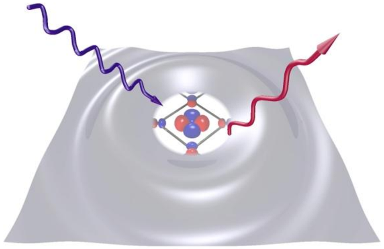
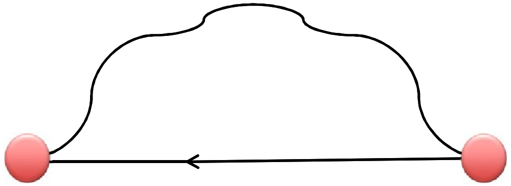
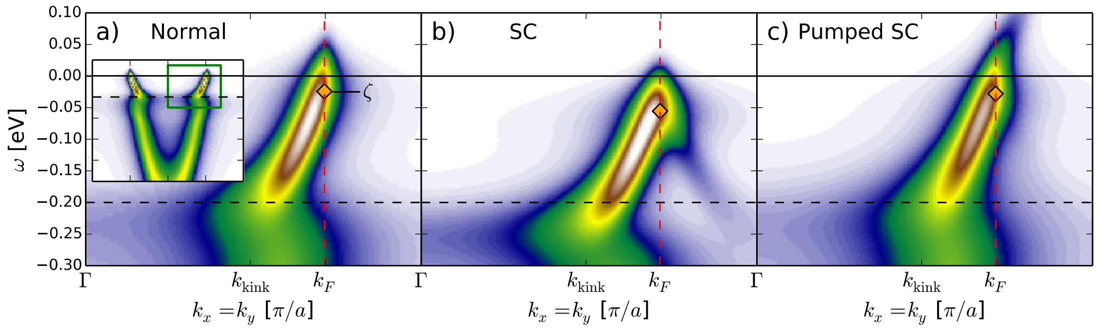
Violation of Matthiessen's rule in pump/probe TR-ARPES







Jim Freericks Georgetown University

Thomas Devereaux (Stanford/SLAC), **Alexander Kemper** (North Carolina State), **Brian Moritz** (SLAC), **Sona Najafi** (Georgetown), and **Michael Sentef** (Hamburg)

Georgetown work supported by DOE, BES, DE-FG02-08ER46542 and McDevitt bequest

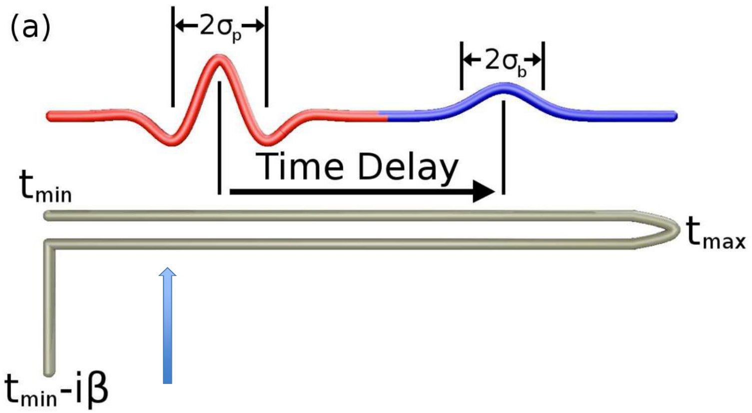
PRL ***102**, 136401 (2009)*
PRL ***111**, 077401 (2013)*
PRX ***3**, 041033 (2013)*
PRB ***87**, 235139 (2013)*
PRB ***90**, 075126 (2014)*
Adv. Imag. Elect. Phys (2015)
PRB ***92**, 224517 (2015)*
Entropy ***18**, 180 (2016)*
Fort. Phys. ***64** (2017).*

Nonequilibrium many-body theory

Lesser Green function: $G_{ij}^{<}(t,t') = iZ_{eq}^{-1}\mathrm{Tr}[e^{-\beta H_{eq}} c_j^{\dagger}(t') c_i(t)]$

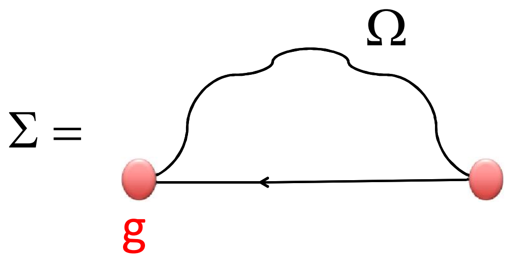
Include the effects of a strong driving field via the Peierls' substitution – $\boldsymbol{k} \rightarrow \boldsymbol{k} - e\mathbf{A}(t)$

for a spatially uniform but time-dependent electric field

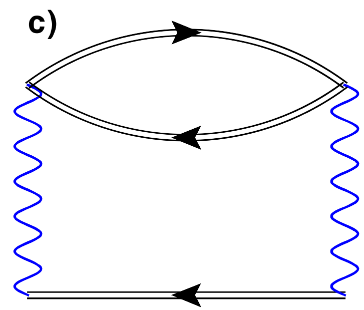


…and about its history

System knows about its thermal initial state…

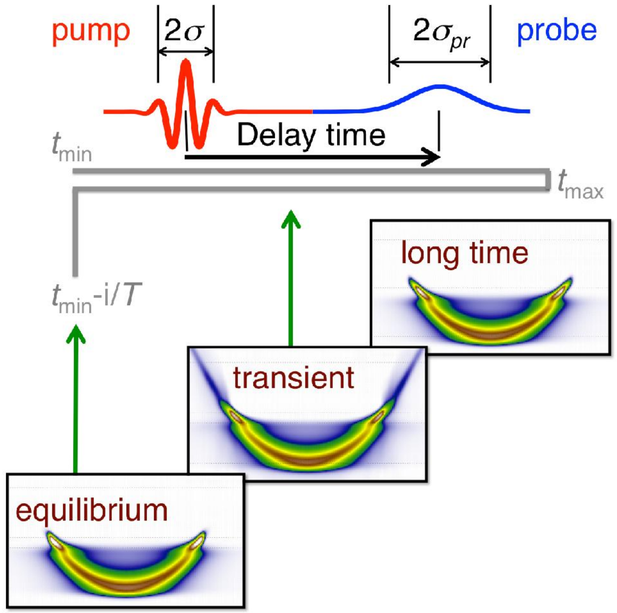


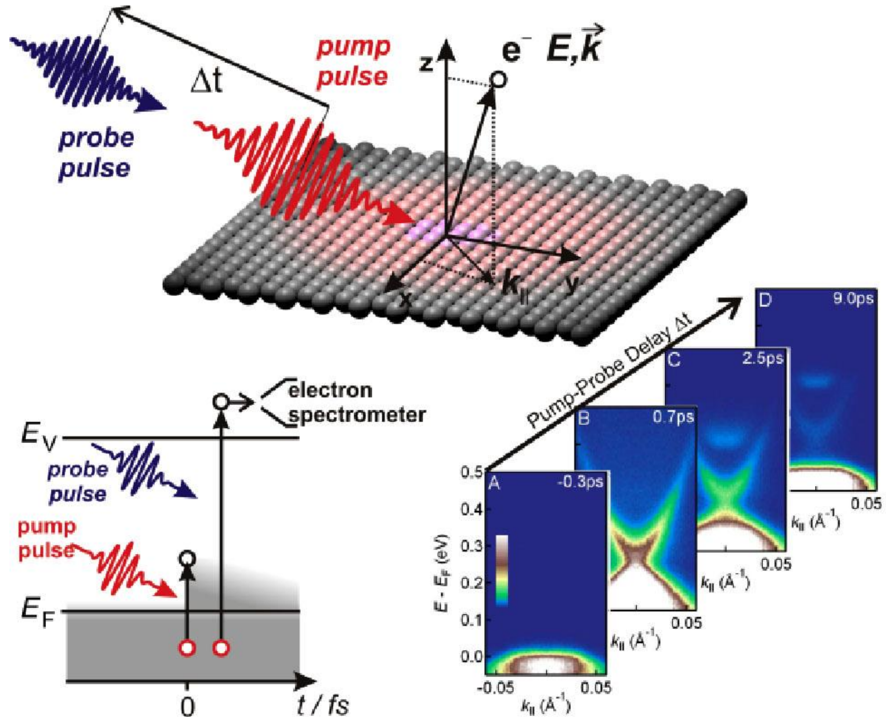
Electron-phonon coupling included via perturbation theory invoking Migdal's theorem



Electron-electron coupling via second-order perturbation theory

Time-resolved ARPES





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

Image source: FHI Berlin

*Freericks, et al., PRL **102**, 136401 (2009); Freericks, et al., Physica Scripta **2015** T165 014012 (2015); Freericks and Krishnamurthy, Photonics **3** 58 (2016)*

Five myths about excitation and relaxation

1. Changes in spectral functions arise from changes in the instantaneous eigenvalues
2. Once excited, a many-body system must relax
3. If it relaxes, the self-energy governs the relaxation rate
4. Electrons thermalize amongst themselves to form a hot thermal state that subsequently relaxes energy to the phonon bath (hot-electron model)
5. Relaxation times are automatically separated in the time domain.

Five realities about excitation and relaxation

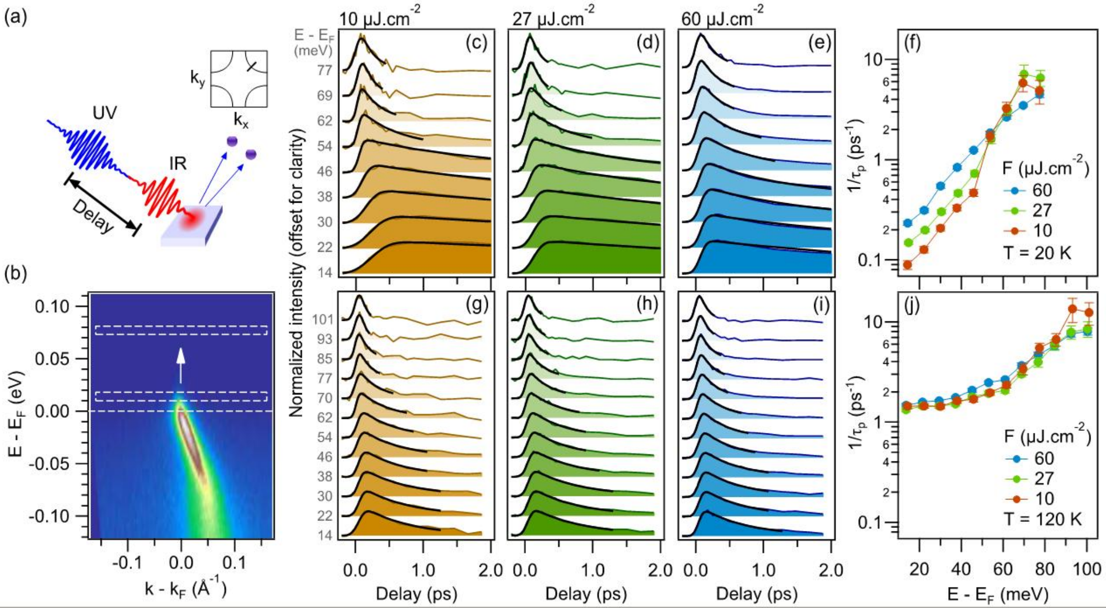
1. Systems without dipolar coupling matrix elements do not change their instantaneous eigenvalues
2. Excited systems can only relax if energy can be removed from them
3. The rate of relaxation is related to the self-energy, but in a complex fashion
4. It is the deviations from hot-electron behavior that determine the relaxation
5. Relaxation times separate in the time domain because of energy bottlenecks and the violation of Mathiessen's rule

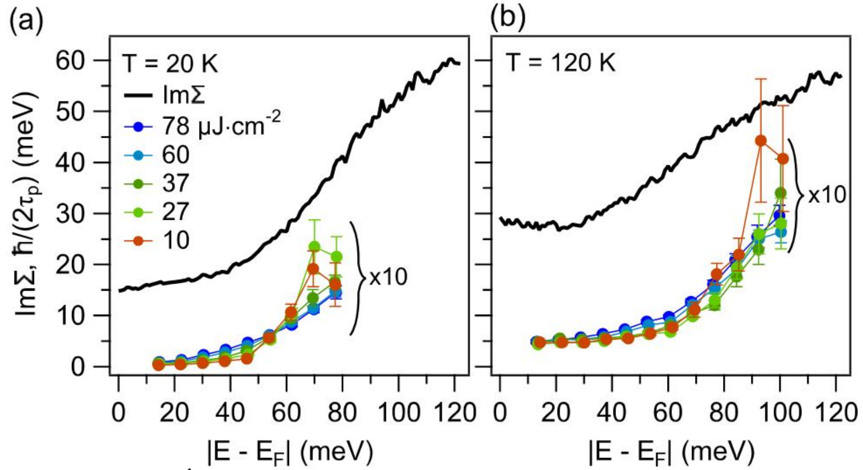
Clear difference between relaxation and line-widths

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

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

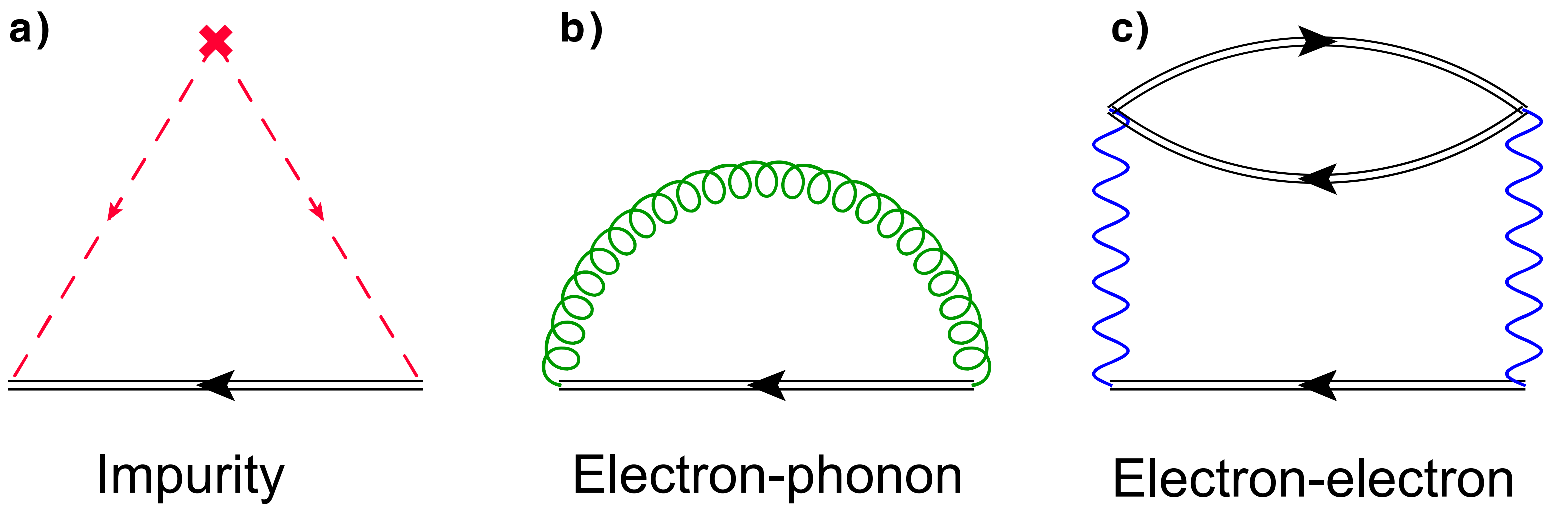
S.-L. Yang,[1,2] J. A. Sobota,[1,3] D. Leuenberger,[1,2] Y. He,[1,2] M. Hashimoto,[4] D. H. Lu,[4] H. Eisaki,[5]
P. S. Kirchmann,[1,*] and Z.-X. Shen[1,2,†]





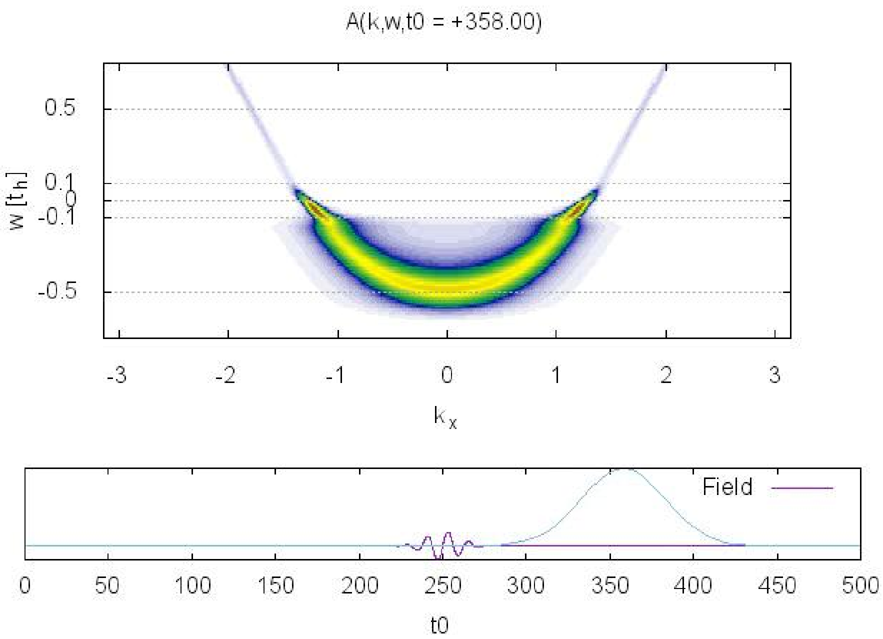
$$\frac{1}{\tau(\omega)} \underset{?}{=} -2\mathrm{Im}\Sigma(\omega = \epsilon_{\mathbf{k}})$$

Three Types of Interactions

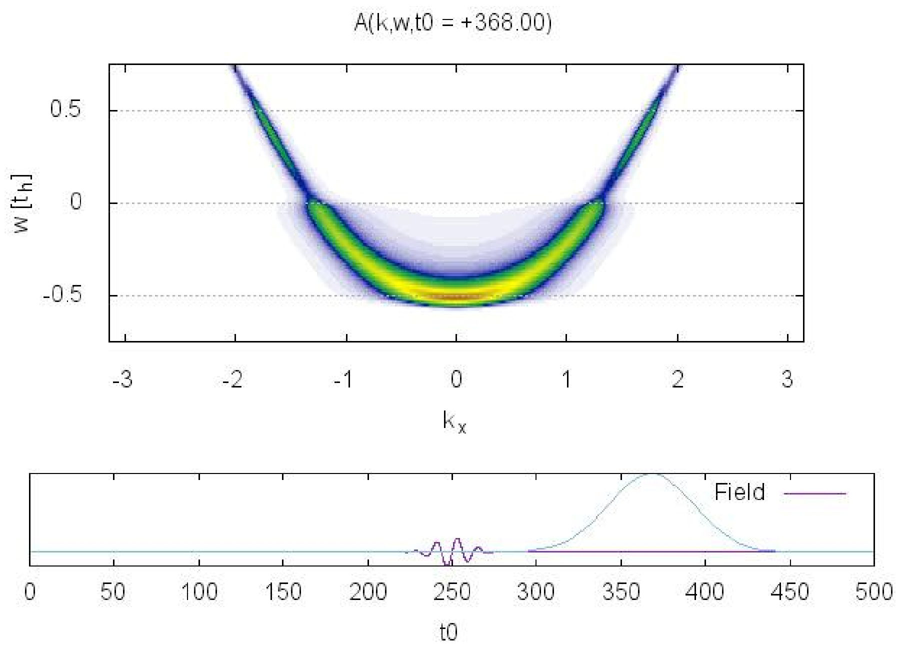


Electron phonon versus impurity

Electron-phonon

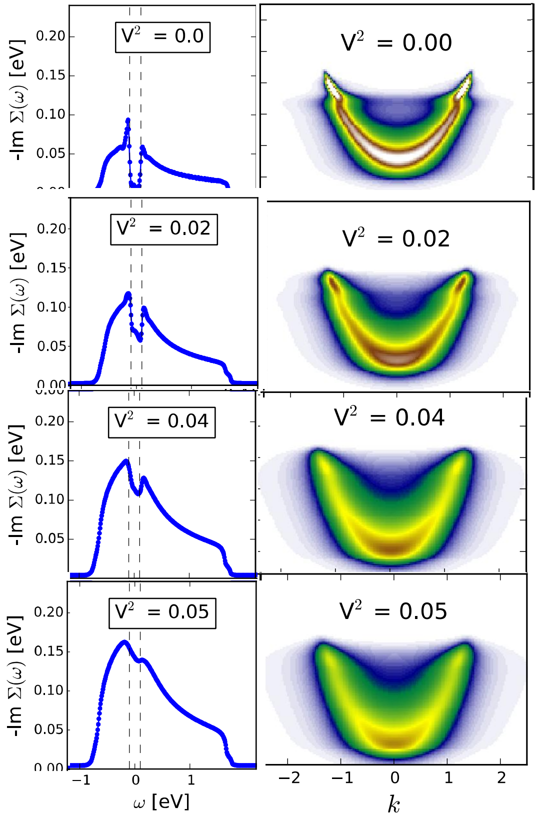


Impurity

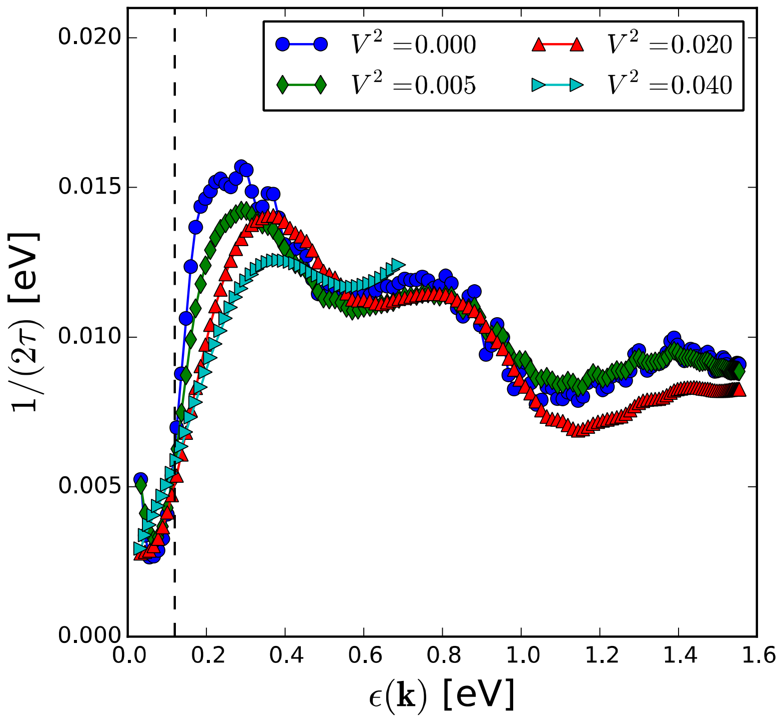


Electron-phonon and impurity

Equilibrium: quasiparticle



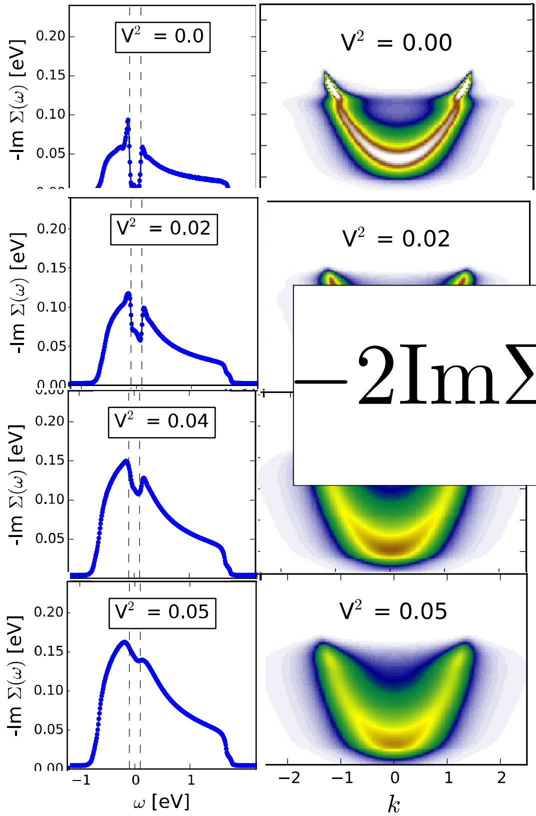
Non-equilibrium: population

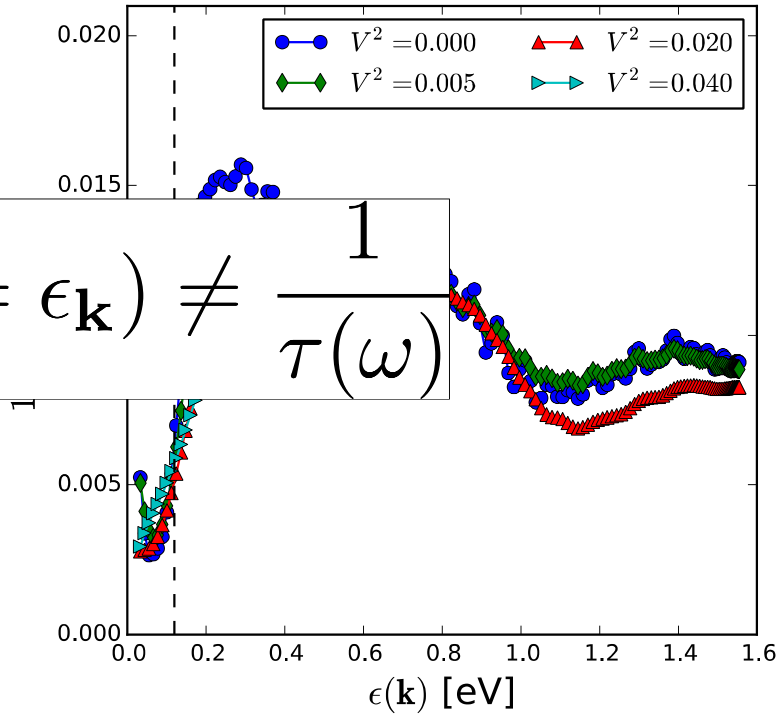


Electron-phonon and impurity

Equilibrium: quasiparticle

Non-equilibrium: population

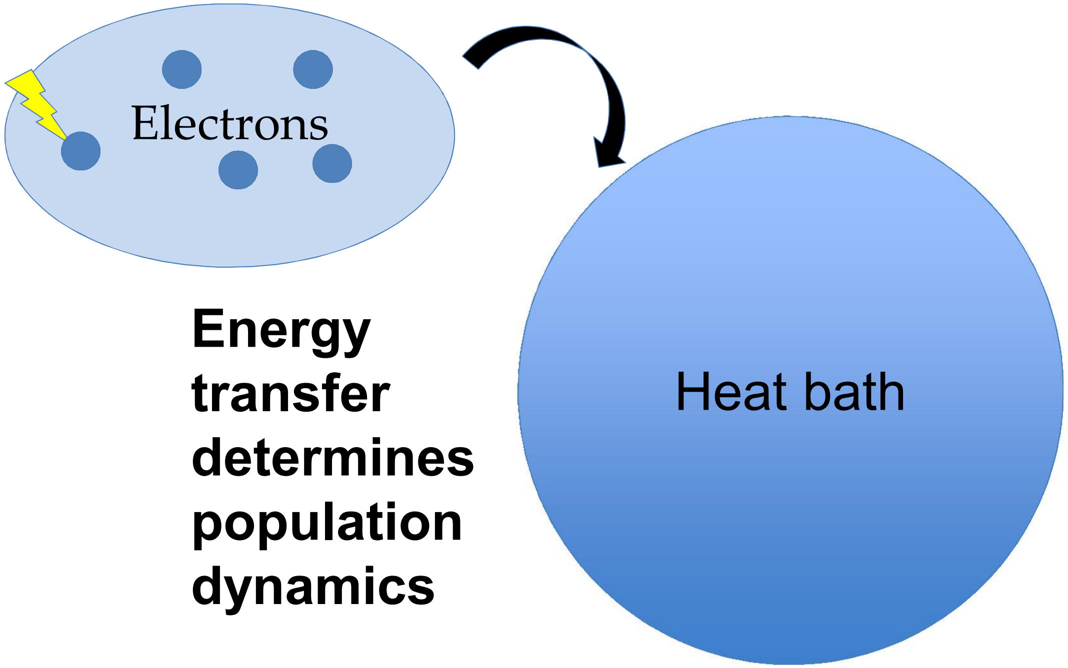




$$-2\mathrm{Im}\Sigma(\omega = \epsilon_{\mathbf{k}}) \neq \frac{1}{\tau(\omega)}$$

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

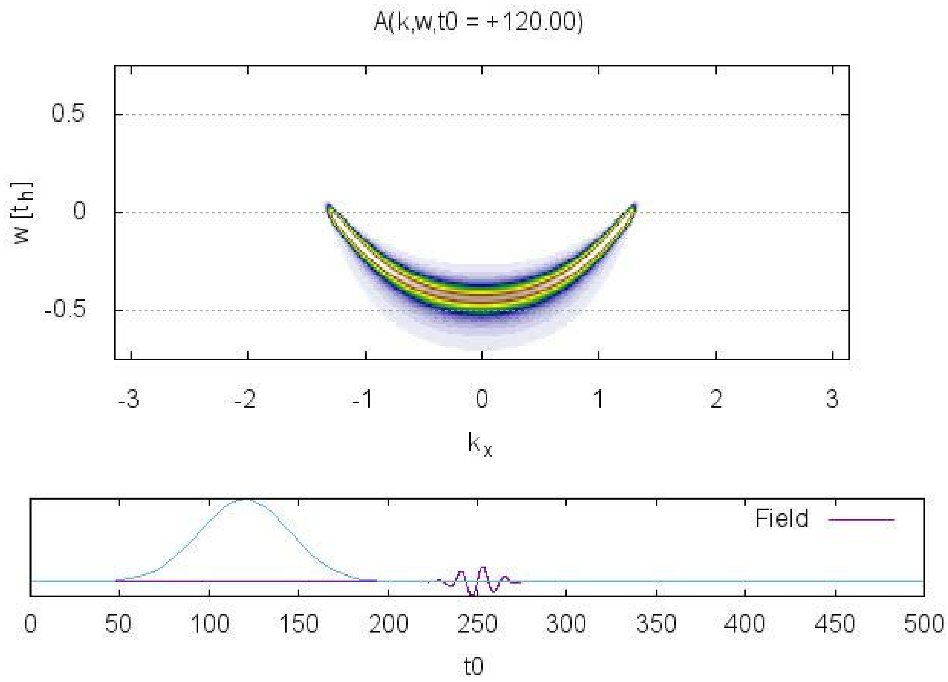
Violation of Matthiessen's rule comes from energy transfer bottlenecks

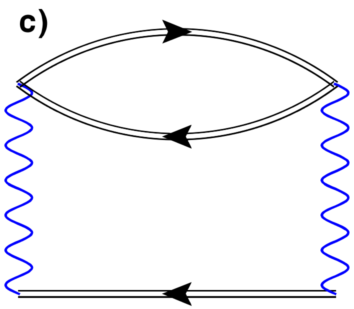


Energy transfer determines population dynamics

Electron-electron interaction

Electron-
electron

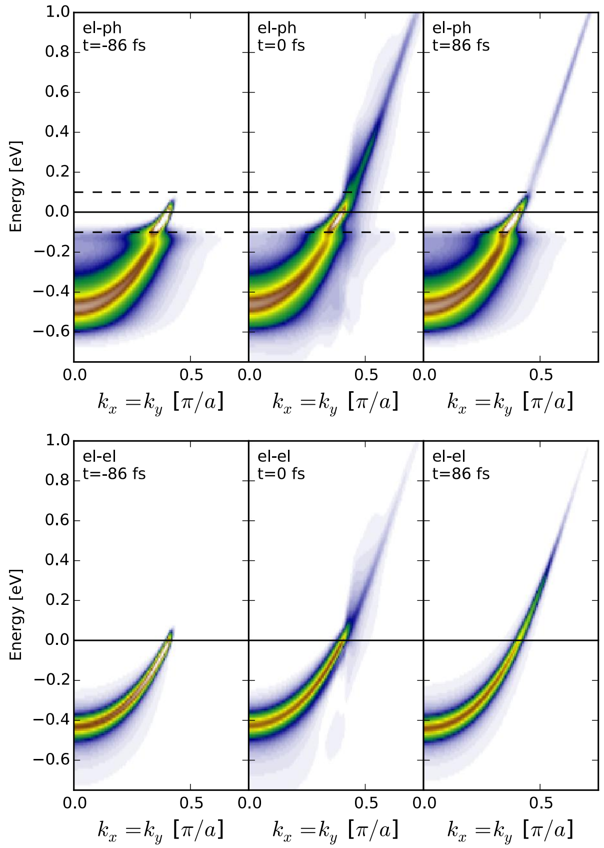




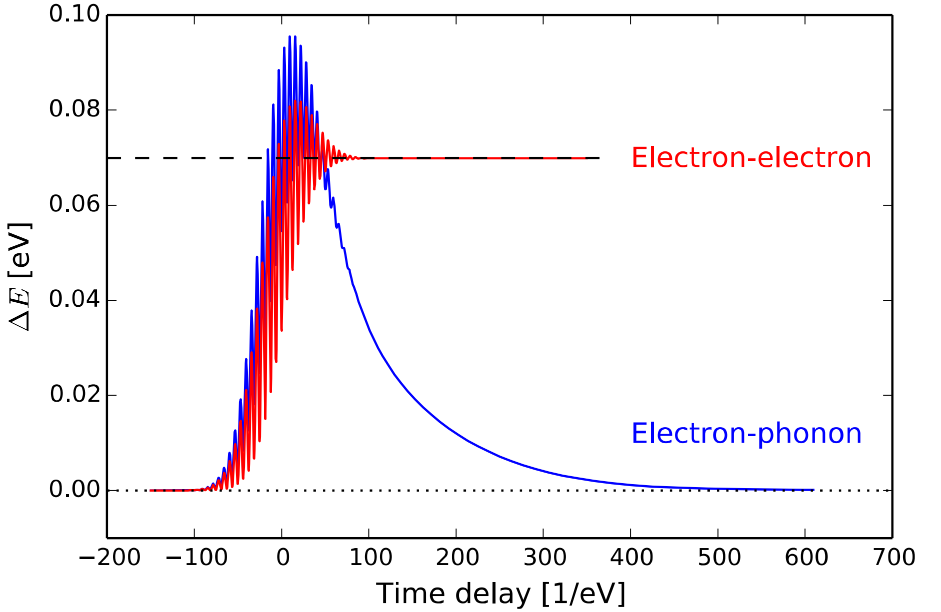
Electron phonon vs electron-electron

Electron-phonon

Electron-electron

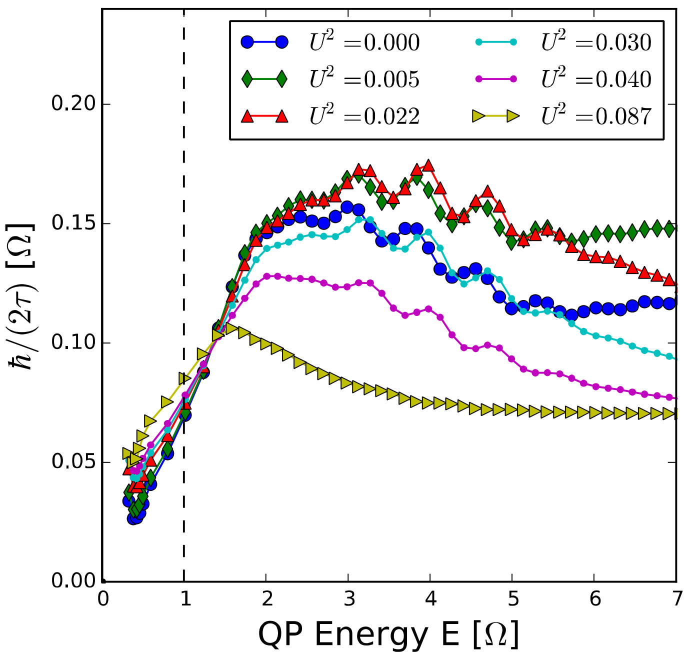


Change in total electronic energy



Electron-phonon and electron-electron

$$g^2 = 0.02$$



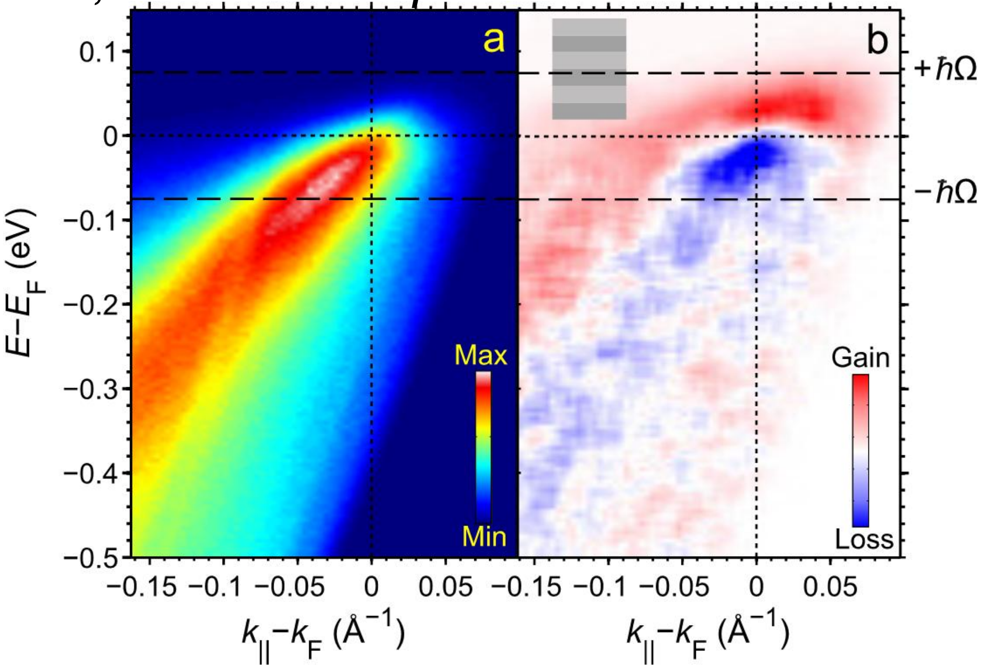
Step in lifetimes remains visible

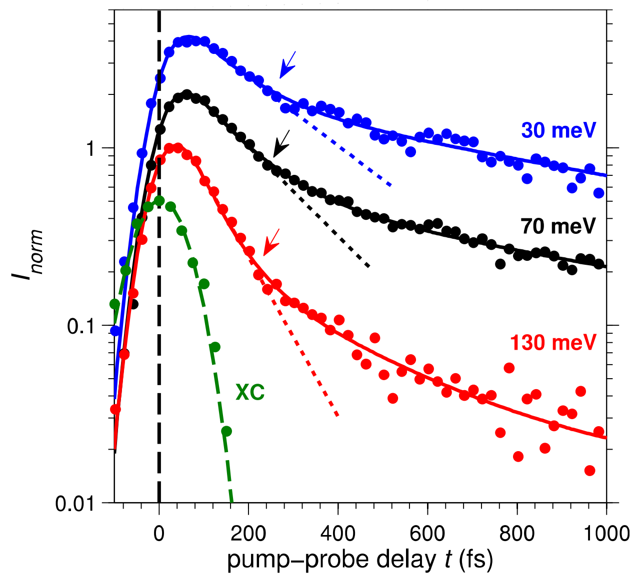
Competition between e-p and e-e scattering

Matthiessen's rule appears not to hold.

TR-ARPES experiment showing this

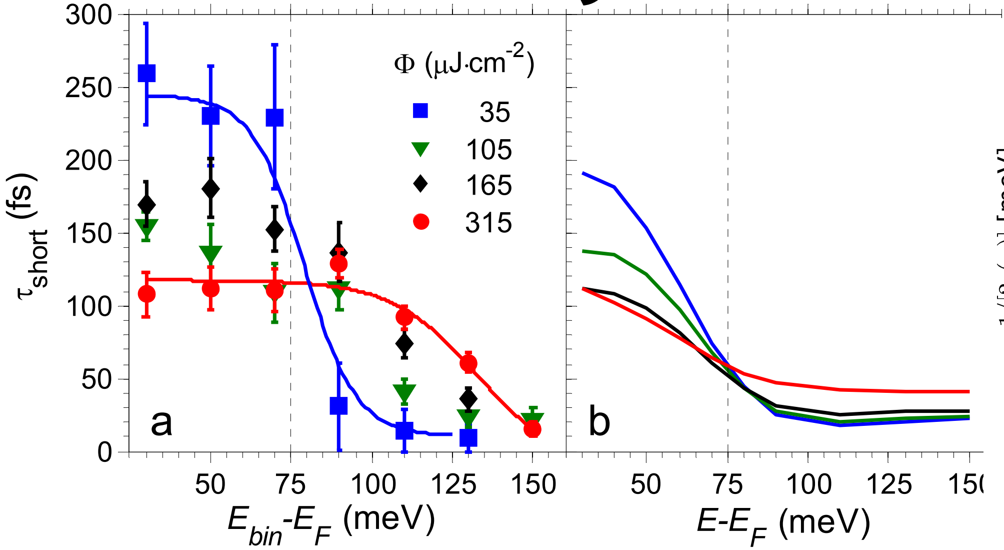
Time-resolved ARPES experiment *by J.D. Rameau, S. Freutel, I. Avigo, M. Ligges, L. Rettig, P.D. Johnson, U. Bovensiepen*

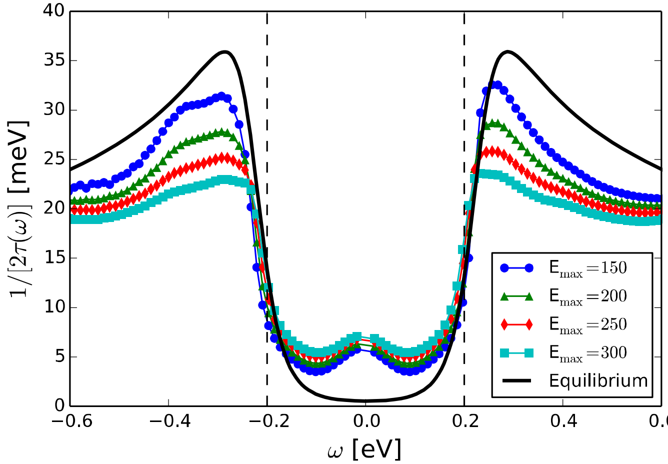




Population decay time in Bi2201 as a function of binding energy

Decay rates depend on fluence



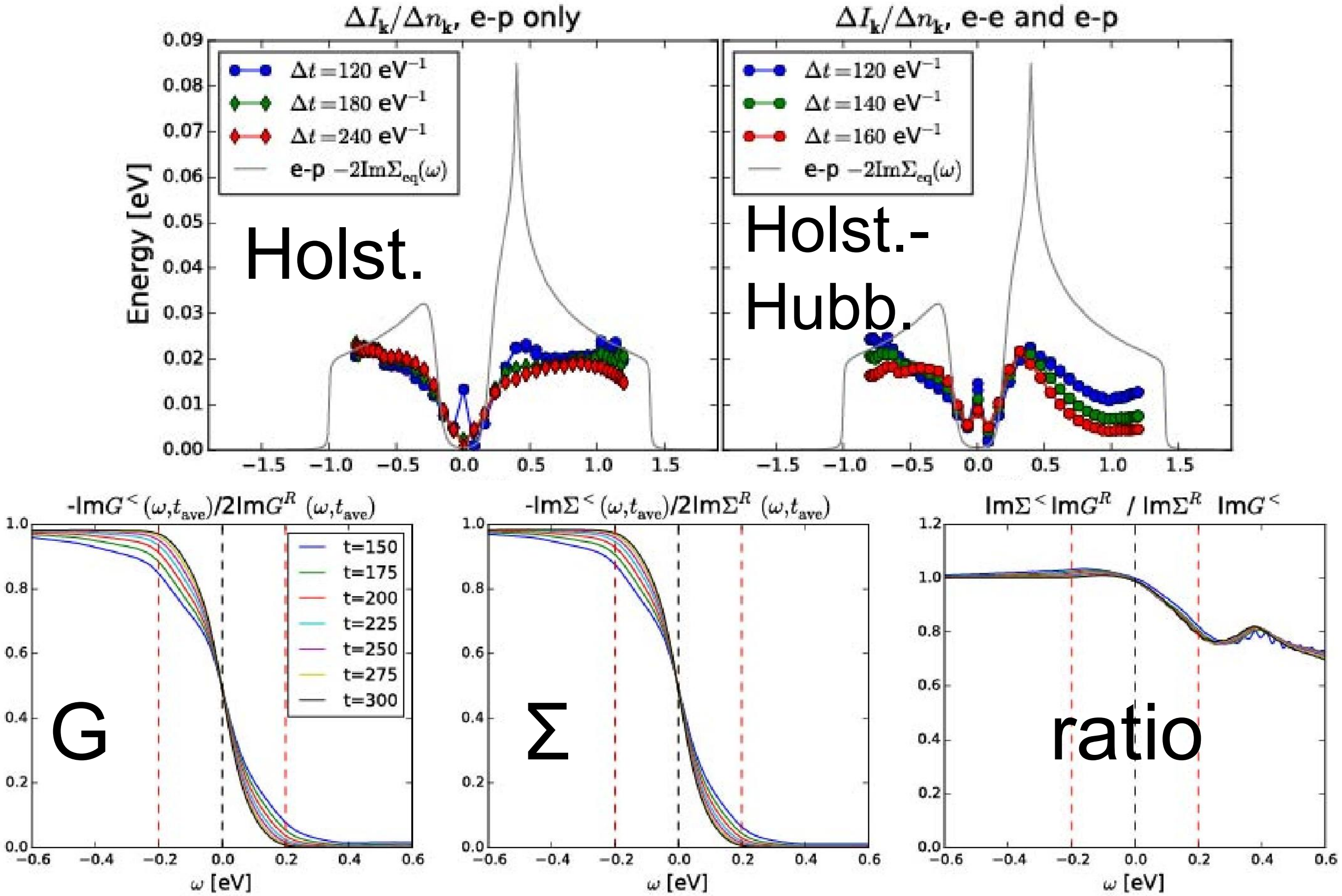


Decay rates are primarily reflective of energy transfer processes

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

Relaxation is complicated—not just hot electrons



Relaxation rate not given just by self-energy… (Entropy **18** 2016)... Different distribution function for G and Σ … (Fort. Phys. **64** 2017)

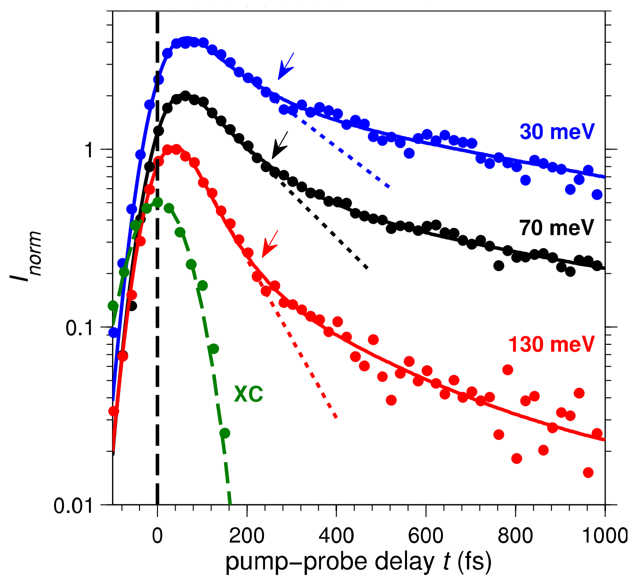
Conclusions

Relaxation is not always determined by equilibrium relaxation rates.

Energy dissipation bottleneck governs the relaxation process. If a boson dissipates energy, you can measure its effective relaxation rate.

Using equilibrium concepts in nonequilibrium can be fraught with peril.

Still need to figure out precisely how relaxation rate is determined. We are making progress, but not there yet.



Thanks to

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