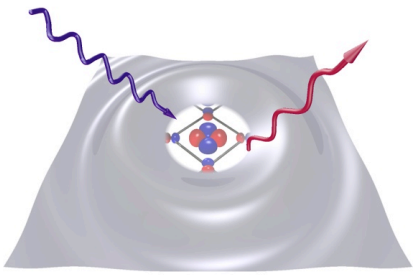
NC STATE UNIVERSITY



Understanding complex materials using non-equilibrium spectroscopy: what can theory tell us?

Alexander F. Kemper
North Carolina State University

IMPACT2016,
Cargèse,
August 29, 2016

PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
PRB 92, 224517 (2015)
PRB 93, 144506 (2016)
arXiv:1505.07055
Entropy 2016, 18(5), 180

M.A. Sentef (MSPD Hamburg)

J.K. Freericks (Georgetown)

B. Nozarewski, B. Moritz, T.P. Devereaux (Stanford/SLAC)

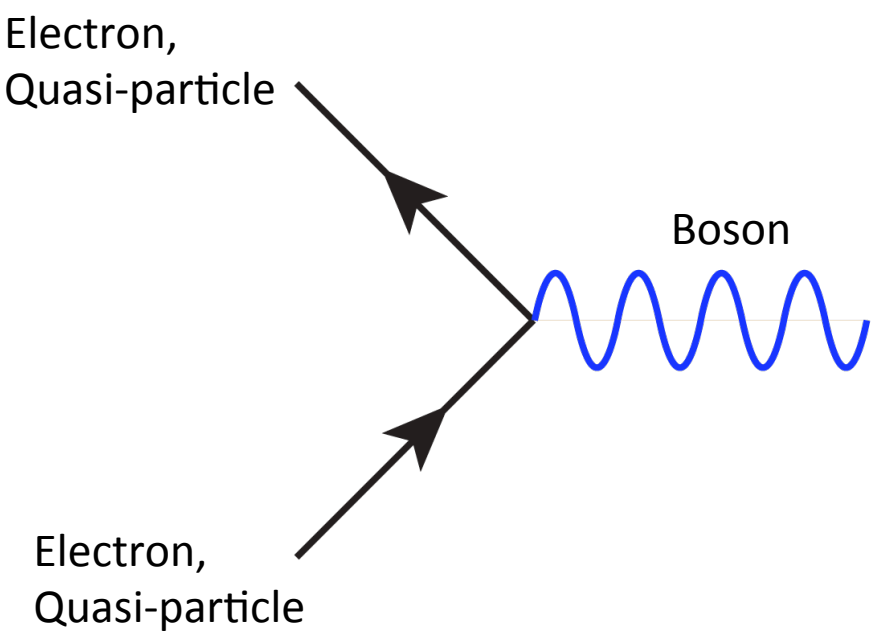
J.D. Rameau, P.D. Johnson (BNL)

S. Freutel, M. Ligges, I. Avigo, U. Bovensiepen (Duisburg-Essen)

Understanding complex materials using non-equilibrium spectroscopy: what can theory tell us?

- Boson interactions and population decay rates
- Violation of Matthiessen's rule in the time domain
- Higgs oscillations in superconductors

Electron-boson coupling

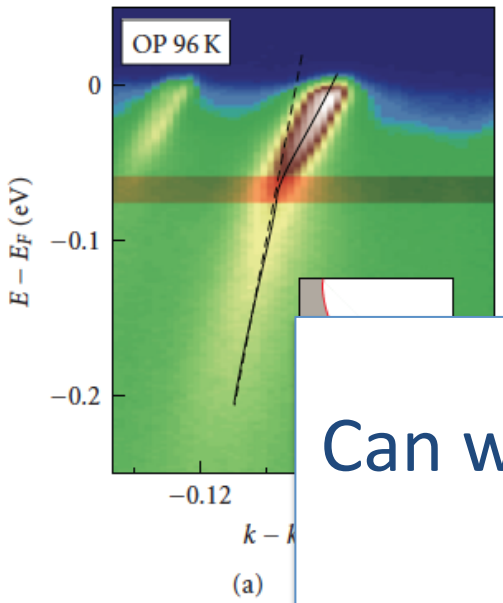


Phonons
Magnons
Spin fluctuations
Plasmons
Orbitons
...

$$H = \sum_{\mathbf{k}\sigma} \epsilon(\mathbf{k}) c^\dagger_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} + \sum_{q} \Omega_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{kq}\sigma} g_{\mathbf{k},\mathbf{q}} c^\dagger_{\mathbf{k}+\mathbf{q},\sigma} c_{\mathbf{k},\sigma} (b_{\mathbf{q}} + b^\dagger_{-\mathbf{q}}) + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$

Electrons | Bosons | Electron-boson coupling | Electron-electron coupling

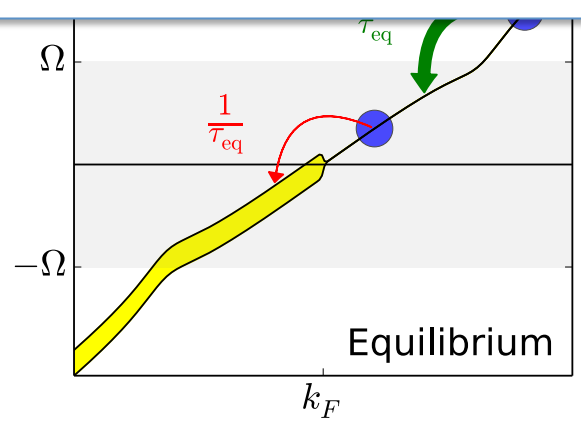
Manifestations of electron-boson interactions in spectra

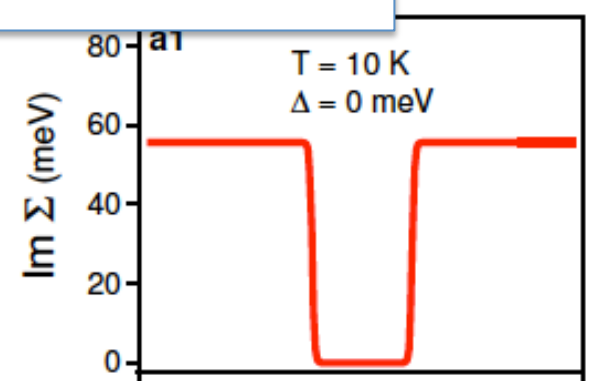


Kinks in dispersion

→ coupling to bosonic modes (phonons, spin fluctuations, …)

Can we measure lifetimes τ directly in the time domain?

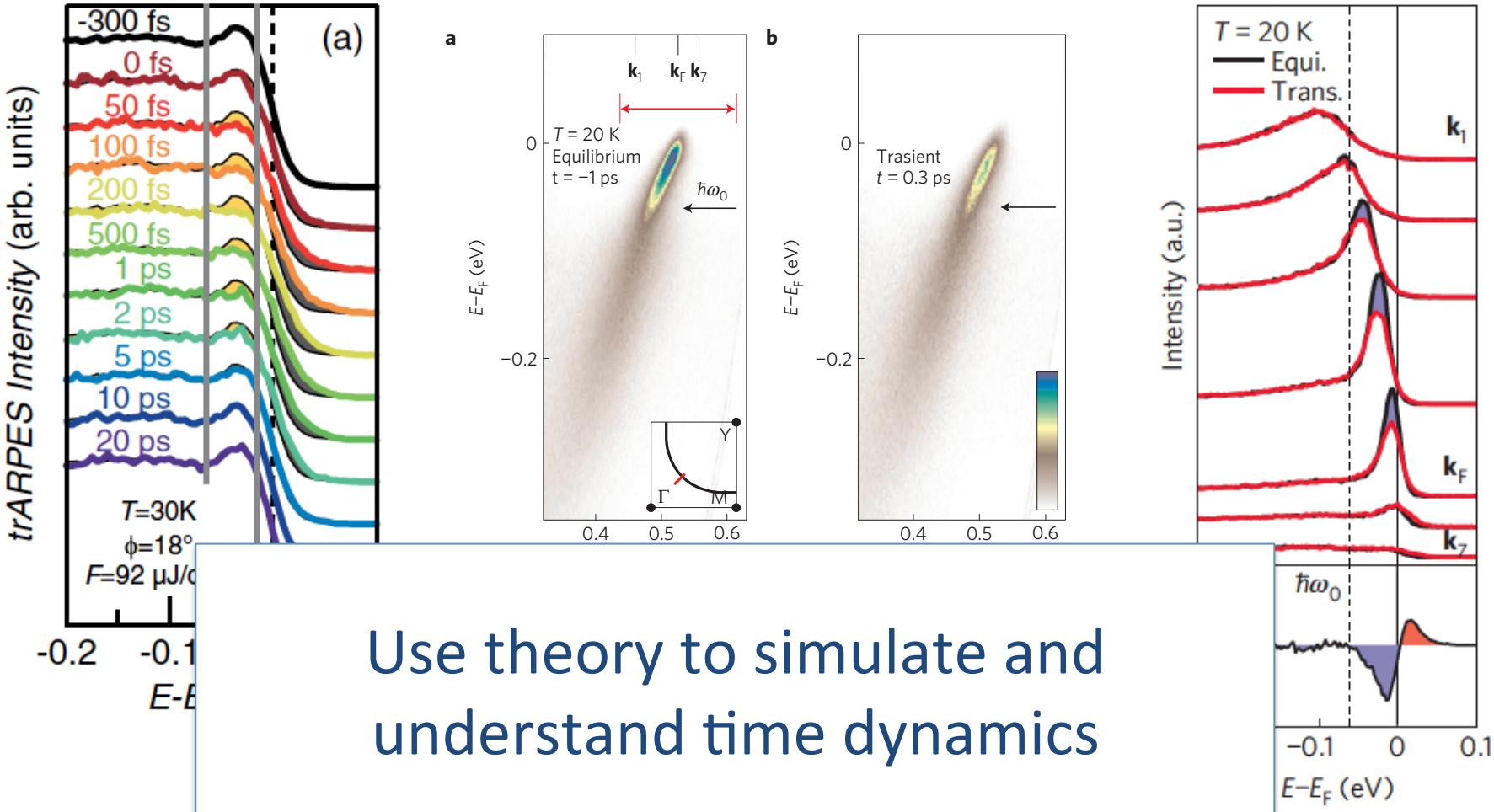




S. Johnston et al., Adv. CMP (2010)

Cuk et al, Physica Scripta (2006)

Manifestations of electron-boson interactions in time-resolved spectra



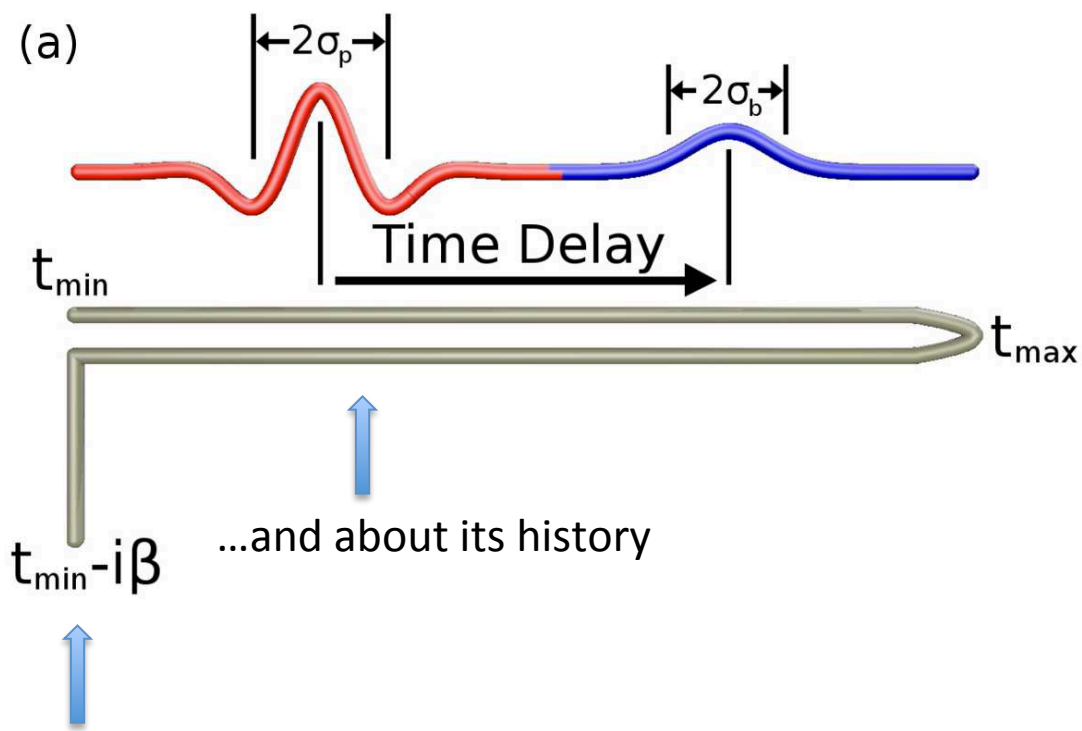
Use theory to simulate and understand time dynamics

R. Cortes et al., PRL (2011)

J. Graf et al., Nat. Phys. (2011)

Non-Equilibrium Keldysh Formalism

$$G_{\mathbf{k}}(t,t') = G^0_{\mathbf{k}}(t,t') + \int dt_1 dt_2\, G^0_{\mathbf{k}}(t,t_1)\,\Sigma(t_1,t_2)\,G_{\mathbf{k}}(t_2,t')$$

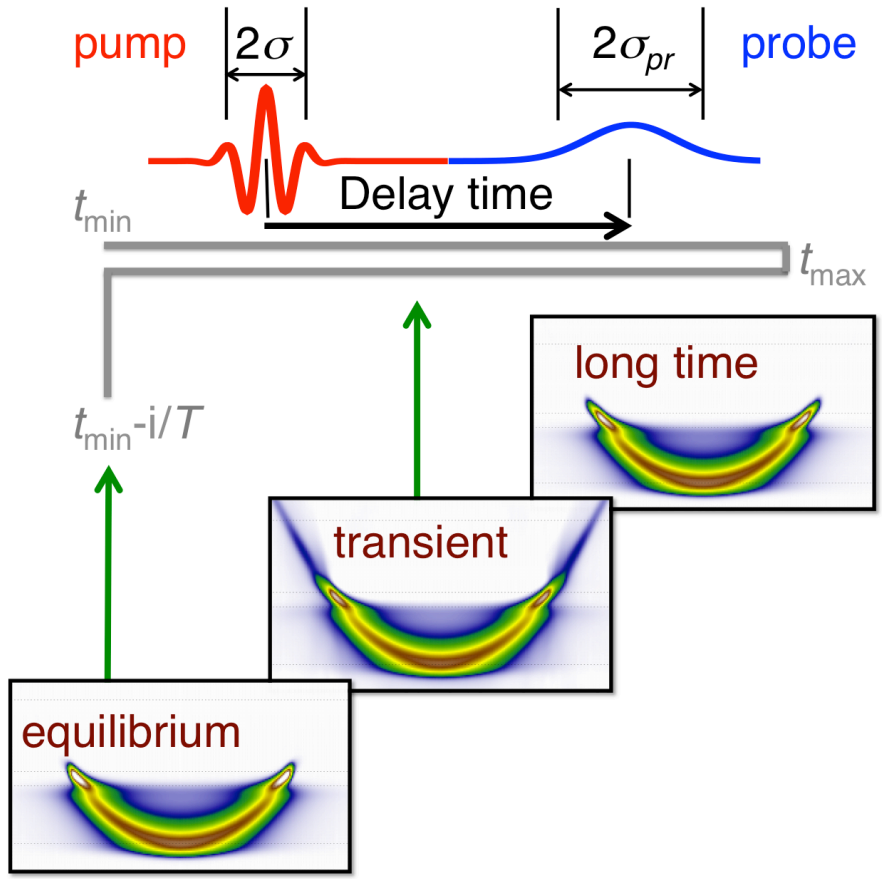


self-energy Σ:
electron-electron scattering
electron-phonon scattering
...

Include the effects of strong driving field through Peierls substitution

$$\boldsymbol{k} \rightarrow \boldsymbol{k} - e\mathbf{A}(t)$$

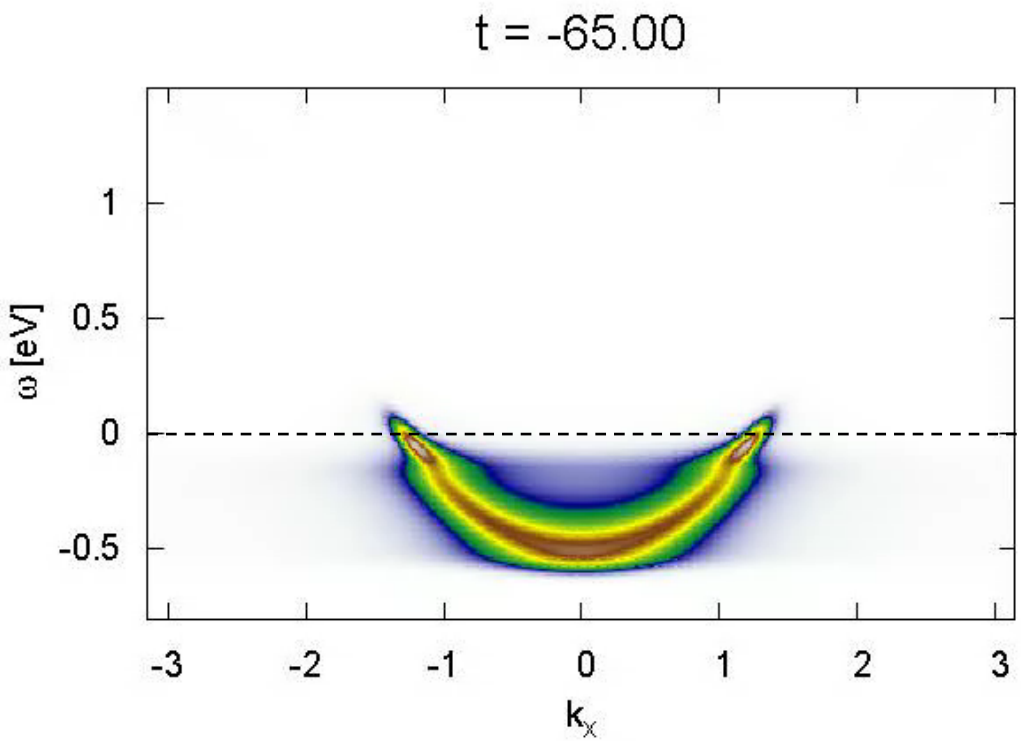
Pump-probe photoemission



$$A_{\mathbf{k}}(\omega, t_0) = \mathrm{Im}\ \frac{1}{2\pi\sigma^2} \int dt dt' G^{<}_{\mathbf{k}}(t,t') e^{-(t-t_0)^2/2\sigma^2} e^{-(t'-t_0)^2/2\sigma^2} e^{i\omega(t-t')}$$

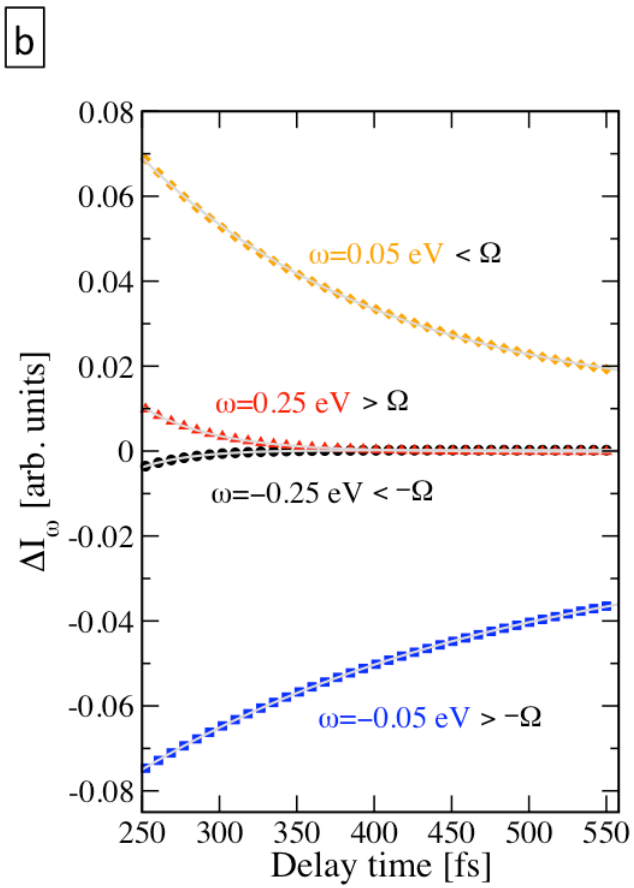
M. Eckstein and M. Kollar, PRB (2008), J. K. Freericks et al., PRL (2009), J. K. Freericks et al.

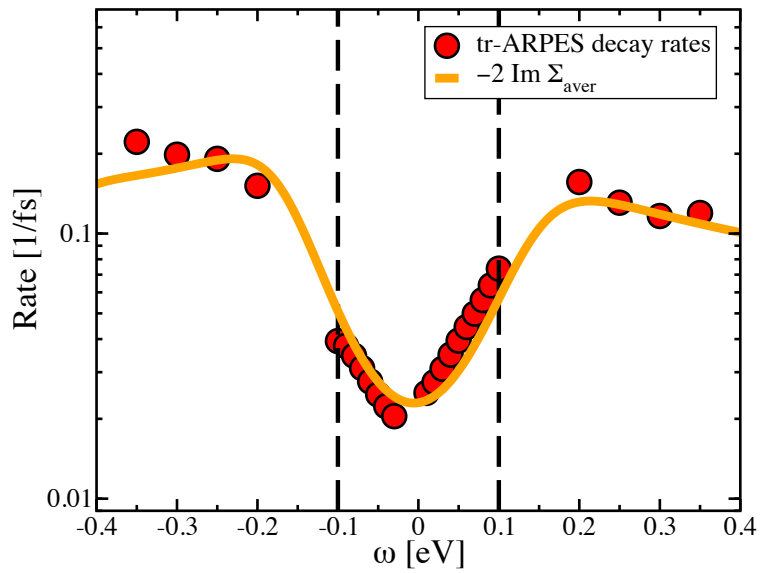
Electron-lattice coupling

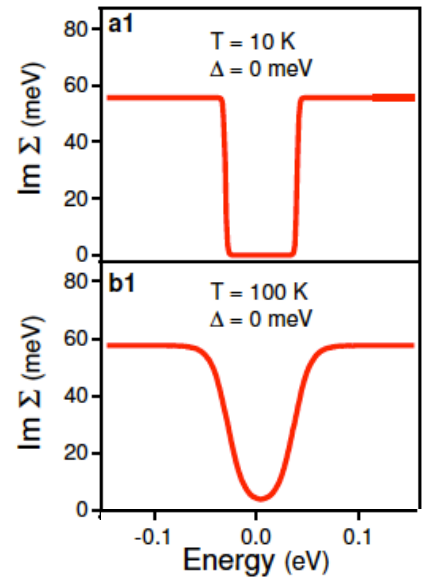


Phys. Rev. X 3, 041033

Electron-lattice coupling







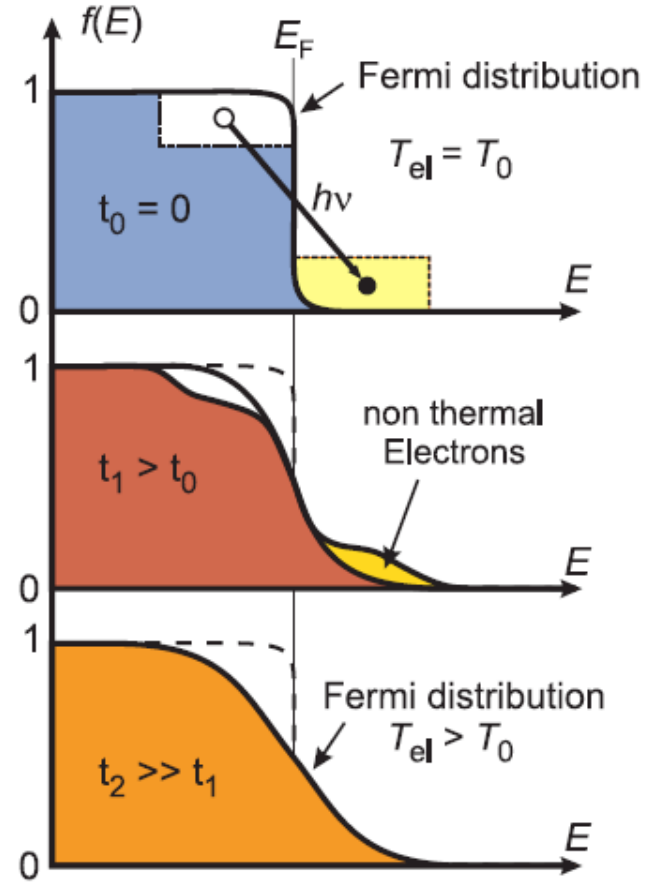
$$1/\tau(\omega)$$

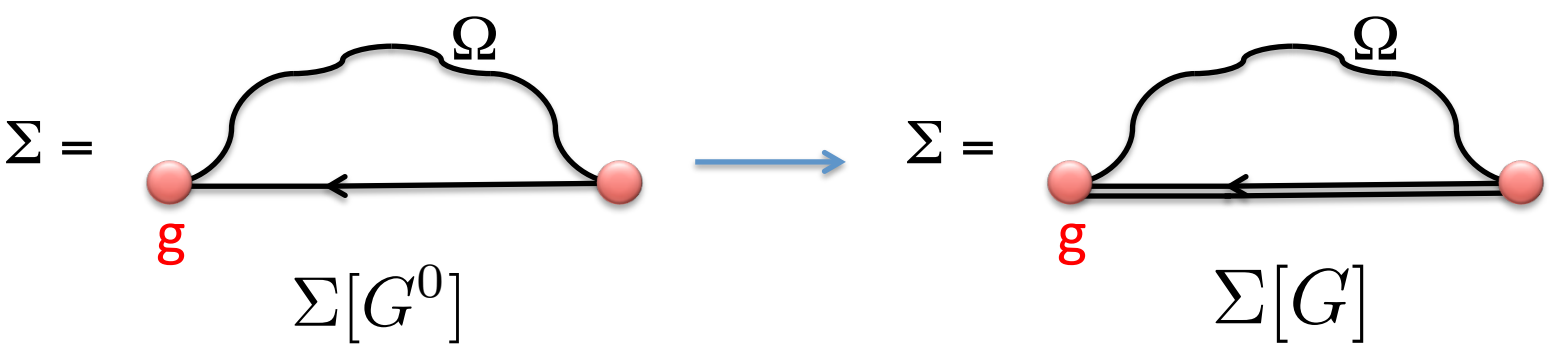
M. Sentef, Phys. Rev. X 3, 041033

How does the pump fluence affect these results?

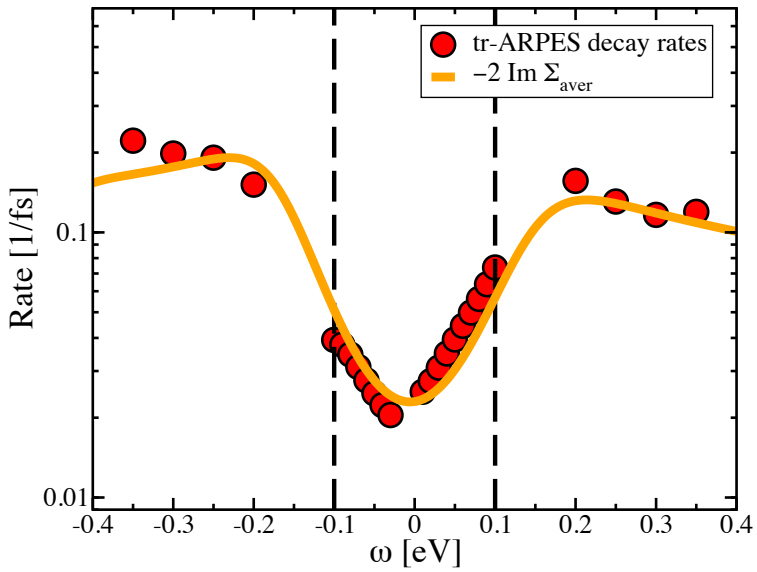
(XKCD)

We should account for the changes in the distribution self-consistently.



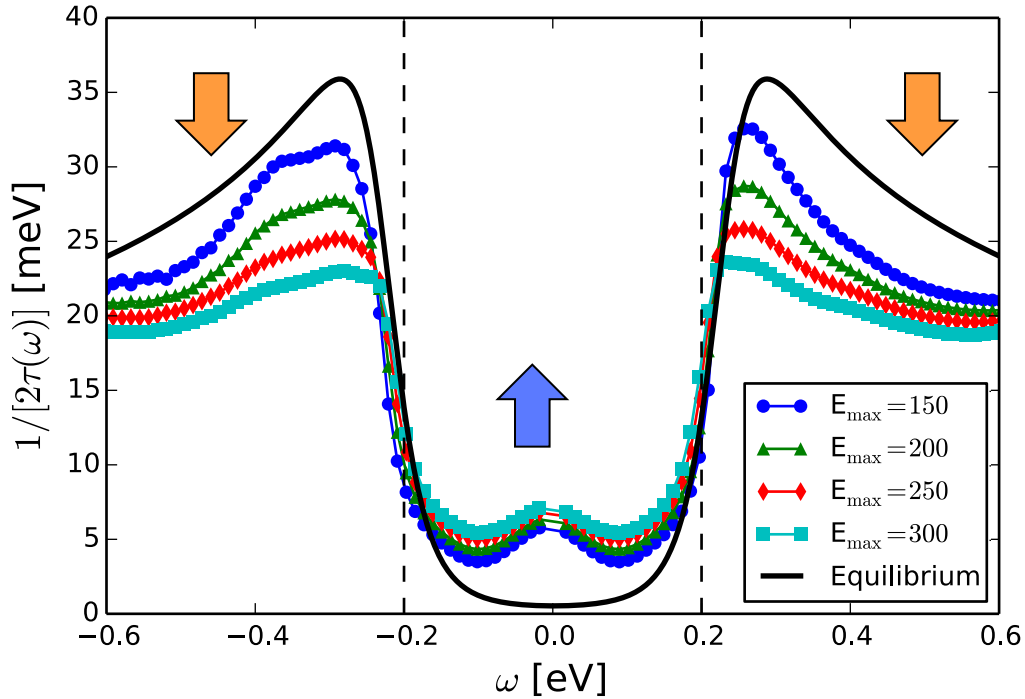


Driving beyond the weak limit



Low fluence limit
Phys. Rev. X 3, 041033

Increased excitation



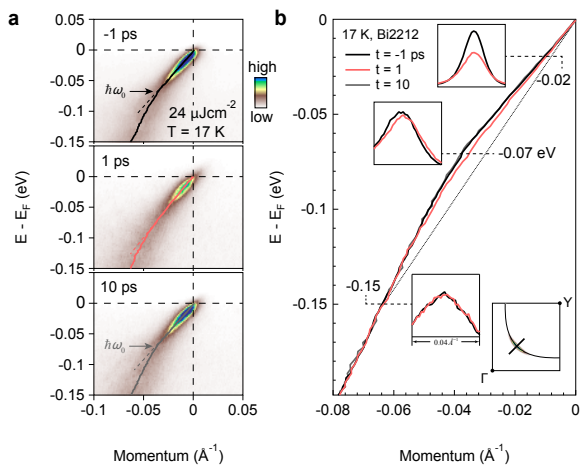
Strong pumping leads to fluence- and time-dependent interactions

$$1/\tau(\omega) \approx -2\mathrm{Im}\Sigma(\omega)$$

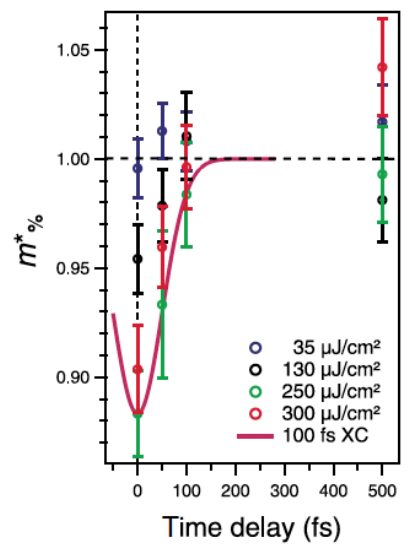
AFK et al PRB 2014

Experimental evidence for modified interactions

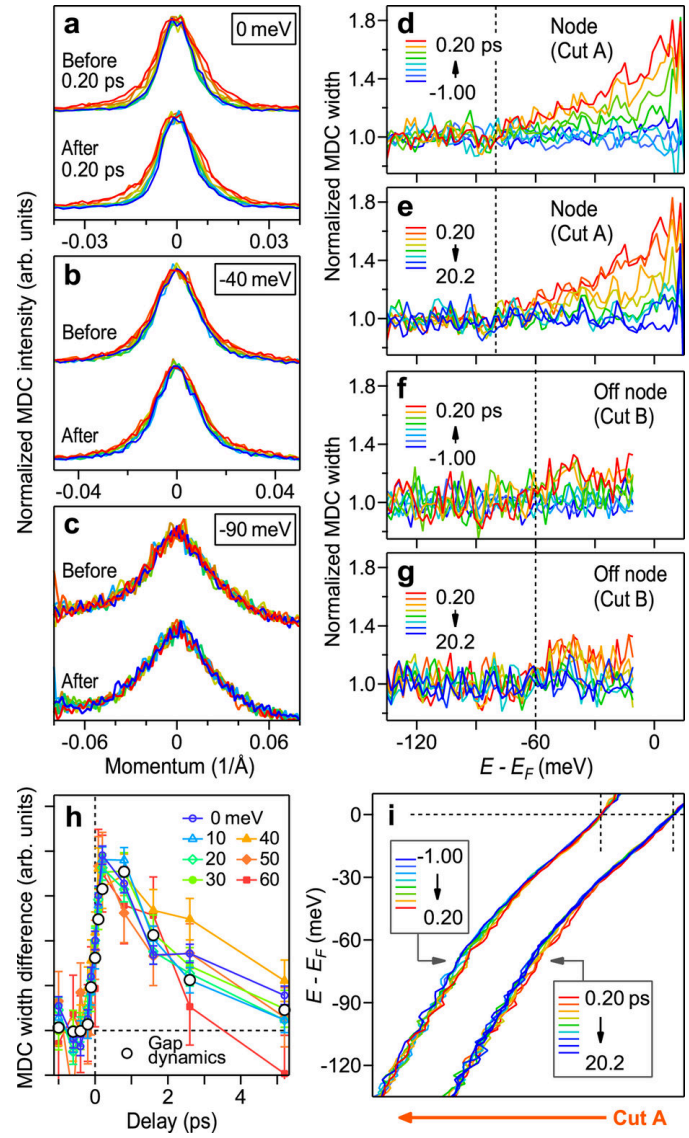
- Increase in MDC width within phonon window after pumping
- Weakening of the kink
- Interpreted as *decoupling* of electrons and phonons



Zhang, Nature Comm. (2014)

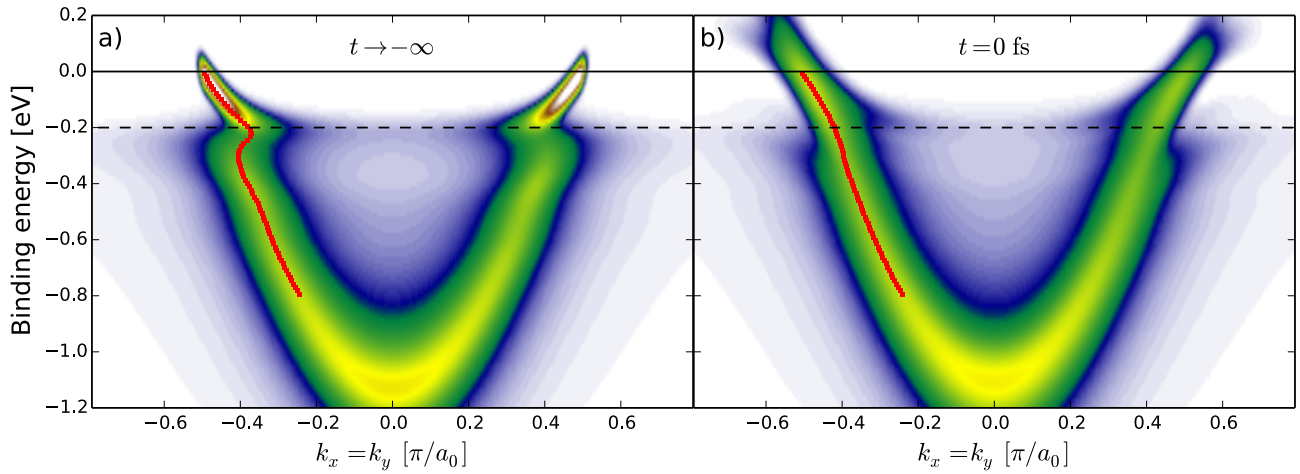


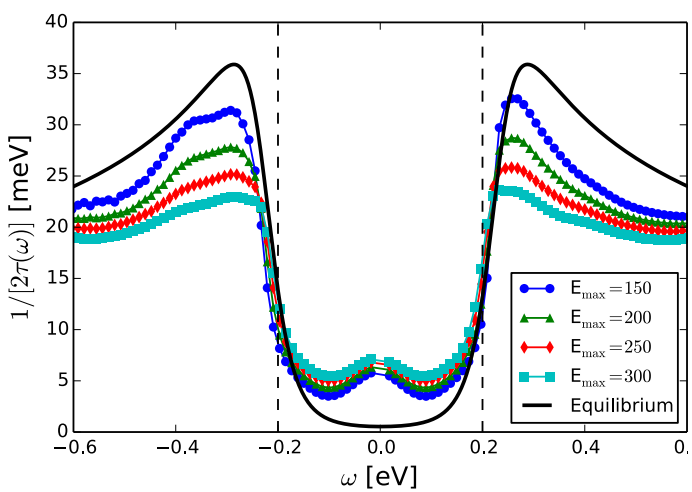
Rameau, PRB (2014)

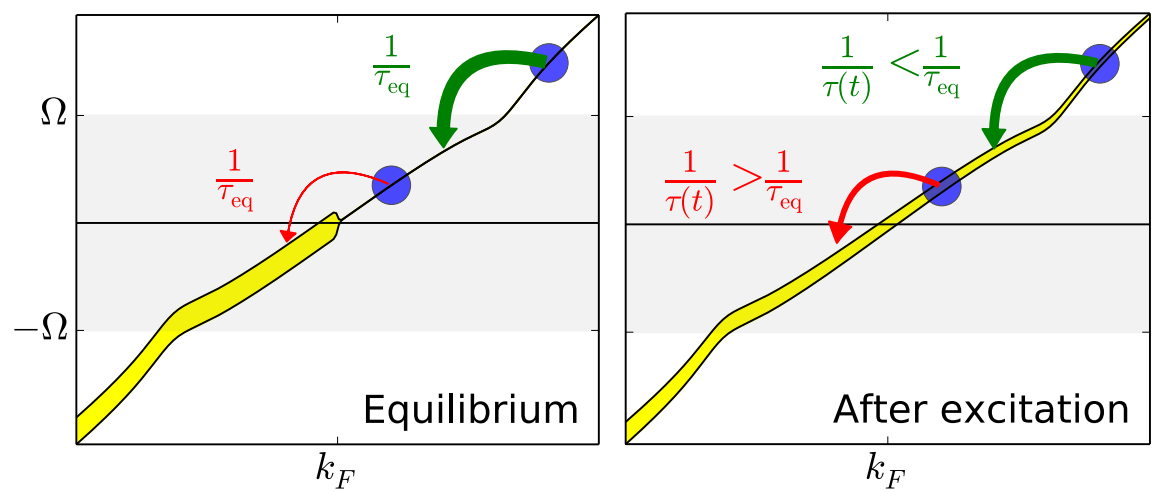


Ishida, Nat. Sci. Reports (2016)

Changes in kink, linewidth, and population decay rate after pumping can be understood by considering population redistribution







Understanding complex materials using non-equilibrium spectroscopy: what can theory tell us?

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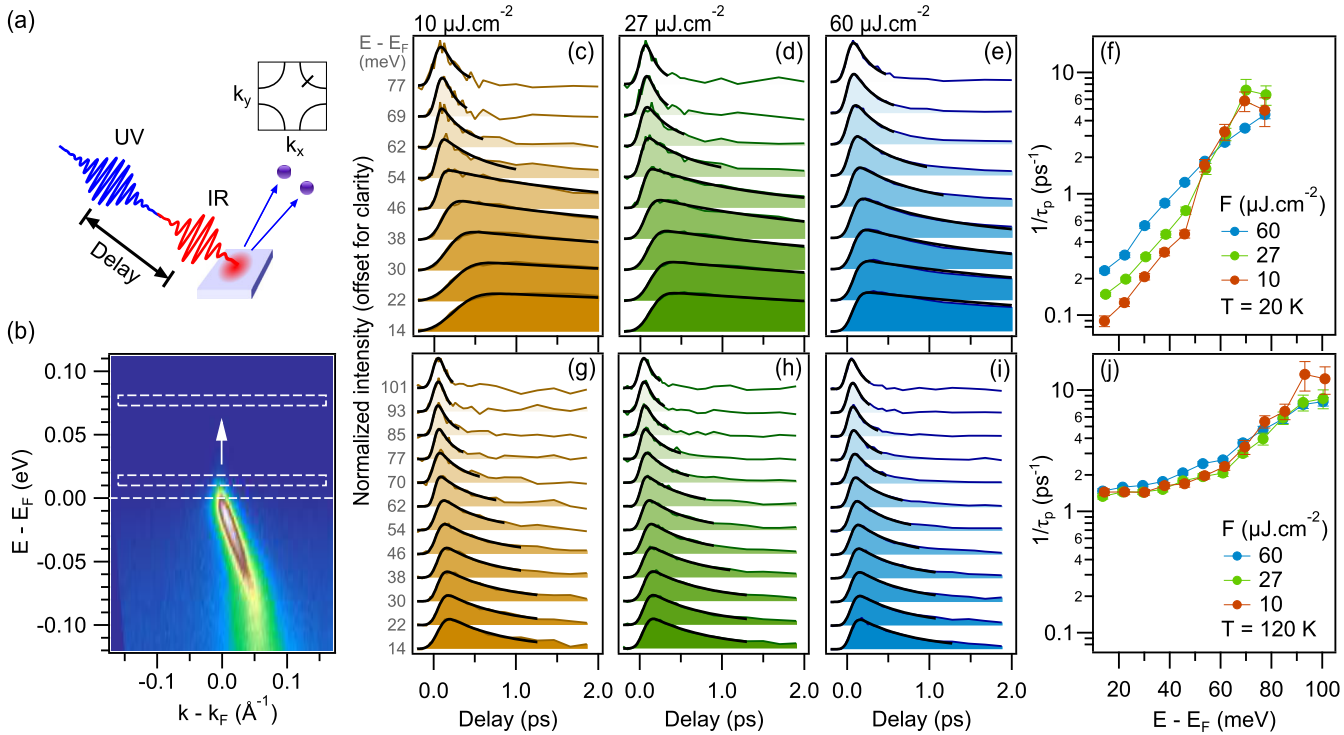
- Higgs oscillations in superconductors

PRL **114,** 247001 (2015) PHYSICAL REVIEW LETTERS week ending 19 JUNE 2015

Inequivalence of Single-Particle and Population Lifetimes in a Cuprate Superconductor

S.-L. Yang,[1,2] J. A. Sobota,[1,3] D. Leuenberger,[1,2] Y. He,[1,2] M. Hashimoto,[4] D. H. Lu,[4] H. Eisaki,[5] P. S. Kirchmann,[1,*] and Z.-X. Shen[1,2,†]



Also: I. Gierz, S. Link, U. Starke, and A. Cavalleri, Faraday Discuss. 171, 311 (2014).

PRL **114,** 247001 (2015) PHYSICAL REVIEW LETTERS week ending 19 JUNE 2015

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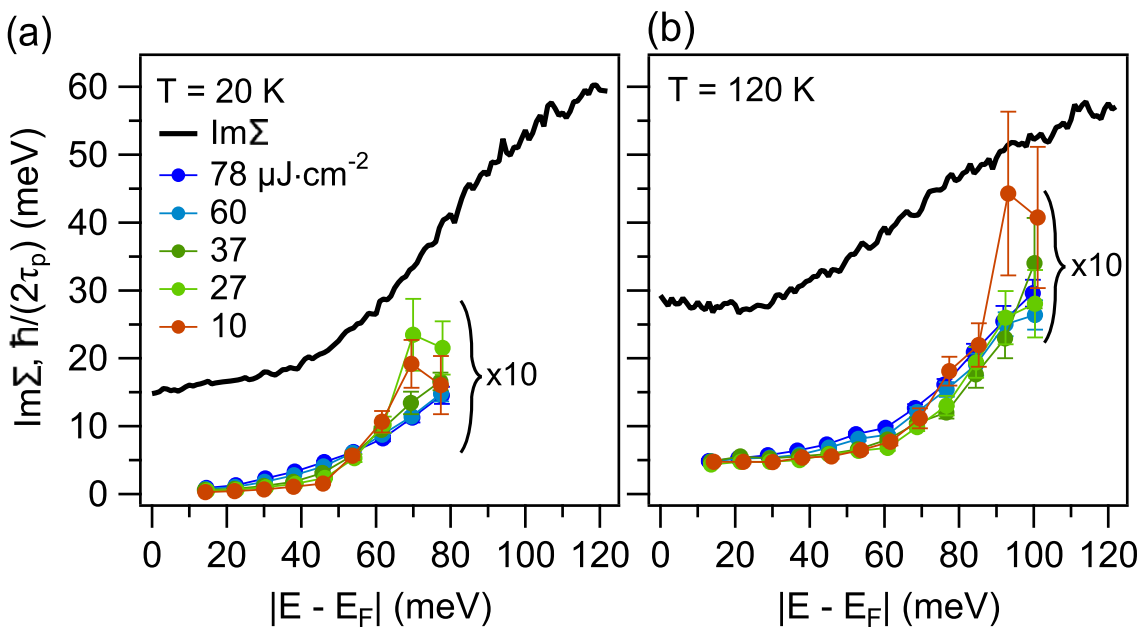
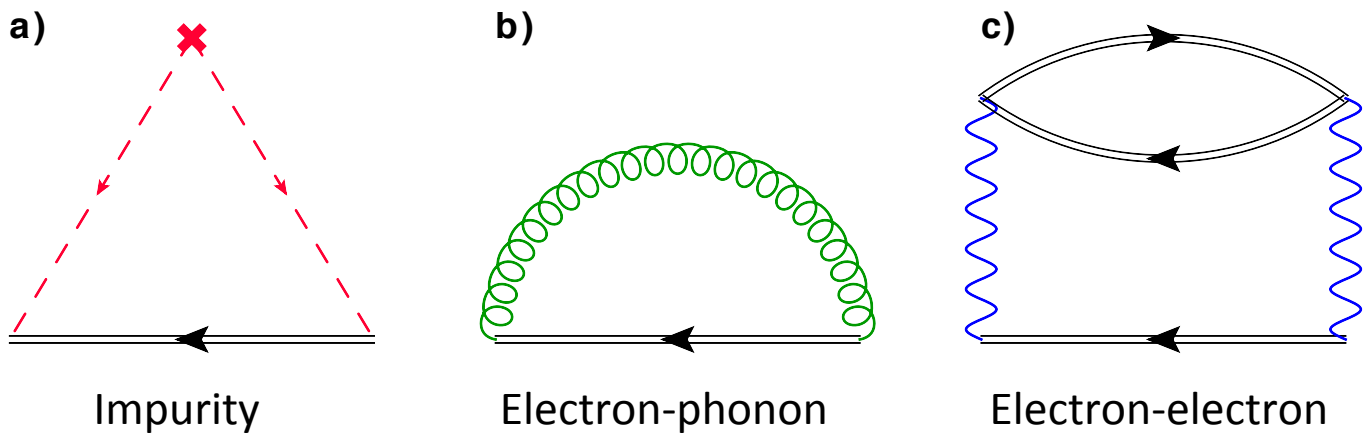
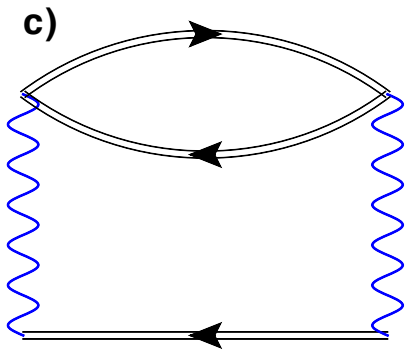


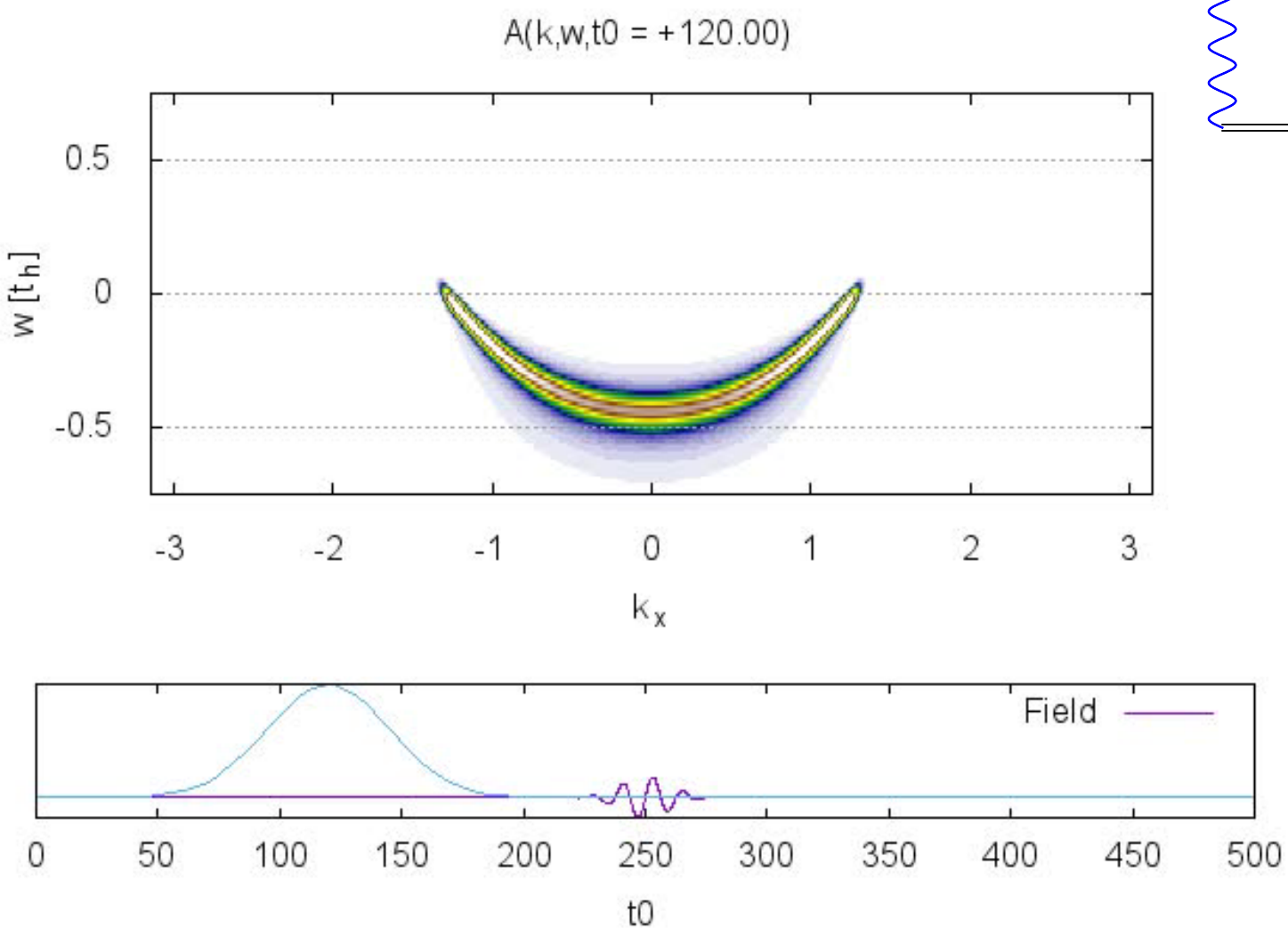
$$\frac{1}{\tau(\omega)} \stackrel{?}{=} -2\mathrm{Im}\Sigma(\omega = \epsilon_{\mathbf{k}})$$

Consider interactions beyond electron-phonon coupling

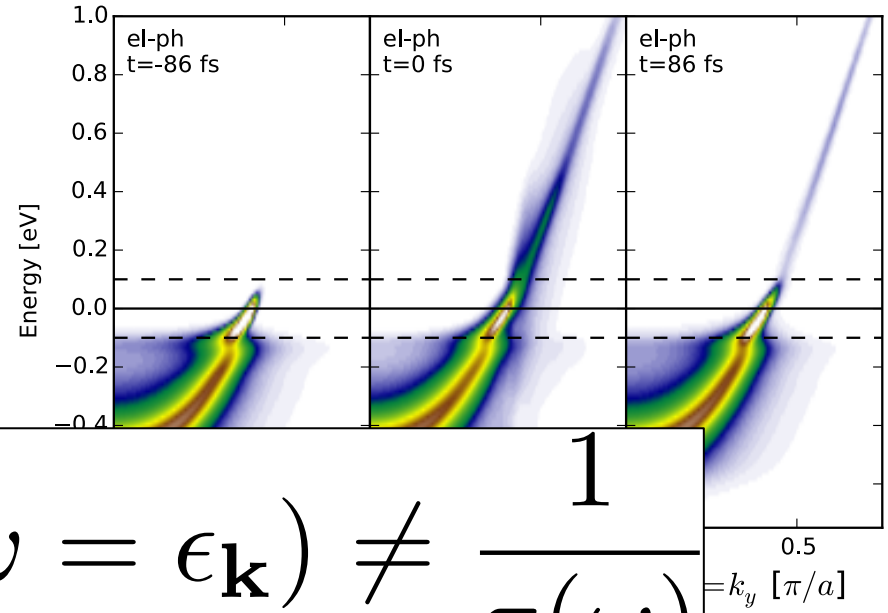


Electron-electron



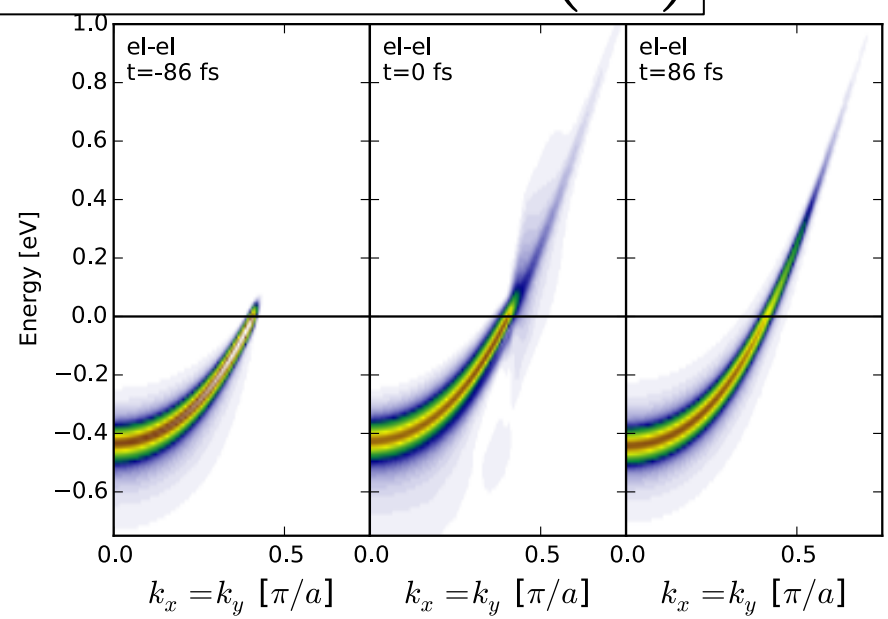


Electron-phonon



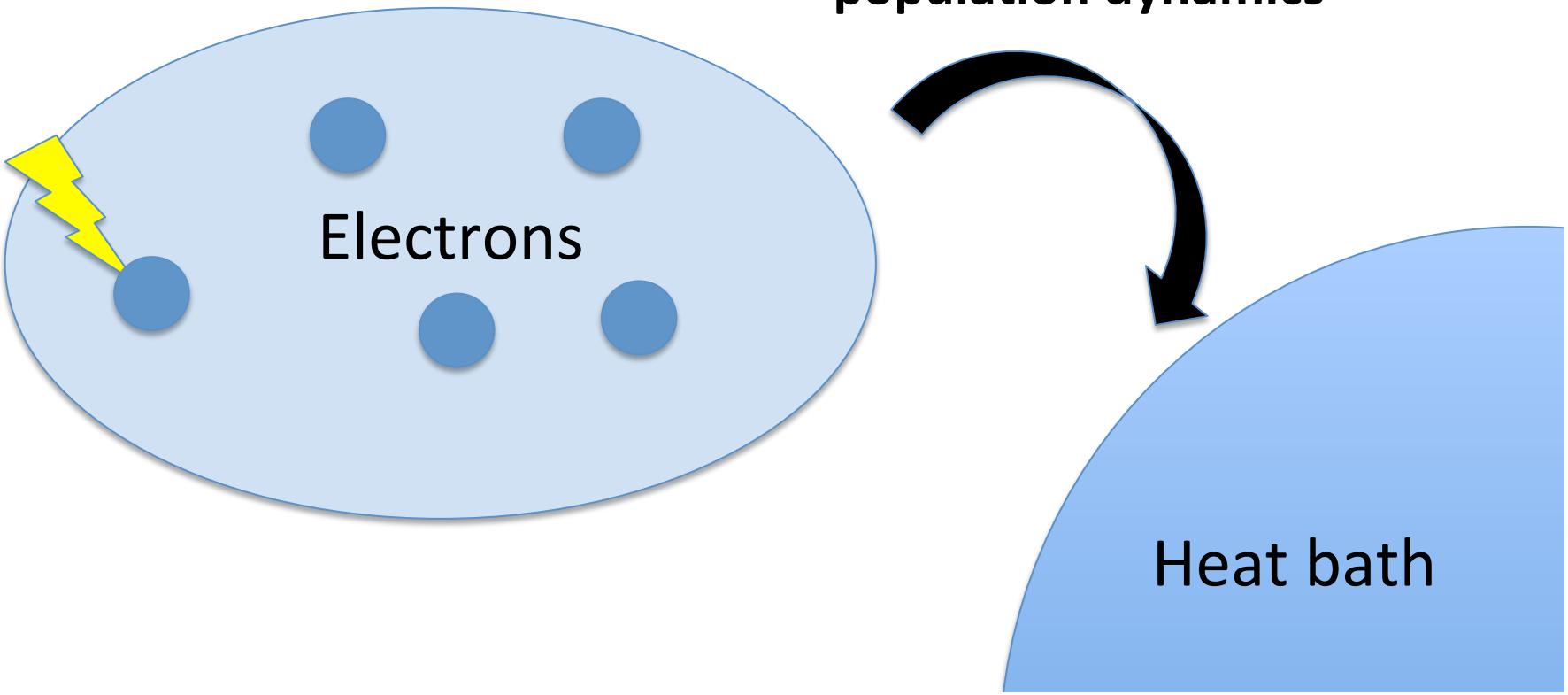
$$-2\mathrm{Im}\Sigma(\omega = \epsilon_{\mathbf{k}}) \neq \frac{1}{\tau(\omega)}$$

Electron-electron

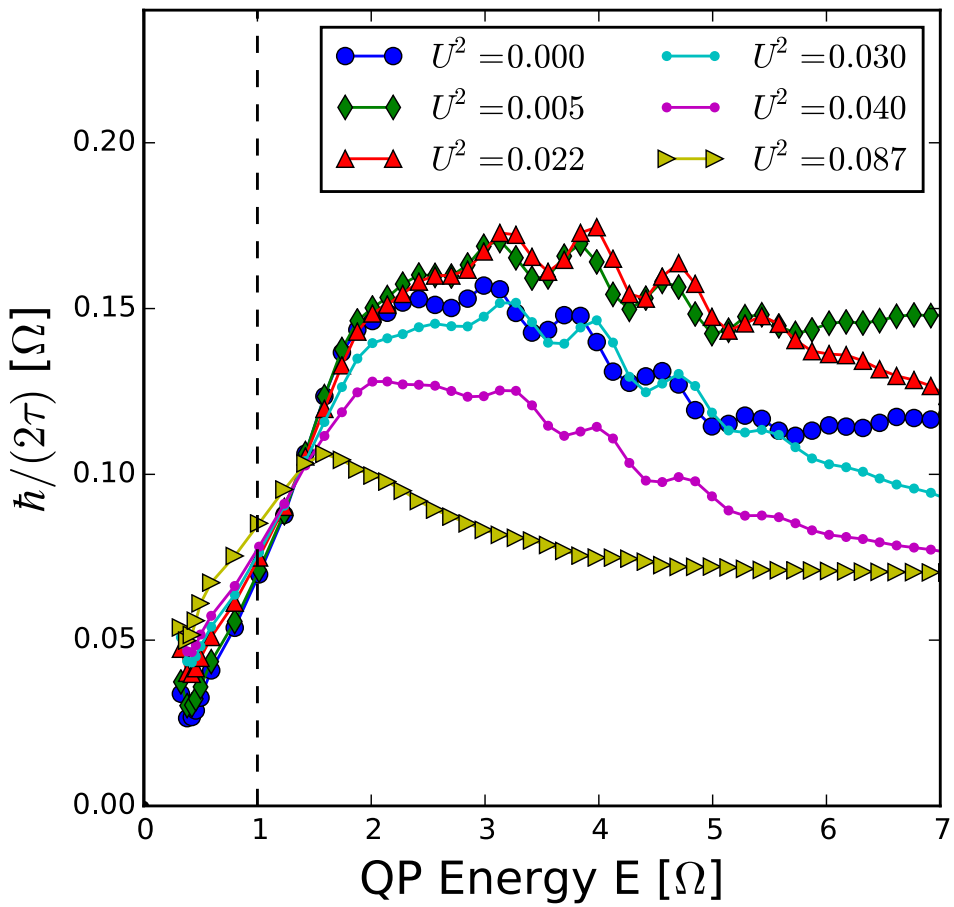


$$-2\mathrm{Im}\Sigma(\omega = \epsilon_{\mathbf{k}}) \neq \frac{1}{\tau(\omega)}$$

Energy transfer determines population dynamics



Combining electron-electron and electron-phonon scattering



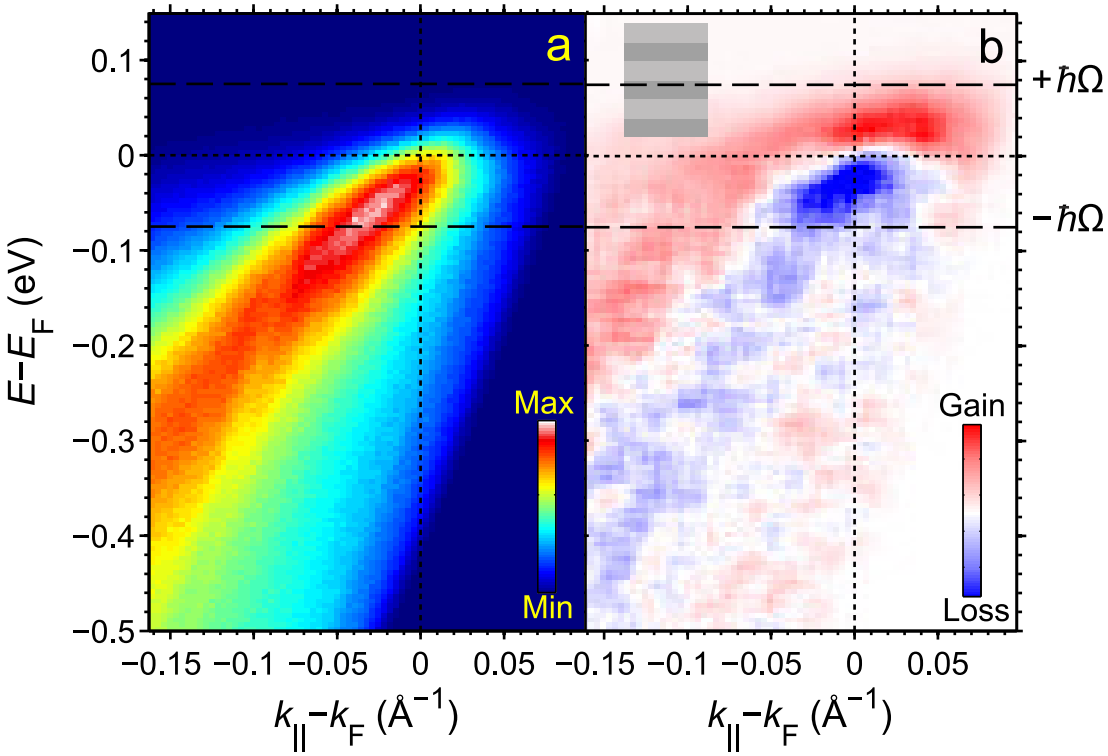
$$g^2 = 0.02$$

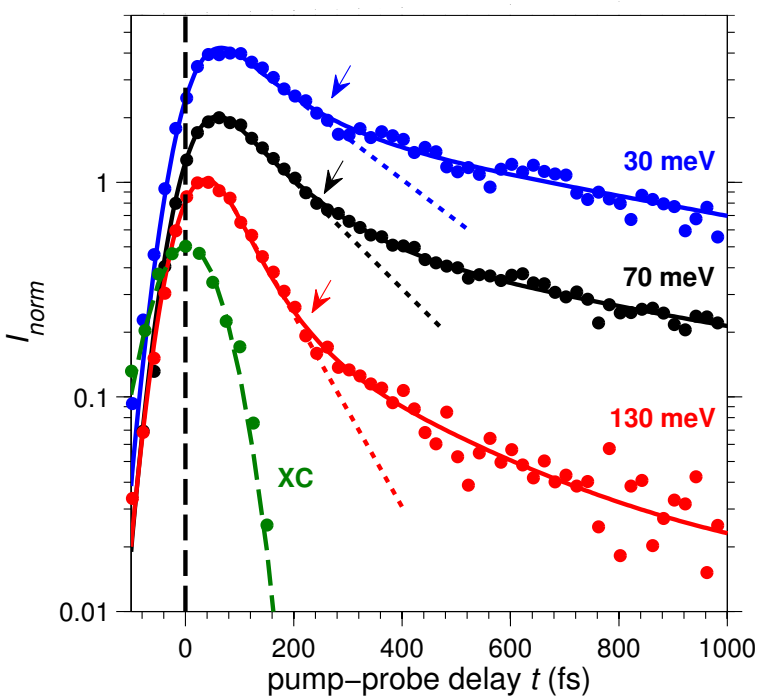
Step in lifetimes remains visible

Competition between e-p and e-e scattering

Mathiessen's rule appears not to hold.

Time-resolved ARPES experiment *by J.D. Rameau, S. Freutel, I. Avigo, M. Ligges, L. Rettig, P.D. Johnson, U. Bovensiepen*



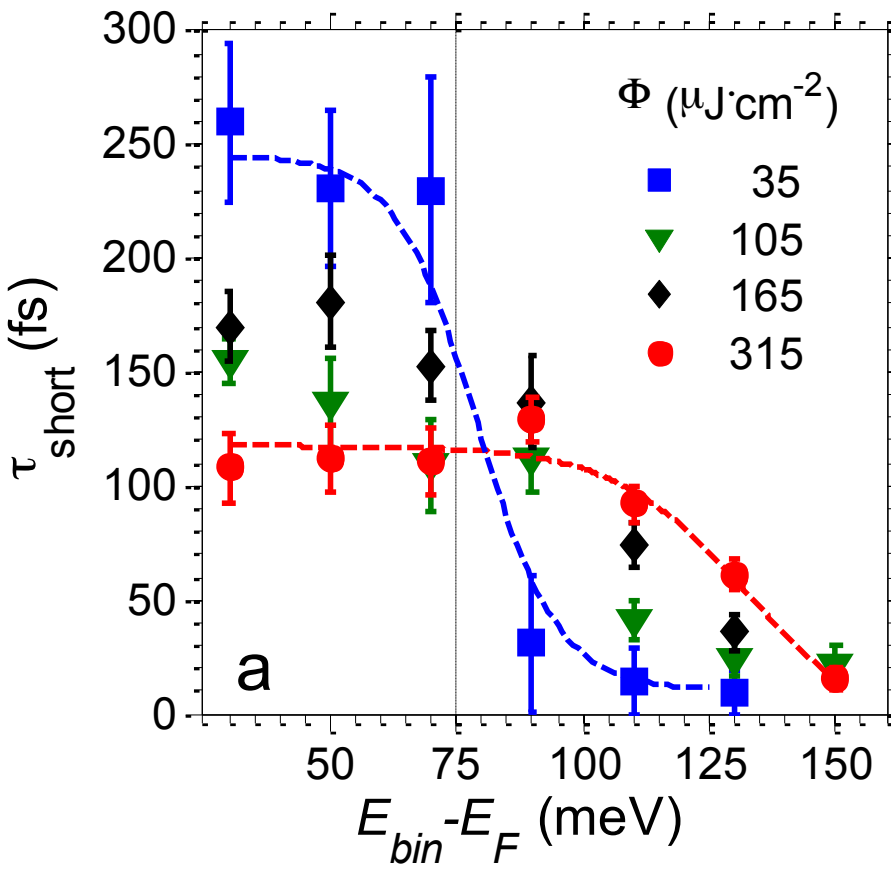


Population decay time in Bi2212 as a function of binding energy

arXiv:1505.07055

Experiment

Theory

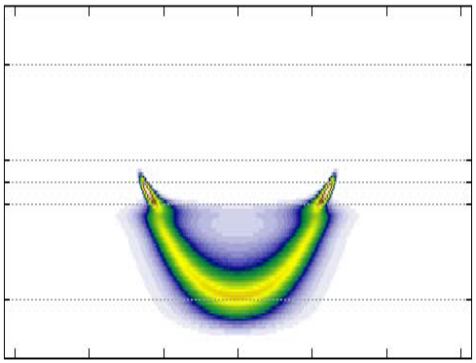


Decay rates are primarily reflective of energy transfer processes

Population dynamics can be understood with a strongly coupled boson at approximately 75 meV and $\lambda \approx 0.2$

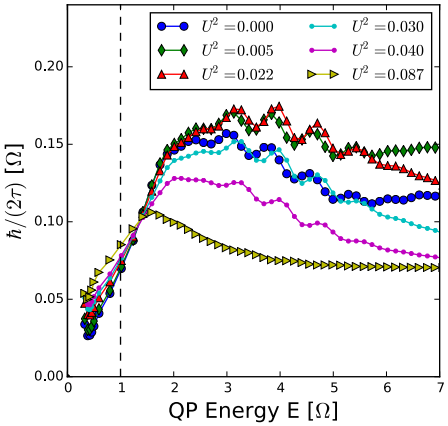
Quantitative agreement between experiment and theory

Summary



- Dynamics in the time domain are not always equivalent to frequency domain – Mathiessen's rule is violated

- Dynamics in the time domain are principally controlled by energy transfer processes



  - We can use this to separate/suppress interactions that can obscure electron-boson interactions

  - Interpreting changes in the spectra using equilibrium language can be erroneous

PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
arXiv:1505.07055

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- Higgs oscillations in superconductors

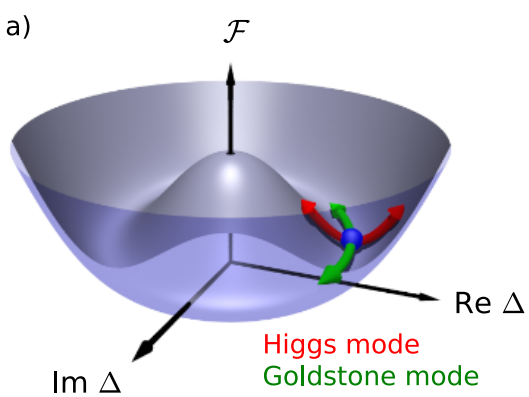
Collisionless relaxation of the energy gap in superconductors

A. F. Volkov and Sh. M. Kogan

Institute of Radio and Electronics, USSR Academy of Sciences

(Submitted June 15, 1973)
Zh. Eksp. Teor. Fiz. **65**, 2038–2046 (November 1973)

Equations are derived for equal-time Green's functions and describe the dynamics of superconductors during a time that is short in comparison with the electron energy relaxation times τ_{ph} and τ_{ee}. The time evolution of small initial perturbations of the order parameter Δ is investigated. It is established that following initial perturbations of a definite type the energy gap relaxes only as a result of inelastic collisions of the electrons during a time on the order of τ_{ph}, τ_{ee}. In the general case, the order parameter at $t \ll \tau_{ph}$, τ_{ee} oscillates with a frequency $\sim 2\Delta$ and with an amplitude that attenuates asymptotically with time in accord with a power law.



"… there is no coupling of ordinary condensed matter probes to it."

-Pekker & Varma, Annual Reviews of Condensed Matter Physics Volume 6 (2015)

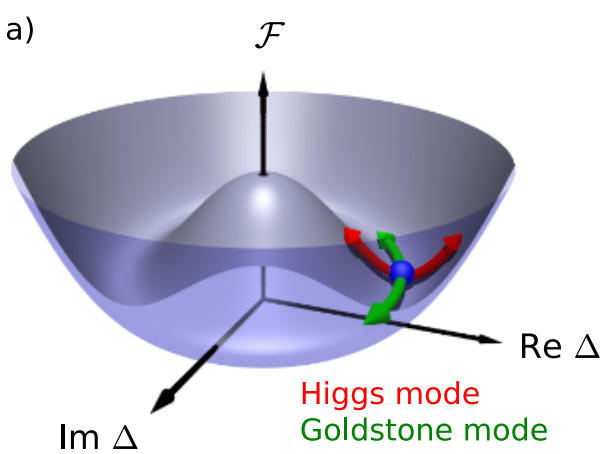
Also: *Barankov PRL 2004, Yuzbashyan PRL 2006, Tsuji 2013/2014, many others*

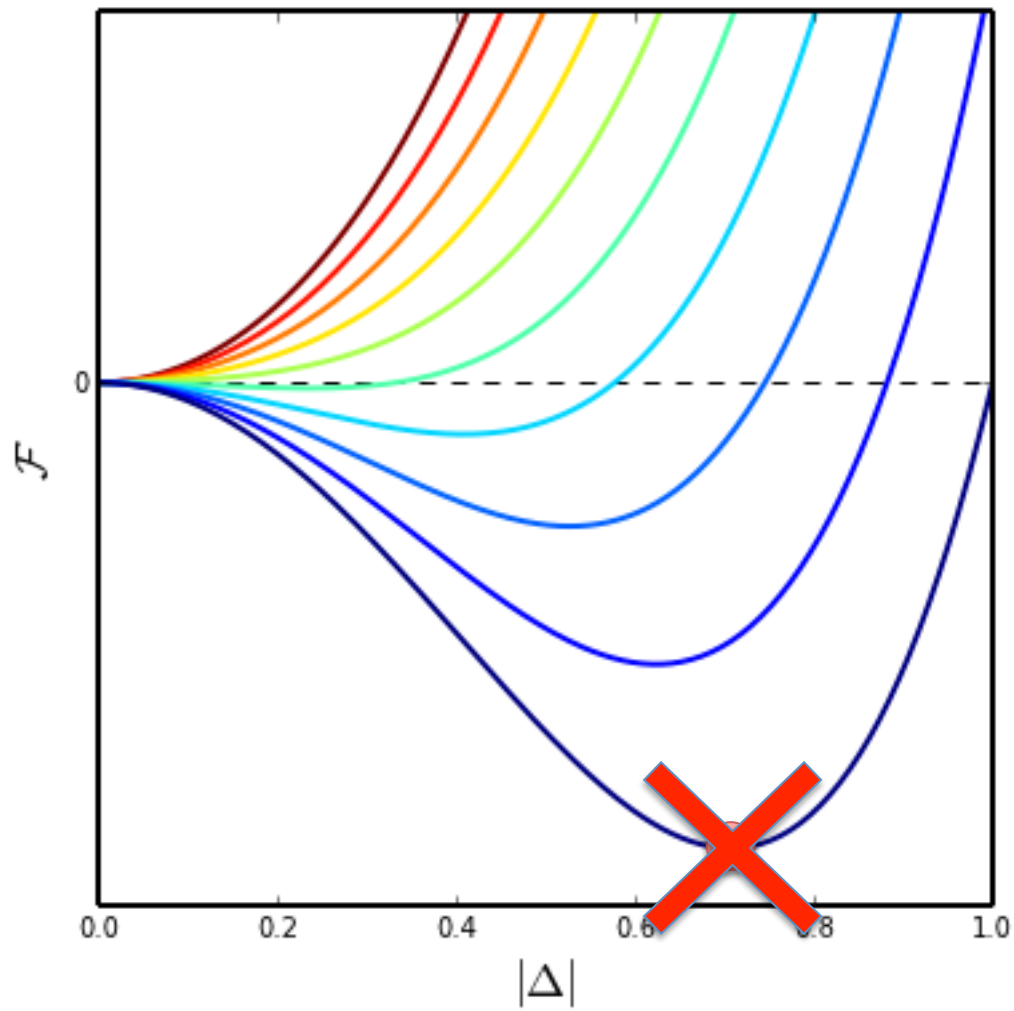
The problem of the Amplitude/Higgs oscillations

Classical theory of phase transitions

$$\mathcal{F} = \int \frac{\alpha}{2}|\Delta|^2 + \beta|\Delta|^4$$

$$\alpha \sim \frac{T - T_c}{T_c}$$

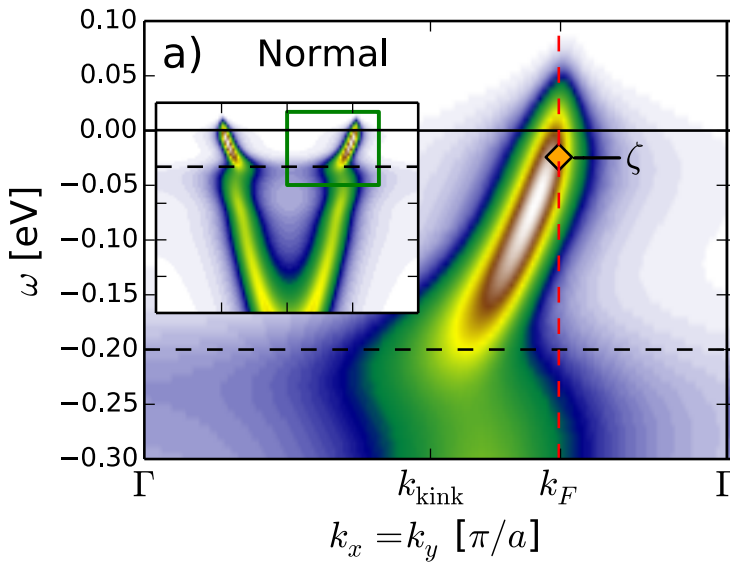




AFK et al., Phys. Rev. B 92, 224517 (2015).

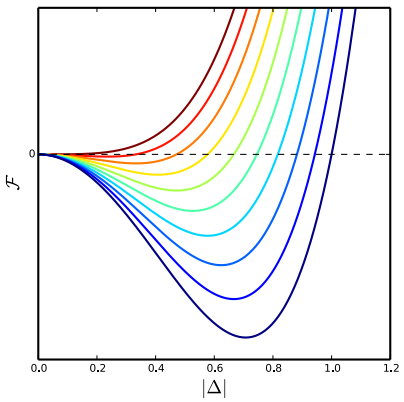
Amplitude mode oscillations in superconductors

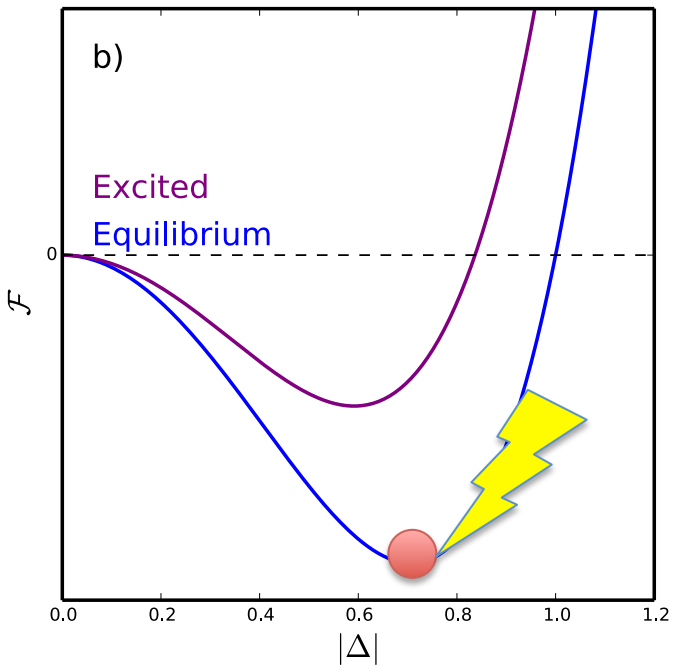
- Non-equilibrium superconductivity
- Migdal-Eliashberg theory of phonon mediated superconductor

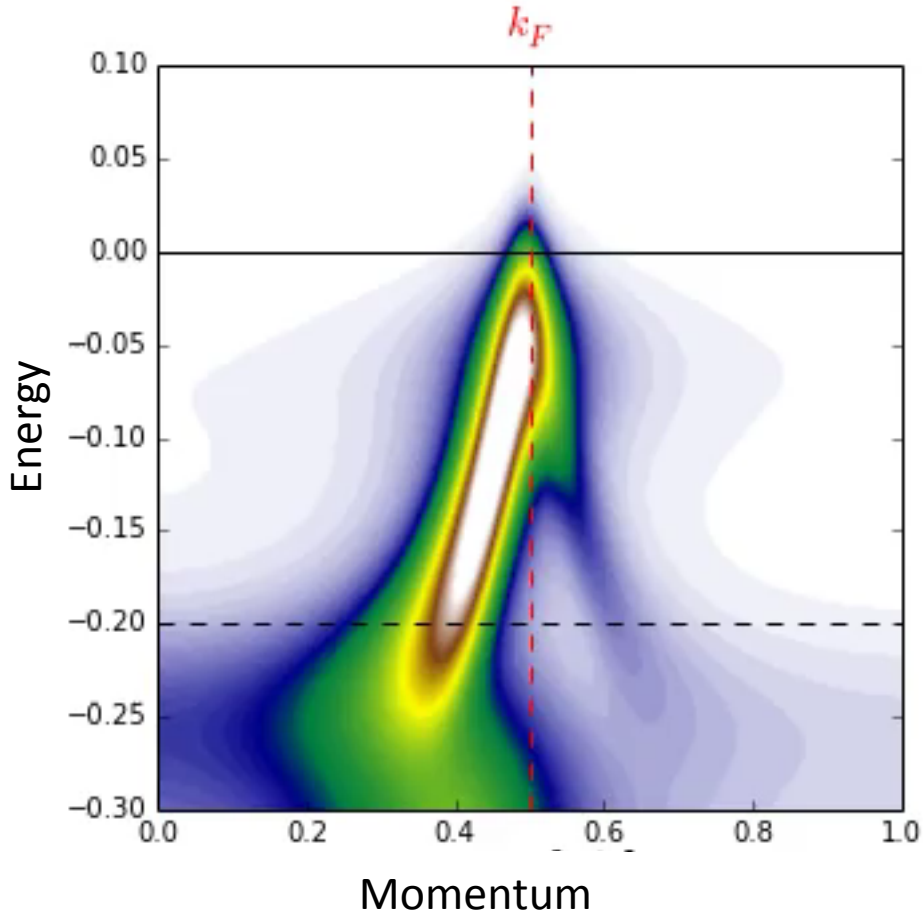


- Real-time driving with electric field via Peierls *k-A(t)*
- Dynamics of the electronically driven superconductor
- Amplitude/Higgs mode?

AFK, PRB (2015)





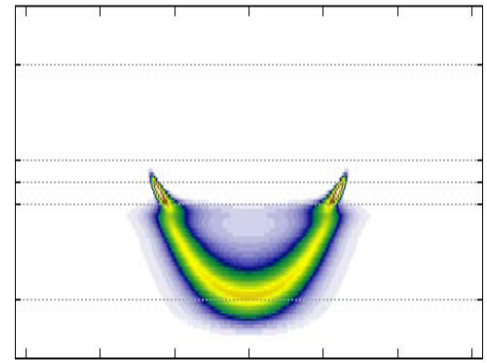


Observing Higgs oscillations only possible in non-equilibrium

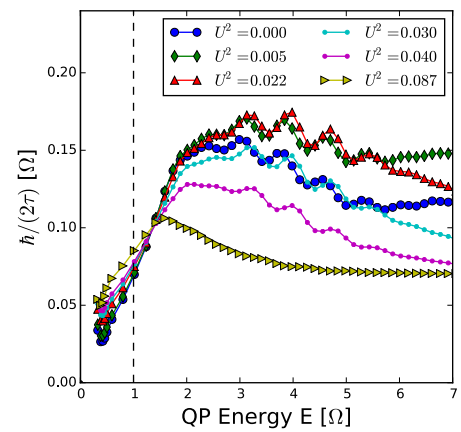
AFK et al., Phys. Rev. B 92, 224517 (2015).

Summary (redux)

- Dynamics in the time domain are not always equivalent to frequency domain domain – Mathiessen's rule is violated



- Dynamics in the time domain are principally controlled by energy transfer processes
  - We can use this to separate/suppress interactions that can obscure electron-boson interactions
  - Interpreting changes in the spectra using equilibrium language can be erroneous



- D-wave superconductors also show Higgs oscillations in pump-probe experiments with an isotropic frequency

PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
PRB 92, 224517 (2015)
PRB 93, 144506 (2016)
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