Enhancing Superconductivity of A_3C_{60} fullerides

arXiv:1606.05796 (2016)

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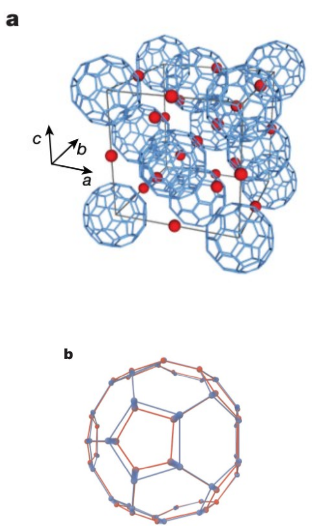
- Inverted Hund's coupling model
- Results
- Analysis
- Conclusion

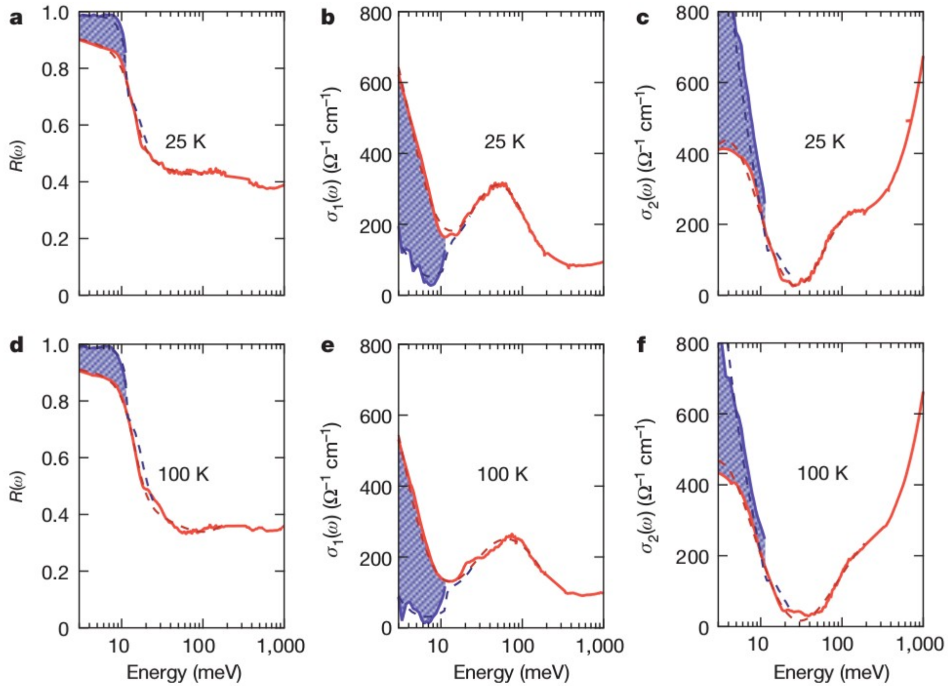
Experiments

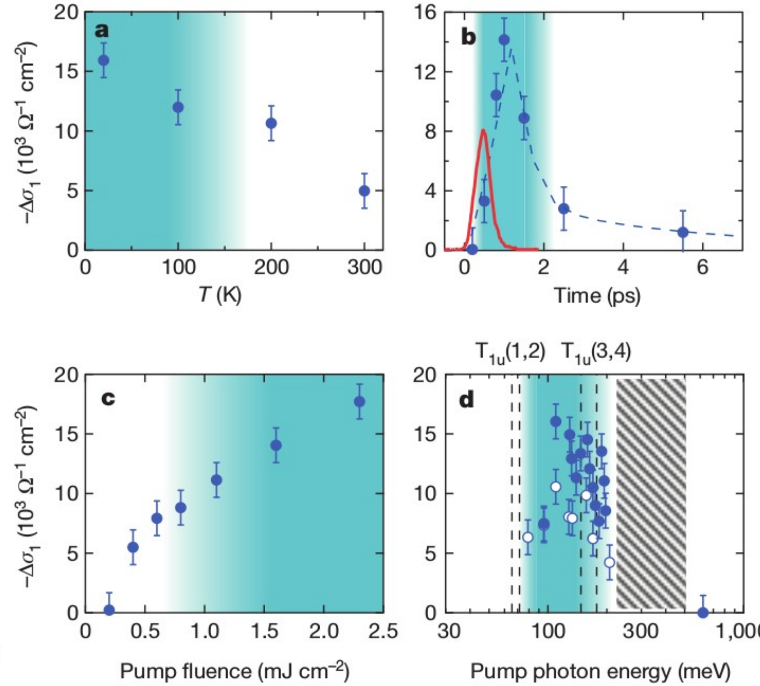
Light-induced superconductivity in K_3C_{60}



Equilibrium

$T_c = 20\,K$

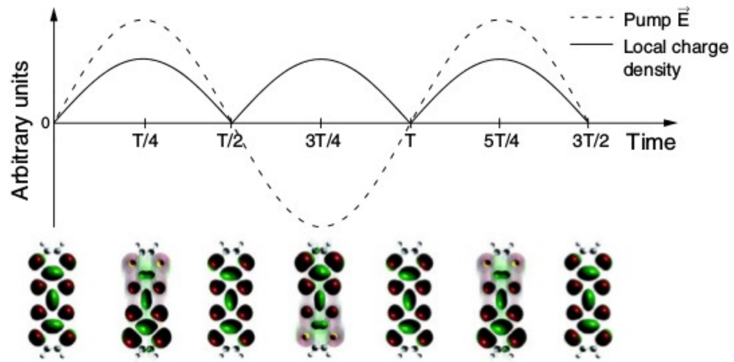




Mitrano et al., Nature (2016)

- Light-induced SC optical property is observed.
- The frequency of the pump light for SC is near that of T_{1u} modes optical phonon of C_{60}.

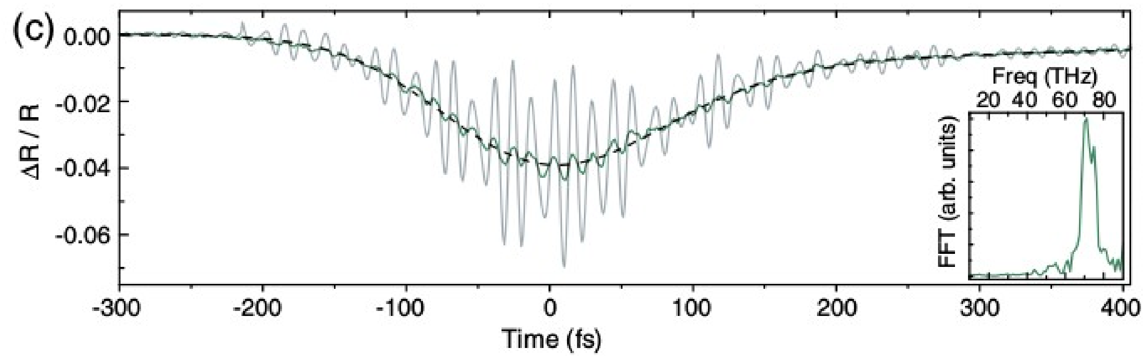
Modulation of Coulomb interaction matrix by THz light

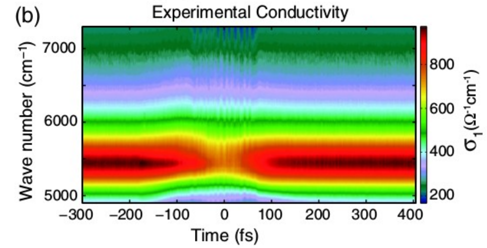


$$\hat{H}_{e\text{-vib}} = \sum_j \hat{n}_j (A_1 q_j + A_2 q_j^2 + \cdots)$$
$$+ \sum_j \hat{n}_{j\uparrow} \hat{n}_{j\downarrow} (B_1 q_j + B_2 q_j^2 + \cdots)$$

$$\hat{H}_{e\text{-vib}} = B_2 q_{\rm IR}^2(\tau) \hat{n}_{j\uparrow} \hat{n}_{j\downarrow} = (C/2) B_2 [1 - \cos(2\Omega_{\rm IR}\tau)] \hat{n}_{j\uparrow} \hat{n}_{j\downarrow}$$

Modulation of Hubbard U in ET-F_2TCNQ

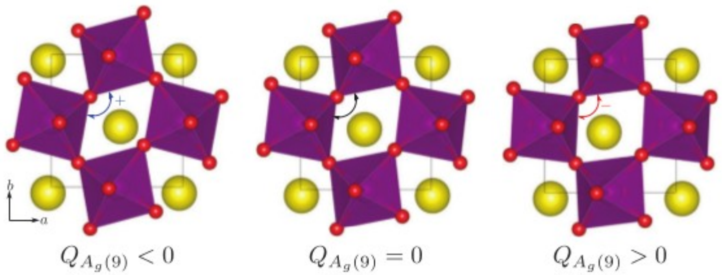


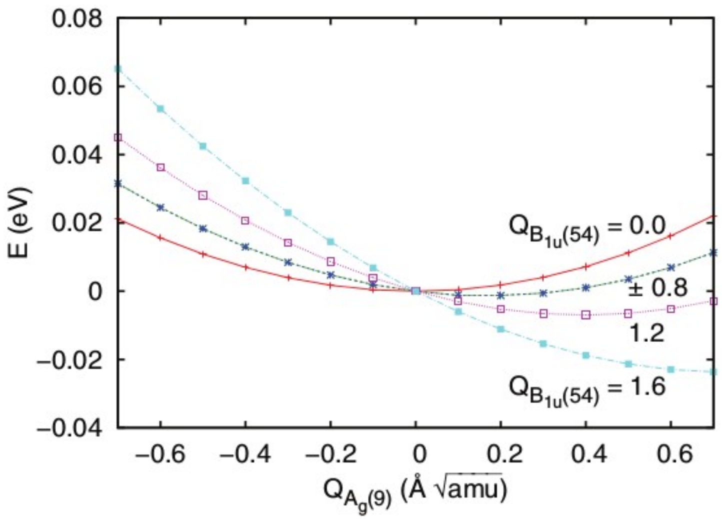


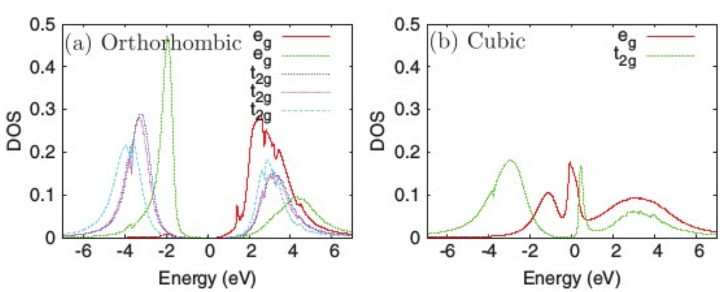
Singla et al.,
Physical Review Letter (2015)

- THz-light induced coherent excitation of a IR mode phonon driven modulation of Coulomb interaction is confirmed.

Nonlinear phononics by THz light







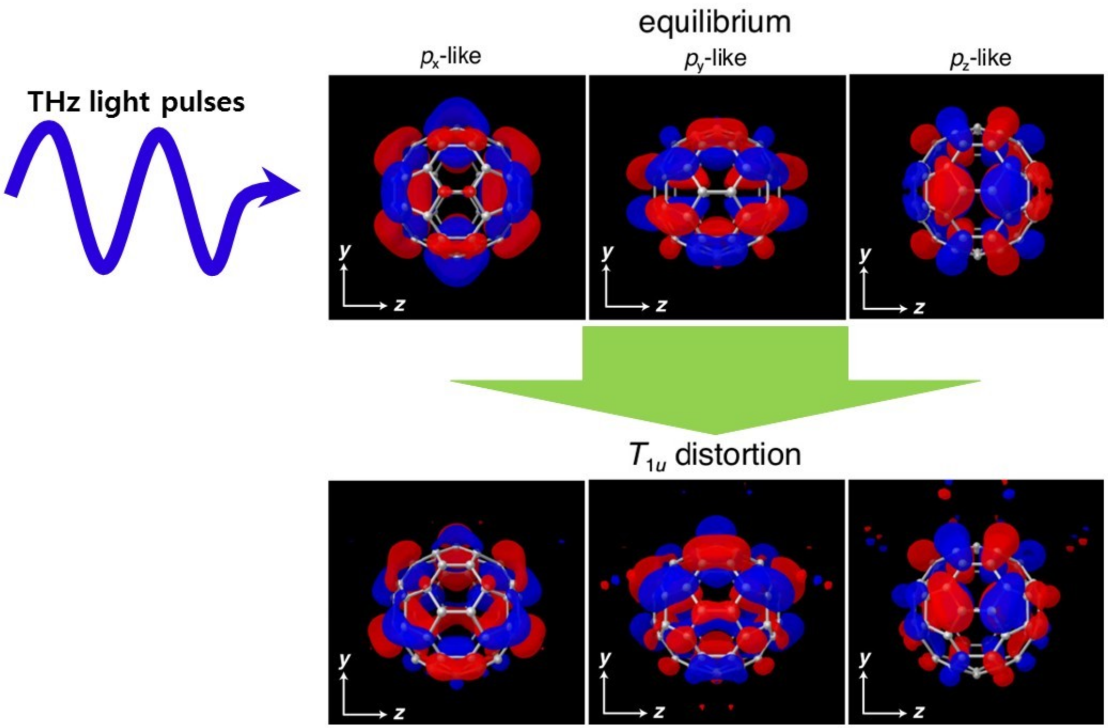
$$V(Q_R, Q_{IR}) = \frac{1}{2}\Omega_R^2 Q_R^2 + \frac{1}{2}\Omega_{IR}^2 Q_{IR}^2 + \frac{1}{3}a_3 Q_R^3 + \frac{1}{4}b_4 Q_{IR}^4 - \frac{1}{2}g Q_R Q_{IR}^2.$$

$$\ddot{Q}_{IR} + \Omega_{IR}^2 Q_{IR} = g\, Q_R Q_{IR} - b_4 Q_{IR}^3 + F(t),$$
$$\ddot{Q}_R + \Omega_R^2 Q_R = \frac{1}{2}g\, Q_{IR}^2 - a_3 Q_R^2.$$

Subedi et al.,
Physical Review B (R) (2014)

- THz-light induced coherent excitation of a IR mode phonon driven structural modulation by Raman mode is possible.

Possible perturbation by pumping $T_{1u}(4)$

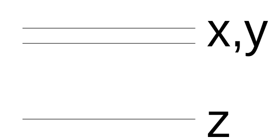


$$Q_{T1u} : \Omega_{IR}$$

$$\bar{U}_a(\tau) = U_a + \Delta U_a(1 - \cos(2\Omega_{\mathrm{IR}}\tau))$$

$$a = \{x, y, z,\}$$

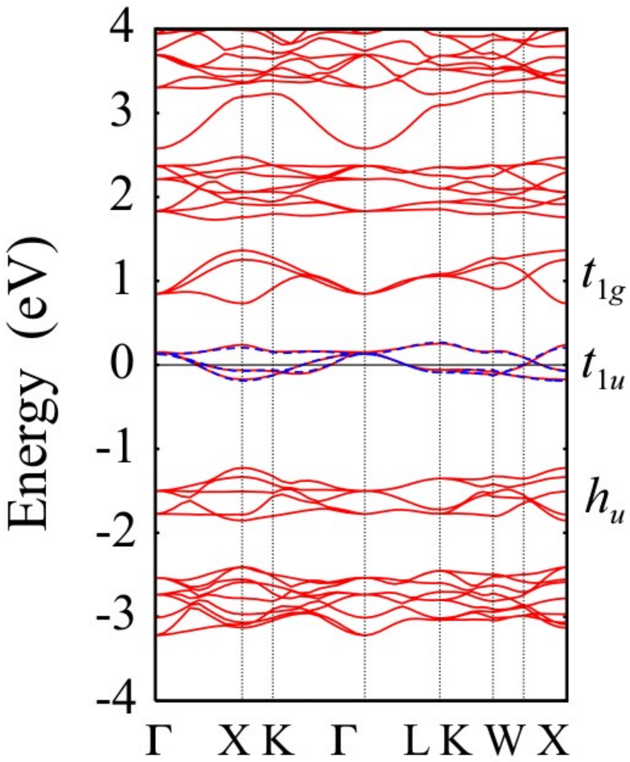
$$q_{Hg} Q_{T1u}^2$$



- Possible perturbation by $T_{1u}(4)$ pumping are
  (a) Modification of Coulomb interaction, and
  (b) H_g Jahn-Teller mode deformation.

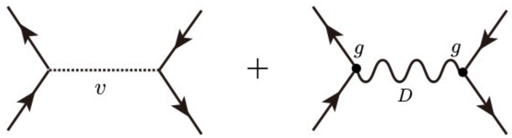
Inverted Hund's coupling model

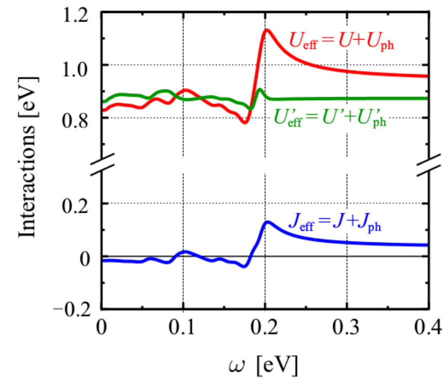
Inverted Hund's coupling model of A_3C_{60}



$$v_{el}=\frac{V}{1-\chi_{cRPA}(\omega)V}$$

$$v_{ph}=g^2_{el-ph,cDFPT}D_{ph,cDFPT}(\omega)$$





K_3C_{60}

$W \sim 0.5\,eV$

$U=U_{el}+V_{el}+U_{ph}\sim W$

$J=J_{el(Hund)}+J_{ph(JT)}\sim -0.04\,W$

Nomura et al.,
Sci. Adv. (2015)
JPCM (2016)

M. Capone et al.,
Review of Modern Physics (2009)

- Construct low energy effective model including el-el, el-ph interaction.
- Sign of J_{eff} is inverted due to el-ph coupling in the H_g JT phonon.
- H_g JT phonons are pairing glues of superconductivity.

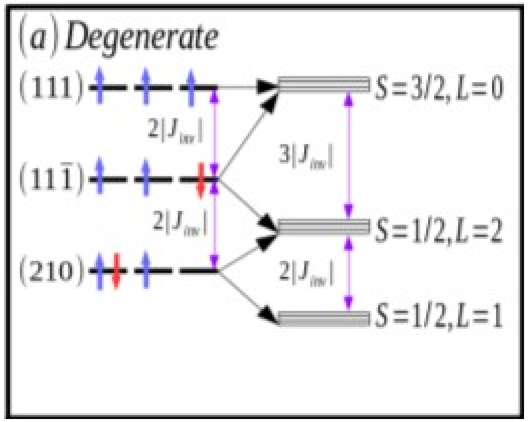
Equilibrium SC of A_3C_{60} in the Inverted Hund's coupling model

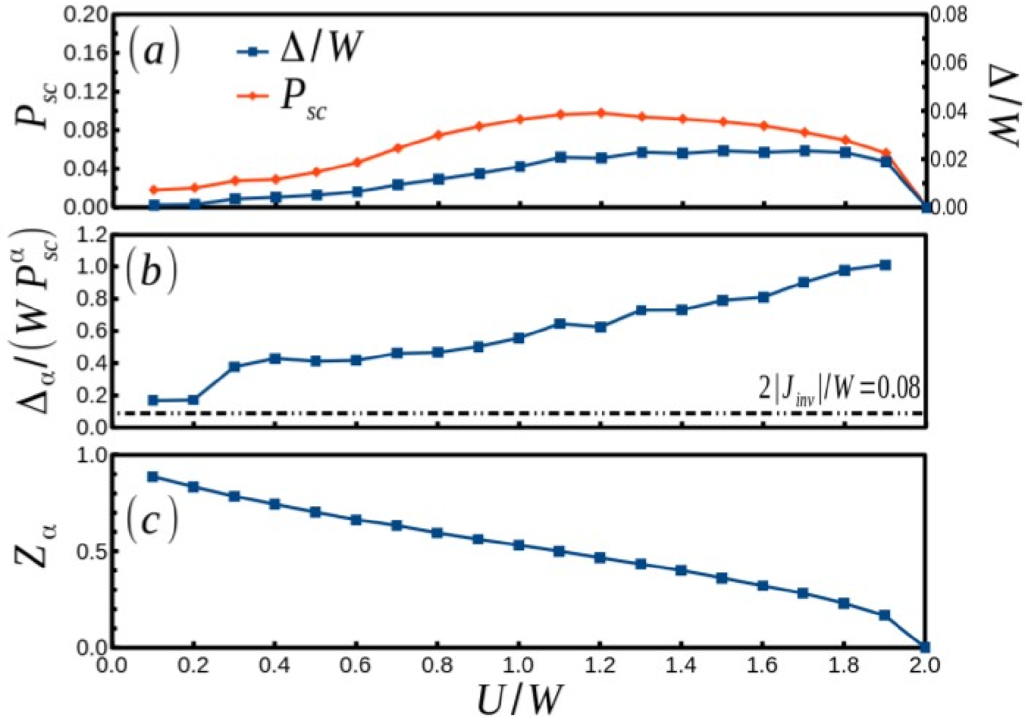
$$H_{\mathrm{int}} = (U - 3J_{\mathrm{inv}})\frac{\hat{N}(\hat{N}-1)}{2} - 2J_{\mathrm{inv}}\vec{S}^2 - \frac{J_{\mathrm{inv}}}{2}\vec{L}^2$$

$$H_{nn} = U\sum_{\alpha}\hat{n}_{\alpha,\uparrow}\hat{n}_{\alpha,\downarrow} + (U-2J_{\mathrm{inv}})\sum_{\alpha\neq\beta}\hat{n}_{\alpha,\uparrow}\hat{n}_{\beta,\downarrow} + (U-3J_{\mathrm{inv}})\sum_{\alpha<\beta,\sigma}\hat{n}_{\alpha,\sigma}\hat{n}_{\beta,\sigma}$$

$$H_{\mathrm{sf}} = -J_{\mathrm{inv}}\sum_{\alpha\neq\beta} d^{\dagger}_{\alpha,\uparrow}d_{\alpha,\downarrow}d^{\dagger}_{\beta,\downarrow}d_{\beta,\uparrow}$$

$$H_{\mathrm{ph}} = J_{\mathrm{inv}}\sum_{\alpha\neq\beta} d^{\dagger}_{\alpha,\uparrow}d^{\dagger}_{\alpha,\downarrow}d_{\beta,\downarrow}d_{\beta,\uparrow}$$



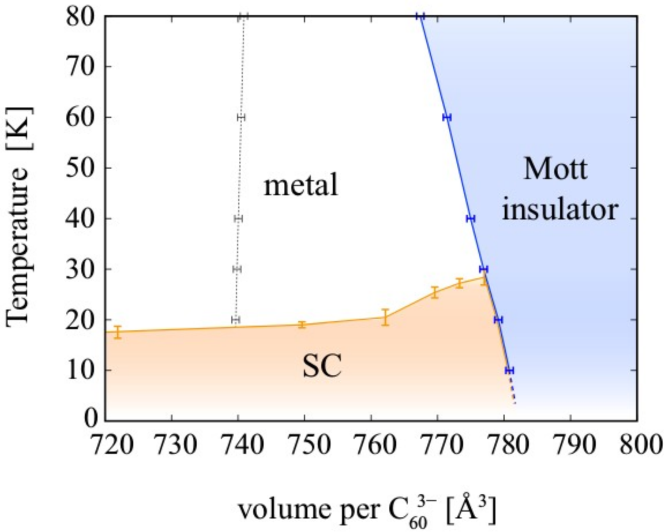


M. Capone et al.,
Review of Modern Physics (2009)

- Low energy effective model including inverted Hund's coupling describes strongly correlated superconductivity of A_3C_{60} in the equilibrium.

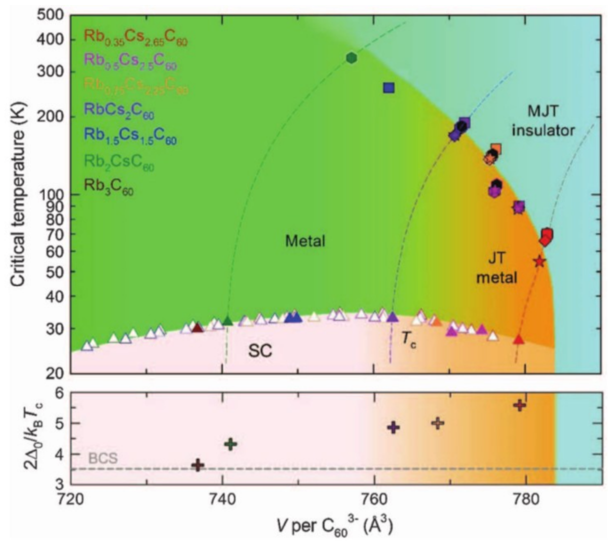
Validity of the inverted Hund's coupling model in fullerides

Extended DMFT



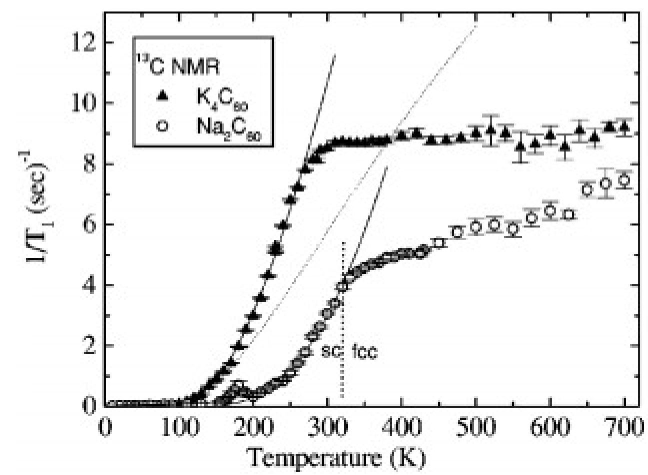
Nomura et al., Sci. Adv. (2015)

Experiments



Zadik et al., Sci. Adv. (2015)

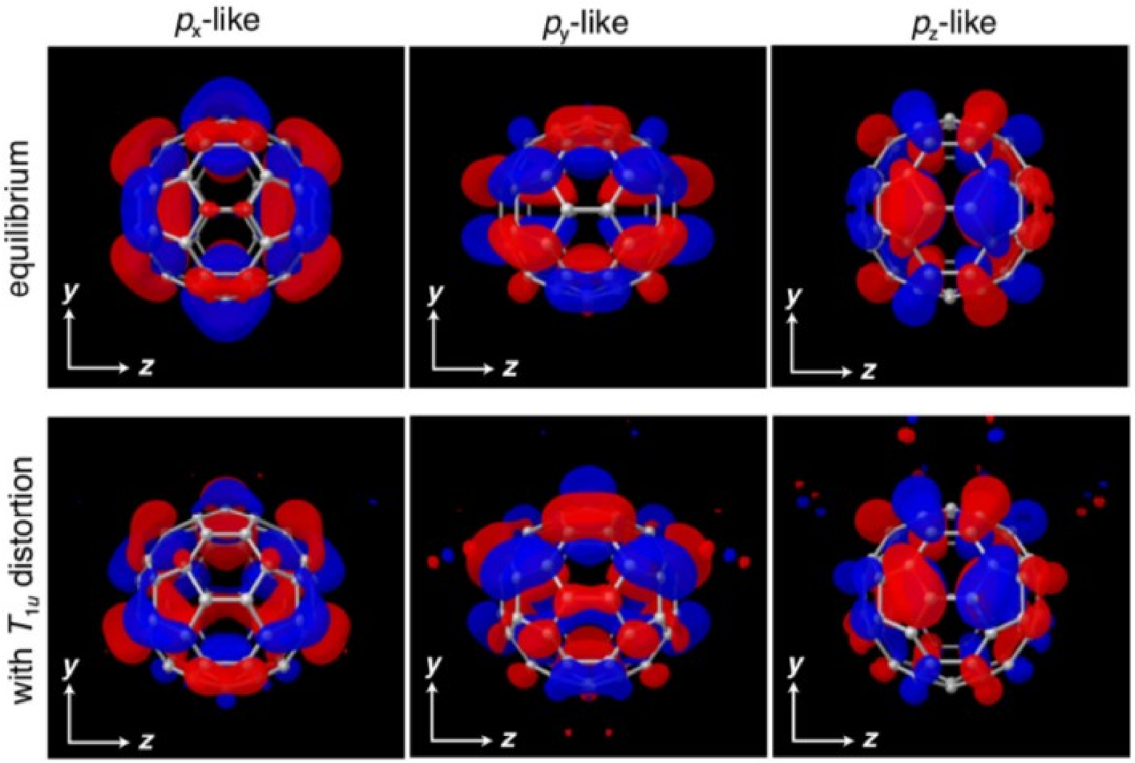
Experiments - NMR



Brouet et al., PRB (2002)

- The first-principle model of inverted Hund's coupling describes experimental phase diagram.
- The spin gap from low-spin to high-spin transition is observed in experiment.

Perturbation in the K_3C_{60}



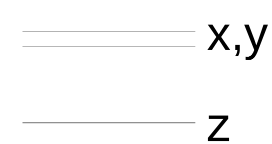
$$Q_{T1u} : \Omega_{IR}$$

$$\bar{U}_a(\tau) = U_a + \Delta U_a(1 - \cos(2\Omega_{\mathrm{IR}}\tau))$$

$$a = \{x, y, z,\}$$

$$H_{dU} = -dU\left(\hat{n}_{x,\uparrow}\hat{n}_{x,\downarrow} + \hat{n}_{y,\uparrow}\hat{n}_{y,\downarrow}\right)$$

$$q_{Hg} Q_{T1u}^2$$



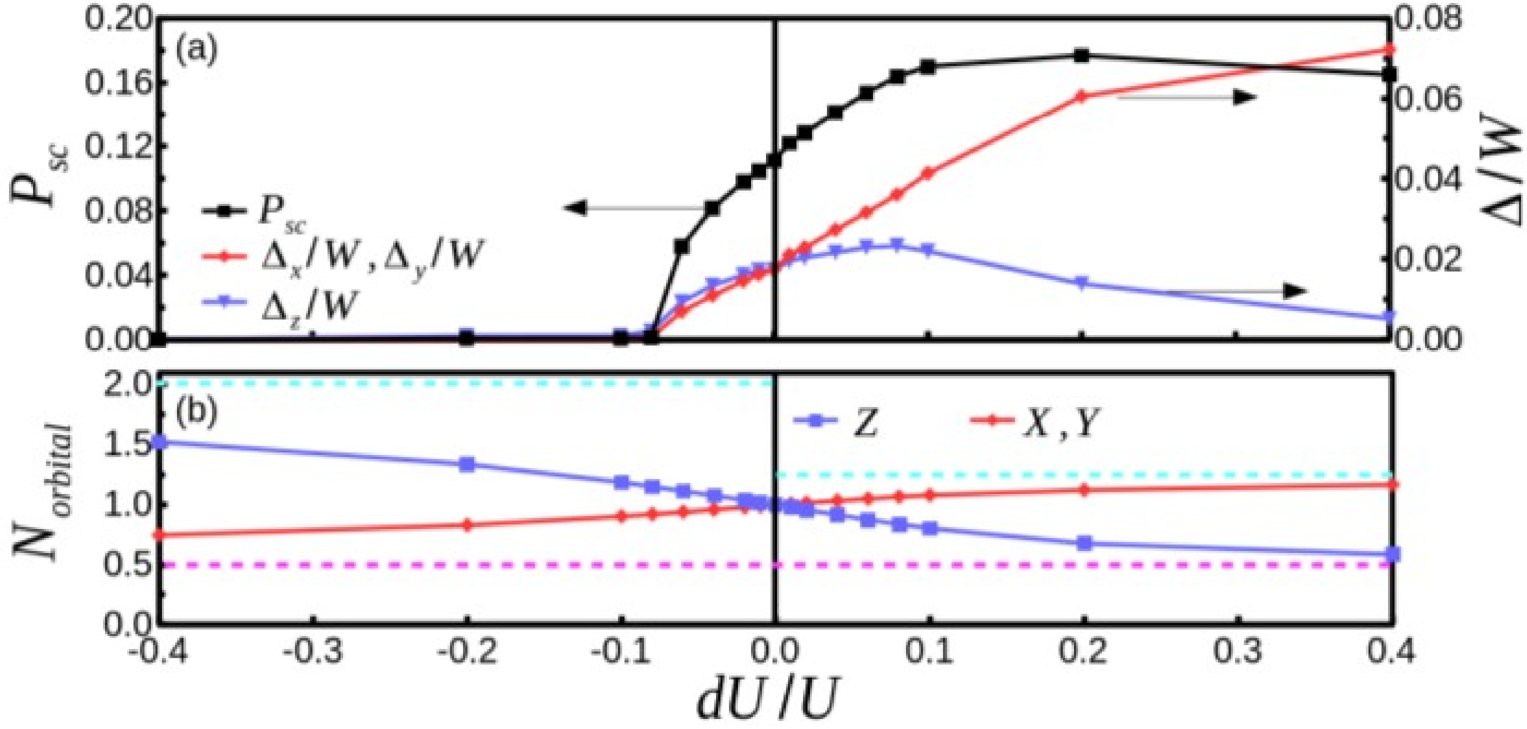
$$H_{\mathrm{CF}} = h_{\mathrm{CF}}\left(\hat{n}_x + \hat{n}_y\right)$$

- From the time scale comparison, $0.5\,Period\left(T_{1u}\right) = 10\,fs \sim 0.01\,ps$, anti-adiabatic deformation of T_{1u} mode is assumed.

$$dU/U \sim 0.04 \qquad h_{CF}/W \sim 0.06$$

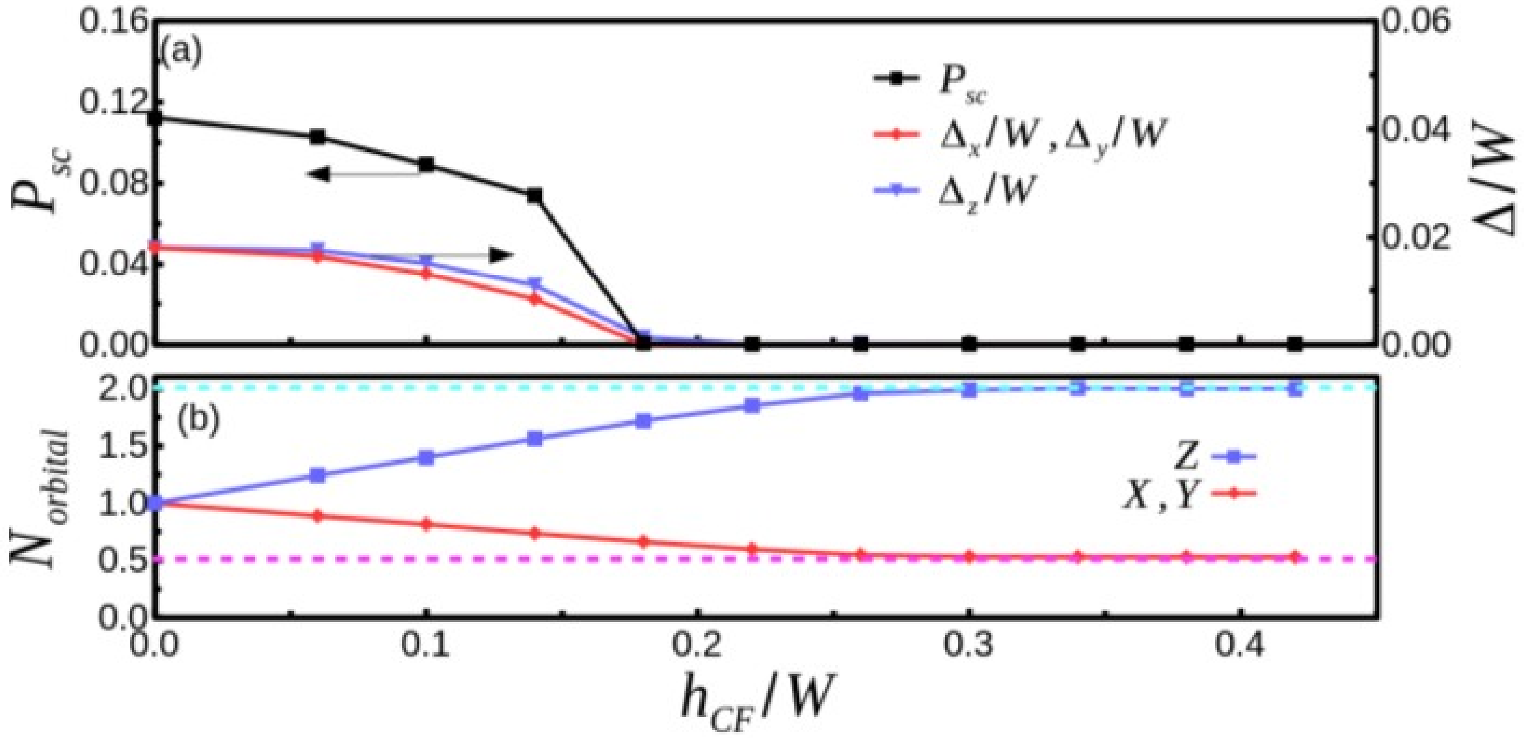
Results

Results : Imbalance of U



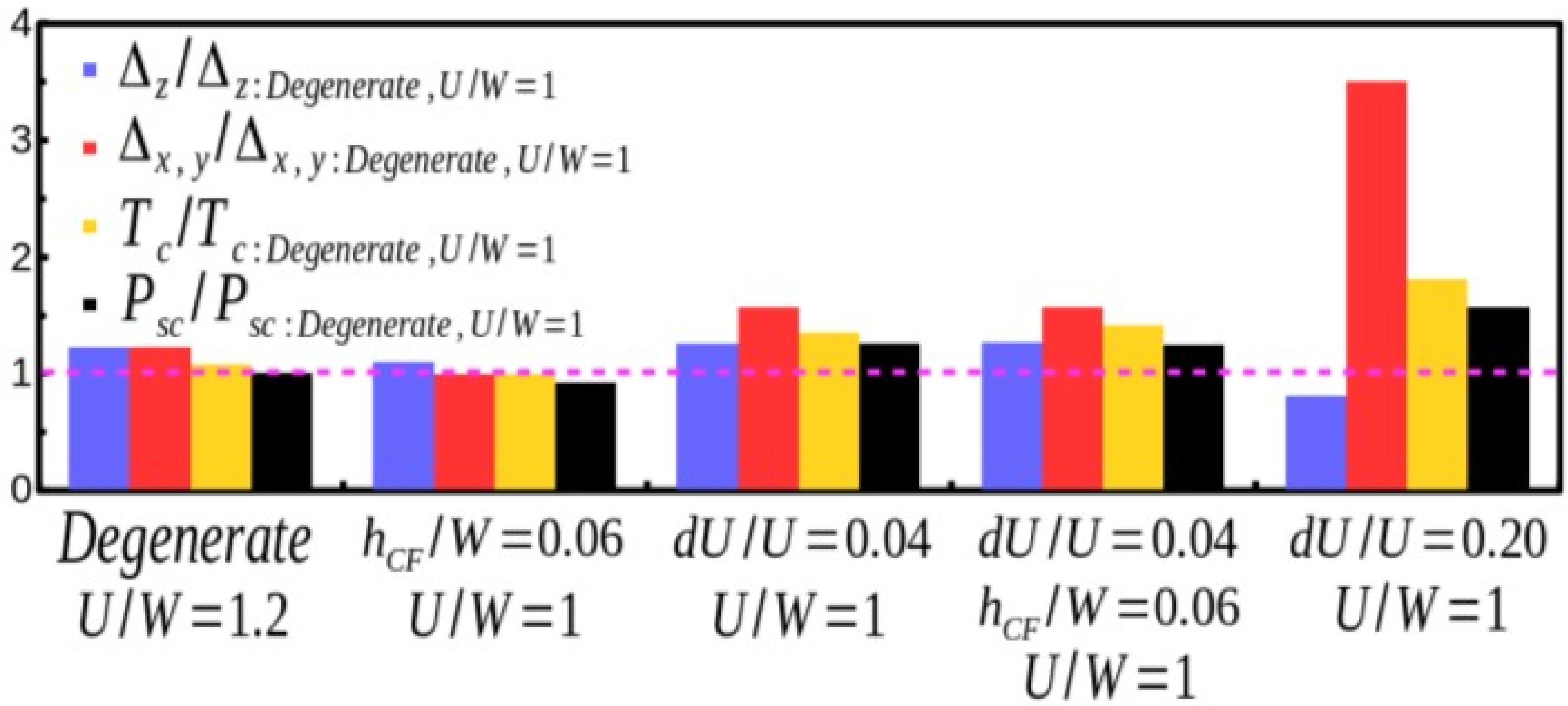
- dU>0 : enhancing SC ($\Delta_{x,y}$ up to factor of 3.5)
- dU<0 : suppresses SC without complete orbital polarization.

Results : Crystal-field



- Crystal-field suppresses SC with complete orbital polarization.

Results : T_c, P_{sc}, and Δ

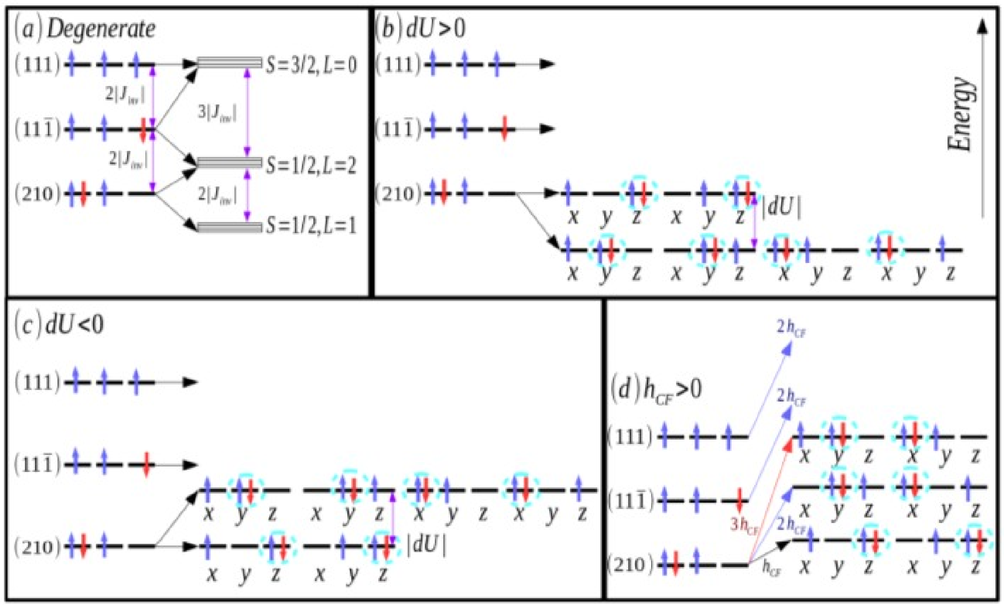


- In the estimated parameters from $T_{1u}(4)$ (2.0 Å$\sqrt{\mathrm{amu}}$)
  T_c was enhanced up to factor of ~1.41.

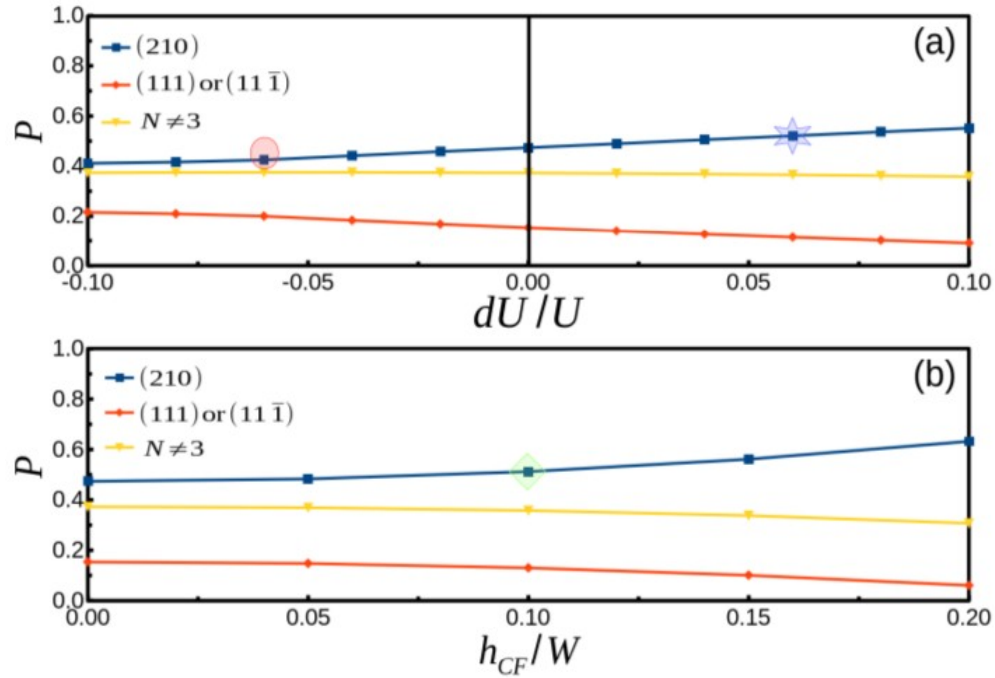
Analysis

Enhancing SC (i): Stabilization of singlet

Multiplets



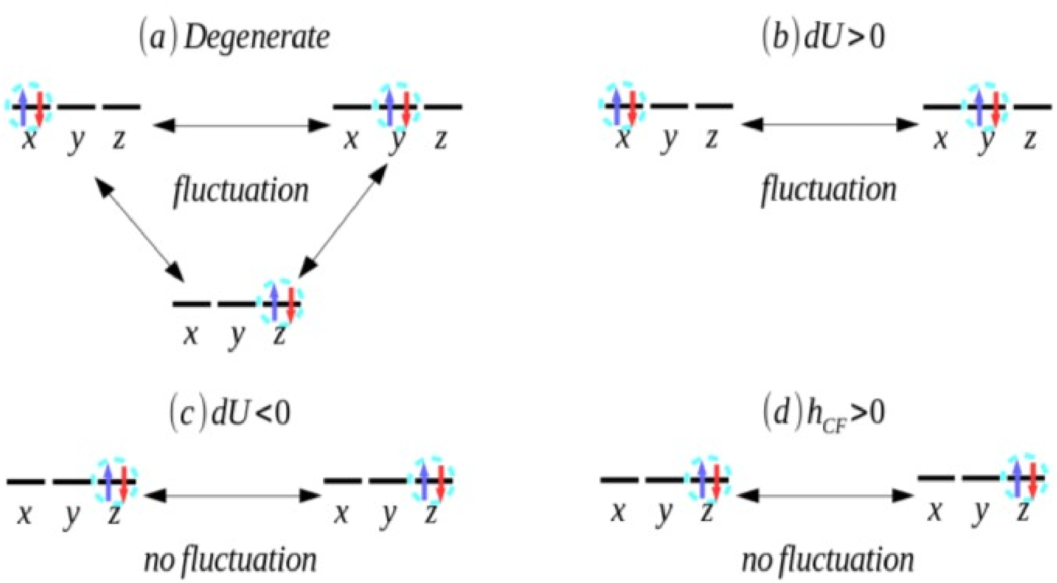
Histogram



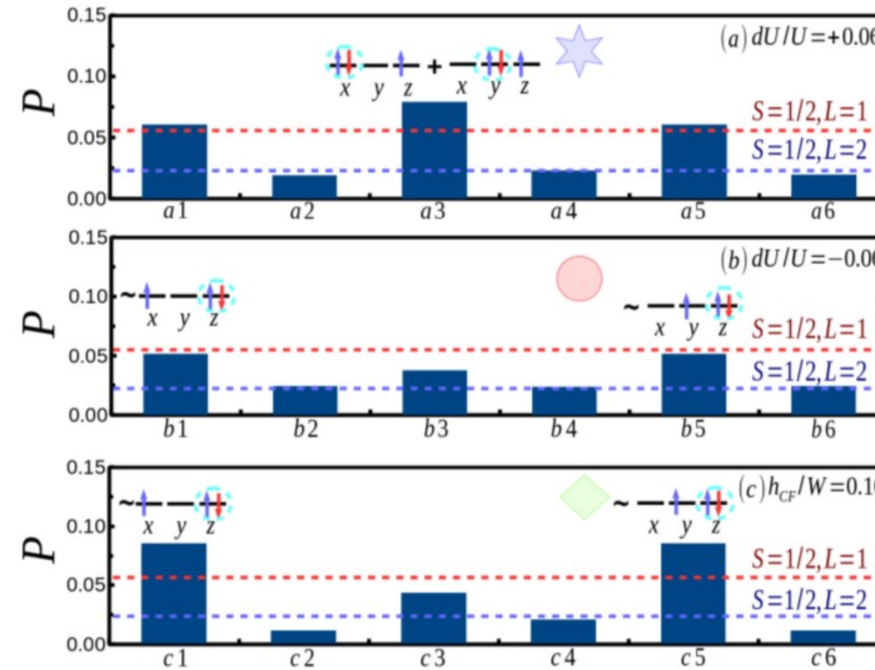
- dU>0 : Singlet state is stabilized & SC is enhanced.
- dU<0 : Singlet state is destabilized & SC is suppressed.
- h_{CF}>0 : Singlet state is stabilized & SC is suppressed?

Enhancing SC (ii): Orbital fluctuation

Possible orbital fluctuation

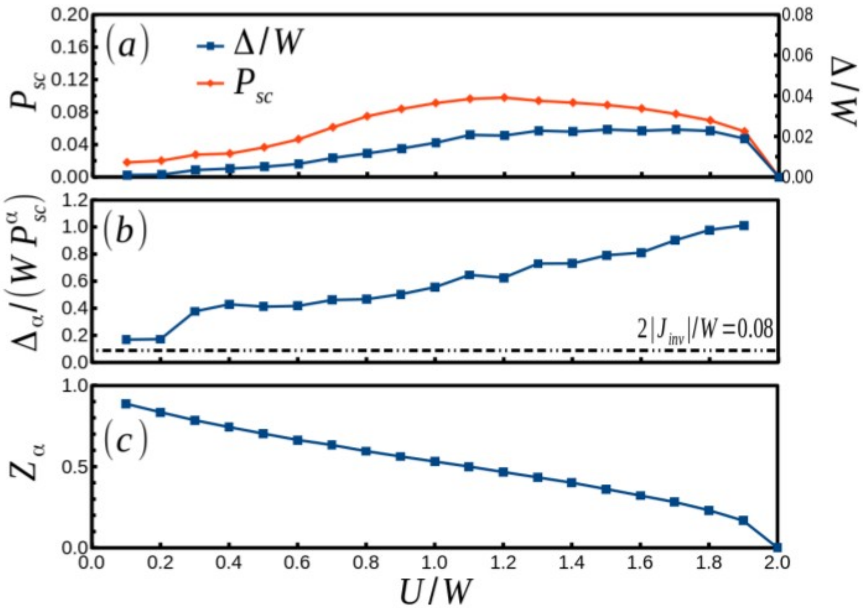


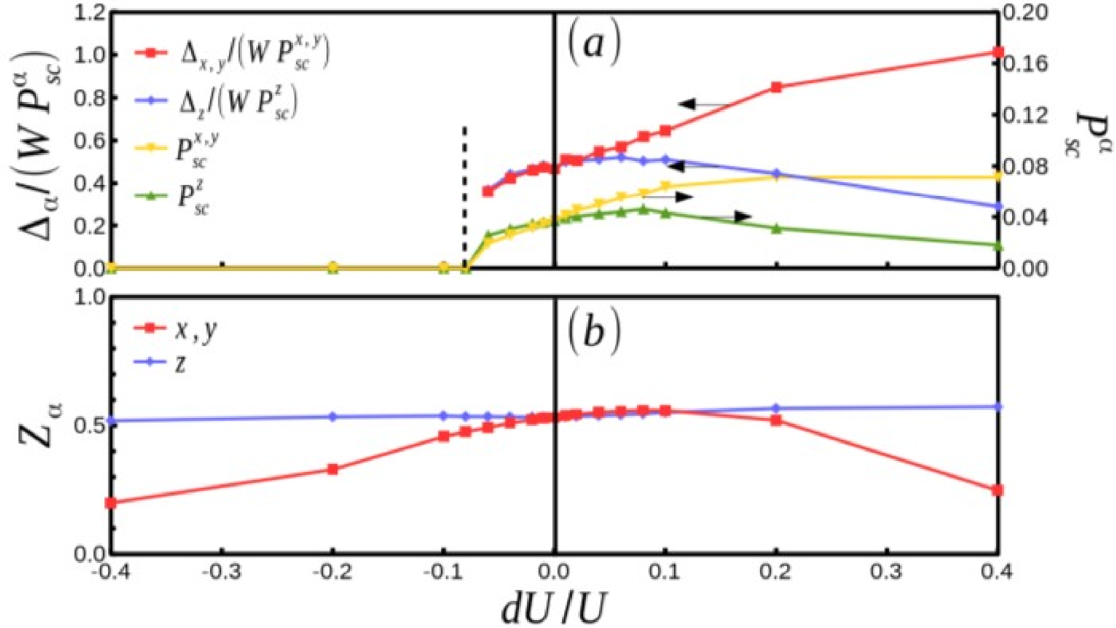
Histogram in the (210) subspace



- Orbital fluctuation is possible in dU>0 case & SC is enhanced.
- Orbital fluctuation is suppressed in dU<0 case & SC is suppressed.
- Orbital fluctuation is suppressed in h_{CF}>0 case & SC is suppressed, even though singlet state is stabilized.

U/W vs dU/U controls





- U/W control (isotropic control of volume) : Strong coupling regime is realized near the metal-insulator transition.
- dU/U control (T_{1u} pumping) : Strong coupling regime is realized without metal-insulator transition. (enters superfluid density)

Conclusion & Questions

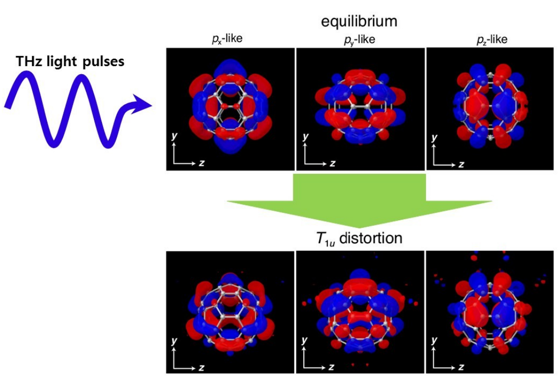
arXiv:1606.05796 (2016)

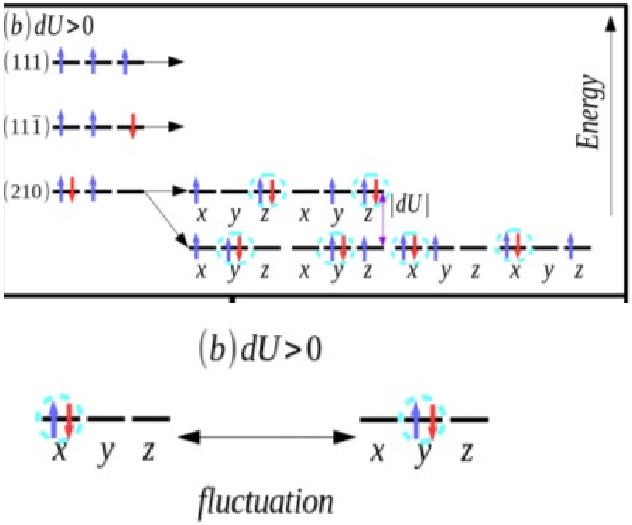
Conclusions

- Perturbation enhancing SC of A_3C_{60} exist.
- This perturbation, dU>0, could be realized by $T_{1u}(4)$ phonon pumping.
- This perturbation satisfies following conditions for enhancing SC of A_3C_{60},

  (a) stabilization of singlet states.

  (b) preserved orbital fluctuation.





Conclusion & Questions

arXiv:1606.05796 (2016)

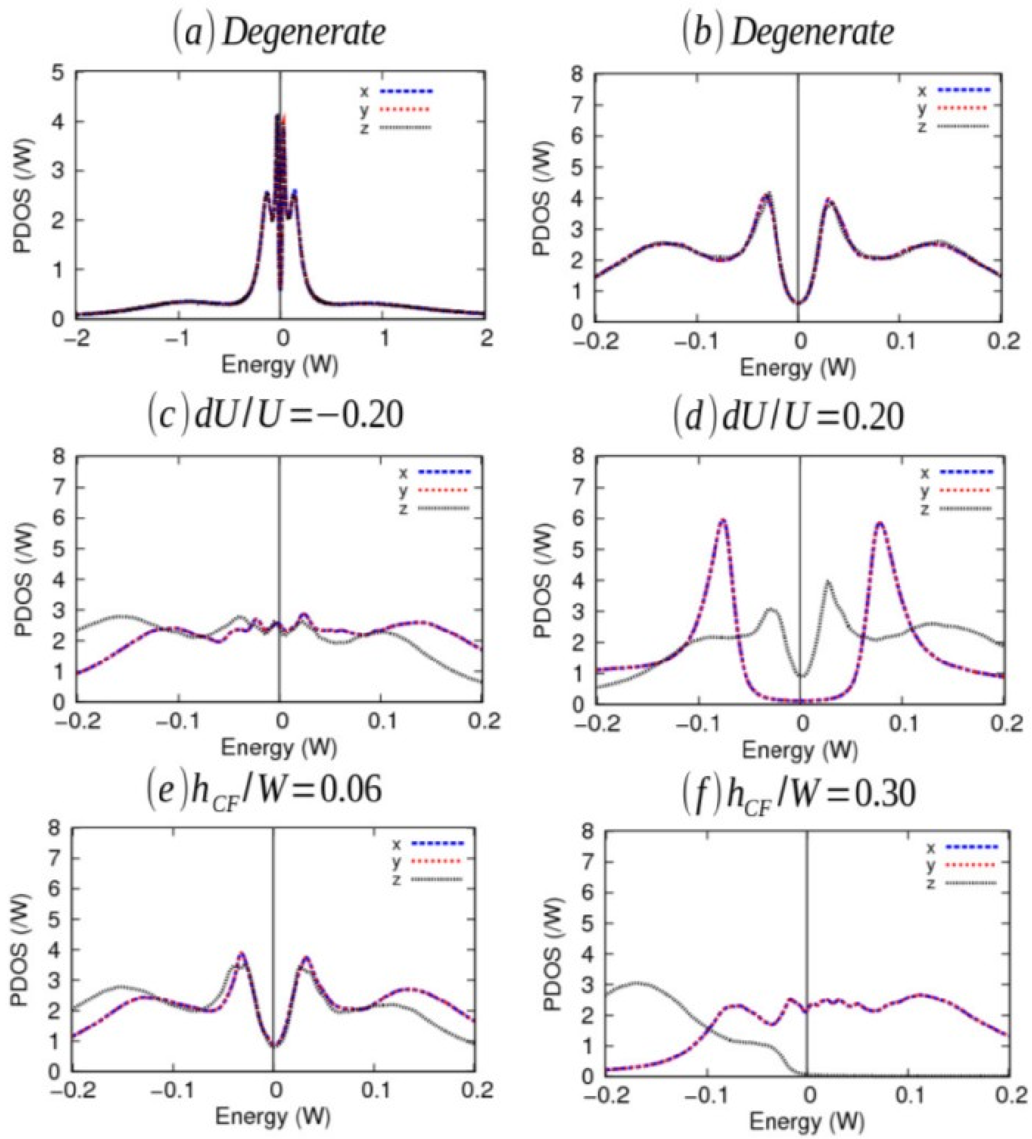
Questions

- Time dependent propagation of states.
- Frequency dependent perturbation beyond Born-Oppenheimer approximation.
- Experimental realization of light-induced structure of C_{60}.

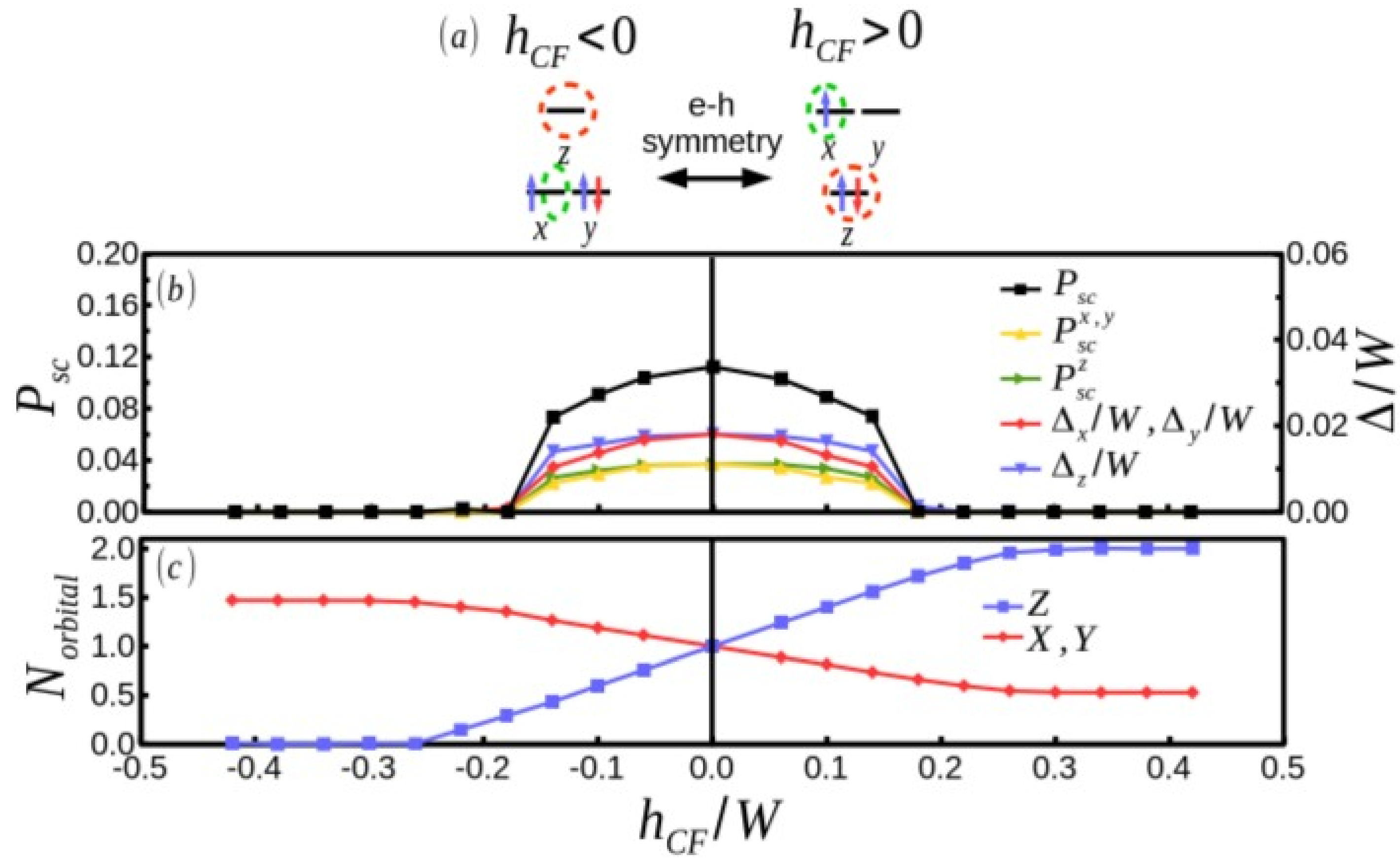
Thanks for your attention

Appendix

Spectral functions

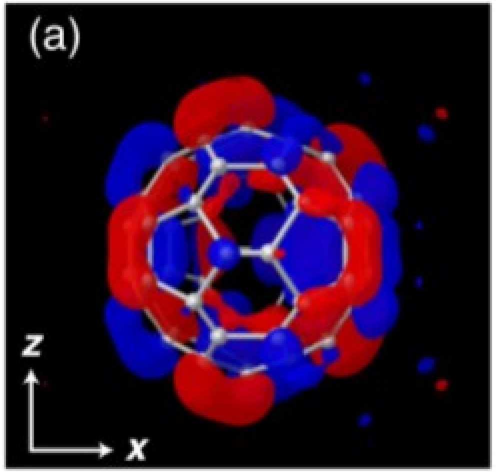


Negative crystal field

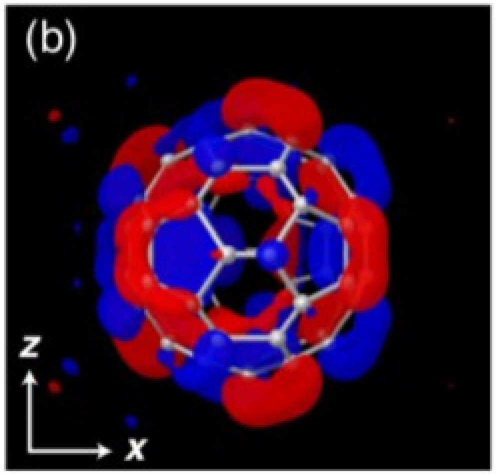


x and y orbital in the T_{1u} pumped structure

x



y



Multiplet states