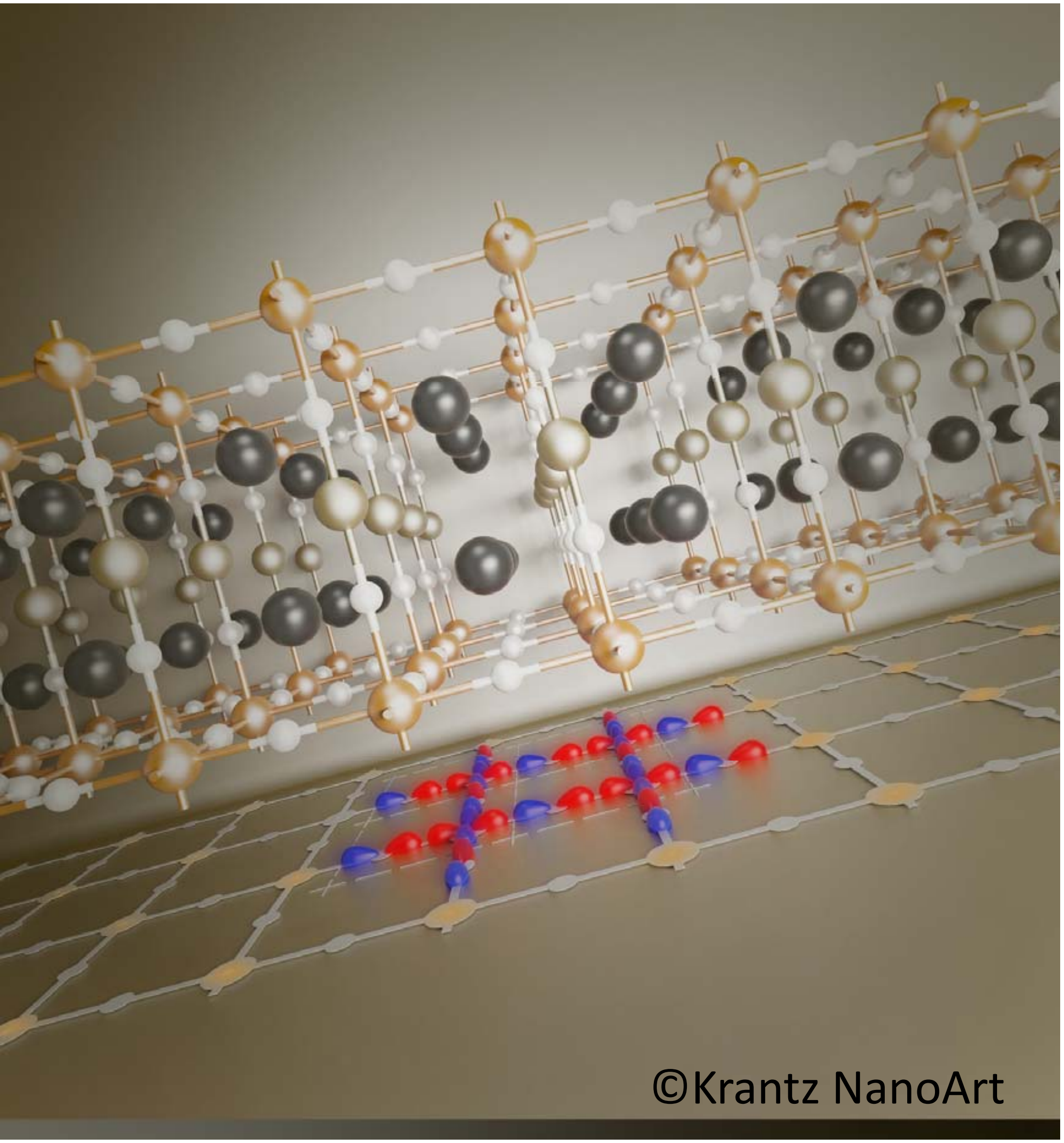
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Mechanism of Superconductivity in Cuprates : oxygen as a witness

André-Marie Tremblay
Université de Sherbrooke
Institut quantique

M2S Vancouver, July 18, 2022

Institut quantique

Université de Sherbrooke

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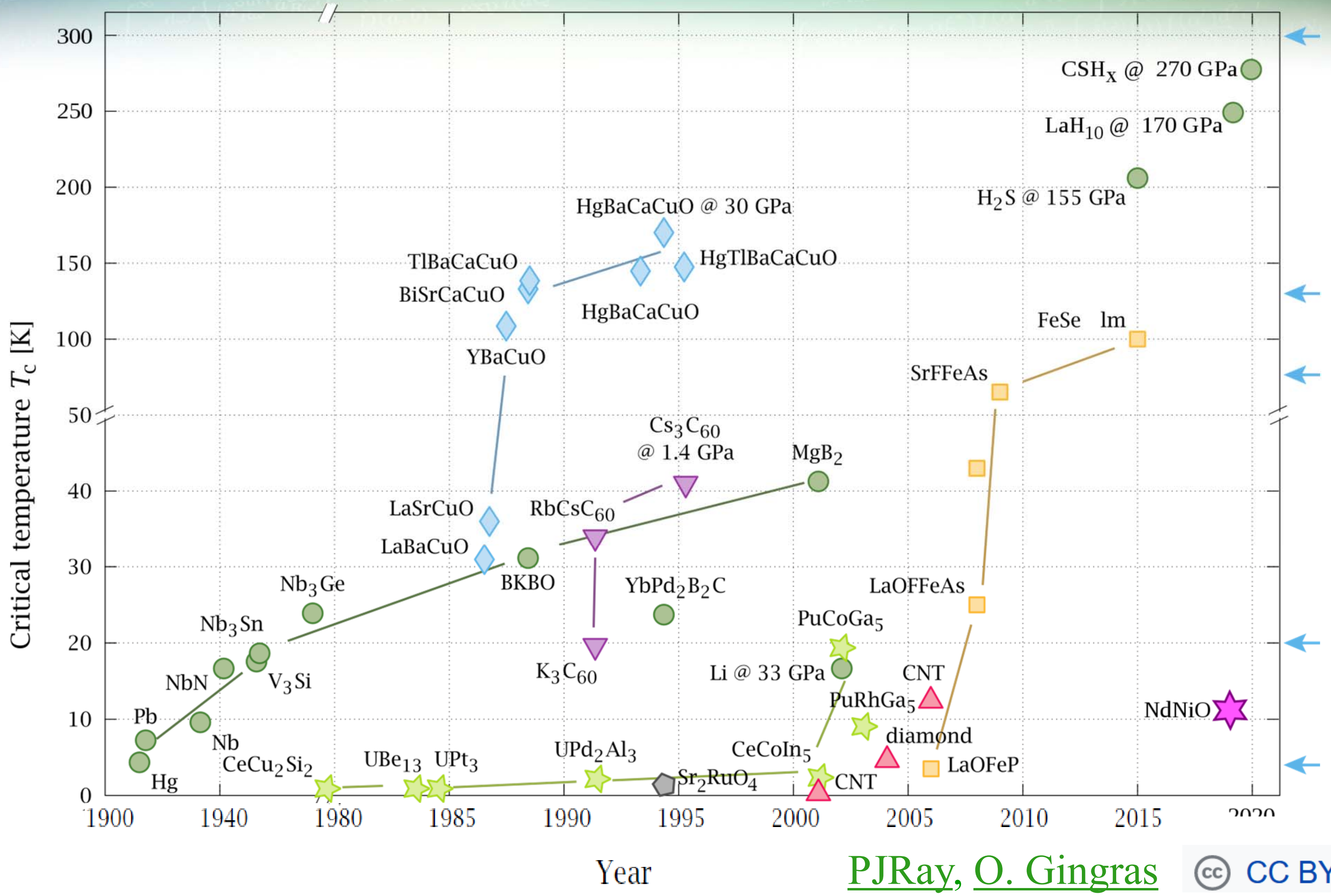
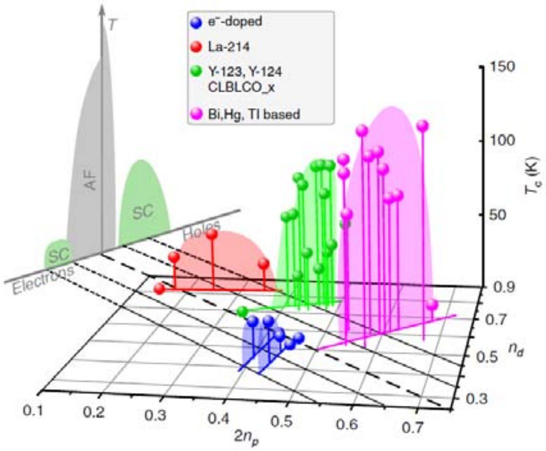
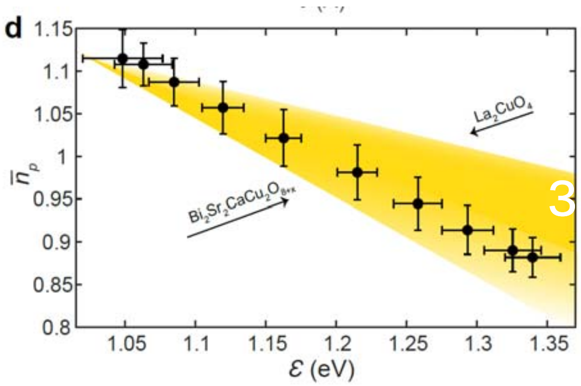


PJRay, O. Gingras

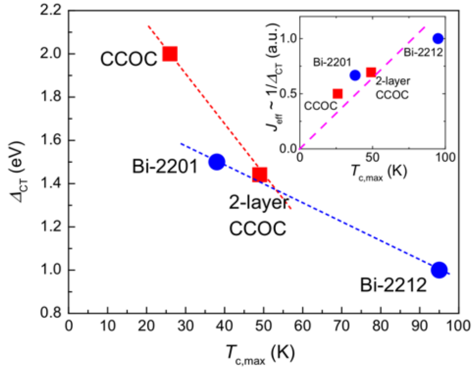
Three experimental observations on optimizing T_c



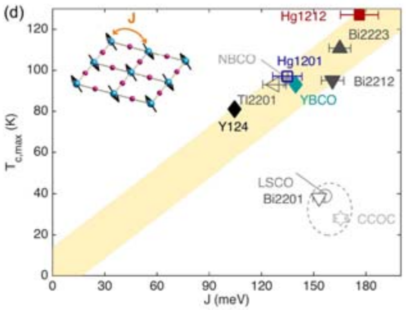
Rybicki, … Haase,
Nat. Comm. 7, 11413
(2016)

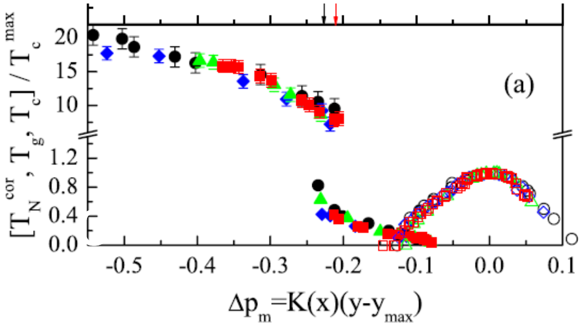


O'Mahony *et al.* arXiv:2108.03655



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



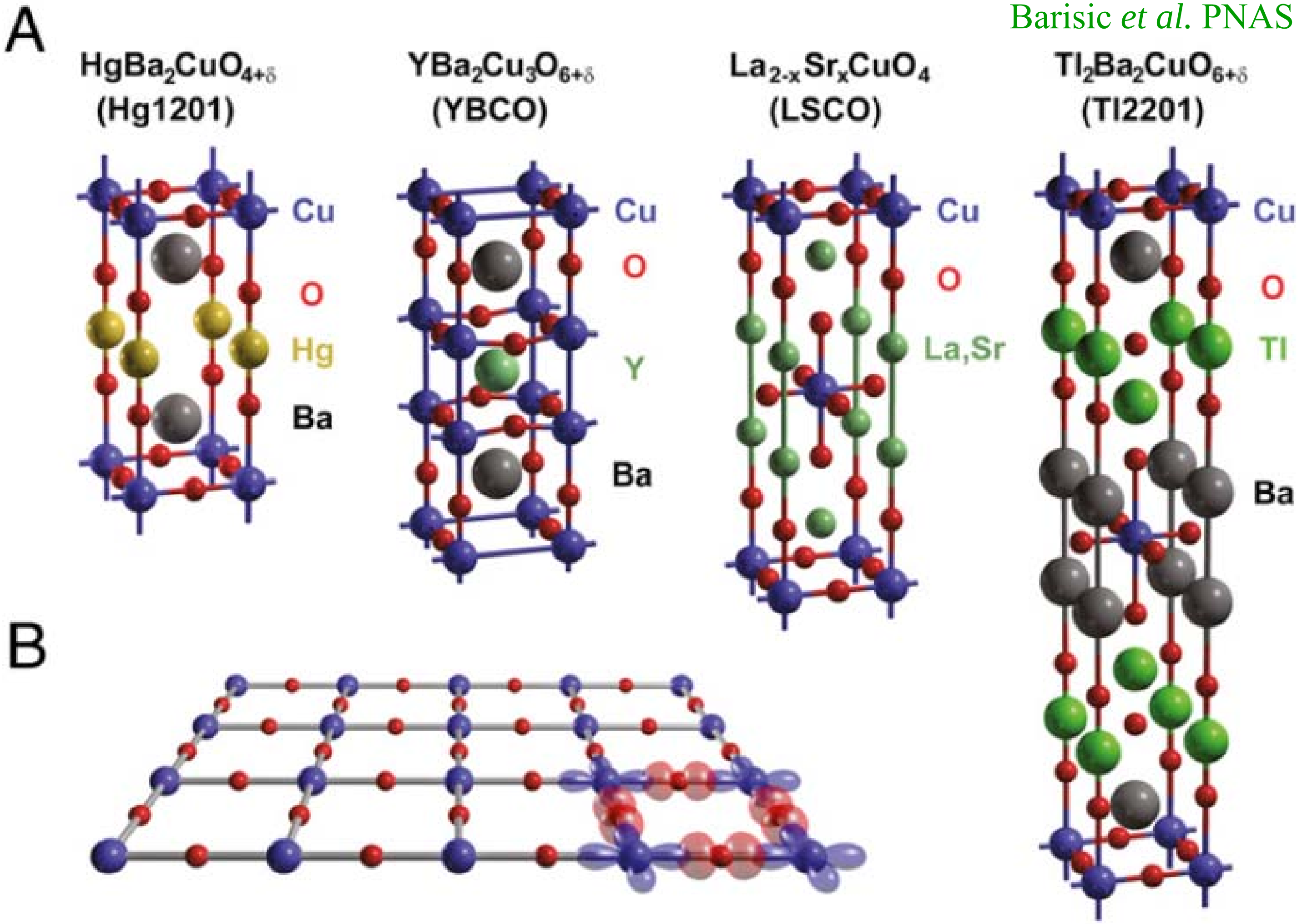


Lichen Wang,
Nat. Comm. **13**, 3163 (2022)

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

There are different kinds of cuprates : All with CuO_2 planes

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



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)

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Outline

- Method
- 3-band Model

- Three experiments that tell us how to optimize Tc.
- Pairing mechanism
- Bonus

- Conclusion

Method
Solving the models

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

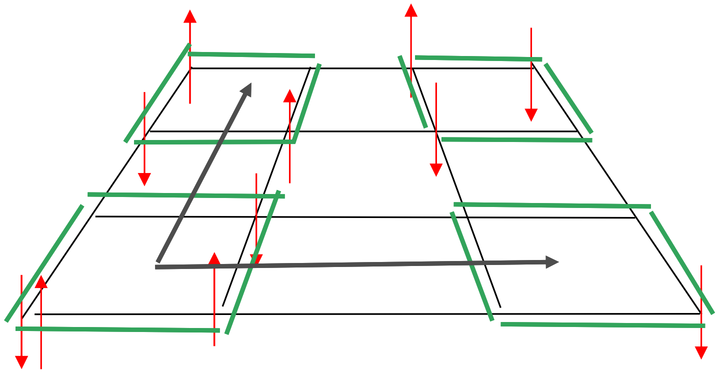
Cluster generalization of
Dynamical Mean-Field Theory : DMFT

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

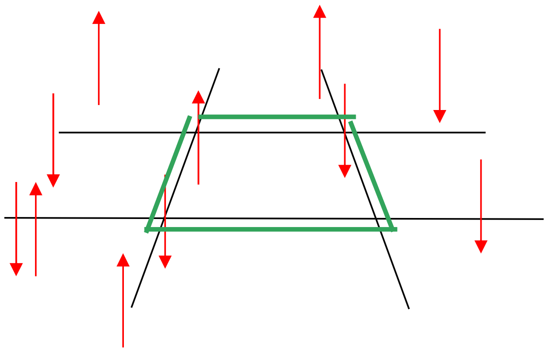
Localized and delocalized pictures C-DMFT

Delocalized



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

Localized



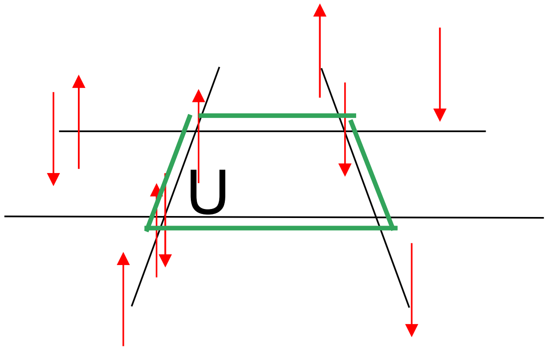
$$G_{ij} = \int \frac{d^d\tilde{k}}{(2\pi)^d} \left(\frac{1}{(i\omega_n+\mu)I-\varepsilon\left(\tilde{k}\right)-\Gamma_O(i\omega_n)-\Sigma(i\omega_n)} \right)_{ij} \quad (G^{-1})_{ij} = (G_0^{-1})_{ij} - \Sigma_{ij}$$

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 (Exact diagonalisation)



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

QCM David Sénéchal

Some groups using these methods for cuprates

- Europe:
  - Georges, Parcollet, Ferrero, Civelli, Fratino (Paris)
  - Sordi (London), 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 of the method: advantages and limitations

+ 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

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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)

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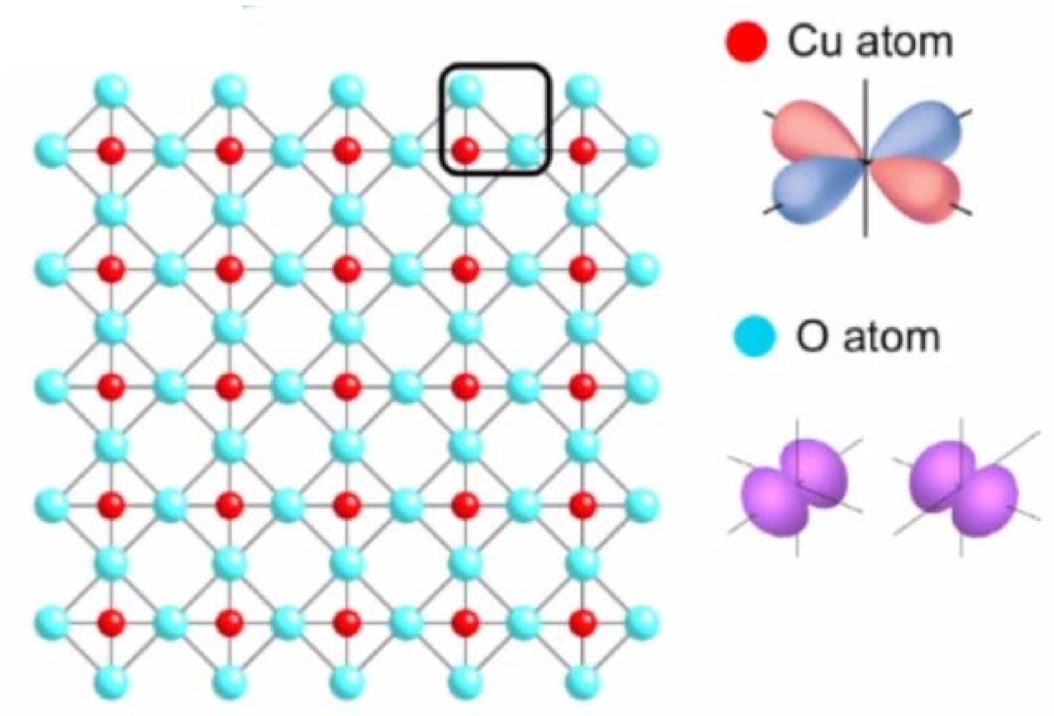
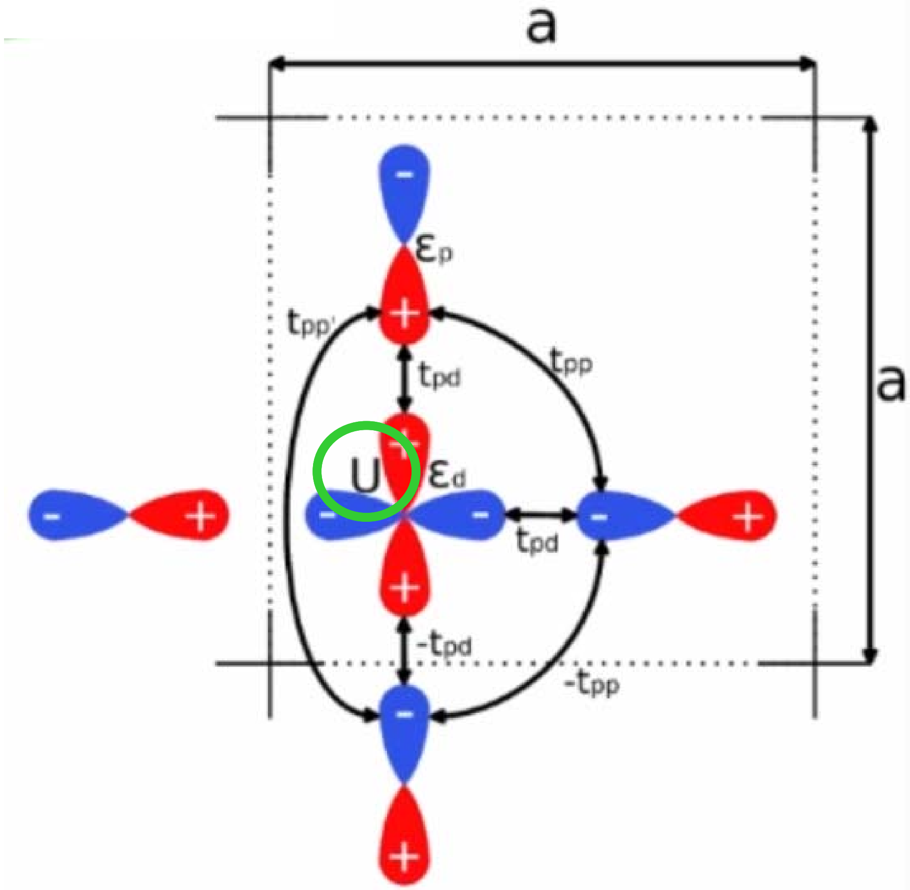
Copper and oxygen planes

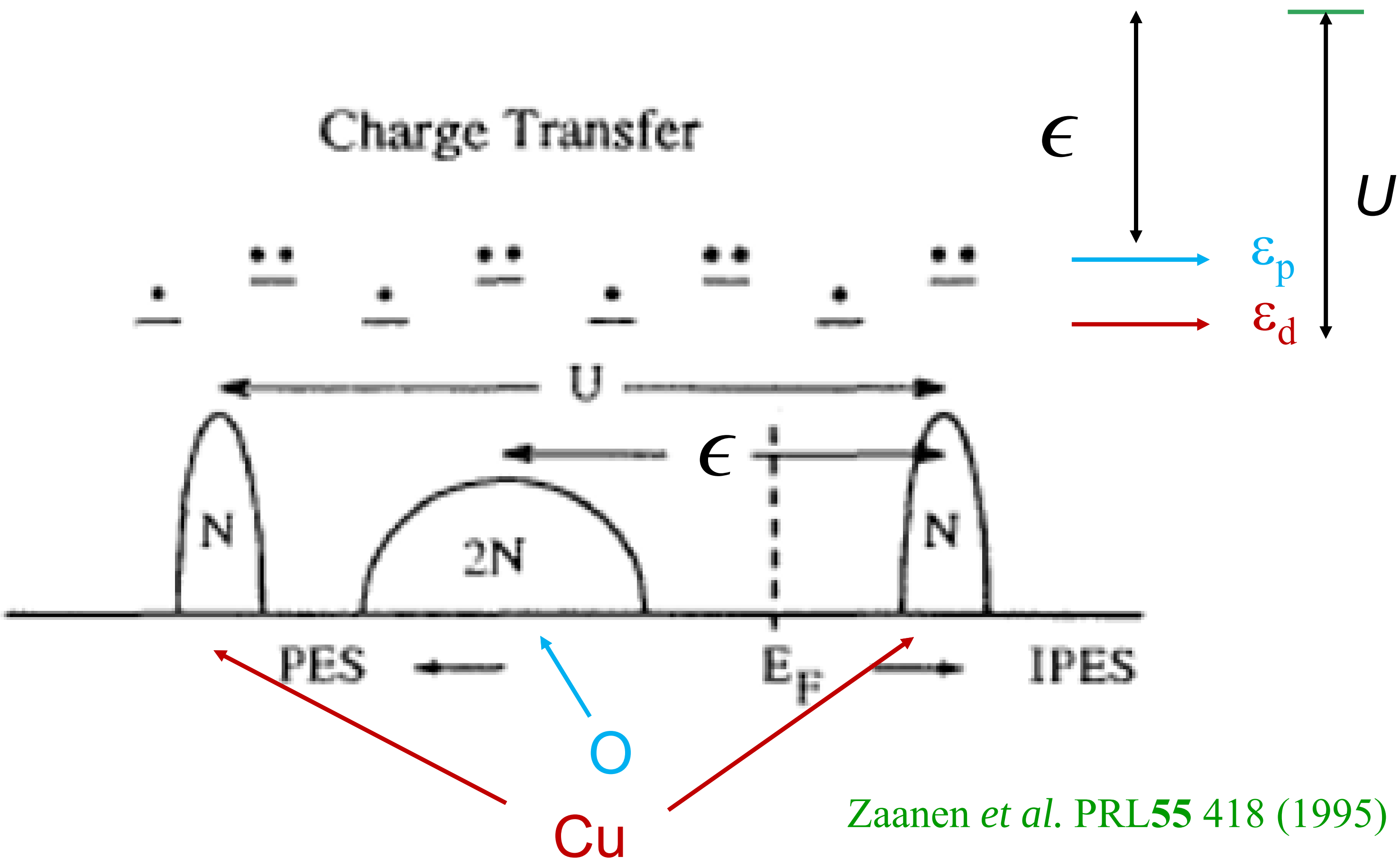


© Nicolas Kowalski



© Nicolas Kowalski

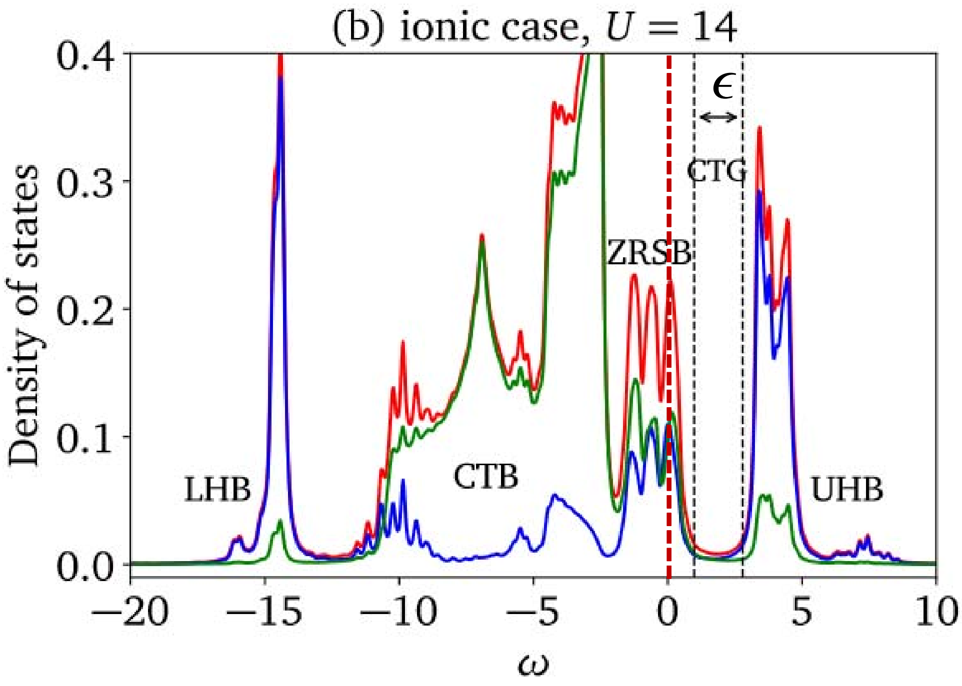
Cartoon of the charge transfer insulator

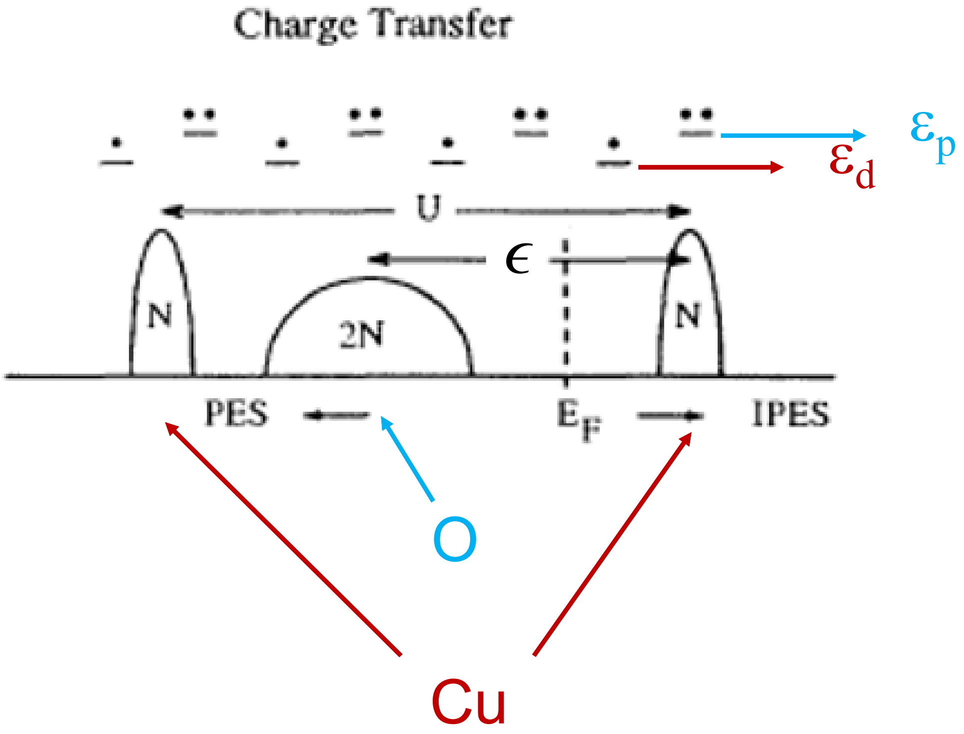


Zaanen *et al*. PRL**55** 418 (1995)

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

"Ionic" limiting case





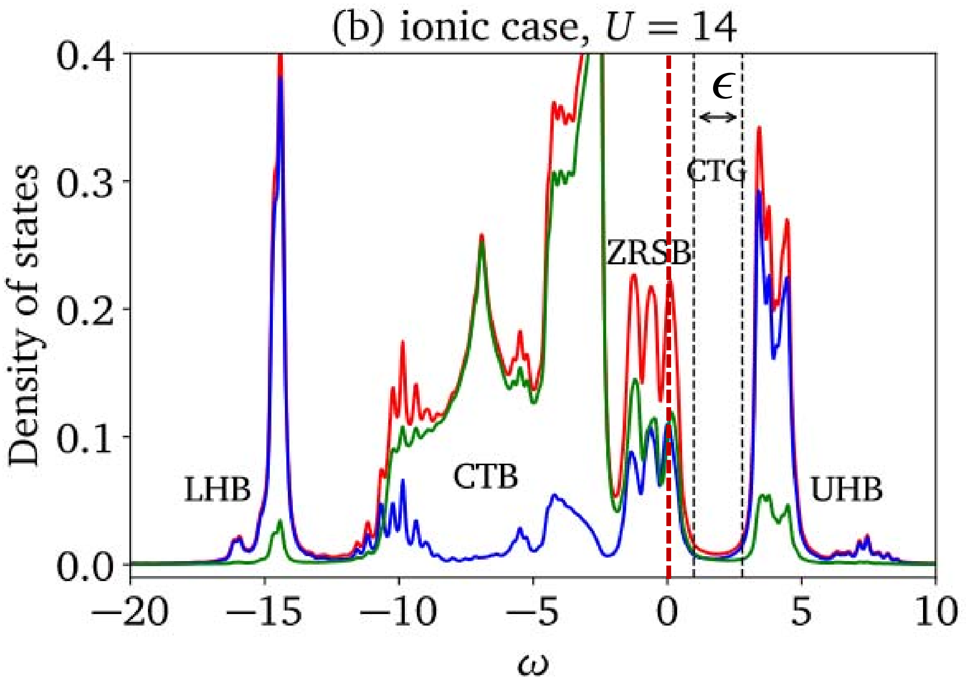
Meinders *et al*. PRB **48**, 3916 (1993)

The strategy

The strategy

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

"Ionic" limiting cases



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

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

"Realistic model"

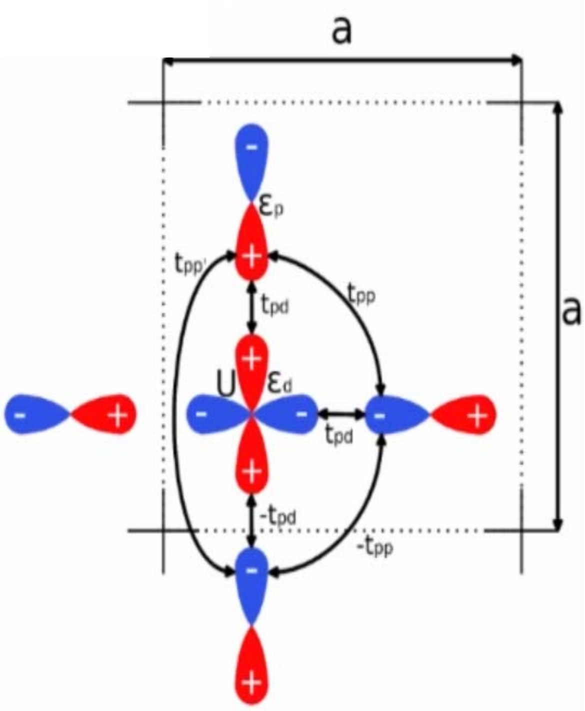
Hohenberg-Kohn : Exchange correlation
Kohn-Sham : Basis set

Density Functional Theory

Electronic structure

	Compound	$\epsilon_d - \epsilon_p$ (eV)	t_{pd} (eV)	t_{pp} (eV)	$t_{pp'}$ (eV)	t'/t	layers	$d^{\mathrm{apical}}_{\mathrm{Cu-O}}$ (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)	$Tl_2BaCaCu_2O_8$	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

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



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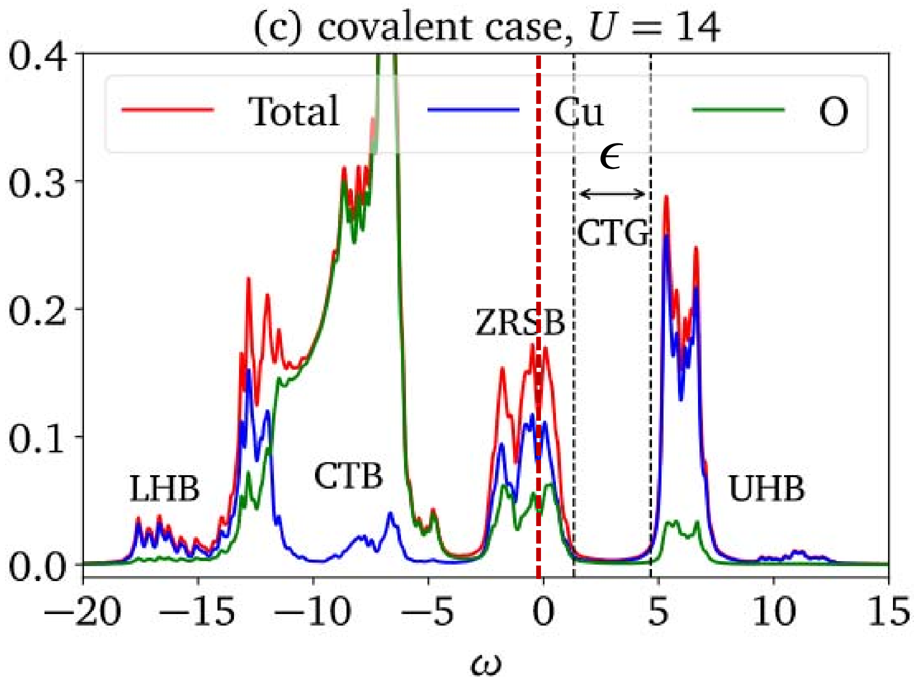
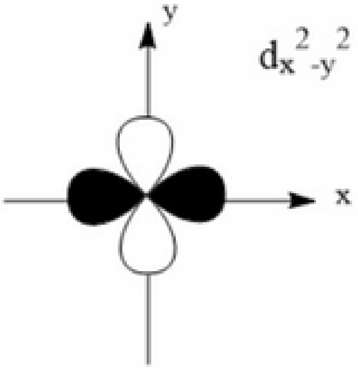
"Covalent" models (*T = 0*)

"Realistic"



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



d-wave Superconductivity

T = 0 exact diagonalization solver : order parameter

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

Reduced wave vector

$$\langle\hat{\Delta}\rangle = \oint \frac{d\omega}{2\pi}\frac{d^2\tilde{\mathbf{k}}}{(2\pi)^2}\, tr\Big[\boldsymbol{\Delta}(\tilde{\mathbf{k}})\mathbf{G}(\tilde{\mathbf{k}},\omega)\Big]$$

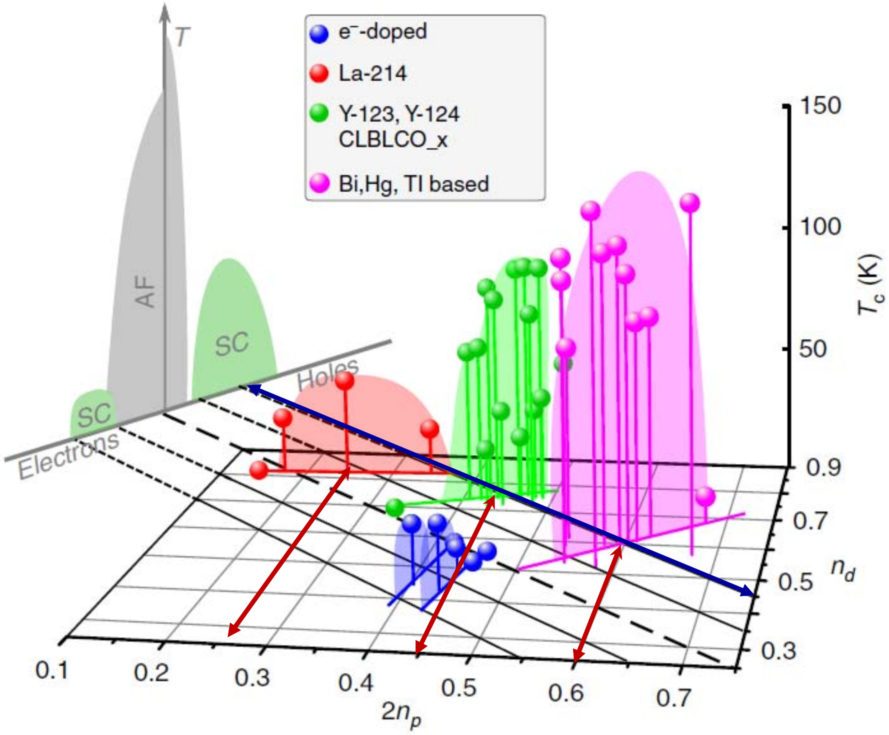
Average per site

Green function from CDMFT

#1 Optimizing T_c with oxygen hole content

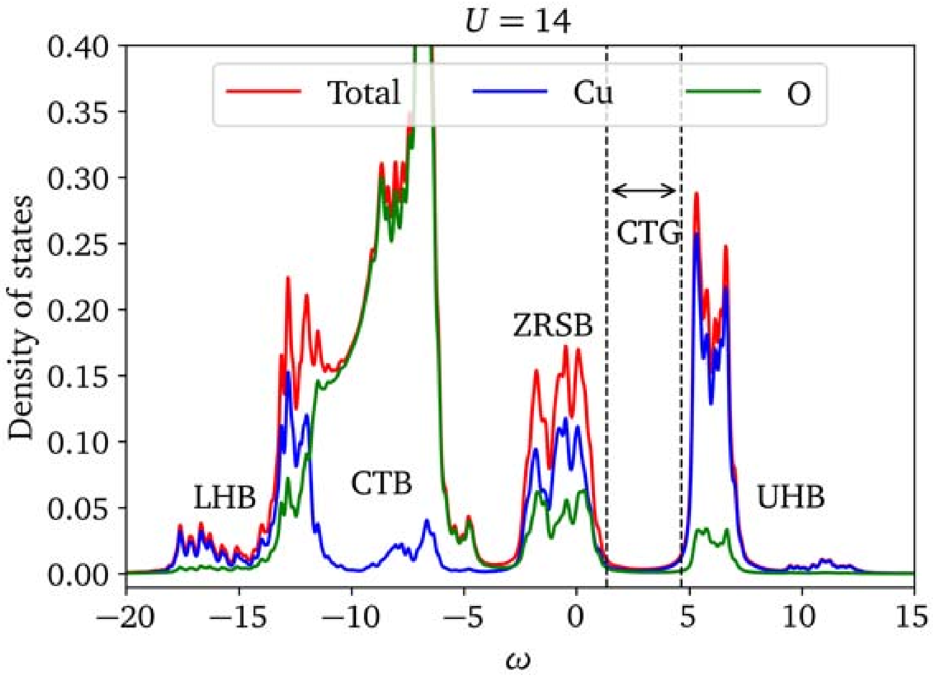
#1 Optimizing T_c with oxygen hole content

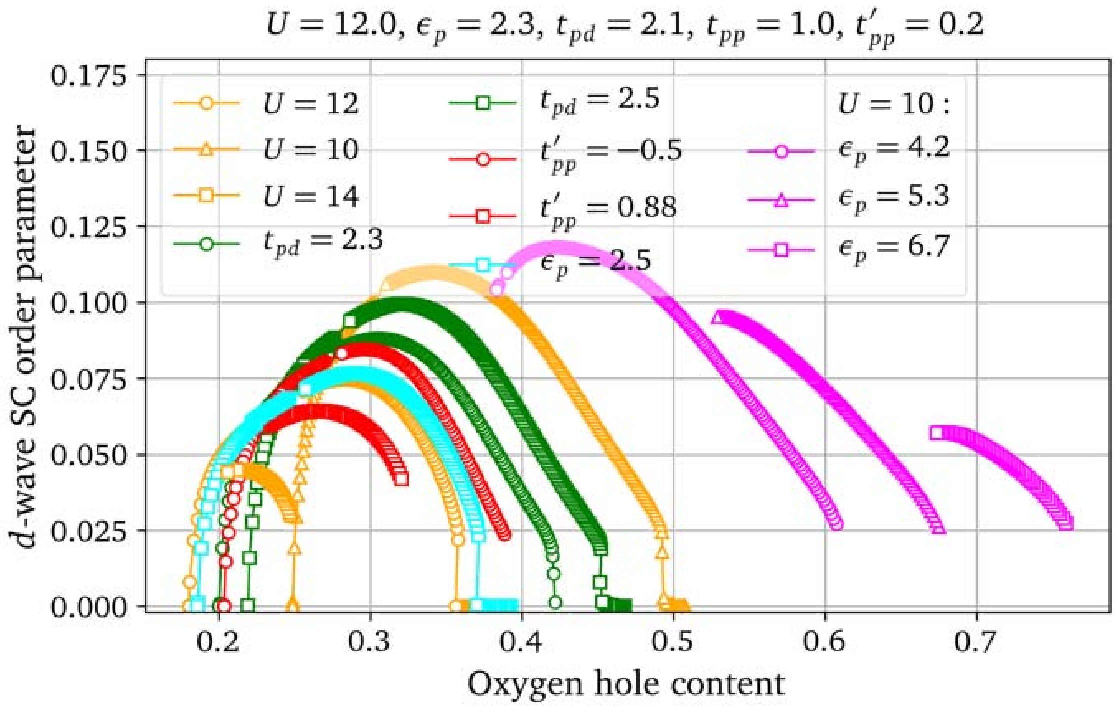
$$n = n_d + 2n_p$$



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

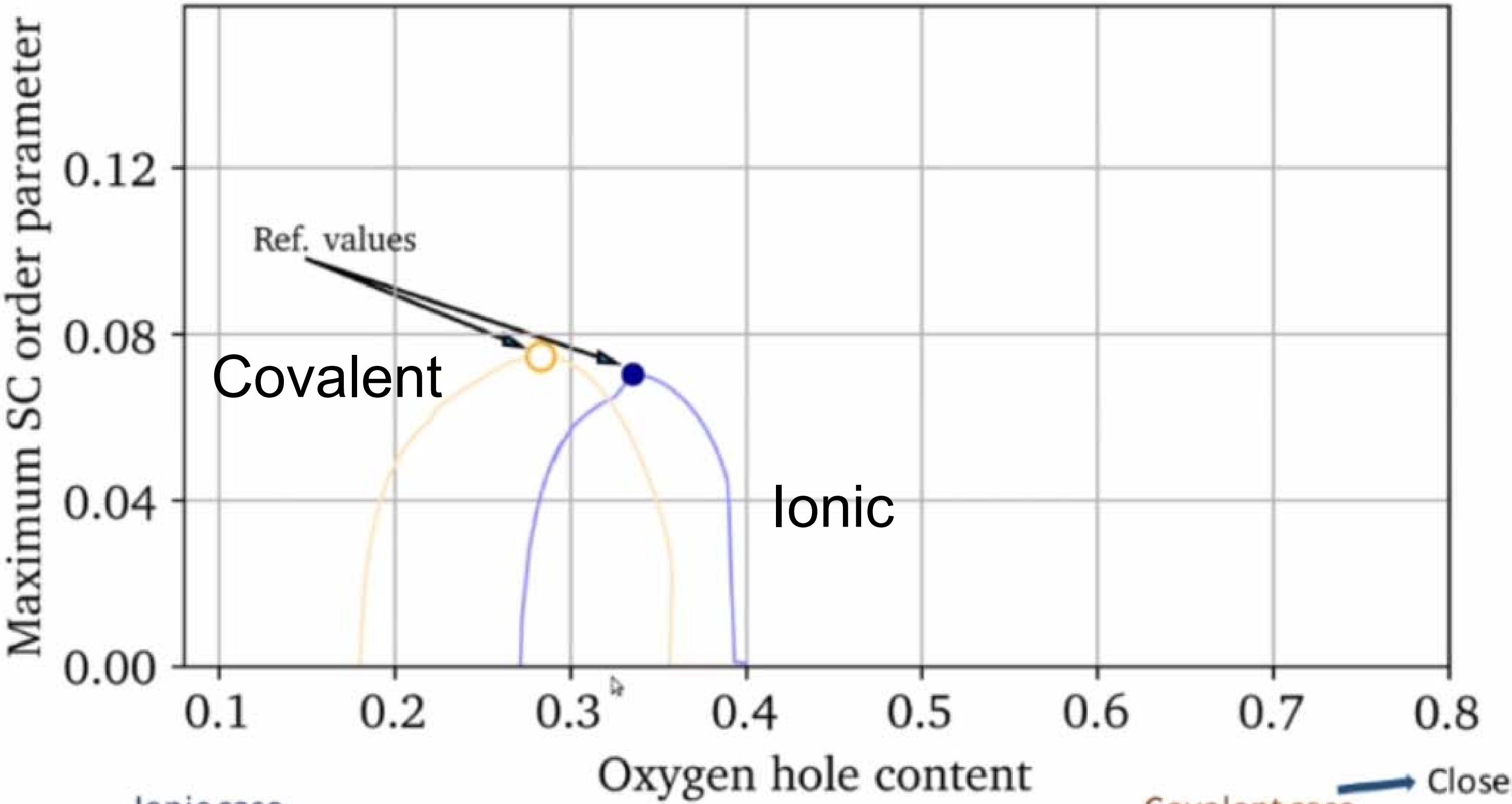
T = *0* superconducting domes for the covalent models





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

T = *0* superconducting domes for the reference models



→ Closer to cuprates

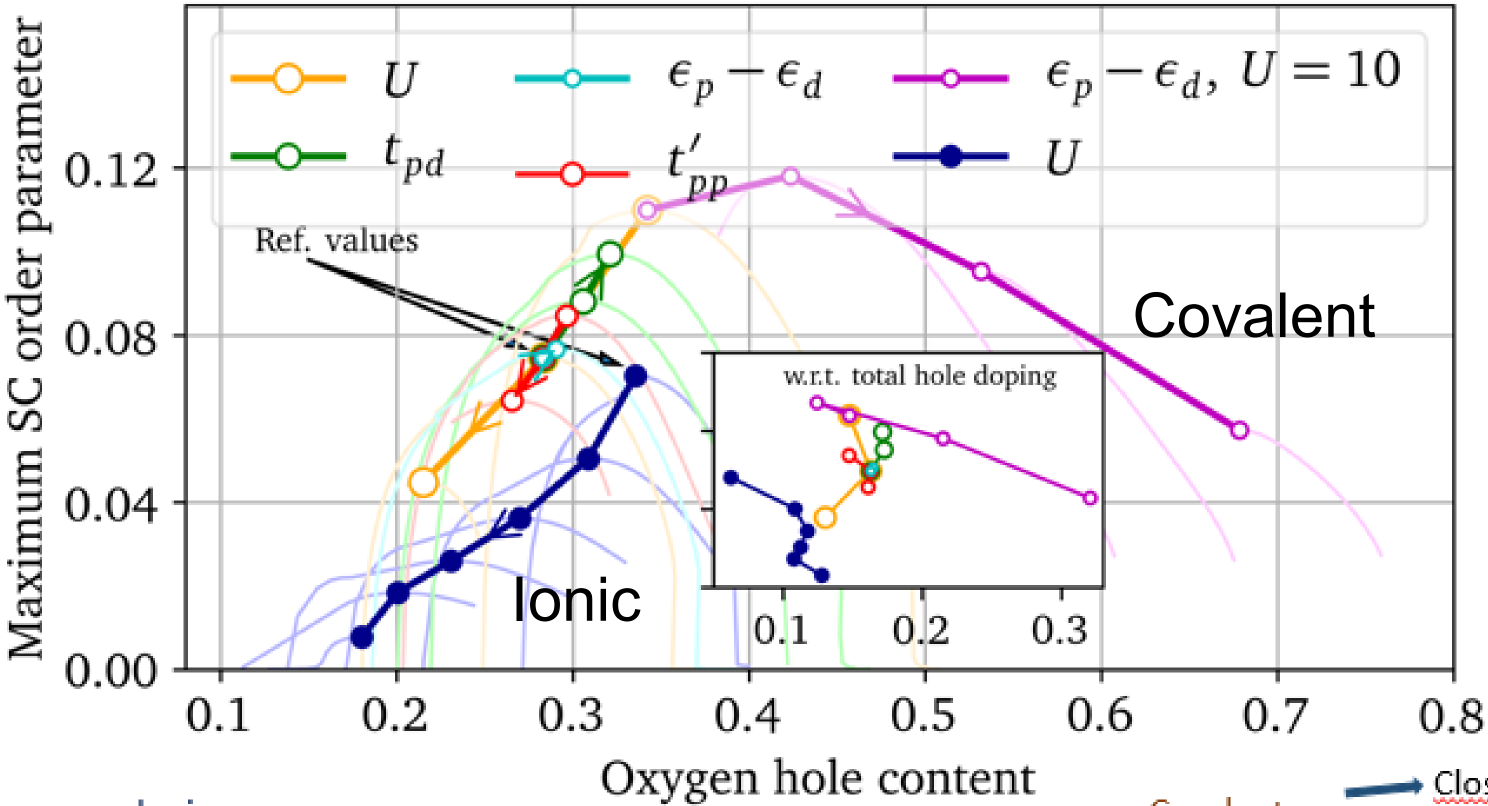
Ionic case

Covalent case

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

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

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



Closer to cuprates

Ionic case

Covalent case

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

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

#2 Optimizing T_c with Charge Transfer gap ϵ

(Oxygen as a witness)

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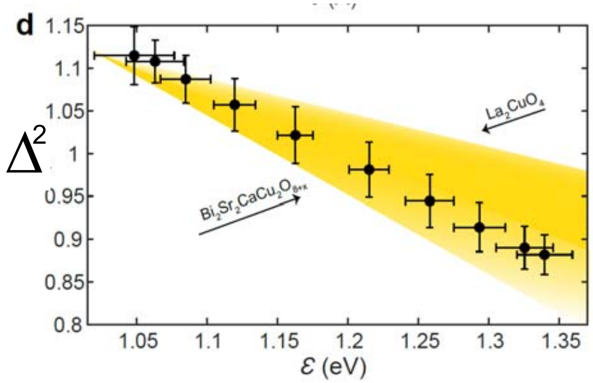
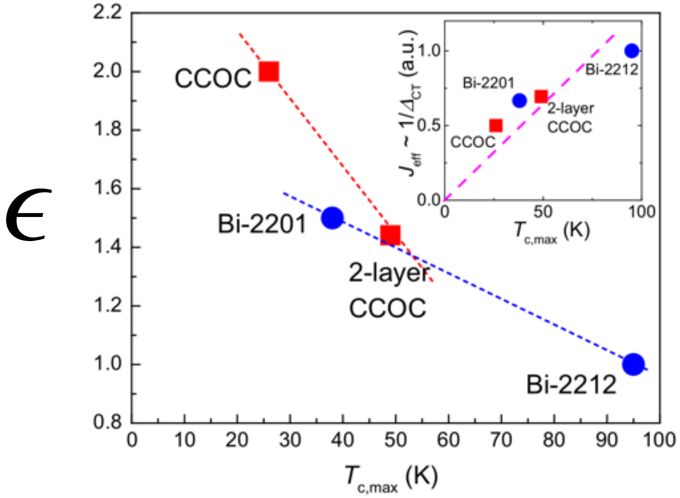
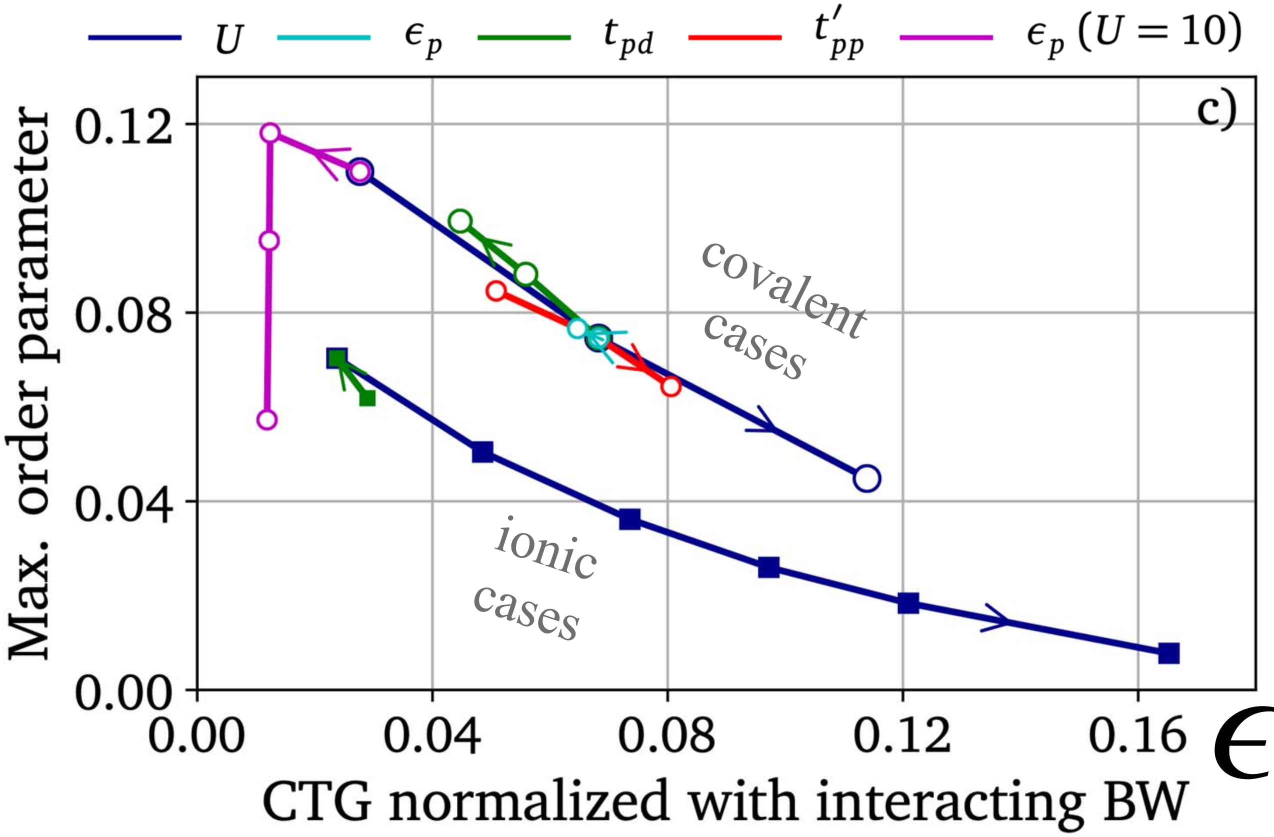
#2 Optimizing T_c with CT gap Δ (Oxygen as a witness)



O'Mahony *et al*. arXiv:2108.03655

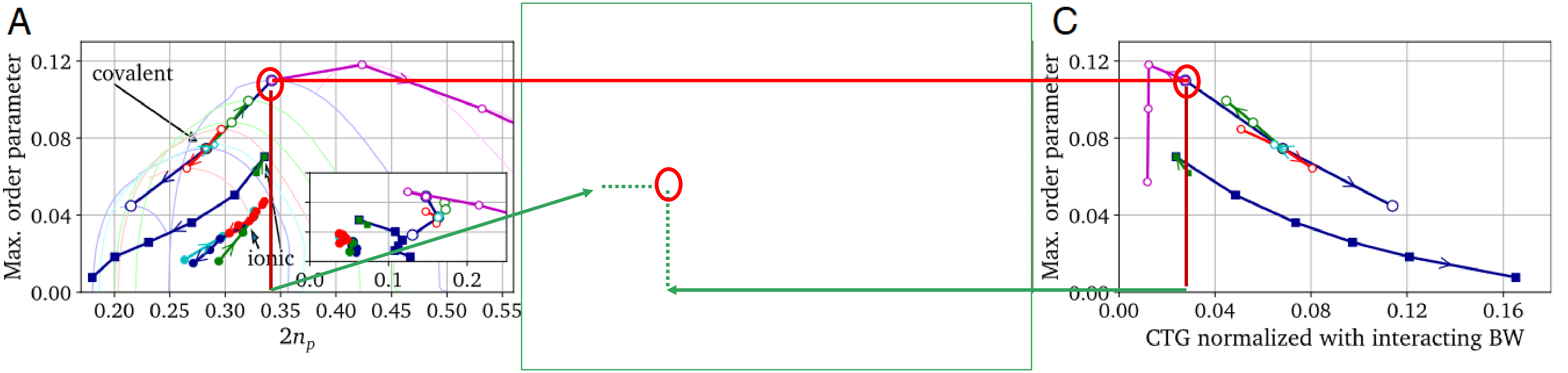


Ruan *et al*. Sci. Bull. **61** (2016)

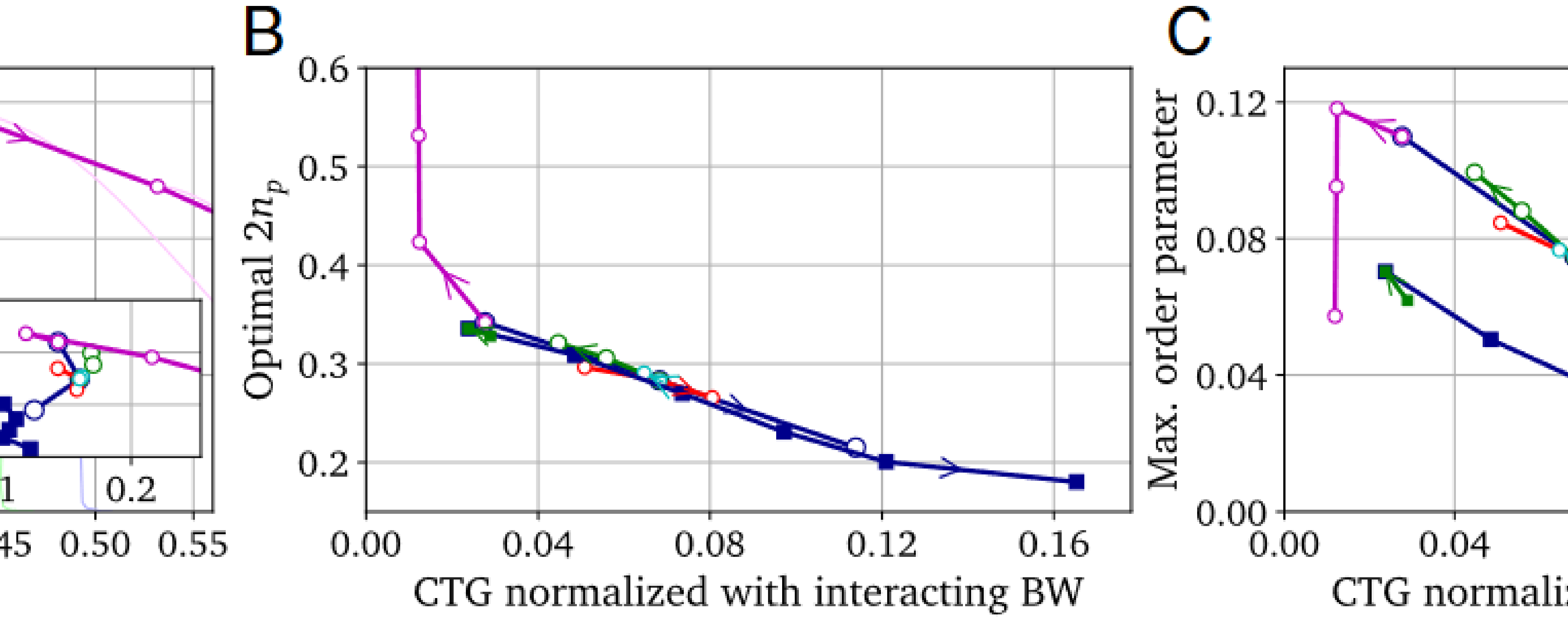


Kowalski, Dash, Sémon, Sénéchal, A-M.T.
PNAS **118** (40) e2106476118 (2021)

Oxygen hole content OR charge transfer gap?



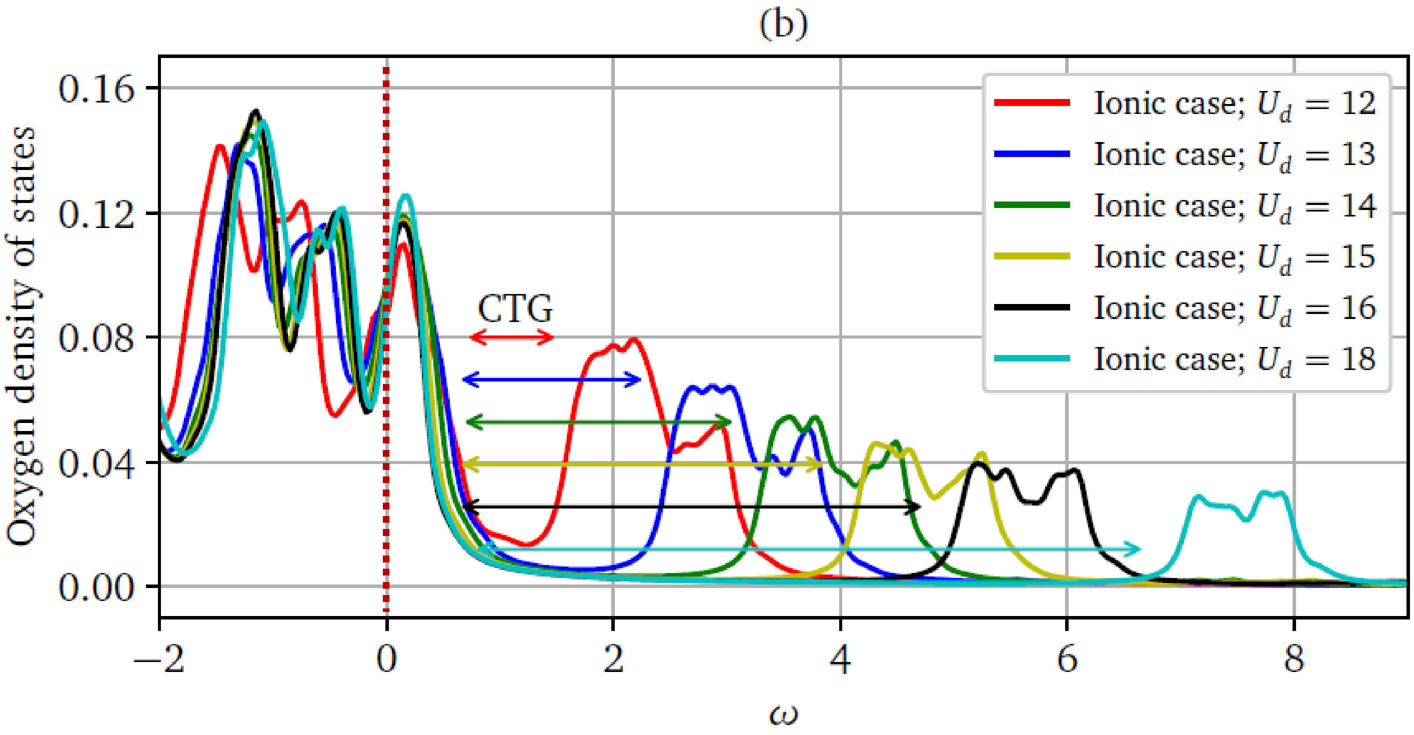
Charge-transfer gap, oxygen hole content



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

Charge transfer gap and oxygen hole content :

Oxygen as a witness



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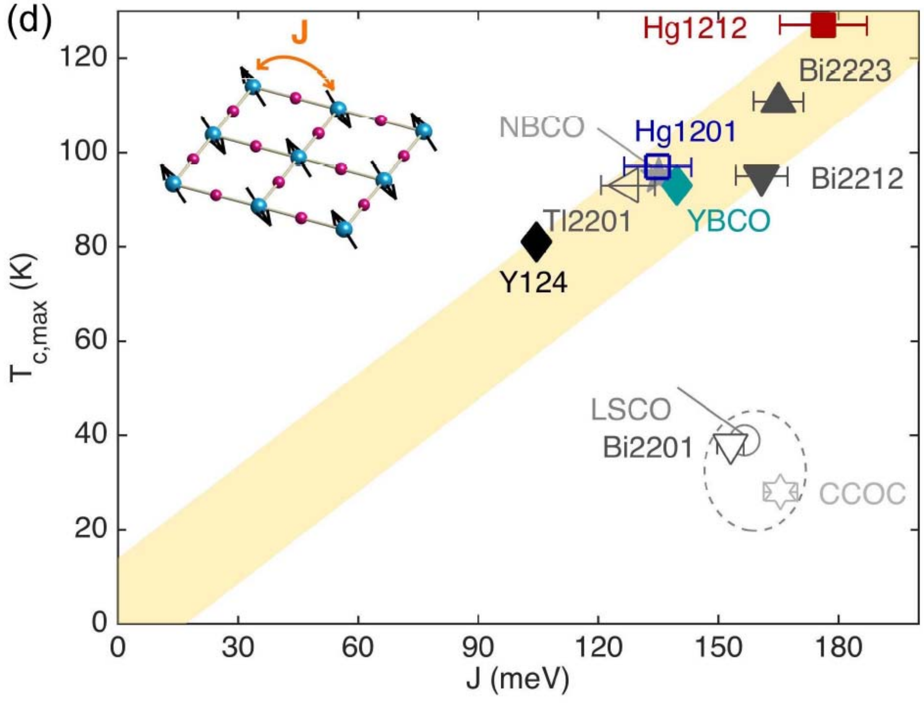
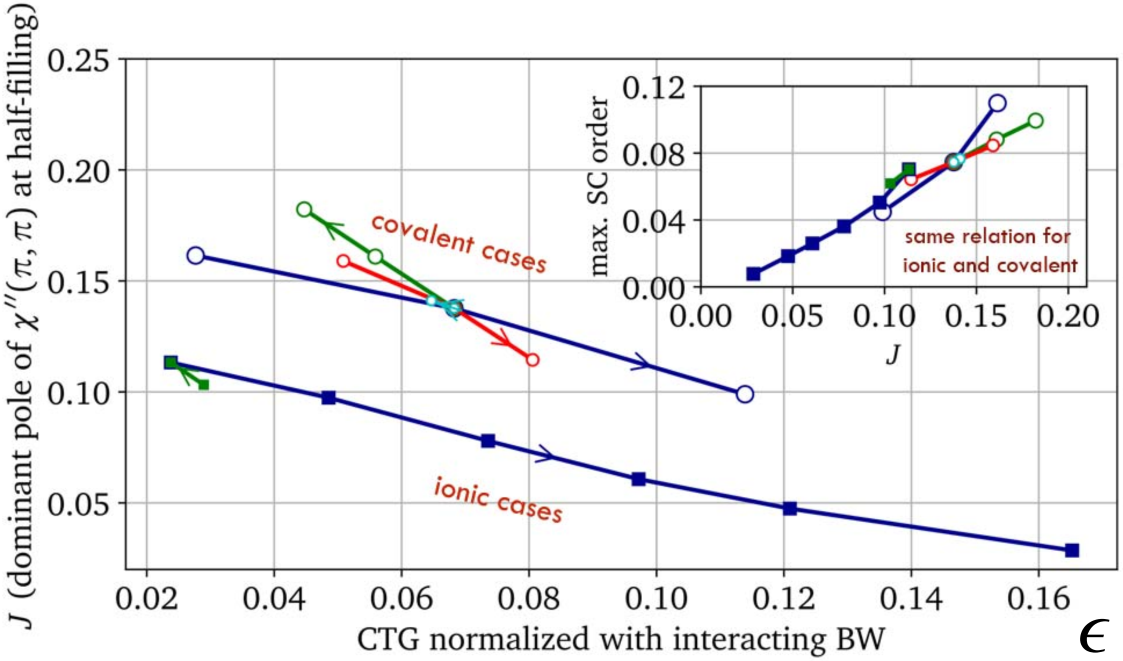
#3 Optimizing T_c with superexchange

#3 Optimizing T_c with superexchange

E. Müller-Hartmann *et al*. Eur. Phys. J. B **28**, 173 (2002)

$$J = \frac{4t_{pd}^4(U+\epsilon)}{U\epsilon^3} \xrightarrow[U\to\infty]{} \frac{4t_{pd}^4}{\epsilon^3}$$



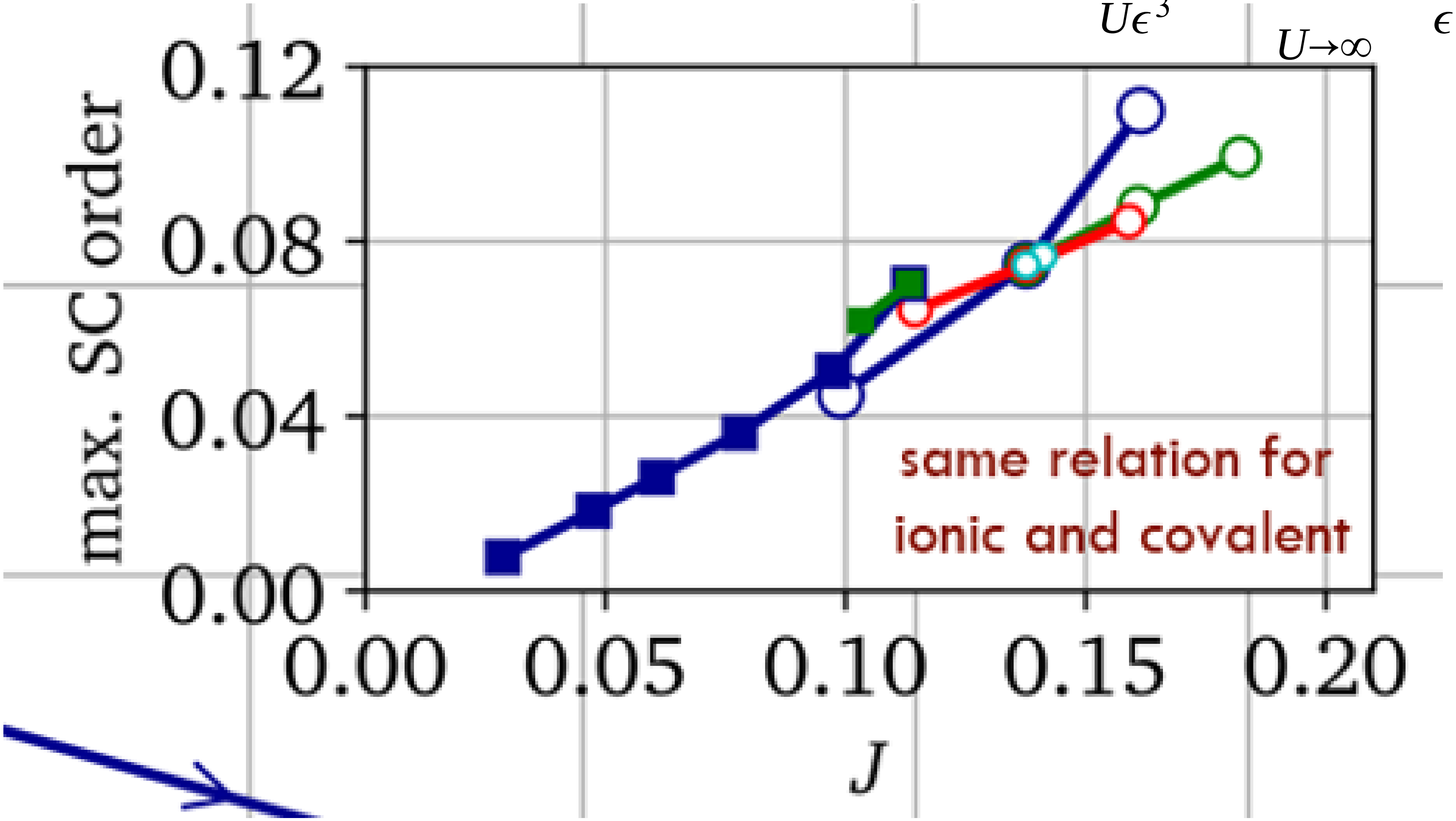


Lichen Wang et al.
Nat. Comm. **13**, 3163 (2022)

Super exchange

E. Müller-Hartmann *et al.* Eur. Phys. J. B **28**, 173 (2002)

$$J = \frac{4t_{pd}^4(U+\epsilon)}{U\epsilon^3} \xrightarrow[U\to\infty]{} \frac{4t_{pd}^4}{\epsilon^3}$$



same relation for
ionic and covalent

- Some work on d-wave in one-band
  - G. Kotliar, J. Liu, Superconducting instabilities in the large-U limit of a generalized Hubbard model. Phys. Rev. Lett. 61, 1784–1787 (1988).
  - N. E. Bickers, D. J. Scalapino, S. R. White, Conserving approximations for strongly correlated electron systems: Bethe-Salpeter equation for the two dimensional Hubbard model. Phys. Rev. Lett. 62, 961–964 (1989)
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- Critique
  - M. Qin et al., Absence of superconductivity in the pure two-dimensional Hubbard model. Phys. Rev. X 10, 031016 (2020)
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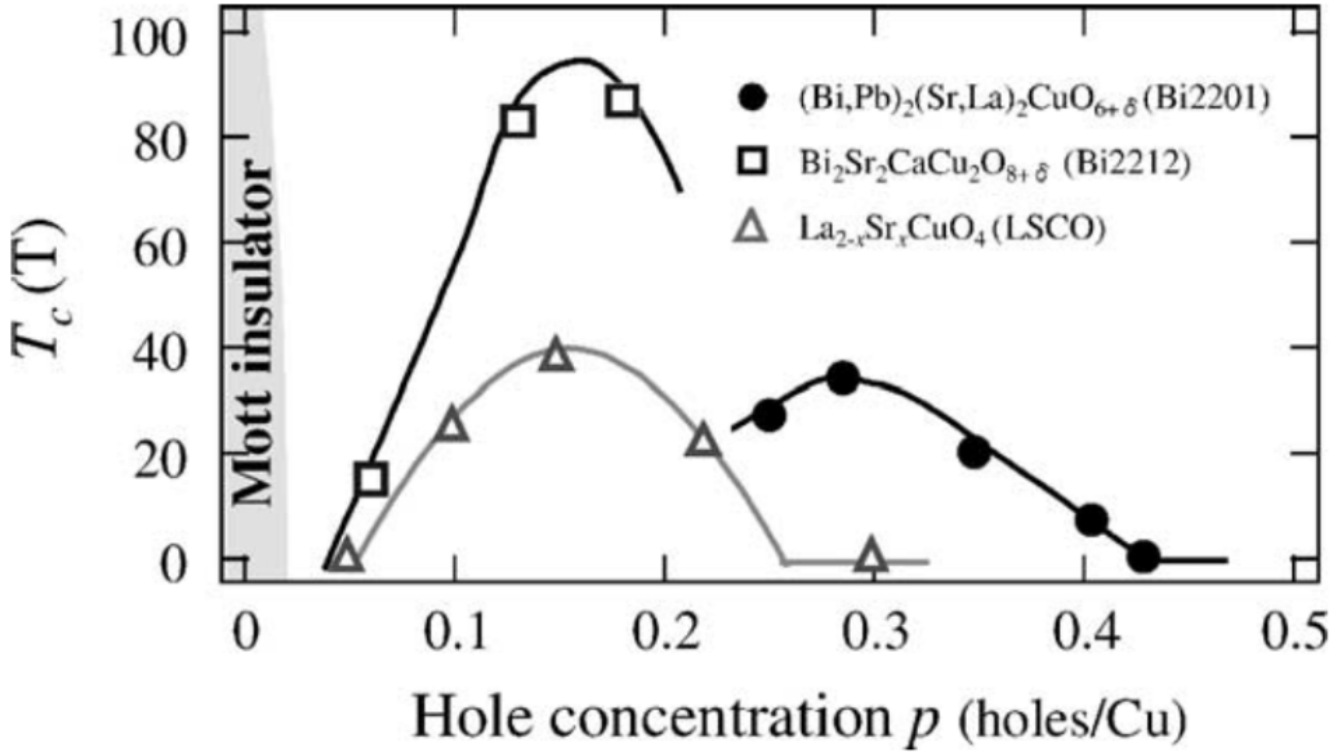
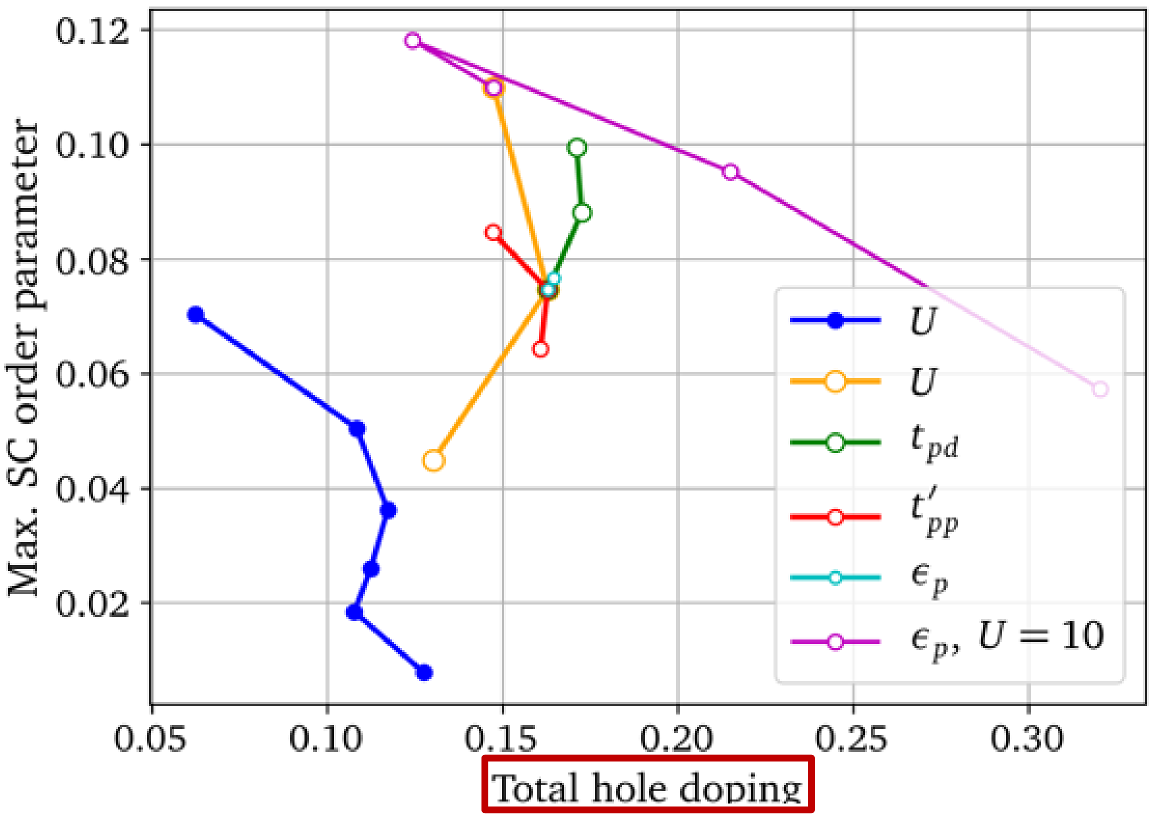
Bonus

T_c and total hole concentration are not well correlated



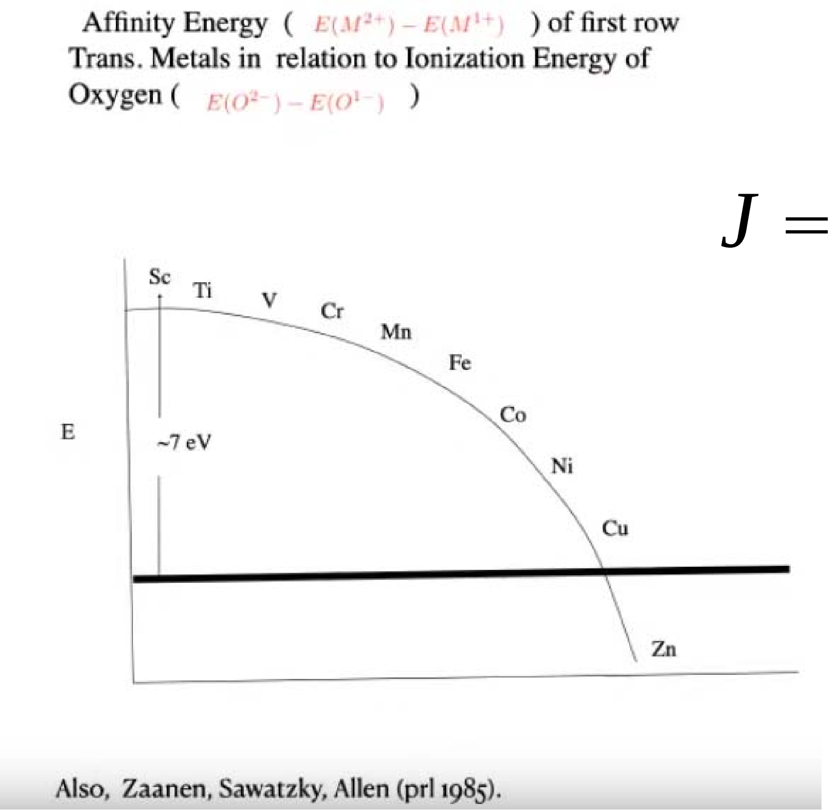
T. Kondo *et al.*
Journal of Electron Spectroscopy and Related Phenomena **137-140**, 663 (2004)

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 ($E(O^{2-}) - E(O^{1-})$)

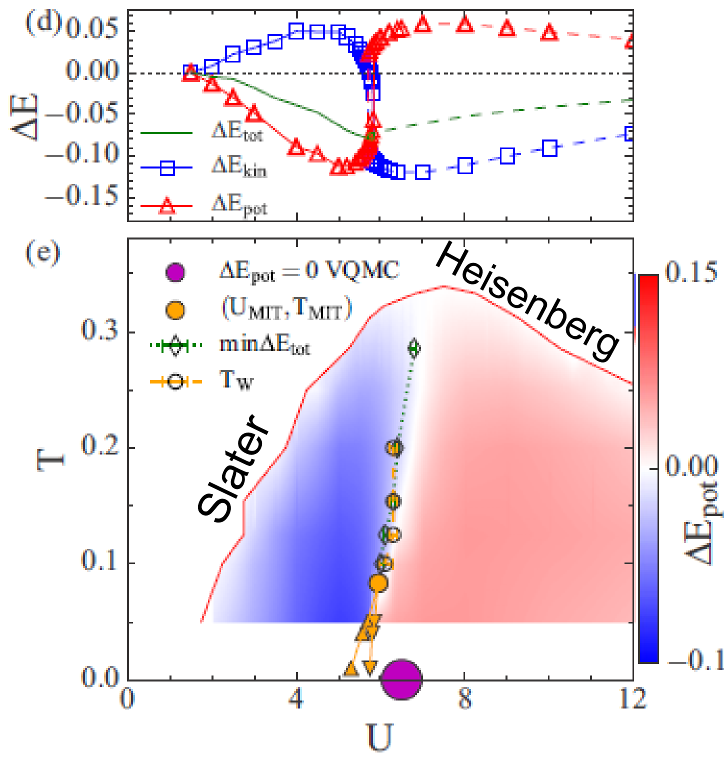
Also, Zaanen, Sawatzky, Allen (prl 1985).

$$J = \frac{4t_{pd}^4(U+\epsilon)}{U\epsilon^3} \xrightarrow[U\to\infty]{} \frac{4t_{pd}^4}{\epsilon^3}$$

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

Summary Conclusion

Pairing at small and large *U* : An analogy



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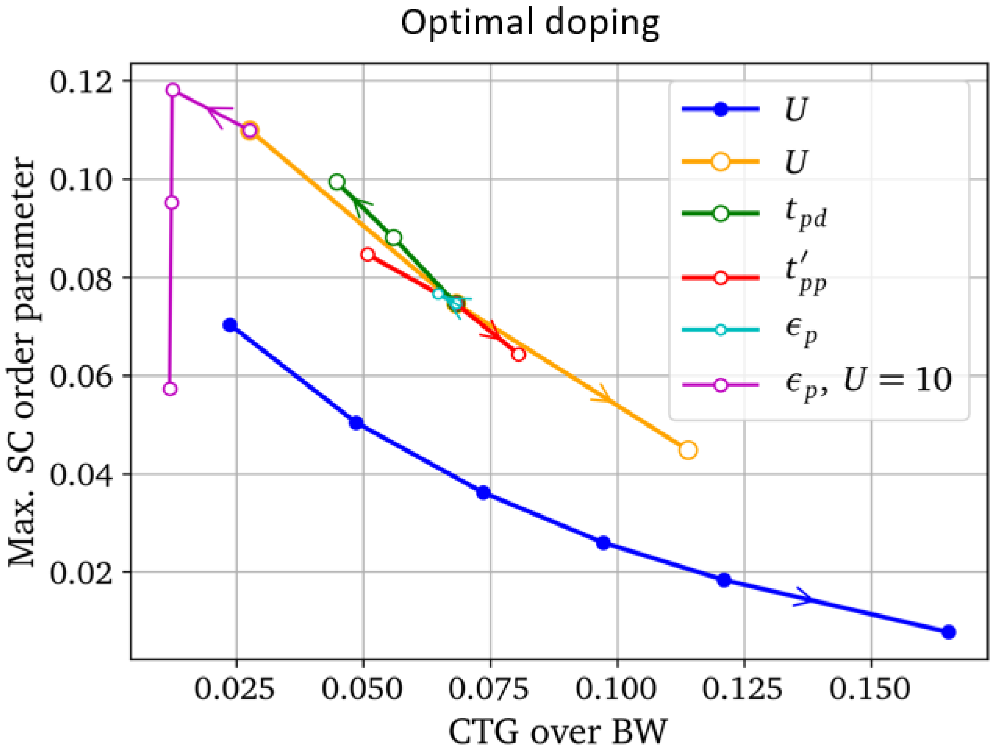

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Weber et al Europhys. Lett. 100, 37001 (2012)
Yee et al Phys. Rev. B 89, 094517 (2014)
Acharya et al Phys. Rev. X 8, 021038 (2018)

Optimizing Tc

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

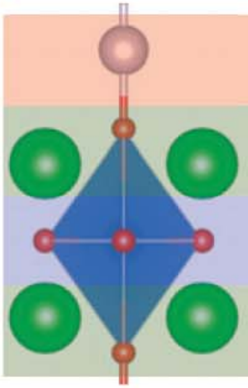
C. Weber, PNAS 2021 Vol. **118** No. 46 e2115874118

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_δ	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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