A Perspective of Sung-Ik Lee's Major Contributions to Cuprate Superconductivity

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1. **Overview**

  -- Materials synthesis; multi-layer cuprates; infinite-layer electron-type cuprates

2. **Multi-layer hole-type cuprate superconductors**

  -- Increasing anisotropy, charge imbalance, competing orders, and quantum & thermal fluctuations with increasing number of CuO_2 layers

3. **Infinite-layer electron-type cuprate superconductors**

  -- Three-dimensional superconductivity; nodeless energy gap; vortex images & intra-vortex pseudogap

4. **Summary**

1. Overview
2. Multi-layer hole-type cuprate superconductors
3. Infinite-layer electron-type cuprate superconductors
4. Summary

1. Overview of Sung-Ik Lee's Research in Cuprate Superconductivity

- **Materials synthesis**

-- Single crystal growth with self-flux method & additional annealing/processing
-- Polycrystalline sample growth w/ solid-state reaction in a high-pressure chamber
-- Chemical substitutions in charge reservoir & quantum impurities in CuO_2 planes
-- Intercalation of organic chains into Bi-2212

- **Multi-layer hole-type cuprate superconductors**

-- Thermodynamic properties
-- Vortex dynamics
-- Increasing anisotropy, charge imbalance, competing order, and thermal & quantum fluctuations with increasing number of CuO_2 planes per unit cell
-- Importance of inter-layer coupling to cuprate superconductivity

- **Infinite-layer electron-type cuprate superconductors**

-- Three-dimensional superconductors (ξ_c > c-axis lattice constant c_0)
-- Node-less energy gap
-- First (and only to-date) observation of vortex images & intra-vortex pseudogap in electron-type cuprates

1. Overview
2. Multi-layer hole-type cuprate superconductors
3. Infinite-layer electron-type cuprate superconductors
4. Summary

2. Multilayer Hole-Type Cuprate Superconductors

- Hole-type multi-layer cuprate superconductors synthesized:

$HgBa_2Ca_{n-1}Cu_nO_{2n+3}$: Hg-12(n-1)n system with n = 1 ~ 5
$TlBa_2Ca_{n-1}Cu_nO_{2n+3}$: Tl-12(n-1)n system with n = 1 ~ 4
$Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$: Tl-22(n-1)n system with n = 1 ~ 4
$CuBa_2Ca_{n-1}Cu_nO_{2n+3}$: Cu-12(n-1)n system with n = 1 ~ 4
$Ca_{1-x}Na_xCuO_2Cl_2$: Na-CCOC system

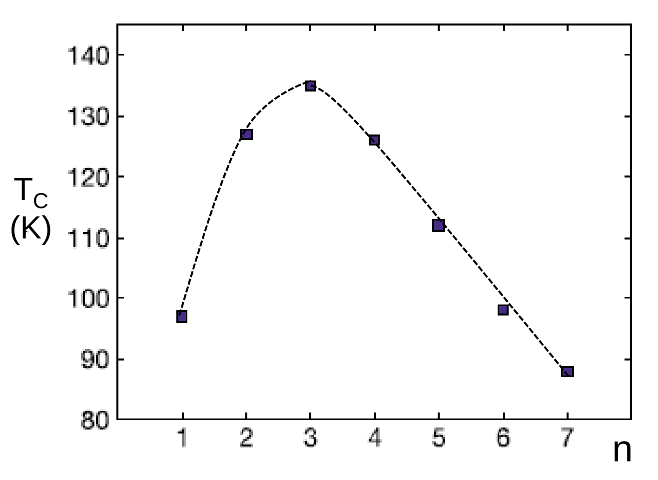
Ba layer ↔ $Ba_{1-x}Sr_x$
Hg layer ↔ $Hg_{1-x}Pb_x$
O layer ↔ $O_{1-x}F_x$

- Hamiltonian for coexisting superconductivity and competing orders:

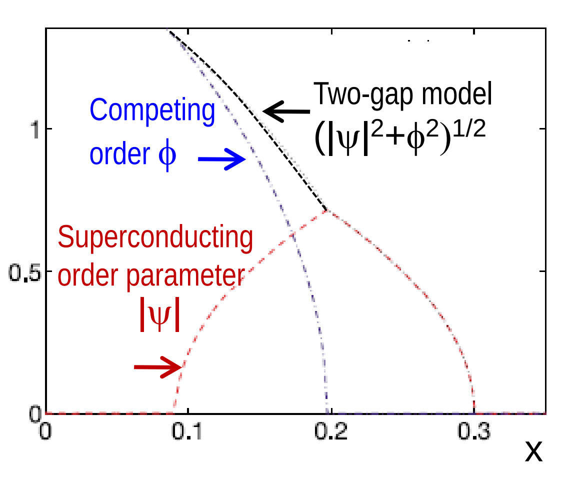
$$\mathcal{H}(\Phi,\Psi) = \mathcal{H}(\Phi) + \mathcal{H}(\Psi) + g|\Phi|^2|\Psi|^2 \qquad (g>0)$$

S. Chakravarty *et al*, Nature 428 (2004)

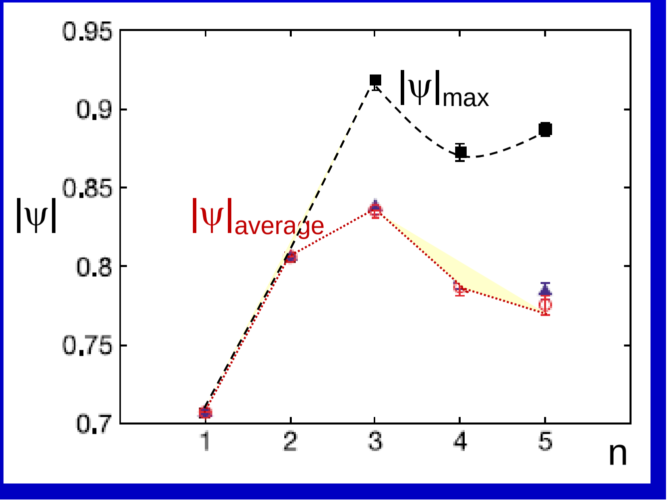
"T_C dome" as a function of n for the same family of cuprates



Order parameter-vs-doping (x) in a single-layer cuprate



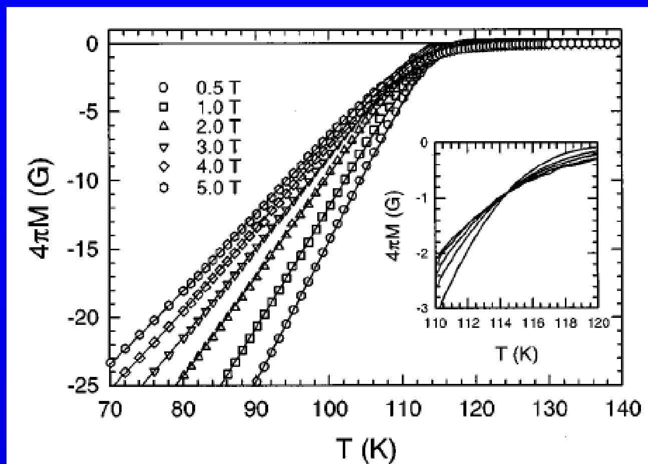
Superconducting parameter-vs-n with charge imbalance



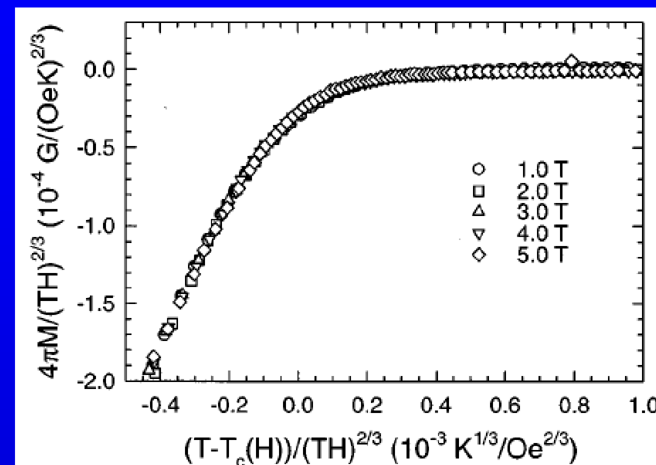
Key findings from investigations of hole-type multi-layer cuprate superconductors

- Increasing anisotropy & thermal fluctuations with increasing n:

Hg-1212



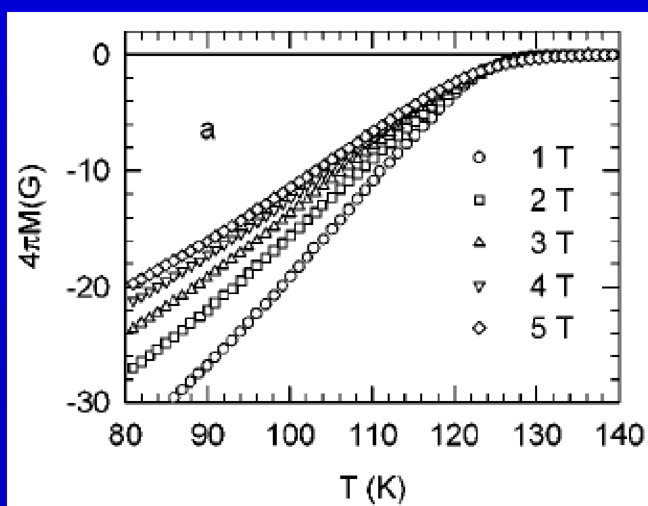
3D critical scaling of M(T,H) →



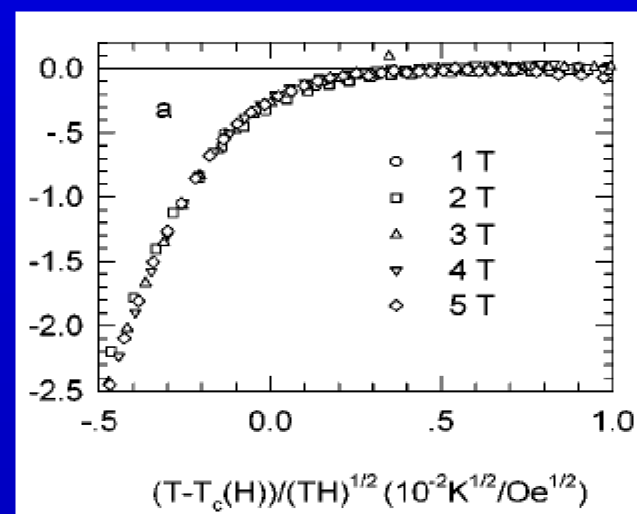
M.S. Kim et al., Phys. Rev. B 53, 9460 (1996)

↕

Hg-1223



2D critical scaling of M(T,H) →



Y. Zhou et al., Phys. Rev. B 60, 13094 (1999)

Key findings from investigations of hole-type multi-layer cuprate superconductors – continued

- Comparative studies of the thermodynamic properties of different systems:

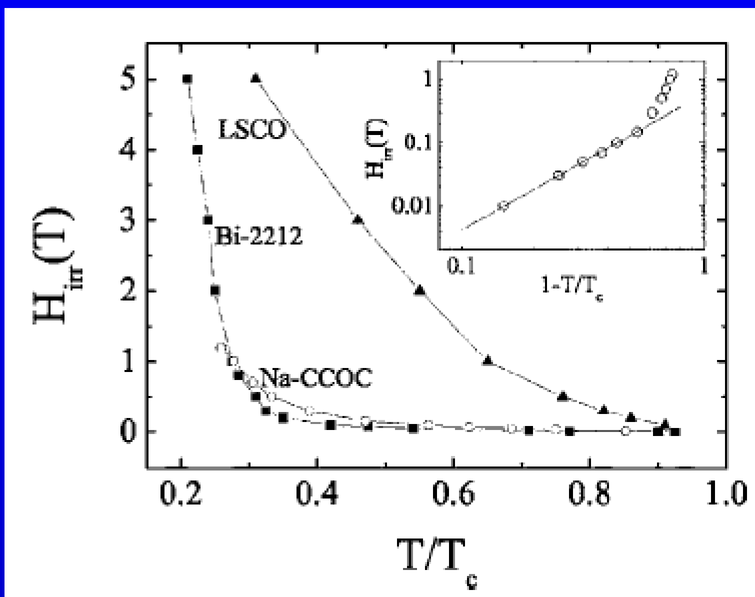
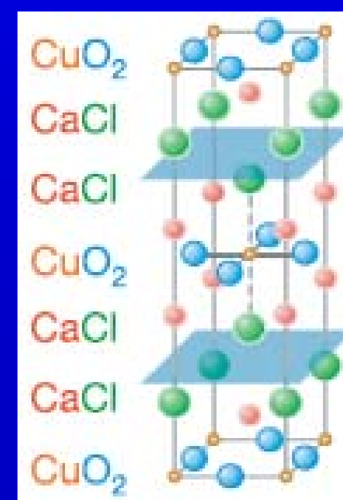


TABLE I. Thermodynamic properties of $Ca_{1.82}Na_{0.18}CuO_2Cl_2$ (Na-CCOC), $La_{1.82}Sr_{0.18}CuO_4$ (LSCO) (Ref. 24), and $Bi_2Sr_2CaCu_2O_{8-\delta}$ (Bi-2212) (Ref. 27).

	T_c^{onset} (K)	κ	$H_c(0)$ (Oe)	$H_{c2}(0)$ (T)	$\lambda_{ab}(0)$ (Å)	ξ_{ab} (Å)
Na-CCOC	27	89.0	1202	16.9	4380	44.1
LSCO	30	75	2170	21	2380	33
Bi-2212	85	266	9753	156	2438	9.5

H.C. Kim et al., Phys. Rev. B 72, 224510 (2005)



→ Na-CCOC is as anisotropic as Bi-2212, with an even lower superfluid density due to Cl at the apical site.

Key findings from investigations of hole-type multi-layer cuprate superconductors – continued

- Cu-NMR & equilibrium magnetization studies show charge imbalance for n ≥ 3.

The hole doping level for the inner CuO_2 planes (δ_i) is always smaller than that for the outer CuO_2 planes (δ_o), $\delta_i < \delta_o$; and (δ_o/δ_i) increases with increasing n.

H. Kotegawa et al., Phys. Rev. B 64, 064515 (2001);
H. Kotegawa et al., Phys. Rev. B 69, 014501 (2004);
H. Mukuda et al., Phys. Rev. Lett. 96, 087001 (2006);
M.S. Kim et al., Physica C 364-365, 228 (2001).

- μSR shows increasing antiferromagnetic fluctuations (& competing orders) for n ≥ 3

The inner CuO_2 planes are generally underdoped and exhibit increasing antiferromagnetic fluctuations with increasing n.

K. Tokiwa et al., Int. J. Mod. Phys. B 17, 3540 (2003).

- Vortex dynamics: The irreversibility field $[H_{irr}(T)/H_{c2}(0)]$ for H || c-axis decreases with increasing n, whereas the static exponent ν associated with the vortex phase transition increases with increasing n, indicating increasing thermal fluctuations with increasing n.

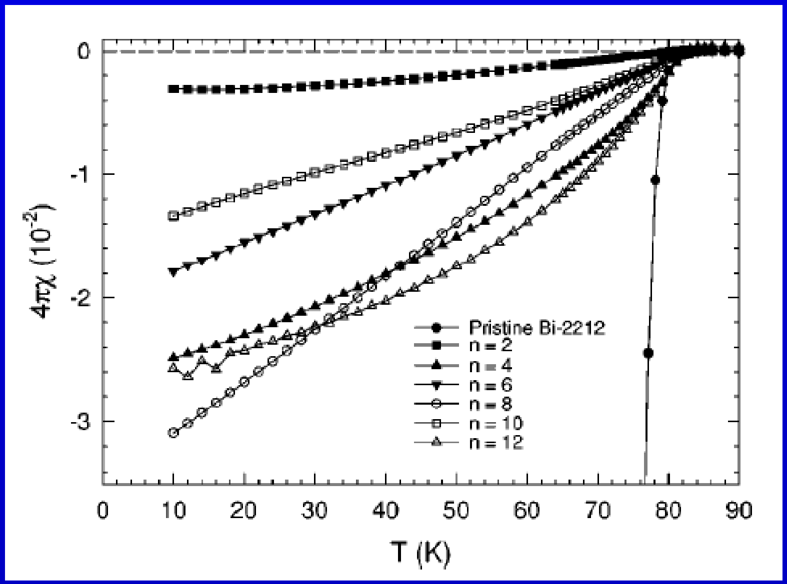
A.D. Beyer et al., Phys. Rev. B 76, 140506(R) (2007).
M.S. Park et al., Phys. Rev. B 77, 024519 (2008).

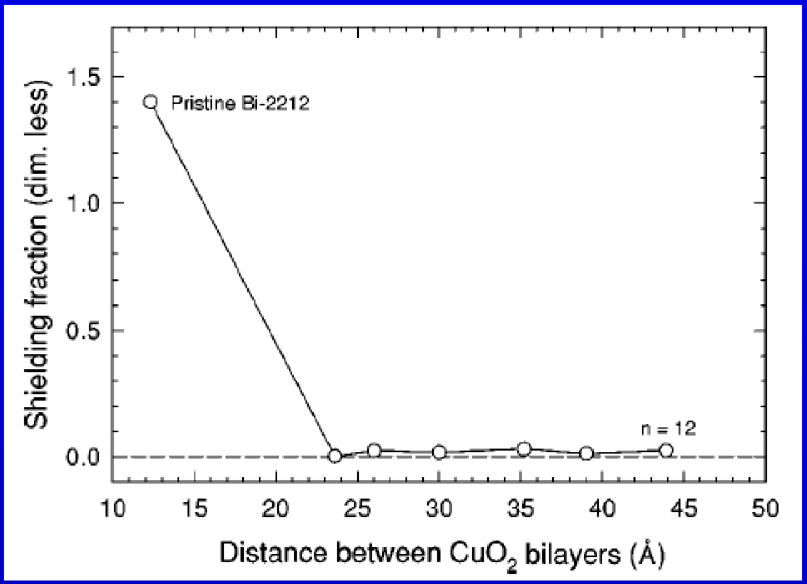
Controlling the CuO_2 interlayer coupling by intercalation of organic chains into Bi-2212

- Evidence of non-existence of superconductivity in an isolated CuO_2 bilayer

Intercalation of organic chains of $[Py\text{-}C_nH_{2n+1}]HgI_4$ into Bi-2212 samples. XRD and HRTEM indicated that the organic chains are intercalated uniformly and commensurately into every Bi-O bilayer and they expand the distance between the CuO_2 bilayers by 11.3 Å, 13.7 Å, 17.7 Å, 22.9 Å, 26.7 Å, and 31.6 Å for n = 2, 4, 6, 8, 10, and 12, respectively.

→ No change in T_c with n, but the shielding fraction diminishes with increasing n, suggesting that an isolated CuO_2 bilayer is not superconducting and that next-nearest-neighbor CuO_2 interlayer coupling is essential for cuprate superconductivity.

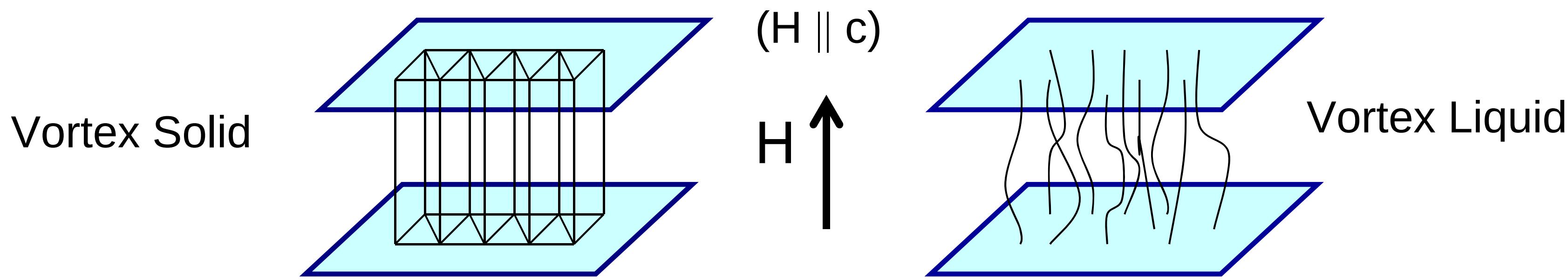




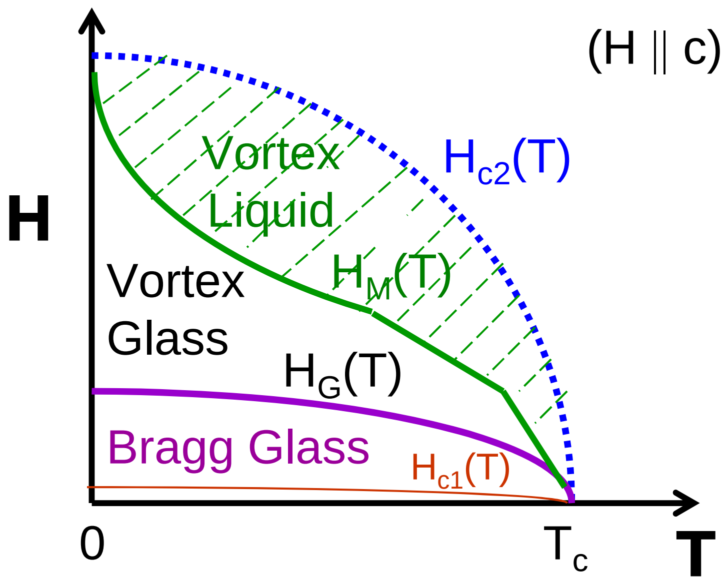
M.S. Kim et al., Phys. Rev. B 63, 092507 (2001).

Consequence of competing orders on the vortex dynamics of cuprate superconductors

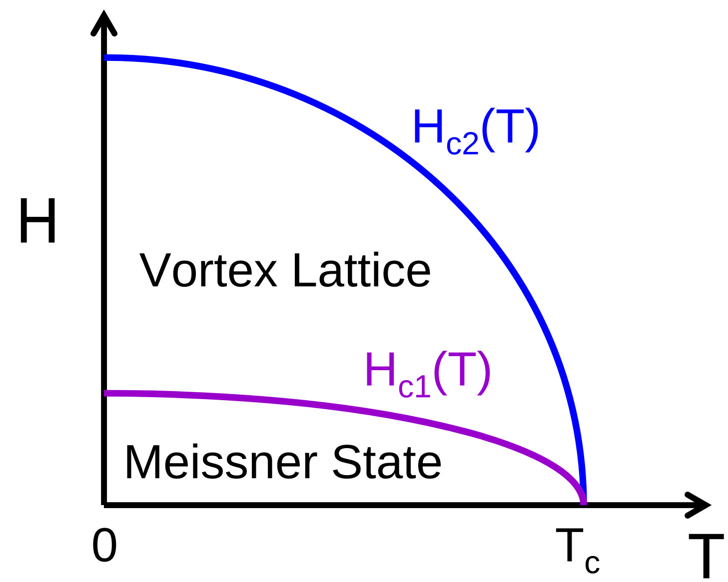
- Competing orders could reduce the superconducting stiffness, giving rise to enhanced thermal and disorder fluctuations in the vortex state → Thermally induced vortex-liquid state and disorder-induced vortex-glass & Bragg glass for H || c.



High-Temperature Superconductors



Conventional Type-II Superconductors

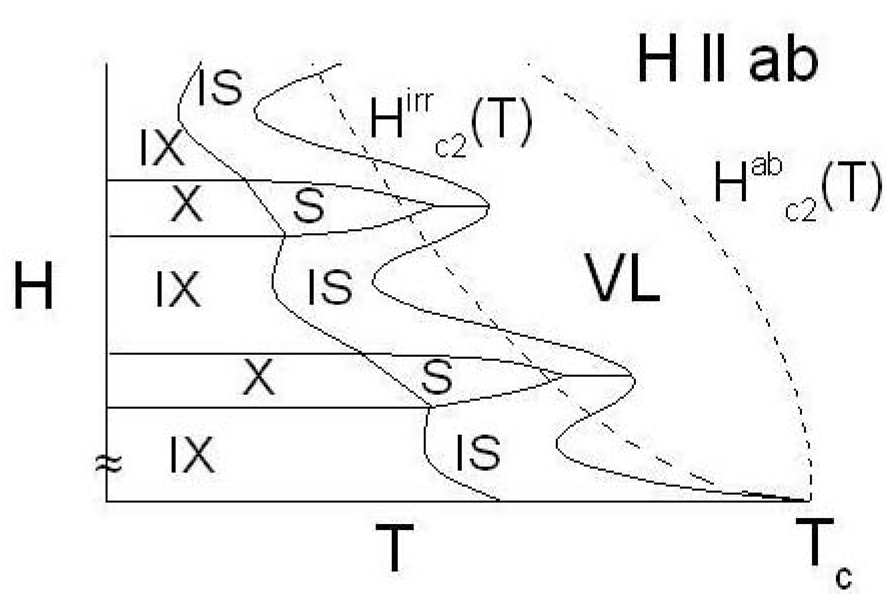


Investigation of the vortex-state quantum fluctuations in cuprate superconductors

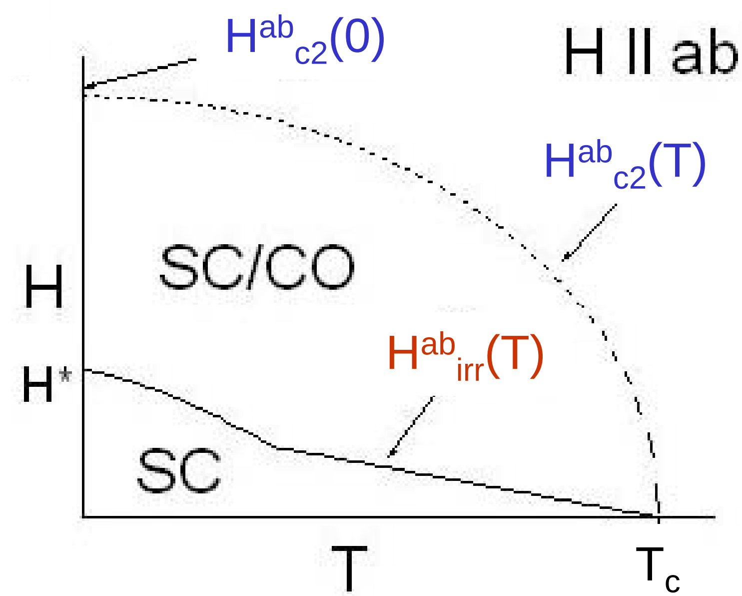
$T \to 0$ (suppressed thermal fluctuations);

H || ab (weak disorder fluctuations due to intrinsic pinning of the CuO_2 planes).

Weak quantum fluctuations



Strong quantum fluctuations



[Balents & Nelson, (1993, 1994)]

S : vortex smectic

X : vortex crystal/glass

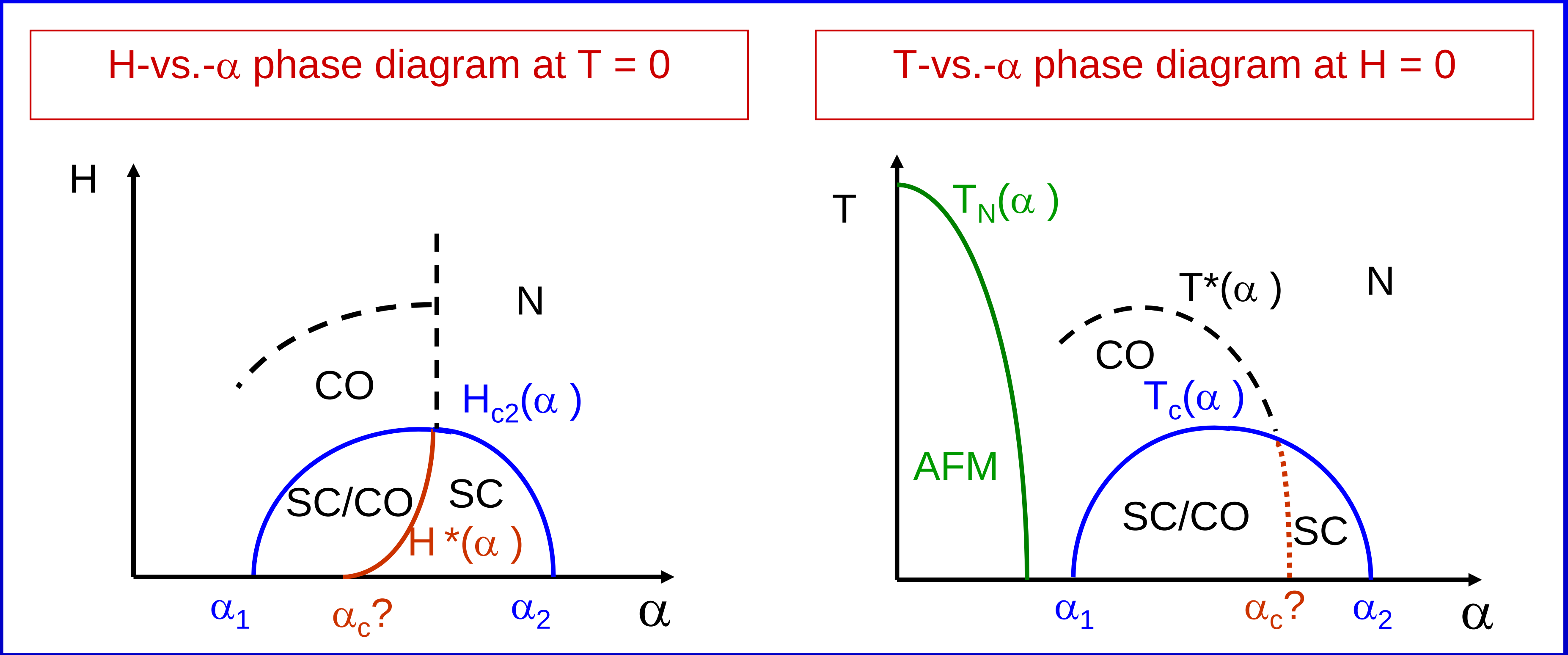
VL: vortex liquid

IS : incommensurate vortex smectic

IX : incommensurate vortex crystal/glass

Natural consequences of competing orders

- Proximity to quantum criticality & quantum fluctuations.
- Reduced superconducting stiffness & extreme type-II nature.
- Unconventional low-energy excitations & pseudogap phenomena.
- Non-universal phenomena among different cuprates.

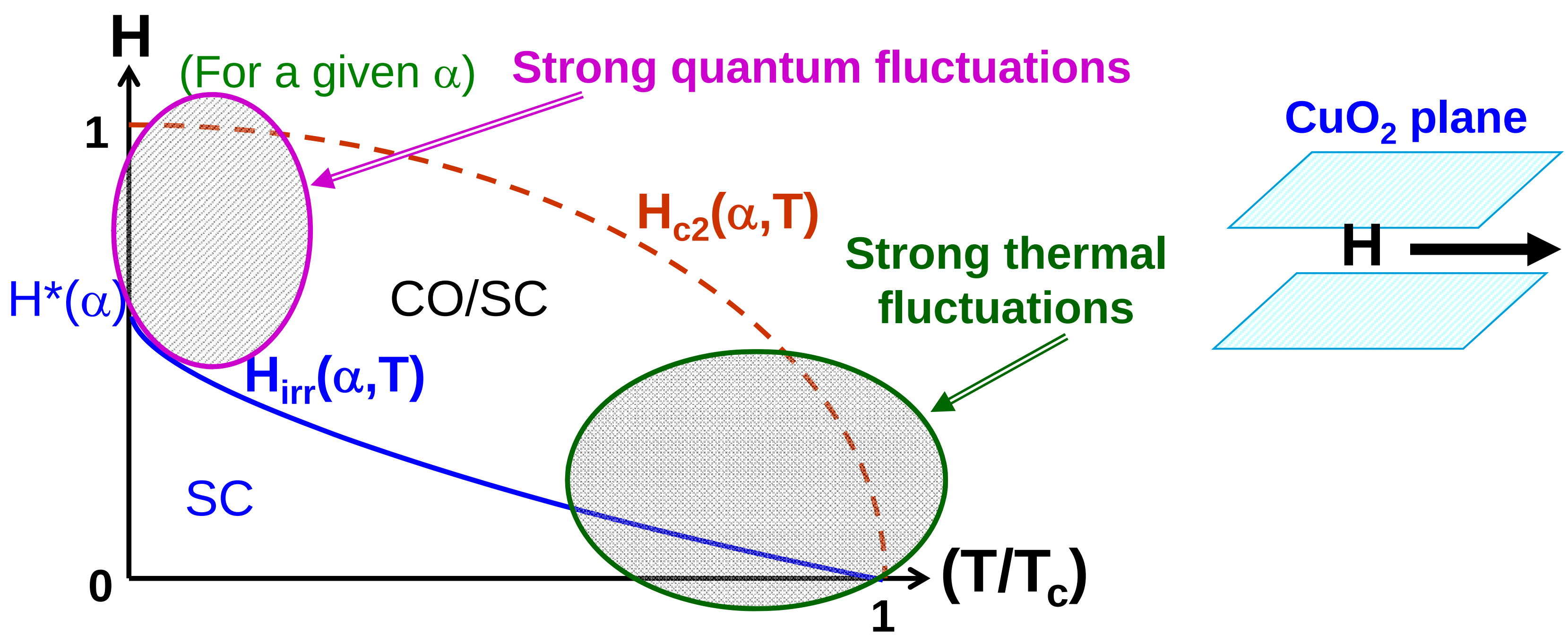


α: material parameter, such as the doping level, anisotropy, number of CuO_2 planes per unit cell, disorder, etc.

Probing quantum fluctuations in cuprate superconductors

To probe the quantum fluctuations induced by competing orders, we take $T \rightarrow 0$ to suppress thermal fluctuations and have H || ab to minimize disorder fluctuations.

H-vs.-(T/T_c) phase diagram (H || ab) for a given material parameter α



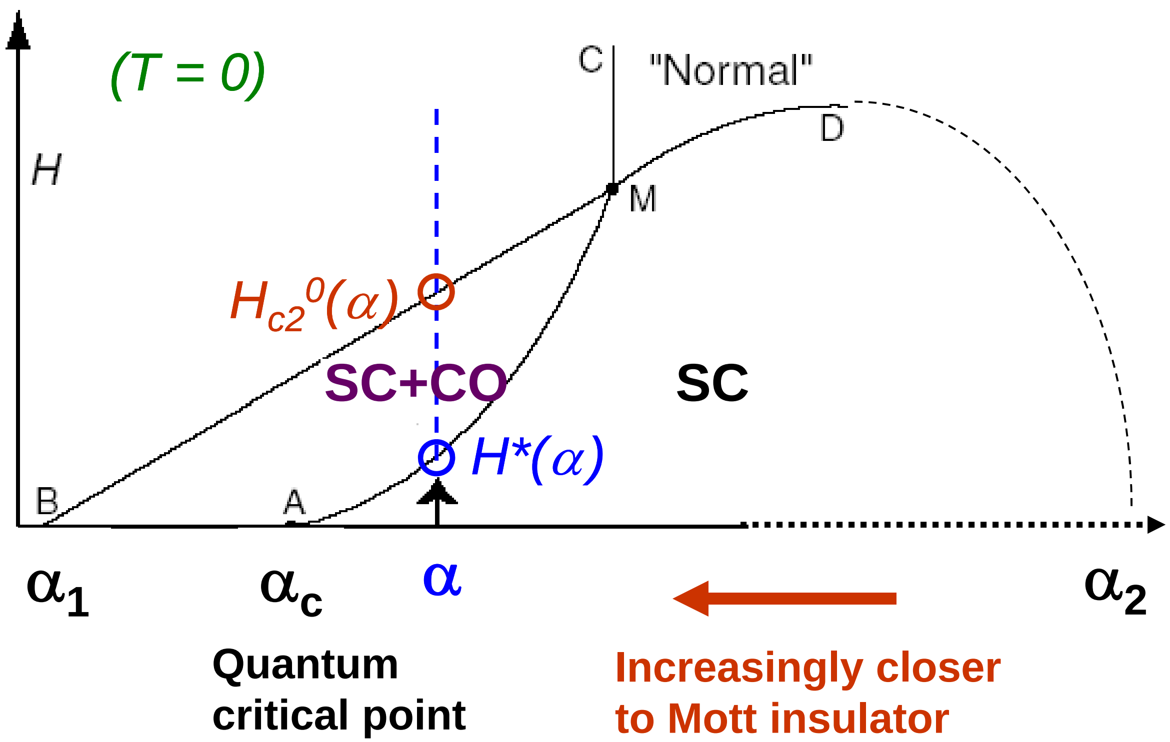
High-field phase diagrams of the cuprates (H || ab)

- Identify a characteristic field H* that separates SC from SC/CO at T = 0:

  ⟹ From the irreversibility field $H_{irr}(T,\alpha)$, with $H^*(\alpha) \equiv H_{irr}(T \to 0,\alpha)$.

- For comparison among different cuprates, find their $H_{c2}(T,\alpha)$ for H || ab:

  ⟹ $H_{c2}^{0}(\alpha) = H_{c2}(T \to 0,\alpha)$.



α: a material-dependent parameter.

How to compare $H^*(\alpha)$ among different cuprates?

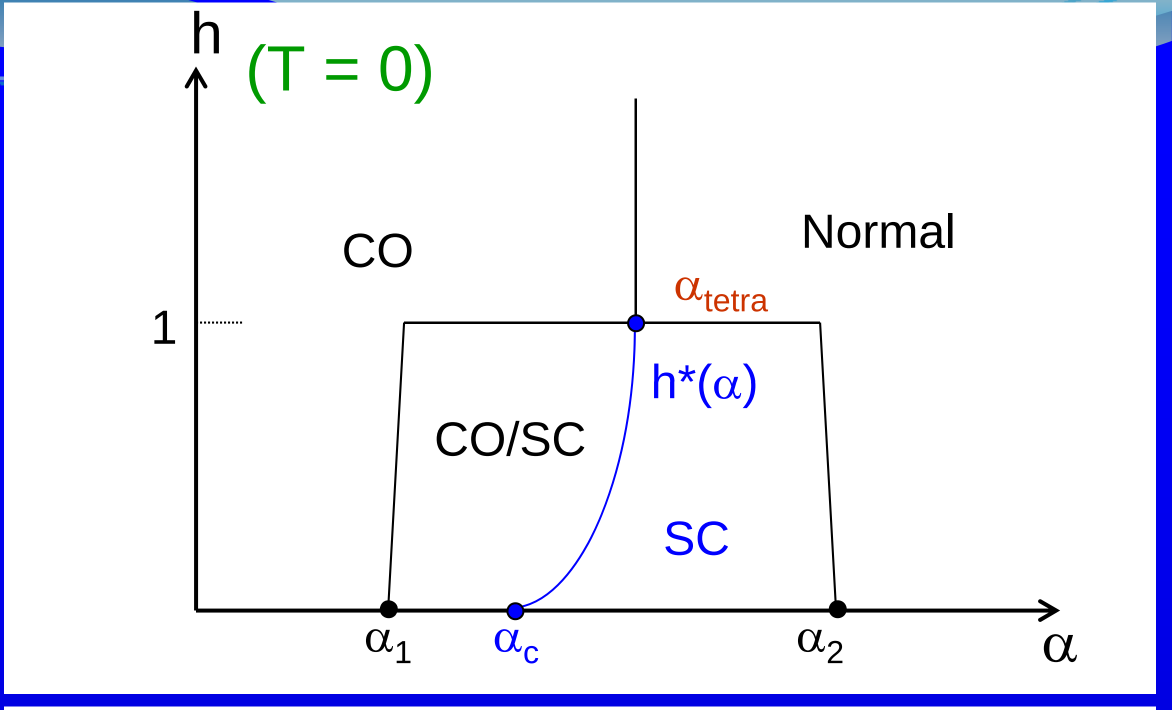
What is α for each cuprate?

Transformed h-vs.-α phase diagram:

$$h^*(\alpha) \equiv H^*(\alpha)/H_{c2}^0(\alpha)$$

$h^* \to 1$: mean-field limit

$h^* \to 0$: strong quantum fluctuations



Reduced h-vs.-(T/T_c) phase diagram:

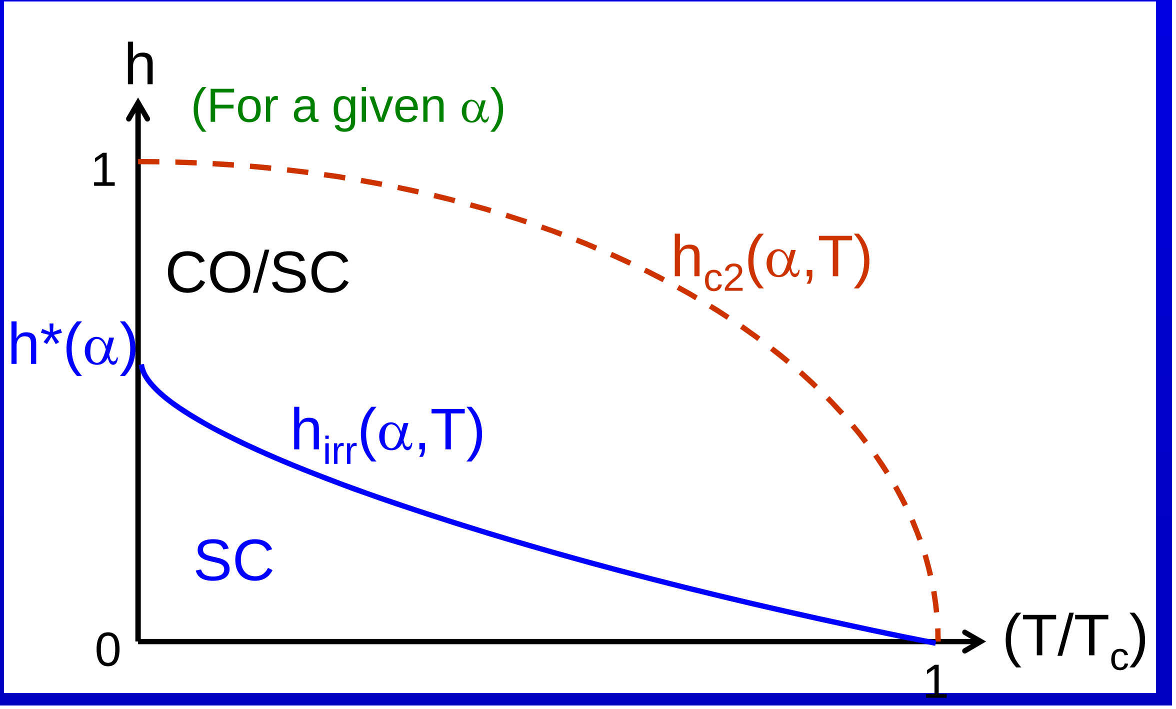
For H || ab:

$H_{c2}^0(\alpha) = H_p(\alpha)$

$= \Delta_{SC}(\alpha)/[2^{1/2}\mu_B]$

$h \equiv H/H_p(\alpha)$;

$h^*(\alpha) \equiv H^*(\alpha)/H_p(\alpha)$;



Physical significance of the material-dependent parameter α

$$\alpha \equiv \gamma^{-1} (\delta_o/\delta_i)^{-(n-2)} \delta, \qquad \text{for } n > 2;$$

$$\alpha \equiv \gamma^{-1} \delta, \qquad \text{for } n \le 2;$$

$\gamma = (\xi_{ab}/\xi_c)$: electronic anisotropy;

n: # of CuO_2 layers per unit cell;

δ : nominal doping level per CuO_2 layer;

(δ_o/δ_i): charge imbalance ratio;

δ_o: doping level per outer layer;

δ_i: doping level per inner layer.

- The outer- and inner-layer doping levels δ_o and δ_i in multilayered cuprate superconductors can be determined from NMR. [Kotegawa et al. (2001)]
- $\delta_o > \delta_i$ for n > 2 and the charge imbalance ratio (δ_o/δ_i) increases with increasing n. [Kotegawa et al. (2001)]
- Charge imbalance in multilayered cuprate superconductors results in stronger competing orders in the inner layer(s). [Chakravarty et al. (2004)]

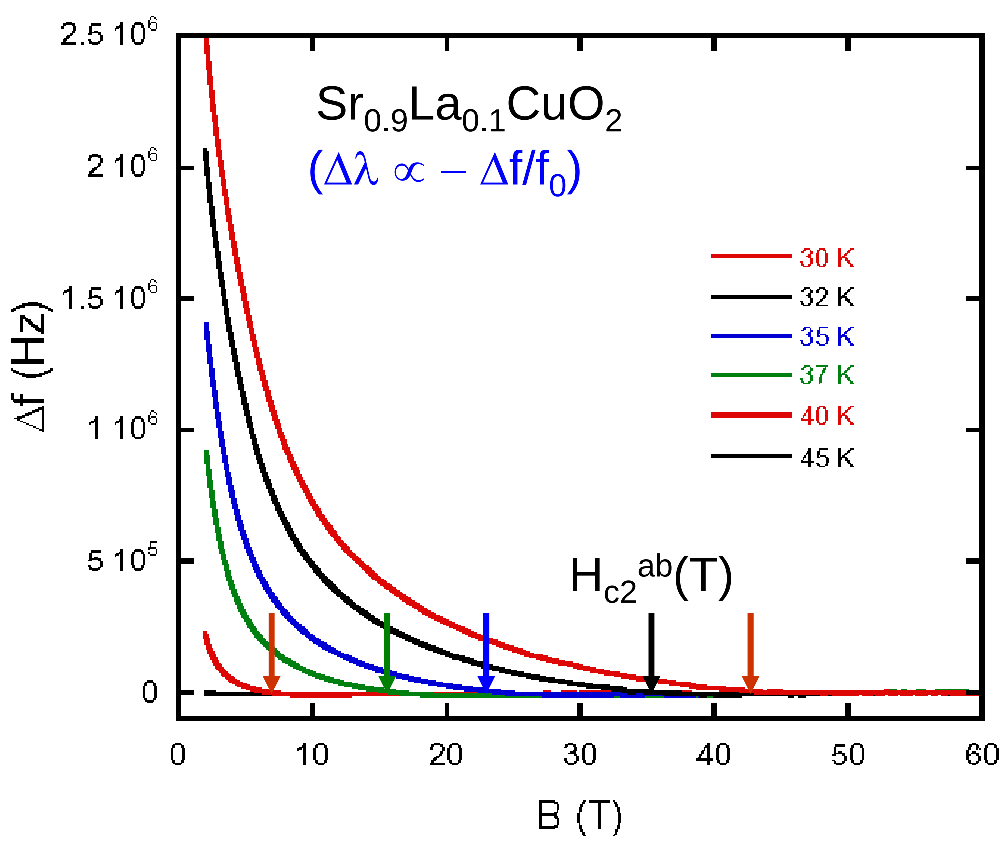
Beyer et al. Phys. Rev. B 76,140506(R) (2007);
Yeh et al., Int. J. Mod. Phys. B 19, 285 (2005).

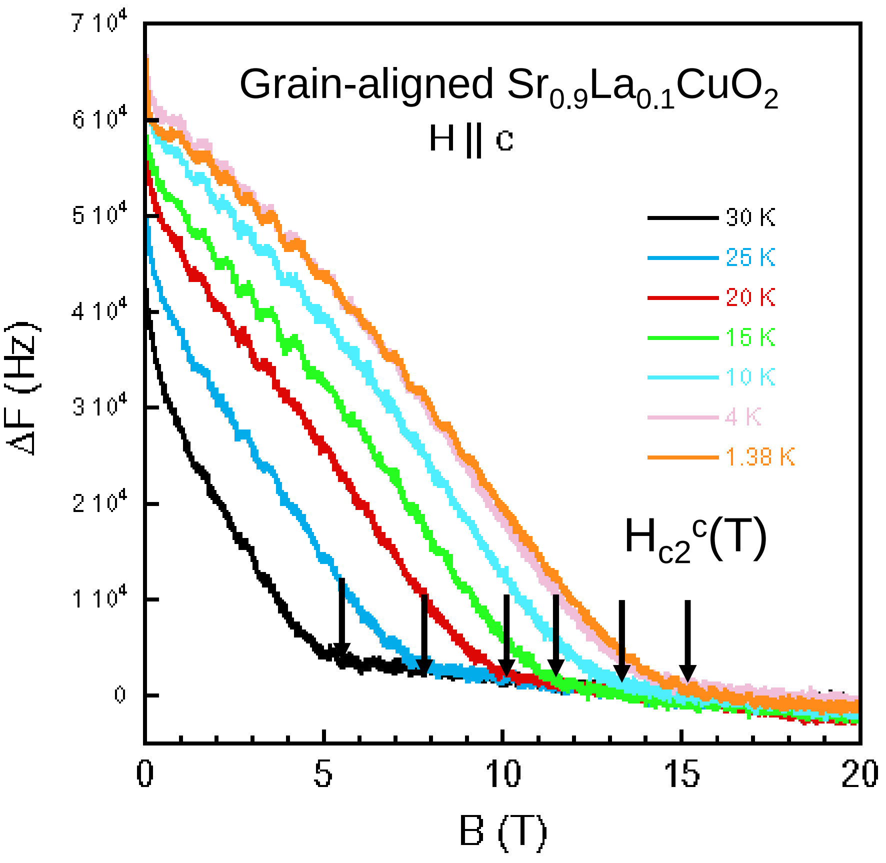
Cuprate Superconductors Investigated

- $HgBa_2Ca_2Cu_3O_x$ (Hg-1223, T_c = 135 K): charge-imbalanced hole-type 3-layer cuprate. [Beyer et al. (2007)]
- $HgBa_2Ca_3Cu_4O_x$ (Hg-1234, T_c = 125 K): charge-imbalanced hole-type 4-layer cuprate. [Beyer et al. (2007)]
- $HgBa_2Ca_4Cu_5O_x$ (Hg-1245, T_c = 110 K): charge-imbalanced hole-type 5-layer cuprate. [Beyer et al. (2007)]
- $La_{0.1}Sr_{0.9}CuO_2$ (La-112, T_c = 43 K): optimally doped electron-type infinite-layer cuprate. [Beyer et al. (2007)]
- $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ (NCCO, T_c = 21 K): optimally doped hole-type 1-layer cuprate. [Yeh et al. (1992)]
- $YBa_2Cu_3O_{7-\delta}$ (Y-123, T_c = 93 & 87 K): optimally and slightly underdoped hole-type 2-layer cuprate. [Yeh et al. (1993); O'Brien et al. (2000)]
- $Bi_2Sr_2CaCu_2O_x$ (Bi-2212, T_c = 60 & 93 K): overdoped hole-typ 2-layer cuprate [Krusin-Elbaum et al. (2004)]

Pulsed-Field Measurements of $H_{c2}(T)$

Determine $H_{c2}(T)$ by measuring the penetration depth (λ) via frequency shifts (Δf) in a tunnel diode oscillator (TDO) resonant tank circuit up to 65 Tesla, with the sample contained in one of the component inductors.



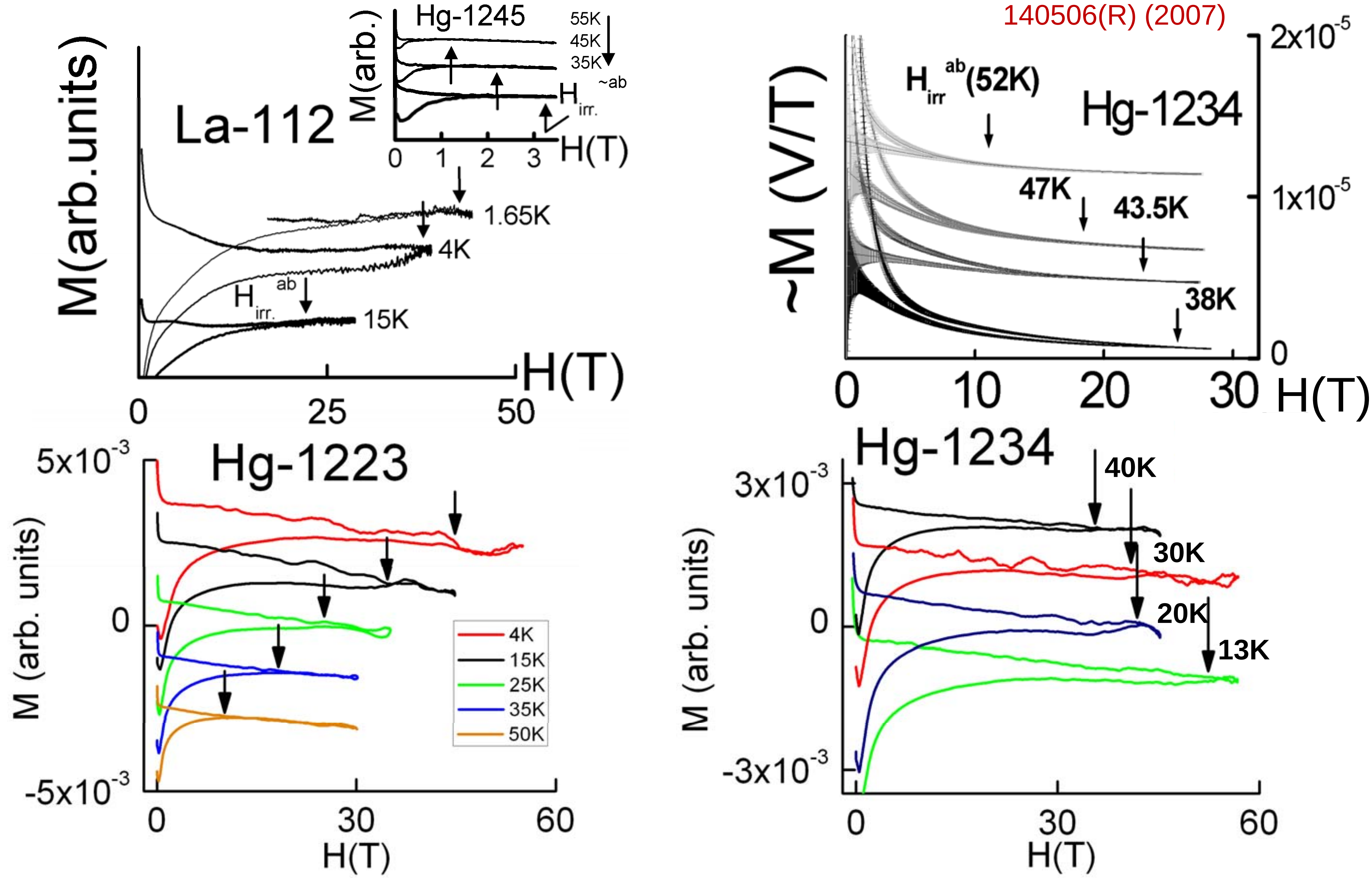


[Zapf et al., Phys. Rev. B 71, 134526 (2005)]

Pulsed & DC-Field Measurements of $H_{irr}(T)$

Examples of M(T,H) measurements for $H_{irr}(T)$ using various techniques, including the cantilever, Hall probe and SQUID magnetometers:

Beyer et al. Phys. Rev. B 76, 140506(R) (2007)

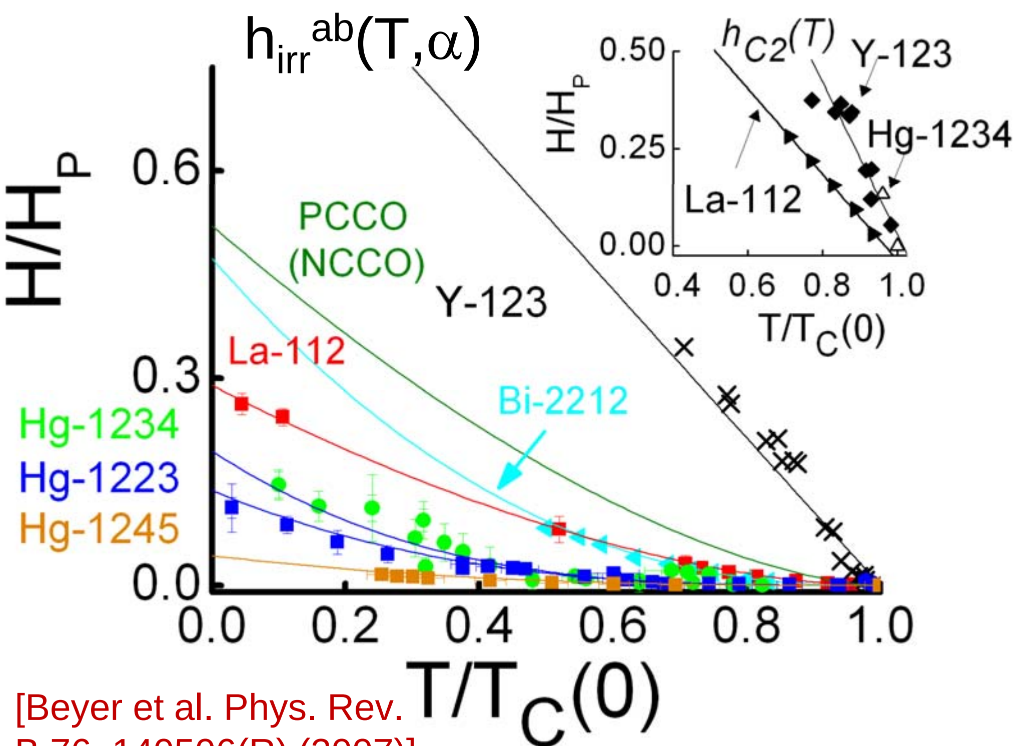


Summary of high-field magnetization measurements

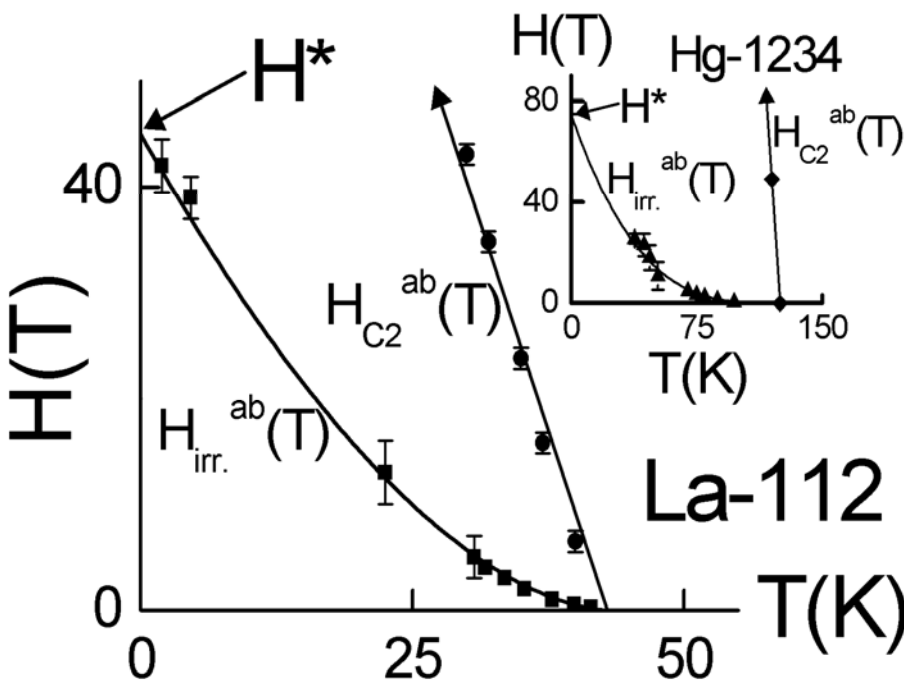
- Field-induced quantum fluctuations exist in all cuprate superconductors.
- Smaller h* implying closer proximity to quantum criticality:

  h*(Hg-1245) << h*(Hg-1234) ~ h*(Hg-1223) < h*(La-112) < h*(PCCO) < h*(Bi-2212) < h*(Y-123)

- $h^*(\alpha) \equiv H^*(\alpha)/H_{c2}^{ab}(0,\alpha) \equiv H_{irr}^{ab}(0,\alpha)/H_{c2}^{ab}(0,\alpha)$



[Beyer et al. Phys. Rev. B 76, 140506(R) (2007)]



[Zapf et al., Phys. Rev. B 71, 134526 (2005)]

Beyer et al. Phys. Rev. B 76,140506(R) (2007); Yeh et al., Int. J. Mod. Phys. B 19, 285 (2005).

Cuprates	T_c (K)	h*	Type	Doping Level
$La_{0.1}Sr_{0.9}CuO_2$ (La-112)	43	0.28	electron-type	optimally doped
$Nd_{1.85}Ce_{0.15}CuO_{4-d}$ (NCCO)	21	0.53	electron-type	optimally doped
$Pr_{1.85}Ce_{0.15}CuO_{4-d}$ (PCCO)	21	0.53	electron-type	optimally doped
$HgBa_2Ca_2Cu_3O_x$ (Hg-1223)	135	0.25	hole-type	charge imbalance
$HgBa_2Ca_3Cu_4O_x$ (Hg-1234)	125	0.20	hole-type	charge imbalance
$HgBa_2Ca_4Cu_5O_x$ (Hg-1245)	110	0.05	hole-type	charge imbalance
$YBa_2Cu_3O_x$ (Y-123)	93	0.85	hole-type	optimally doped
$YBa_2Cu_3O_x$ (Y-123)	87	0.6	hole-type	underdoped
$Bi_2Sr_2CaCu_2O_x$ (Bi-2212)	60	0.45	hole-type	overdoped

Universal correlation of h*(α) with quantum fluctuations

$h^*(\alpha) \equiv [H_{irr}^{ab}(0,\alpha)/H_{c2}^{ab}(0,\alpha)] \propto (\alpha-\alpha_c)^a$; (a ~ 0.5)

$\alpha \equiv \gamma^{-1} (\delta_o/\delta_i)^{-(n-2)} \delta$ [for n > 2]

$\gamma = (\xi_{ab}/\xi_c)$: electronic anisotropy;

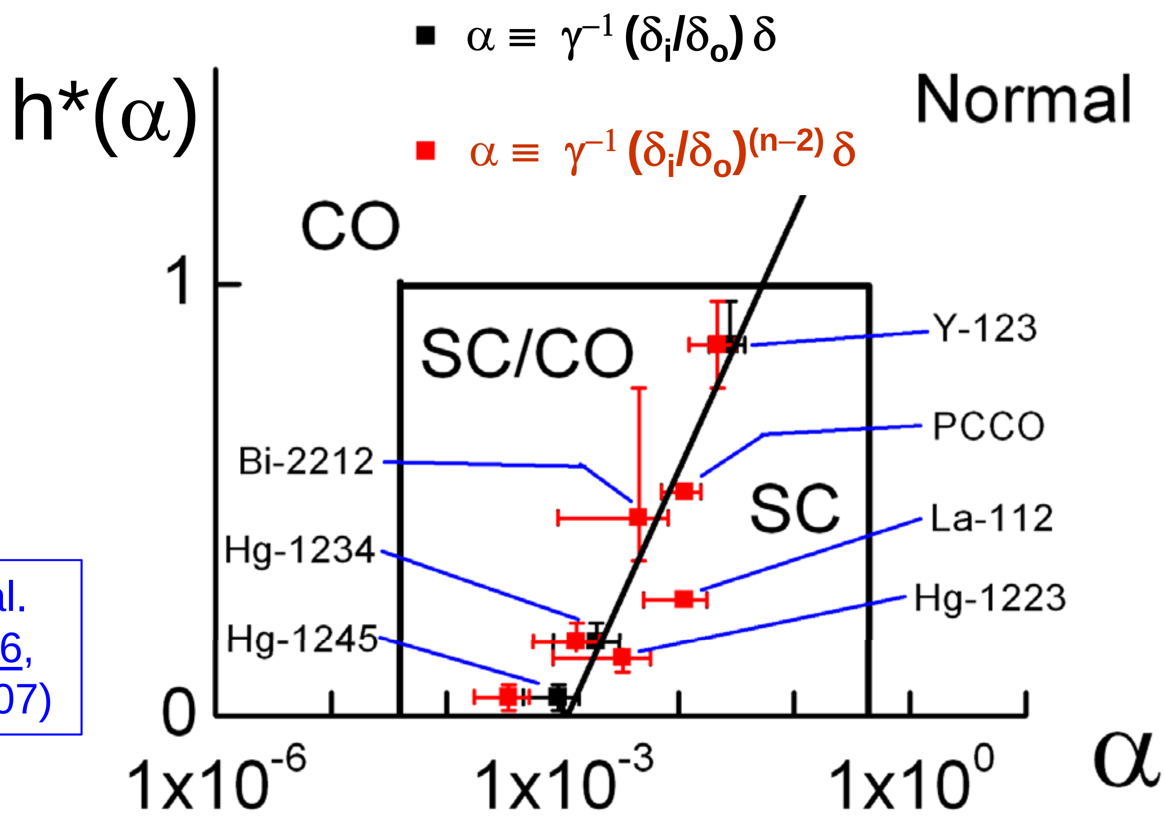
δ : nominal doping level per CuO_2 layer;

δ_o: doping level per outer layer;

n: # of CuO_2 layers per unit cell;

(δ_o/δ_i): charge imbalance ratio;

δ_i: doping level per inner layer.



A.D. Beyer et al. Phys. Rev. B 76, 140506(R) (2007)

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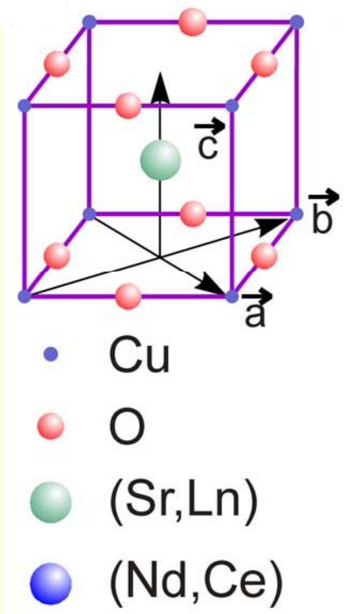
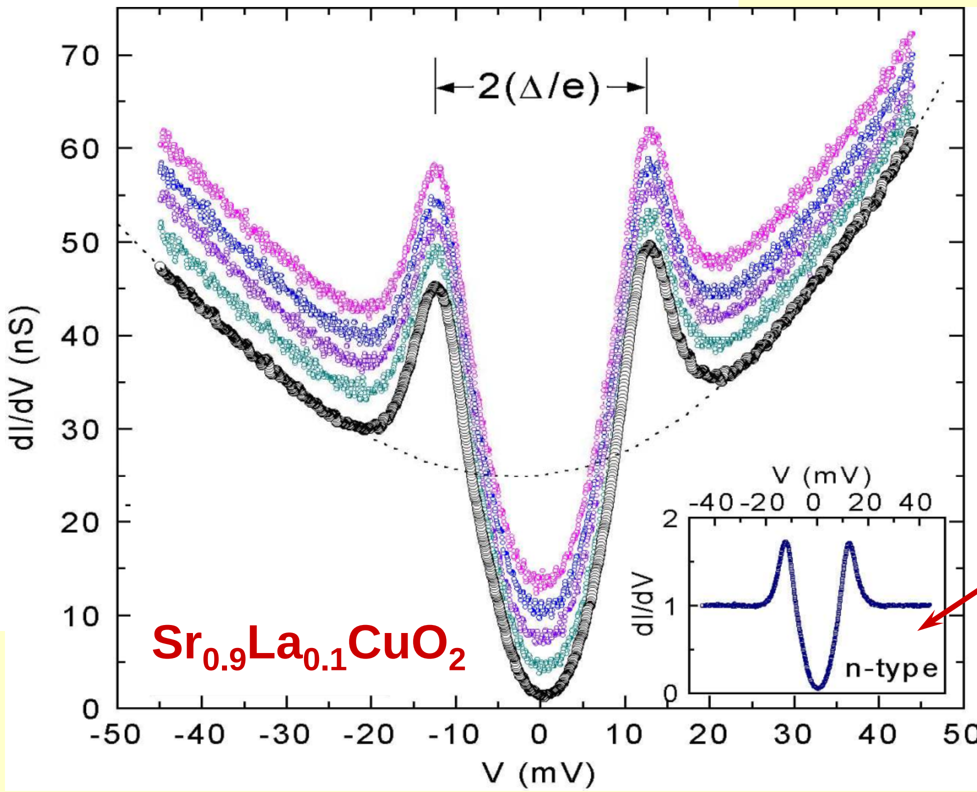
3. Scanning Tunneling Spectroscopic Studies of the Infinite-Layer Electron-Type Cuprates $Sr_{0.9}La_{0.1}CuO_2$





(T_c = 43 K)

normalized spectra

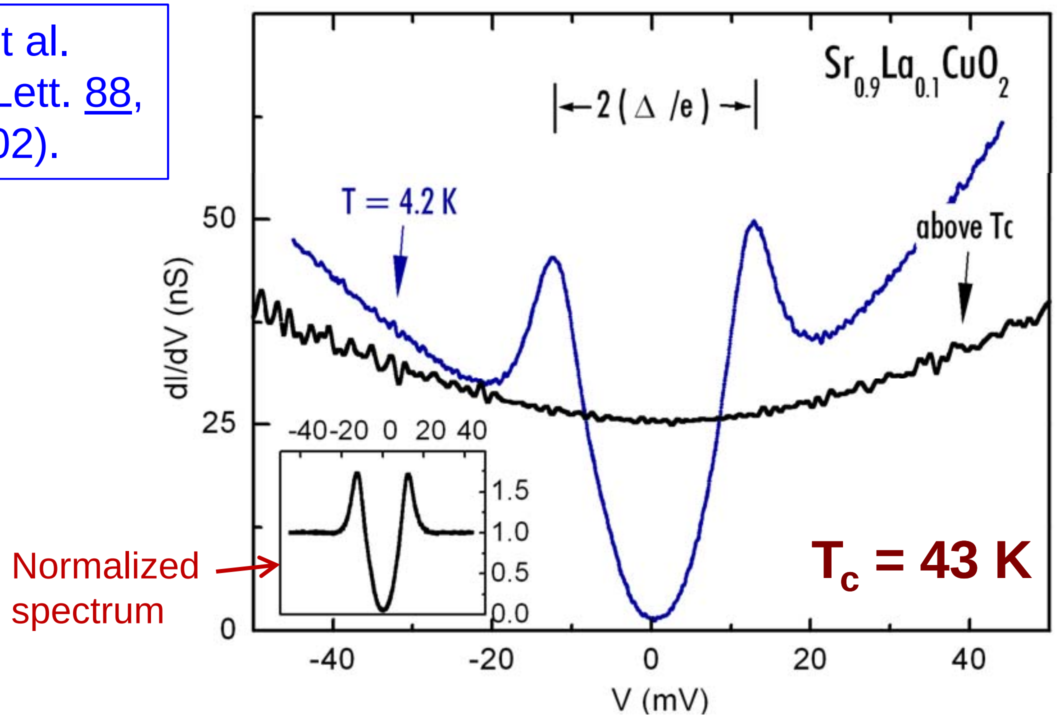
C.T. Chen et al. Phys. Rev. Lett. 88, 227002 (2002).

- The tunneling spectra taken over 200 randomly oriented grains appear to be the same without showing any zero-bias anomaly → s-wave pairing symmetry?

Absence of pseudogap above T_c & strong correlation

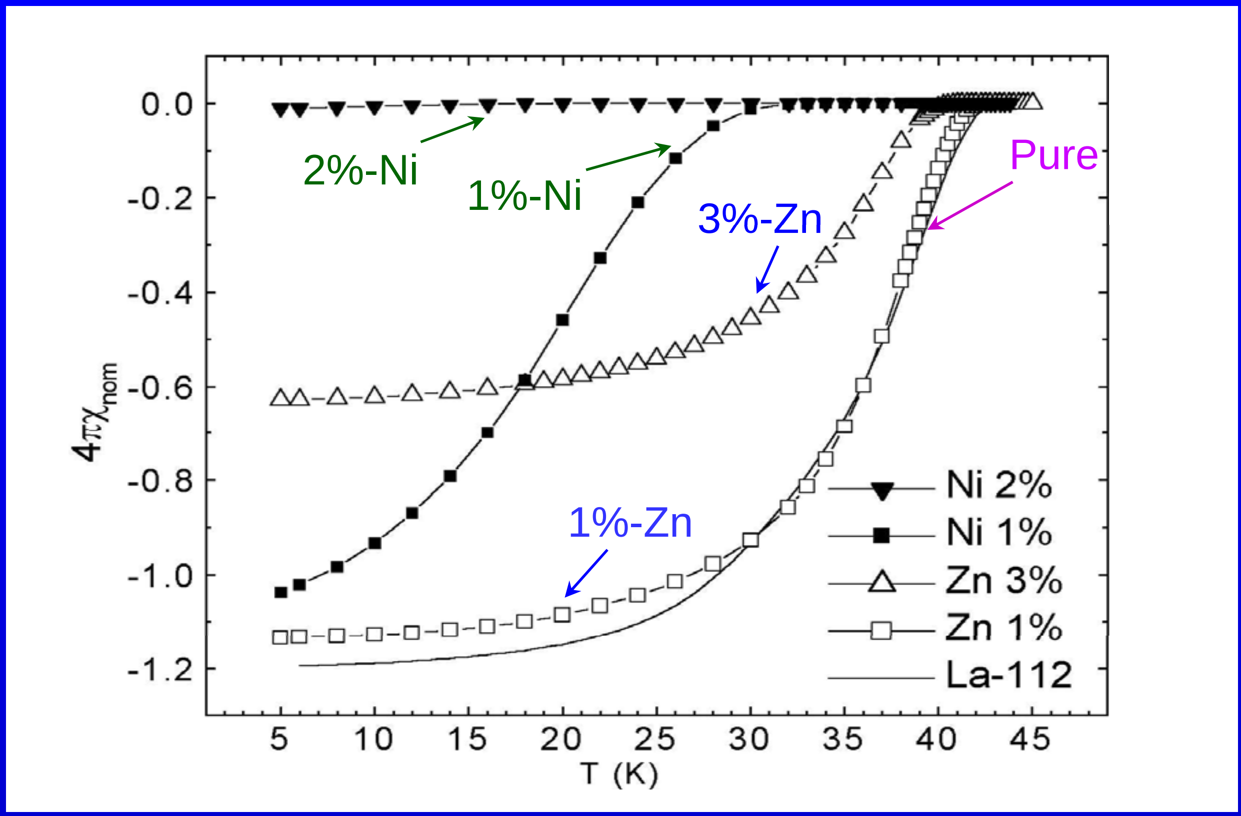
- The tunneling gap completely vanishes at T_c, indicating no pseudogap above T_c.
- Substitution of La by Gd yields the same T_c = 43 K for $Sr_{0.9}Gd_{0.1}CuO_2$.
- $(2\Delta/k_BT_c)$ ~ 7 >> BCS ratio ↔ strong correlation?

C.T. Chen et al.
Phys. Rev. Lett. 88,
227002 (2002).



- $Sr_{0.9}Ln_{0.1}CuO_2$ (Ln = La, Gd) are three-dimensional superconductors and are the only cuprate system with c-axis superconducting coherence length ξ_c (= 0.53 nm) > c-axis lattice constant c_0 (= 0.347 nm).

Global effect of quantum impurities on $Sr_{0.9}La_{0.1}CuO_2$

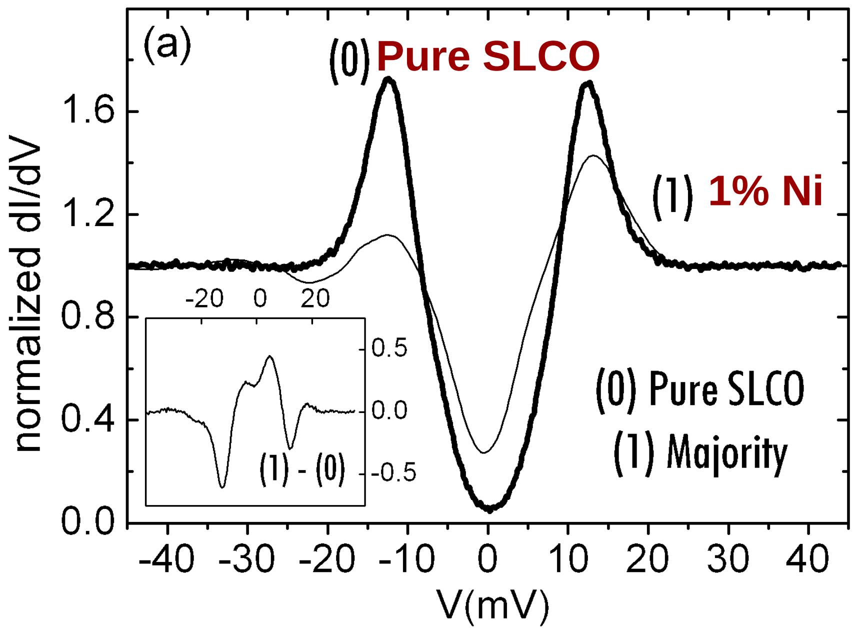


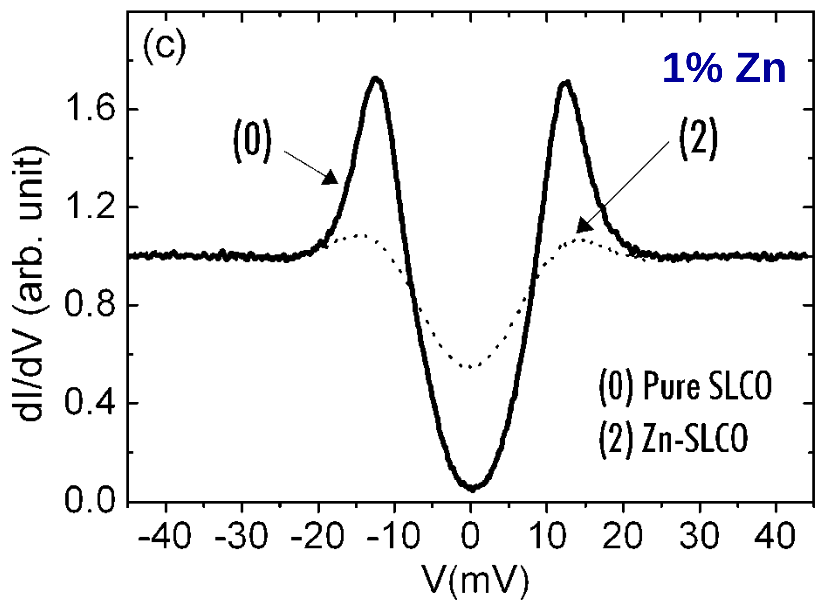
C.U. Jung et al. Phys. Rev. B65, 172501 (2002).

- Spinless Zn^{2+} impurities: weak effects on T_c.
- Magnetic Ni^{2+} impurities: strong suppression of T_c.

⟹ Consistent with conventional superconductors.

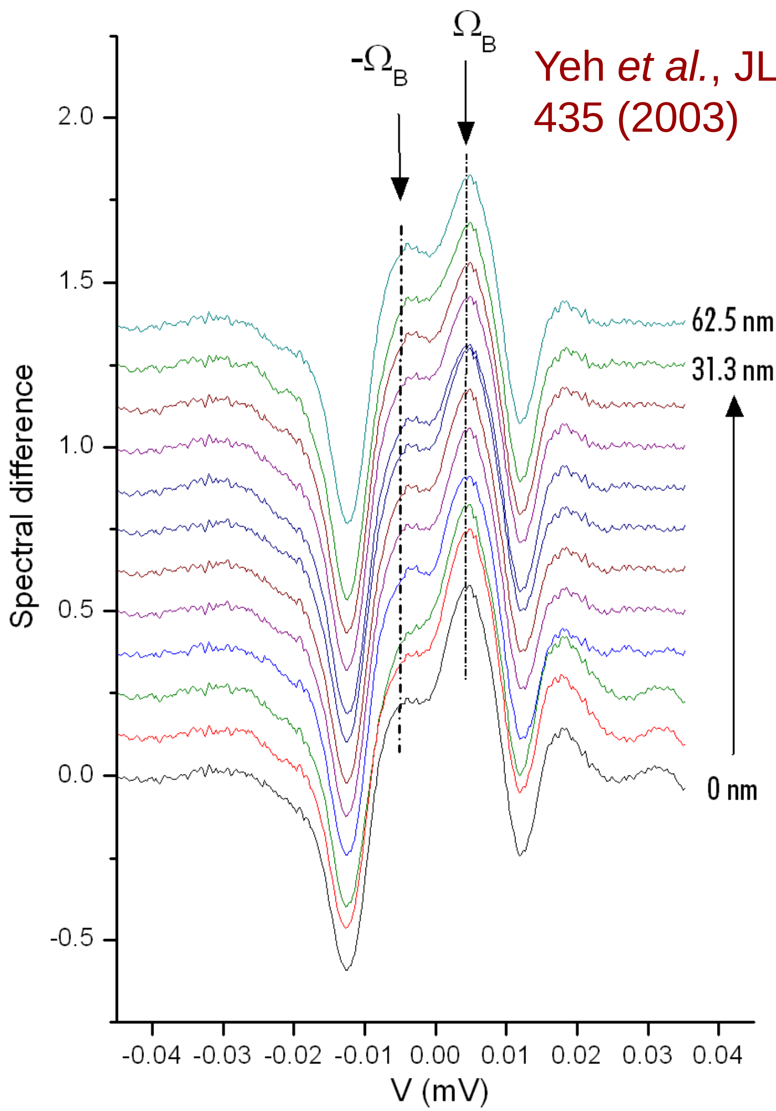
Local effects of quantum impurities on n-type $Sr_{0.9}La_{0.1}CuO_2$



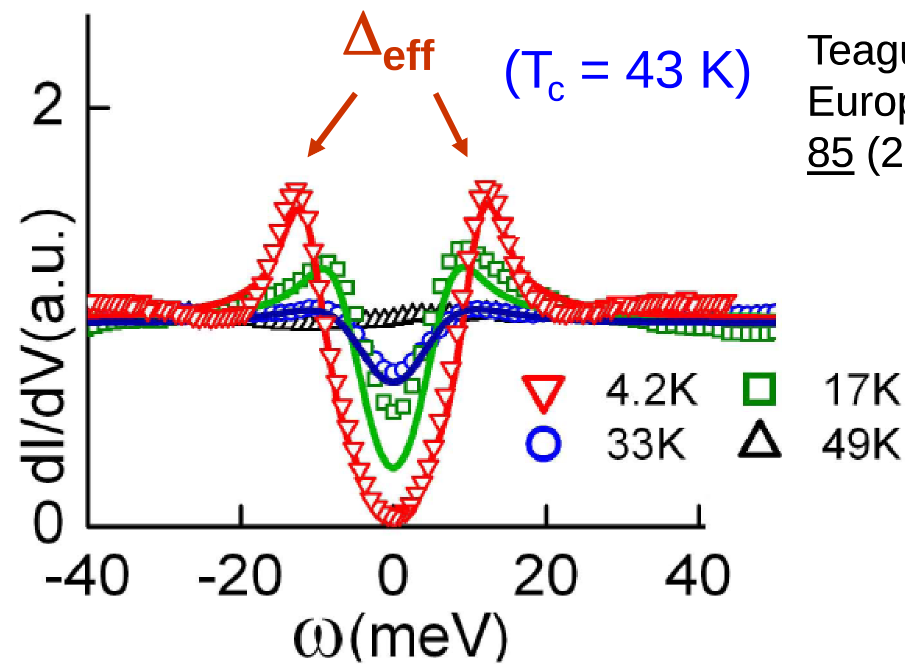


Long-range Ni-induced Shiba states
→ consistent with s-wave pairing.

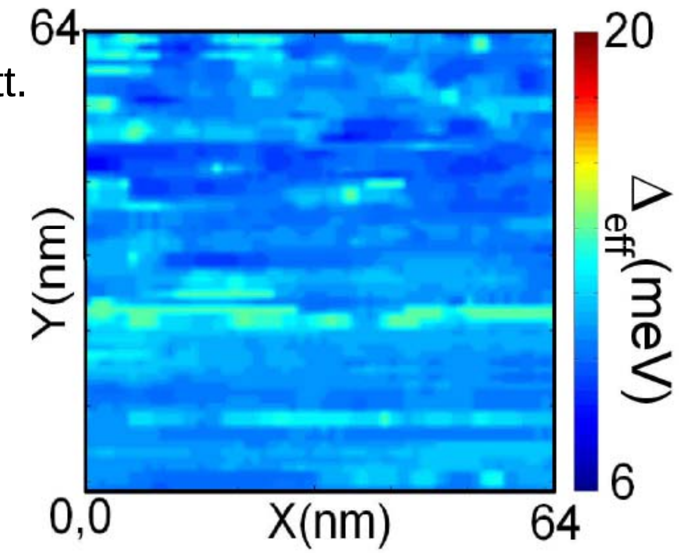
Yeh *et al.*, JLTP 131, 435 (2003)

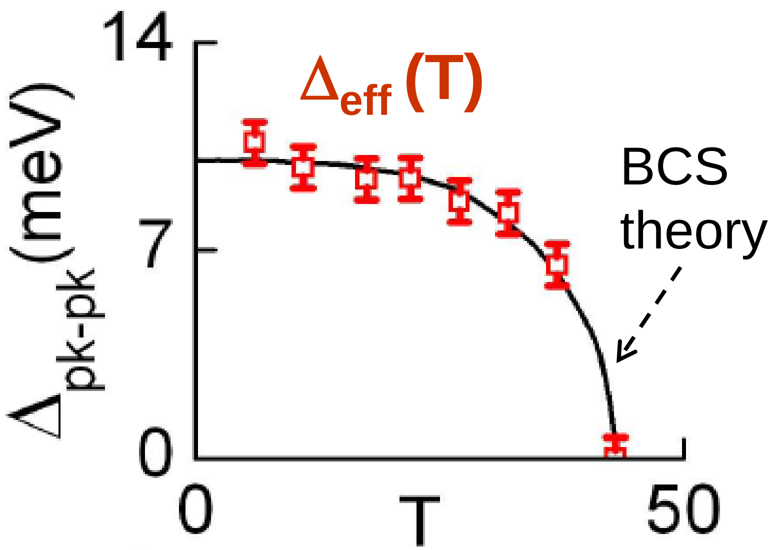


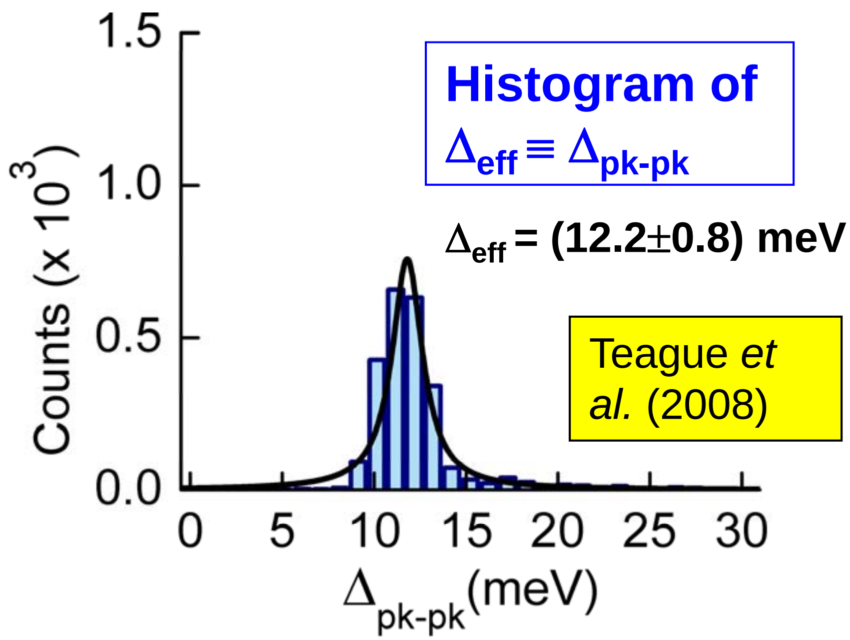
Zero-Field Quasiparticle Tunneling Spectra in La-112:



Teague et al, Europhys. Lett. 85 (2009)

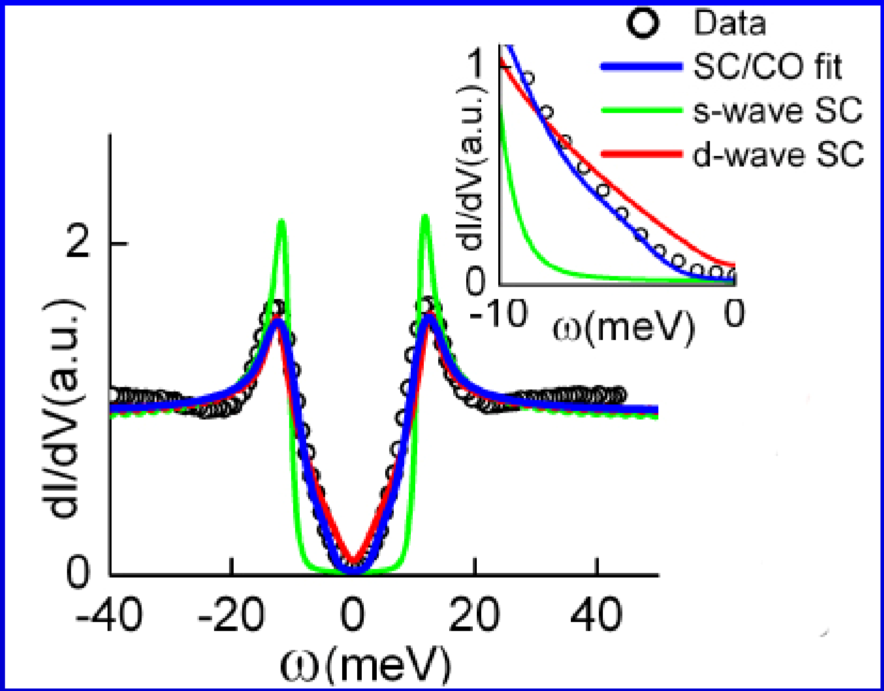






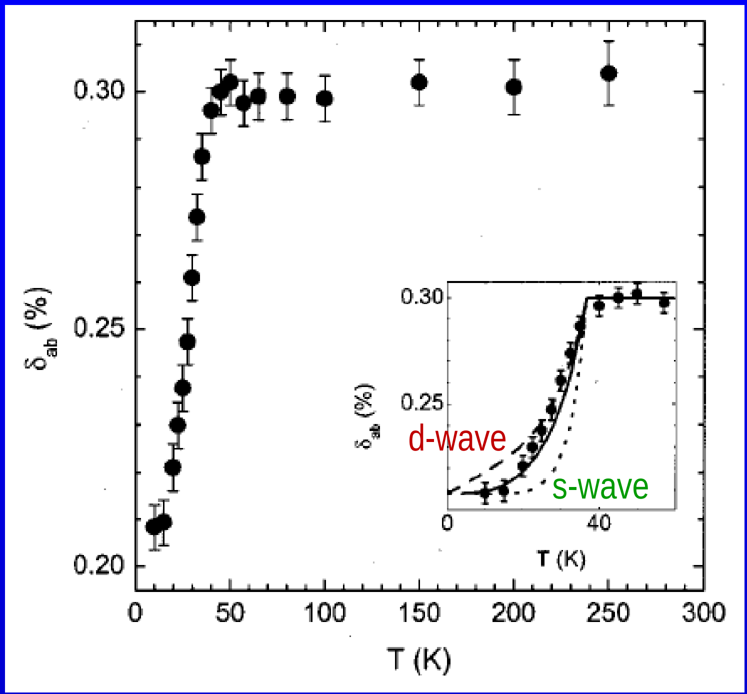
Anomalous tunneling spectra & Knight shifts in La-112 due to coexisting superconductivity & competing orders:

Model fitting to the STS spectra



M.L. Teague et al, Europhys. Lett. 85, 17004 (2009); C.-T. Chen et al, Solid State Commun. 143, 447 (2007).

T-dependent Cu-NMR shifts



G.V.M. Williams et al, Phys. Rev. B 65, 224520 (2002)

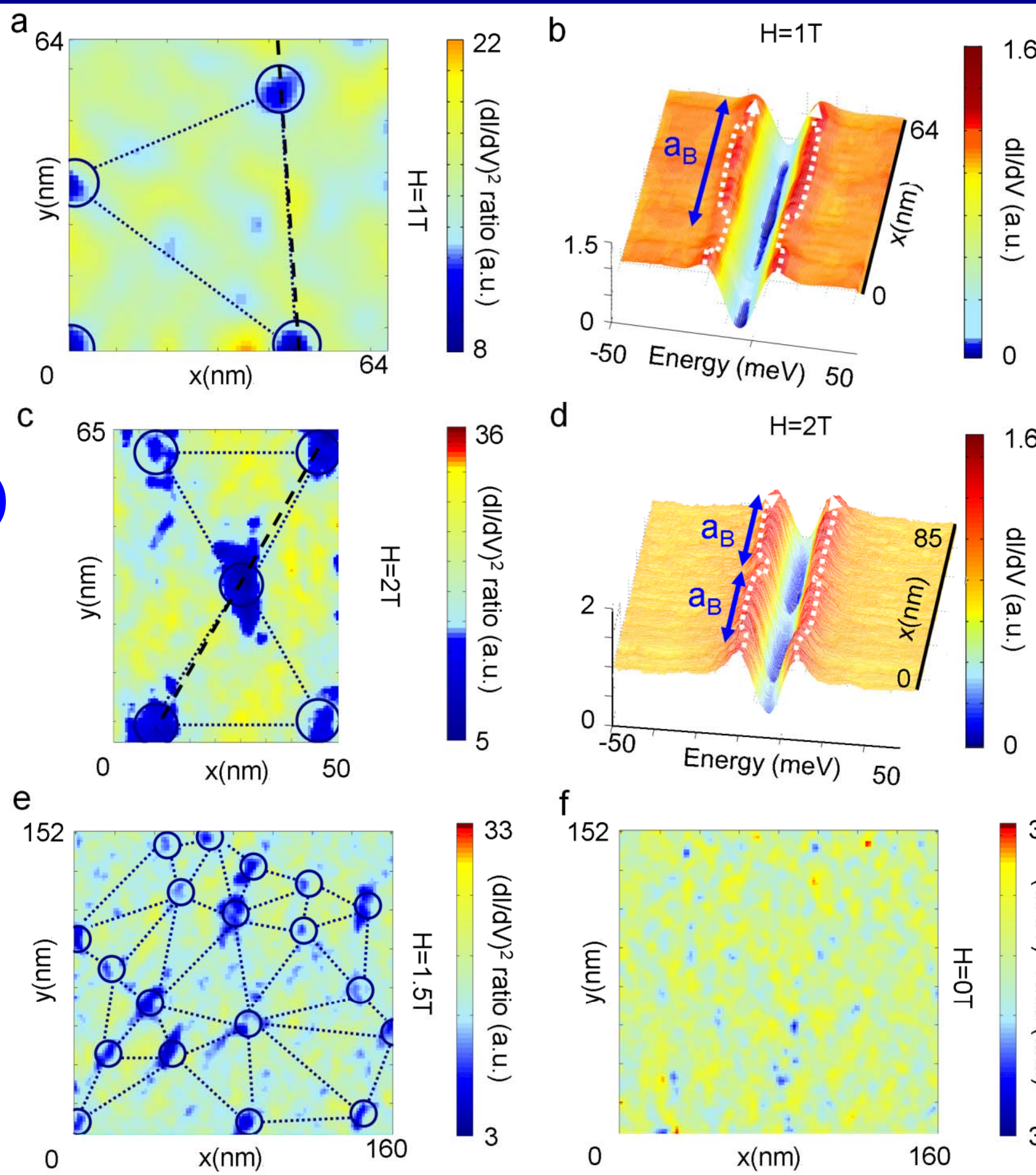
- Experimental results may be understood in terms of a nearly isotropic competing order energy gap V_{CO}~ 8 meV and a d-wave superconducting gap Δ_{SC} ~ 12 meV; further confirmed by STM studies of the vortex-state quasiparticle spectra.

Vortex-State Quasiparticle Tunneling Spectra in La-112

(H = 1.0 T)

(H = 2.0 T)

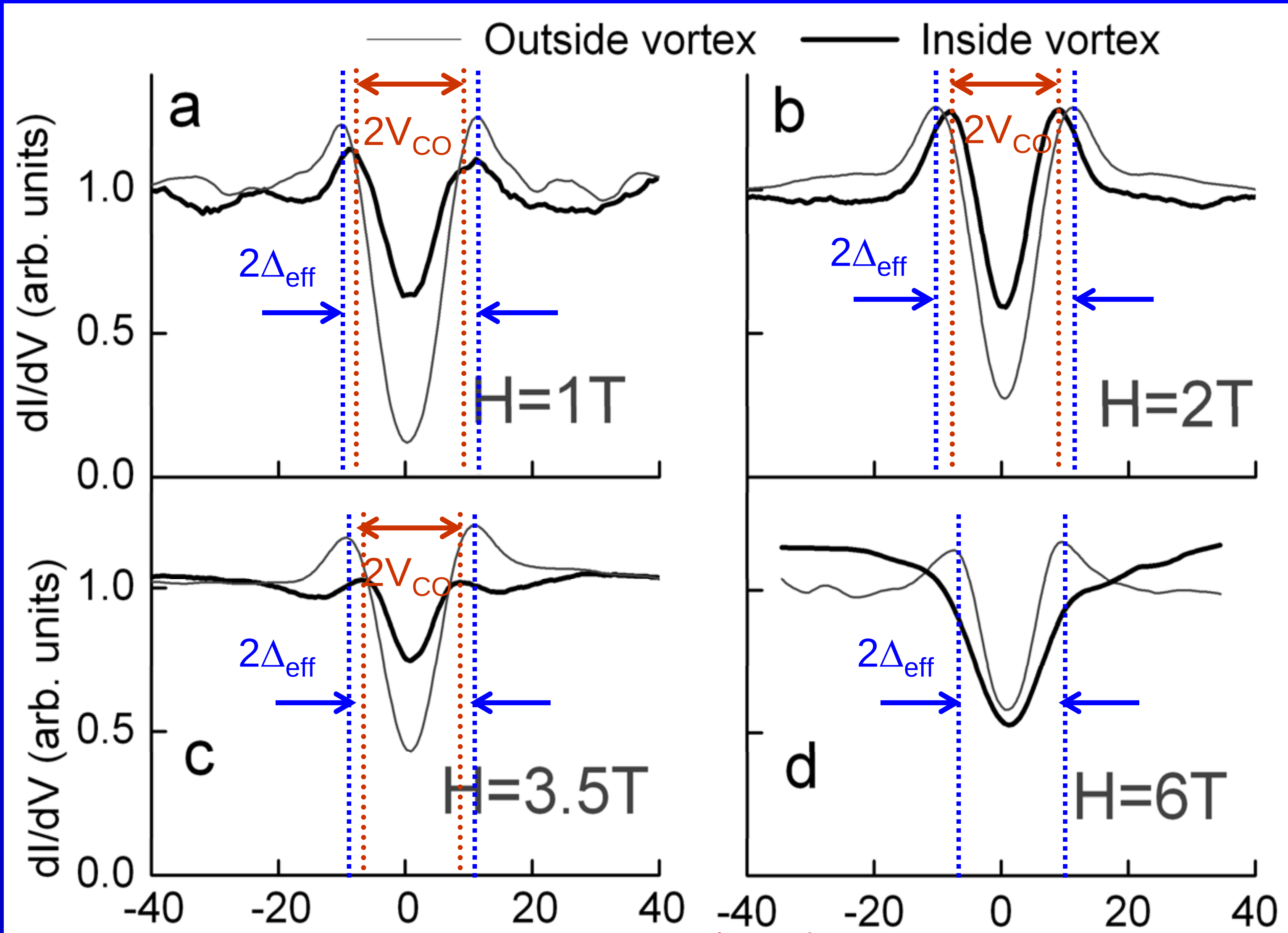
(H = 1.5 T)



(H = 0)

Teague *et al.*, Europhys. Lett. 85, 17004(2009)

Intra- and Inter-Vortex Spectral Comparison & Field-evolution (Infinite-layer electron-type La-112)

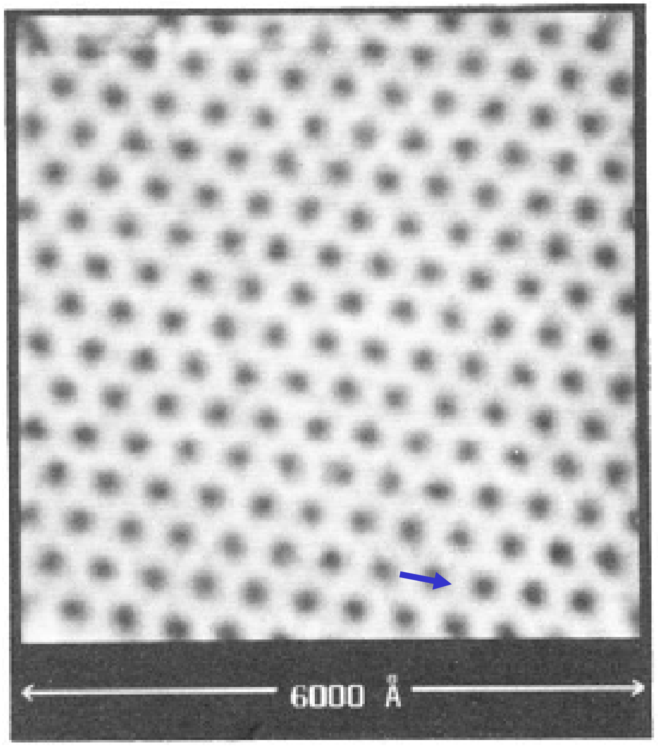


Teague *et al*, Europhys. Lett. 85, 17004 (2009)

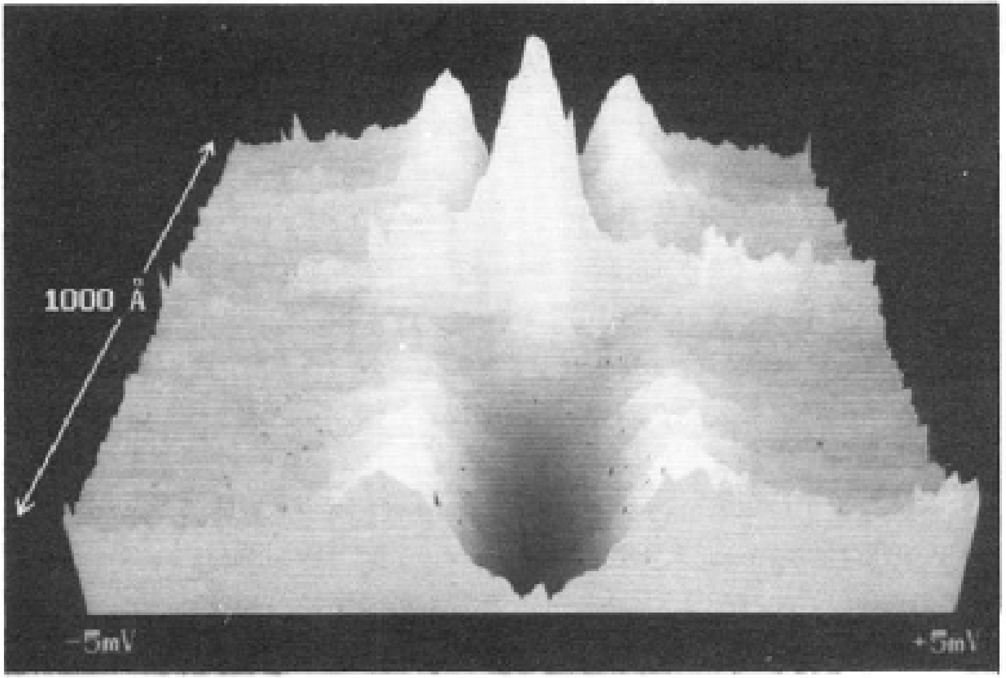
Vortex-state quasiparticle spectra in conventional superconductors (w/o ground-state competing orders)

Hess et al., Phys. Rev. Lett. 62 (1989).

Bound states inside the vortex core with energies $E_\mu = \mu(\Delta^2/E_F)$, μ = 1/2, 3/2, …, E_F = Fermi energy.



Conductance image of the Abrikosov vortex lattice produced by 1 Tesla magnetic field in $NbSe_2$ at 1.8 K



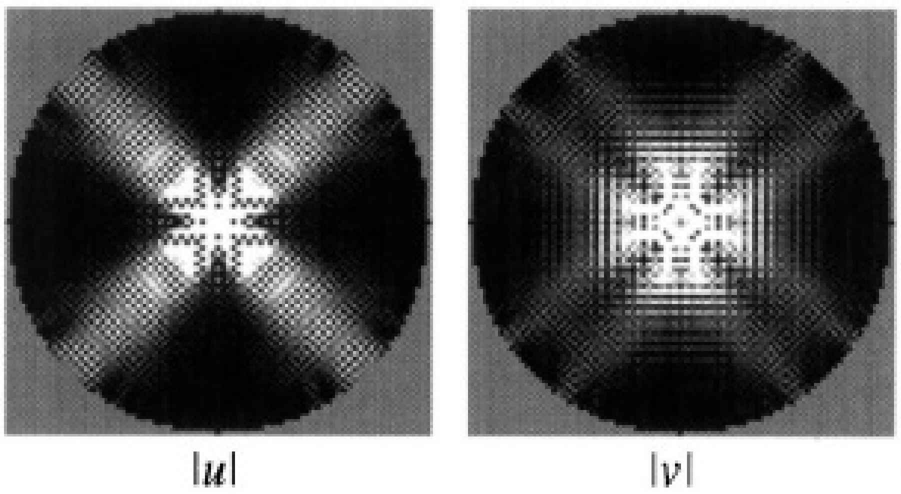
Perspective image of dI/dV-vs.-V as a function of position along a line that intersects a vortex, as represented by the blue arrow on the left figure.

Predictions for the vortex-state quasiparticle spectra in cuprate superconductors (w/o ground-state competing orders)

Premise: Pure $d_{x^2-y^2}$ SC order parameter

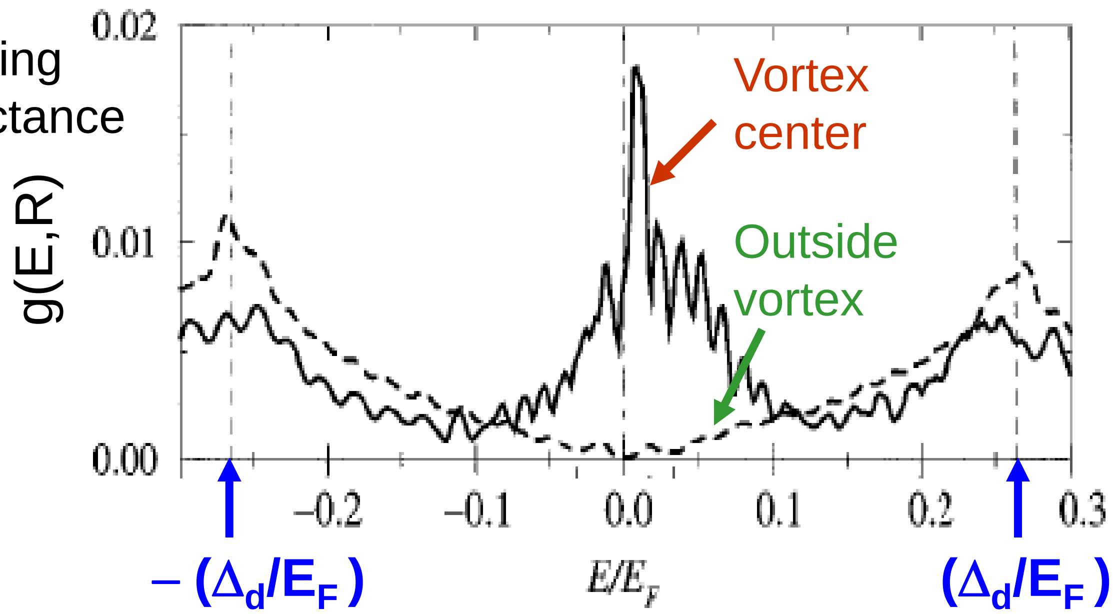
|*u*|: electron-like quasiparticle amplitude;

|*v*|: hole-like quasiparticle amplitude;



Franz & Tesanovic PRL **80**, (1998).

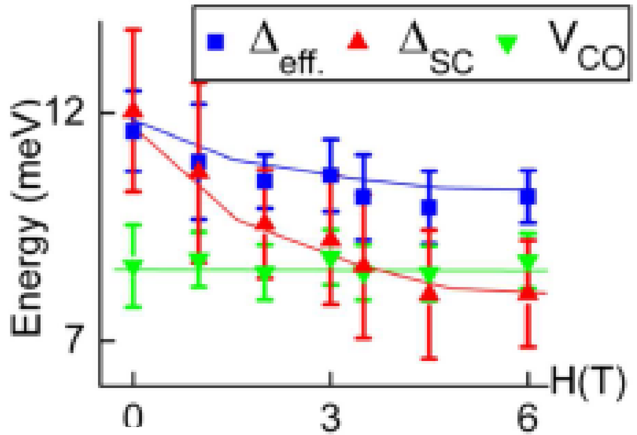
Tunneling conductance



Unconventional Spectral Evolution with Magnetic Field

La-112

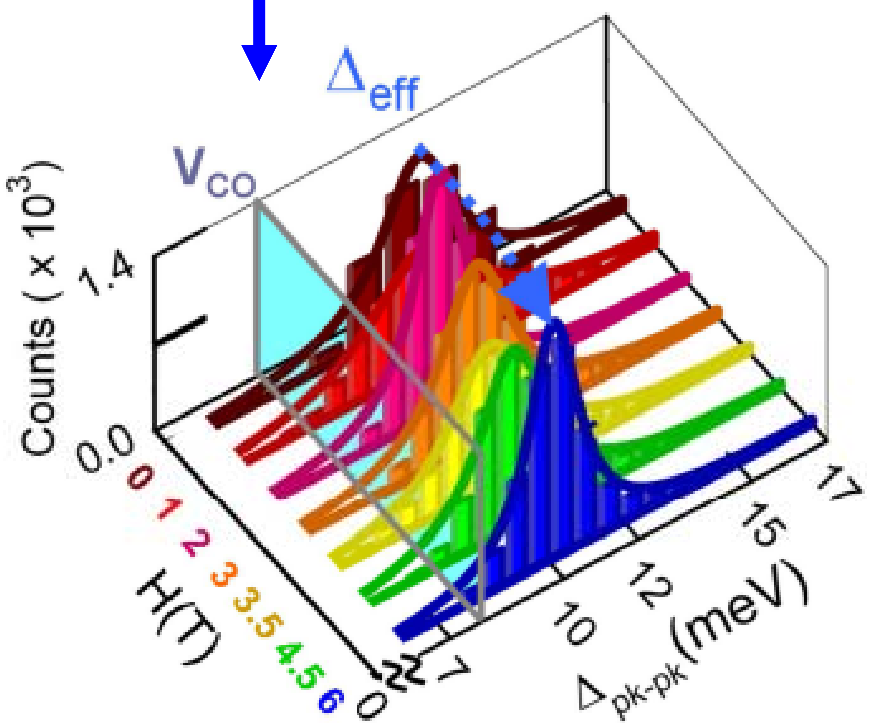
- At H = 0, $\Delta_{SC} > V_{co}$.

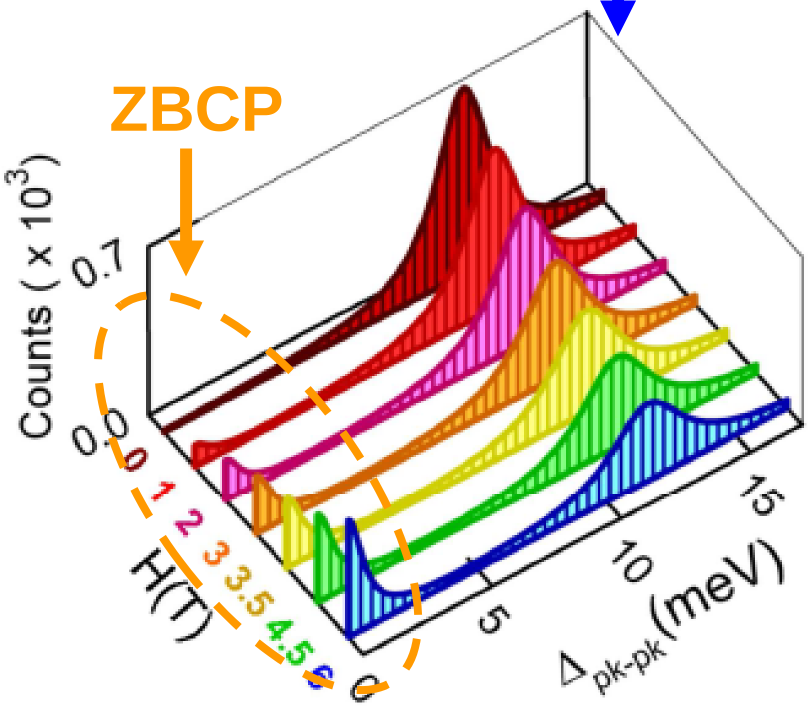


M.L. Teague et al, Europhys. Lett. 85, 17004 (2009)

Energy histograms of electron-type La-112 w/ cutoff @ V_{co}

Energy histograms of conventional type-II SC w/ ZBCP



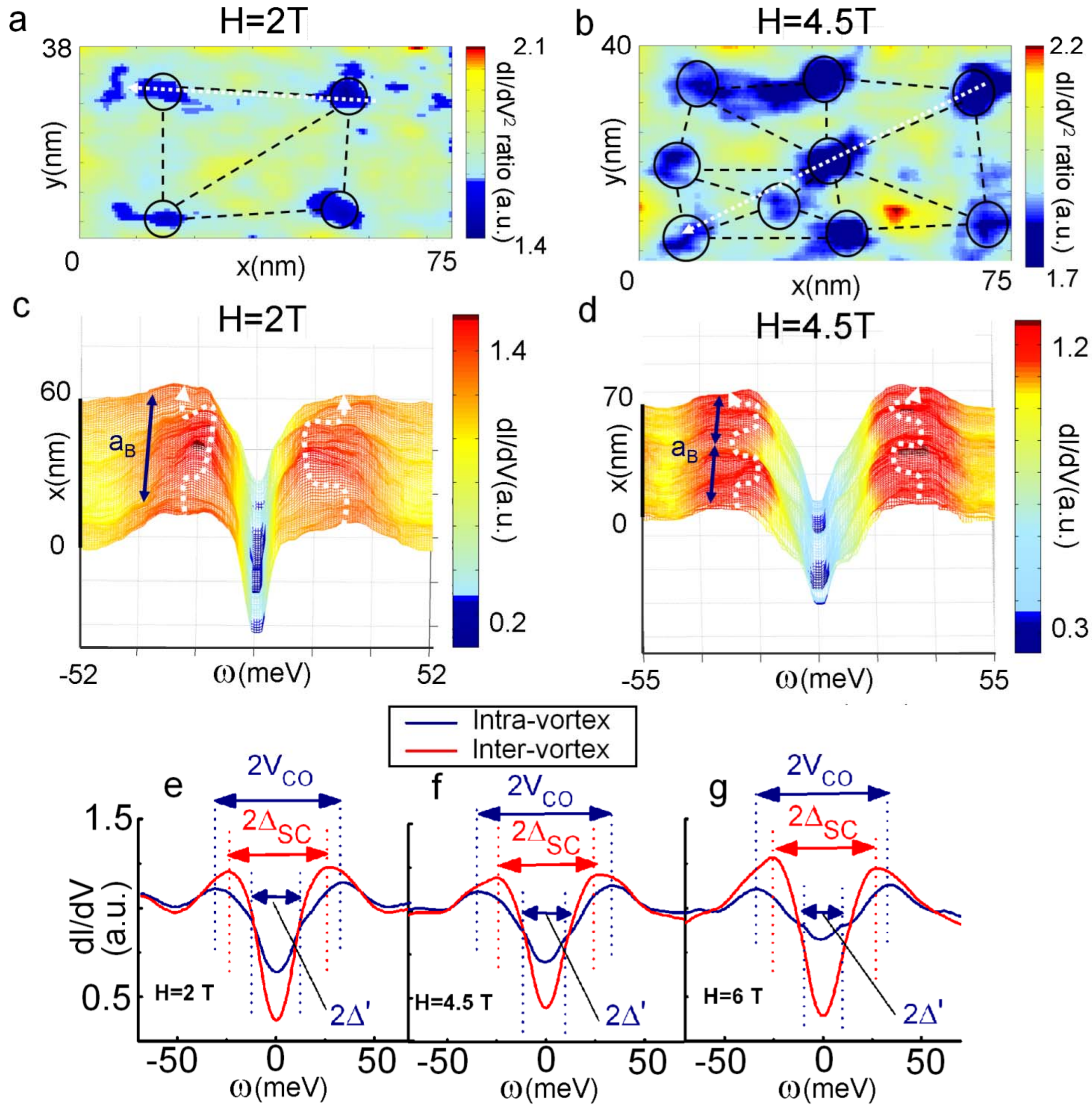


Spatially resolved quasiparticle spectra in Y-123: H > 0

Disordered “vortex halos” with radius $\xi_{halo} \sim 10\ \xi_{SC}$.

A. D. Beyer *et al.*, Europhys. Lett. 87, 37005 (2009)

Gapped features everywhere in the vortex-state spectra.

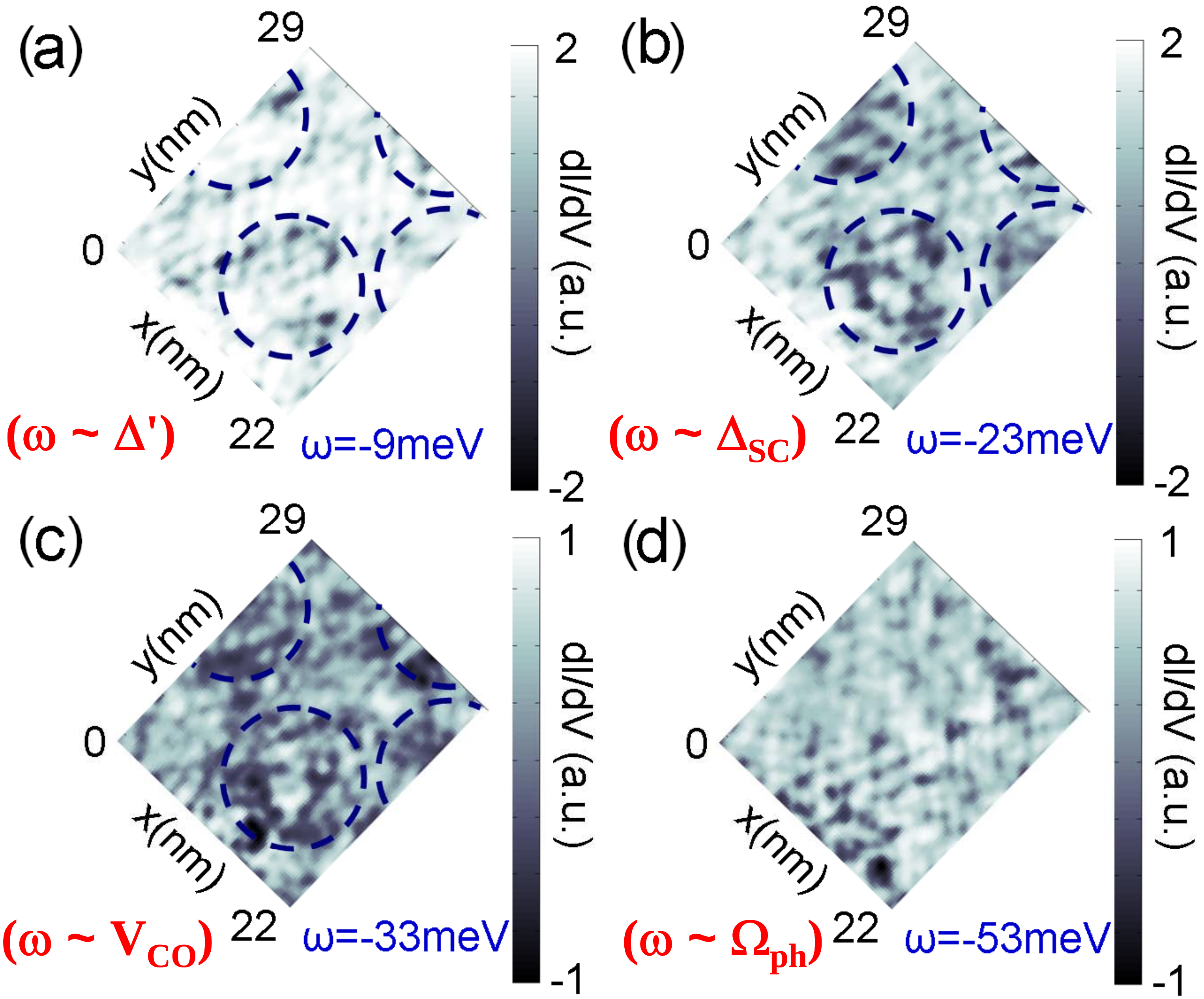


Appearance of two new energy scales inside vortices:

V_{CO} & Δ'

Vortex-state local density of states (LDOS) modulations

H = 5 T



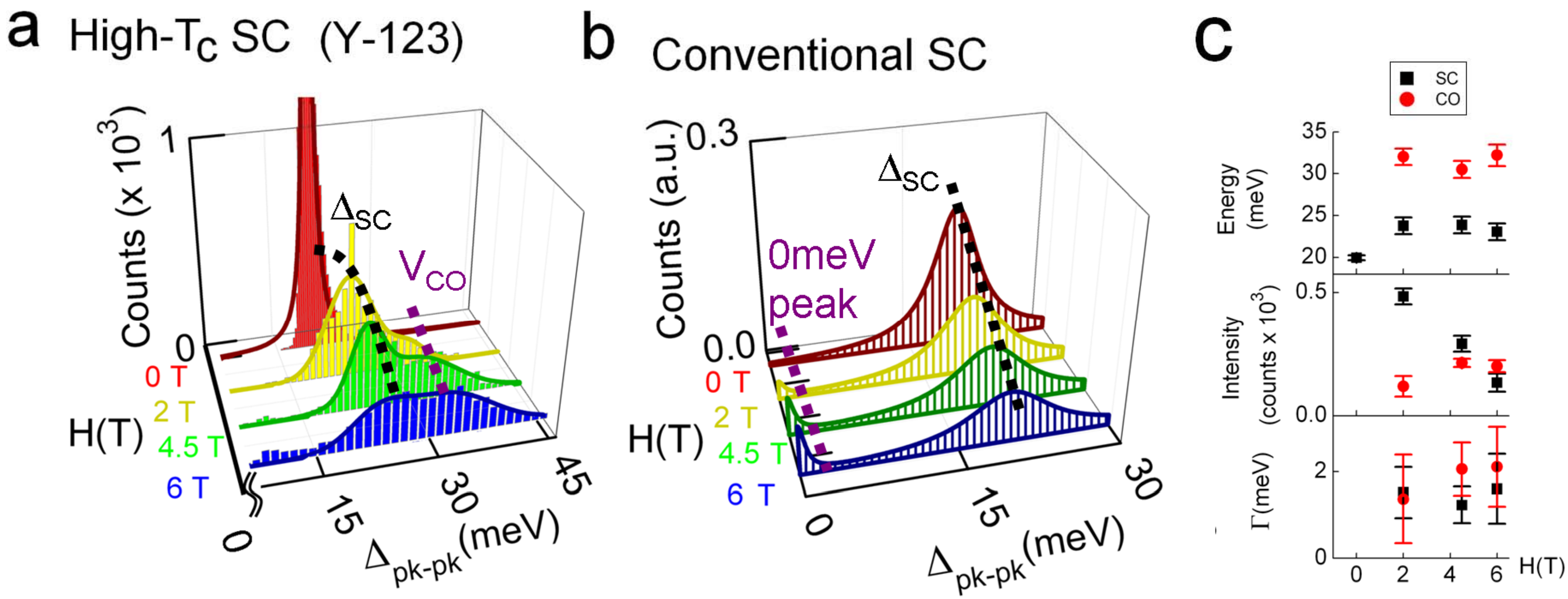
Ω_{ph}: optical Cu-O phonon frequency

A. D. Beyer *et al.*, Europhys. Lett. 87, 37005 (2009)

Energy histograms & spectral evolution with field in Y-123

Energy histogram of Y-123

Energy histogram of conventional type-II SC

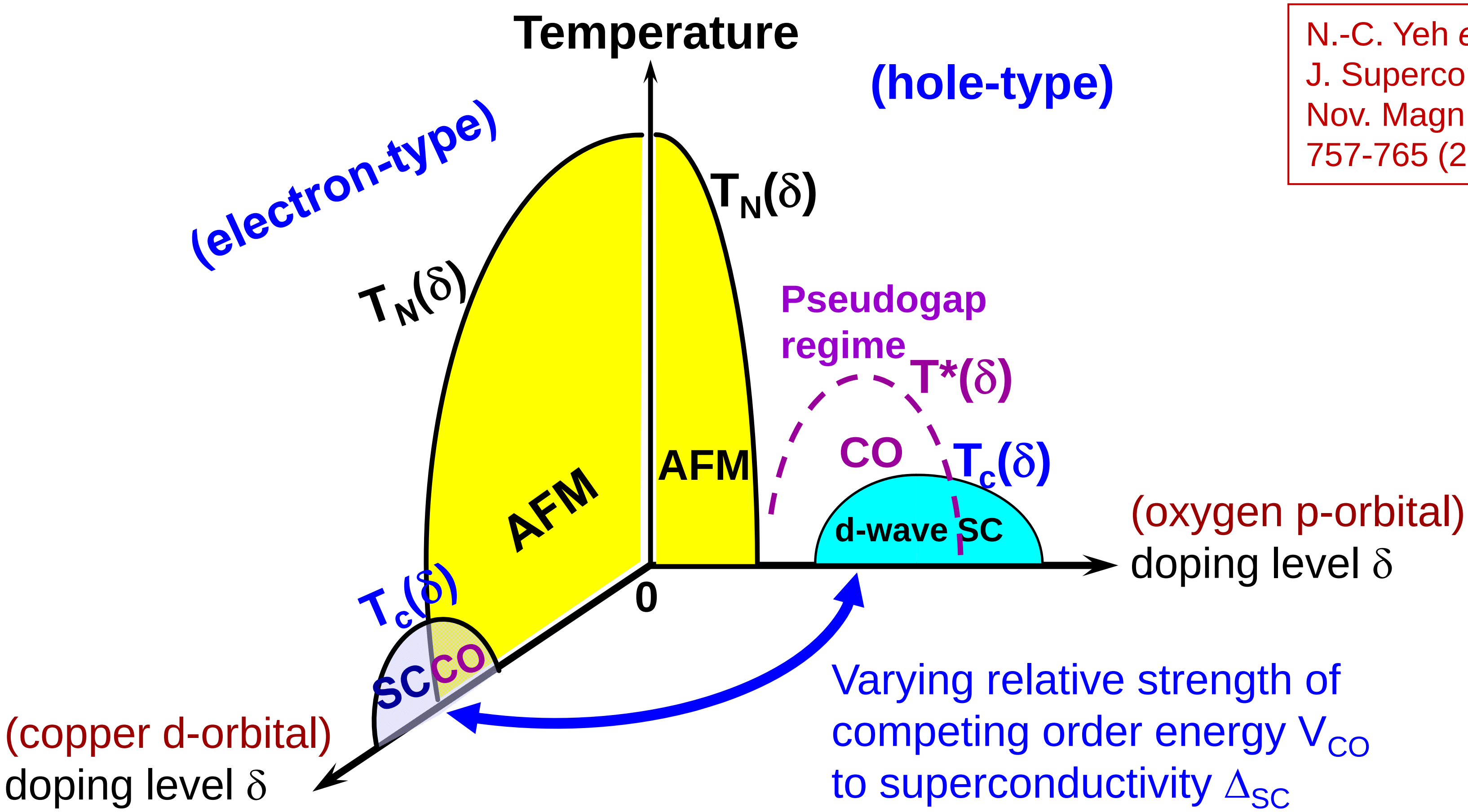


A.D. Beyer *et al.*, Europhys. Lett. 87, 37005 (2009)

Possible physical origin for asymmetric phase diagrams of cuprate superconductors between hole- and electron-doping :

* The coexisting competing order has an energy scale V_{CO} larger than the superconducting gap Δ_{SC} in the under- and optimally doped hole-type cuprate superconductors, whereas $V_{CO} < \Delta_{SC}$ for all electron-type cuprate superconductors, hence no pseudogap above T_C in the latter.

N.-C. Yeh *et al.*, J. Supercond. Nov. Magn. 23, 757-765 (2010)



1. Overview
2. Multi-layer hole-type cuprate superconductors
3. Infinite-layer electron-type cuprate superconductors
4. Summary

4. Summary

Professor Sung-Ik Lee had made many important contributions to the field of cuprate superconductivity, particularly in the following areas:

- **Materials synthesis**

-- Developed novel methods for synthesizing high-quality & unique materials.

- **Multi-layer hole-type cuprate superconductors**

-- Determined thermodynamic properties and vortex dynamics.
-- Established that the anisotropy, charge imbalance, competing order, and thermal & quantum fluctuations increase with increasing number of CuO_2 planes per unit cell.
-- Importance of next-nearest neighbor inter-layer coupling to the occurrence of cuprate superconductivity

- **Infinite-layer electron-type cuprate superconductors**

-- Three-dimensional superconductors (ξ_c > c-axis lattice constant c_0)
-- Node-less energy gap due to coexisting competing order & superconductivity
-- First (and the only) observation of vortex images & intra-vortex pseudogap in electron-type cuprates to date.

To my friend Professor 李星翊

哲人日已遠，

典型在夙昔．

風簷展書讀，

古道照顏色．

-- 文天祥“正氣歌”

Although those great philosophers and scholars in history have long left us,

their fine examples remain in our minds ‘till this day.

As I unfolded books and began reading in the gentle breeze under the eaves,

I seemed to have seen the images of the great characters shine through the passage of time.

Although Sung-Ik is no longer with us physically, his legacy lives on…

In his fine publications;

In the minds of his loved ones and friends;

and

In the life of all those young people that he had kindly touched upon.