Many-body physics approach to time-resolved photoemission spectroscopy in strongly correlated electron materials

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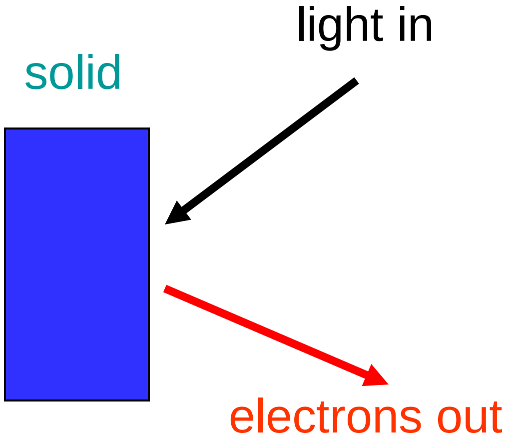
Funding from NSF (formalism development) and DOE (TR-PES data)

Thanks to Tom Devereaux, Sasha Joura, H. Krishnamurthy, Brian Mortiz, Thomas Pruschke, Volodmyr Turkowski and Veljko Zlatic' for collaborating on this work.

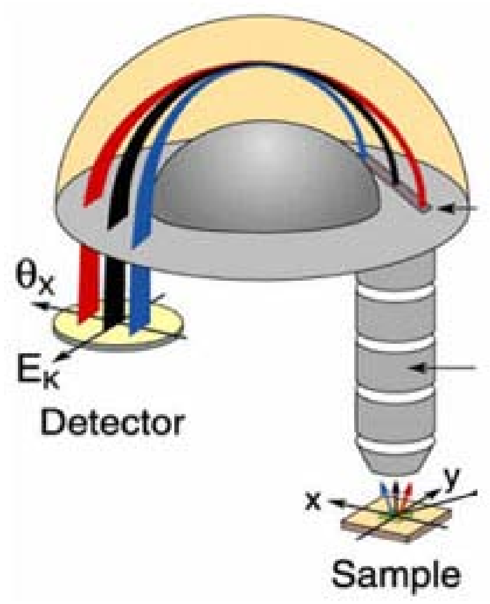
Time resolved PES

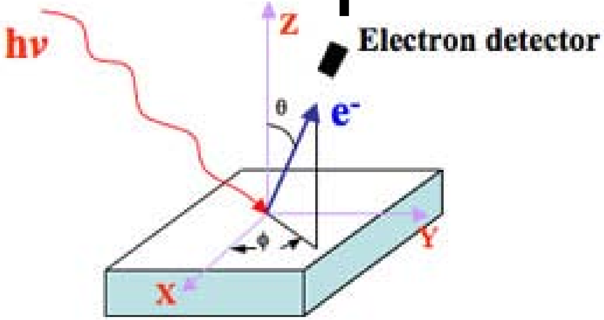
Photoelectric effect

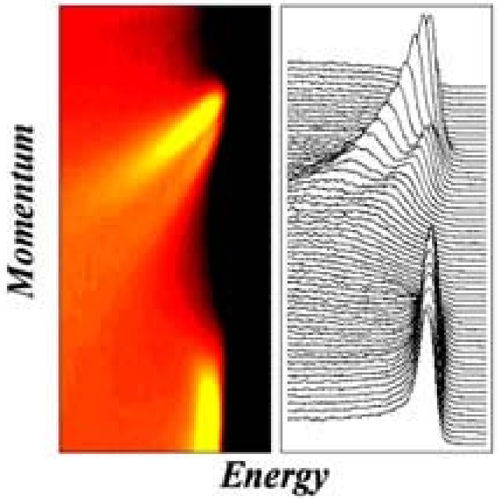
Einstein's Nobel prize winning work used the "old theory" of quantum mechanics to explain the photoelectric effect: Light carries discrete packets of energy related to its color. Energetic enough photons can eject electrons from the material.



Modern angle resolved photoelectron spectroscopy



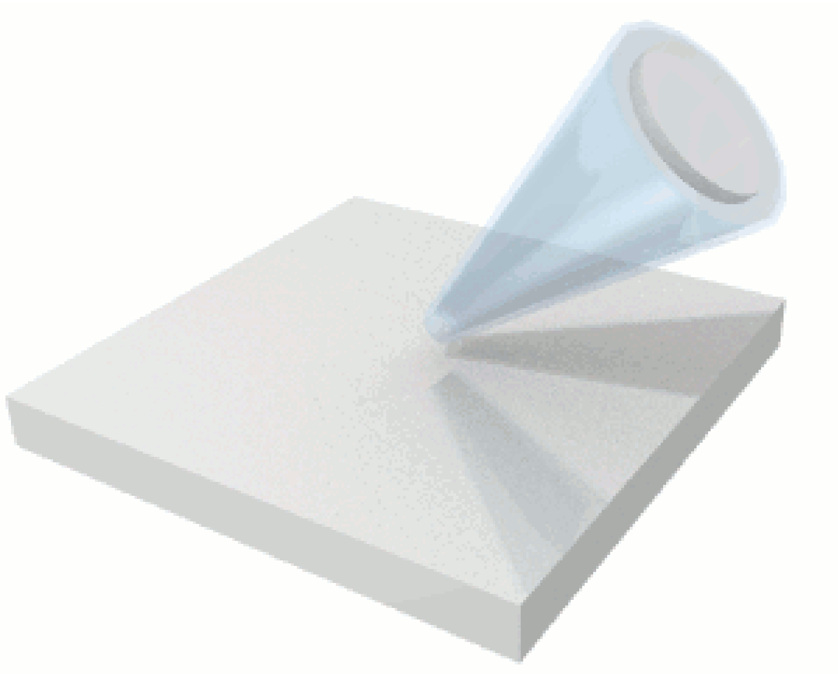




But continuous beam ARPES only measures information about the occupied states in equilibrium.

(Images from Z.-X. Shen's group)

Time-resolved pump/probe photoelectron spectroscopy



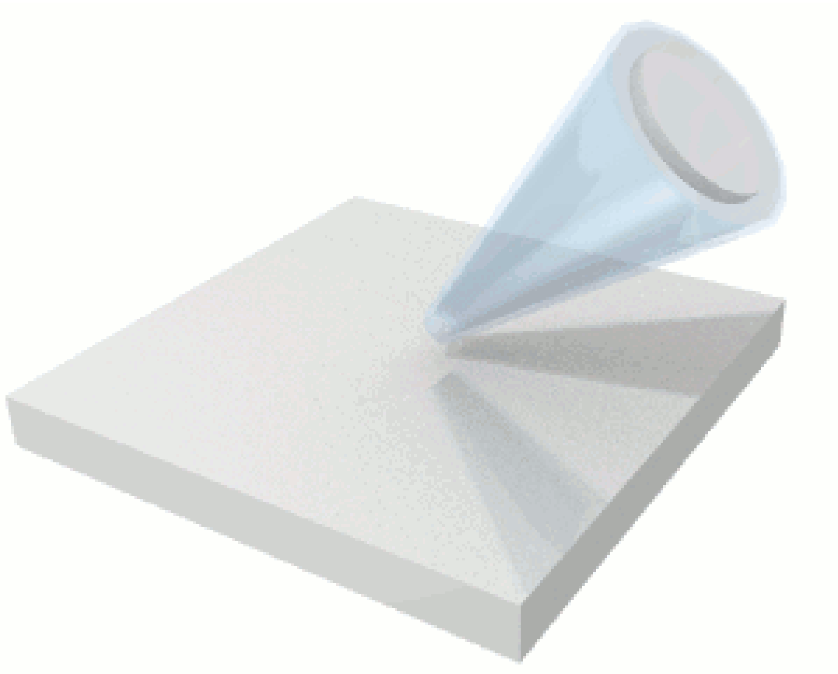
Schematic of a TR-PES experiment

(from Z.-X. Shen's group)

Pump the system into an excited nonequilibrium state with an intense pulse of light.

Probe with a short pulse of light energetic enough to photo-emit electrons.

Time-resolved pump/probe photoelectron spectroscopy



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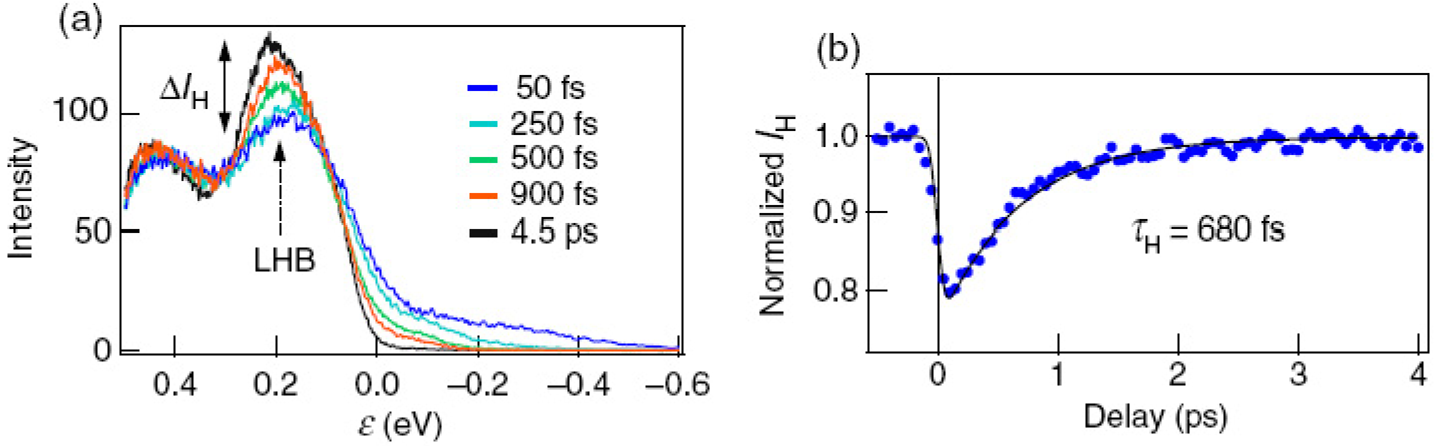
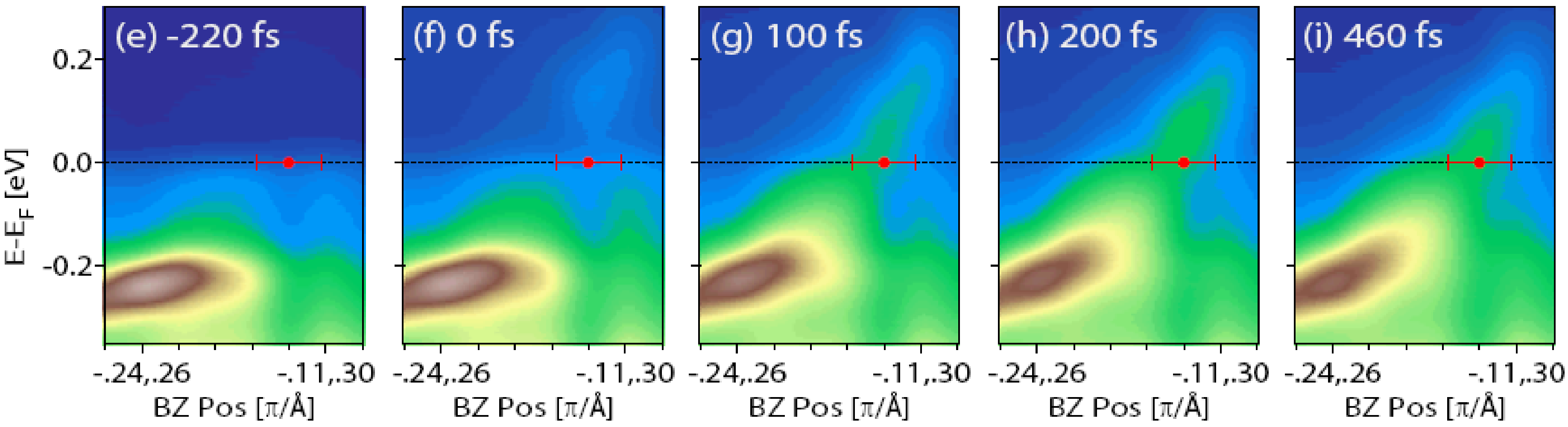
TR-PES on TaS_2



Experimental results by **Perfetti et al.** New Journal of Physics (2008)

One can clearly see that for short time delays the signal has much more high-energy spectral weight, which then relaxes to equilibrium at long times.

TR-PES on $TbTe_3$



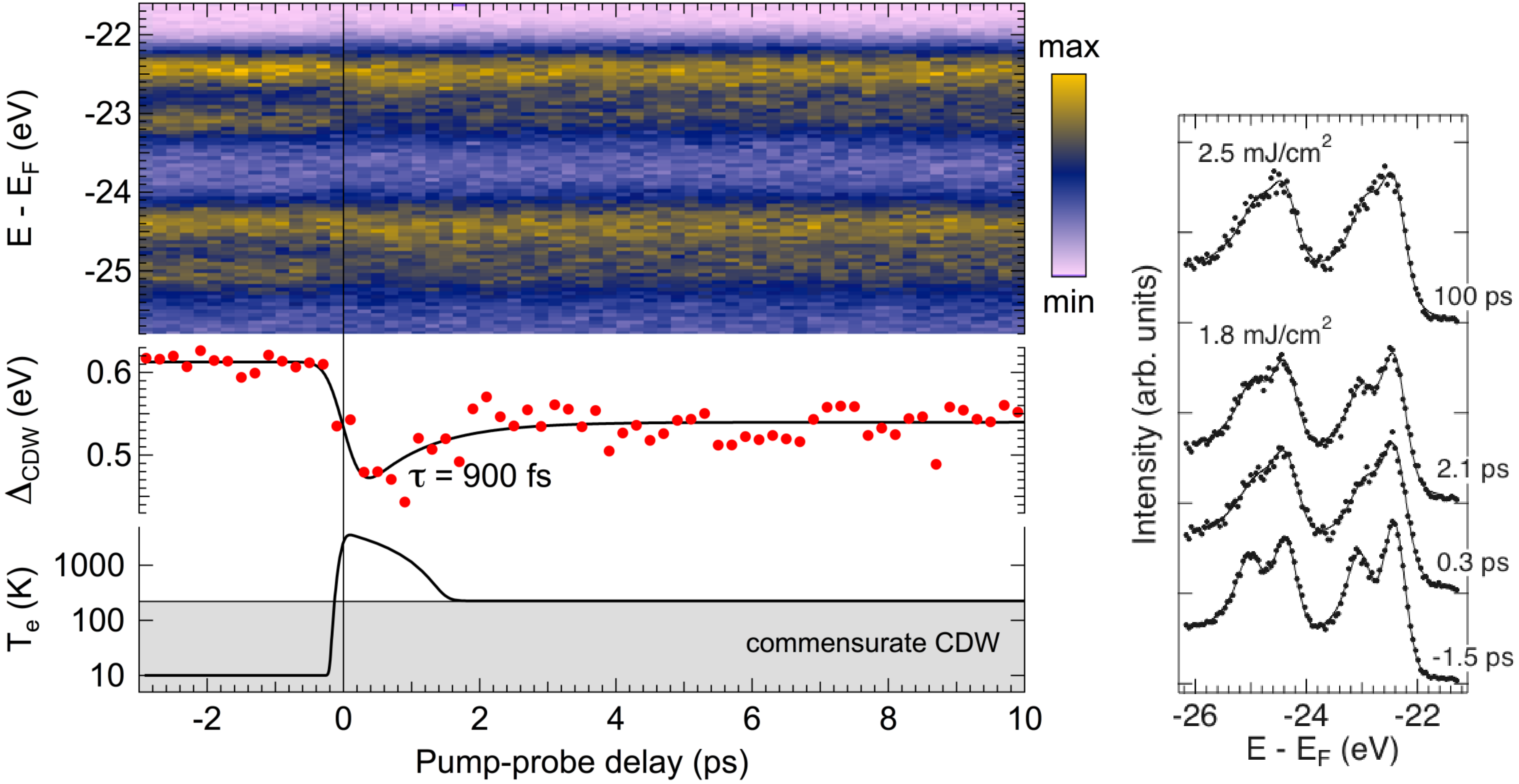
Experimental results by **Schmitt et al.** Science (2008)

Here one sees excitations above the Fermi energy at specific locations in the Brillouin zone.

Ultrafast melting of a CDW (TaS_2)

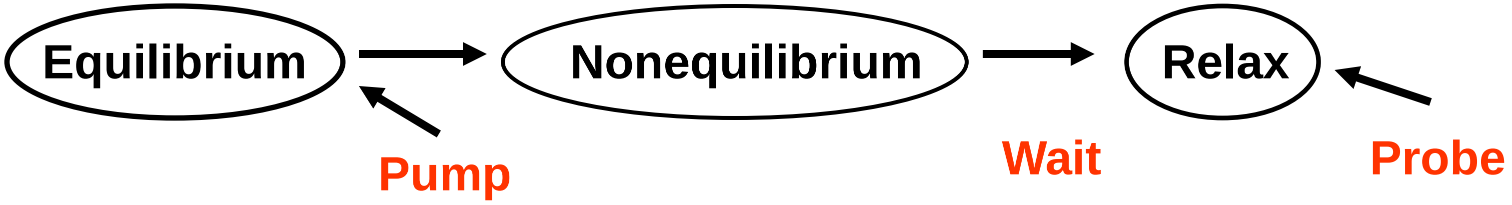
Beamline PG2, FLASH, Hamburg, **Expt by Rossnagel's group**

$T = 10$ K. $\hbar\omega = 156$ eV. $\Delta E = 300$ meV. $\Delta\tau = 700$ fs



TR-PES formalism

We use a scattering state approach with the sudden approximation. (More exact developments are also possible, but won’t be described here.)


Equilibrium
Nonequilibrium
Relax
Pump
Wait
Probe

TR-PES formalism

Time-resolved photoemission spectroscopy is found by performing a Fourier transform of the nonequilibrium two-time lesser Green's function weighted by the probe envelope functions.

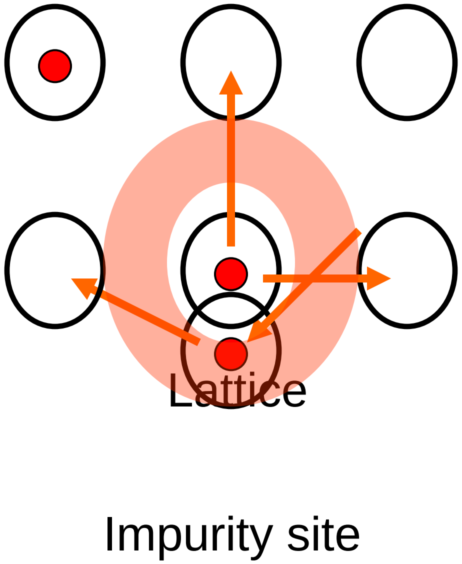
$$P_{TR-PES}(t) = -i\int_0^t dt' \int_0^t dt'' s(t')s(t'')e^{i\omega(t'-t'')}G^{<}_{loc}(t',t'')$$

Broad probes--good energy, poor time.
Narrow probes--good time, poor energy.

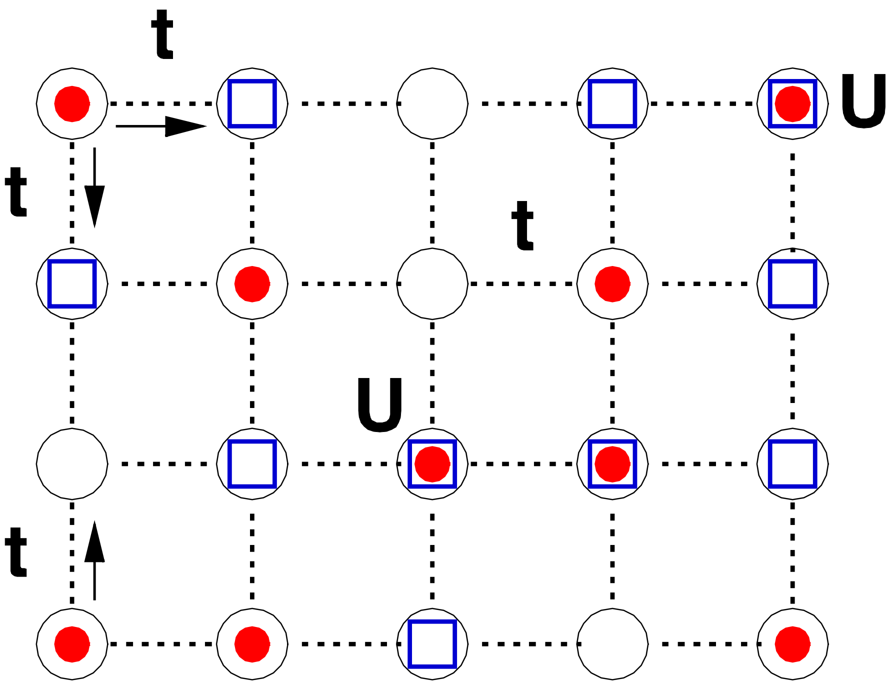
Many-body physics and the dynamical mean-field theory approach to nonequilibrium problems

Dynamical mean field theory

- Models of strongly correlated materials are difficult to solve.
- Significant progress has been made over the past 20 years by examining the limit of **large spatial dimensions**.
- In this case, the lattice problem can be mapped onto a self-consistent impurity (single-site) problem, in a time-dependent field that **mimics the hopping of electrons onto and off of the lattice sites**.

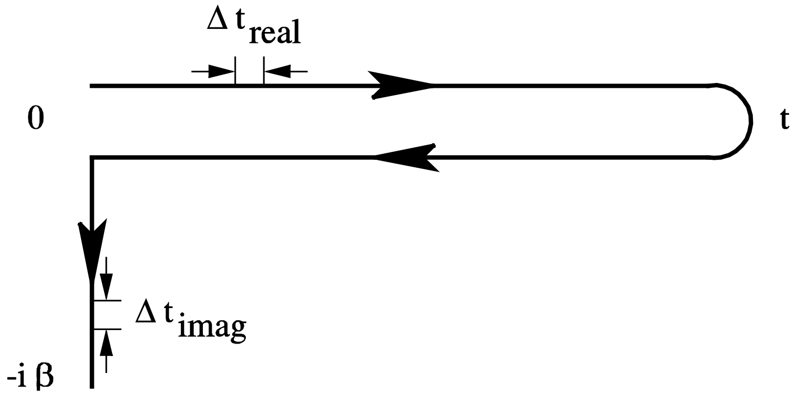


Falicov-Kimball Model



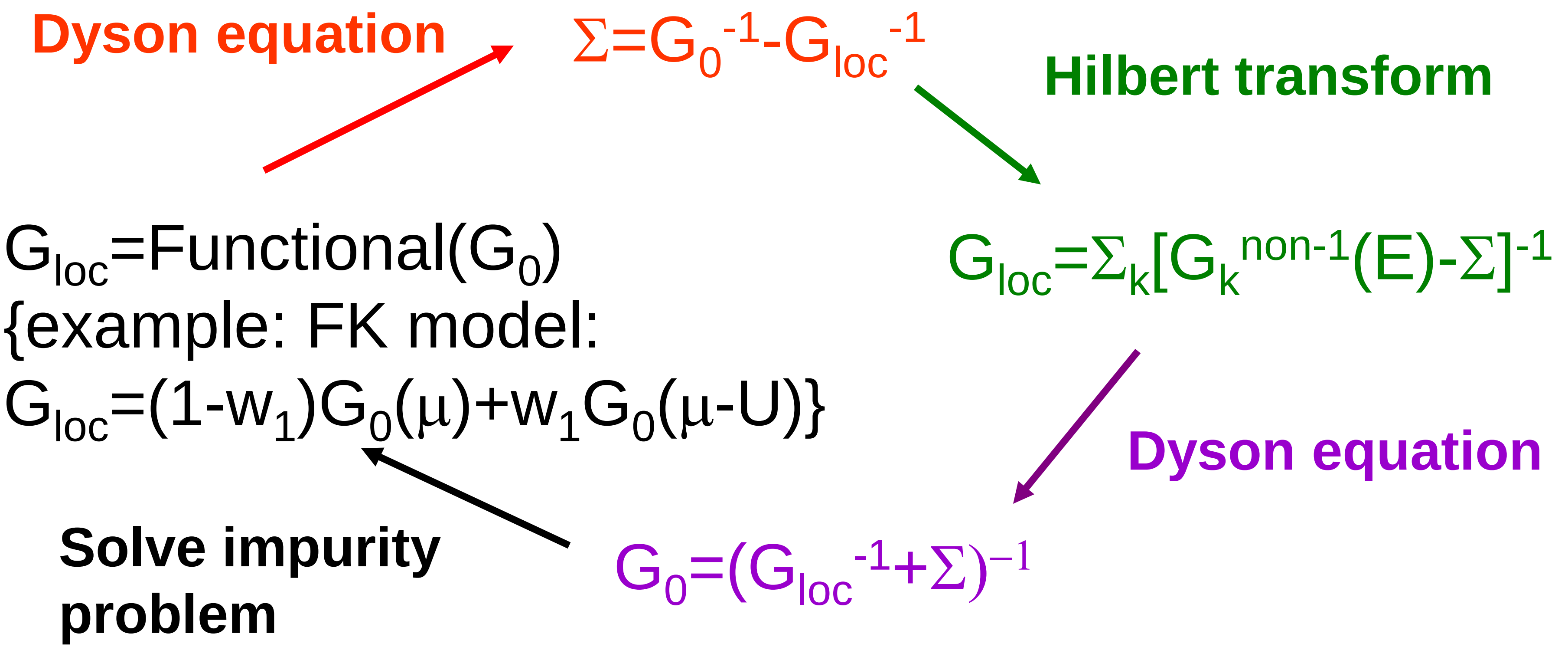
- Two kinds of particles: (i) **mobile electrons** and (ii) **localized electrons**.
- When both electrons are on **the same site** they interact with a correlation energy U.
- Many-body physics enters from an **annealed average over all localized electron configurations**.

Kadanoff-Baym-Keldysh formalism



- Problems without time-translation invariance can be solved with a so-called **Keldysh formalism**.
- Green’s functions are defined with time arguments that run over the **Kadanoff-Baym-Keldysh contour**.
- The electrons evolve in the fields **forwards** in time, then de-evolve in the fields **backwards** in time (we use the **Hamiltonian gauge, where the scalar potential vanishes**).
- **Functional derivatives** are then used to determine the Green’s functions and other correlation functions of interest.

Dynamical mean-field theory algorithm



Dyson equation

$\Sigma = G_0^{-1} - G_{loc}^{-1}$

Hilbert transform

$G_{loc} = \Sigma_k [G_k^{non-1}(E) - \Sigma]^{-1}$

Dyson equation

$G_0 = (G_{loc}^{-1} + \Sigma)^{-1}$

Solve impurity problem

G_{loc}=Functional(G_0)
{example: FK model:
$G_{loc} = (1-w_1)G_0(\mu) + w_1 G_0(\mu - U)$}

All objects (G and Σ) are **matrices** with each time argument lying on the contour.

Peierl’s substitution and the Hilbert transform

The band structure is a sum of cosines on a hypercubic lattice:

$$\varepsilon(k) = -\frac{t^*}{2\sqrt{d}}\sum_{i=1}\cos k_i \Rightarrow -\frac{t^*}{2\sqrt{d}}\sum_{i=1}\cos[k_i - eA_i(t)] = \varepsilon\cos[eA(t)] + \bar{\varepsilon}\sin[eA(t)]$$

which becomes the sum of two “band energies” when the field lies in the diagonal direction after the Peierl’s substitution.

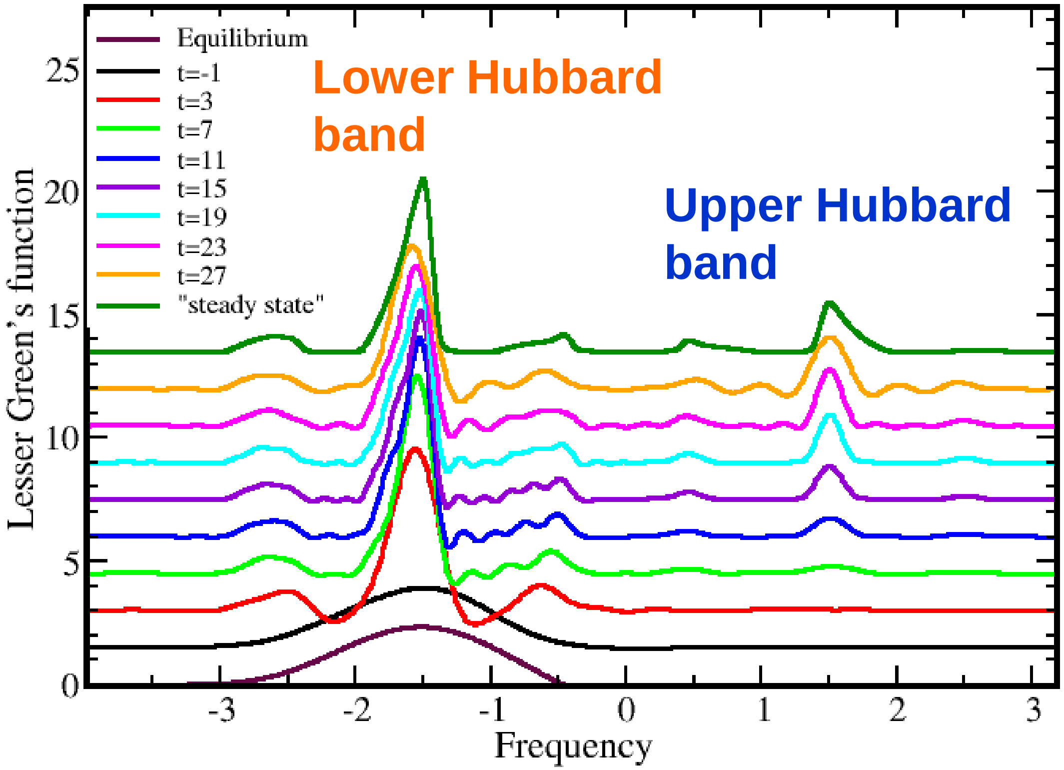
These band energies have a joint Gaussian density of states, so a summation over the Brillouin zone can be replaced by a two-dimensional Gaussian-weighted integral.

Probing the formation of a nonequilibrium state

Constant pump

- Turn the pump on and leave on (we use a large amplitude dc field).
- Probe the system with different time delays from when the pump was originally turned on.
- Now one can probe the transient development of a nonequilibrium steady state and the properties of the steady state.

Example: Mott insulator with large uniform electric field as a pump



(Exact solution for the Falicov-Kimball model in large dimensions and in the presence of a driving electric field.)

Lesser Green's function at average time rapidly evolves from equilibrium to a steady state, with a distribution function similar to a Fermi function at higher T.

Vary the field, small interactions

- We see clear Bloch oscillations in the TR-PES signal as a function of the time delay. Probe pulse width is narrow here---1/hopping (1 fs).
- As E increases, the oscillation period goes down. Beats begin to develop.

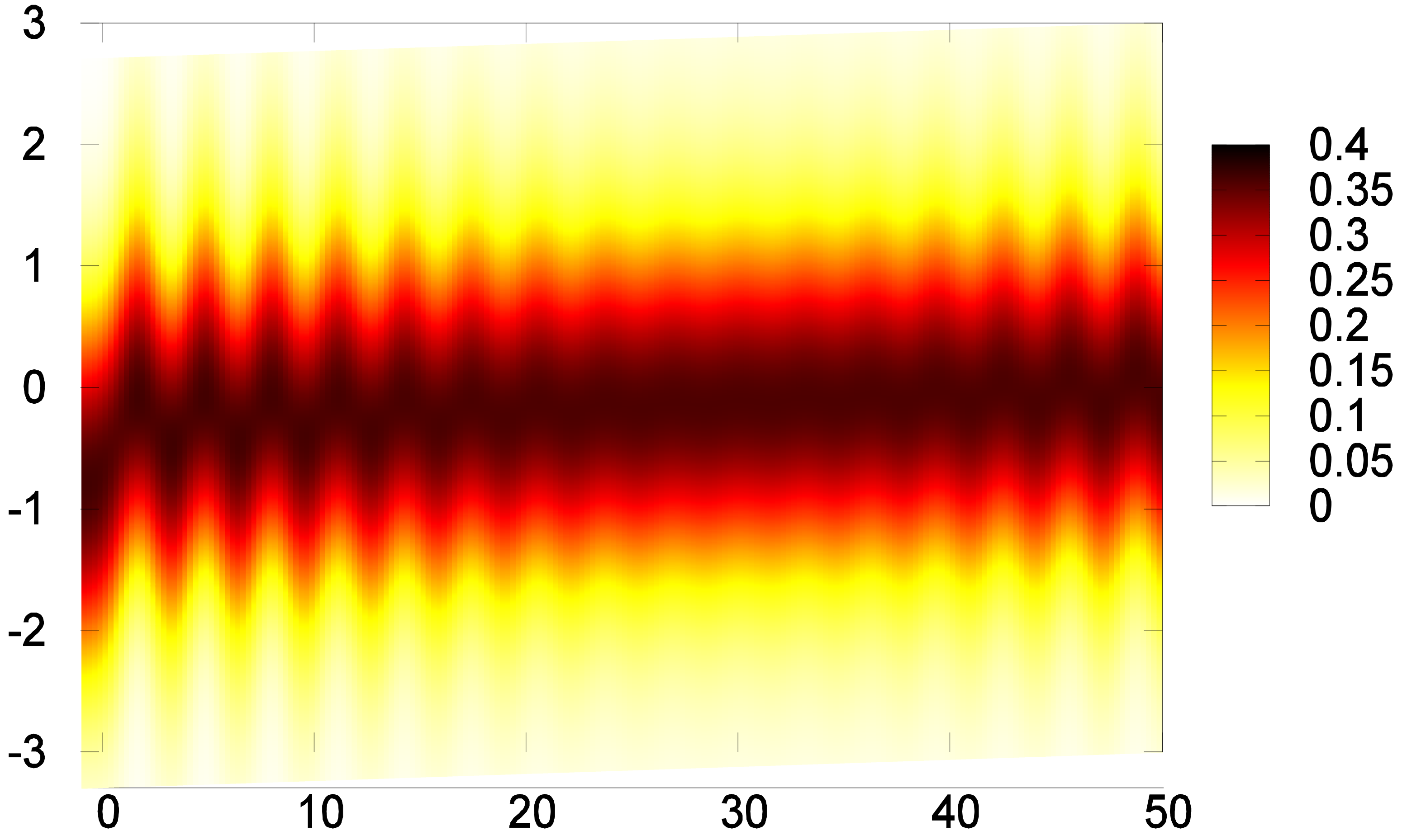
E=0.5

E=1

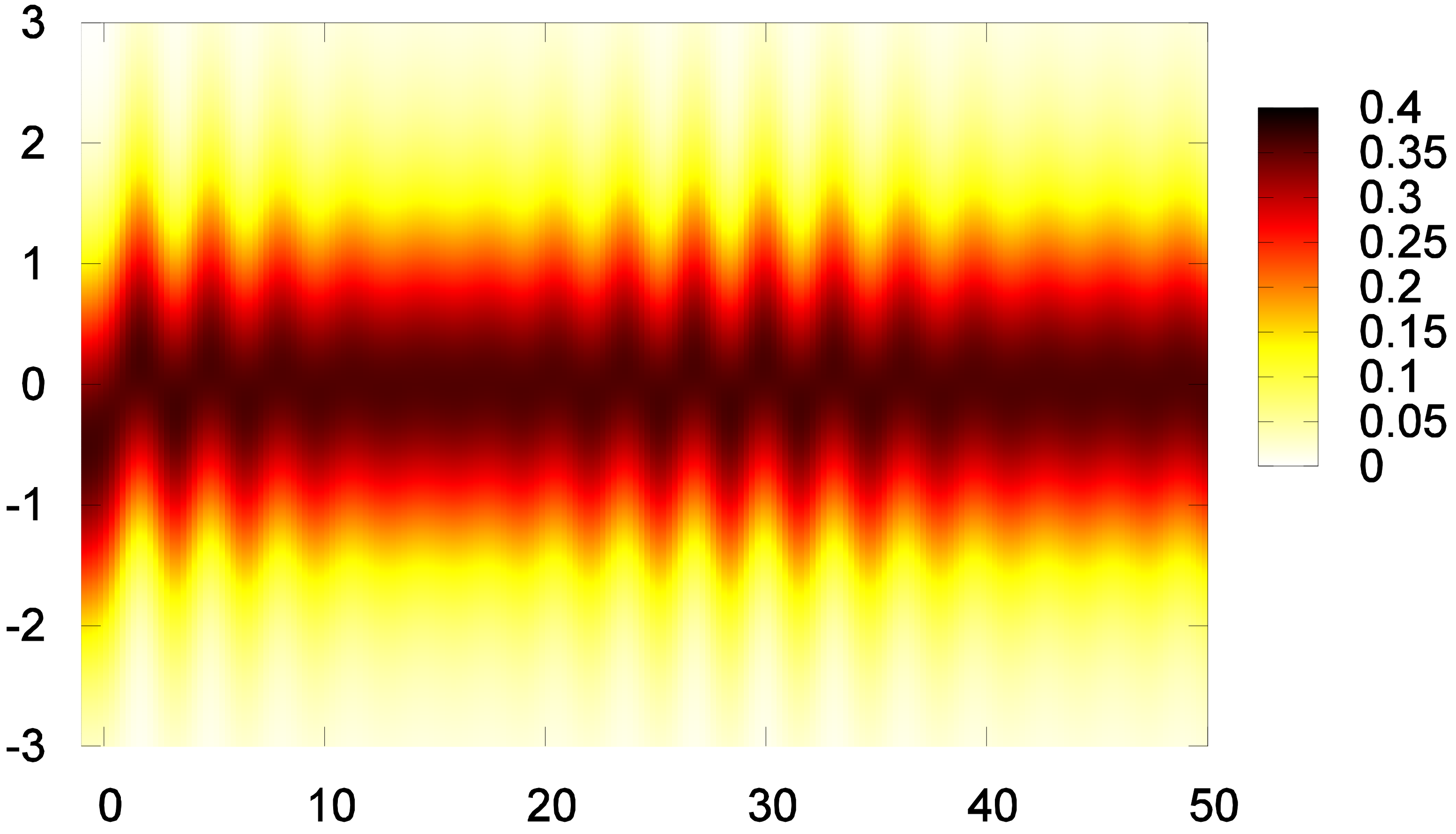
E=2

Examine beat phenomena for E=2

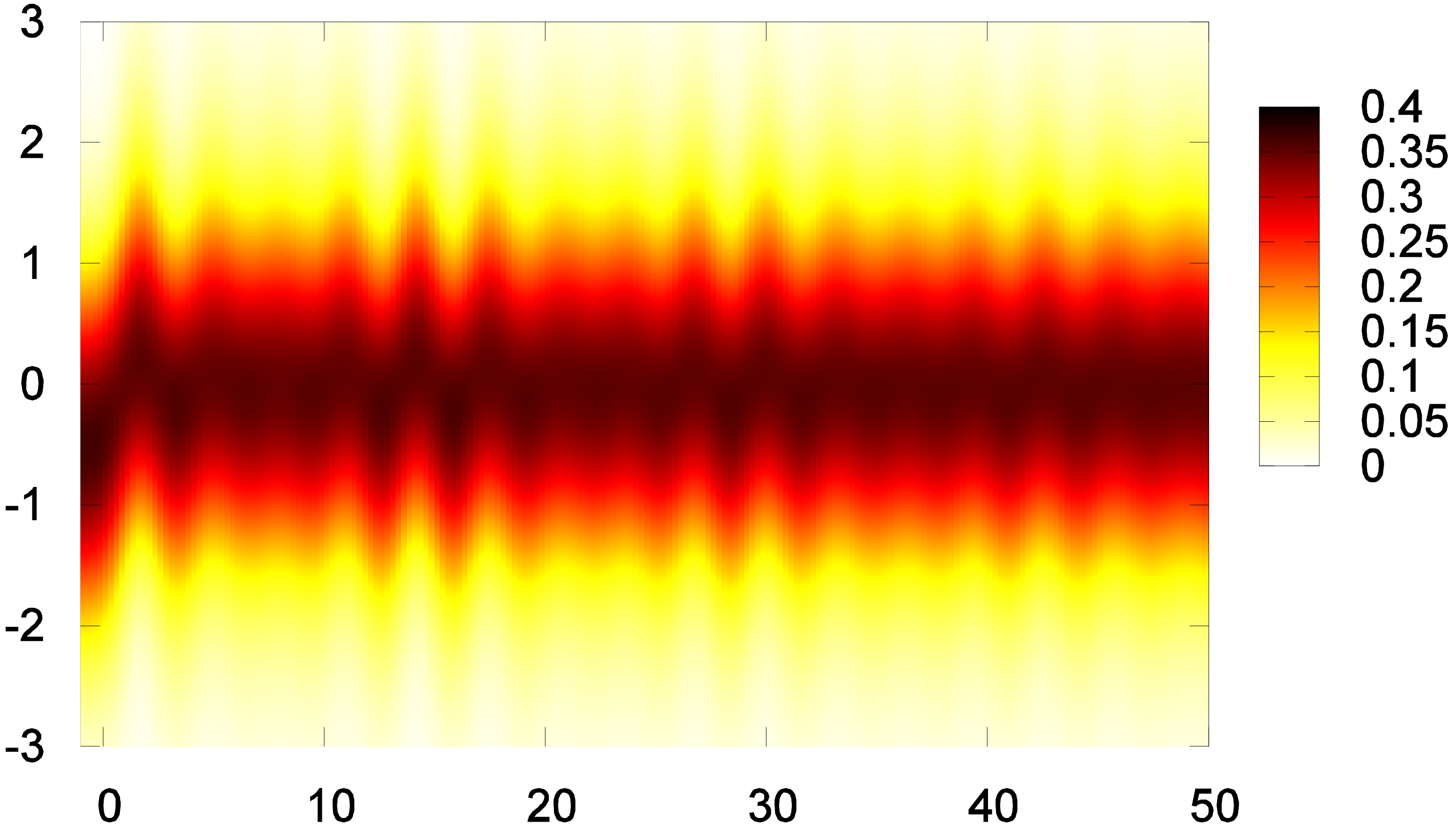
E=2, U=0.125 (close up)



E=2, U=0.25 (close up)



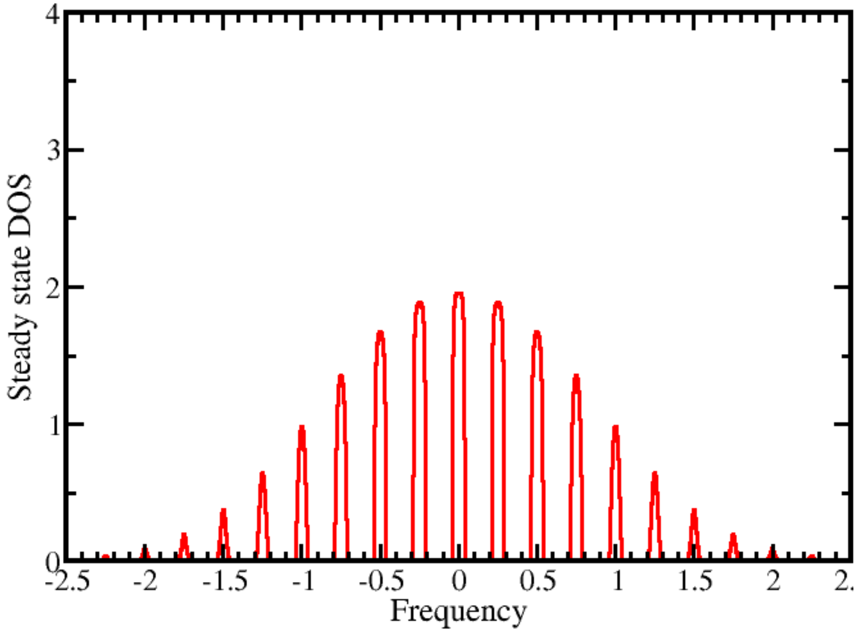
E=2, U=0.5 (close up)



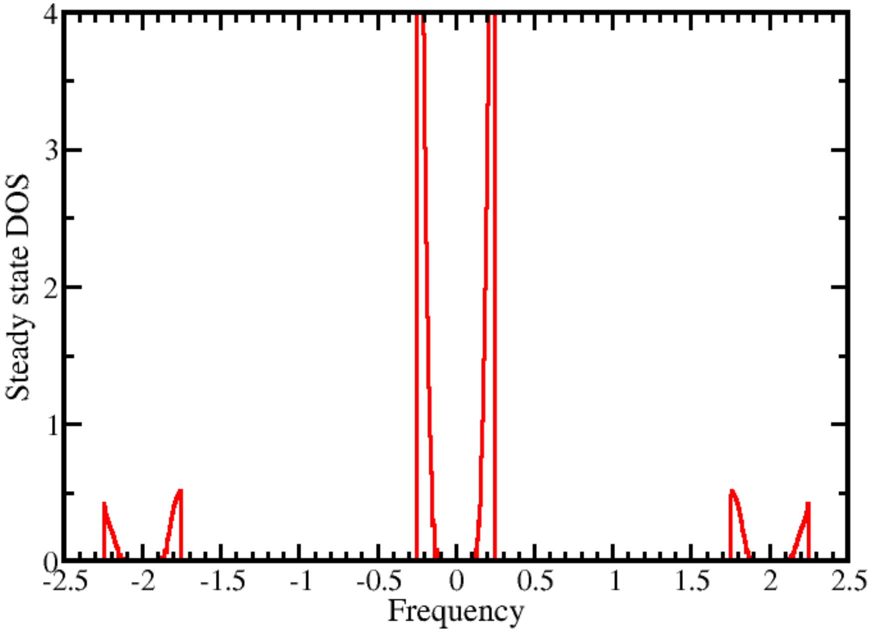
Period of the envelope function is proportional to 1/U

Destruction of the Wannier-Stark ladder

E=0.25, U=0.125 (small E)



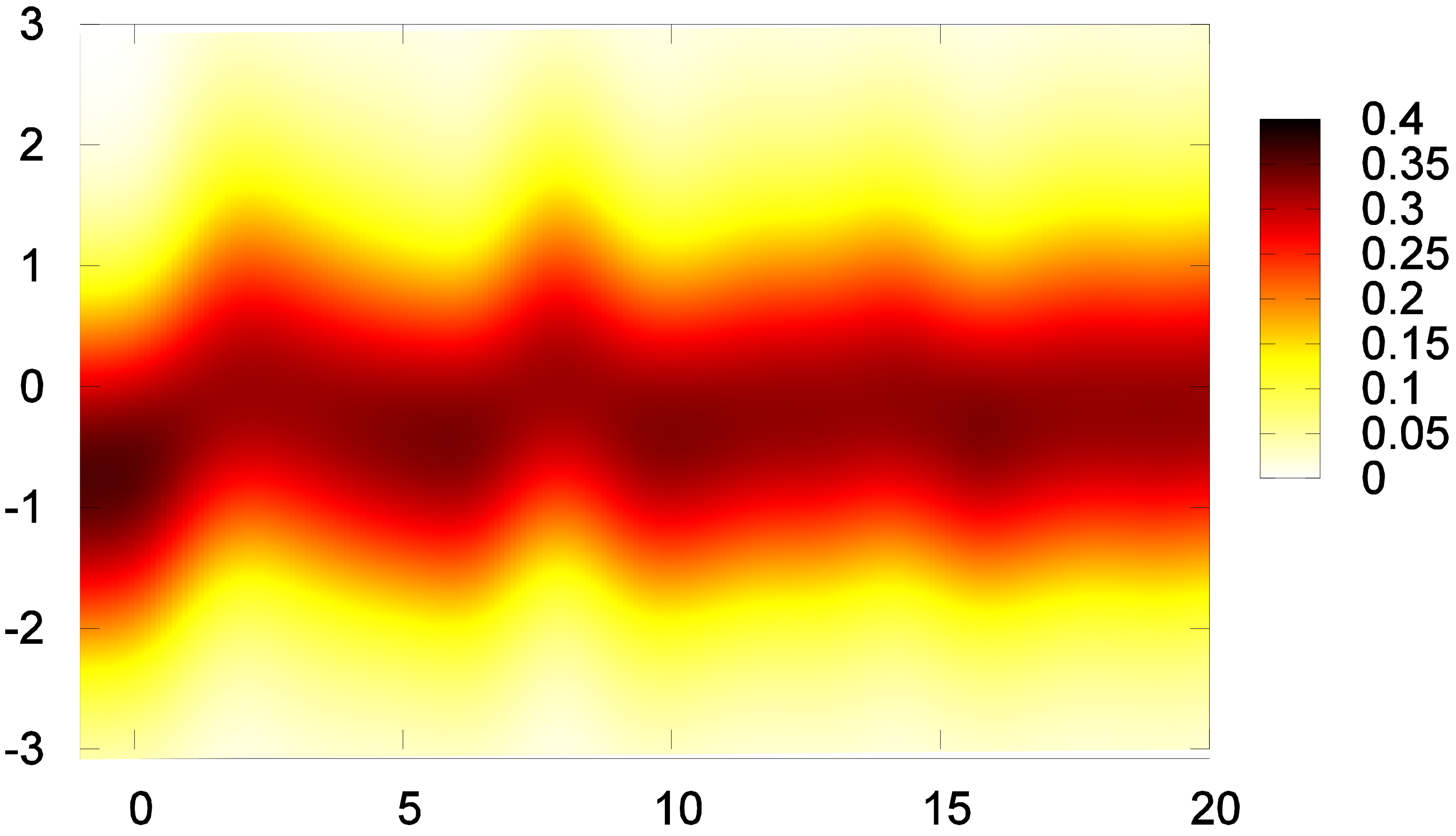
E=2, U=0.5 (large E)



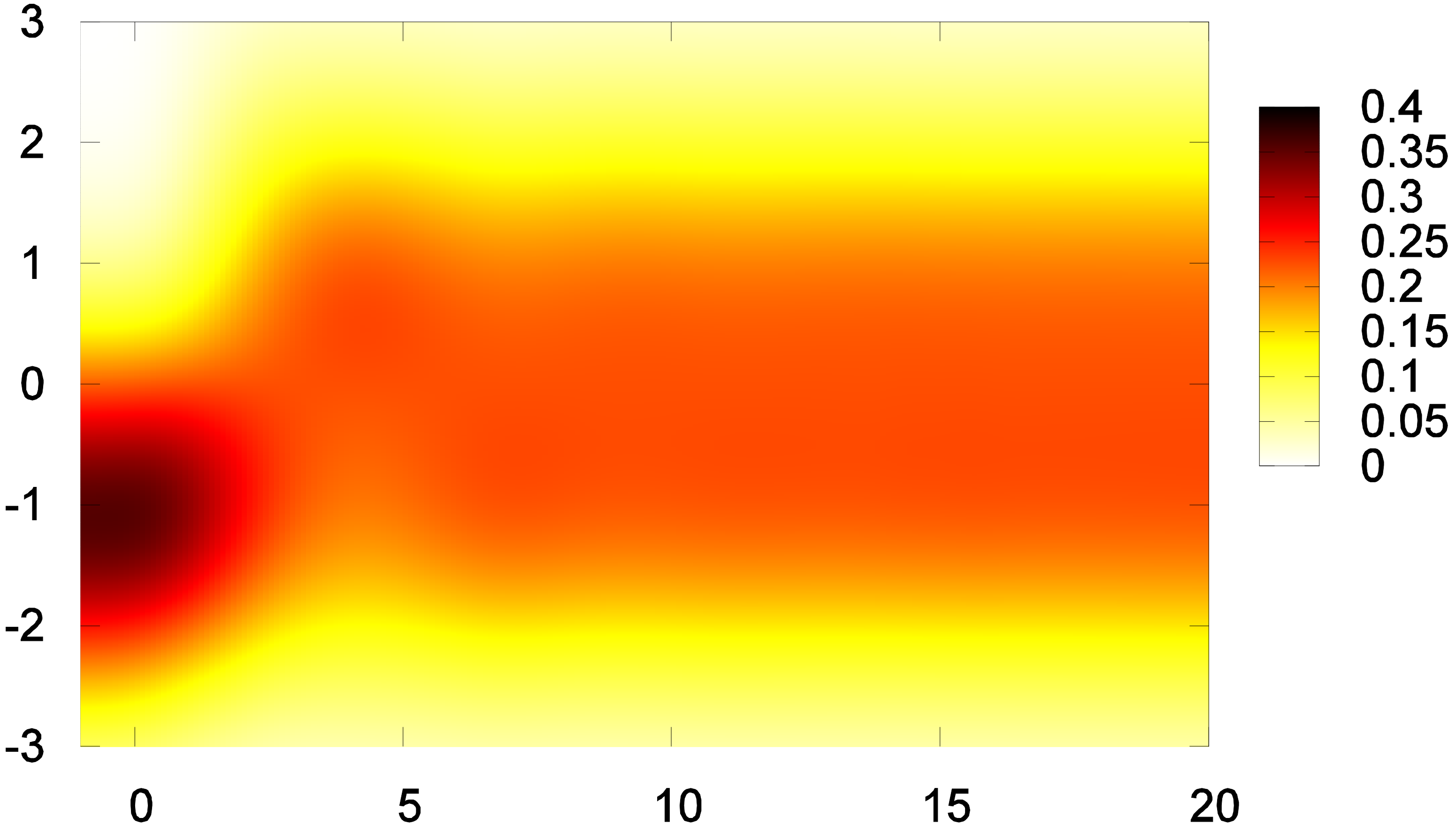
Either the delta function peaks broaden (small E) or the peaks split like a “mini” metal-insulator transition (large E)

Examine damping of oscillations as we go through the MIT for E=2

E=2, U=1 (anomalous metal)

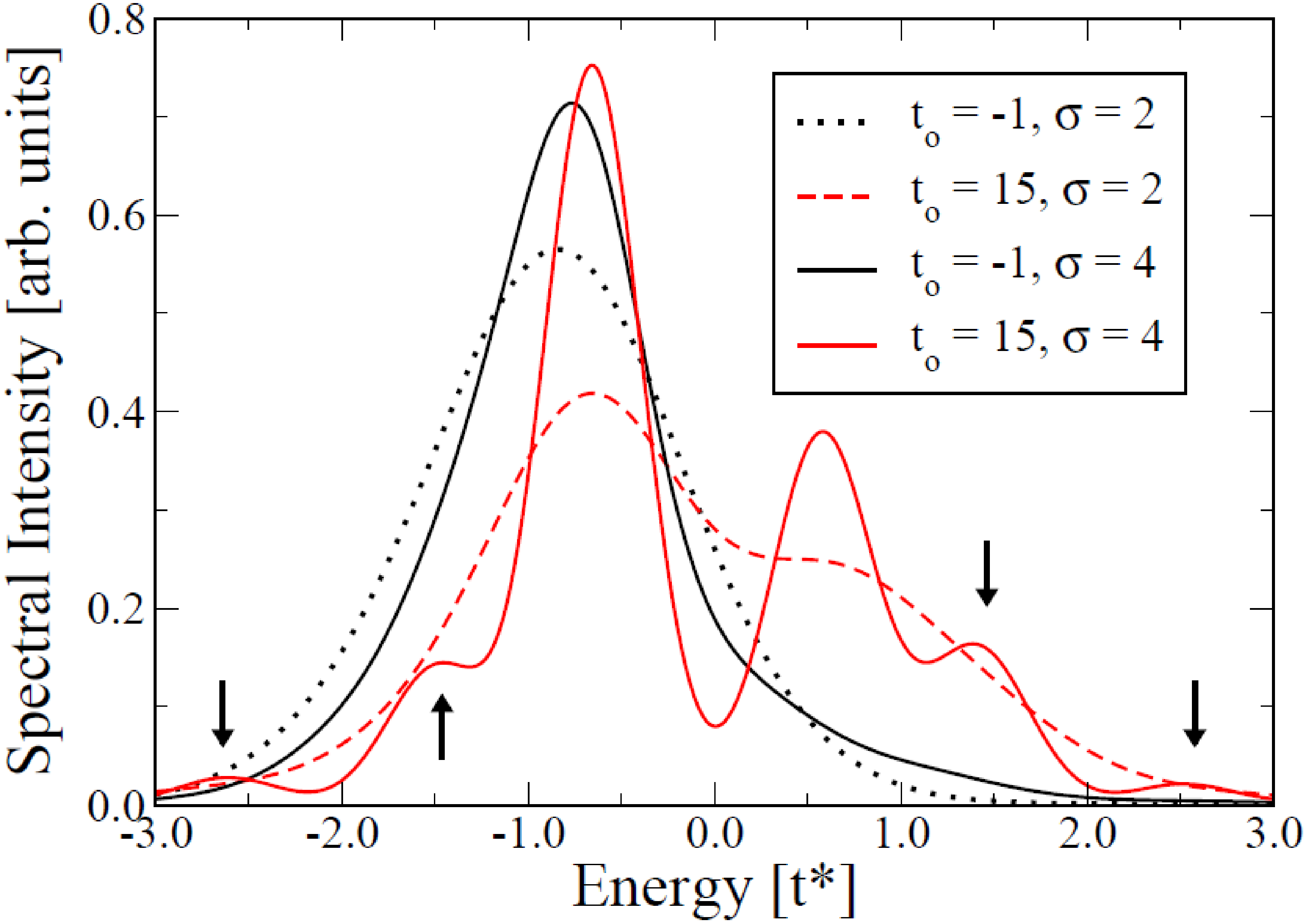


E=2, U=2 (insulator)



- Oscillations gone, probe too narrow to show spectral features

Hot-electron model does not work

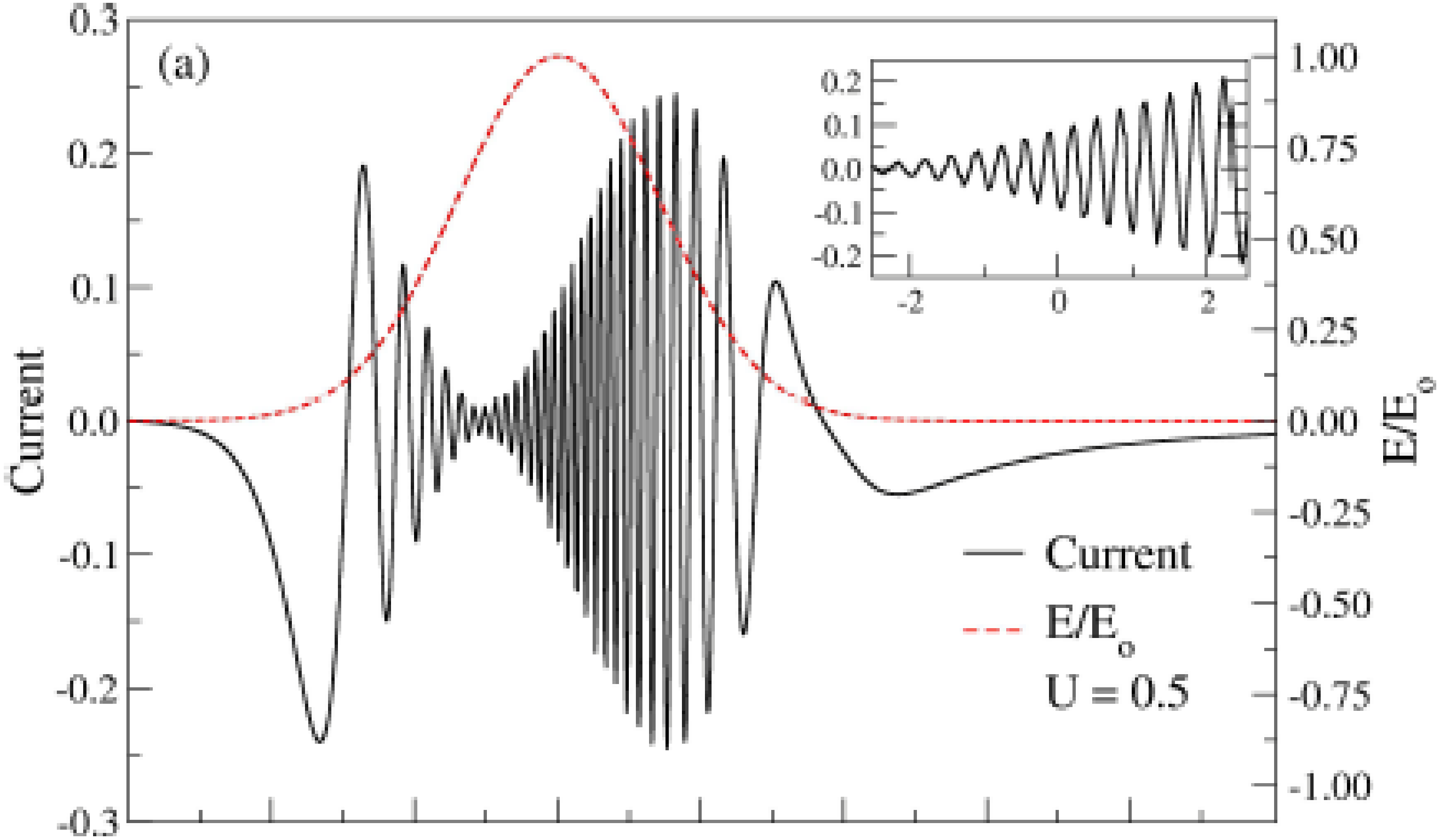


Gauge-invariant momentum distribution, metal U=0.5, E=1

Gauge-invariant momentum distribution, insulator U=2, E=1

Pumped Pulse

Current from a pulse

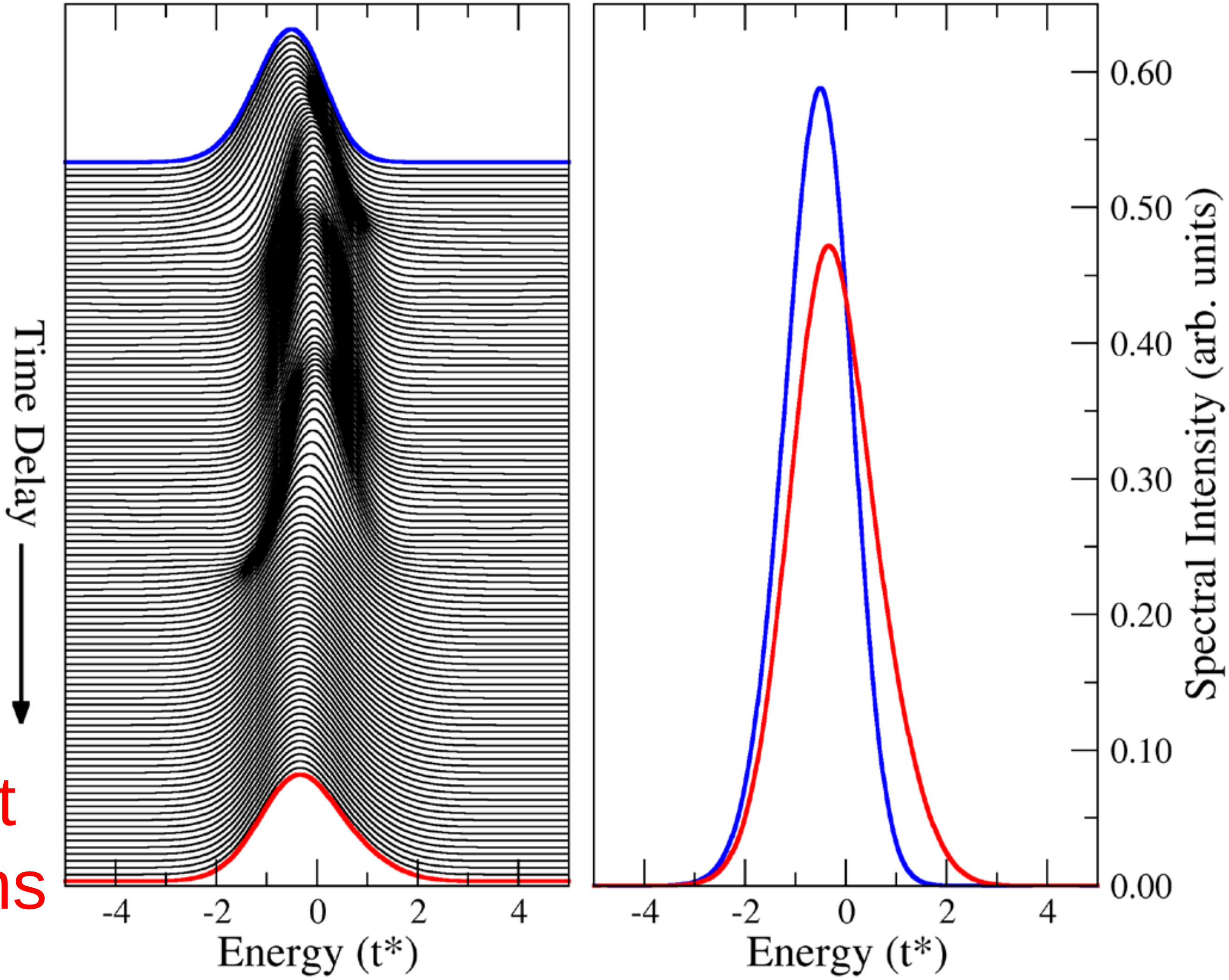


Current is irregular here, with varying amplitude and Varying frequency (like a chirp).

TR-PES response

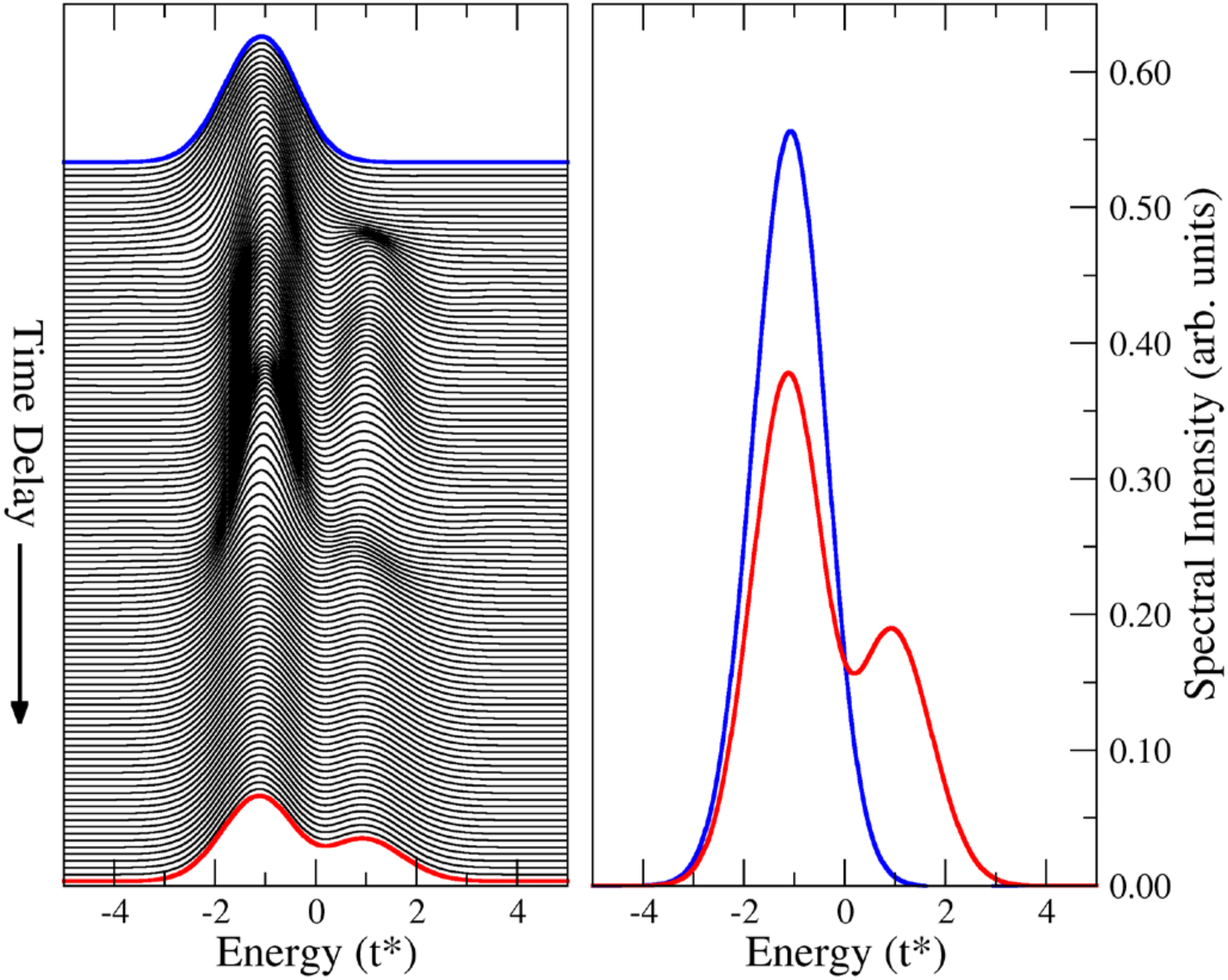
U=0.5

No significant oscillations



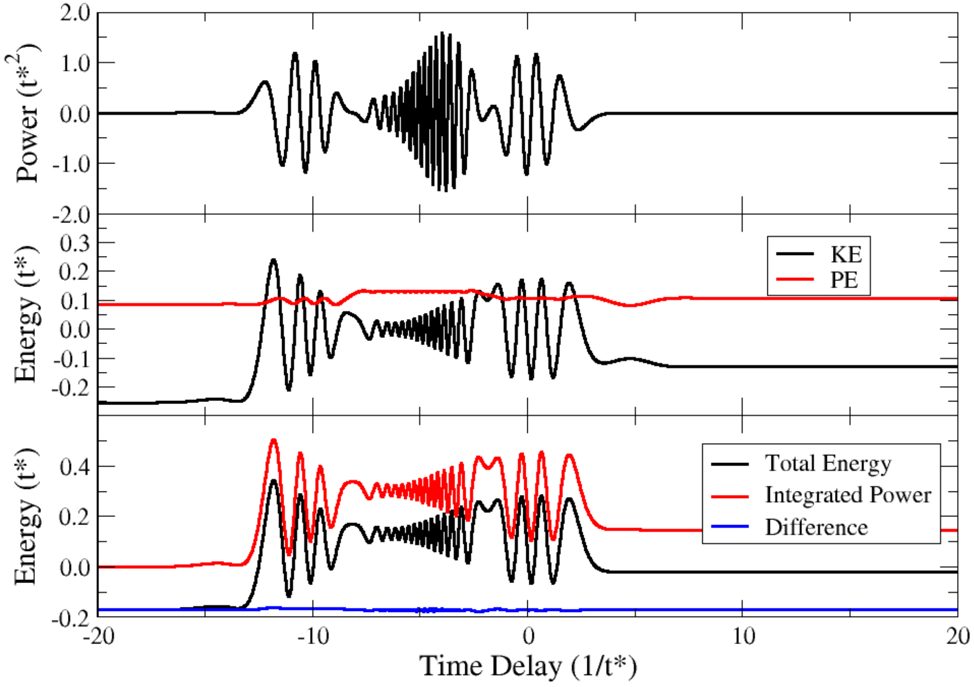
TR-PES response

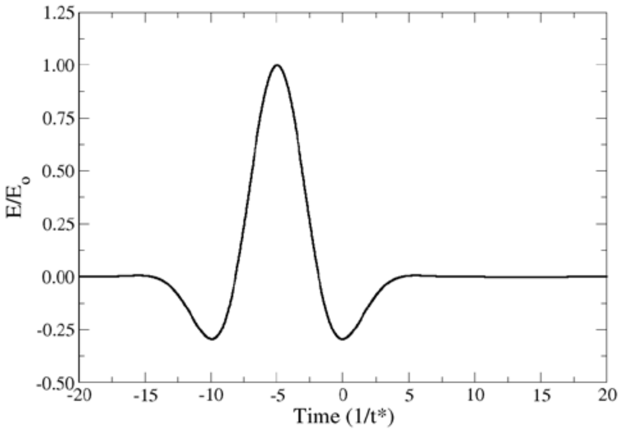
U=2



Evolution saturates, not clear that it is just hot electrons.

Energy from a modulated pulse

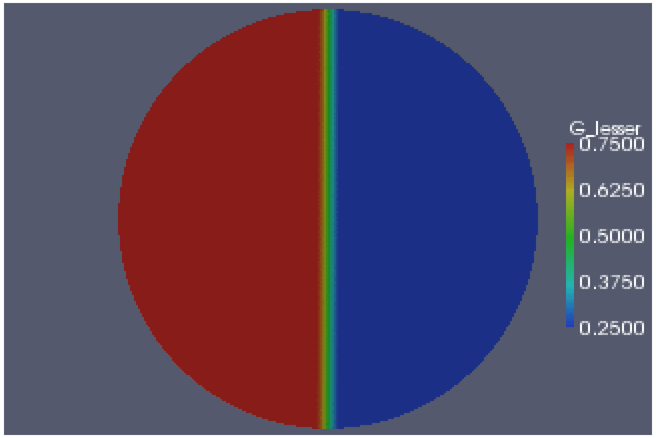


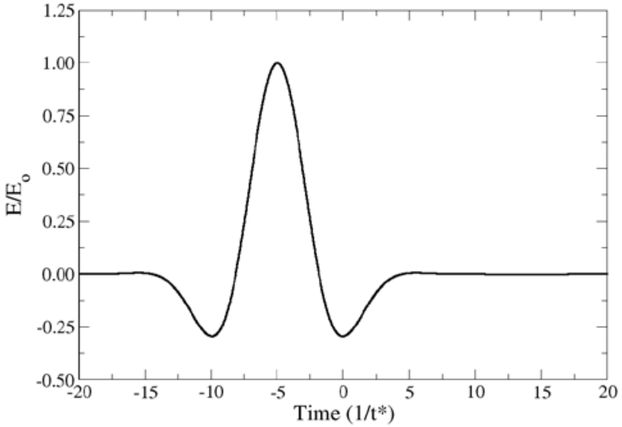


Energy increases due to the pulse, but final state is not yet in equilibrium at a higher temperature.

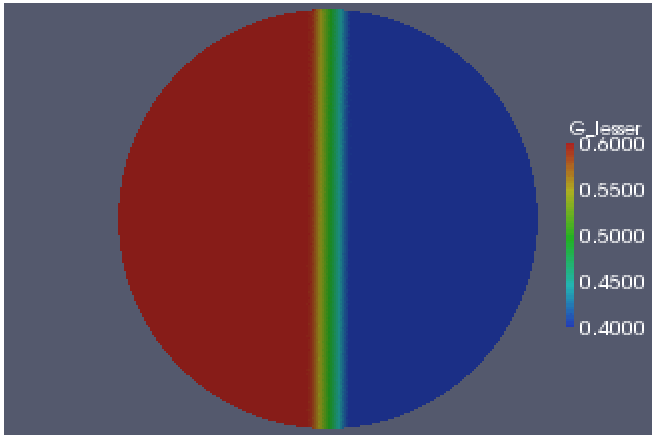
Distribution functions

U=0.5





U=2



Distribution functions

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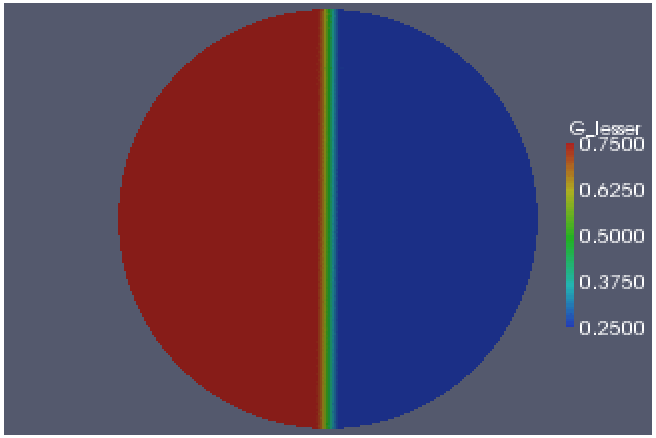

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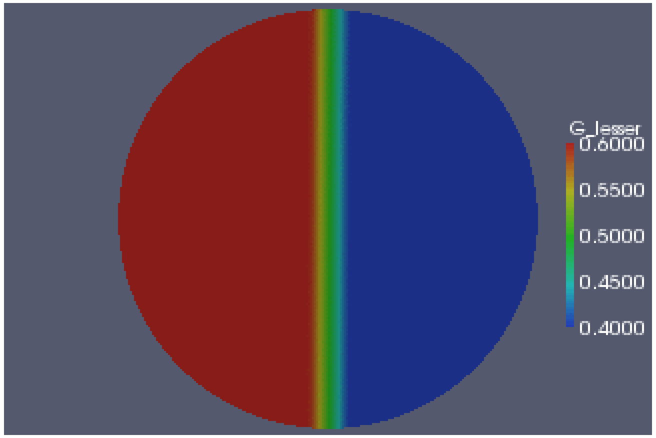

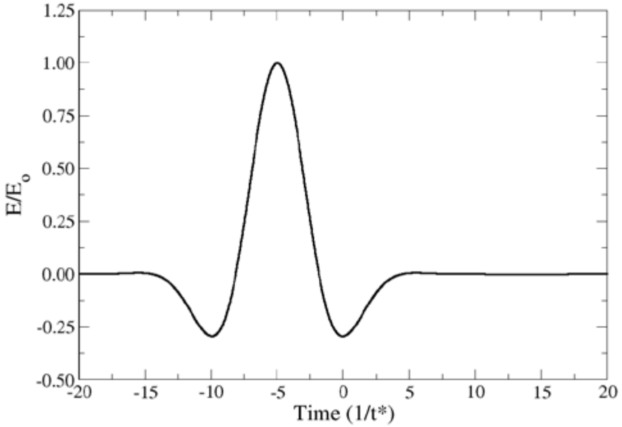

Conclusions

- Showed how to solve the nonequilibrium many-body problem with dynamical mean field theory.
- Using the Falicov-Kimball model, we examined the time resolved PES response.
- Showed that the hot electron model does not typically work at short times.
- This work is just the tip of the iceberg in what can be done with nonequilibrium DMFT. Future work will focus on charge density wave systems and on more complex models (Hubbard).
- Need long-run impurity solvers in the time domain!