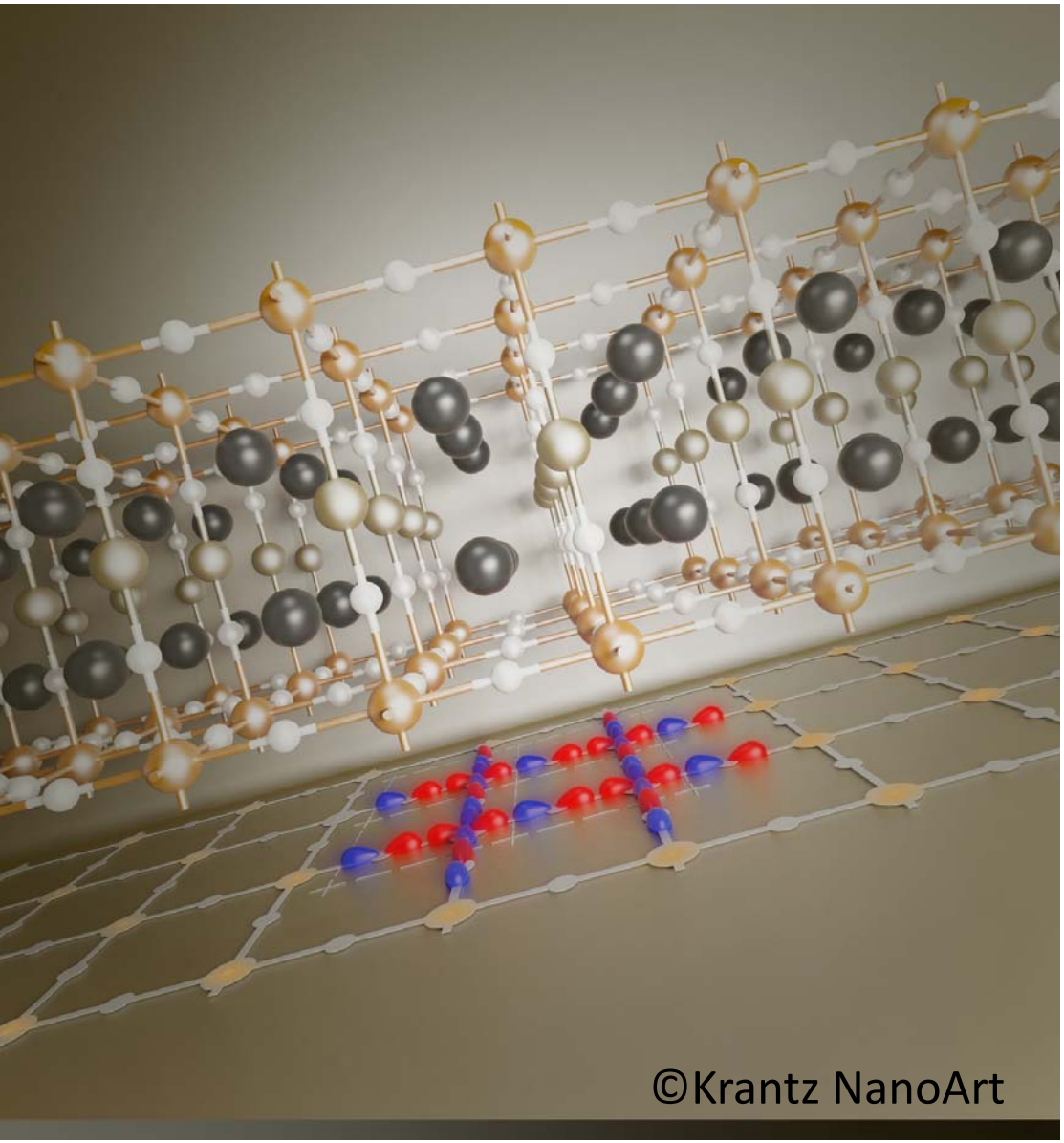
©Krantz NanoArt

Mechanism of Superconductivity in Cuprates: Optimizing T_c

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CIFAR meeting, Toronto, May 3, 2022

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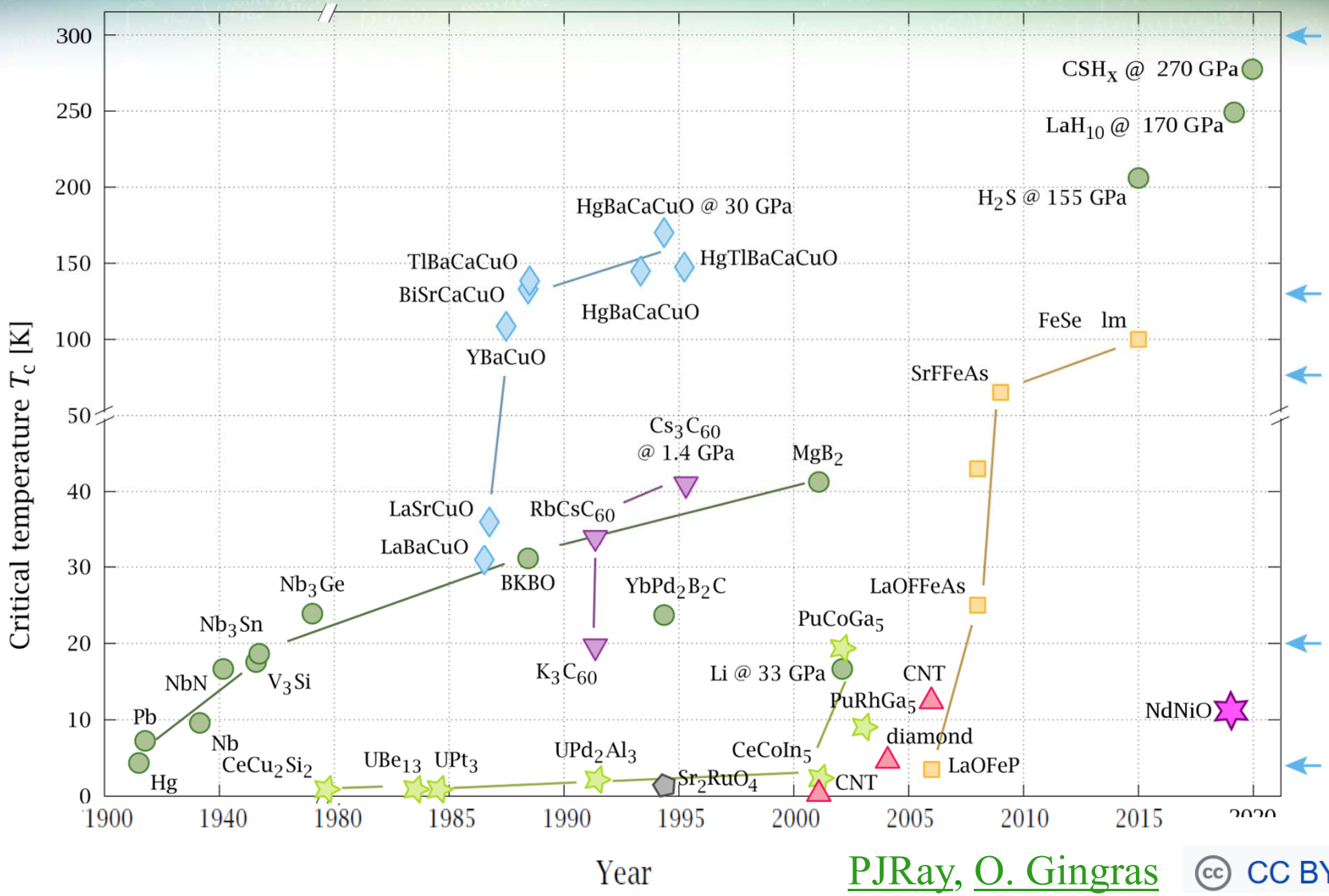
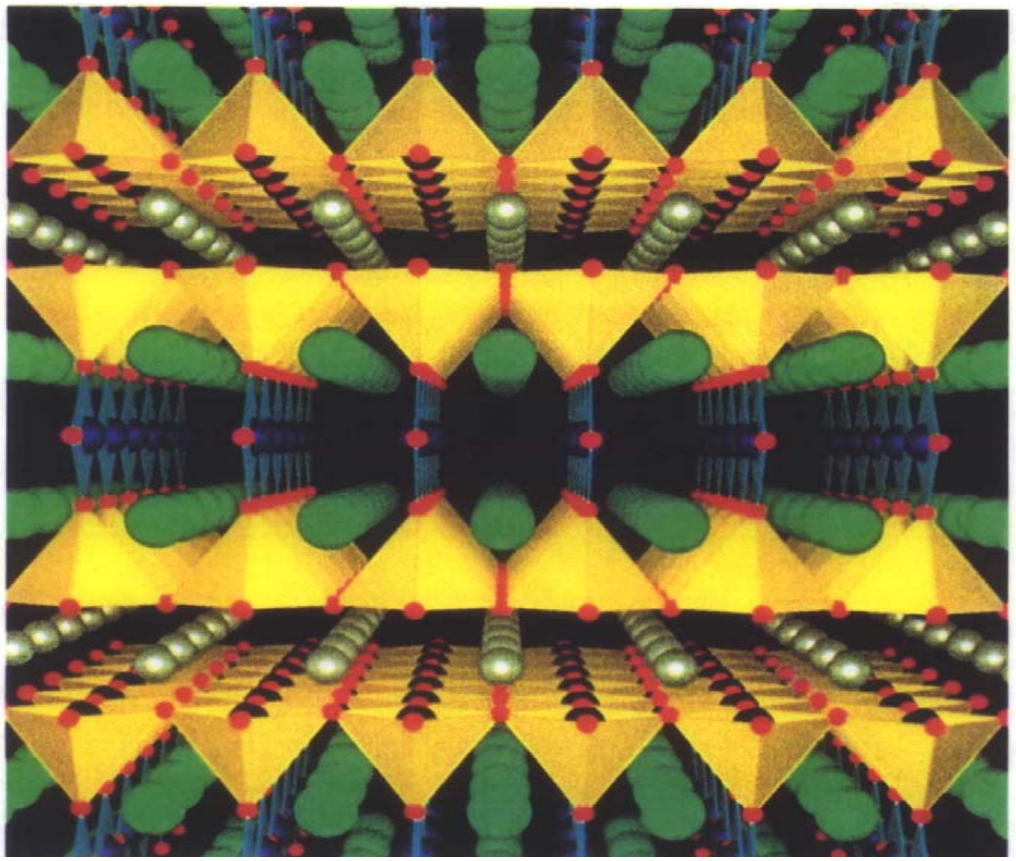
Université de Sherbrooke

USHERBROOKE.CA/IQ



PJRay, O. Gingras

Cuprates : Atomic structure



Outline

- Method
- 3-band Model

- Three experiments
- Pairing mechanism
- Bonus

- Conclusion

Method :
The precursors

Hohenberg-Kohn : Exchange correlation
Kohn-Sham : Basis set

Density Functional Theory

Method

Metzner, Vollhardt PRL **62**, 324 (1989)
Georges, Kotliar, PRB **45,** 6479 (1992)
Jarrell PRL **69,** 168 (1992)
Review: Georges, Kotliar, Krauth, Rozenberg, RMP **68**, 13 (1996)

Dynamical Mean-Field Theory : DMFT

Method

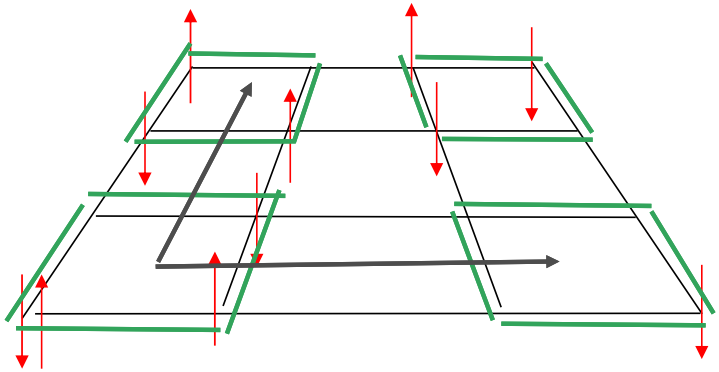
Cluster generalization of
Dynamical Mean-Field Theory : DMFT

REVIEWS
Maier, Jarrell et al., RMP. (2005)
Kotliar *et al.* RMP (2006)
AMST *et al.* LTP (2006)

Lichtenstein *et al.*,PRB 2000
Kotliar *et al.*, PRB 2000
M. Potthoff, EJP 2003

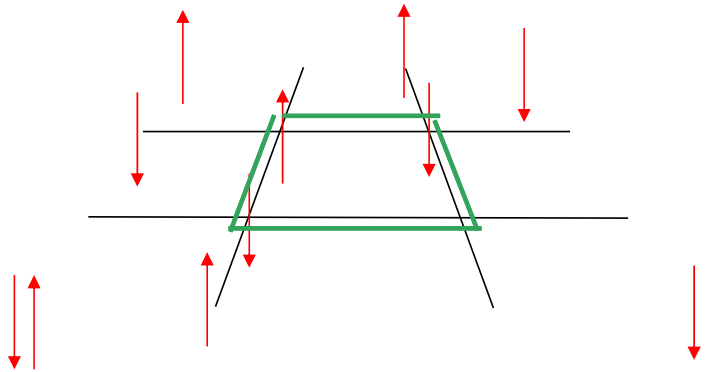
Localized and delocalized pictures C-DMFT

Delocalized



$$\mathbf{R} \rightarrow \tilde{k}$$

Localized



$$G_{ij} = \int \frac{d^d\tilde{k}}{(2\pi)^d} \left(\frac{1}{(i\omega_n+\mu)I-\varepsilon(\tilde{k})-\Sigma} \right)_{ij}$$

$$(G^{-1})_{ij} = (G_0^{-1})_{ij} - \Sigma_{ij}$$

REVIEWS
Maier, Jarrell et al., RMP. (2005)
Kotliar *et al.* RMP (2006)
AMST *et al.* LTP (2006)

Lichtenstein *et al.*,PRB 2000
Kotliar *et al.*, PRB 2000
M. Potthoff, EJP 2003

Impurity solvers

Impurity solver : continuous-time quantum Monte Carlo

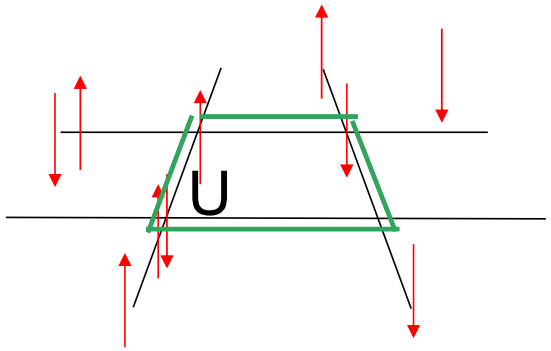
$$Z = \int \mathcal{D}[\psi^\dagger, \psi]\, e^{-S_c - \int_0^\beta d\tau \int_0^\beta d\tau' \sum_{\mathbf{K}} \psi^\dagger_{\mathbf{K}}(\tau)\Delta_{\mathbf{K}}(\tau,\tau')\psi_{\mathbf{K}}(\tau')}$$

Hybridization expansion :

Werner Millis PRB **74**, 155107 (2006)
Werner Millis B **75**, 085108 (2007)
Haule, PRB **75**, 155113 (2007)
Sémon, Sordi, AMST PRB **89**, 165113 (2014)
Sémon, Yee, Haule, AMST PRB **90,** 075149 (2014)

triqs **ALPSCore / CT-HYB** **iQIST** **ComCTQMC**

Impurity solver (Exact diagonalisation)



Caffarel, Krauth, PRL **72** 1545 (1994)

QCM David Sénéchal

Some groups using these methods for cuprates

- Europe:
  - Georges, Parcollet, Ferrero, Civelli (Paris)
  - Lichtenstein, Potthoff, (Hamburg) Aichhorn (Graz), Liebsch (Jülich) de Medici (Grenoble) Capone (Italy)
- USA:
  - Gull (Michigan) Millis (Columbia)
  - Kotliar, Haule (Rutgers) (Haule, Kotliar PRB **76**, 104509 (2007))
  - Jarrell (Louisiana)
  - Maier, Okamoto (Oakridge)
- Japan
  - Imada (Tokyo) Sakai, Tsunetsugu, Motome
- China
  - Wei Wu …

Critique

+ and -

- Long range order:
  - No mean-field factorization on the cluster
  - Symmetry breaking allowed in the bath
- Included exactly:
  - Short-range dynamical and spatial correlations

- Missing:
  - Long wavelength p-h and p-p fluctuations
  - Hence good when the corresponding correlation lengths are small

Three-band (Emery VSA) Hubbard model

Sidhartha Dash

Nicolas Kowalski

Patrick Sémon

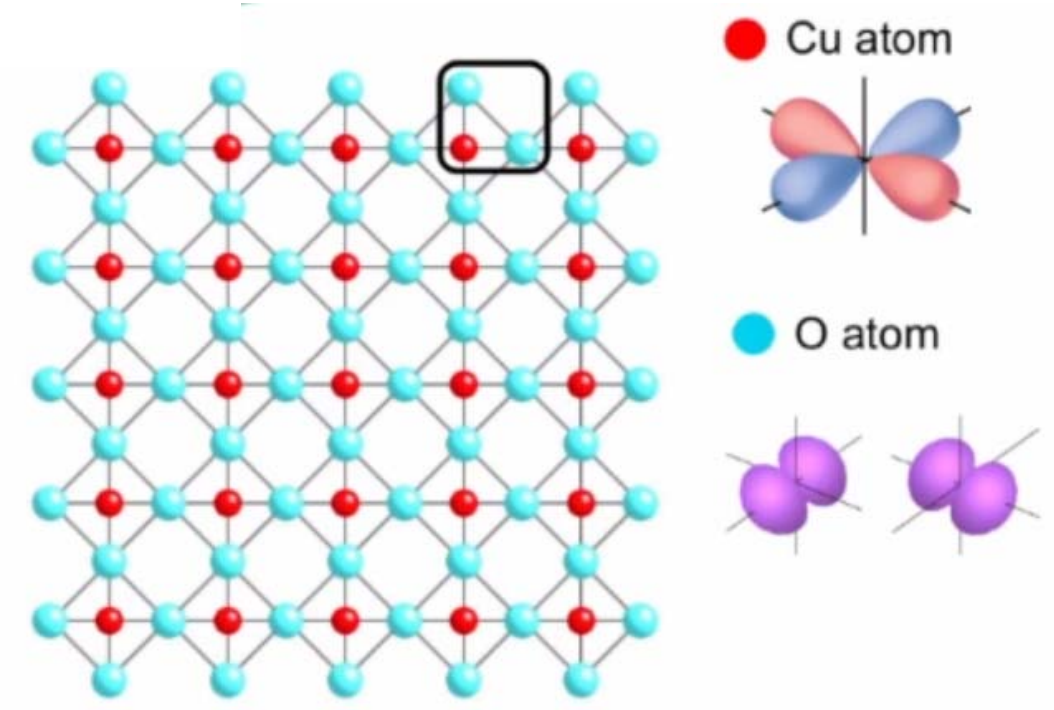
David Sénéchal

V. J. Emery, Phys. Rev. Lett. **58**, 2794 (1987)

C. M. Varma, S. Schmitt-Rink, and E. Abrahams, Solid State Communications **62**, 681–685 (1987), ISSN 0038-1098,

PNAS **118** (40) e2106476118 (2021)

Copper and oxygen planes



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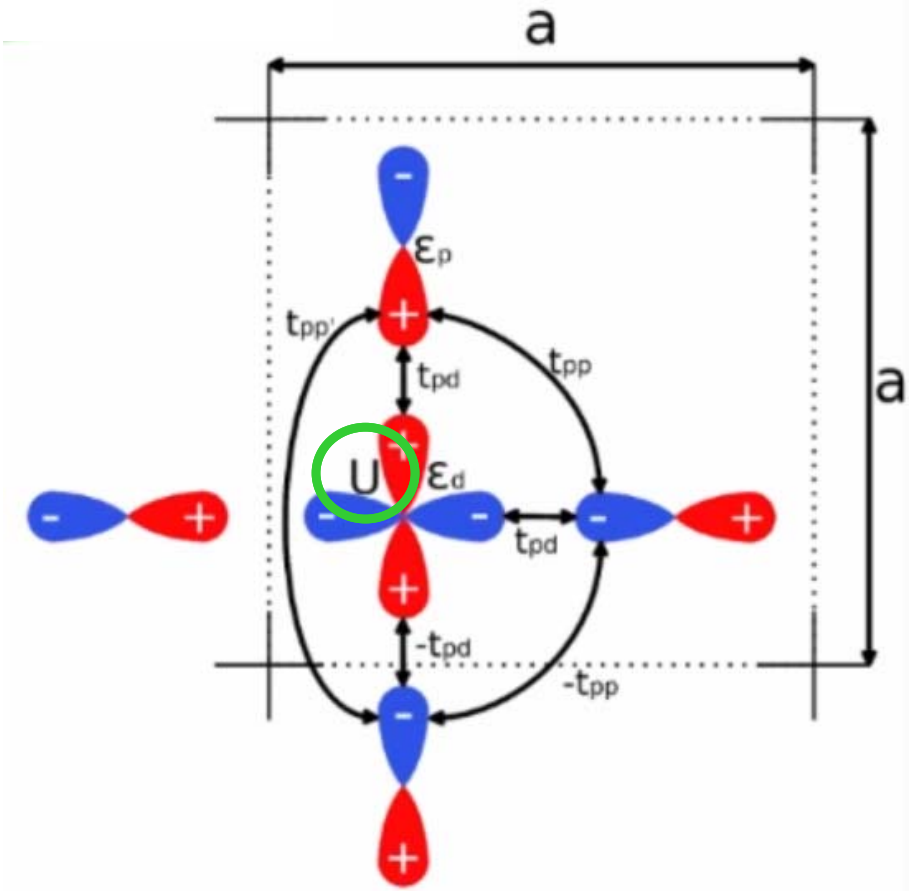

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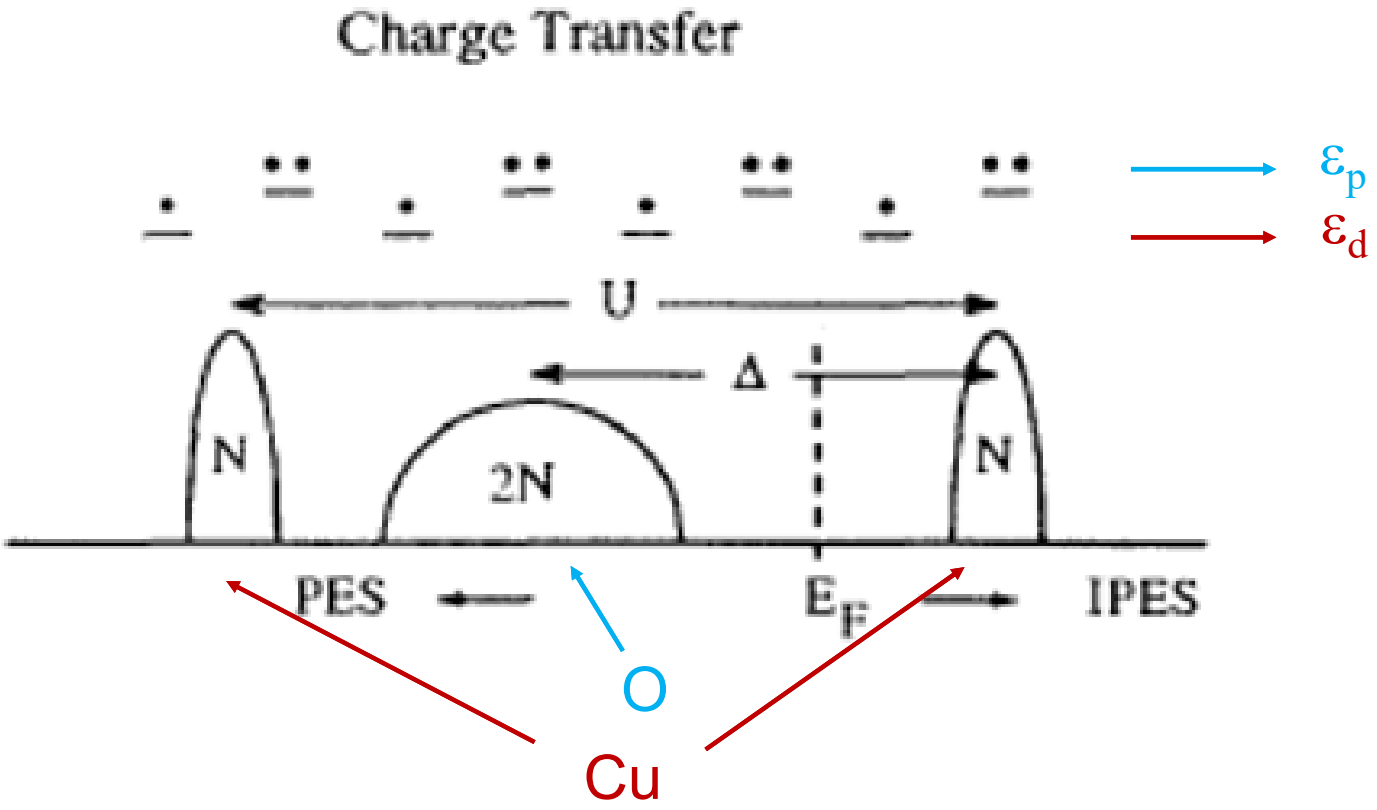
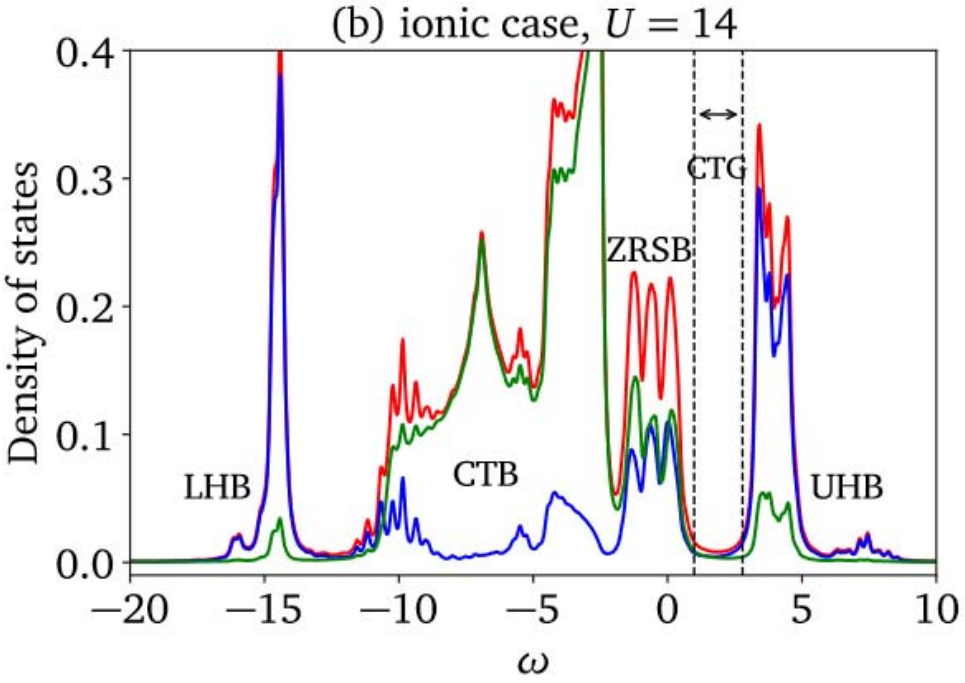
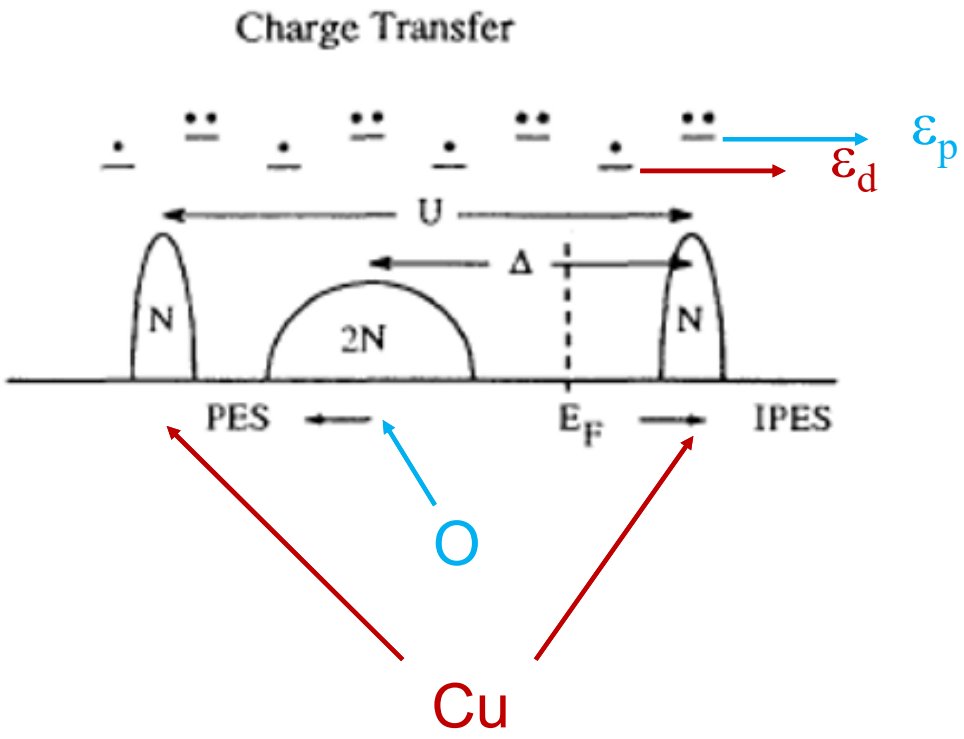
Effect of the various parameters



From Meinders *et al.* PRB **48**, 3916 (1993)

"Ionic" limiting cases with manageable sign problem

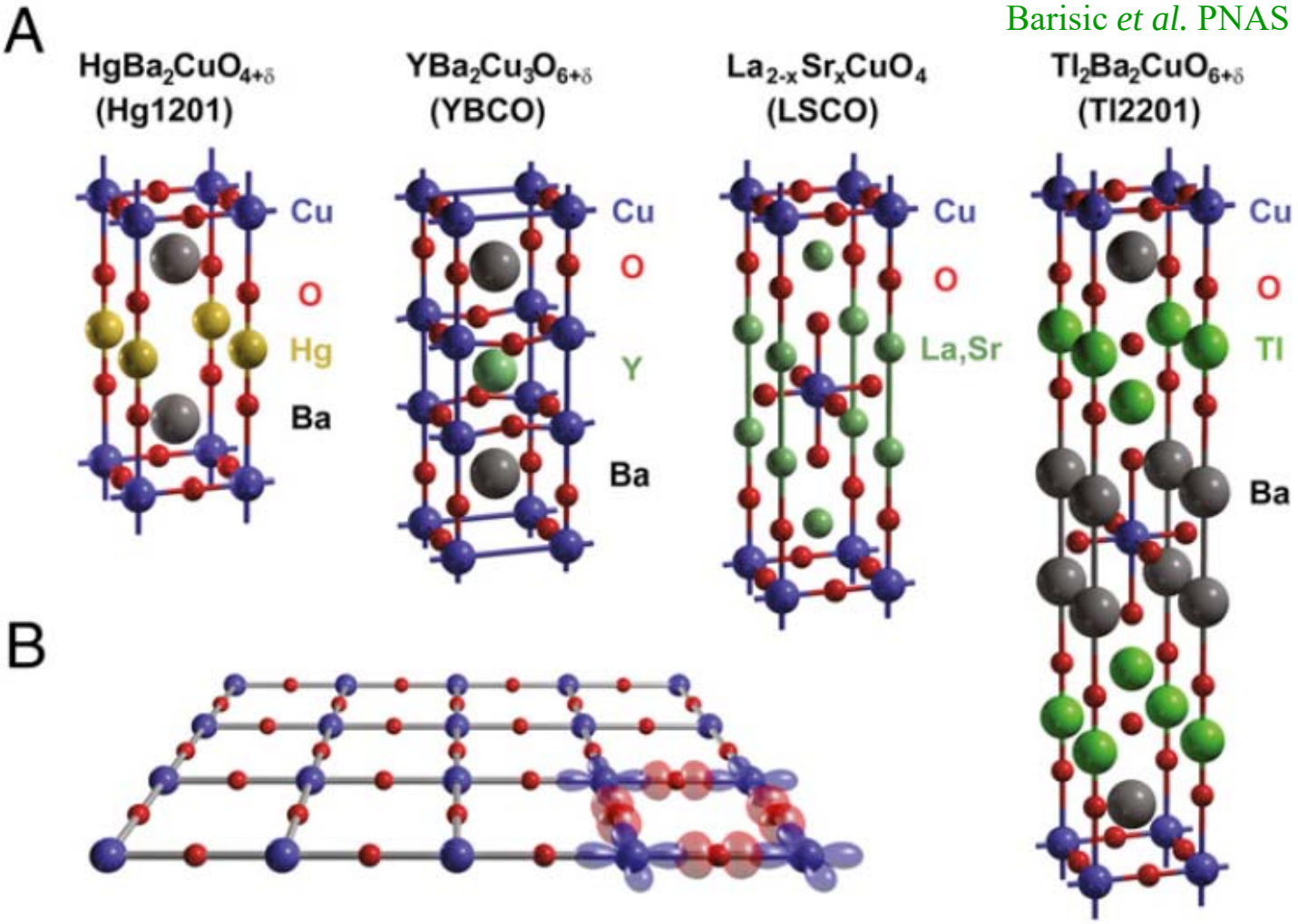




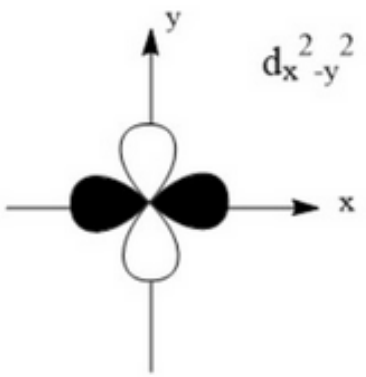
Meinders *et al.* PRB **48**, 3916 (1993)

There are different kinds of cuprates : All with CuO_2 planes

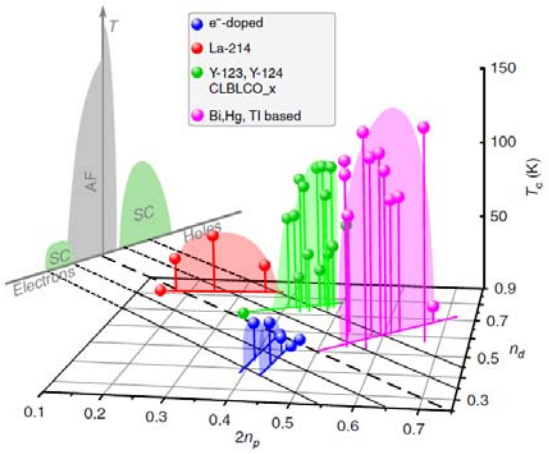
Barisic *et al.* PNAS **110**, 12235 (2013)



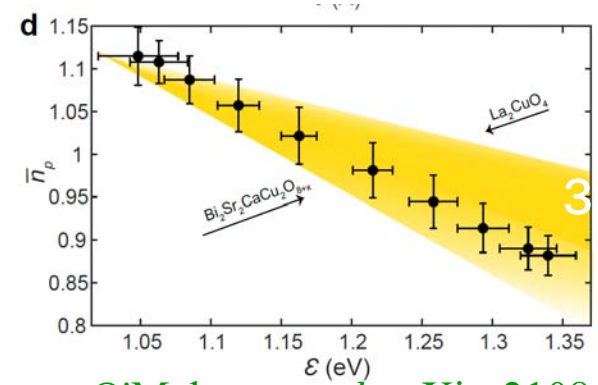
d-wave Superconductivity



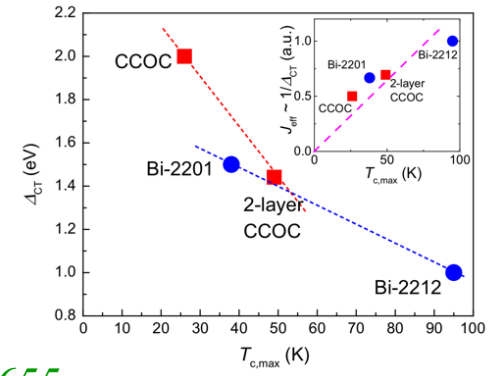
Three experimental observations on optimizing T_c



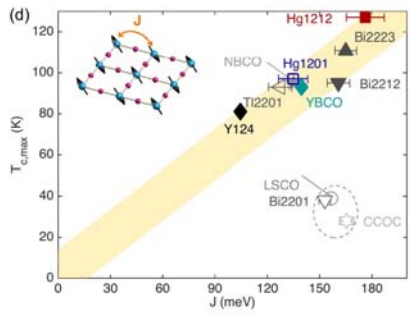
Rybicki, … Haase,
Nat. Comm. 7, 11413
(2016)



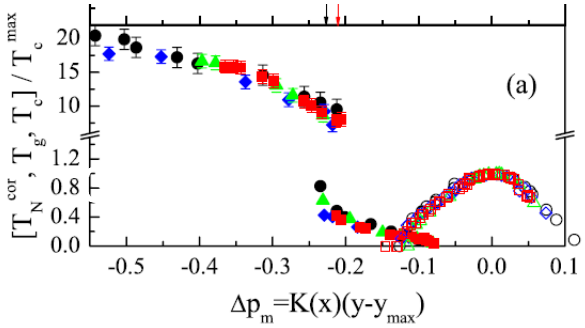
O'Mahony *et al.* arXiv:2108.03655



Ruan *et al.*
Sci. Bull. **61** (2016)



Lichen Wang, *et al.*
arXiv 2011.05029



A. Keren New J. Phys. **11** 065006

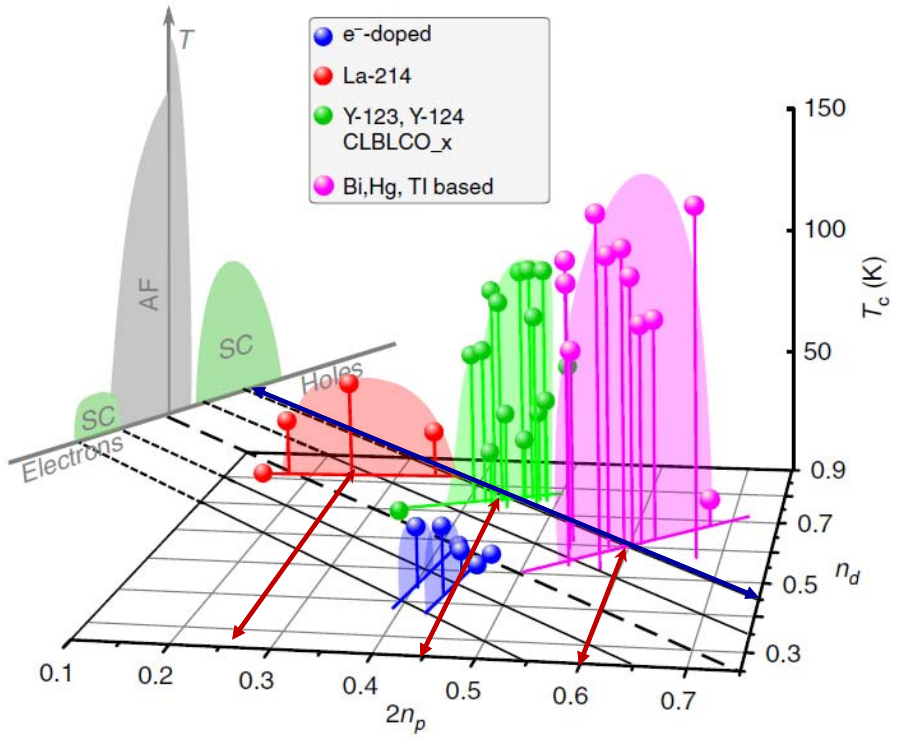
The strategy

The strategy

- Variations in microscopic parameters in Hamiltonian
  - "Ionic" models
    - Large value of $\varepsilon_p - \varepsilon_d$
  - "Covalent" models
    - Smaller and more realistic value of $\varepsilon_p - \varepsilon_d$

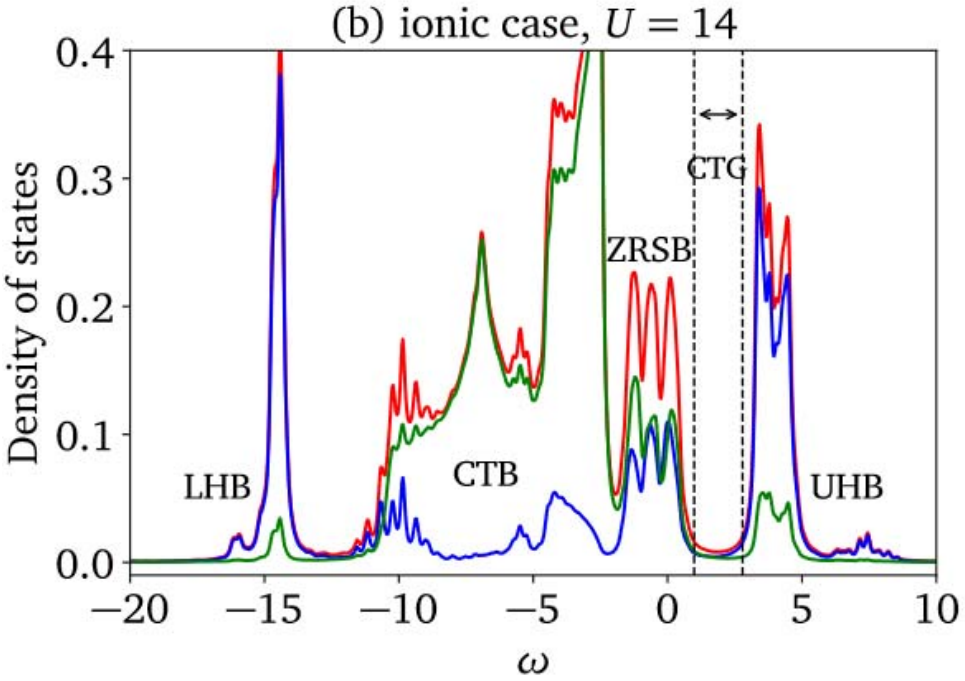
#1 Optimizing T_c with oxygen hole content

#1 Optimizing T_c with oxygen hole content



Rybicki,,, Haase, Nat. Comm. 7, 11413 (2016)

"Ionic" limiting cases with manageable sign problem



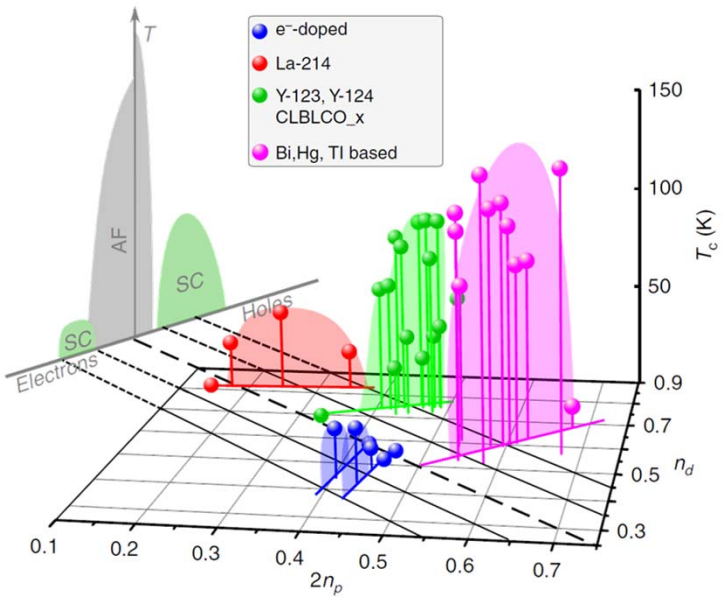
- $\varepsilon_p - \varepsilon_d = 7.0,\ t_{pd} = 1.5,\ t_{pp} = 1.0,\ t'_{pp} = 1.0$

Also, Fratino, Sémon, Sordi, AMT, PRB **93**, 245147 (2016)

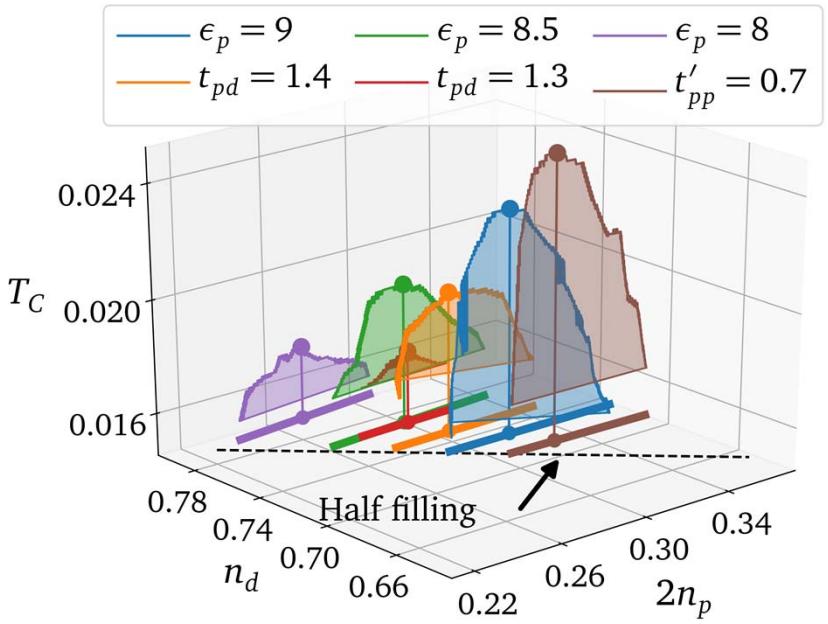
Results

Critical Temperature

- $\epsilon_p - \epsilon_d = 7.0$, $t_{pd} = 1.5$, $t_{pp} = 1.0$, $t'_{pp} = 1.0$



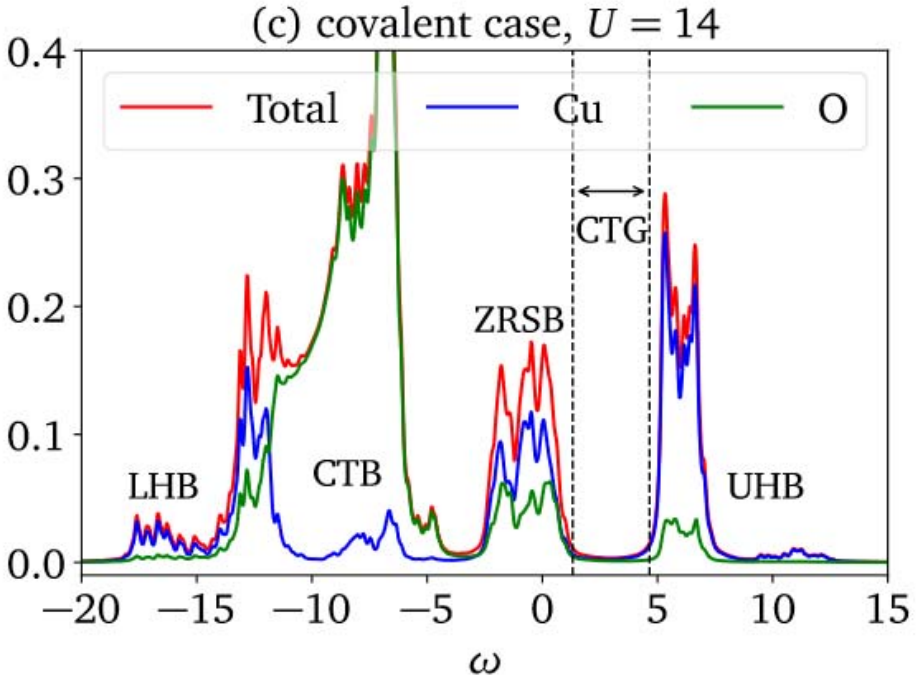
D. Rybicki et al. "Perspective on the phase diagram of cuprate high-temperature superconductors," Nature Communications, vol. 7, p. 11413, 2016



Kowalski, Dash, Sémon, Sénéchal, A-M.T. PNAS **118** (40) e2106476118 (2021)

"Covalent" models ($T = 0$)

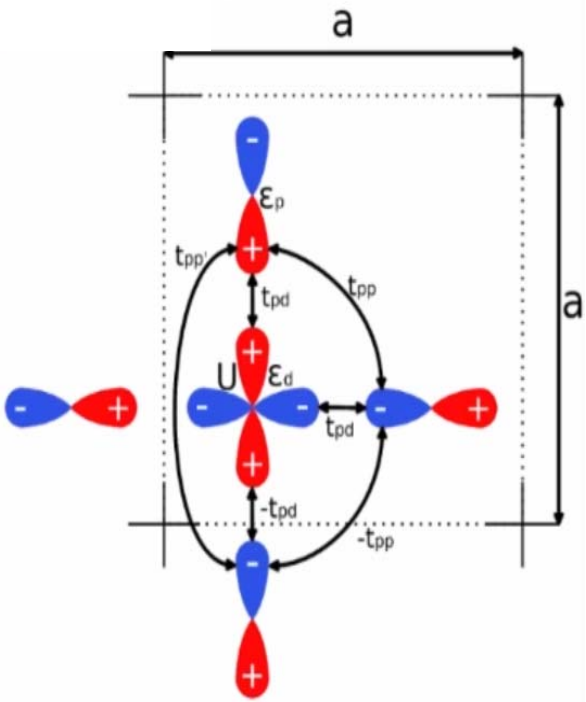
"Realistic"



- $\varepsilon_p - \varepsilon_d = 2.3$, $t_{pd} = 2.1$, $t_{pp} = 1.0$, $t'_{pp} = 0.2$

Electronic structure

	Compound	$\epsilon_d - \epsilon_p$ (eV)	t_{pd} (eV)	t_{pp} (eV)	$t_{pp'}$ (eV)	t'/t	layers	$d^{\text{apical}}_{\text{Cu-O}}$ (Å)	T_c (K)
(1)	La_2CuO_4	2.61	1.39	0.640	0.103	0.070	1	2.3932	38
(2)	$Pb_2Sr_2YCu_3O_8$	2.32	1.30	0.673	0.160	0.108	2	2.3104	70
(3)	$Ca_2CuO_2Cl_2$	2.21	1.27	0.623	0.132	0.085	1	2.7539	26
(4)	$La_2CaCu_2O_6$	2.20	1.31	0.644	0.152	0.120	2	2.2402	45
(5)	$Sr_2Nd_2NbCu_2O_{10}$	2.10	1.25	0.612	0.144	0.110	2	2.0450	28
(6)	$Bi_2Sr_2CuO_6$	2.06	1.36	0.677	0.153	0.105	1	2.5885	24
(7)	$YBa_2Cu_3O_7$	2.05	1.28	0.673	0.150	0.110	2	2.0936	93
(8)	$HgBa_2CaCu_2O_6$	1.93	1.28	0.663	0.187	0.133	2	2.8053	127
(9)	$HgBa_2CuO_4$	1.93	1.25	0.649	0.161	0.122	1	2.7891	90
(10)	$Sr_2CuO_2Cl_2$	1.87	1.15	0.590	0.140	0.108	1	2.8585	30
(11a)	$HgBa_2Ca_2Cu_3O_8$ (outer)	1.87	1.29	0.674	0.184	0.141	3	2.7477	135
(11b)	$HgBa_2Ca_2Cu_3O_8$ (inner)	1.94	1.29	0.656	0.167	0.124	3	2.7477	135
(12)	$Tl_2Ba_2CuO_6$	1.79	1.27	0.630	0.150	0.121	1	2.7143	90
(13)	$LaBa_2Cu_3O_7$	1.77	1.13	0.620	0.188	0.144	2	2.2278	79
(14)	$Bi_2Sr_2CaCu_2O_8$	1.64	1.34	0.647	0.133	0.106	2	2.0033	95
(15)	$Tl_2Ba_2CaCu_2O_8$	1.27	1.29	0.638	0.140	0.131	2	2.0601	110
(16a)	$Bi_2Sr_2Ca_2Cu_3O_{10}$ (outer)	1.24	1.32	0.617	0.159	0.138	3	1.7721	108
(16a)	$Bi_2Sr_2Ca_2Cu_3O_{10}$ (inner)	2.24	1.32	0.678	0.198	0.121	3	1.7721	108



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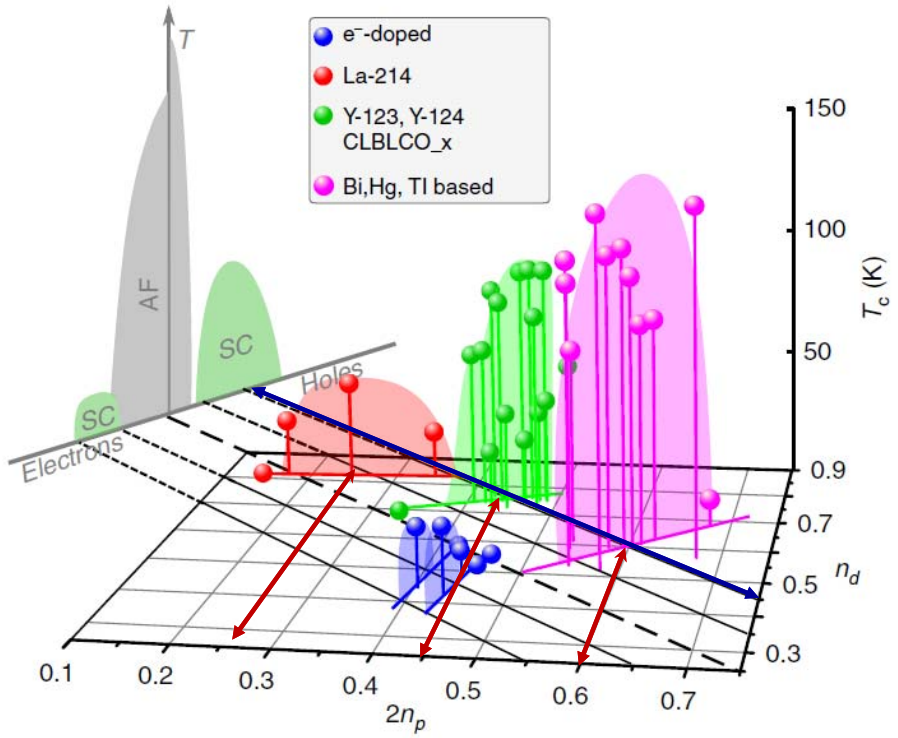
Weber, Yee, Haule, Kotliar, EPL 100, 2012

#1 Optimizing T_c with oxygen hole content

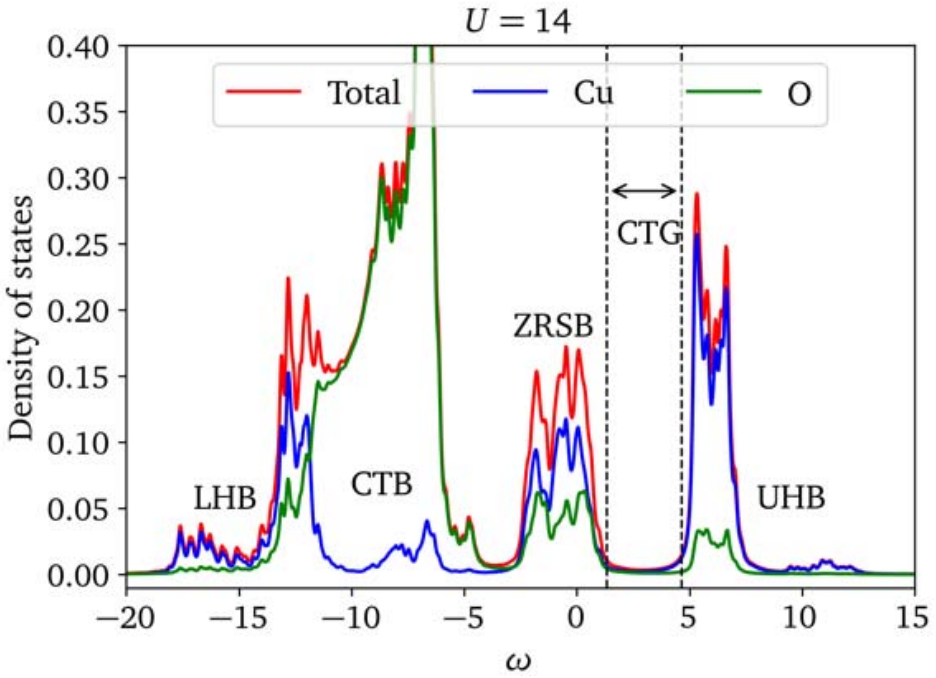


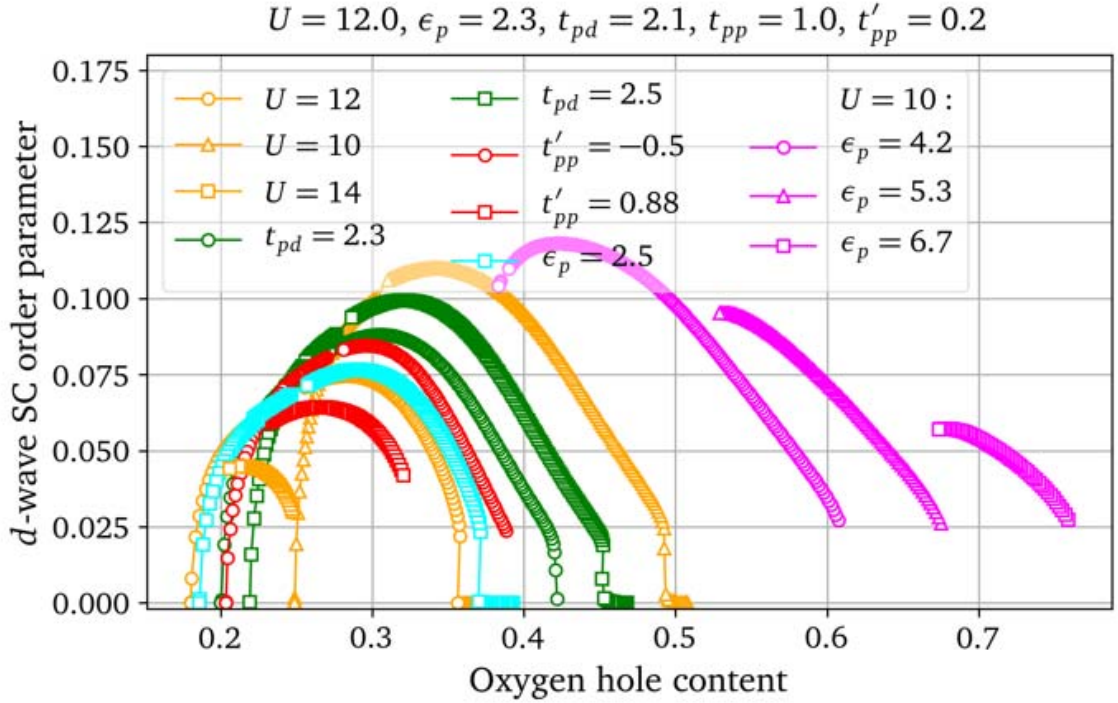
Rybicki,,, Haase, Nat. Comm. 7, 11413 (2016)

T = 0 exact diagonalization solver : order parameter

$$2\hat{\Delta} = \sum_{\langle ij \rangle_x} \left(d_{i,\uparrow} d_{j,\downarrow} - d_{i,\downarrow} d_{j,\uparrow} \right) - \sum_{\langle ij \rangle_y} \left(d_{i,\uparrow} d_{j,\downarrow} - d_{i,\downarrow} d_{j,\uparrow} \right) + \text{H.c.},$$

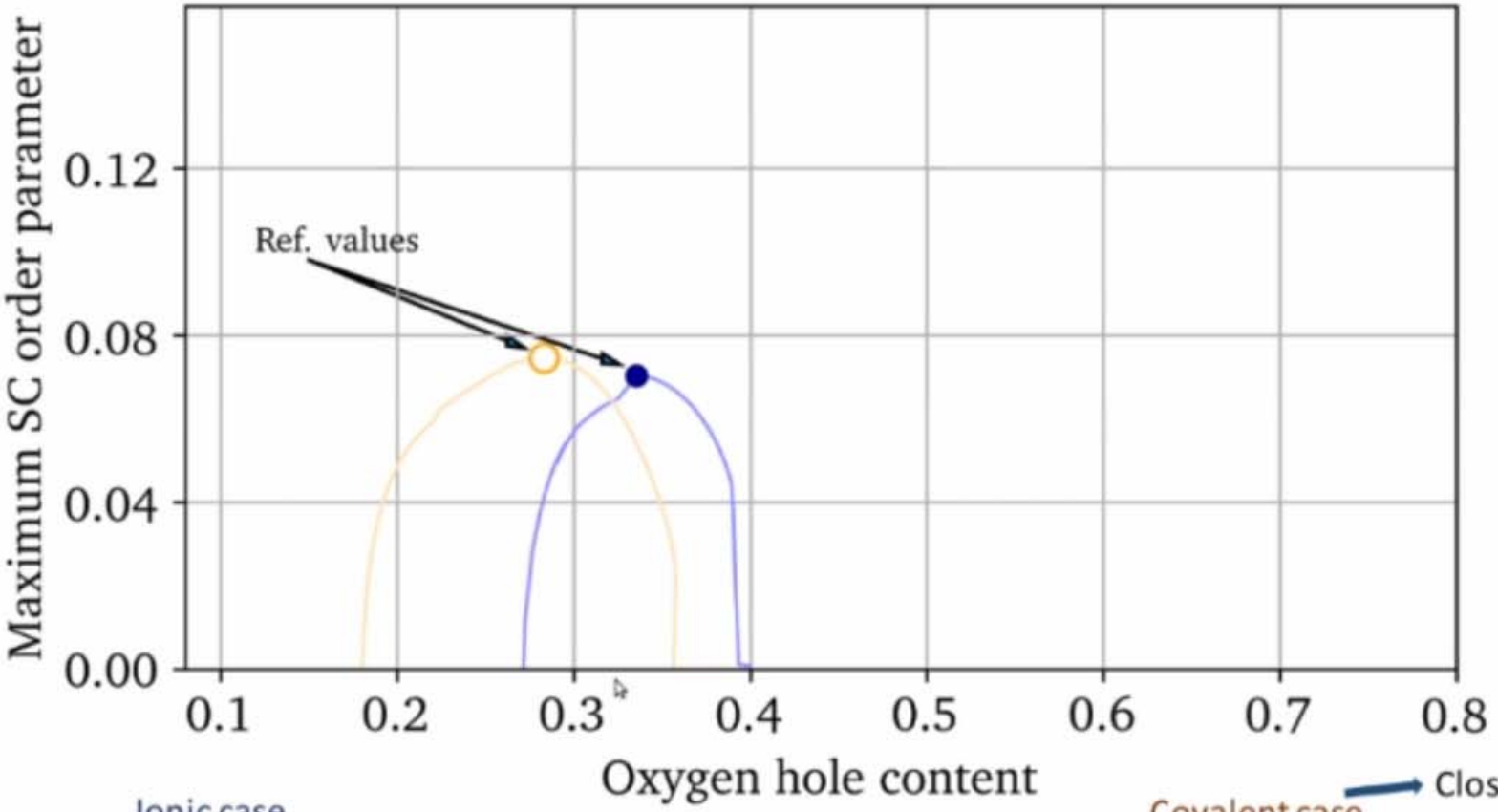
$T = 0$ superconducting domes for the covalent models





Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

T = 0 superconducting domes for the reference models



Ionic case

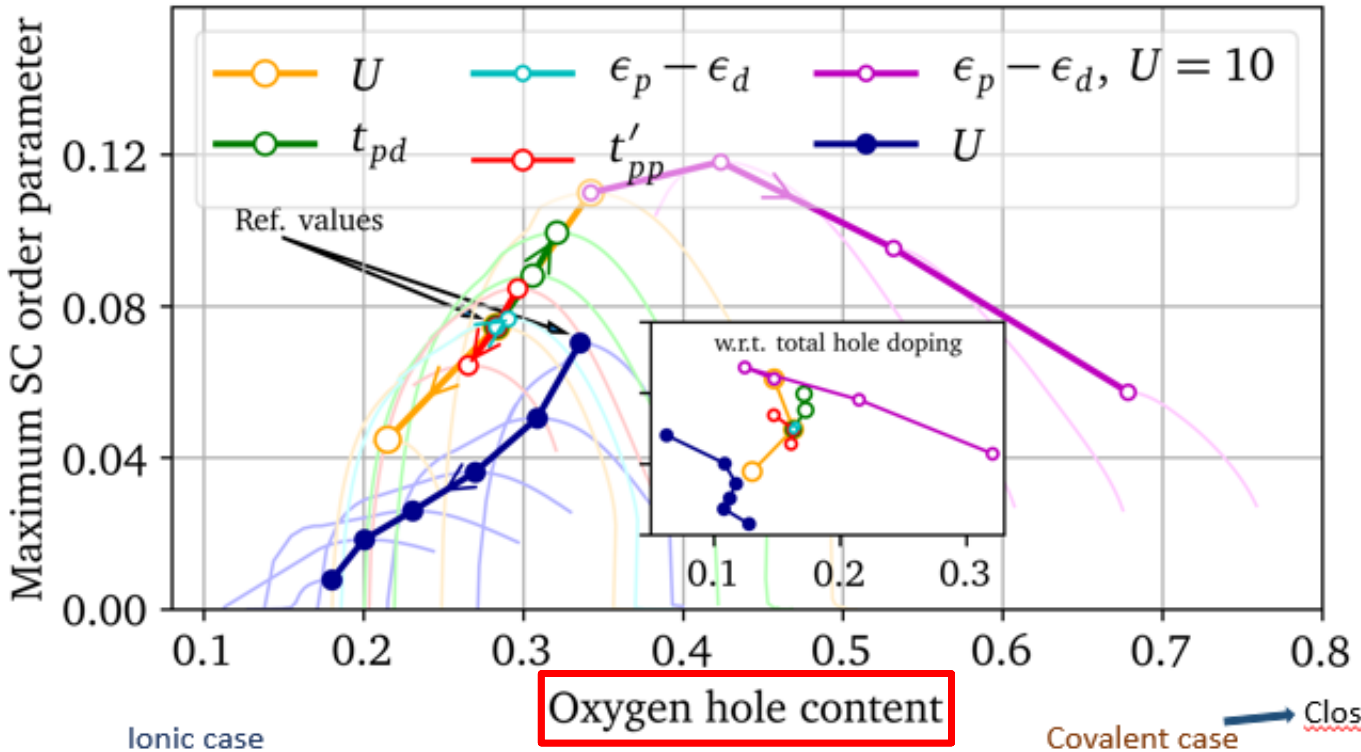
● $U = 12, \boxed{\epsilon_p - \epsilon_d = 7.0}, t_{pd} = 1.5, t_{pp} = 1.0, t'_{pp} = 1.0$

Covalent case

○ $U = 12, \boxed{\epsilon_p - \epsilon_d = 2.3}, t_{pd} = 2.1, t_{pp} = 1.0, t'_{pp} = 0.2$

Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

$T = 0$ max order parameter for the two models



Ionic case

Covalent case

Closer to cuprates

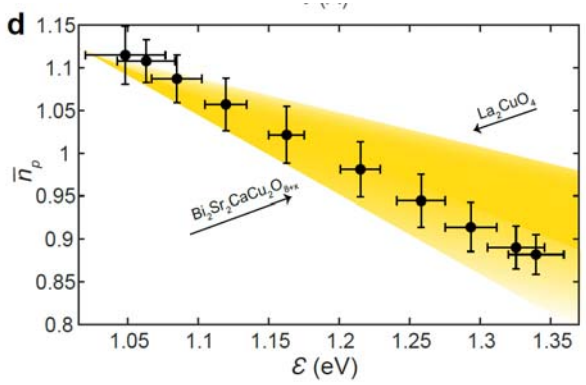
● $U = 12$, $\epsilon_p - \epsilon_d = 7.0$, $t_{pd} = 1.5$, $t_{pp} = 1.0$, $t'_{pp} = 1.0$ ○ $U = 12$, $\epsilon_p - \epsilon_d = 2.3$, $t_{pd} = 2.1$, $t_{pp} = 1.0$, $t'_{pp} = 0.2$

Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

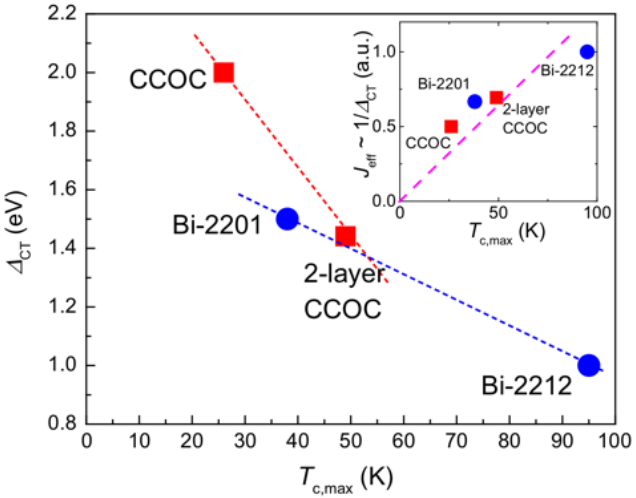
#2 Optimizing T_c with Charge Transfer gap Δ

(Oxygen as a witness)

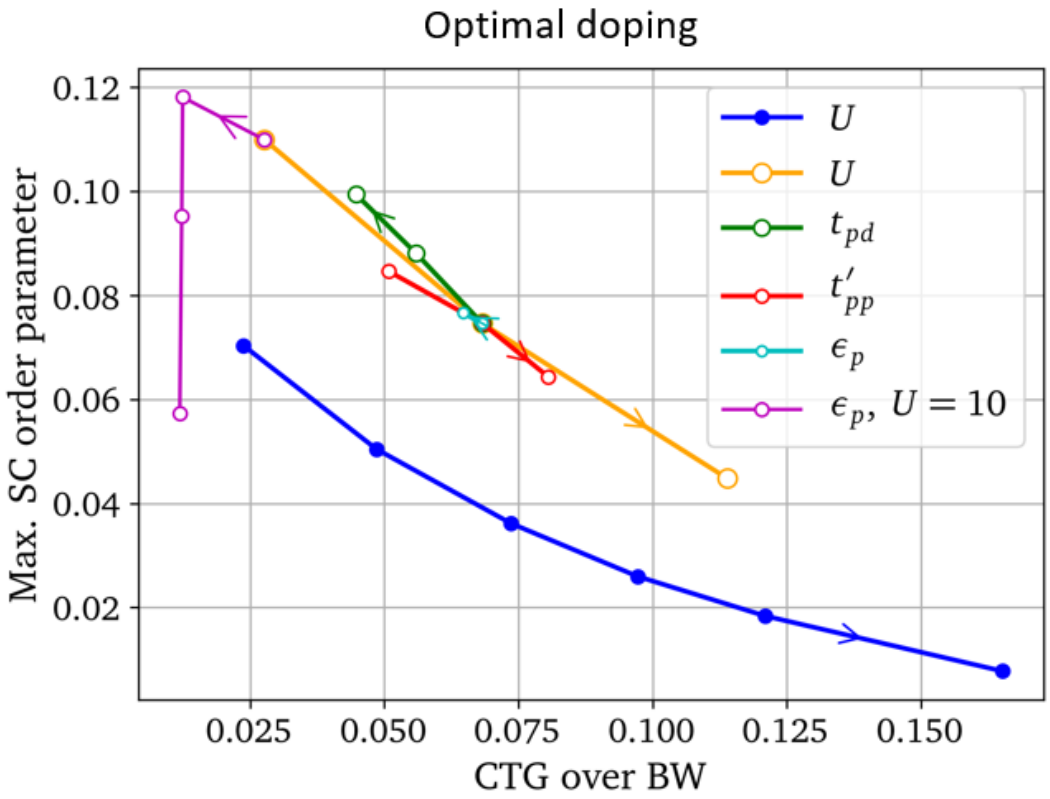
#2 Optimizing T_c with CT gap Δ (Oxygen as a witness)



O'Mahony *et al.* arXiv:2108.03655

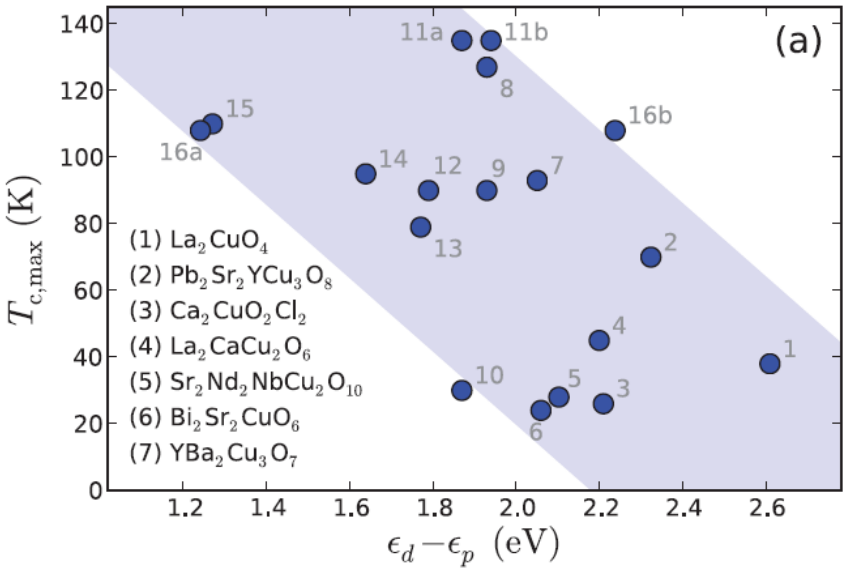


Ruan *et al.* Sci. Bull. **61** (2016)

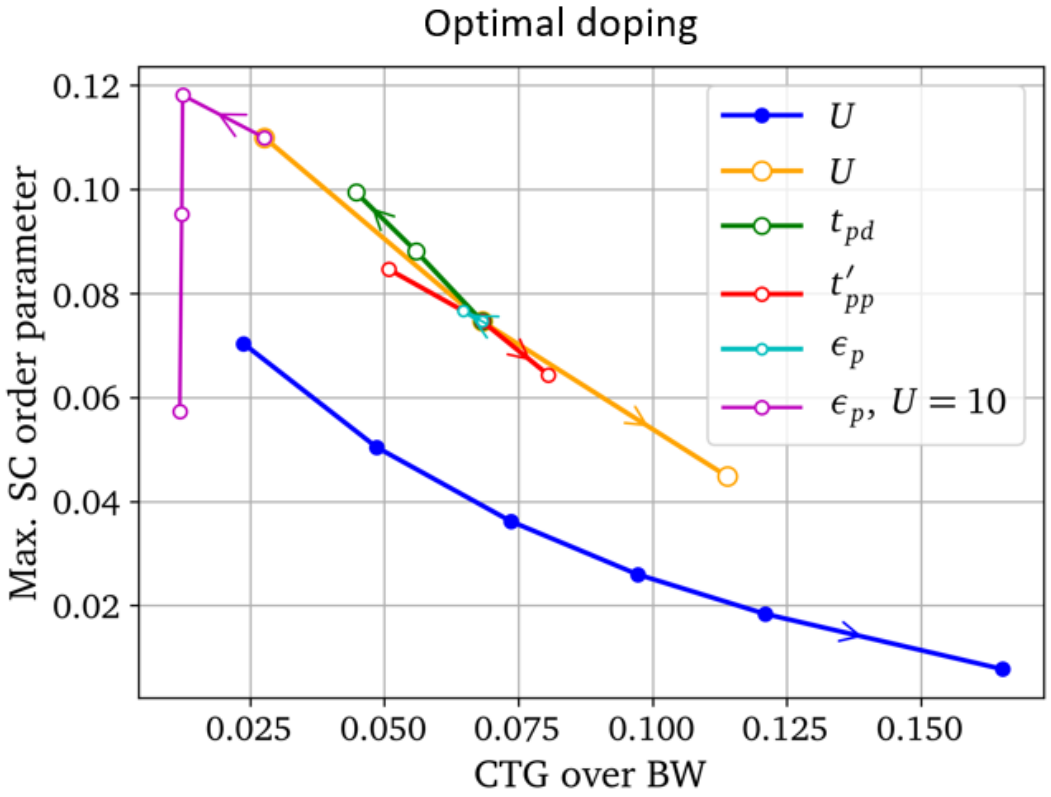


Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

Experimental puzzle #2 with Charge Transfer Gap

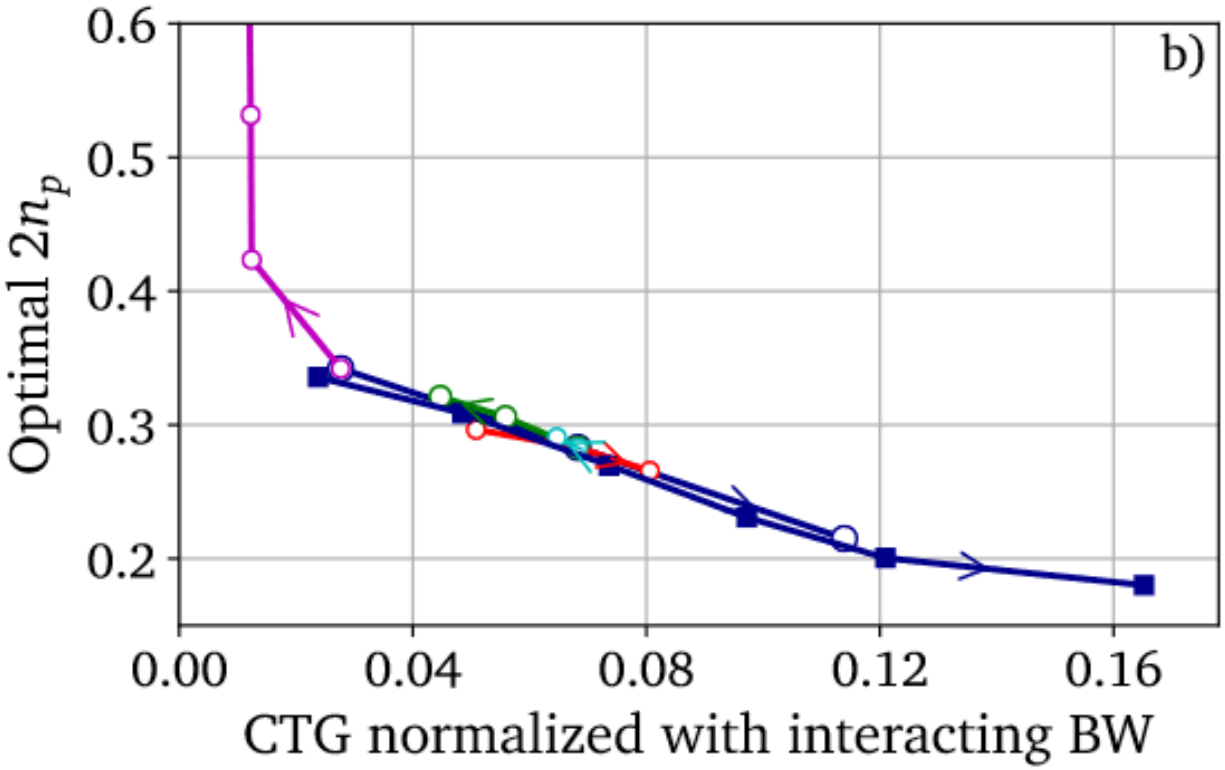


Weber, Yee, Haule, Kotliar, EPL 100, 2012



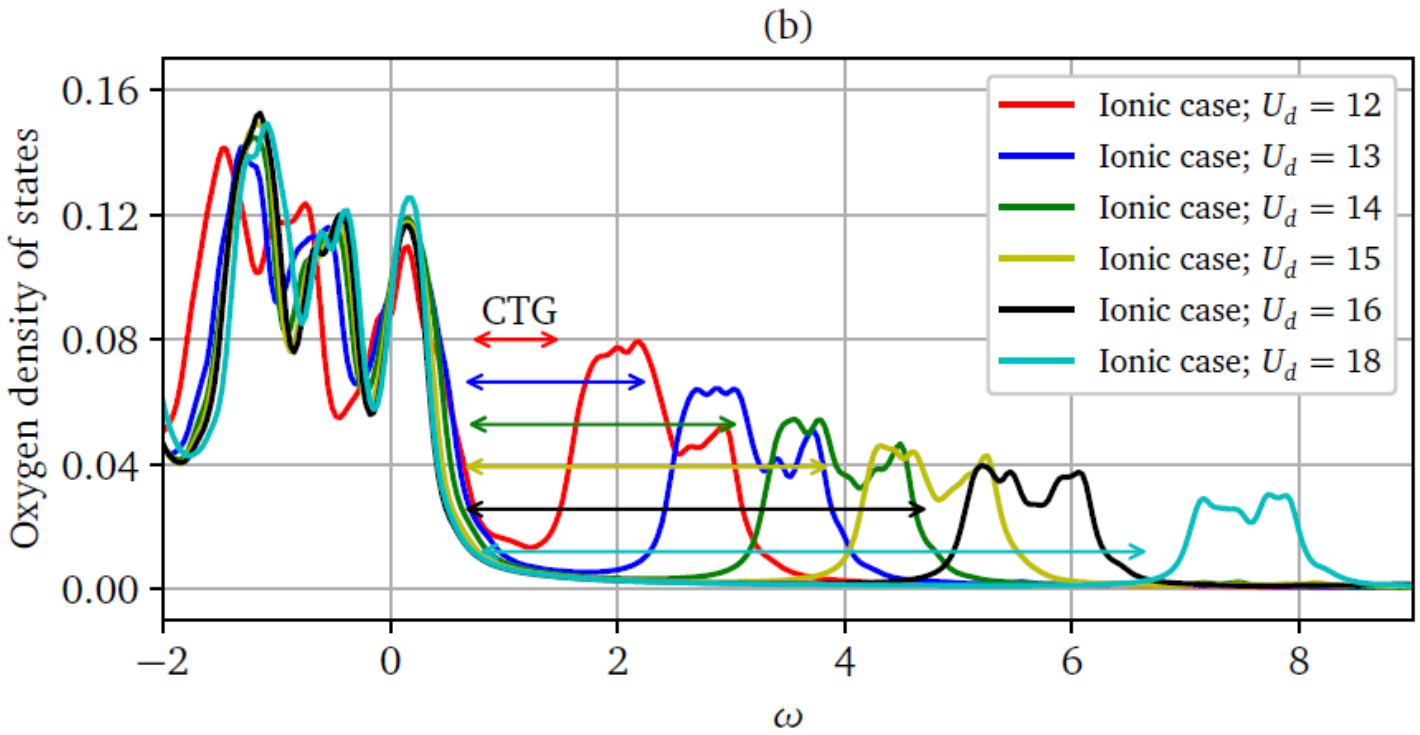
Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

Charge-transfer gap, oxygen hole content



Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

Charge transfer gap and oxygen hole content : Oxygen as a witness



#3 Optimizing T_c with superexchange

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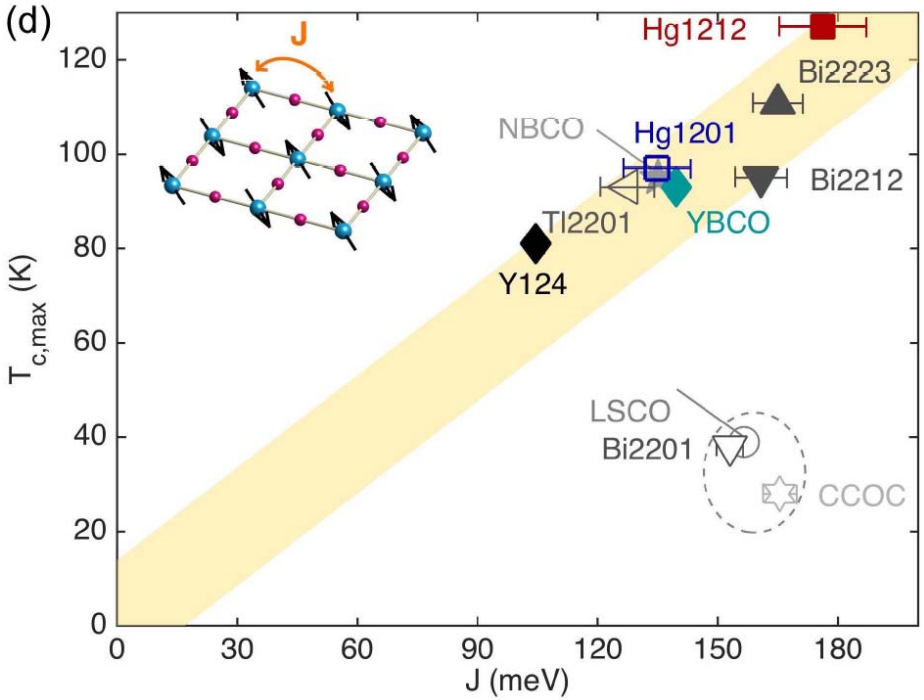
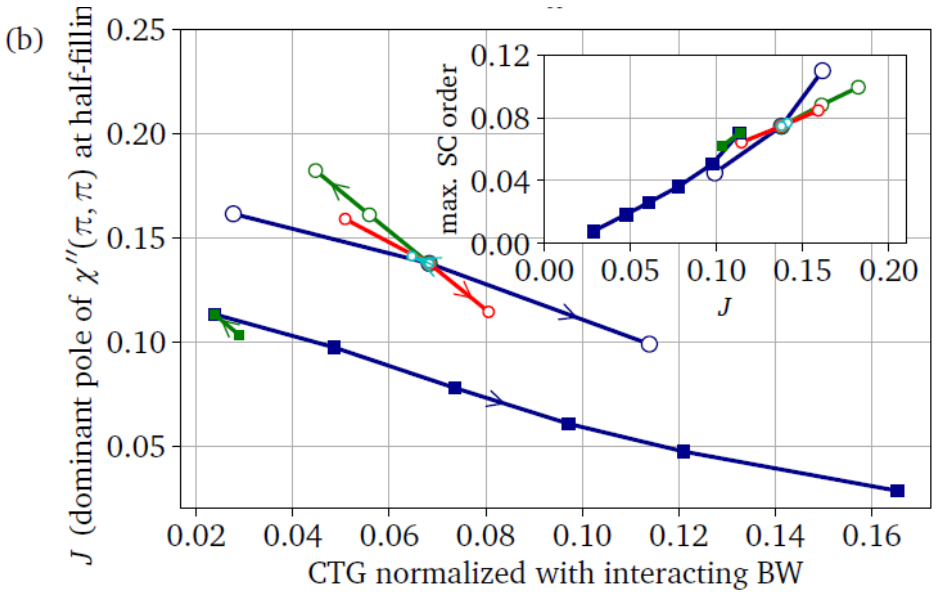




Lichen Wang, *et al.* arXiv 2011.05029

$J = 4t^2/U$

Copper pairing mechanism : superexchange

A cartoon strong correlation picture

$$\hat{\mathcal{H}}_{modèle\ t-J} = -t \sum_{\langle i,j\rangle\ \sigma} \hat{P}\left(\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + c.h\right)\hat{P} + J\sum_{\langle i,j\rangle}\left(\vec{\hat{S}}_i.\vec{\hat{S}}_j - \frac{1}{4}\hat{n}_i\hat{n}_j\right)$$

$$\begin{aligned}
J\hat{S}^z_i\hat{S}^z_j &= J(\hat{n}_{i\uparrow} - \hat{n}_{i\downarrow})(\hat{n}_{j\uparrow} - \hat{n}_{j\downarrow}) \\
&= J(\hat{c}^{\dagger}_{i\uparrow}\hat{c}_{i\uparrow} - \hat{c}^{\dagger}_{i\downarrow}\hat{c}_{i\downarrow})(\hat{c}^{\dagger}_{j\uparrow}\hat{c}_{j\uparrow} - \hat{c}^{\dagger}_{j\downarrow}\hat{c}_{j\downarrow}) \\
&= -J(\hat{c}^{\dagger}_{i\downarrow}\hat{c}_{i\downarrow}\hat{c}^{\dagger}_{j\uparrow}\hat{c}_{j\uparrow} + \hat{c}^{\dagger}_{i\uparrow}\hat{c}_{i\uparrow}\hat{c}^{\dagger}_{j\downarrow}\hat{c}_{j\downarrow}) + \dots \\
&= -J(\hat{c}^{\dagger}_{j\uparrow}\hat{c}^{\dagger}_{i\downarrow}\hat{c}_{i\downarrow}\hat{c}_{j\uparrow} + \hat{c}^{\dagger}_{i\uparrow}\hat{c}^{\dagger}_{j\downarrow}\hat{c}_{j\downarrow}\hat{c}_{i\uparrow}) + \dots
\end{aligned}$$

Hartree-Fock :

$$d^* = \langle \hat{c}^{\dagger}_{j\uparrow}\hat{c}^{\dagger}_{i\downarrow}\rangle_{\mathcal{H}_{modèle\ t-J}}$$

$$\langle J\hat{S}^z_i\hat{S}^z_j\rangle = -2J\,d^*d + \dots$$

Pitaevskii Brückner:
Pair state orthogonal to repulsive core of Coulomb interaction

P.W. Anderson Science 317, 1705 (2007)

Miyake, Schmitt–Rink, and Varma P.R. B **34**, 6554-6556 (1986)

More sophisticated Slave Boson: Kotliar Liu PRB 1988

D. Sénéchal

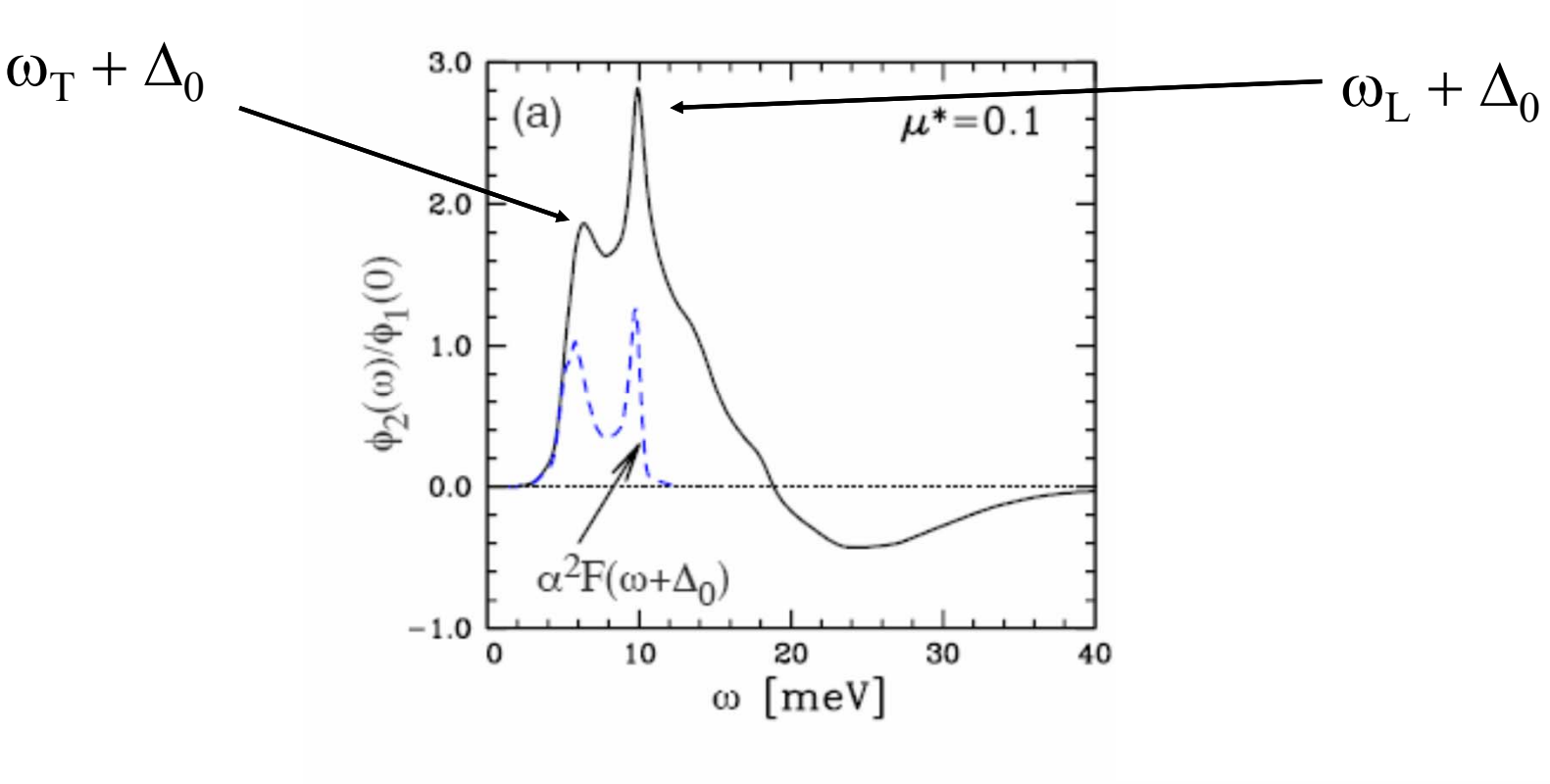
Bumsoo Kyung

The glue

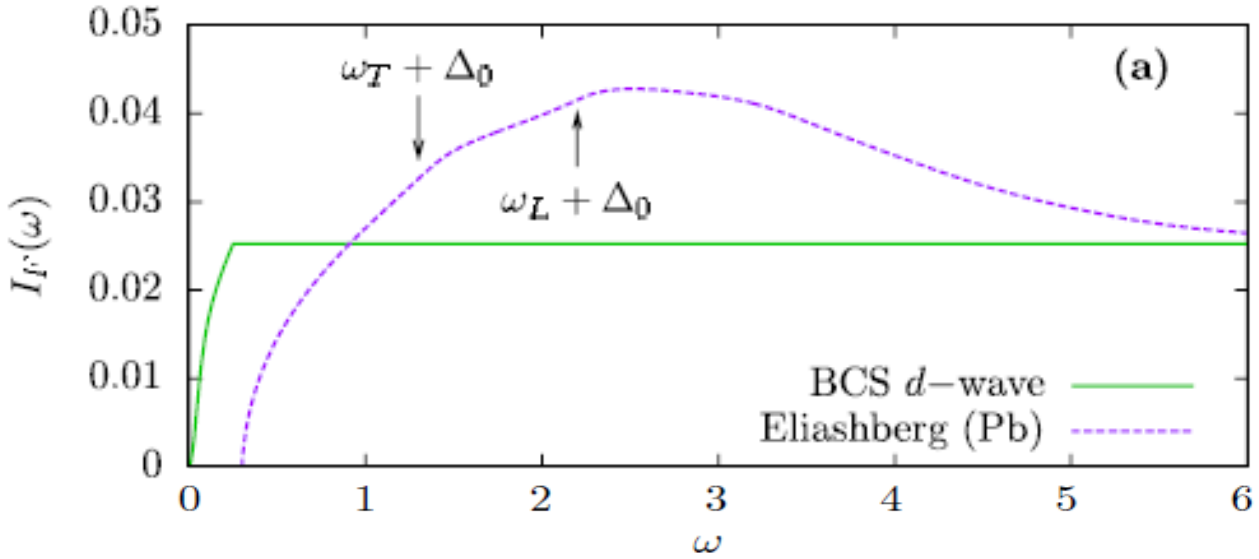
Kyung, Sénéchal, Tremblay, Phys. Rev. B **80,** 205109 (2009)
Sénéchal, Day, Bouliane, AMST, Phys. Rev. B **87**, 075123 (2013)
A. Reymbaut *et al.* PRB **94** 155146 (2016)

Im Σ_{an} and electron-phonon in Pb

Maier, Poilblanc, Scalapino, PRL (2008)



Another way to look at this

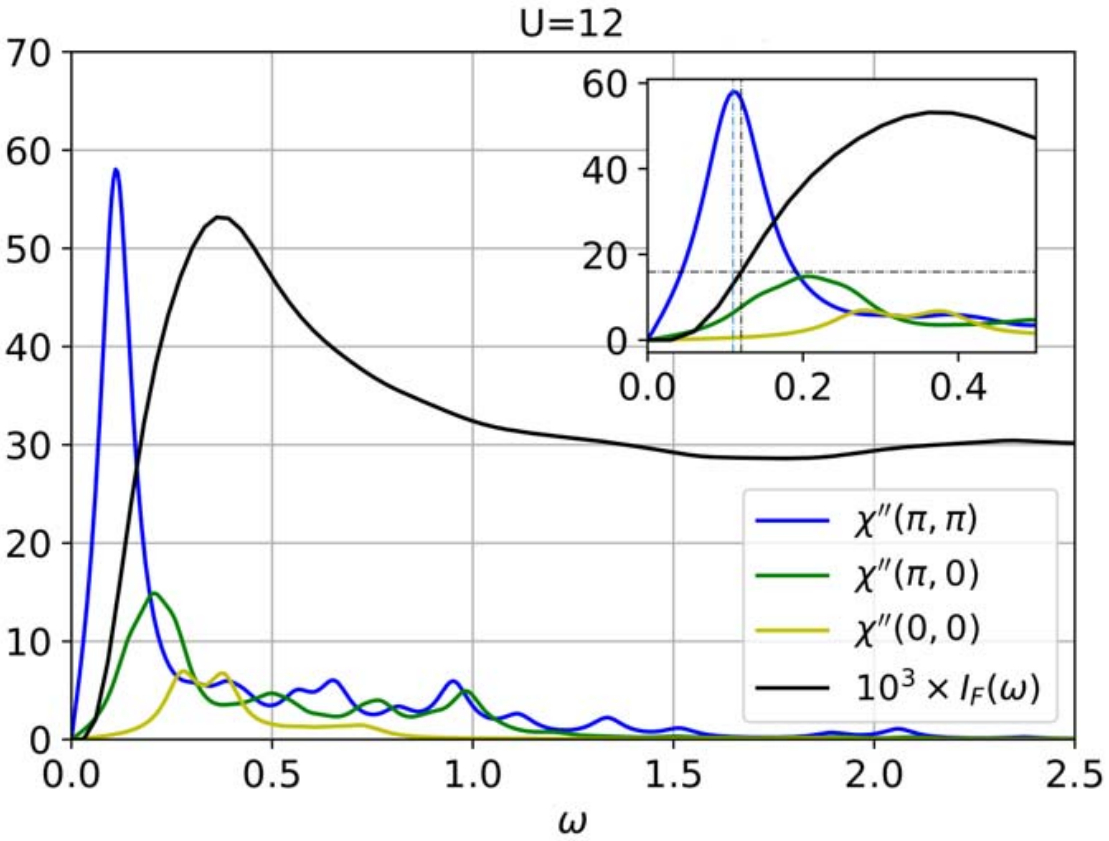


$$I_F(\omega) \equiv -\int_0^{\omega} \frac{d\omega'}{\pi} \mathrm{Im}\, F_{ij}^R(\omega')$$

$$F_{ij} \equiv -\langle T c_{i\uparrow}(\tau) c_{j\downarrow}(0) \rangle$$

Kyung, Sénéchal, Tremblay, Phys. Rev. B **80,** 205109 (2009)
Sénéchal, Day, Bouliane, AMST, Phys. Rev. B **87**, 075123 (2013)
A. Reymbaut *et al.* PRB **94** 155146 (2016)

Spin fluctuations on copper in the three-band model



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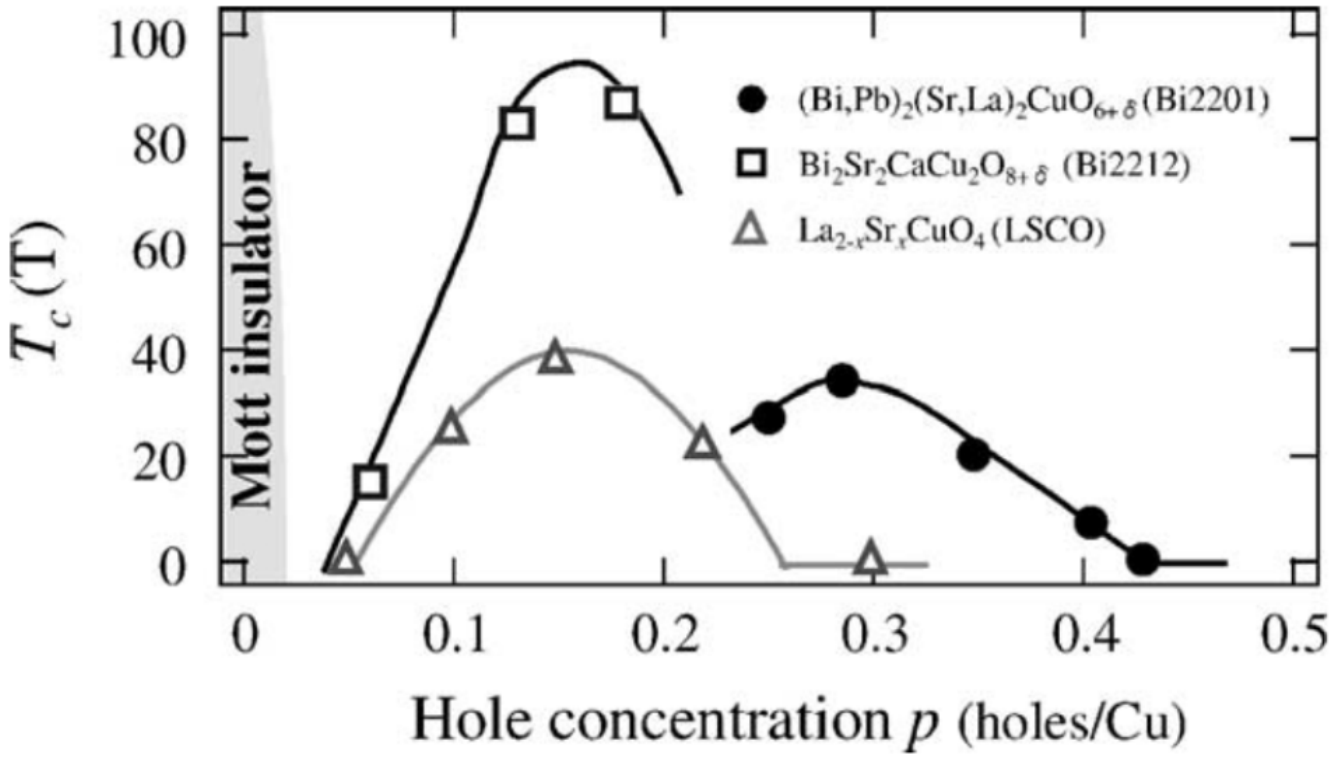
$$F_{ij} \equiv -\langle T c_{i\uparrow}(\tau) c_{j\downarrow}(0) \rangle$$

Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

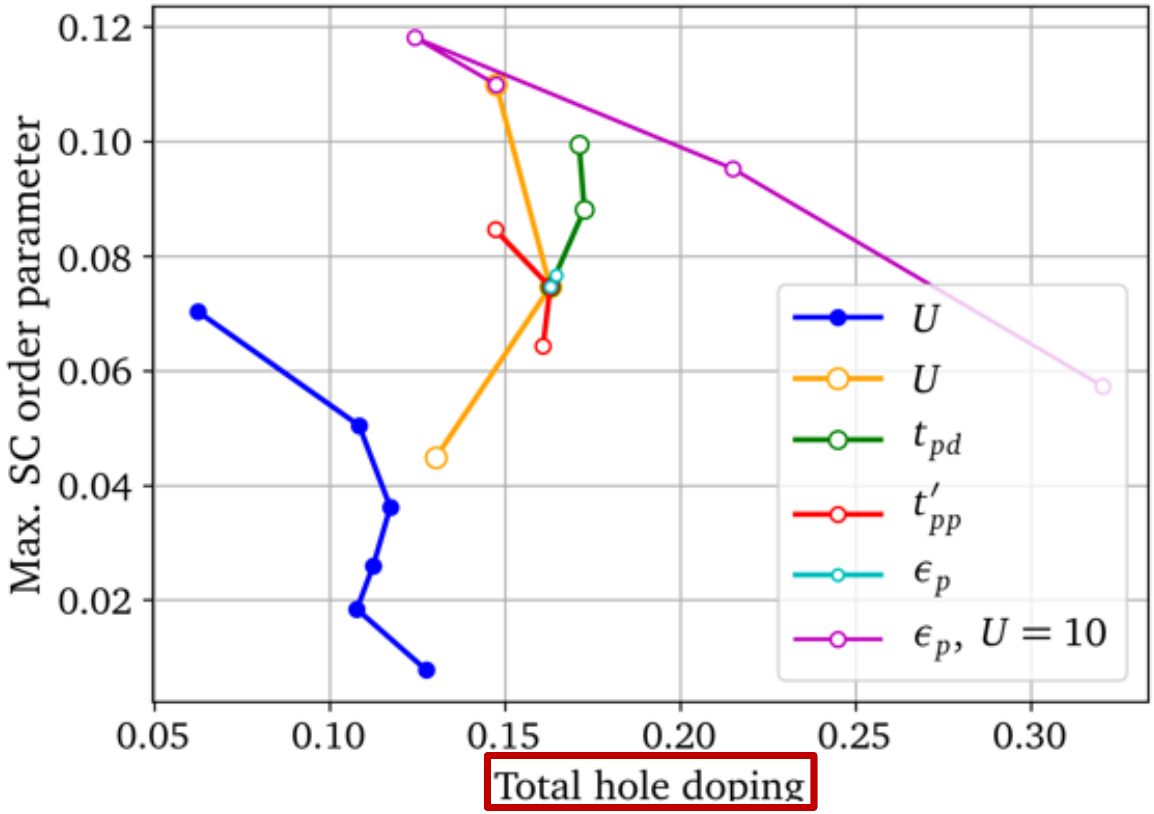
Bonus

T_c and total hole concentration are not well correlated



T. Kondo *et al.*
Journal of Electron Spectroscopy and Related Phenomena **137-140**, 663 (2004)

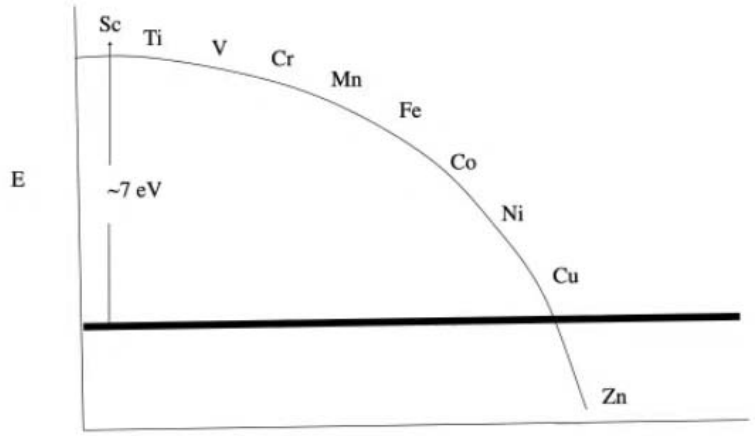
Bonus: total hole doping does not explain max order parameter for the two classes of models



Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

Bonus : Importance of covalency

Affinity Energy ($E(M^{2+}) - E(M^{1+})$) of first row Trans. Metals in relation to Ionization Energy of Oxygen ($E(O^{2-}) - E(O^{1-})$)



Also, Zaanen, Sawatzky, Allen (prl 1985).

C. M. Varma and T. Giamarchi, *Model for copper oxide metals and superconductors* (Elsevier Science B.V., 1995).

Summary Conclusion

Optimizing Tc

- Spin ½
- One band
- Two-dimensions
- Strong covalency between chalchogen and transition metal.
  - Chalcogen screens U
- Charge-transfer gap just opening (intermediate interactions).
- Large *J* at half-filling
- … and more

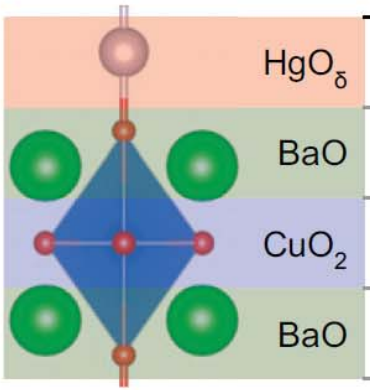
C. Weber, PNAS 2021 Vol. **118** No. 46 e2115874118

Chuck-Hou Yee *et al EPL* **111** 17002 (2015)

Stanev *et al.*, npj Computational Materials **4**, 29 (2018)

Liu *et al.* APL Materials **8**, 061104 (2020)

Optimizing T_c



	charge	dopants	structure	hamiltonian
HgO$_\delta$	balances -2 charge	supplies	harbors dopants	tunes chemical potential
BaO	neutral	inert	protects CuO$_2$ from disorder	tunes in-plane *t*, *t'*, *U*
CuO$_2$	-2 charge/u.c.	accepts	roughly sets lattice const.	superconducts
BaO			(same as other CaS layer)	

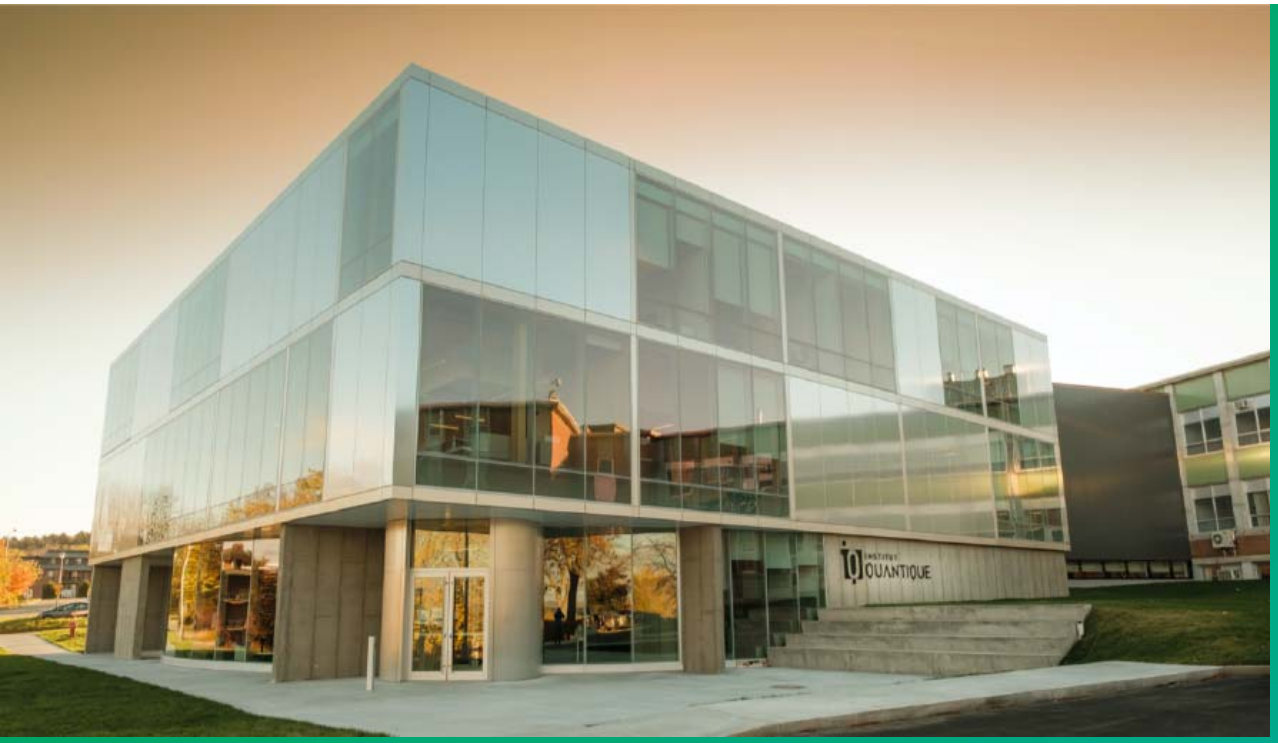
$$\mathrm{Hg(CaS)_2CuO_2}$$

Chuck-Hou Yee *et al EPL* **111** 17002 (2015)

Take home messages

- A detailed picture of the origin of superconductivity in cuprates follows from a model that takes into account Cu, O, kinetic energy and repulsion

- We need to look beyond traditional tools of solid state physics to work this out.



Merci
Thank you

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Fratino, et al.
PRB **93**, 245147 (2016)