NGSCES 2015
15/09/2015

Fully *ab initio* calculation of transition temperature for alkali-doped fullerene superconductors

Yusuke Nomura

Univ. of Tokyo (Japan) → École polytechnique
(From April 2015)

In collaboration with
Shiro Sakai, Massimo Capone, and Ryotaro Arita

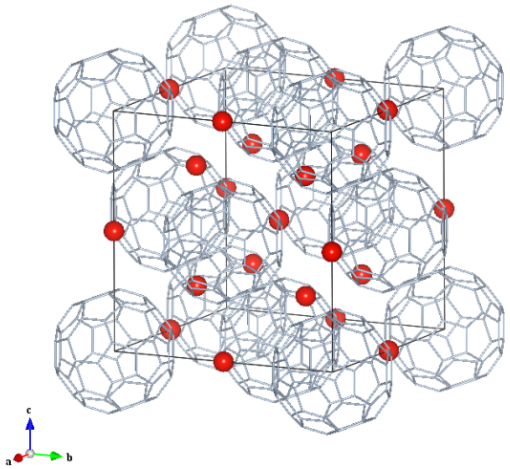
SciAdv AAAS

Ref: YN *et al*., Science Advances 1, e1500568 (2015)

Science Advances: new journal in Science family (established in 2015)

Alkali-doped fullerides

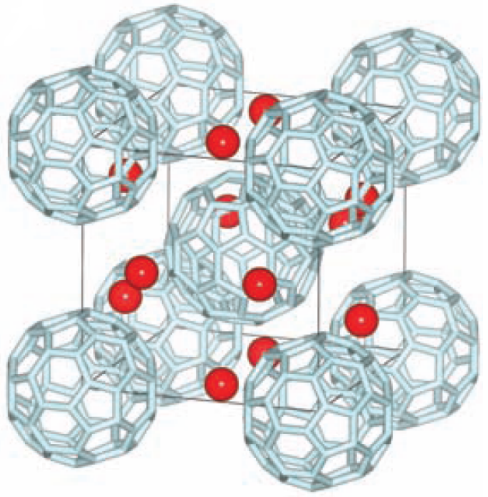
- fcc A_3C_{60} (A=K, Rb, Cs)



K_3C_{60} : T_c = 19 K
Rb_3C_{60} : T_c = 29 K
Cs_3C_{60} : T_c = 35 K

O.Gunnarsson Rev.Mod.Phys. 69, 575 (1997)
Ganin et al, Nature 466,221(2010)

- A15 Cs_3C_{60}



T_c = 38 K

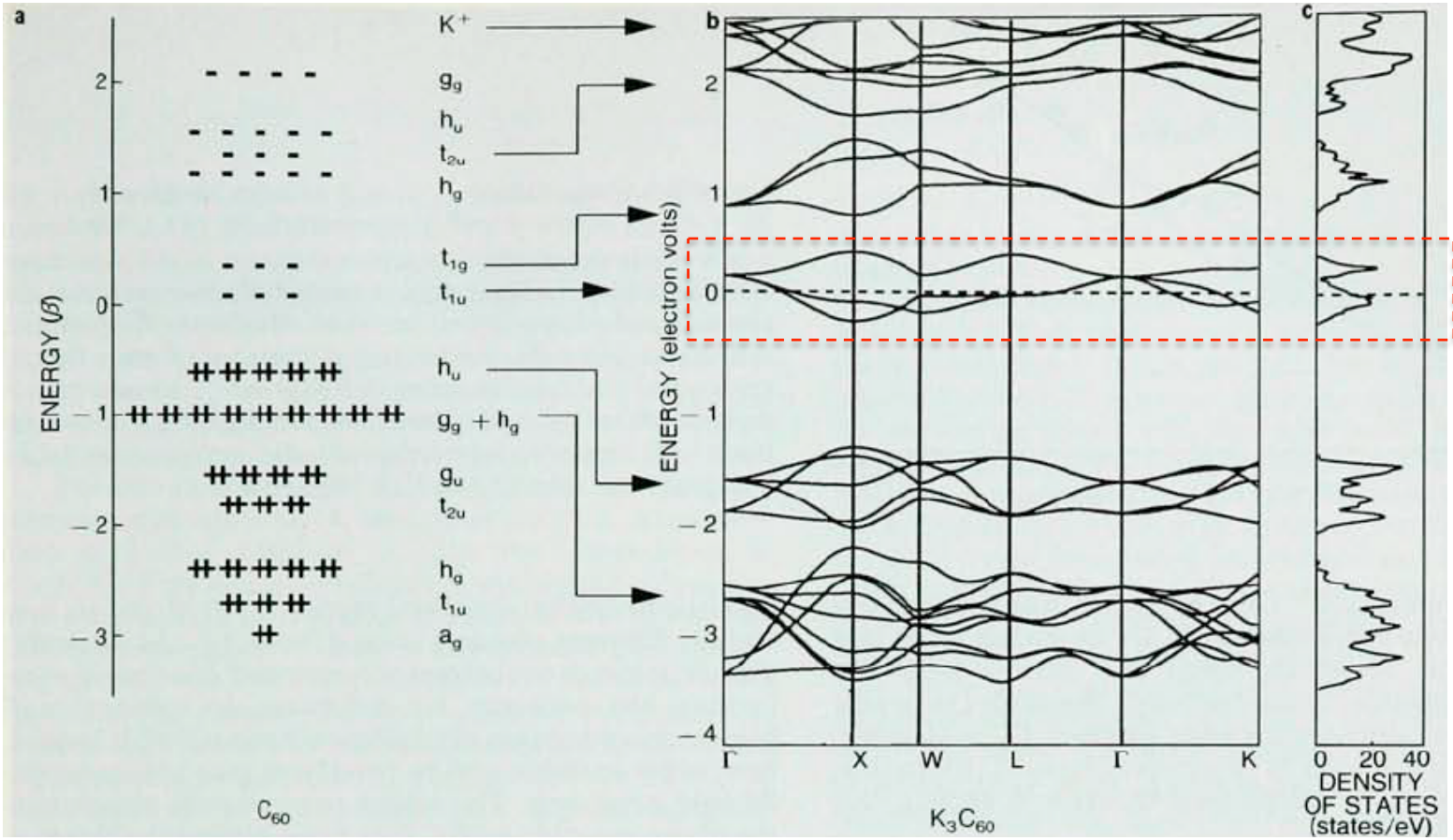
A.Ganin et al Nature Mater. 7,367-371(2008)
Y.Takabayashi et al Science 323,1285-1590(2009)

✓ Highest T_c among molecular superconductors

Electronic structure

S. C. Erwin, W. E. Pickett, Science 254, 842 (1991); A. F. Hebard, Physics Today 45, 26 (1992)

Band structure (fcc system)

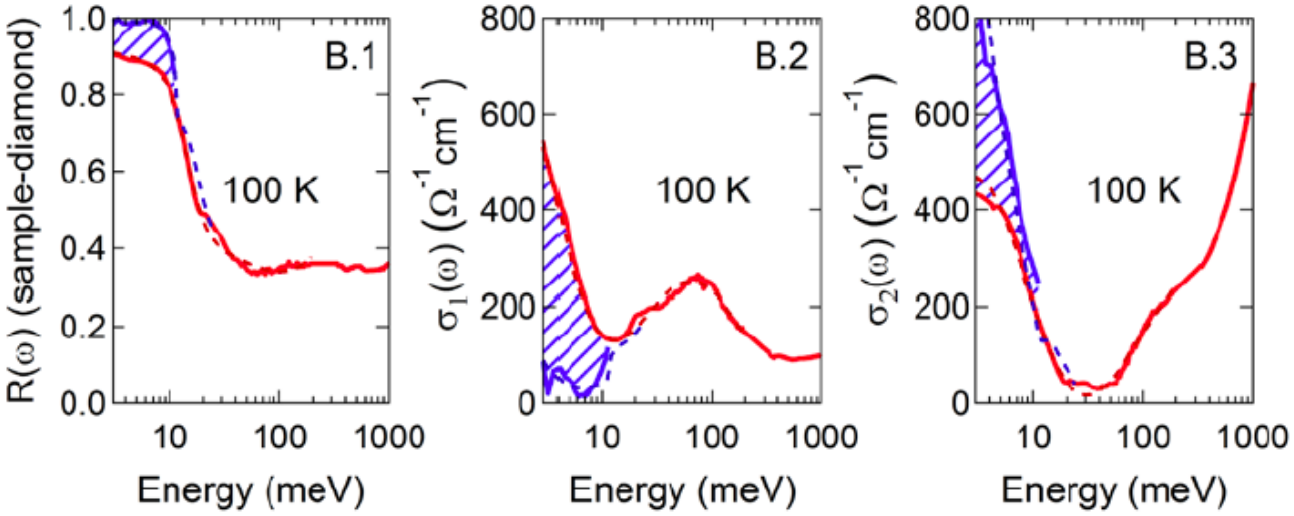


LUMO band
$W \sim 0.5$eV

- ✓ 3 orbital, half-filled
- ✓ Molecular orbital + small hopping between them

Light-induced superconducting-like phase in K_3C_{60}

M. Mitrano et al., arXiv: 1505.04529



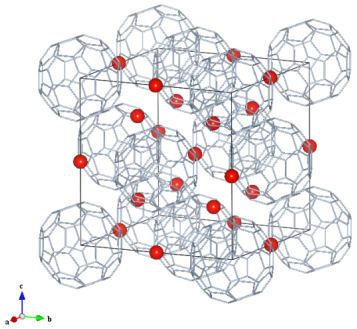
- ✓ Talk by Alice Cantaluppi (this morning)
- ✓ Study for nonequilibrium SC by Michael Sentef (tomorrow)

I focus on equilibrium properties !

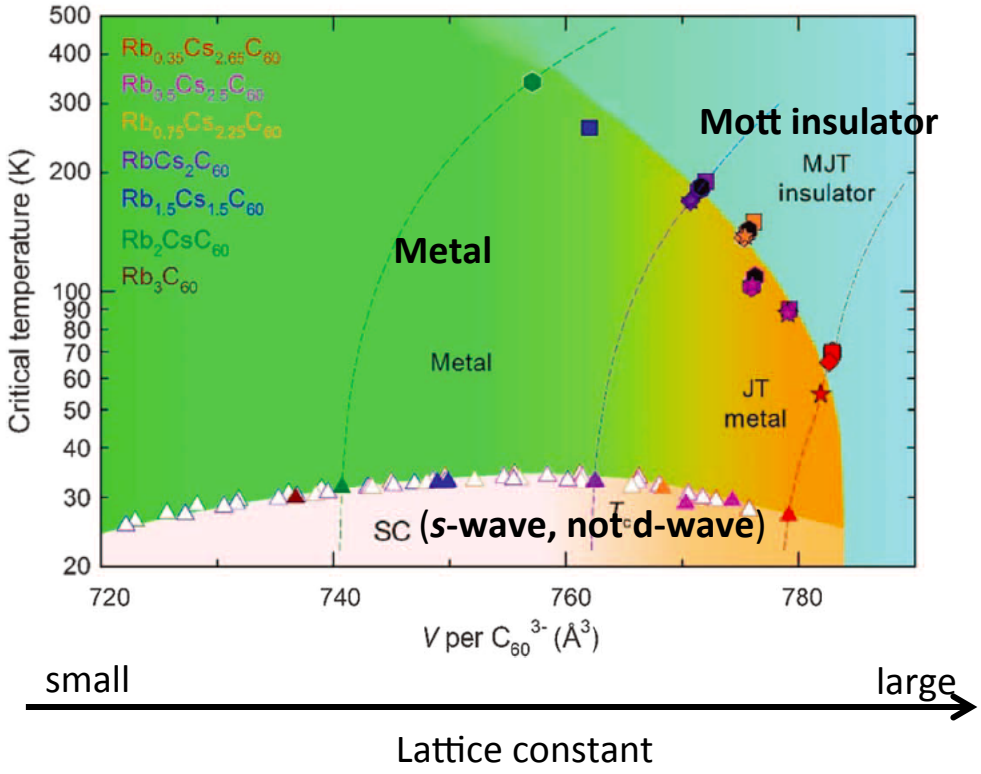
Superconductivity in alkali-doped fullerides

Zadik *et al.*, Sci. Adv. **1**, e1500059 (2015).

Cystal structure



phase diagram (fcc system)



- Mott insulating phase: induced by repulsive interaction
- *s*-wave superconductivity (T_c ~ 35K, very high for small bandwidth): induced by attractive interaction
- Low-spin state and dynamical Jahn-Teller effect in Mott phase (positive Hund's coupling should favor high-spin state)

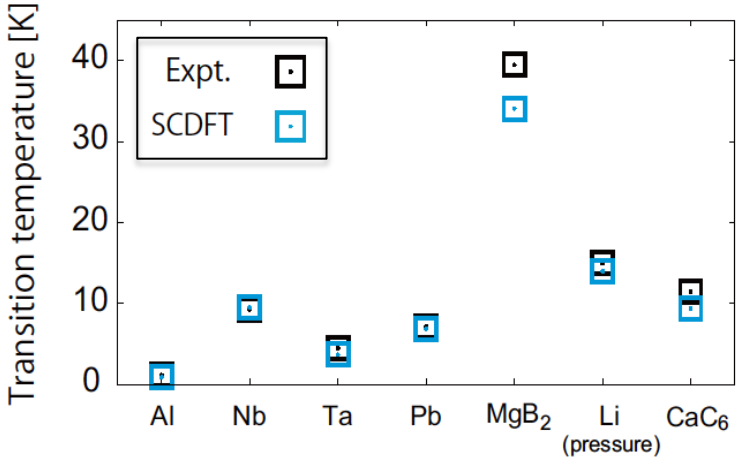
Difficulty in predicting T_c

Density functional theory for superconductors (SCDFT)

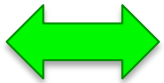
— purely theoretical method to calculate T_c without any empirical parameter such as μ^*

L. N. Oliveira *et al.*, PRL **60**, 2430 (1988); T. Kreibich and E. K. U. Gross, PRL **86**, 2984 (2001); Lueders *et al.*, PRB **72**, 024545 (2005); Marques *et al.*, PRB **72**, 024546 (2005)

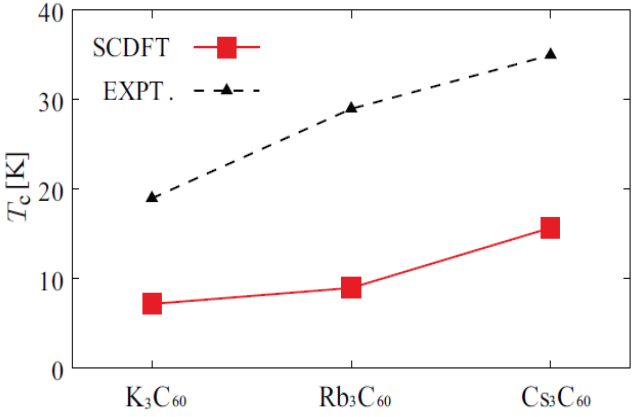
✓ Success in Conventional SC



Ryosuke Akashi, Doctor Thesis



× Failure in C_{60} superconductors



R. Akashi and R. Arita, PRB **88**, 054510 (2013)

The way to calculate T_c for **unconventional** superconductors has not been established

Motivation & Outline

- Unified description of the phase diagram
  - Pairing mechanism?
  - Why s-wave? (naïvely, strong correlation is incompatible with *s*-wave)
  - Origin of low-spin state?

- Fully *ab initio* calculation of superconducting transition temperature T_c
  - Previous methods have often employed empirical parameters

we employ the combination of density functional theory (DFT) and model-calculation method (DMFT)

We show that effectively inverted Hund's rule is realized
→ unusual cooperation between strong correlations and phonons

Unconventional mechanism !

M. Capone *el al*., RMP 81, 943 (2009); M. Capone *et al*., Science 296, 2364 (2002).
M. Capone *et al*,. PRL 86, 5361-5364 (2001); YN *et al*, Sci. Adv. 1, e1500568 (2015).

Multi-energy-scale *ab initio* scheme for correlated electrons (MACE)

	Density functional theory (DFT)	Model calculation [e.g. dynamical mean field theory (DMFT)]
Realistic	☺	☹
Strong correlation	☹	☺

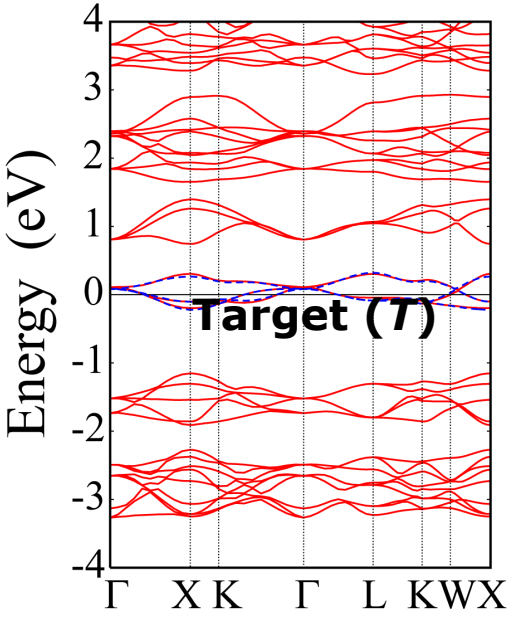
⇒ **Combination of DFT and model-calculation method**

Procedure

1. Perform the DFT band structure calculation
2. *Ab initio* derivation of low-energy Hamiltonian including phonon (lattice vibration) degrees of freedom
3. Unbiased analysis with the extended DMFT
   → reproduction of experimental phase diagram at a quantitative level

Ab initio derivation of the low-energy model

- Low-energy Hamiltonians for C_{60} superconductors (3 orbital, half-filled)



$$\mathcal{H} = \sum_{\mathbf{k}} \sum_{ij} \left[\mathcal{H}_0^{(\mathrm{w})}(\mathbf{k})\right]_{ij} c_{i\mathbf{k}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} + \sum_{\mathbf{q}} \sum_{\mathbf{k}\mathbf{k}'} \sum_{ij,i'j'} \sum_{\sigma\sigma'} U_{ij,i'j'}^{(p)}(\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{i'\mathbf{k}'}^{\sigma'\dagger} c_{j'\mathbf{k}'+\mathbf{q}}^{\sigma'} c_{j\mathbf{k}}^{\sigma}$$
$$+ \sum_{\mathbf{q}\nu} \sum_{\mathbf{k}} \sum_{ij} \sum_{\sigma} g_{ij}^{(p)\nu}(\mathbf{k},\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} \left(b_{\mathbf{q}\nu} + b_{-\mathbf{q}\nu}^{\dagger}\right) + \sum_{\mathbf{q}\nu} \omega_{\mathbf{q}\nu}^{(p)} b_{\mathbf{q}\nu}^{\dagger} b_{\mathbf{q}\nu}$$

- Electronic one-body part (red): maxloc Wannier
- Coulomb interaction part (green): cRPA method
  → talks by Priyanka, Philipp (yesterday)
- Phonon (lattice vibration) part (blue): cDFPT method

All the parameters are determined by *ab initio* calculations, not by hand

YN, K. Nakamura, and R. Arita, PRB **85**, 155452 (2012); **YN**, K. Nakamura, and R. Arita, PRL **112**, 027002 (2014)

Ab initio downfolding for electron-phonon coupled systems

Low-energy models for electron-phonon coupled systems:

Electronic one-body part

$$\mathcal{H} = \sum_{\mathbf{k}} \sum_{ij} \left[\mathcal{H}_0^{(\mathrm{w})}(\mathbf{k})\right]_{ij} c_{i\mathbf{k}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} + \sum_{\mathbf{q}} \sum_{\mathbf{k}\mathbf{k}'} \sum_{ij,i'j'} \sum_{\sigma\sigma'} U_{ij,i'j'}^{(p)}(\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{i'\mathbf{k}'}^{\sigma'\dagger} c_{j'\mathbf{k}'+\mathbf{q}}^{\sigma'} c_{j\mathbf{k}}^{\sigma}$$
$$+ \sum_{\mathbf{q}\nu} \sum_{\mathbf{k}} \sum_{ij} \sum_{\sigma} g_{ij}^{(p)\nu}(\mathbf{k},\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} \left(b_{\mathbf{q}\nu} + b_{-\mathbf{q}\nu}^{\dagger}\right) + \sum_{\mathbf{q}\nu} \omega_{\mathbf{q}\nu}^{(p)} b_{\mathbf{q}\nu}^{\dagger} b_{\mathbf{q}\nu}$$

i,j: orbital (Wannier) indices (w): Wannier gauge σ : spin index

$O^{(p)}$: the quantity with constraint (partially screened)

Maximally localized Wannier function

N. Marzari and D. Vanderbilt, Phys. Rev. B. 56 12847 (1997) ; I. Souza et al., ibid. 65, 035109 (2001)

Maximally localized Wannier function

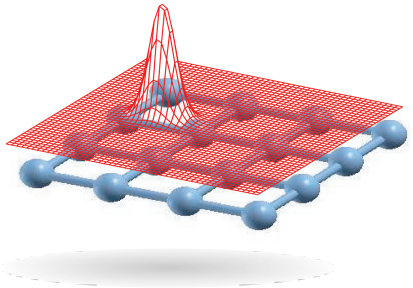
N. Marzari and D. Vanderbilt, Phys. Rev. B. 56 12847 (1997)
I. Souza et al., ibid. 65, 035109 (2001)

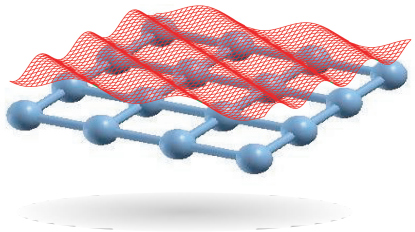
$$|w_{n\mathbf{R}}\rangle = a^{\dagger}_{n\mathbf{R}}|0\rangle$$

Wannier State

$$|\psi_{\alpha\mathbf{k}}\rangle = c^{\dagger}_{\alpha\mathbf{k}}|0\rangle$$

Bloch state





$$|w_{n\mathbf{R}}\rangle = \frac{1}{\sqrt{N}} \sum_{\alpha\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{R}} U_{\alpha n,\mathbf{k}} |\psi_{\alpha\mathbf{k}}\rangle$$

The unitary matrix *U* is decided to minimize the spread functional Ω :

$$\Omega = \sum_n \left[\langle r^2 \rangle_n - \bar{\mathbf{r}}_n^2 \right]$$

where,

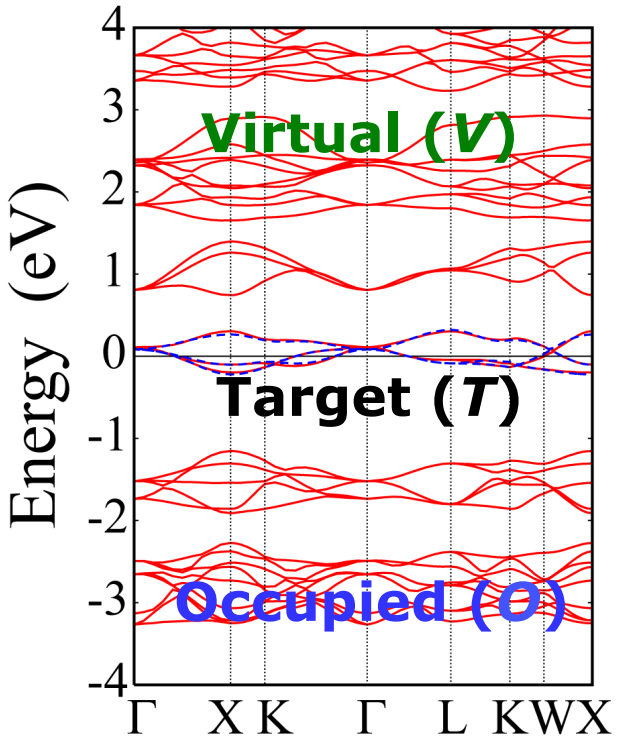
$$\bar{\mathbf{r}}_n = \langle w_{n\mathbf{0}} | \mathbf{r} | w_{n\mathbf{0}} \rangle$$

$$\langle r^2 \rangle_n = \langle w_{n\mathbf{0}} | r^2 | w_{n\mathbf{0}} \rangle$$

One body part of the Hamiltonian

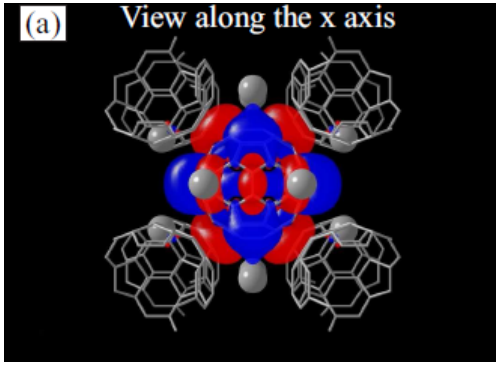
N. Marzari and D. Vanderbilt, Phys. Rev. B. 56 12847 (1997)
I. Souza et al., ibid. 65, 035109 (2001)

Band structure of fcc K_3C_{60}

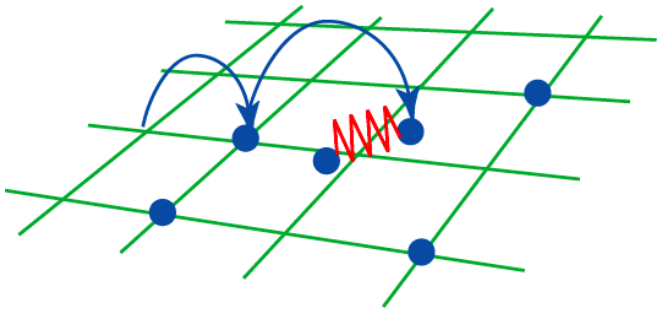


LUMO band
W ~ 0.5 eV

Max loc Wannier



molecular-orbital like



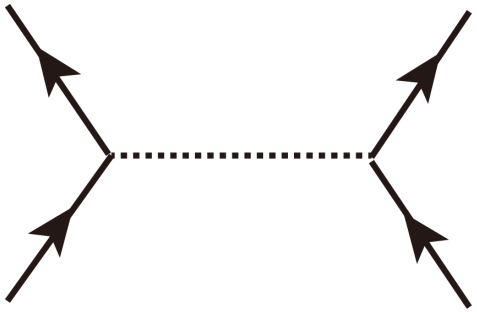
Hopping between molecular orbital

$$t_{m\mathbf{R}n\mathbf{R}'} = \langle w_{m\mathbf{R}} | \mathcal{H}_{KS} | w_{n\mathbf{R}'} \rangle$$

YN-Nakamura-Arita, Phys. Rev. B 85, 155452 (2012)

Interaction between electrons

Coulomb interaction

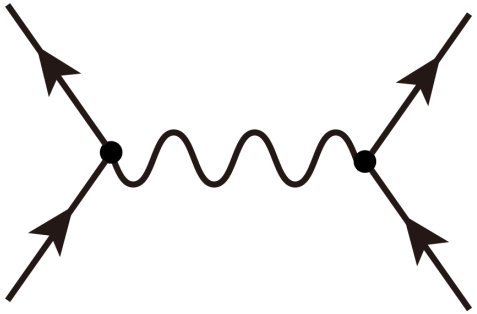


U: intraorbital
U': interorbital
J_H: exchange

repulsive

+

Interaction mediated by phonons (lattice vibration)

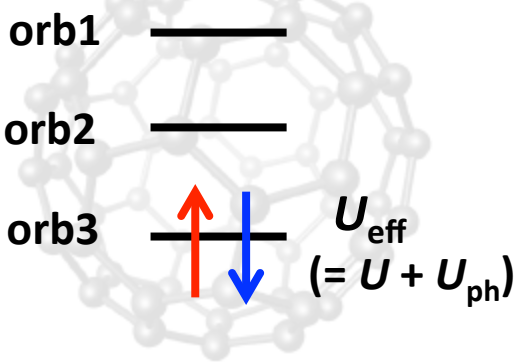


U_{ph}: intraorbital
U'_{ph}: interorbital
J_{ph}: exchange

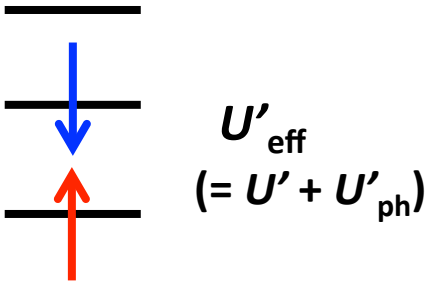
attractive

Types of intramolecular interaction

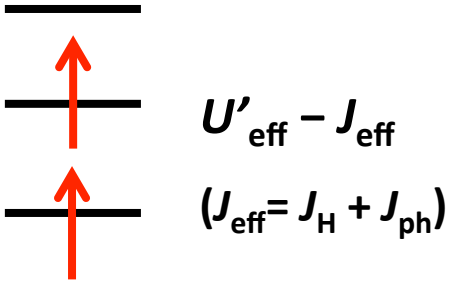
- Intraorbital



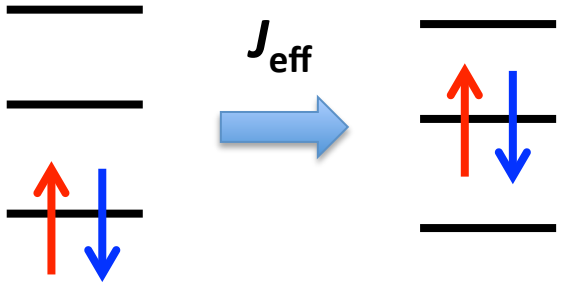
- Interorbital (opposite spin)



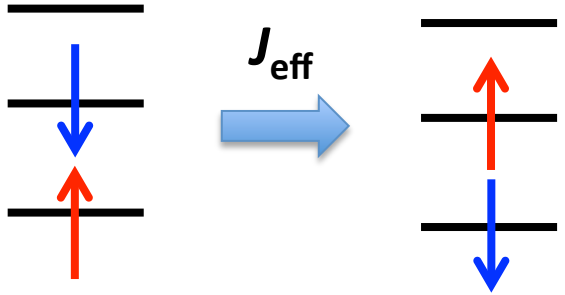
- Interorbital (same spin)



- Pair hopping



- Spin flip



Ab initio downfolding for electron-phonon coupled systems

Low-energy models for electron-phonon coupled systems:

Coulomb interaction part

$$\mathcal{H} = \sum_{\mathbf{k}} \sum_{ij} \left[\mathcal{H}_0^{(\mathrm{w})}(\mathbf{k})\right]_{ij} c_{i\mathbf{k}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} + \sum_{\mathbf{q}} \sum_{\mathbf{k}\mathbf{k}'} \sum_{ij,i'j'} \sum_{\sigma\sigma'} U_{ij,i'j'}^{(p)}(\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{i'\mathbf{k}'}^{\sigma'\dagger} c_{j'\mathbf{k}'+\mathbf{q}}^{\sigma'} c_{j\mathbf{k}}^{\sigma}$$
$$+ \sum_{\mathbf{q}\nu} \sum_{\mathbf{k}} \sum_{ij} \sum_{\sigma} g_{ij}^{(p)\nu}(\mathbf{k},\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} \left(b_{\mathbf{q}\nu} + b_{-\mathbf{q}\nu}^{\dagger}\right) + \sum_{\mathbf{q}\nu} \omega_{\mathbf{q}\nu}^{(p)} b_{\mathbf{q}\nu}^{\dagger} b_{\mathbf{q}\nu}$$

i,j: orbital (Wannier) indices (w): Wannier gauge σ : spin index

$O^{(p)}$: the quantity with constraint (partially screened)

Constrained random phase approximation (cRPA)

F. Aryasetiawan et al., Phys. Rev. B. 70 19514 (2004)

Constrained RPA

F. Aryasetiawan et al., Phys. Rev. B. 70 19514 (2004)

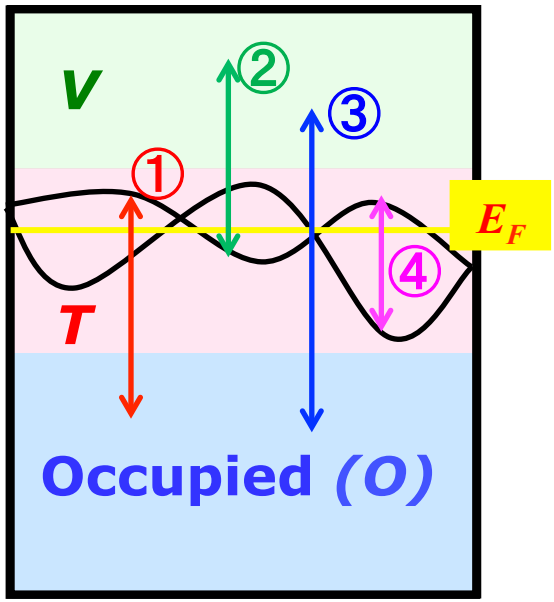
- Screened Coulomb interaction *W* (RPA)

$$W = (1 - v\chi_0)^{-1} v$$

$$\chi_0(\mathbf{r}, \mathbf{r}') = \sum_{i,j} \frac{f_i(1-f_j)}{\epsilon_i - \epsilon_j} \left[\phi_i^*(\mathbf{r})\phi_j(\mathbf{r})\phi_j^*(\mathbf{r}')\phi_i(\mathbf{r}') + \text{c.c.}\right]$$

$$\chi_0 = \sum_{O\leftrightarrow T} + \sum_{T\leftrightarrow V} + \sum_{O\leftrightarrow V} + \sum_{T\leftrightarrow T}$$

① ② ③ ④



$$\chi_{\text{cRPA}} = ① + ② + ③ + \cancel{④}$$

exclude the contribution from $T \leftrightarrow T$ scattering

This screening process should be considered when we solve the low-energy effective model

$$W_{\text{eff}} = (1 - v\chi_{\text{cRPA}})^{-1} v$$

$$U_{\mu\nu} = \int d\mathbf{r} \int d\mathbf{r}' \, |\psi_\mu(\mathbf{r})|^2 W_{\text{eff}}(\mathbf{r}, \mathbf{r}') |\psi_\nu(\mathbf{r}')|^2$$

***V*:virtual**

***T*:target**
(= C_{60} LUMO)

v: bare Coulomb interaction

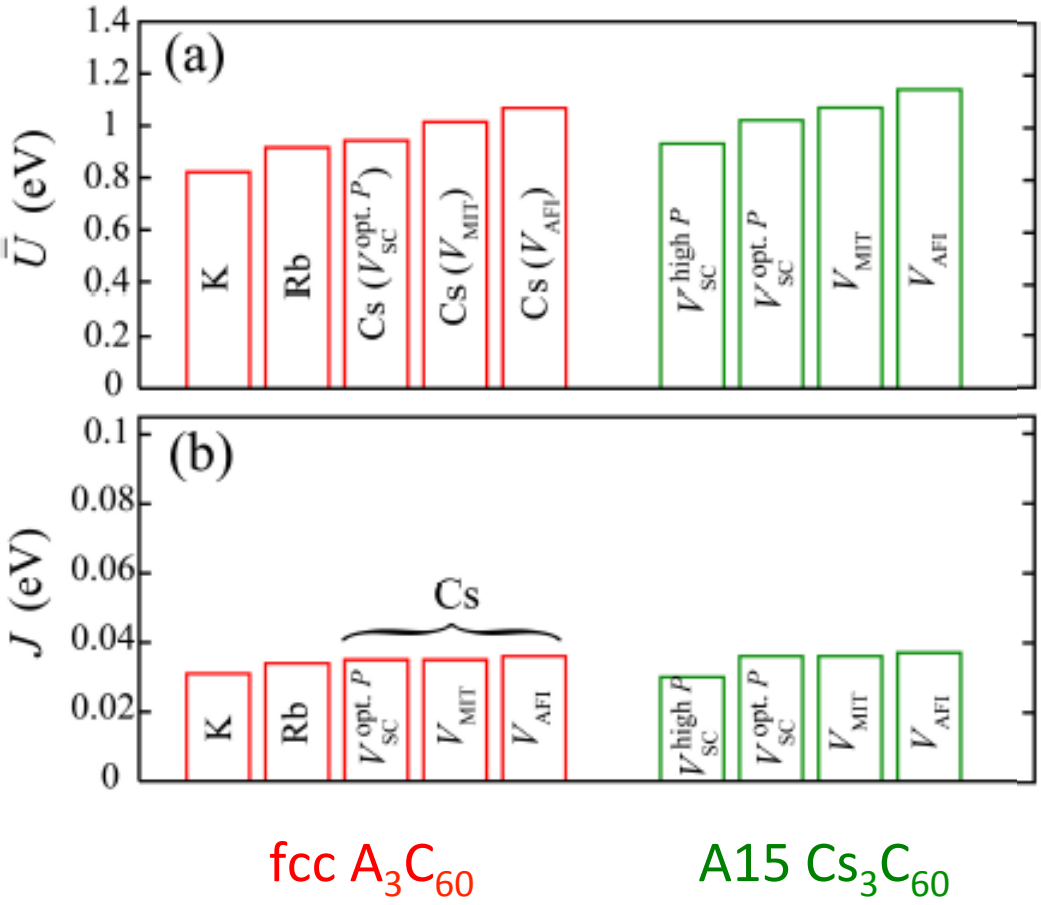
Ψ: Wannier function

Interaction parameters for C_{60} superconductors

YN-Nakamura-Arita, Phys. Rev. B 85, 155452 (2012)

Hubbard interaction

Hund's coupling



fcc A_3C_{60} A15 Cs_3C_{60}

- U ~1 eV > W ~ 0.5 eV → strongly correlated
- J_H ~ 0.035 eV → very small compared to U
- V ~ 0.25-0.30 eV

Ab initio downfolding for electron-phonon coupled systems

Low-energy models for electron-phonon coupled systems:

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$$+ \boxed{\sum_{\mathbf{q}\nu} \sum_{\mathbf{k}} \sum_{ij} \sum_{\sigma} g_{ij}^{(p)\nu}(\mathbf{k},\mathbf{q}) c_{i\mathbf{k}+\mathbf{q}}^{\sigma\dagger} c_{j\mathbf{k}}^{\sigma} \left(b_{\mathbf{q}\nu} + b_{-\mathbf{q}\nu}^{\dagger}\right) + \sum_{\mathbf{q}\nu} \omega_{\mathbf{q}\nu}^{(p)} b_{\mathbf{q}\nu}^{\dagger} b_{\mathbf{q}\nu}}$$

phonon part

i,j: orbital (Wannier) indices (w): Wannier gauge σ : spin index

$O^{(p)}$: the quantity with constraint (partially screened)

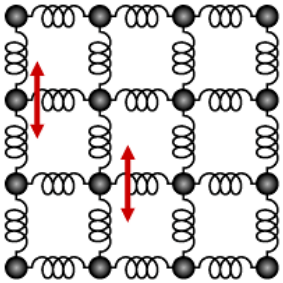
Constrained density-functional perturbation theory (cDFPT)

YN, K. Nakamura, and R. Arita, Phys. Rev. Lett. 112, 027002 (2014)
YN and R. Arita, arXiv:1509.01138.

Density-functional perturbation theory (DFPT)

S. Baroni *et al*, Rev. Mod. Phys. **73**, 515 (2001).

- phonon frequency

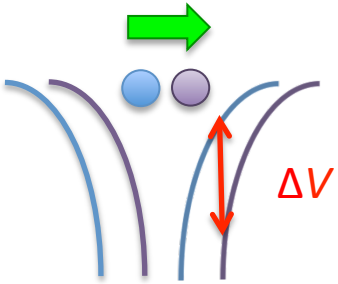


$$\omega = \sqrt{\frac{k}{m}}$$

$$k = k_{\mathrm{ion}} + k_{\mathrm{el}}$$

http://monoist.atmarkit.co.jp/feledev/articles/heat/01/netsu01_05.gif

- electron-phonon coupling



$$g_{\mu\nu} = \langle \psi_\mu | \Delta V | \psi_\nu \rangle$$

$$\Delta V = \Delta V_{\mathrm{ion}} + \Delta V_{\mathrm{el}}$$

k_{el} and ΔV_{el} are related with the electron-density response to ionic displacement

⇒ density-functional perturbation

S. Baroni *et al*, Rev. Mod. Phys. **73**, 515 (2001).

Phonon frequency and electron-phonon coupling

Phonon frequencies $\omega_{\mathbf{q}\nu}$ and **electron-phonon couplings** $g^{\nu}_{n'n}(\mathbf{k},\mathbf{q})$ are given by
(for simplicity we consider the case where there is one atom with mass *M* in the unit cell)

$$D(\mathbf{q})\mathbf{e}_\nu(\mathbf{q}) = \omega^2_{\mathbf{q}\nu}\mathbf{e}_\nu(\mathbf{q})$$

Dynamical matrix

$$g^{\nu}_{n'n}(\mathbf{k},\mathbf{q}) = \left\langle \psi_{n'\mathbf{k}+\mathbf{q}} \middle| u_0 \mathbf{e}_\nu(\mathbf{q}) \cdot \frac{\partial V_{SCF}(\mathbf{r})}{\partial \mathbf{u}(\mathbf{q})} \middle| \psi_{n\mathbf{k}} \right\rangle$$

Potential change due to the ionic displacement

v: phonon mode
u : displacement of the ion

$u_0 = \sqrt{\frac{\hbar}{2M\omega_{\mathbf{q}\nu}}}$: characteristic length scale

where

$$\left[D(\mathbf{q})\right]_{\alpha\alpha'} = \frac{1}{M}\frac{\partial^2 E}{\partial u^*_\alpha(\mathbf{q})\partial u_{\alpha'}(\mathbf{q})} = \frac{1}{M}\left[C(\mathbf{q})\right]_{\alpha\alpha'}$$

Interatomic force constant (~spring constant)

E : electron ground-state energy
α: cartesian coordinates (*x*,*y*,*z*)
n(**r**): electron density

$$\left[C(\mathbf{q})\right]_{\alpha\alpha'} = \frac{1}{N}\left[\int \left(\frac{\partial n(\mathbf{r})}{\partial u_\alpha(\mathbf{q})}\right)^* \frac{\partial V_{ion}(\mathbf{r})}{\partial u_{\alpha'}(\mathbf{q})} d\mathbf{r} + \int n(\mathbf{r}) \frac{\partial^2 V_{ion}(\mathbf{r})}{\partial u^*_\alpha(\mathbf{q})\partial u_{\alpha'}(\mathbf{q})} d\mathbf{r} + \frac{\partial E_{\mathrm{N}}}{\partial u^*_\alpha(\mathbf{q})\partial u_\alpha(\mathbf{q})}\right]$$

renormalizing (softening) — bare

$$\frac{\partial V_{SCF}(\mathbf{r})}{\partial \mathbf{u}(\mathbf{q})} = \frac{\partial V_{ion}(\mathbf{r})}{\partial \mathbf{u}(\mathbf{q})} + \int \left(\frac{e^2}{|\mathbf{r}-\mathbf{r}'|} + \frac{dV_{xc}(\mathbf{r})}{dn}\delta(\mathbf{r}-\mathbf{r}')\right)\frac{\partial n(\mathbf{r}')}{\partial \mathbf{u}(\mathbf{q})} d\mathbf{r}'$$

bare — Hartree + exchange correlation terms (screening)

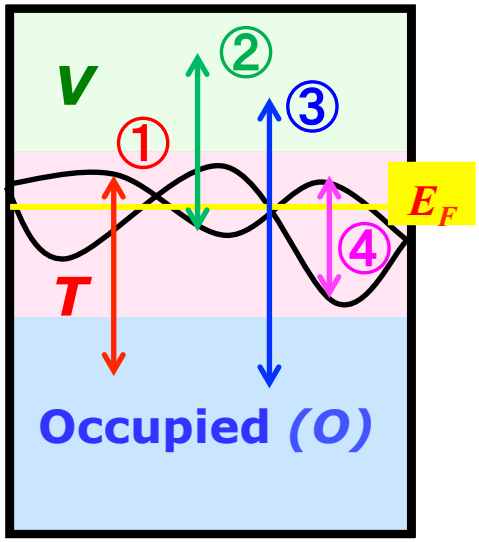
Constrained DFPT

YN, K. Nakamura, and R. Arita, PRL **112**, 027002 (2014)
YN and R. Arita, arXiv:1509.01138.

In the metallic case, $\frac{\partial n(\mathbf{r})}{\partial \mathrm{u}(\mathbf{q})}$ is given by

$$\frac{\partial n(\mathbf{r})}{\partial \mathrm{u}(\mathbf{q})} = 2\sum_{nm}\sum_{\mathbf{k}} \frac{f_{n\mathbf{k}} - f_{m\mathbf{k}+\mathbf{q}}}{\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}+\mathbf{q}}} \psi^*_{n\mathbf{k}}(\mathbf{r})\psi_{m\mathbf{k}+\mathbf{q}}(\mathbf{r}) \langle \psi_{m\mathbf{k}+\mathbf{q}} | \frac{\partial V_{SCF}(\mathbf{r})}{\partial \mathrm{u}(\mathbf{q})} | \psi_{n\mathbf{k}} \rangle$$

n,m: band indices



$$\sum_{O\leftrightarrow T} + \sum_{T\leftrightarrow V} + \sum_{O\leftrightarrow V} + \cancel{\sum_{T\leftrightarrow T}}$$

① ② ③ ④

exclude the target-target processes

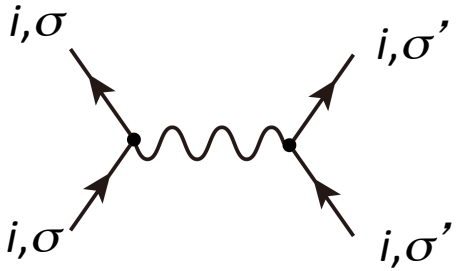
Partially screened quantities such as $g^{(p)\nu}_{ij}(\mathbf{k},\mathbf{q})$ and $\omega^{(p)}_{\mathbf{q}\nu}$

(excluding the target-subspace renormalization effect)

***V*:virtual** ***T*:target** (= C_{60} LUMO)

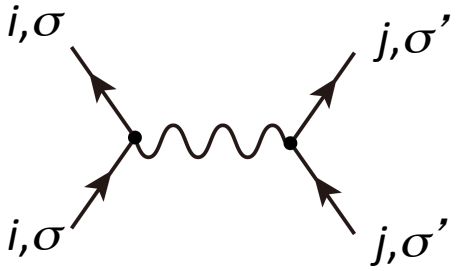
Phonon-mediated interactions

U_{ph}



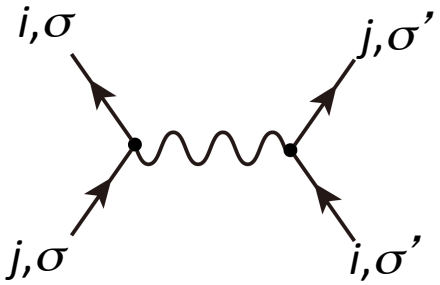
$$U_{\mathrm{ph}}(i\omega_n) = -\sum_{\nu}(g_{ii}^{\nu})^2 D(i\omega_n) = -\sum_{\nu}(g_{ii}^{\nu})^2 \frac{2\omega_\nu}{\omega_n^2+\omega_\nu^2}$$

U'_{ph}

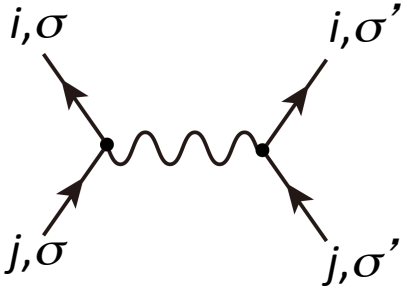


$$U'_{\mathrm{ph}}(i\omega_n) = -\sum_{\nu} g_{ii}^{\nu} g_{jj}^{\nu} D(i\omega_n) = -\sum_{\nu} g_{ii}^{\nu} g_{jj}^{\nu} \frac{2\omega_\nu}{\omega_n^2+\omega_\nu^2}$$

J_{ph}



or



$$J_{\mathrm{ph}}(i\omega_n) = -\sum_{\nu} g_{ij}^{\nu} g_{ji}^{\nu} \frac{2\omega_\nu}{\omega_n^2+\omega_\nu^2}$$

$$= -\sum_{\nu} g_{ij}^{\nu} g_{ji}^{\nu} \frac{2\omega_\nu}{\omega_n^2+\omega_\nu^2}$$

Phonon-mediated interaction for fcc A_3C_{60}

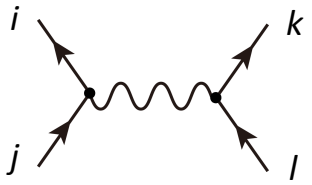
YN *et al.*, Science Advances **1**, e1500568 (2015)

small → Lattice constant → large

	K_3C_{60}	Rb_3C_{60}	Cs_3C_{60}	Cs_3C_{60}	Cs_3C_{60}
U_{ph} (0) [eV]	-0.15	-0.14	-0.11	-0.12	-0.13
U'_{ph} (0) [eV]	-0.053	-0.042	-0.013	-0.022	-0.031
J_{ph} (0) [eV]	-0.050	-0.051	-0.051	-0.051	-0.052

$U_{ph} = V_{ii,ii}$, $U'_{ph} = V_{ii,jj}$, $J_{ph} = V_{ij,ij}$

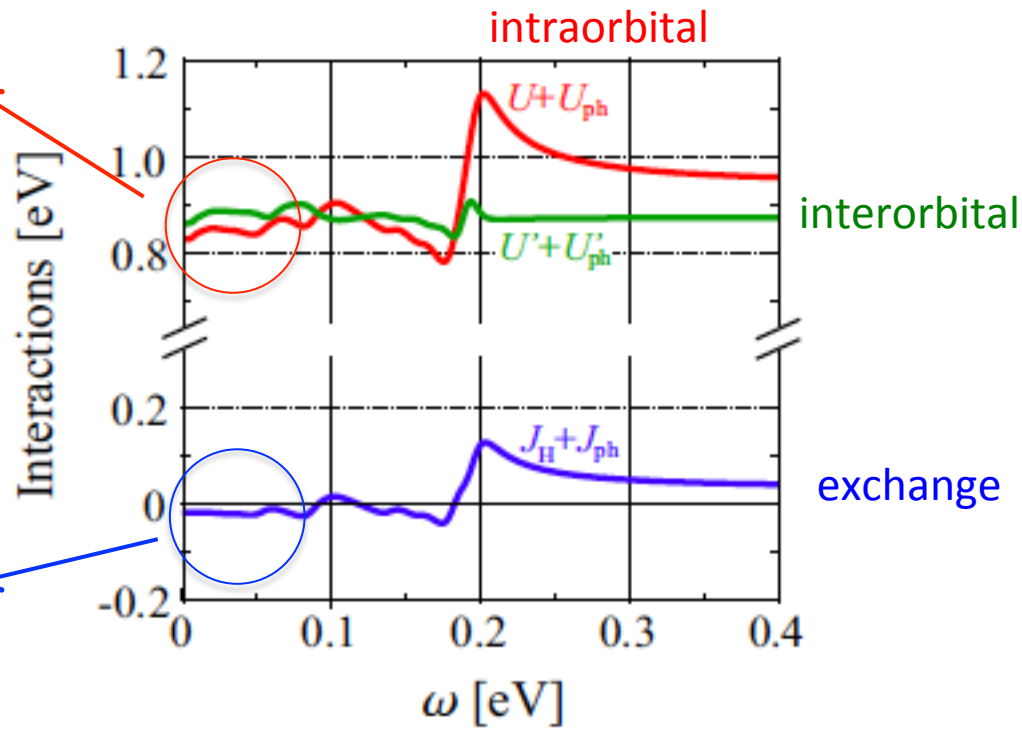
$V_{ij,kl}$ =



- $|J_{ph}(0)| \sim 0.05$ eV > $J_H \sim 0.035$ eV → negative exchange interaction (Inverted Hund's rule)
- The relation $U'_{ph} \sim U_{ph} - 2J_{ph}$ holds

Effective intramolecular interaction

Repulsive interaction
(Coulomb wins)
Effectively, $U' > U$



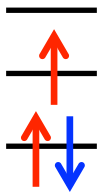
Effectively, $J < 0$
(phonon wins)
Attractive interaction

snap shot

$U > U', J > 0$

(111) configuration

$U < U', J < 0$



(210) configuration

Model Analysis

- Extended dynamical mean-field theory (extended DMFT)
  - — DMFT+ dynamical screening coming from off-site Coulomb interactions

A. M. Sengupta and A. Georges, PRB **52**, 10295 (1995); Q. Si and J. L. Smith, PRL **77**, 3391 (1996).

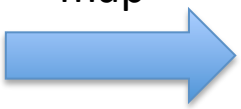
Lattice model

Onsite repulsion : U

Non-local Coulomb int. : V

Phonon-mediated int.: U_{ph}

map



Anderson Impurity model

Interaction : $U_{imp} = U + U_{ph} + U_v$

dynamical screening

Solver: continuous-time quantum Monte Carlo (strong coupling expansion)

+ self consistent conditions

- ✓ describes the local correlation (intramolecular quantum fluctuations) accurately
- ✓ One of the most reliable methods to study three-dimensional *s*-wave SC

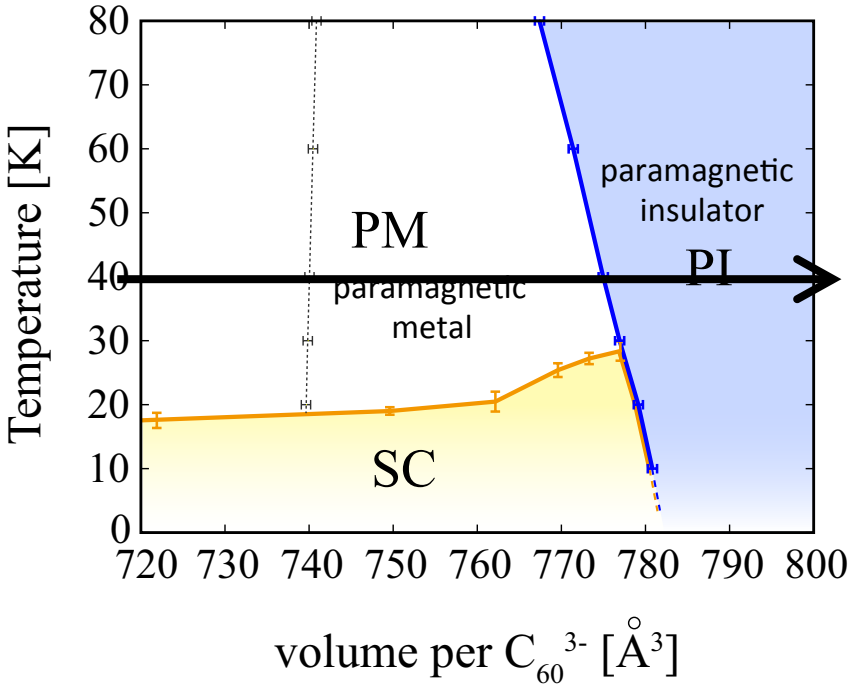
DMFT analysis using realistic Hamiltonian

→ Jan's talk

Phase diagram

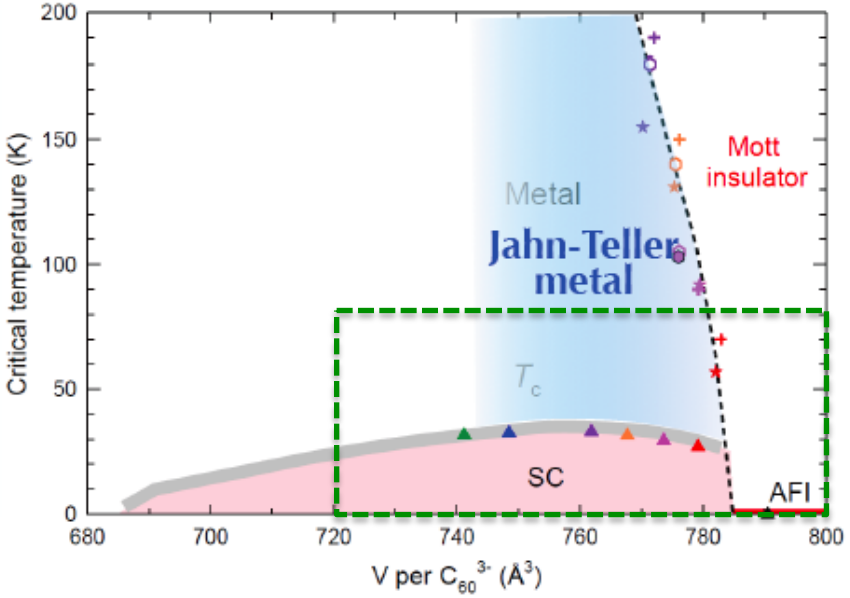
Theoretical (fcc)

DMFT analysis using realistic Hamiltonian



YN *et al.*, Science Advances **1**, e1500568 (2015).

Experimental (fcc)



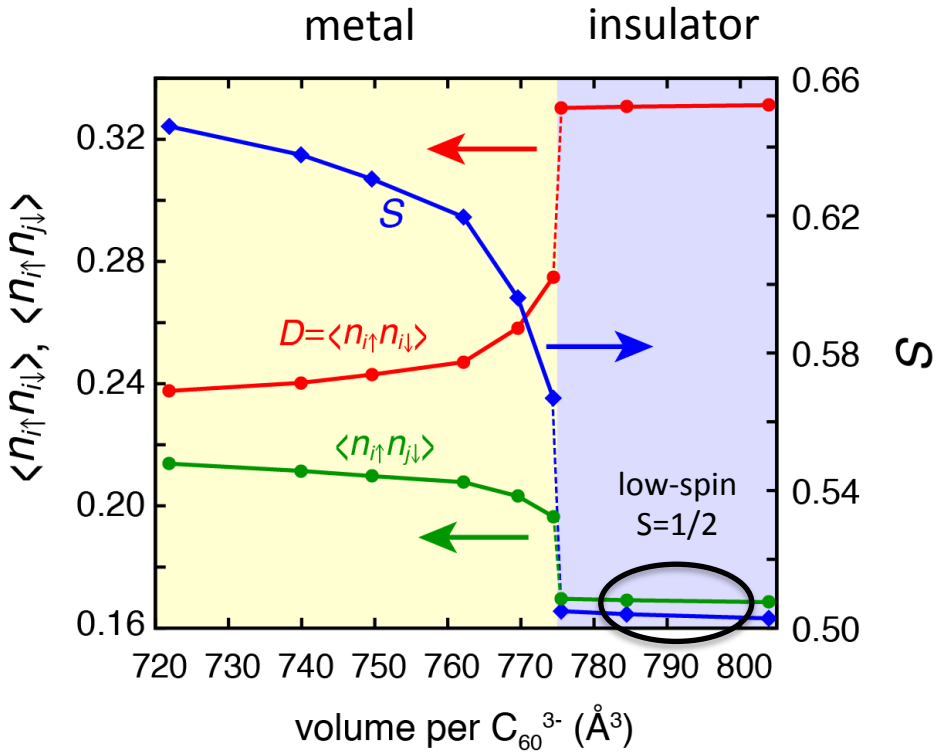
Courtesy of Y. Kasahara

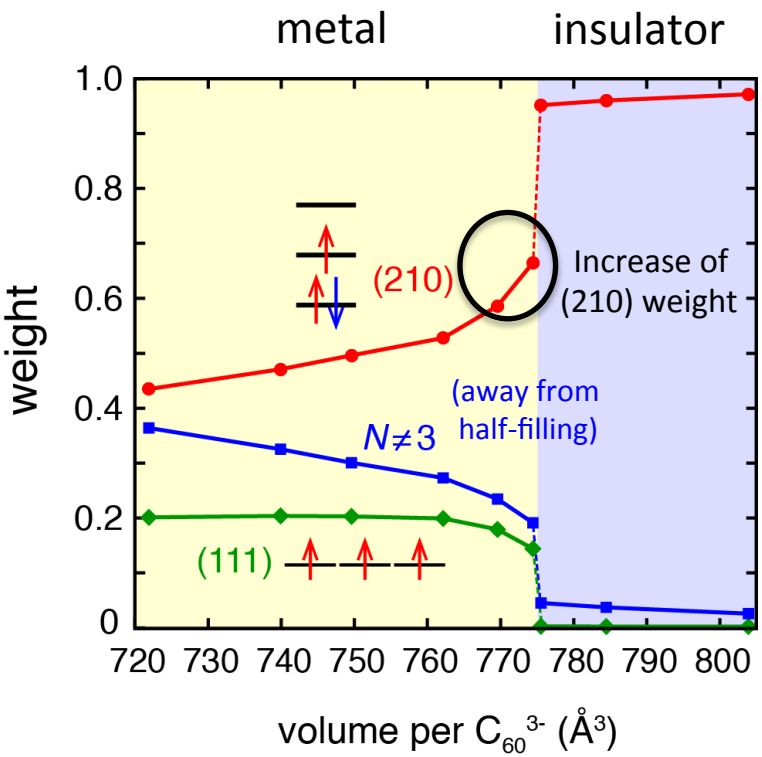
Zadik *et al.*, Sci. Adv. **1**, e1500059 (2015).

- ✓ *s*-wave SC next to Mott phase with T_c ~ 30 K
- ✓ Critical volume
- ✓ Slope between PM and PI

Property of metal-insulator transition at 40 K (above T_c)

YN *et al.*, Science Advances **1**, e1500568 (2015).





- (210) configurations dominate (because of $U' > U$)
- Mott physics: filling is (nearly) fixed at half-filling in the insulating phase

no ferro-orbital order, i.e., six types of (210) configurations
($\{n_1,n_2,n_3\}$ ={2,1,0}, {0,2,1}, {1,0,2}, {2,0,1}, {1,2,0}, {0,1,2}) are degenerate

Superconducting mechanism

YN *et al.*, Science Advances **1**, e1500568 (2015).

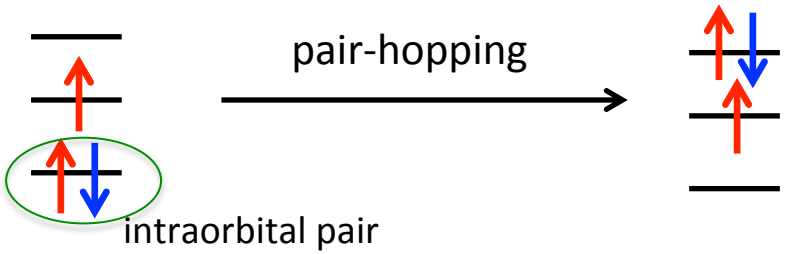
- Stability of superconductivity at 10 K

realistic	(pair hopping)=0	(spin flip)=0	$U'_{eff} < U_{eff}$ (U'_{ph}(new) = U_{ph})
SC	Non SC	SC	Non SC

- The crucial factors for *s*-wave superconductivity are

1. Generation of intraorbital pair by $U'_{eff} > U_{eff}$
   strong correlation helps it by suppressing kinetic energy
2. Tunneling of the pairs due to pair-hopping term (Suhl-Kondo mechanism)

H. Suhl et al. (1959); J. Kondo (1963)



strong electron correlations and phonons cooperatively work for SC (unconventional)

Summary

- *Ab initio* derivation of low-energy model
  - ✓ $U, U' > W \rightarrow$ strongly correlated
  - ✓ $J \rightarrow$ effectively inverted
- Unbiased EDMFT analysis
  - ✓ Treat both Coulomb and electron-phonon interactions
- Quantitative and qualitative reproduction of the phase diagram
  - ✓ $T_c \sim 30$K
  - ✓ Mott phase next to SC phase
- Superconducting mechanism
  - ✓ Generation of intraorbital pairs by $U' > U$ (strong correlation helps)
  - ✓ Tunneling of the pair due to the pair-hopping term (Suhl-Kondo mechanism)

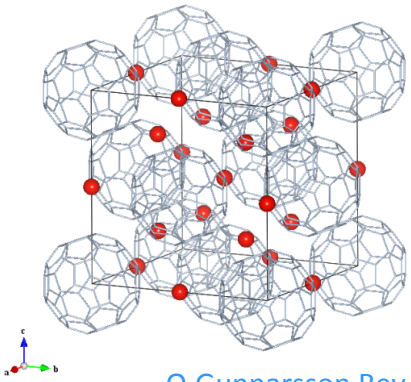
YN *et al.*, Science Advances 1, e1500568 (2015).

Outlook: light-induced superconducting-like phase (Tc > 100 K) in K_3C_{60}

M. Mitrano et al., arXiv: 1505.04529

Alkali-doped fullerides

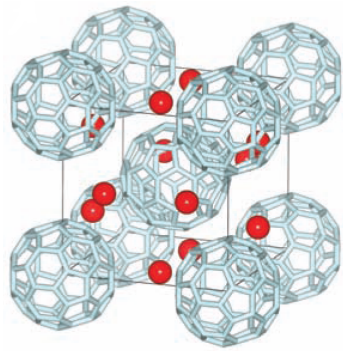
- fcc A_3C_{60} (A=K, Rb, Cs)



K_3C_{60} : T_c = 19 K
Rb_3C_{60} : T_c = 29 K
Cs_3C_{60} : T_c = 35 K

O.Gunnarsson Rev.Mod.Phys. 69, 575 (1997)
Ganin et al, Nature 466,221(2010)

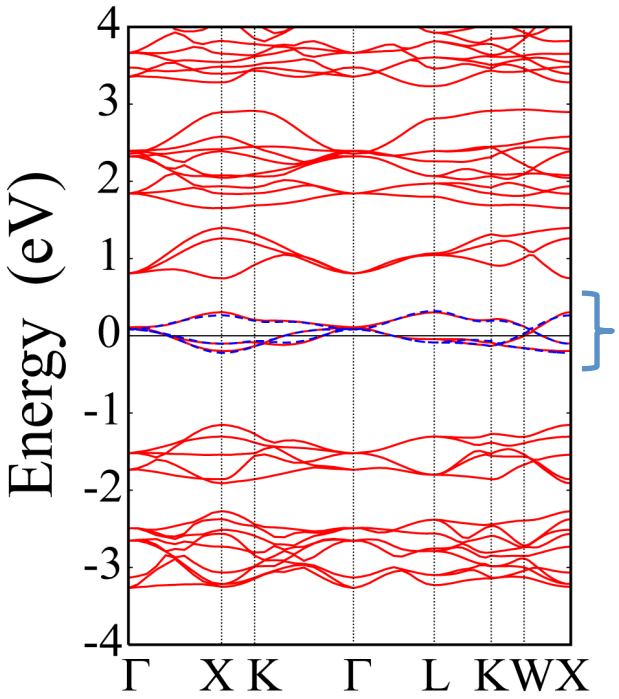
- A15 Cs_3C_{60}



T_c = 38 K

A.Ganin et al Nature Mater. 7,367-371(2008)
Y.Takabayashi et al Science 323,1285-1590(2009)

- band structure (fcc K_3C_{60})



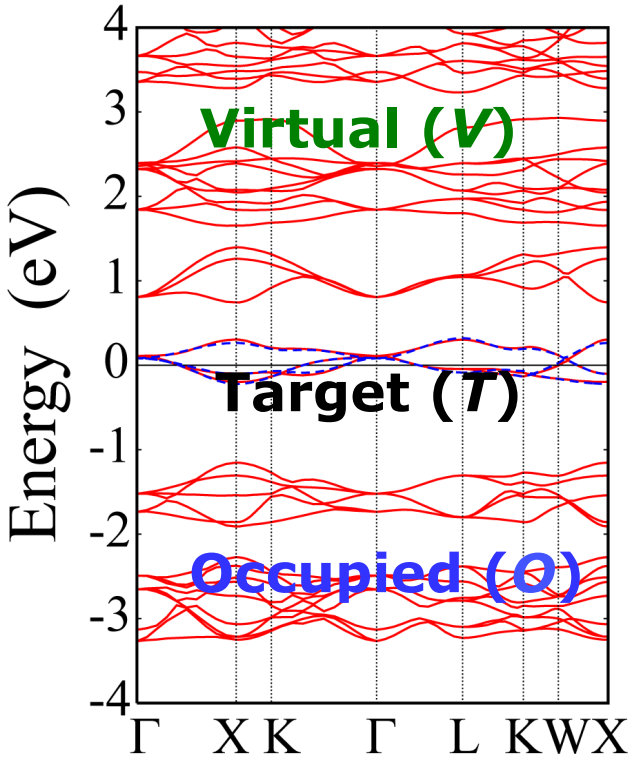
✓ 3 orbital, half-filling
✓ Molecular orbital
+ small hopping between them

Effective low-energy Hamiltonian

Ab initio Hamiltonian

$$H_{FP} = \sum_i \left(-\frac{\hbar^2}{2m}\Delta_i - \sum_\alpha \frac{Z_\alpha e^2}{|\mathbf{R}_\alpha - \mathbf{r}_i|} \right) + \frac{1}{2}\sum_{ij}\frac{e^2}{r_{ij}} + \sum_{\alpha<\beta}\frac{Z_\alpha Z_\beta e^2}{|\mathbf{R}_\alpha - \mathbf{R}_\beta|}$$

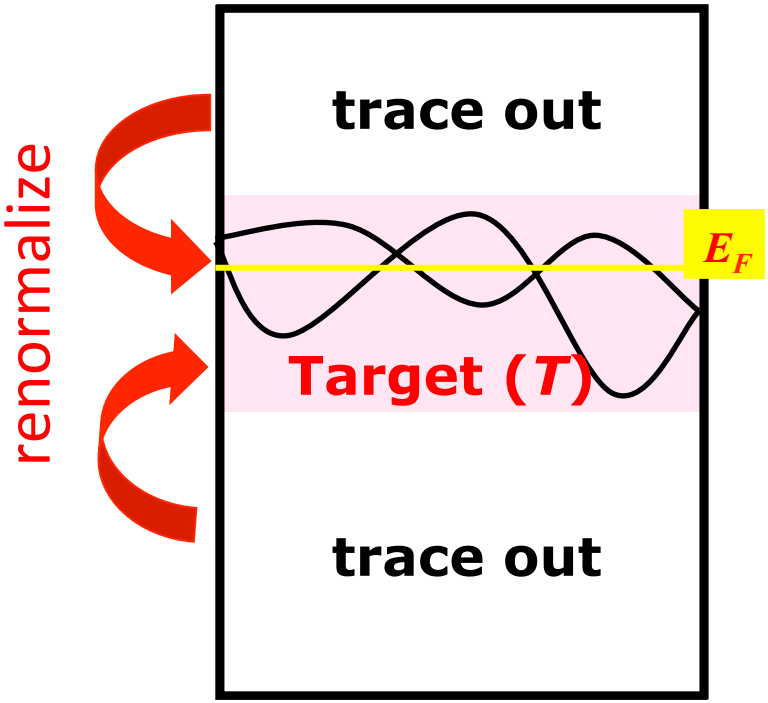
global energy scale (> 100,000 K ~ 10 eV)



Effective Hamiltonian

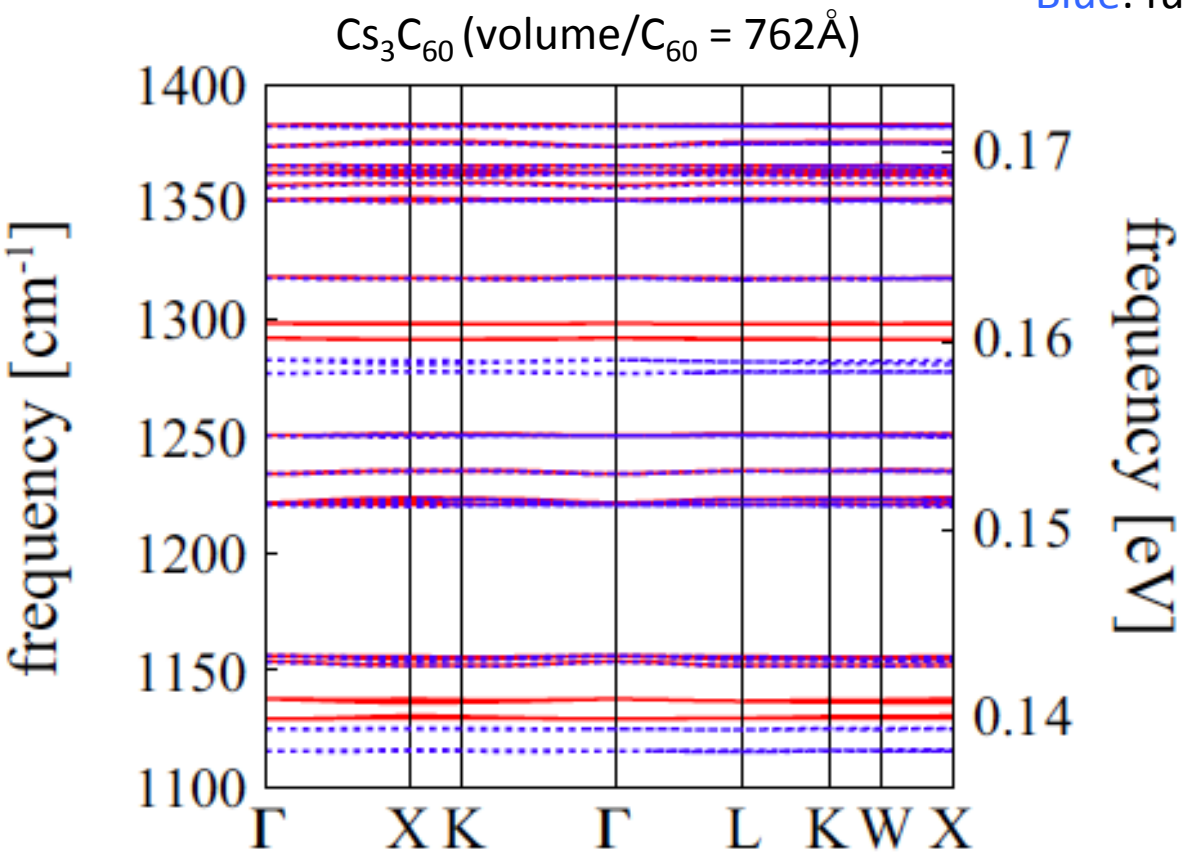
$$H_{eff} = \sum_\sigma \sum_{i\neq j} t_{ij} a^\dagger_{i\sigma} a_{j\sigma} + \sum_{ijkl}\sum_{\sigma\rho} W_{ijkl} a^\dagger_{i\sigma} a^\dagger_{k\rho} a_{l\rho} a_{j\sigma}$$

low-energy phenomena (~ 50 K)



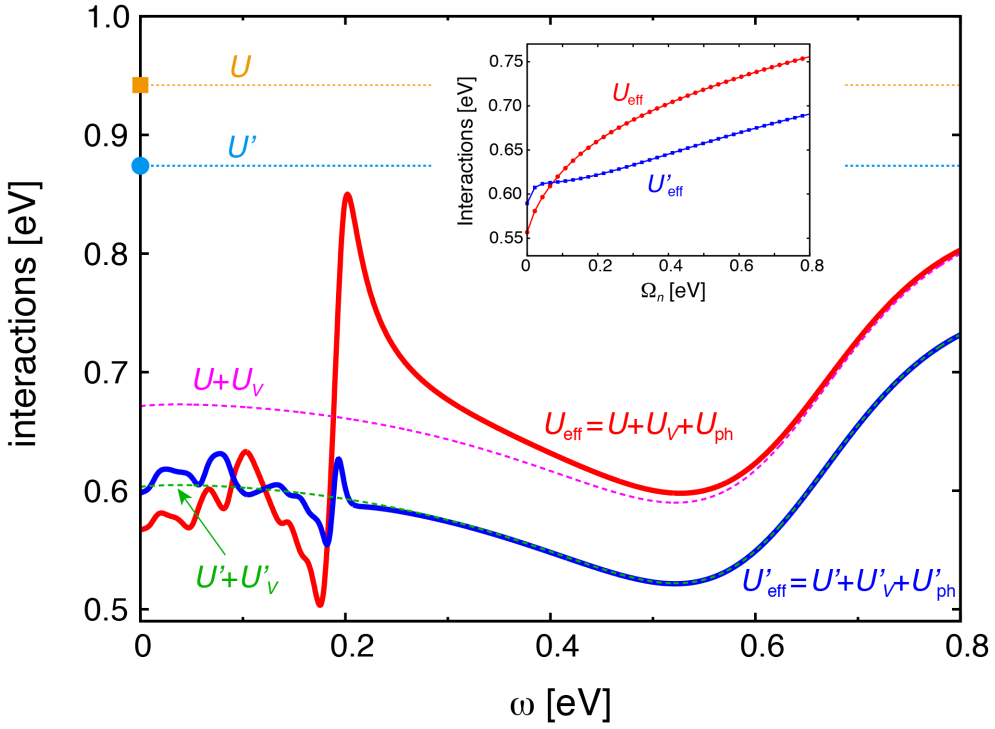
Phonon frequency

Red: partially renormalized
Blue: fully renormalized



✓ Frequencies of the phonons coupled to t_{1u} electrons are renormalized
✓ Tiny momentum dependence → Einstein-like phonon

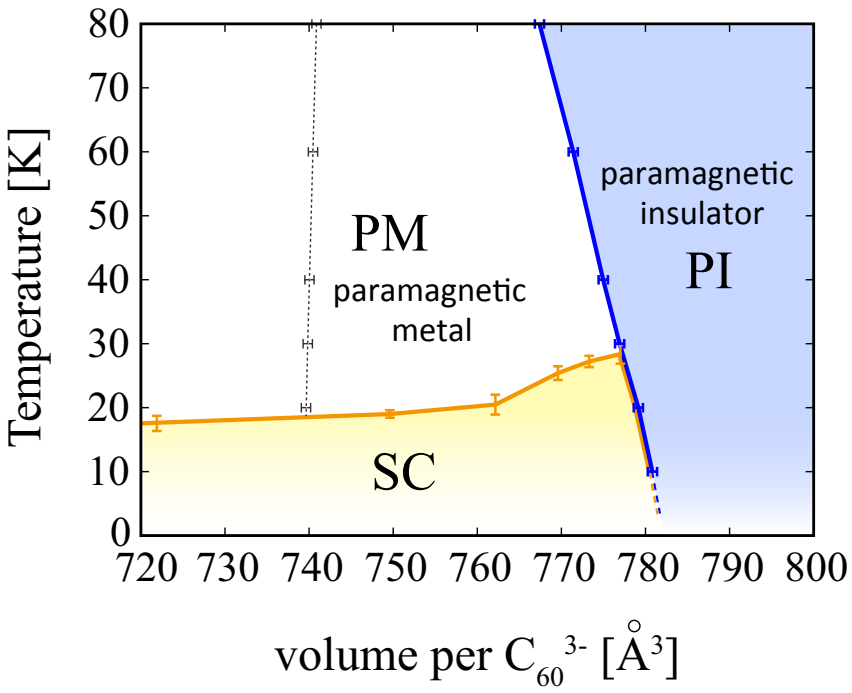
Dynamical structure of U_{eff} and U'_{eff}



- ✓ $U'_{eff} > U_{eff}$ for $\omega \lesssim 0.2$ eV (difference is small ~ 5 %)
- ✓ Consider the dynamical screening effects through off-site interactions and electron phonon coupling (we use static $J_{eff} = J_{ph}(0)+J_H$)
- ✓ Effects of off-site Coulomb interactions
  → U and U' are reduced by ~ 0.27 eV

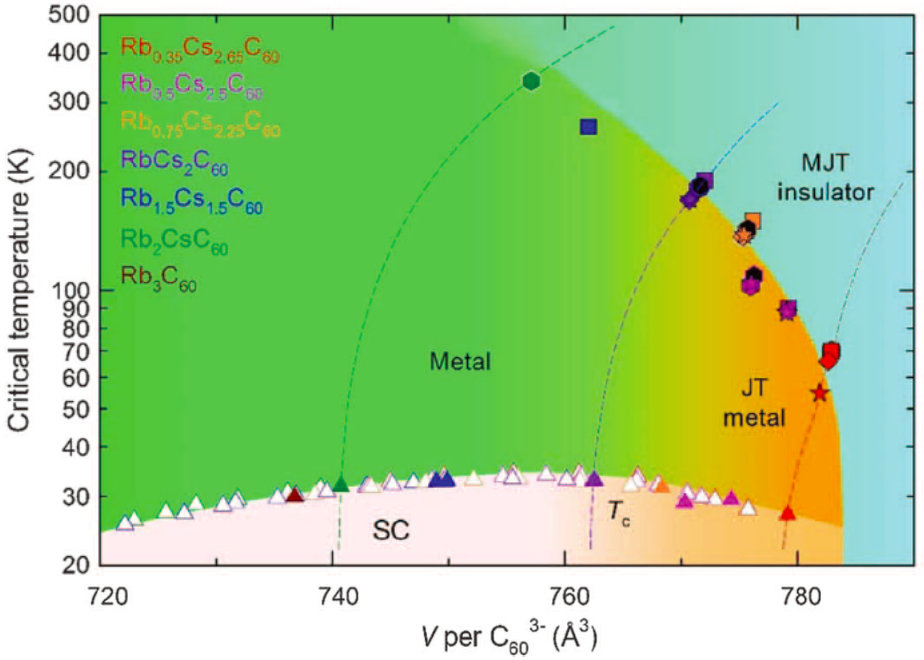
Phase diagram

Theoretical (fcc)



YN *et al.*, Science Advances **1**, e1500568 (2015).

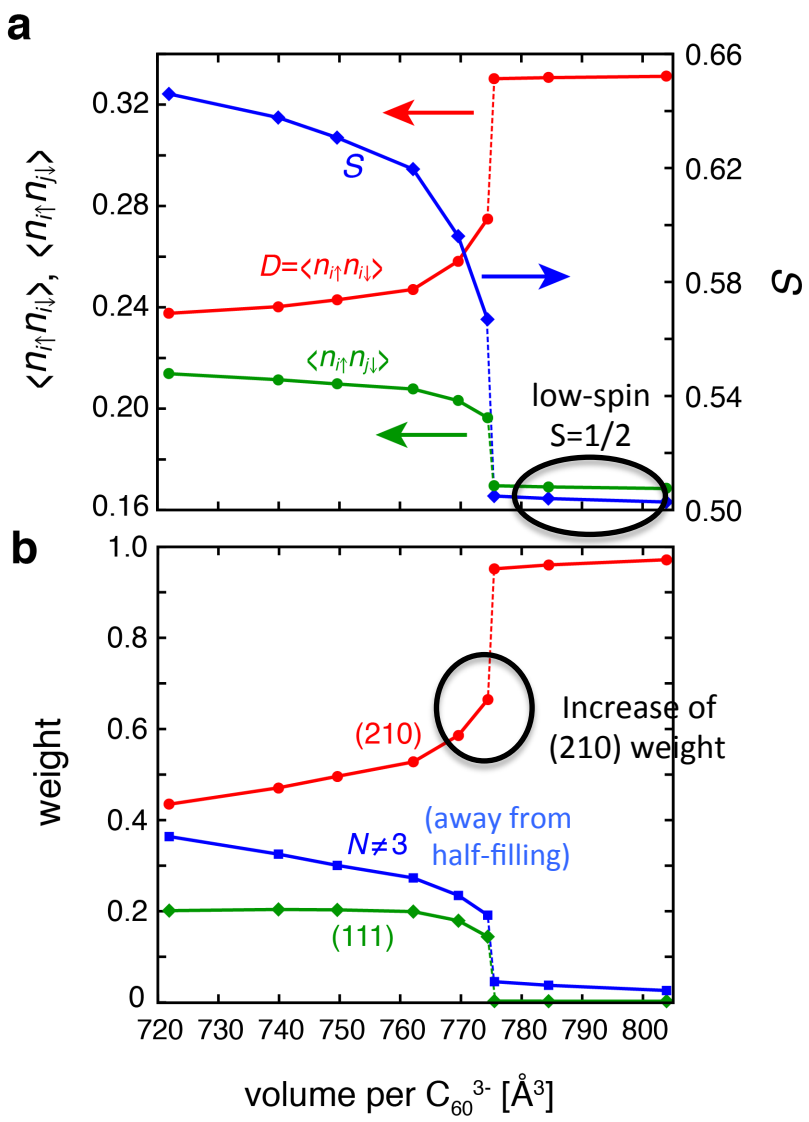
Experimental (fcc)

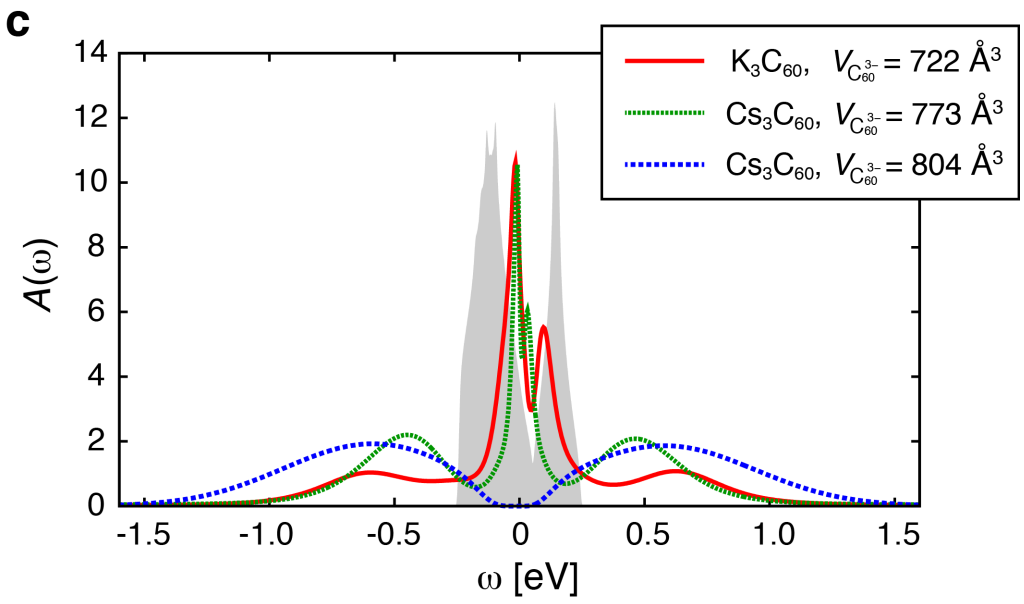


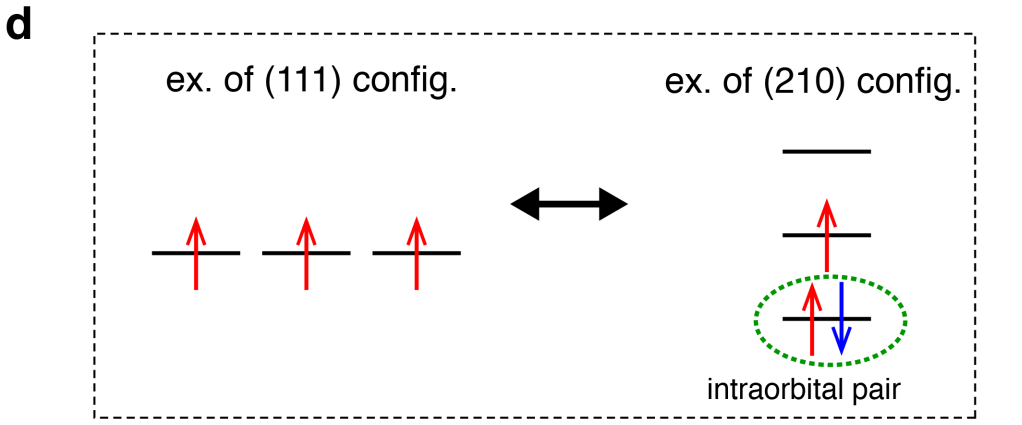
Zadik *et al.*, Sci. Adv. **1**, e1500059 (2015).

- ✓ *s*-wave SC next to Mott phase with T_c ~ 30 K
- ✓ Critical volume
- ✓ Slope between PM and PI

Quantity at 40 K (above T_c)







YN *et al.*, Science Advances **1**, e1500568 (2015).

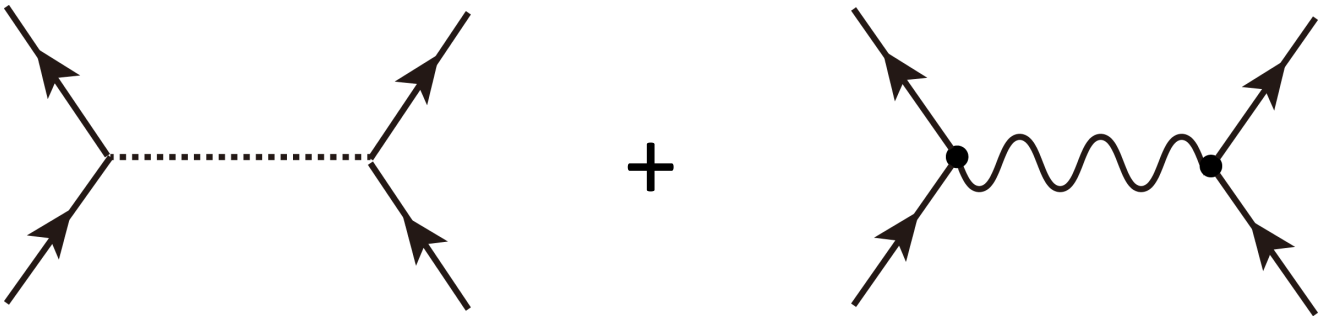
Interaction between electrons

Coulomb interaction

repulsive

Interaction mediated by phonons (lattice vibration)

attractive



U ~ 1 eV (> W ~ 0.5 eV)
J_H ~ 0.035 eV
($U' \sim U - 2J_H$)

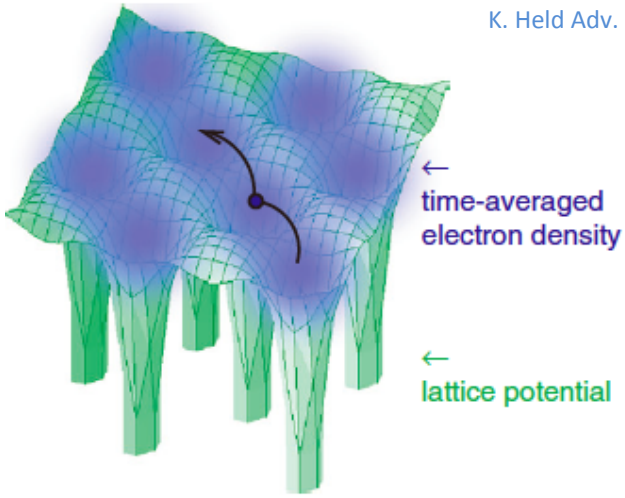
U_{ph} ~ −0.1 eV
J_{ph} ~ −0.05 eV
($U'_{ph} \sim U_{ph} - 2J_H$)

➢ $|J_{ph}|$ ~ 0.05 eV > J_H ~ 0.035 eV → **Inverted Hund's rule**

Density functional theory (DFT)

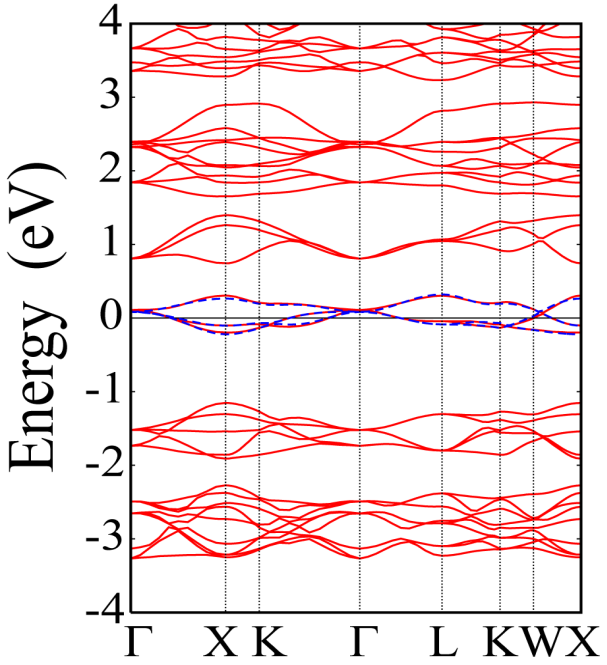
- Good approximation for *ab initio* Hamiltonian

$$H_{FP} = \sum_i \left(-\frac{\hbar^2}{2m}\Delta_i - \sum_\alpha \frac{Z_\alpha e^2}{|\mathbf{R}_\alpha - \mathbf{r}_i|} \right) + \frac{1}{2}\sum_{ij}\frac{e^2}{r_{ij}} + \sum_{\alpha<\beta}\frac{Z_\alpha Z_\beta e^2}{|\mathbf{R}_\alpha - \mathbf{R}_\beta|}$$



K. Held Adv. Phys. 56 829 (2007).

- band structure (fcc K_3C_{60})



☺ Weakly correlated material

☺ Material dependence (e.g. difference between K_3C_{60} and Rb_3C_{60})

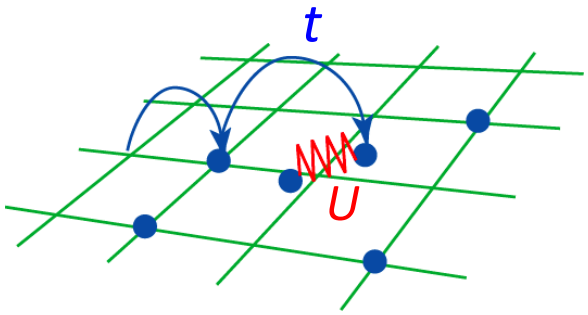
☹ Strongly correlated material

model calculation

- Solving the lattice model (e.g. Hubbard model) accurately

$$\mathcal{H} = -t \sum_{\langle i,j \rangle} \sum_{\sigma} (\hat{c}^{\dagger}_{i\sigma} \hat{c}_{j\sigma} + \hat{c}^{\dagger}_{j\sigma} \hat{c}_{i\sigma}) + U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

hopping Coulomb repulsion



Solver: dynamical mean-field theory (DMFT), variational Monte Carlo,
path-integral renormalization group, tensor network,
density matrix renormalization group ….

☺ Describes the effect of strong electron correlations (e.g. Mott insulator)

☹ Realistic calculation (what is an appropriate value for *t*, *U*?)