Enhancing Superconductivity of $A_3C_{60}$ fullerides


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Experiments
Light-induced superconductivity in $K_3C_{60}$

- 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 $C_{60}$.

Mitrano et al., Nature (2016)

Equilibrium $T_c = 20 \, K$
Modulation of Coulomb interaction matrix by THz light

\[ \hat{H}_{e-vib} = \sum_j \hat{n}_j (A_1 q_j + A_2 q_j^2 + \cdots) \]
\[ + \sum_j \hat{n}_j \uparrow \hat{n}_j \downarrow (B_1 q_j + B_2 q_j^2 + \cdots) \]
\[ \hat{H}_{e-vib} = B_2 q_{IR}^2 (\tau) \hat{n}_j \uparrow \hat{n}_j \downarrow = (C/2) B_2 [1 - \cos(2\Omega_{IR}\tau)] \hat{n}_j \uparrow \hat{n}_j \downarrow \]

- 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 $T_{1u}(4)$

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

\[
Q_{T1u} \cdot \Omega_{IR} \\
\tilde{U}_a(t) = U_a + \Delta U_a(1 - \cos(2\Omega_{IR}t)) \\
a = \{x, y, z,\} \\
q_{Hg} Q_{T1u}^2 \quad \underline{x, y} \quad \underline{z}
\]
Inverted Hund's coupling model
Inverted Hund’s coupling model of $A_3C_{60}$

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

\[ v_{el} = \frac{V}{1 - \chi_{\text{crPA}}(\omega)V} \]
\[ v_{ph} = g_{el-ph, cDFPT}^2 D_{ph, cDFPT}(\omega) \]
\[ U = U_{\text{el}} + V_{\text{el}} + U_{ph} \sim W \]
\[ J = J_{el(Hund)} + J_{ph(JT)} \sim -0.04W \]
Equilibrium SC of A$_3$C$_{60}$ in the Inverted Hund's coupling model

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

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

\[H_{sf} = -J_{\text{inv}} \sum_{\alpha \pm \beta} d_{\alpha,\uparrow}^{\dagger} d_{\alpha,\downarrow} d_{\beta,\uparrow}^{\dagger} d_{\beta,\downarrow}^{\dagger}\]

\[H_{ph} = J_{\text{inv}} \sum_{\alpha \neq \beta} d_{\alpha,\uparrow}^{\dagger} d_{\alpha,\downarrow}^{\dagger} d_{\beta,\uparrow} d_{\beta,\downarrow}\]

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

- Low energy effective model including inverted Hund's coupling describes strongly correlated superconductivity of A$_3$C$_{60}$ in the equilibrium.
Validity of the inverted Hund's coupling model in fullerides

- 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.


Brouet et al., PRB (2002)
Perturbation in the $K_3C_{60}$

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

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

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

$$H_{dU} = -dU(\hat{n}_x, \hat{n}_x, \downarrow + \hat{n}_y, \hat{n}_y, \downarrow)$$

$$q_{Hg} Q_{T1u}^2 \quad \begin{array}{c} x, y \\ \hline \end{array}$$

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

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

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

- In the estimated parameters from $T_{1u}(4) \left( 2.0 \text{ Å}\sqrt{\text{amu}} \right)$, $T_c$ was enhanced up to factor of $\sim 1.41$. 
Analysis
Enhancing SC (i): Stabilization of singlet

- $dU>0$: 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

- 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)
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.
Questions

- Time dependent propagation of states.
- Frequency dependent perturbation beyond Born-Oppenheimer approximation.
- Experimental realization of light-induced structure of C₆₀.
Thanks for your attention
Appendix
Spectral functions

(a) Degenerate

(b) Degenerate

(c) $dU/U = -0.20$

(d) $dU/U = 0.20$

(e) $h_{CF}/W = 0.06$

(f) $h_{CF}/W = 0.30$
Negative crystal field
x and y orbital in the $T_{1u}$ pumped structure
Multiplet states