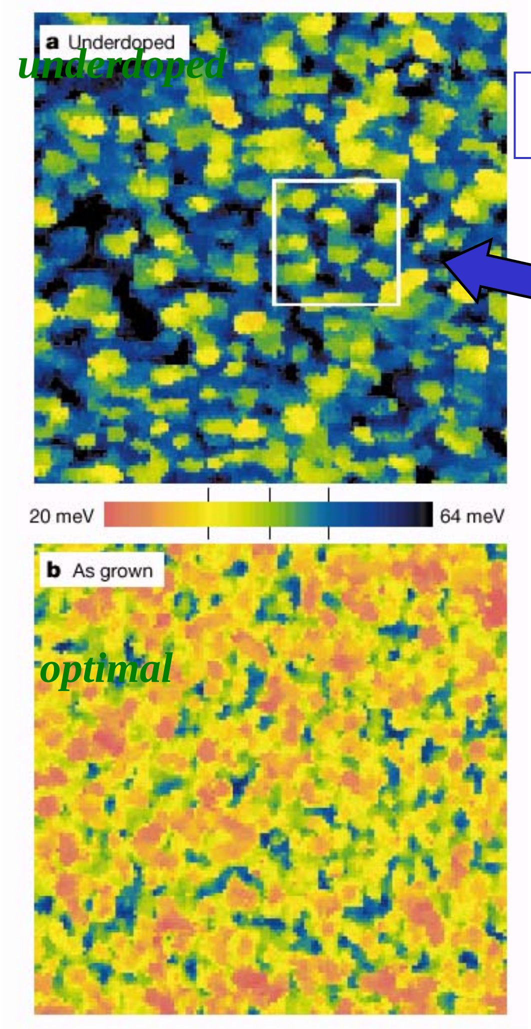
Inhomogeneities in BiSCCO viewed by STM



560 A

Lang et al, Nature 412, 415 (2002)

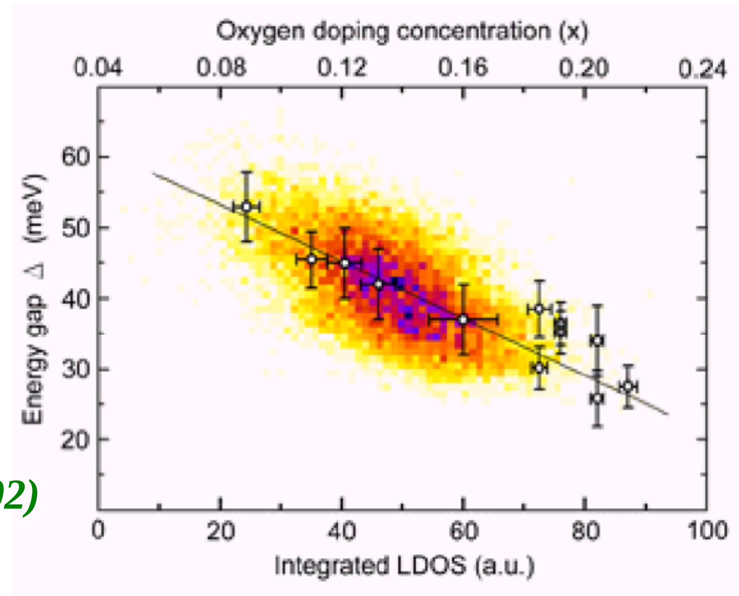
Cren et al, PRL 84,147 (2000); Howald et al PRB 64 10054-1(2001)

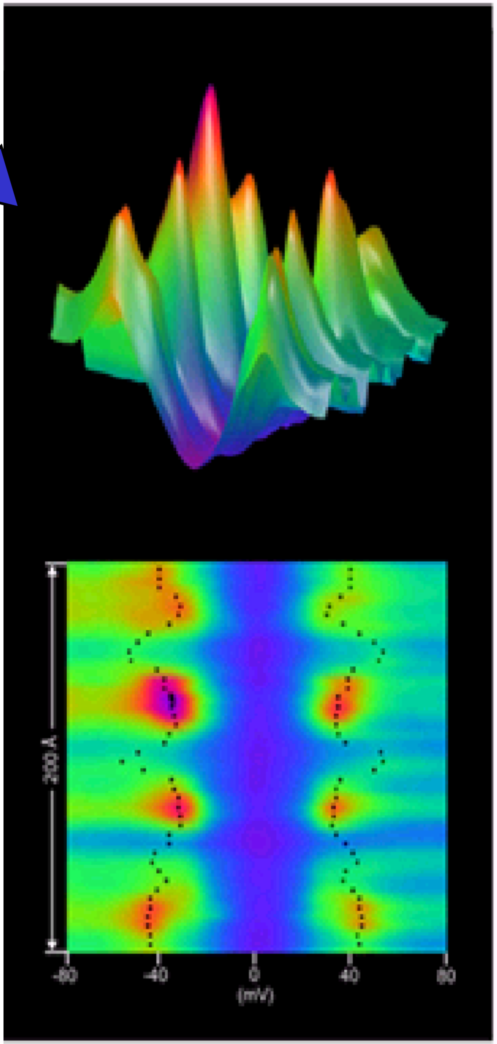
DOS depends on the STM tip location :

2D maps of the gap magnitude

Pan et al, Nature 413, 282 (2001)

Local distribution of hole doping



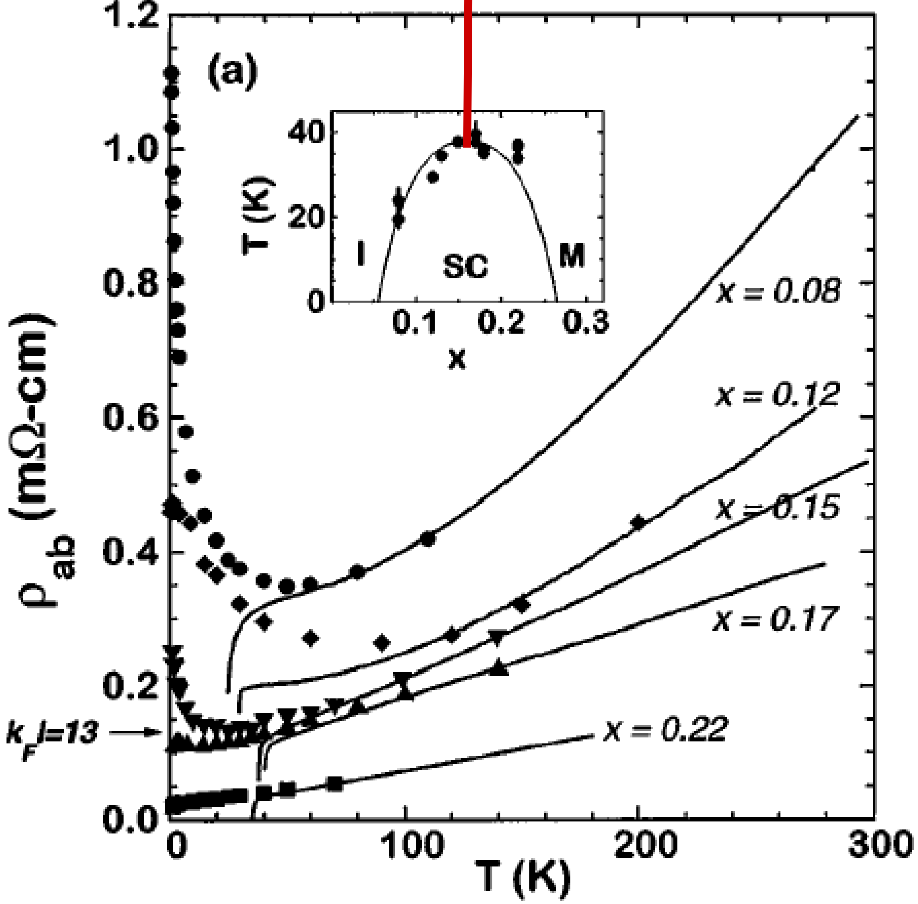


H. Alloul, Cours A. Georges CDF, 9/11/2010

Metal insulator transition

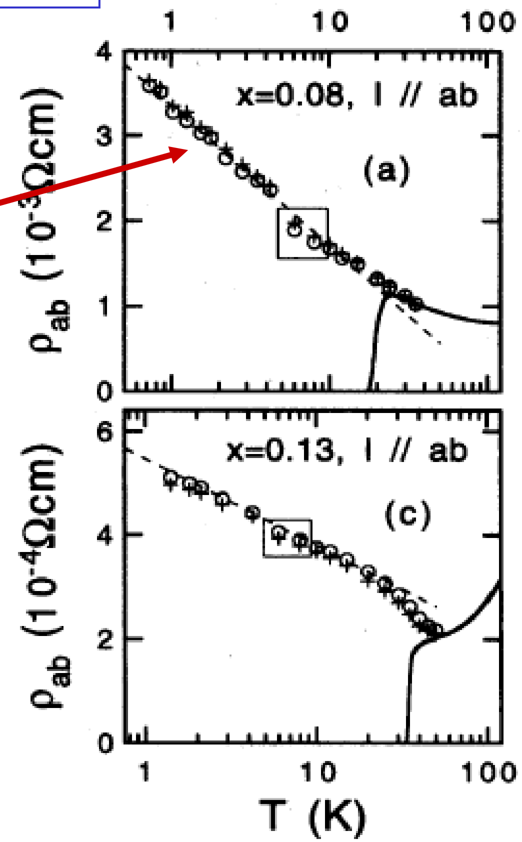
$La_{2-x}Sr_xCuO_4$

High field measurements



G.S. Boebinger, Y. Ando et al PRL 1996

Log *T*

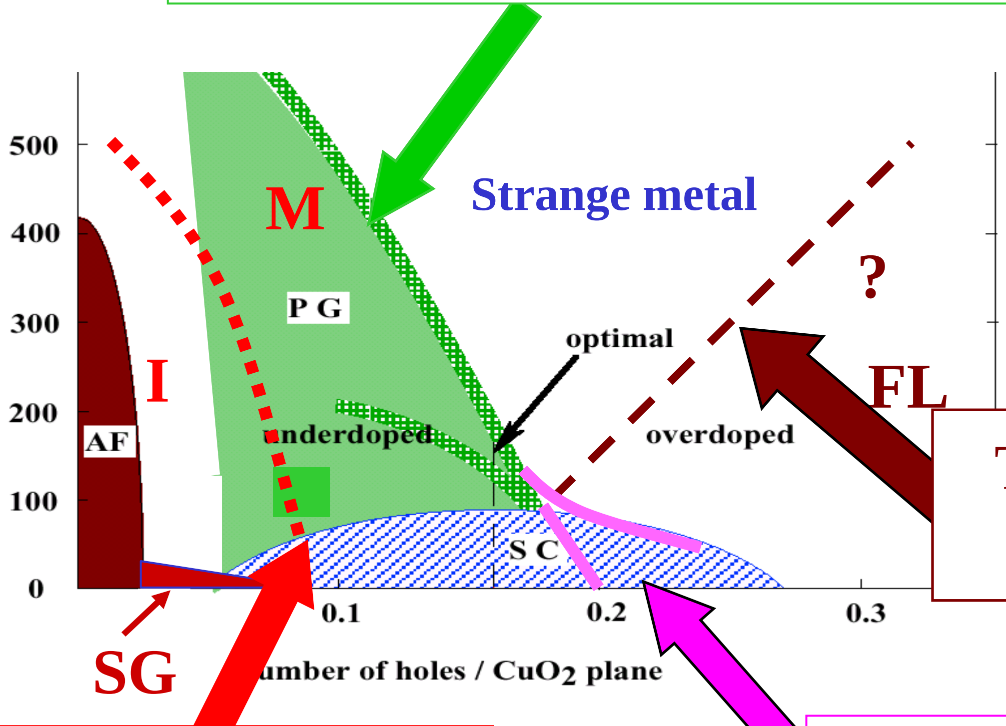


Y. Ando, G.S. Boebinger et al PRL 1995

Insulating behaviour at optimal dpoping

Questions About the Phase Diagram

Pseudogap: Preformed pairs?
Phase transition? Crossover? Order parameter



Transition to a Fermi liquid?

MIT and Disorder?

Pseudogap joins Tc curve or QCP ??

NMR studies of cuprates :
pseudogap, correlations, phase diagram: past and future?

- *Magnetic spin susceptibilities in NMR :*
  - Usual metals and superconductors
  - The case of cuprates:
  - Singlet spin pairing
  - Single spin fluid in the normal state
- *Dynamic susceptibilities and spin lattice relaxation :*
  - Magnetic correlations in the phase diagram
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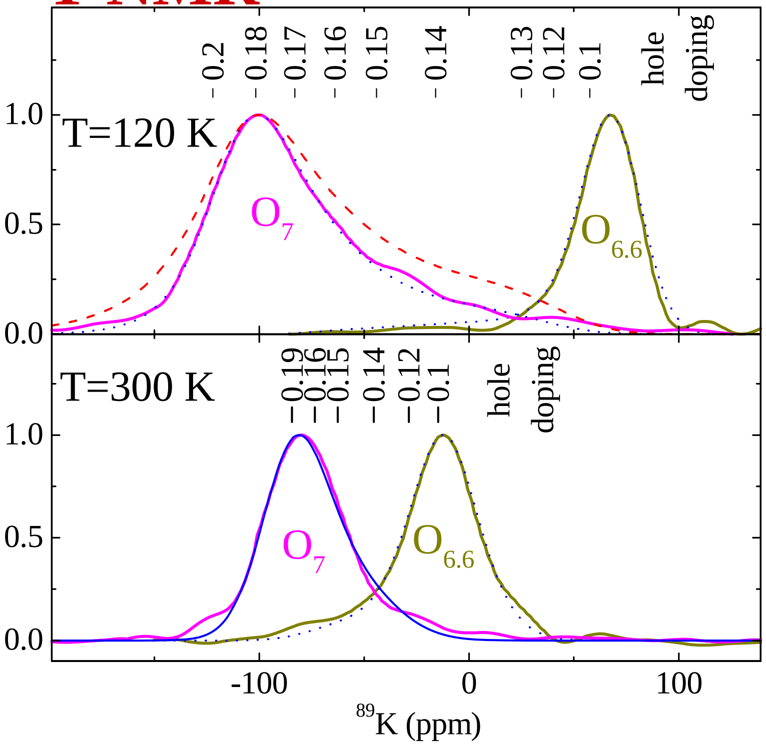
- *The pseudogap and questions on the phase diagram*
  - NMR as a local magnetic probe
- ***Pseudogap, MIT and disorder***
  - **NMR and high field transport measurements**
- *SC Fluctuations and pseudogap*
  - Some answers about the phase diagram

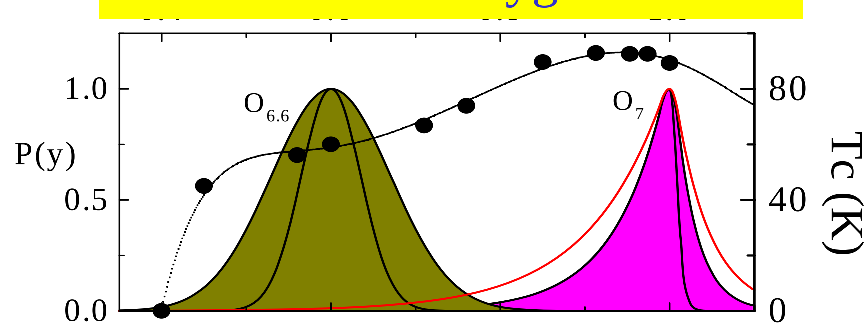
NMR Spectra: histograms of the hole content

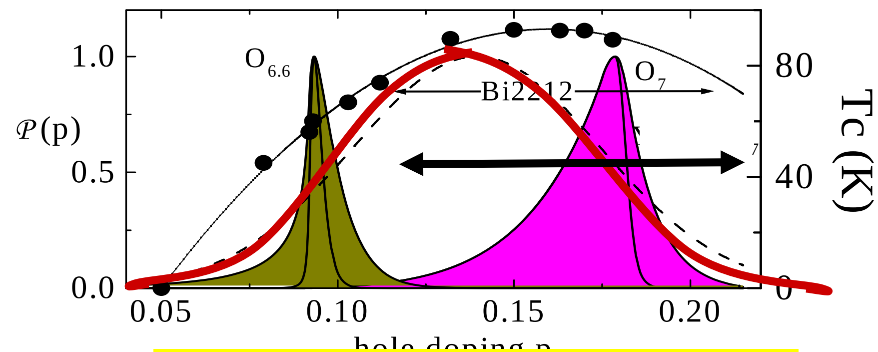
^{89}Y NMR



J. Bobroff, H.A,… PRL 2002

Distribution of oxygen content





Distribution of hole content

Detailed analysis of the spectra versus *T*

The maximum distribution of hole content is

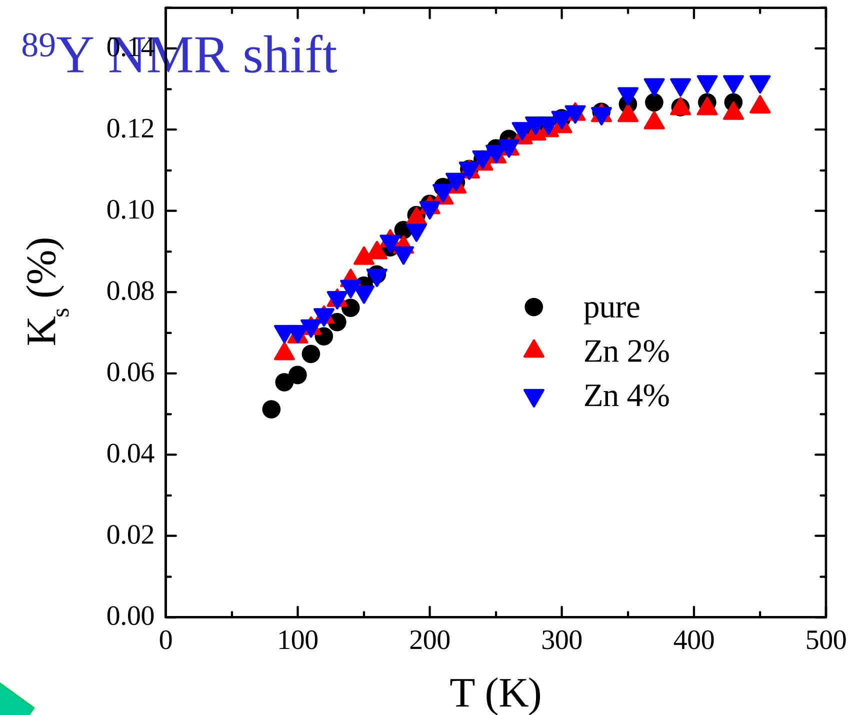
- much narrower than in Bi2212 (STM) or LSCO (RMN)
- seen **on large samples** (0.5g)
- likely of macroscopic origin

YBCO is very homogeneous
Only weak charge disorder

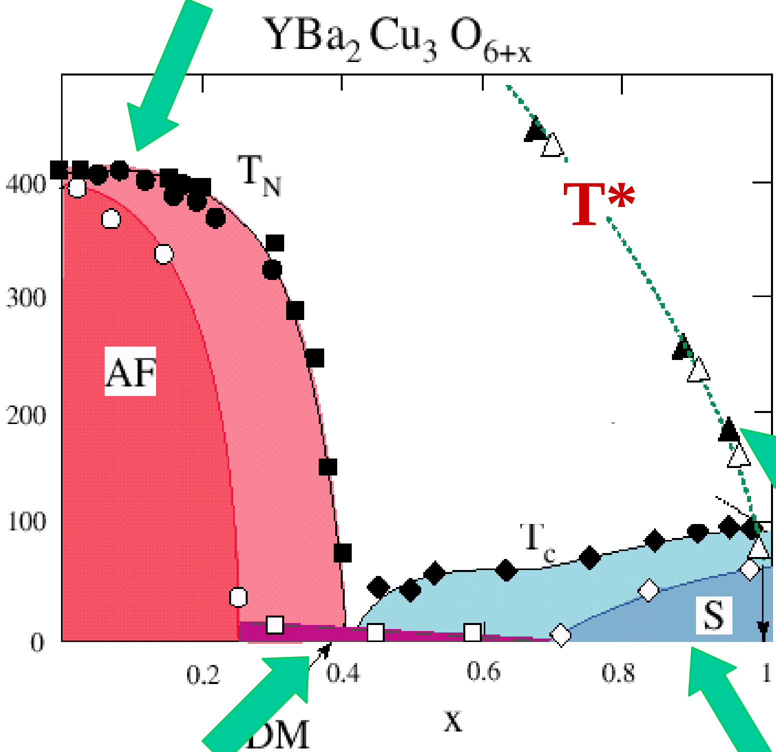
Influence of defects on T_c and on the pseudogap

$YBCO_{6+x}$ + 4%Zn → $YBCOZ_{6+x}$

No change of hole doping



Dilution effect on T_N



No change of T* : the pseudogap is not sensitive to disorder

H. Alloul , P. Mendels et al , PRL 67, 3140 (1991)

Increase of the disordered magnetism range

Large depression of T_c

Correlations between Magnetic and Superconducting Properties of Zn-Substituted $YBa_2Cu_3O_{6+x}$

H. Alloul,[1] P. Mendels,[1] H. Casalta,[1] J. F. Marucco,[2] and J. Arabski[1]

[1]*Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France*

[2]*Laboratoire des Composés Non Stoechiométriques, Université Paris-Sud, 91405, Orsay, France*

(Received 8 August 1991)

T_c and T_N (Néel) have been measured for a series of $YBa_2(Cu_{0.96}Zn_{0.04})_3O_{6+x}$ samples. The T variations of the homogeneous susceptibility χ_s of the CuO_2 planes, given by the shift of the ^{89}Y NMR line, are found to be nearly unchanged with respect to pure samples for $x > 0.5$, which implies that the charge transfer is negligibly modified by Zn, and that the magnetic pseudogap is not associated with superconducting pairing. Detection of an unusual Curie contribution to the ^{89}Y NMR width for $x \geq 0.5$ provides evidence that Zn induces magnetic moments in the CuO_2 planes, which play a role in the depression of T_c.

PACS numbers: 74.70.Hk, 75.20.Hr, 75.30.Kz, 76.60.Cq

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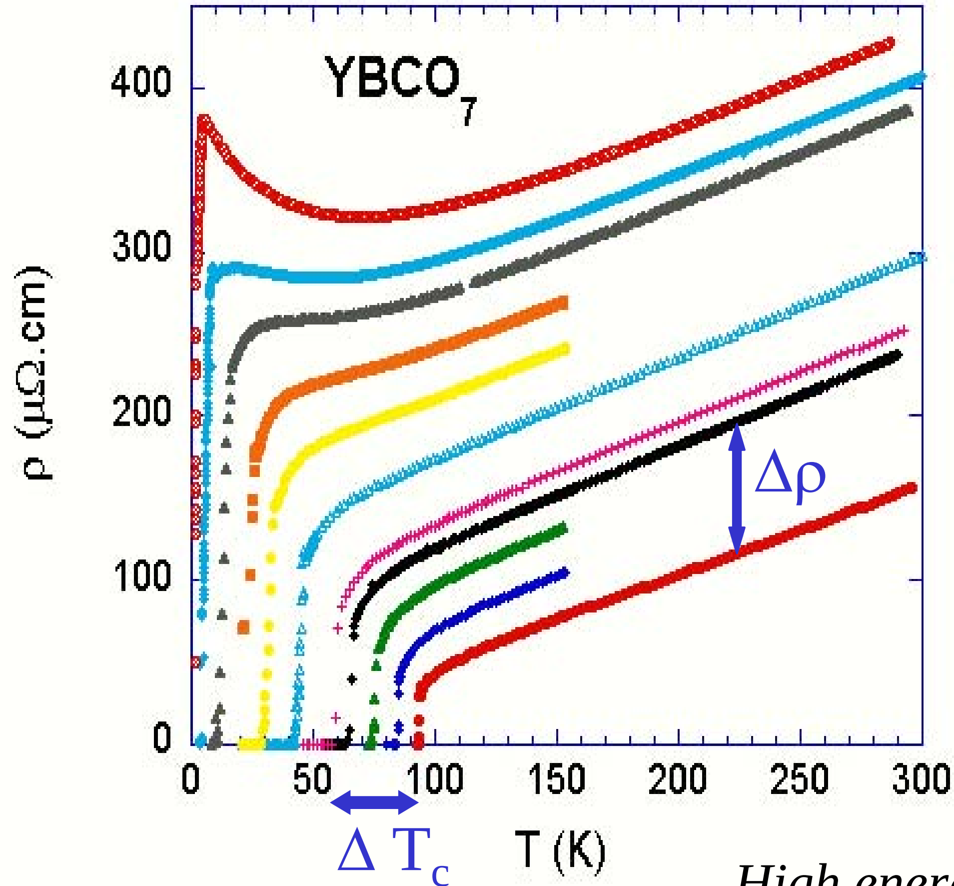
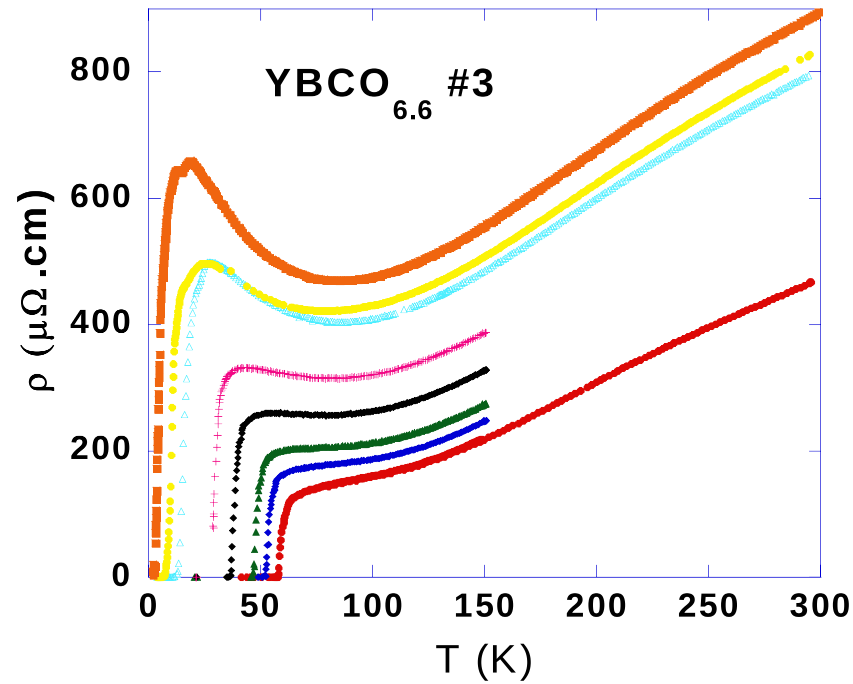
Inflence of irradiation defects on transport properties





Same single crystal

High energy (MeV) electron irradiation at low T

Cu and O vacancies in the CuO_2 Planes

The transition curves remain very sharp → **Homogeneous damage**

Excellent control of defect content

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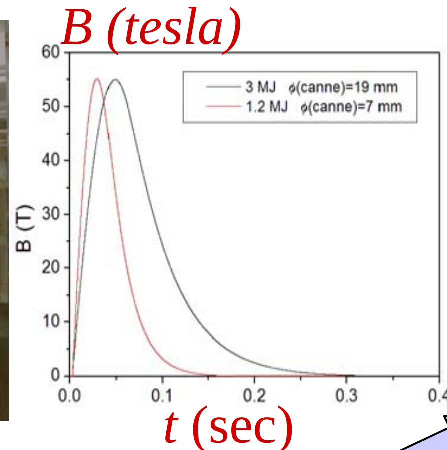
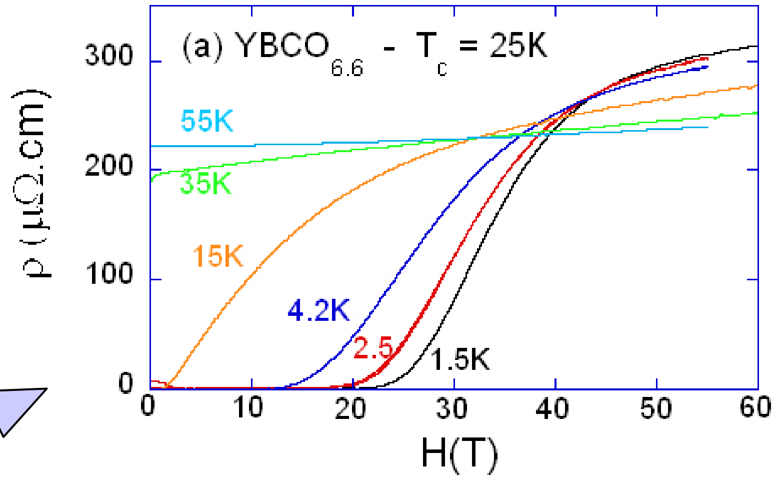
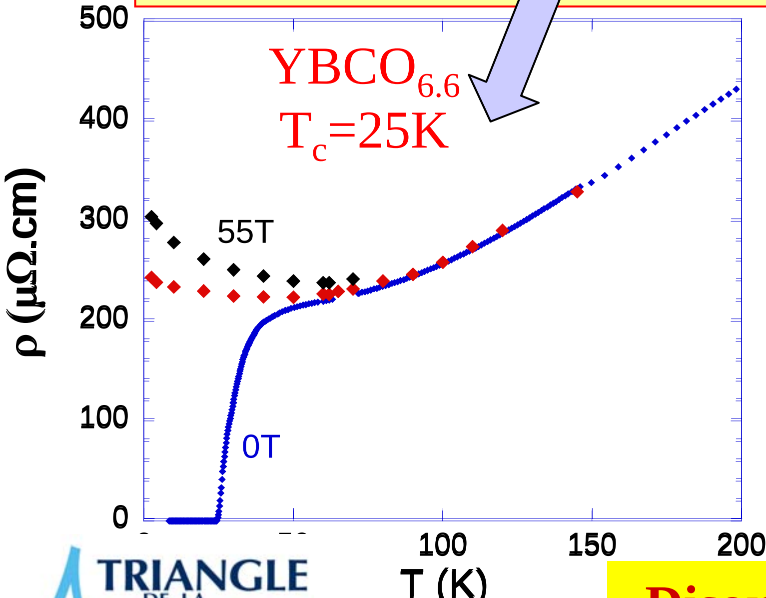
H. Alloul, Cours A. Georges CDF, 9/11/2010

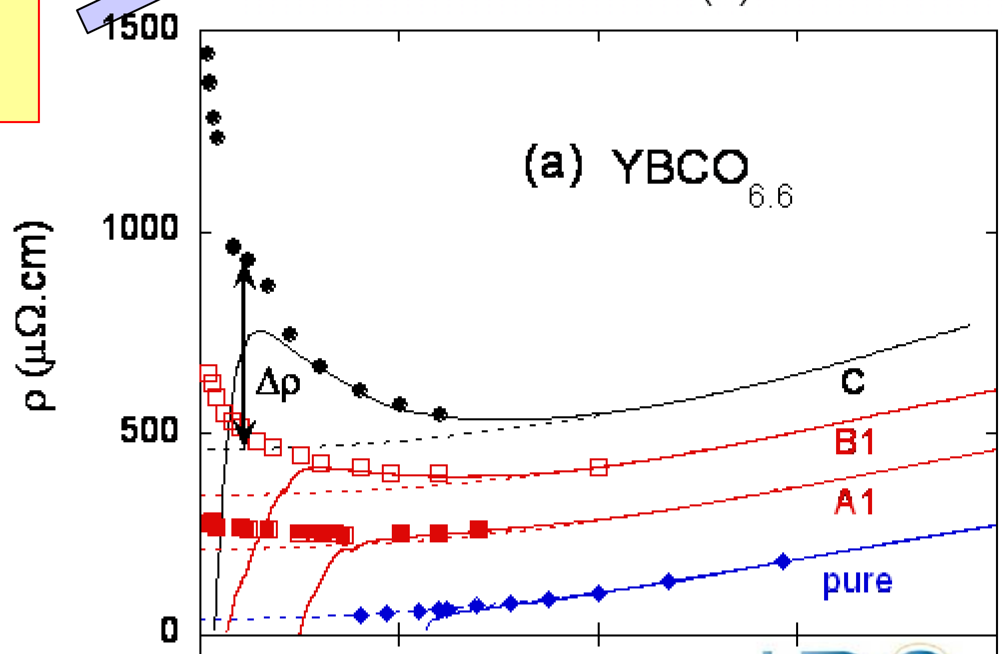
Pulsed magnetic field facility in Toulouse





High field suppresses SC reveals resistivity upturns





Disorder Induces MIT ?

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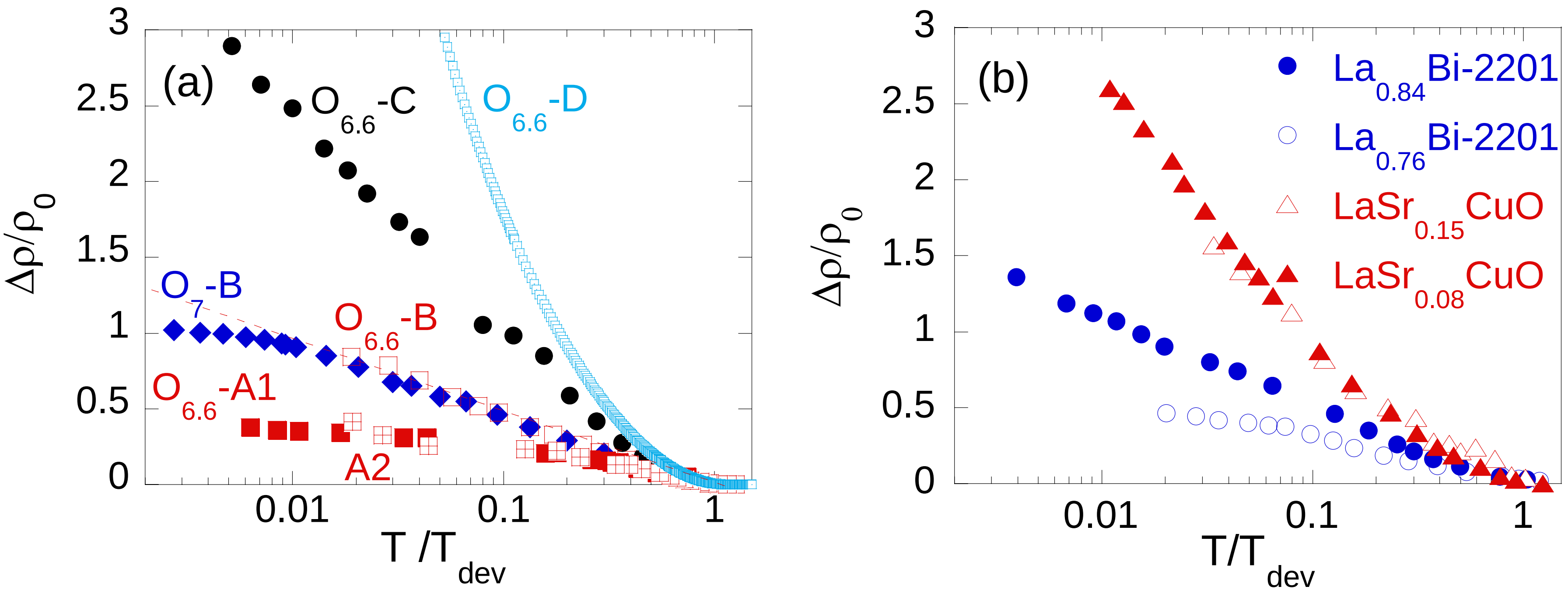
Irradiated YBCO

« Pure» low T_c Cuprates



Controlled disorder

System Specific disorder reduces T_c

The upturns are quantitatively similar

The disorder is not generic
MIT is driven by disorder

F. Rullier-Albenque, H. A. et al , EPL 2008.

TRIANGLE DE LA PHYSIQUE

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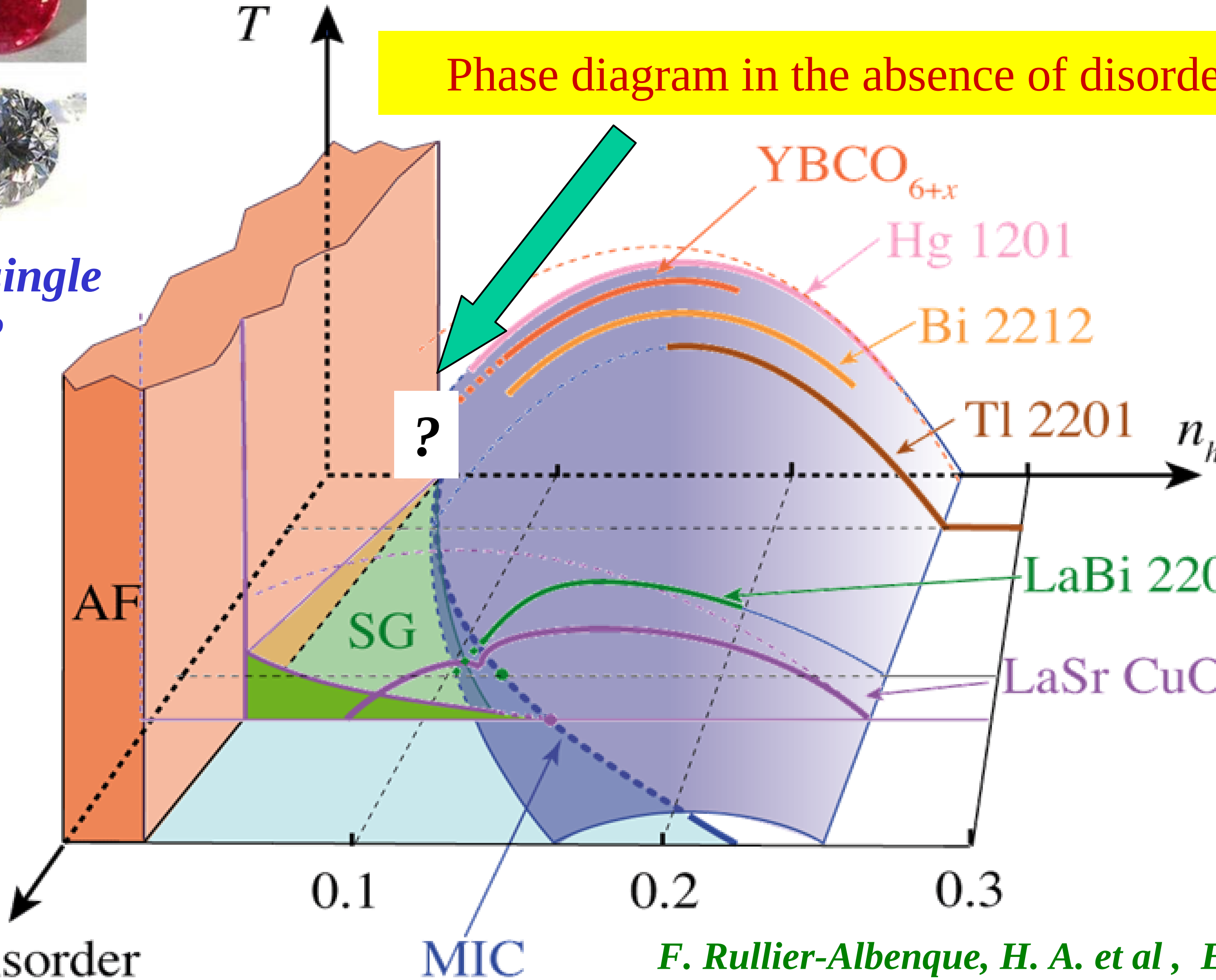
The various cuprate families

High quality single crystals ?

Phase diagram in the absence of disorder ??



F. Rullier-Albenque, H. A. et al , EPL 2008.

SG and MIT are determined by disorder

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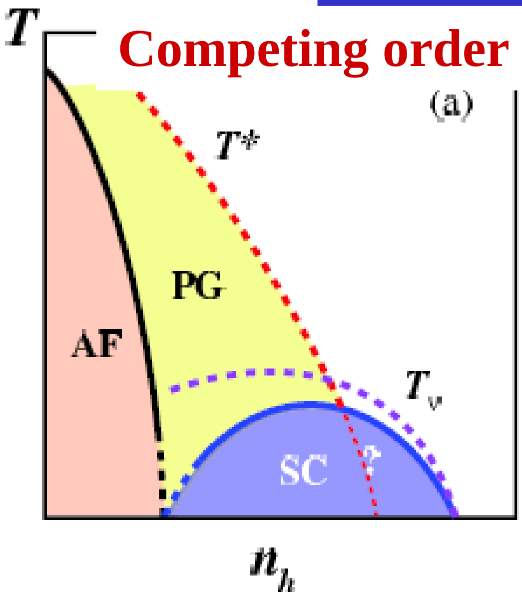
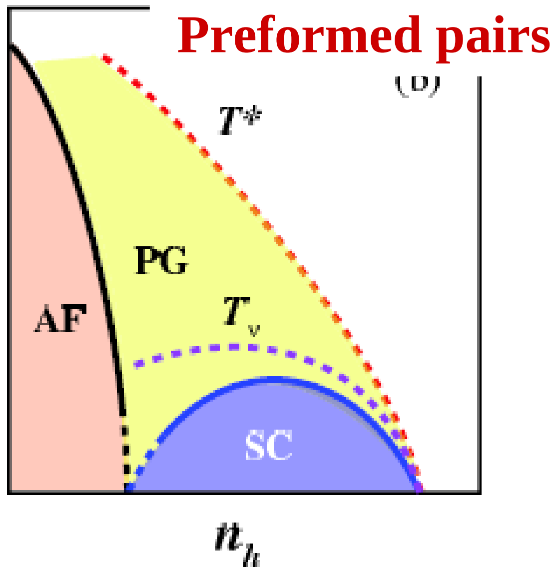
  - NMR and high field transport measurements
- ***SC Fluctuations and pseudogap***
  - **Some answers about the phase diagram**

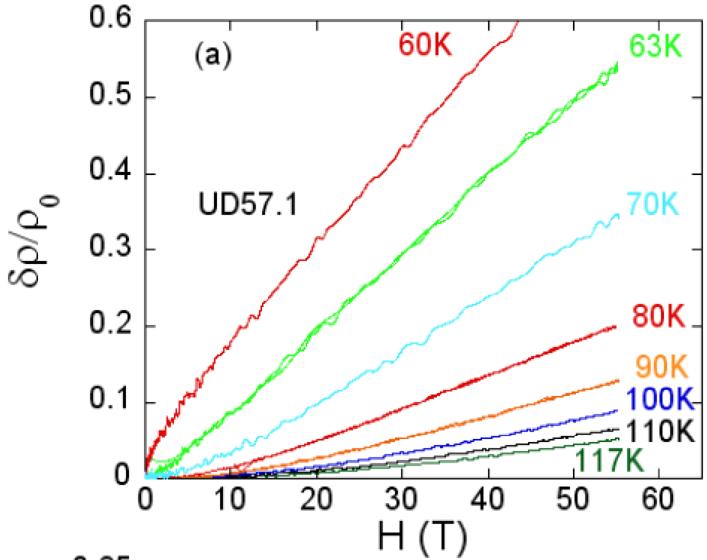
TRIANGLE DE LA PHYSIQUE

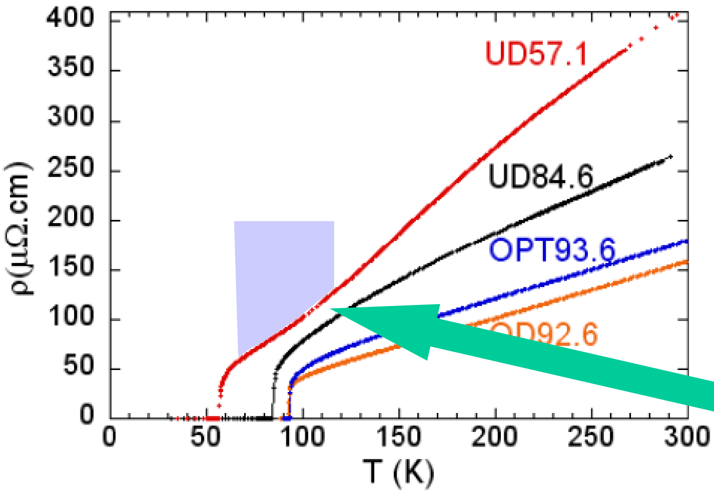
LPS ORSAY

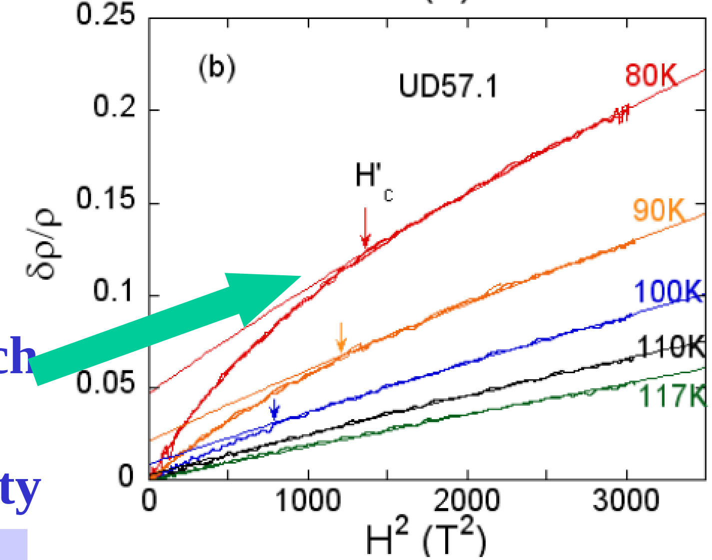
Superconducting fluctuations and pseudogap









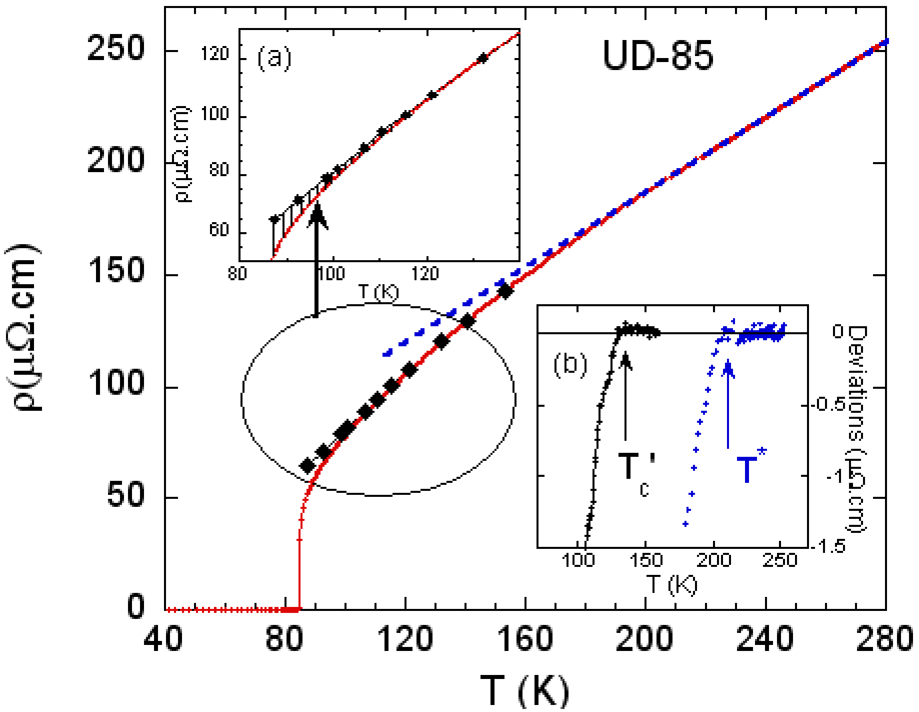


A new approach to determine SCF conductivity

Total suppression of SC fluctuations with large fields

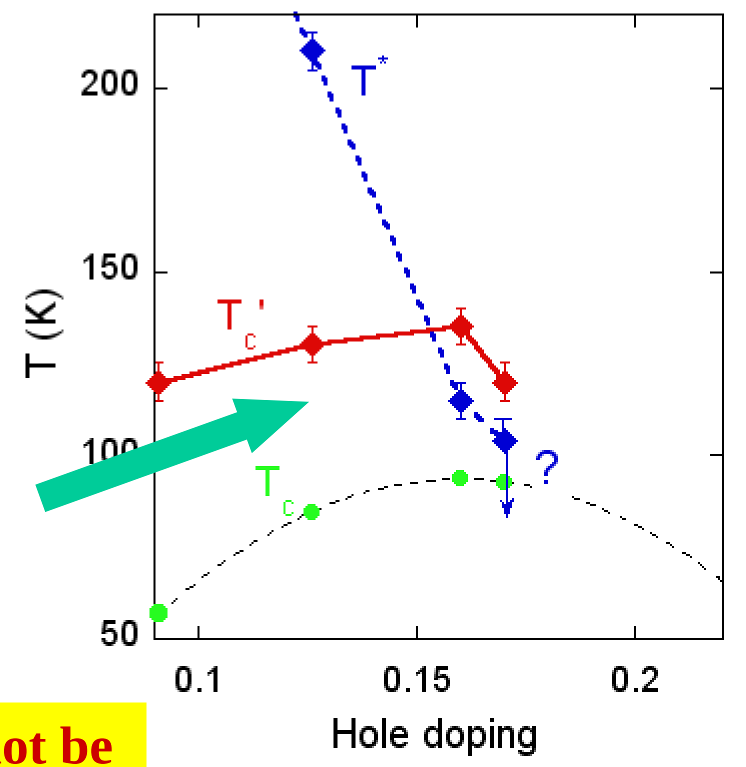
F. Rullier-Albenque, H. A. et al , PRL 2007

Determination of T^* and T'_c onset of SC Fluctuations

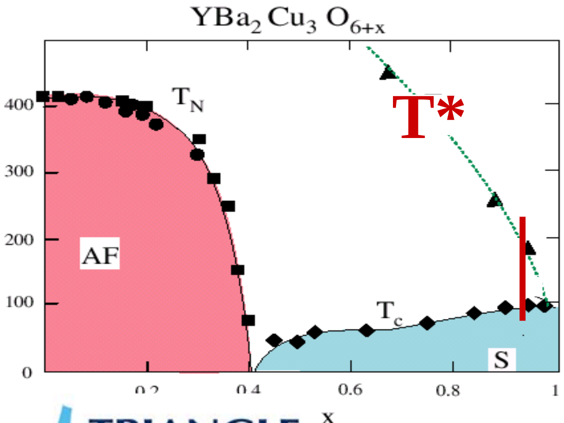


T^* and T'_c and σ_{SCF} (T,H) can be measured in a single experiment

H.A, F. Rullier-Albenque, EPL 2010



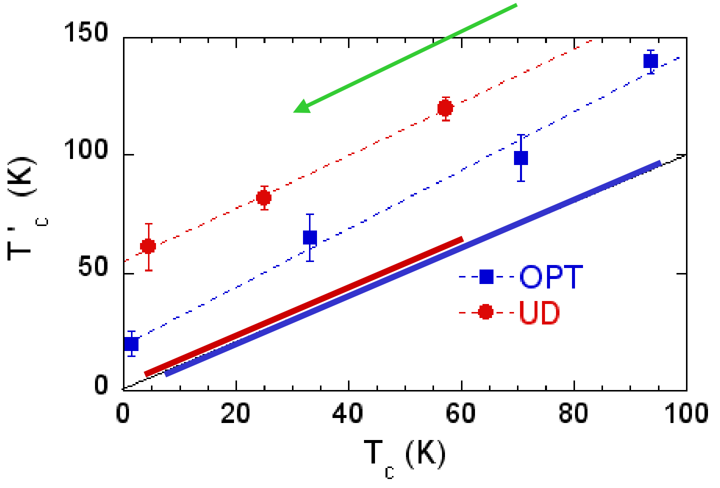
T^* crosses T'_c at optimal doping



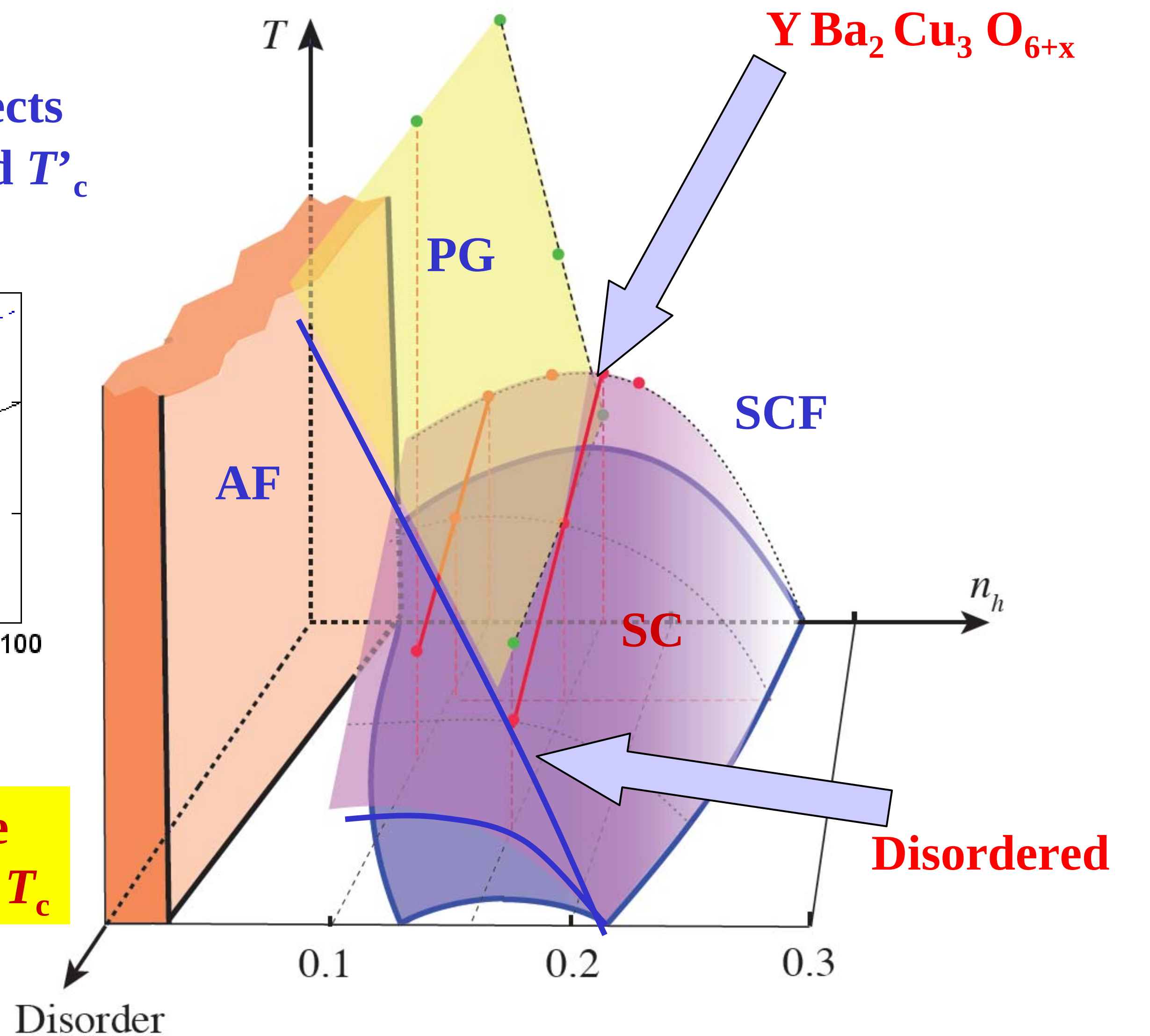
The pseudogap cannot be the onset of pairing

The real phase diagram of cuprates

Irradiation defects decrease of T_c and T'_c with disorder



SCF range does not change but increases with respect to T_c



H.A, F. Rullier-Albenque… EPL 2010

F. Rullier-Albenque, H.A et al to be published…

Some conclusions

- *Magnetic spin susceptibilities in NMR :*

  Singlet spin pairing

  Single spin fluid in the normal state

  The pseudogap is generic and robust to disorder

- *Dynamic susceptibilities and spin lattice relaxation :*

  Magnetic correlations up to the Optimal doping

  Metallic like at q=0, AF correlations for $q=(\pi/a , \pi/a)$

  d- wave SC

- *The pseudogap and questions on the phase diagram*

  Importance of disorder in the phase diagram

  MIT and SG phases governed by disorder

- *SC Fluctuations and pseudogap*

  SC Fluctuations follow Tc versus hole doping , remain with disorder

  A preformed pair scenario does not apply

  Pseudogap is intimately linked with magnetism (competing order?)

  NMR will be helpful to check possible models