

# Dynamical Cooper pairing in non-equilibrium electron-phonon systems

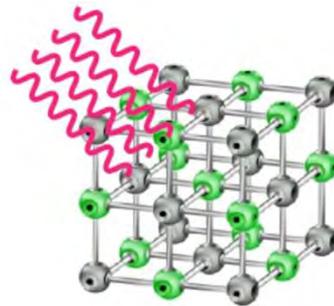
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**Tao Shi** (MPQ), **Ignacio Cirac** (MPQ)



\$\$ NSF DMR, Simons Foundation,  
MURI QUANTUM MATTER, ITS ETH

# Outline

Physical picture: enhanced electron-phonon interaction in systems with driven phonons

Floquet-Migdal-Eliashberg analysis

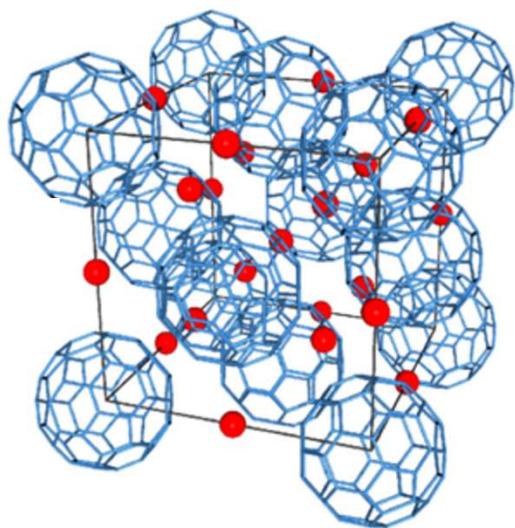
Optical response of photo-excited superconductor:  
Nonequilibrium dynamics with variational states

Conclusions

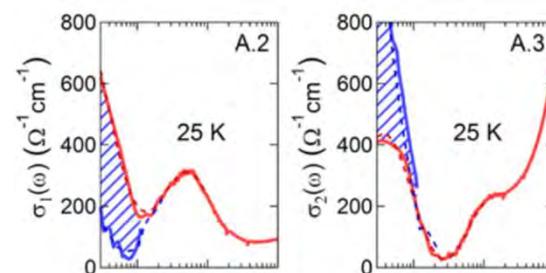
# Motivation: photoinduced superconductivity in K3C60

M. Mitrano et al.,  
Nature 530, 461 (2016)

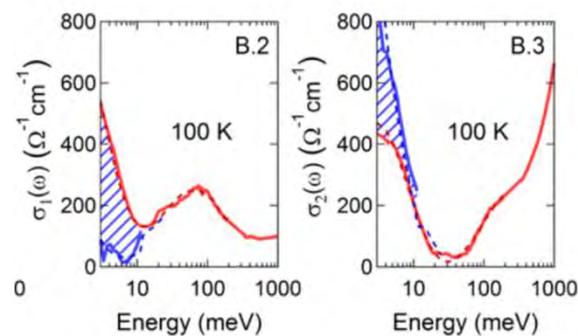
$T_c=20\text{K}$



## Response of photoexcited system

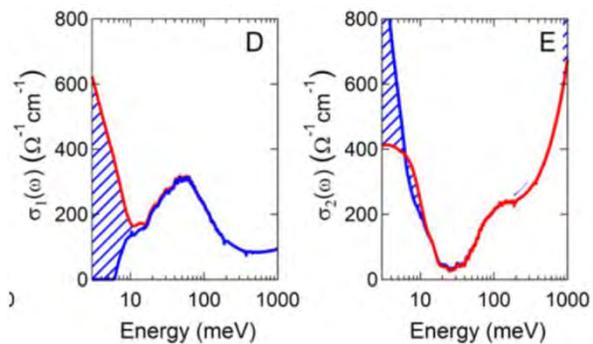


$T=25\text{ K}$



$T=100\text{ K}$

Equilibrium:  $T=25\text{K}$  (red) and  $T=10\text{ K}$  (blue)



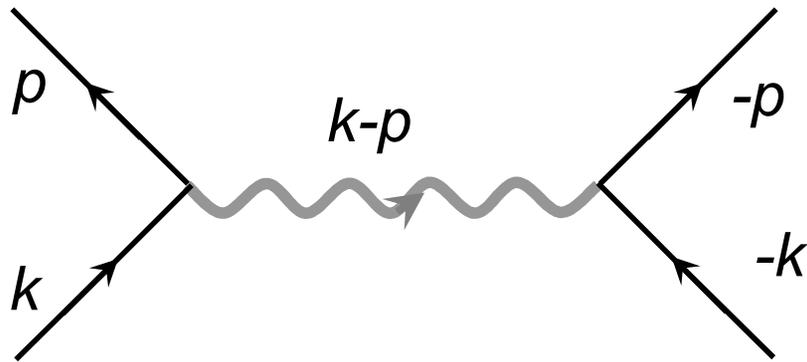
How to increase electron-phonon interaction  
and enhance superconductivity, CDW, ...  
by creating a non-equilibrium state of phonons

This talk: focus on superconductivity

Other experiments: optical control of Mott insulators,  
charge and spin density wave states,  
superconductivity in high  $T_c$  cuprates

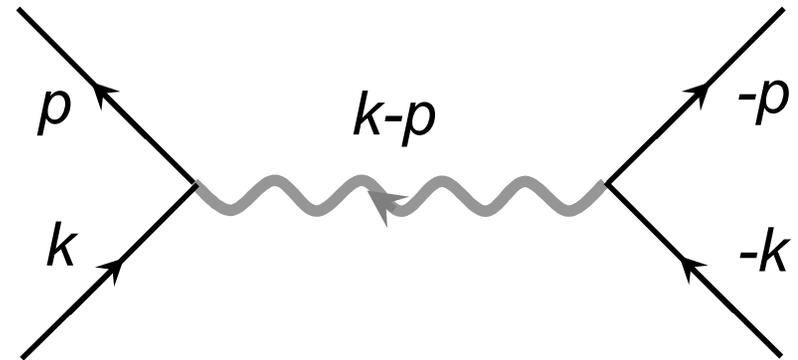
Other proposals for photoinduced superconductivity:  
Jaksch, Galitski, Komnik, Thorwart, Georges,  
Kollath, Mathey, Millis, Chamon, Sentef, ...

# Can one gain from non-equilibrium state of phonons?



interaction via phonon emission

$$-\frac{g^2(1+n)}{\epsilon_p + \omega_{k-p} - \epsilon_k} \approx -\frac{g^2(1+n)}{\omega_{ph}}$$



interaction via phonon absorption

$$-\frac{g^2 n}{\epsilon_p - \omega_{p-k} - \epsilon_k} \approx +\frac{g^2 n}{\omega_{ph}}$$

## Effective interaction

$$V_{\text{eff}} = -\frac{g^2}{\omega_{ph}}$$

This is the usual argument that real photons do not help to increase effective pairing strength

# Electron-phonon interactions in systems with driven phonons

$$\mathcal{H} = \sum_q \left( \frac{P_q^2}{2M} + \frac{M\omega_q^2}{2} Q_q^2 \right) + \mathcal{H}_{\text{drive}}(t)$$

Example: parametric drive of phonons

$$\mathcal{H} = \sum_q \left( \frac{P_q^2}{2M} + \frac{M\Omega_q^2(t)}{2} Q_q^2 \right)$$

$$\Omega_q^2(t) = \omega_q^2 (1 + 2\alpha \cos(2\Omega_{\text{drv}}t))$$

Effective electron-electron interaction assuming slow electron dynamics

$$\mathcal{H}_{\text{eff}} = U(t) \hat{\rho}_{\text{el}}(t) \hat{\rho}_{\text{el}}(t)$$

$$U(t) = \frac{|\tilde{g}_{\mathbf{q}}|^2}{\hbar} \int_{-\infty}^t dt' \mathcal{D}_{\hat{Q}\hat{Q}}^R(t, t')$$

$$\mathcal{D}_{\hat{Q}\hat{Q}}^R(t, t') = -i\theta(t - t') \langle \hat{Q}(t) \hat{Q}(t') - \hat{Q}(t') \hat{Q}(t) \rangle$$

# Electron-phonon interactions in systems with driven phonons

Compare to

$$\mathcal{H}' = \mathcal{H} - \phi \hat{Q}$$
$$\langle \hat{Q} \rangle = \chi \phi \quad \text{Fluctuation-dissipation theorem} \quad \chi = \langle \hat{Q} \hat{Q} \rangle$$
$$\Delta E = -\frac{\chi \phi^2}{2}$$

From electron-phonon coupling to effective electron-electron interaction

$$\mathcal{H}_{\text{el-phon}} = \sum_{k q \sigma} g_q Q_q c_{k-q\sigma}^\dagger c_{k\sigma}$$

$$\mathcal{H}_{\text{eff}} = U(t) \hat{\rho}_{\text{el}}(t) \hat{\rho}_{\text{el}}(t)$$

$$U(t) = \frac{|\tilde{g}_{\mathbf{q}}|^2}{\hbar} \int_{-\infty}^t dt' \mathcal{D}_{QQ}^R(t, t')$$

$$\mathcal{D}_{QQ}^R(t, t') = -i\theta(t - t') \langle \hat{Q}(t) \hat{Q}(t') - \hat{Q}(t') \hat{Q}(t) \rangle$$

## Phonon response function

Parametric drive  $\mathcal{H} = \sum_q \left( \frac{P_q^2}{2M} + \frac{M\Omega_q^2(t)}{2} Q_q^2 \right)$

Harmonic oscillator equations of motion  $\frac{d\hat{Q}(t)}{dt} = \frac{\hat{P}(t)}{M}$

$$\frac{d\hat{P}(t)}{dt} = -M\omega_q^2 [1 + 2\alpha \cos(2\Omega_{\text{drv}}t)] \hat{Q}(t)$$

From linearity of equations  $\hat{Q}(t) = \mathfrak{M}_{QQ}(t, t') \hat{Q}(t') - \mathfrak{M}_{QP}(t, t') \frac{\hat{P}(t')}{M\Omega_{\text{drv}}}$

Response function  $\mathcal{D}_{QQ}^R(t, t') = -i\theta(t - t') \langle [\hat{Q}(t'), \hat{P}(t')] \rangle \times \frac{-\mathfrak{M}_{QP}(t, t')}{M\Omega_{\text{drv}}}$   
 $= -\frac{\hbar}{M\Omega_{\text{drv}}} \theta(t - t') \mathfrak{M}_{QP}(t, t')$

Response function does not depend on the initial state of phonons.

It is determined by the Hamiltonian only. It is enhanced near parametric resonance.

# Electron-phonon interactions in systems with driven phonons

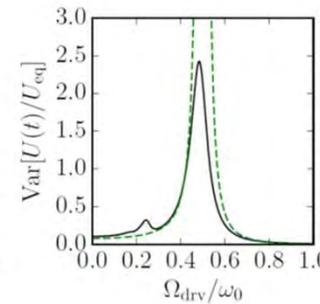
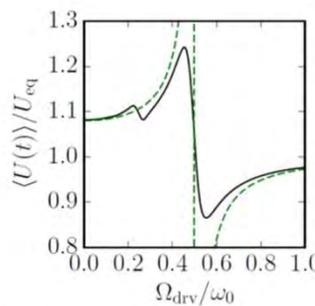
Parametrically driven phonons  $\Omega_q^2(t) = \omega_q^2 (1 + 2\alpha \cos(2\Omega_{\text{drv}}t))$

$$\frac{U(t)}{U_{\text{eq}}} = 1 - \frac{2\alpha\omega_q^2 \cos(2\Omega_{\text{drv}}t)}{\omega_q^2 - 4\Omega_{\text{drv}}^2} + \frac{2\alpha^2\omega_q^2 [\omega_q^2 - 16\Omega_{\text{drv}}^2 + \omega_q^2 \cos(4\Omega_{\text{drv}}t)]}{(\omega_q^2 - 16\Omega_{\text{drv}}^2)(\omega_q^2 - 4\Omega_{\text{drv}}^2)} + \mathcal{O}(\alpha^4)$$

Effective interaction: time average and variance.  $\alpha = 0.2$

$$\left\langle \frac{U(t)}{U_{\text{eq}}} \right\rangle = 1 + \frac{2\alpha^2\omega_q^2}{\omega_q^2 - 4\Omega_{\text{drv}}^2} + \mathcal{O}(\alpha^4)$$

$$\text{Var} \left[ \frac{U(t)}{U_{\text{eq}}} \right] = \frac{2\alpha^2\omega_q^4}{(\omega_q^2 - 4\Omega_{\text{drv}}^2)^2} + \mathcal{O}(\alpha^4)$$



Strong enhancement near resonance. Large response function near “instability”

“Naïve” averaging

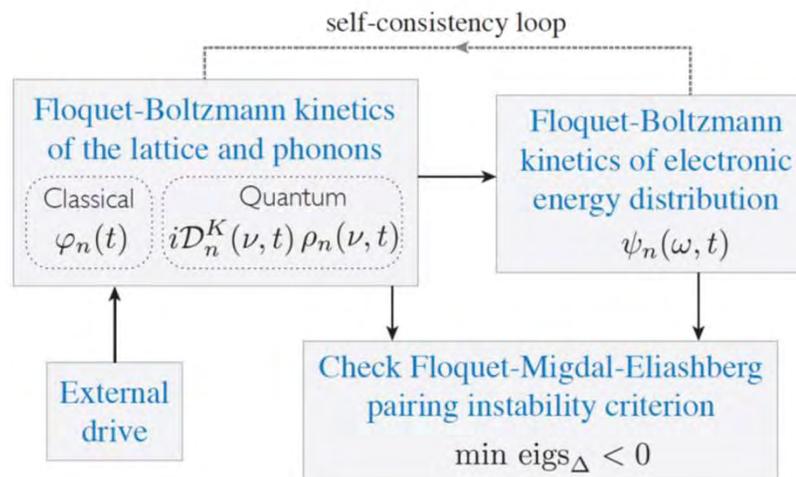
$$\left\langle \frac{T_c^{\text{BCS}}(t)}{T_{c,\text{eq}}^{\text{BCS}}} \right\rangle = \left\langle e^{-\frac{1}{\nu(0)U_{\text{eq}}} + \frac{1}{\nu(0)U(t)}} \right\rangle$$

Exponential dependence of  $T_c$ : gain on increase in  $U$  is larger than suppression due to decrease

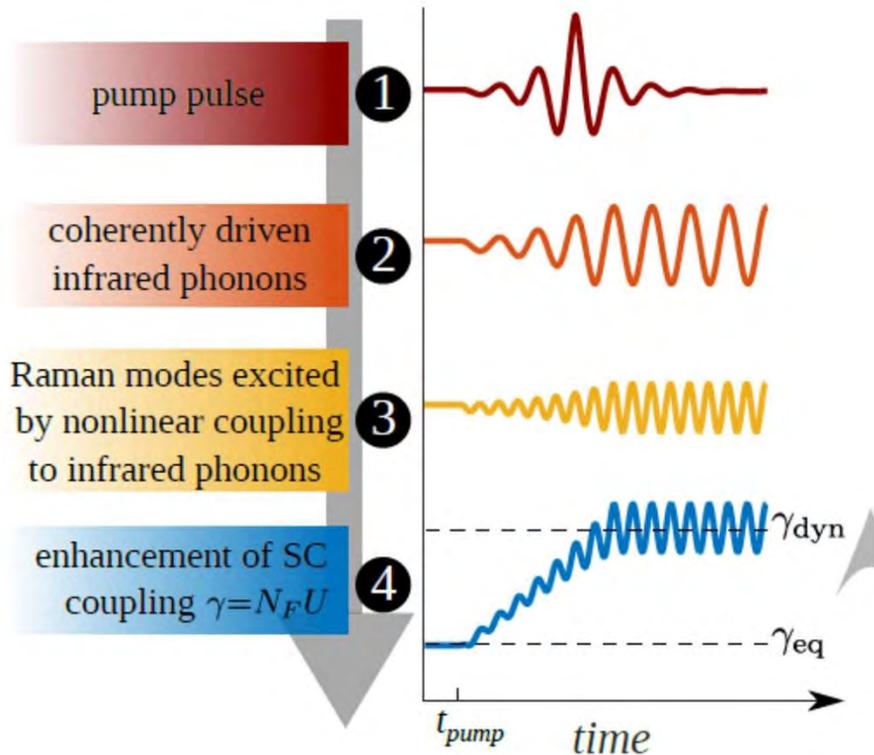
# Photo-induced superconductivity

## Floquet-Keldysh-Migdal-Eliashberg approach

M. Babadi, M. Knap, G. Refael, I. Martin, E. Demler



# Driven electron-phonon system



$$\tau_{\text{decoherence}} \gg \tau_{\text{pairing}}, \tau_{\text{therm.}}$$

This talk: focus on dynamical effects

## Microscopic model

$$\begin{aligned} \mathcal{L}[\varphi, \Psi](t) = & \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^{\dagger} (i\partial_t \mathbb{I} - \xi_{\mathbf{k}} \hat{\sigma}_3) \Psi_{\mathbf{k}} - \frac{1}{2} \sum_{\mathbf{q}} \frac{1}{2\omega_{\mathbf{q}}} \varphi_{\mathbf{q}} (\partial_t^2 + \omega_{\mathbf{q}}^2) \varphi_{-\mathbf{q}} \\ & - \sum_{j \in \text{lattice}} \mathcal{V}^{\text{ph}}(\varphi_j) - \frac{1}{\sqrt{N}} \sum_{\mathbf{k}, \mathbf{k}'} g_{\mathbf{k}, \mathbf{k}'} \varphi_{\mathbf{k}-\mathbf{k}'} \Psi_{\mathbf{k}'}^{\dagger} \hat{\sigma}_3 \Psi_{\mathbf{k}} + \frac{\Lambda}{2} |F(t)|^2 \sum_{j \in \text{lattice}} \varphi_j \end{aligned}$$

Phonons are driven only at  $q=0$ : e.g.  $\Lambda \varphi_{\text{IR}, q=0}^2(t) \varphi_{q=0}$ .

This analysis can be applied to the IR modes themselves, if they couple to electrons

We assume phonon-nonlinearities given by  $\kappa_3$  and  $\kappa_4$ .

$$\mathcal{V}^{\text{ph}}(\varphi) = -\frac{\kappa_3}{3!} \varphi^3 - \frac{\kappa_4}{4!} \varphi^4$$

We assume finite small dissipation  $\gamma_0$  for phonons at  $q=0$  due to other modes.

Additional dissipation is generated by electrons

# Phonon dynamics

Equations of motion for phonons: coherent part and fluctuations

$$\frac{1}{2\omega_0} (\partial_t^2 + \omega_0^2 + \gamma_0 \partial_t) \varphi(t) - \frac{\kappa_4}{6} \varphi^3(t) - \frac{\kappa_3}{2} \varphi^2(t) - \frac{\kappa_4}{2} \chi(t) \varphi(t) = \frac{\Lambda}{2} |F(t)|^2 + \frac{\kappa_3}{2} \chi(t)$$

$$\chi(t) \equiv \frac{1}{N} \sum_{\mathbf{q}} i\mathcal{D}_{\mathbf{q}}(t, t) \quad \text{Force from finite } \mathbf{q} \text{ phonons}$$

$$-\frac{1}{2\omega_{\mathbf{q}}} [\partial_{t_1}^2 + \omega_{\mathbf{q}}^2] \mathcal{D}_{\mathbf{q}}(t_1, t_2) = \delta_{\mathcal{C}}(t_1, t_2) + V(t_1) \mathcal{D}_{\mathbf{q}}(t_1, t_2) + \int_{\mathcal{C}} d\tau \Pi_{\mathbf{q}}(t_1, \tau) \mathcal{D}_{\mathbf{q}}(\tau, t_2)$$

$$V(t) \equiv -\frac{\kappa_4}{2} \chi(t) - \frac{\kappa_4}{2} \varphi^2(t) - \kappa_3 \varphi(t)$$

Drive from  $\mathbf{q}=0$  phonon

$$\Pi_{\mathbf{q}}(t_1, t_2) = \frac{1}{N} \sum_{\mathbf{k}} |g_{\mathbf{k}, \mathbf{k}+\mathbf{q}}|^2 \text{tr} \left[ \hat{\mathcal{G}}_{\mathbf{k}+\mathbf{q}}(t_1, t_2) \hat{\sigma}_3 \hat{\mathcal{G}}_{\mathbf{k}}(t_2, t_1) \hat{\sigma}_3 \right]$$

Force from electrons including damping and frequency renormalization

# Floquet-Wigner representation

Wigner transform—

$$\mathcal{D}(t_1, t_2) \rightarrow \mathcal{D}(\omega, T) \equiv \int dt e^{-i\omega t} \mathcal{D}(T + t/2, T - t/2)$$

center-of-mass (COM) time

$$T = (t_1 + t_2)/2$$

Usually:  
 Slow COM time evolution  
 Fast relative time.  
 Not applicable here b/c  
 of the fast drive

$$\varphi_{\text{IR}}^2(t) = \mathcal{A}(t) \cos^2(\Omega t)$$

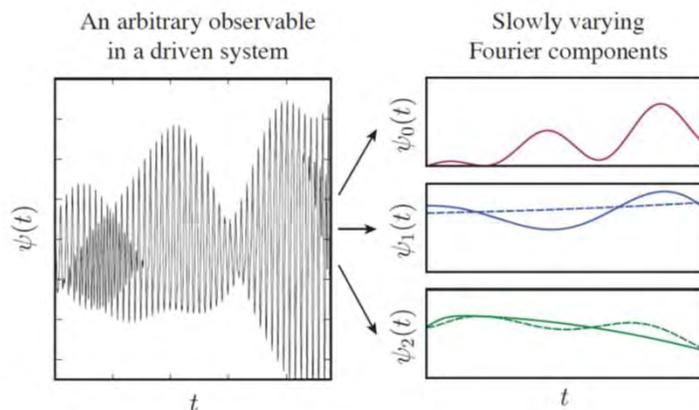
$$\partial_t \mathcal{A}(t) / \mathcal{A}(t) \ll \Omega^{-1}, \bar{\Omega}_0^{-1}$$



"Floquet-Wigner" representation

$$\mathcal{D}^{R/A/K}(\omega, T) = \sum_n \mathcal{D}_n^{R/A/K}(\omega; T) e^{-in\Omega T}$$

*slowly-varying  
 Floquet components*



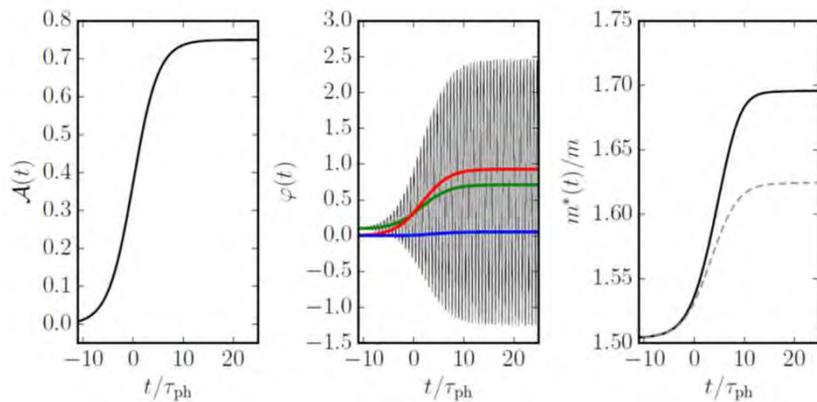
Earlier work on Floquet-Wigner:  
 Tsuji et al., PRB (2008),  
 Genske, Rosch, PRA (2015).

**This work: full quantum kinetic equation  
 beyond quasiparticle Boltzmann approximation**

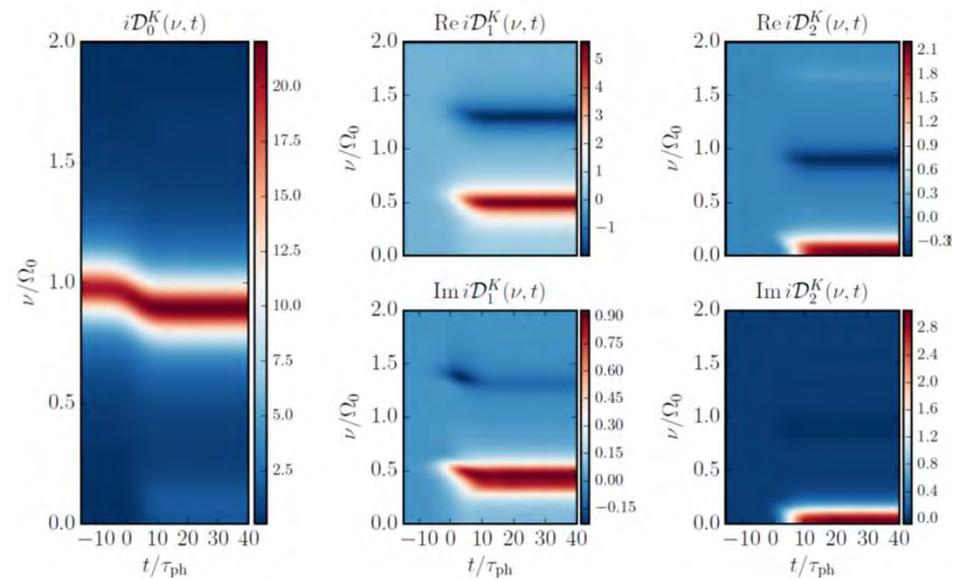
# Evolution of phonons

Ramped-up external drive from with  $A_{\max}=0.75$

Coherent amplitude at  $q=0$



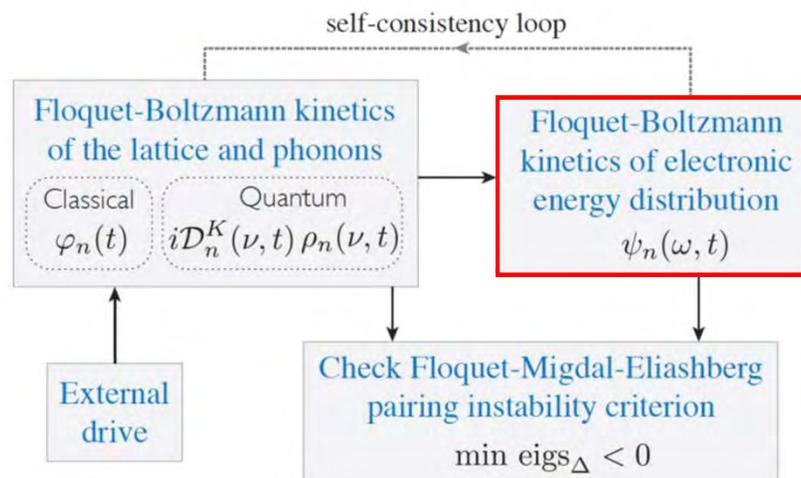
Propagators for finite  $q$



Note the red shift in the spectrum of phonons: contributes to SC enhancement

# The real time Migdal-Eliashberg theory

We want to compare enhancement of electron-phonon interaction with shortening of electron lifetime

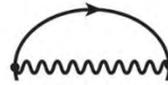


# The real time Migdal-Eliashberg theory

No superconductivity yet

Renormalization of electron energy, quasiparticle weight, lifetime

$$\hat{\Sigma}_{\mathbf{k}}(t, t') = \frac{i}{N} \sum_{\mathbf{k}'} \hat{\sigma}_3 \hat{\mathcal{G}}_{\mathbf{k}'}(t, t') \hat{\sigma}_3 |g_{\mathbf{k}\mathbf{k}'}|^2 D_{\mathbf{k}-\mathbf{k}'}(t, t')$$



spectral/Keldysh decomposition

$$i\hat{\mathcal{G}}_{\mathbf{k}}^>(\omega, T) = \frac{1}{2} [i\hat{\mathcal{G}}^K(\omega, T) + \hat{A}(\omega, T)]$$

$$i\hat{\mathcal{G}}_{\mathbf{k}}^<(\omega, T) = \frac{1}{2} [i\hat{\mathcal{G}}^K(\omega, T) - \hat{A}(\omega, T)]$$

$$\hat{\Sigma}^R(\omega, T) = \frac{1}{2} \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{+\infty} \frac{d\nu}{\omega - \omega' - \nu + i0^+} \left\{ iF^K(\nu, T) \check{A}(\omega', T) + F^\rho(\nu, T) i\check{\mathcal{G}}^K(\omega', T) \right\},$$

Renormalization of effective mass

$$i\hat{\Sigma}^K(\omega, T) = \frac{1}{2} \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{+\infty} d\nu (2\pi) \delta(\omega - \omega' - \nu) \left\{ iF^K(\nu, T) i\check{\mathcal{G}}^K(\omega', T) + F^\rho(\nu, T) \check{A}(\omega', T) \right\}$$

Coherence of quasiparticles

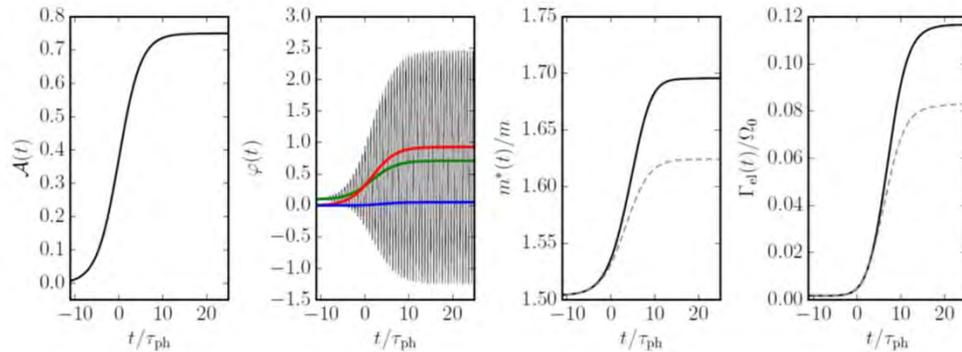
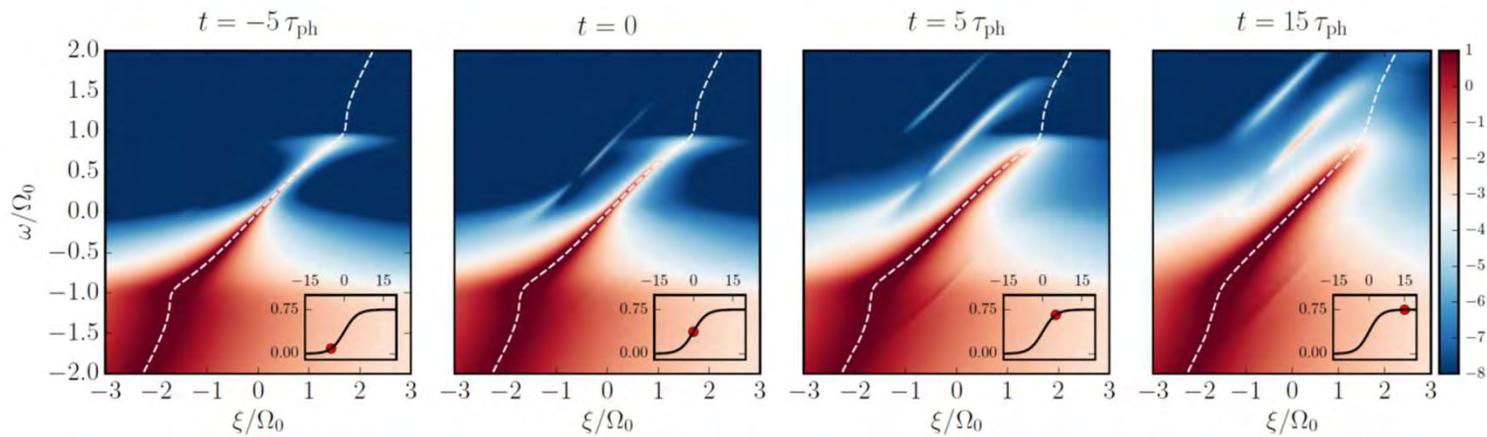
spectral Eliashberg  $F_{\xi, \xi'}^\rho(\nu, T) \equiv \frac{\nu(0)}{\nu(\xi)\nu(\xi')} \frac{1}{N^2} \sum_{\mathbf{k}, \mathbf{k}'} |g_{\mathbf{k}\mathbf{k}'}|^2 \frac{1}{2\pi} \rho_{\mathbf{k}-\mathbf{k}'}(\nu, T) \delta(\xi_{\mathbf{k}} - \xi) \delta(\xi_{\mathbf{k}'} - \xi')$ ,

Out of equilibrium phonons

Keldysh Eliashberg  $iF_{\xi, \xi'}^K(\nu, T) \equiv \frac{\nu(0)}{\nu(\xi)\nu(\xi')} \frac{1}{N^2} \sum_{\mathbf{k}, \mathbf{k}'} |g_{\mathbf{k}\mathbf{k}'}|^2 \frac{1}{2\pi} iD_{\mathbf{k}-\mathbf{k}'}^K(\nu, T) \delta(\xi_{\mathbf{k}} - \xi) \delta(\xi_{\mathbf{k}'} - \xi')$

# The real time Migdal-Eliashberg theory

## Predictions for ARPES

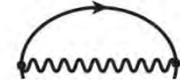


Mass enhancement indicates increase in effective electron-phonon interaction: good for pairing

Decrease of electron coherence: detrimental for pairing

# Onset of pairing in non-equilibrium Floquet system

Introduce off-diagonal component of self-energy. Require self-consistency.



1. Calculate the Q-matrix—

$$(2\omega - m\Omega) \delta\mathcal{F}_{n,m}^R = -2\pi i \phi_{n,m} + \sum_{n'=-N_D}^{N_D} (\Sigma_{n',m-n+n'}^R \delta\mathcal{F}_{n-n',m+n'}^R + \Sigma_{n',m+n-n'}^R \delta\mathcal{F}_{n-n',m-n'}^R)$$

$$\delta\mathcal{F}_{n,m}^R(\omega) = \sum_{n',m'} \mathbf{Q}_{n',m'}^{n,m}(\omega) \phi_{n',m'}(\omega)$$

Floquet cutoff (approx.)

2. Solve the functional eigenvalue equation—

$$\Delta_n(\omega) = \frac{i\omega}{2\pi} \sum_{n'=-N_\phi}^{N_\phi} \sum_{n''=-N_D}^{N_D} \sum_{m'} \left\{ \mathbf{Q}_{n',m'}^{n,0}(\omega) \int_0^{+\infty} \frac{d\omega'}{\omega'} K_{n''}(\omega - m'\Omega/2, \omega') \Delta_{n'-n''}(\omega') \right.$$

$$\left. - \left[ \mathbf{Q}_{n',m'}^{-n,0}(\omega) \right]^* \int_0^{+\infty} \frac{d\omega'}{\omega'} K_{n''}^*(\omega - m'\Omega/2, \omega') \Delta_{n'-n''}^*(\omega') \right\}$$

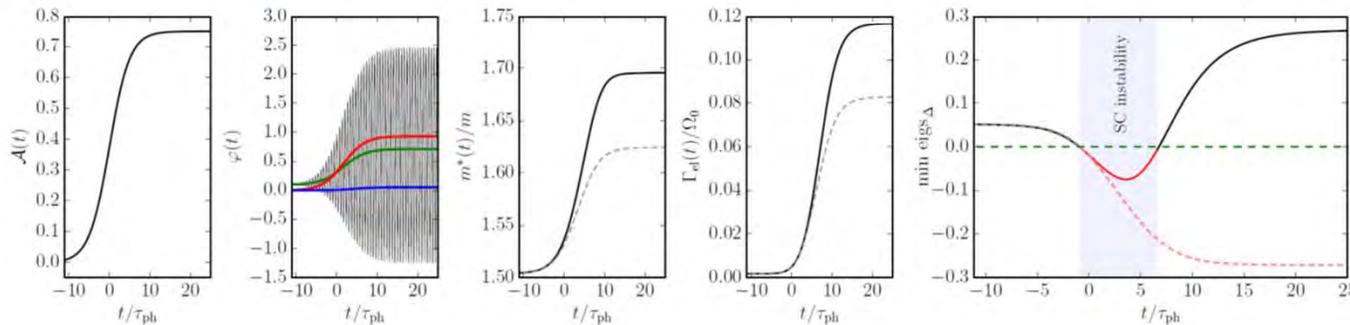
retarded interaction

dynamical self-energy effects (scattering, qp renormalization)

Compare to simple BCS

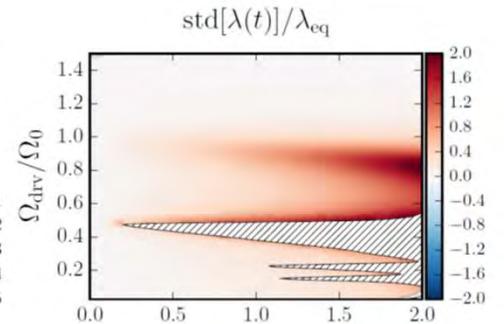
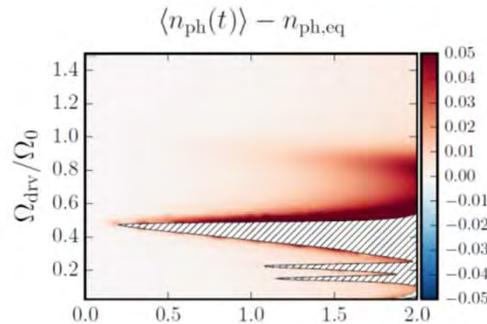
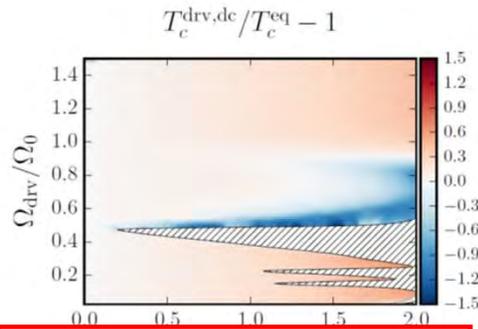
$$\Delta = -V\nu(0) \int d\xi \frac{\Delta}{|\xi|}$$

$$V = -\frac{g^2}{\omega_{\text{ph}}}$$

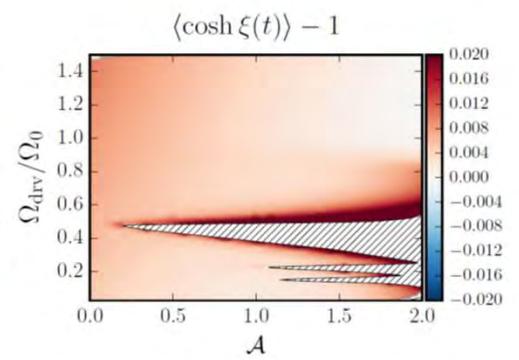
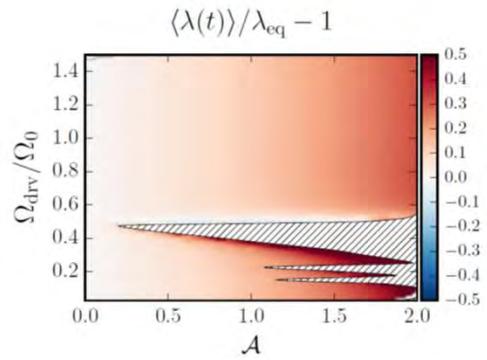
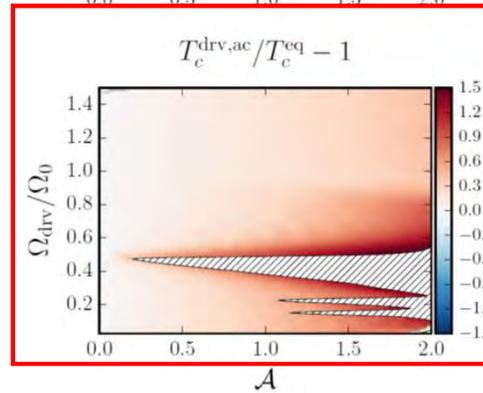


# Pairing instability in a driven electron-phonon system

Neglecting Floquet components. Use time averaged values



Include all Floquet components



# Optical response of photo-excited superconductor

## Mean-field as Gaussian states

Bogoliubov theory of the superfluid state of bosons

$$|\Psi_{\text{Bogoliubov}}\rangle = e^{i\alpha_0 b_{k=0}^\dagger + \sum_k f_k b_k^\dagger b_{-k}^\dagger} |0\rangle$$

BCS theory of superconductivity

$$|\Psi_{\text{BCS}}\rangle = \prod_k (u_k + v_k c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger) |0\rangle = e^{\sum_k \frac{v_k}{u_k} c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger} |0\rangle$$

# Mean-field as Gaussian states

## General Gaussian state of bosons

$$R = \begin{pmatrix} b_b^\dagger + b_1 \\ i(b_1^\dagger - b_1) \\ \dots \\ b_N^\dagger + b_N \\ i(b_N^\dagger - b_N) \end{pmatrix}$$

$$|\Psi_{\text{Boson Gauss}}\rangle = e^{iR^T v - \frac{i}{4} R^T \xi_b R}$$

$$\langle R_i \rangle = v_i$$

$$\delta R_i = R_i - \langle R_i \rangle$$

$$\langle \{\delta R_i, \delta R_j\}_+ \rangle = e^{i\xi_b}$$

## General Gaussian state of fermions

$$A = \begin{pmatrix} c_1^\dagger + c_1 \\ i(c_1^\dagger - c_1) \\ \dots \\ c_N^\dagger + c_N \\ i(c_N^\dagger - c_N) \end{pmatrix}$$

$$|\Psi_{\text{Fermion Gauss}}\rangle = e^{\frac{i}{4} R A^T \xi_m A}$$

$$\langle [A_i, A_j]_- \rangle = e^{i\xi_m}$$

## General Gaussian state

$$|\Psi_{\text{Gauss}}\rangle = |\Psi_{\text{Boson Gauss}}\rangle \times |\Psi_{\text{Fermion Gauss}}\rangle$$

## Beyond Gaussian states

$$\mathcal{H} = -t \sum \langle ij \rangle \sigma c_{i\sigma}^\dagger c_{j\sigma} - \mu \sum_{i\sigma} c_{i\sigma}^\dagger c_{i\sigma} + \sum_{nm} \omega_{nm} b_n^\dagger b_m + g \sum_{nm} c_{n\sigma}^\dagger c_{n\sigma} (b_n + b_n^\dagger)$$

Flow in imaginary time to find the ground state    Tao Shi, Ignacio Cirac, ED

$$|\Psi_{\text{Variational}}(\tau)\rangle = e^{iS(\tau)} \times |\Psi_{\text{Gauss}}(\tau)\rangle$$

$$S(\tau) = i \sum_{nm} \lambda_{nm}(\tau) (b_n - b_n^\dagger) c_{m\sigma}^\dagger c_{m\sigma}$$

Lang-Firsov transformation

Geometrical approach to variational imaginary time flow

$$|\Psi_j\rangle = \partial_{\xi_j} |\Psi(\xi)\rangle$$

$$|\mathbf{R}_\Psi\rangle = -(H - E) |\Psi(\xi)\rangle$$

$$\mathbf{G}_{ij} = \langle \Psi_i | \Psi_j \rangle$$

$$d_\tau \xi_i = \sum_{j=1} \mathbf{G}_{ij}^{-1} \langle \Psi_j | \mathbf{R}_\Psi \rangle$$

# Beyond Gaussian states

Variational approach to real time dynamics

$$|\Psi_{\text{Variational}}(t)\rangle = e^{iS(t)} \times |\Psi_{\text{Gauss}}(t)\rangle$$

$$\mathcal{H} = -t \sum \langle ij \rangle \sigma c_{i\sigma}^\dagger c_{j\sigma} - \mu \sum_{i\sigma} c_{i\sigma}^\dagger c_{i\sigma} + \sum_{nm} \omega_{nm} b_n^\dagger b_m + g \sum_{nm} c_{n\sigma}^\dagger c_{n\sigma} (b_n + b_n^\dagger) + \mathcal{H}_J(t)$$

$$H_J(t) = 2J \sum_{k,\sigma,\alpha} A_\alpha(t) \sin k_\alpha c_{k\sigma}^\dagger c_{k\sigma},$$

Calculation of optical conductivity in parametrically driven superconductors

Work in progress ...

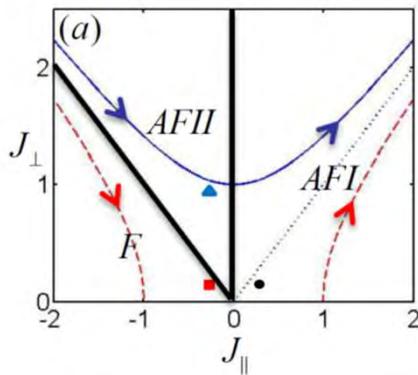
# Beyond Gaussian states

Variational approach to real time dynamics in the Kondo model    Tao Shi, Ignacio Cirac, ED

$$|\Psi_{\text{Variational}}(t)\rangle = e^{iS(t)} \times |\Psi_{\text{Gauss}}(t)\rangle$$

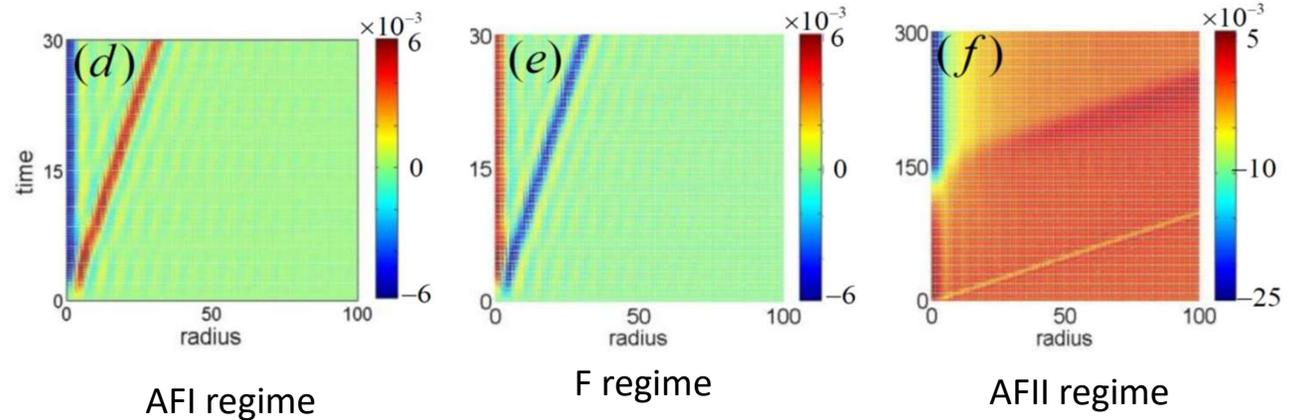
$$H_K = \sum_{k\sigma} kc_{k\sigma}^\dagger c_{k\sigma} + \frac{J_\perp}{2} [\sigma_+ \psi_\downarrow^\dagger(0) \psi_\uparrow(0) + \text{H.c.}] + \frac{J_\parallel}{4} \sigma_z [\psi_\uparrow^\dagger(0) \psi_\uparrow(0) - \psi_\downarrow^\dagger(0) \psi_\downarrow(0)]$$

Equilibrium phase diagram



Non-equilibrium dynamics is  
Challenging problem due to many crossovers

Fermion spin structure corresponding to spin up impurity state



## Conclusions

External drive leads to enhancement of electron-phonon interaction. It can be understood as parametric amplification. This leads to an increase in the effective BCS coupling constant

Nonequilibrium state of phonons also results in additional scattering of electrons that leads to pair-breaking

We find that increase in BCS coupling can dominate and find possible increase of instability temperature by 150%. Floquet aspects are crucial.













