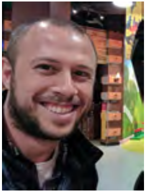
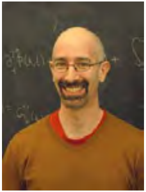
Electronic Squeezing of Pumped Phonons: Negative U and Transient Superconductivity

[1]D.M. Kennes

with [1]E.Y. Wilner, [2]D.R. Reichman and [1]A.J. Millis

[1]Columbia University, Department of Physics
[2]Columbia University, Department of Chemistry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Controlling Electronic Properties with Light?

e.g. superconducting-like response in K_3C_{60}:

LETTER

doi:10.1038/nature16522

Possible light-induced superconductivity in K_3C_{60} at high temperature

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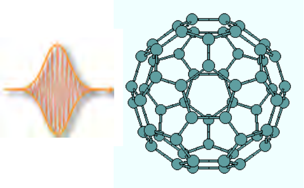

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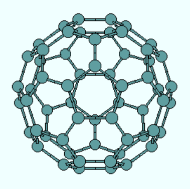

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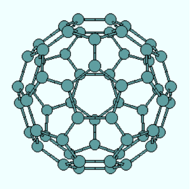
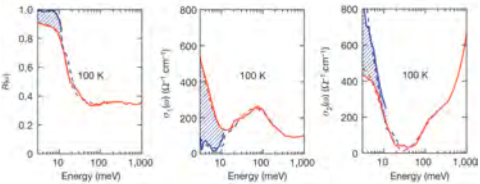


superconduct.-like response (above T_c):



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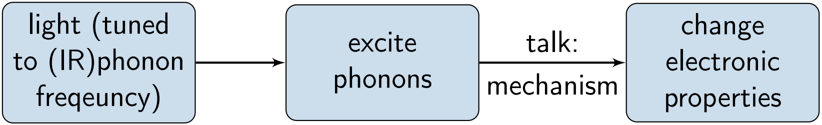


- phonon in K_3C_{60} experiment coupled DIRECTLY to electrons only via gradient ($T_{1u} \otimes T_{1u}$ no coupling)

$$H_{\mathrm{ep}} = \vec{q} \cdot \sum_k \Psi_k^\dagger \vec{\nabla} \Psi_k$$

  very weak!

- $\Rightarrow$ seek non-linear coupling?

Non-Linear Phononics

different proposals

- Light-induced change in lattice structure via quartic (IR)phonon–(Raman)phonon coupling [Subedi, et. al. PRB **89**, 220301(R) (2014)]
- Periodic modulation of interaction by direct coupling [Singla, et. al. PRL **115**, 187401 (2015)]
- ⇒ variant applied to changes in shape of C_{60} molecule gives orbital dependence of U promoting SC [Kim, et. al. PRB **94**, 155152 (2016)]
- dynamical modulations of other phonons via (IR)phonon–(Raman)phonon coupling [Knap, et. al. PRB **94**, 214504 (2016); M. Babadi et. al. arXiv:1702.02531)]
- Interesting different proposal: effective cooling (not Phonons) [Nava, et. al. arXiv:1704.05613]

Non-Linear Phononics

different proposals: common ground

[Subedi, et. al.; Singla, et. al.; Kim, et. al.; Knap, et. al.] light induced changes in electronic properties (distribution func., electron or electron-phonon Hamiltonian)

here: what about the phonon energetics?

the phonon properties depend on the electronic state: could this (partially) be the transduction mechanism?

simple model (symmetry allowed):

$$H_{\mathrm{e-ph}} = gK \sum_i \hat{n}_i \hat{Q}_i^2$$

with small dimensionless coupling $g \sim 0.05 - 0.1$.

$\Rightarrow$ restoring force for phonon depends on electron density $\hat{n}$

simple model

$$H = H_{\mathrm{e}} + H_{\mathrm{e-ph}} + H_{\mathrm{ph}}$$

electron-phonon coupling

$$H_{\mathrm{e-ph}} = gK \sum_i \hat{n}_i \hat{Q}_i^2$$

bare phonon Hamiltonian

$$H_{\mathrm{ph}} = \sum_i \frac{K}{2} \hat{Q}_i^2 + \frac{1}{2M} \hat{P}_i^2 = \omega_0 \sum_i \left(b_i^\dagger b_i + \frac{1}{2} \right)$$

phonon energetics

$$H_{\mathrm{e-ph}} \quad \Rightarrow \quad K \to K' = (1 + 2g\,\hat{n}_i) K$$

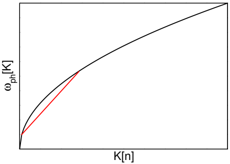
$$\omega_0 \to \omega_0 \sqrt{1 + 2g\,\hat{n}_i}$$

$\omega_0 \to \omega_0\sqrt{1+2g\hat{n}_i}$

- shift in oscillator frequency: energy of m-th eigenstate $E = \omega_0(m+\frac{1}{2})$ depends on local electron occupancy
- electron pairing energy: $U_{\rm eff} = E(n=2) + E(n=0) - 2E(n=1)$

$$U_{\rm eff}[m] = U + \left(m+\frac{1}{2}\right)\omega_0\left(\sqrt{1+4g}+1-2\sqrt{1+2g}\right)$$
$$\stackrel{g\ll 1}{\approx} U - \left(m+\frac{1}{2}\right)\omega_0 g^2$$

- effective interactions are:
  - negative because square root is concave down
  - increases linearly with number of phonons m
  - proportional to g^2



Minimal Model: Hubbard Model + Optical Phonons

$$H = -\sum_{ij\sigma} J_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U_{\mathrm{elec}} \sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow} + \sum_i \left(\frac{K}{2}\hat{x}_i^2 + \frac{1}{2M}\hat{p}_i^2 \right)$$

$$+gK \sum_i \hat{n}_i \hat{x}_i^2$$

electron-phonon coupling via electron density dependence of osc. stiffness

treating $\hat{n}\hat{x}^2 \sim \hat{n}\left(b^{\dagger} + b\right)^2$ difficult

(unitary) squeezing transform $e^S H e^{-S}$ with $e^{\hat{S}} = e^{\frac{i}{2}\sum_j \zeta_j \left(\hat{x}_j \hat{p}_j + \hat{p}_j \hat{x}_j\right)}$ and

$$\zeta_j = -\frac{1}{4} \ln\left[1 + 2g\left(\hat{n}_{j\uparrow} + \hat{n}_{j\downarrow}\right)\right]$$

$$\Rightarrow H_{\mathrm{eff}} = -\sum_{\langle i,j \rangle \sigma} J^{\star}_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U_{\mathrm{elec}} \sum_i n_{i\uparrow} n_{i\downarrow} + \omega_0 \sqrt{1 + 2g\hat{n}_i} \left(\beta_i^{\dagger}\beta_i + \frac{1}{2} \right)$$

Onsite Terms: Expand in g

$$\omega_0\sqrt{1+2g\hat{n}_i}\left(\beta_i^\dagger\beta_i+\frac{1}{2}\right) \approx \omega_0\left(\beta_i^\dagger\beta_i+\frac{1}{2}\right) + \frac{g\omega_0}{2}\left(1-\frac{g}{2}\right)\left(2\beta_i^\dagger\beta_i+1\right)n_{i\sigma} - \frac{g^2\omega_0}{2}\left(2\beta_i^\dagger\beta_i+1\right)n_{i\uparrow}n_{i\downarrow}$$

Three contribution:

1. usual phonon
2. onsite potential (will lead to effective disorder)
3. onsite attraction (will promote SC in the right parameter regime)

Hopping Term: Complicated

$$\tilde{H}_{hop}\left[\{n_i, n_j\}\right] = -\sum_{ij\sigma} J_{ij} c^\dagger_{i\sigma} c_{j\sigma} e^{i(\zeta[n_i+1]-\zeta[n_i])\mathcal{O}_i} e^{i\left(\zeta[n_j-1]-\zeta[n_j]\right)\mathcal{O}_j}$$

$$\text{with:} \quad \mathcal{O}_j = \frac{\beta^\dagger_j \beta^\dagger_j - \beta_j \beta_j}{2}$$

- hopping process involves pair creation or destruction of phonons
- because $\mathcal{O}$ is quadratic in phonon operators the usual Feynman disentangling does not work
- checked in exactly solvable 2-site version of model $\Rightarrow$ assume light field puts oscillators in coherent state that instantly looses coherence between different m sectors
- $\zeta[n_i \pm 1] - \zeta[n_i] = \pm \frac{g}{2} + \mathcal{O}(g^2 \hat{n})$ and expanding to leading contribution (with non-zero expectation value):

$$J^\star = e^{-\frac{g^2}{8}\left(n_{\mathrm{B}}^2 + 2n_{\mathrm{B}} + 1\right)} J.$$

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$$\beta_i^\dagger \beta_i^\dagger - \beta_i \beta_i$$

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_m \frac{\alpha^m}{\sqrt{m!}} |m\rangle$$

$$\rho_\alpha = |\alpha\rangle\langle\alpha| = e^{-|\alpha|^2} \sum_{m,m'} \frac{\alpha^m \alpha^{m'}}{\sqrt{m! m'!}} |m\rangle\langle m'|$$

$$\rho_{\mathrm{deph}} = e^{-|\alpha|^2} \sum_m \frac{\alpha^{2m}}{m!} |m\rangle\langle m|$$

ce

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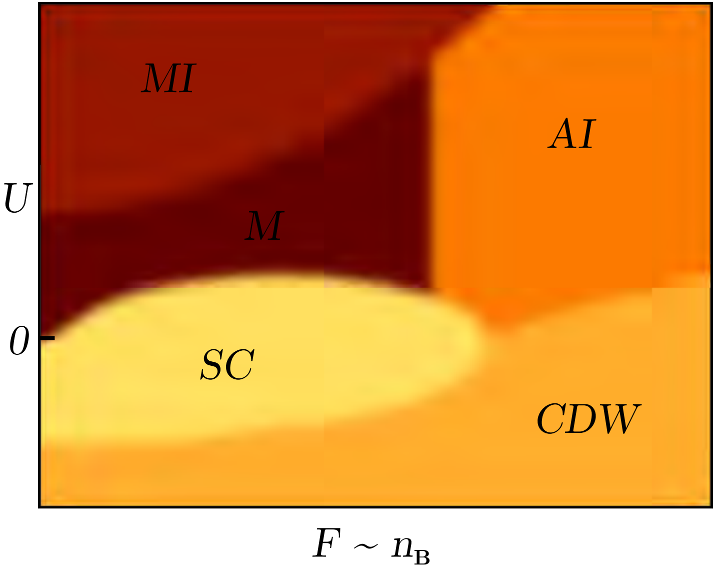
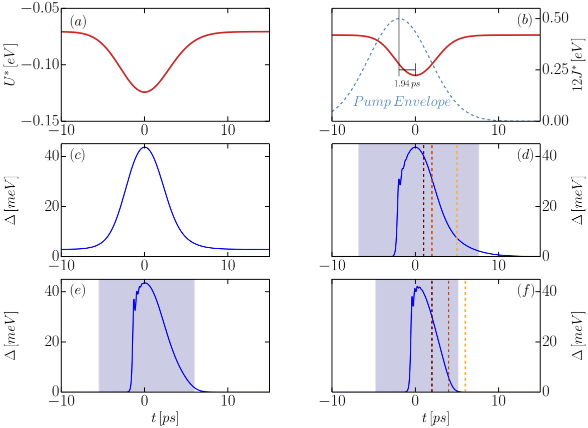
"Phases"



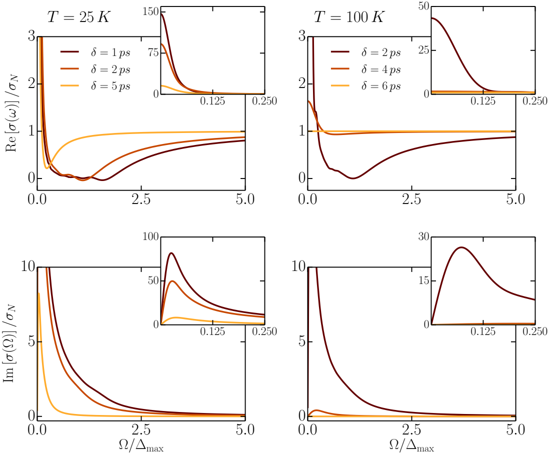
Experimental Parameters

- neglect disorder
- BCS treatment (+ phenomenological electron and phonon relaxation mechanism)



Optical Conductivity: Adiabatic Approximation

- define as Fourier transform of $j(t) = \int dt' \sigma(t, t') E_{\rm probe}(t')$ with $E_{\rm probe}(t) = E_P \delta(t - t_D)$
- adia. approx. for conductivity: $\sigma(T, t_{rel}) \approx \sigma^{\Delta(T)}_{equil}(t_{rel})$



Summary and Outlook

so far:

- new generic mechanism connecting phonon drive to changed interaction parameters
- plausible for light induced SC, but our value of g is a factor of 2-4 to large compared to the one estimated from experiments!
- see also M.A. Sentef [Phys. Rev. B **95**, 205111 (2017)]

many open issues:

- Energy flow and heating?
  - DMRG and perturbation theory
- calculation of the non-equilibrium conductivity (not adiabatic)
  - arXiv:1703.07248
- inclusion of other phonon modes
- interplay with other mechanisms
- Application to other materials (optical control of Mott insulators)

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