

Nonlinear Response of a Mott Insulator

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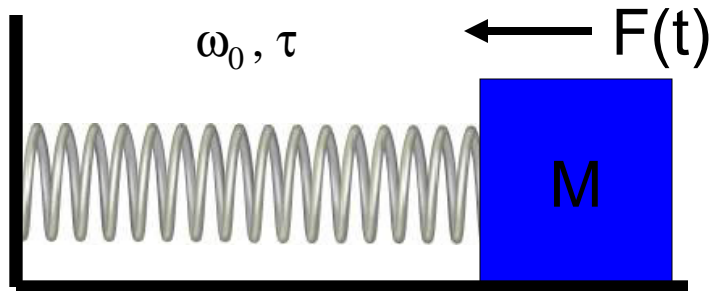
Georgetown University

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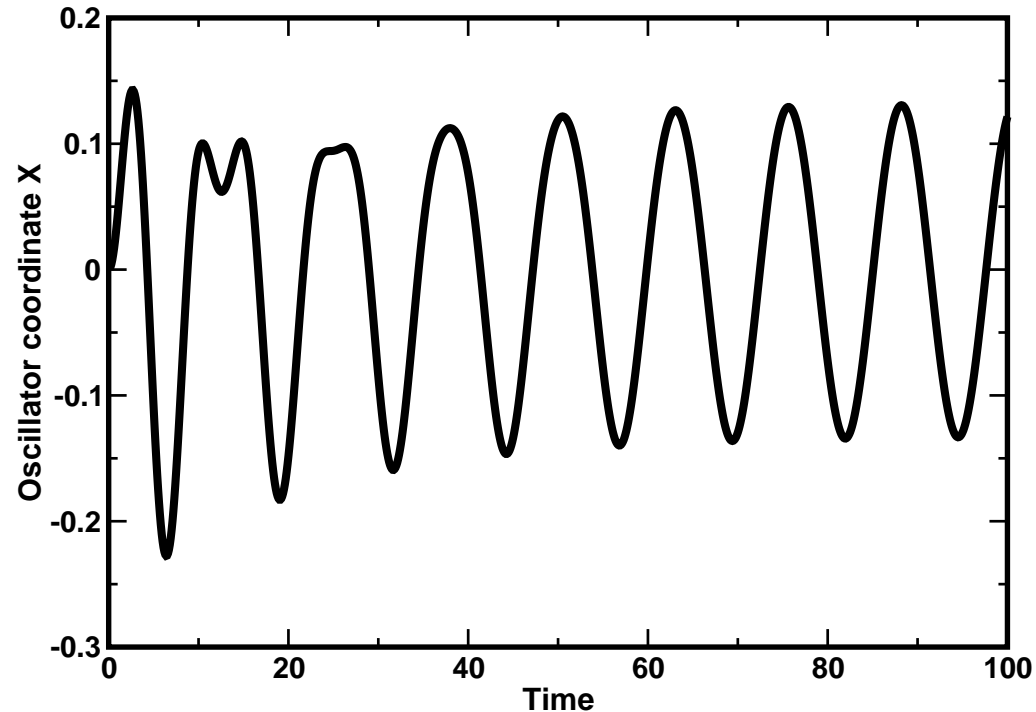
Cray T3E and X1 time from ARSC and ERDC

Thanks to Volodmyr Turkowski and Veljko Zlatic' for collaborating on this work.

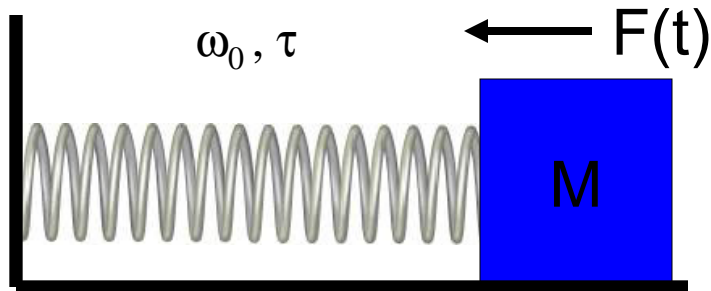
Driven damped simple harmonic oscillator



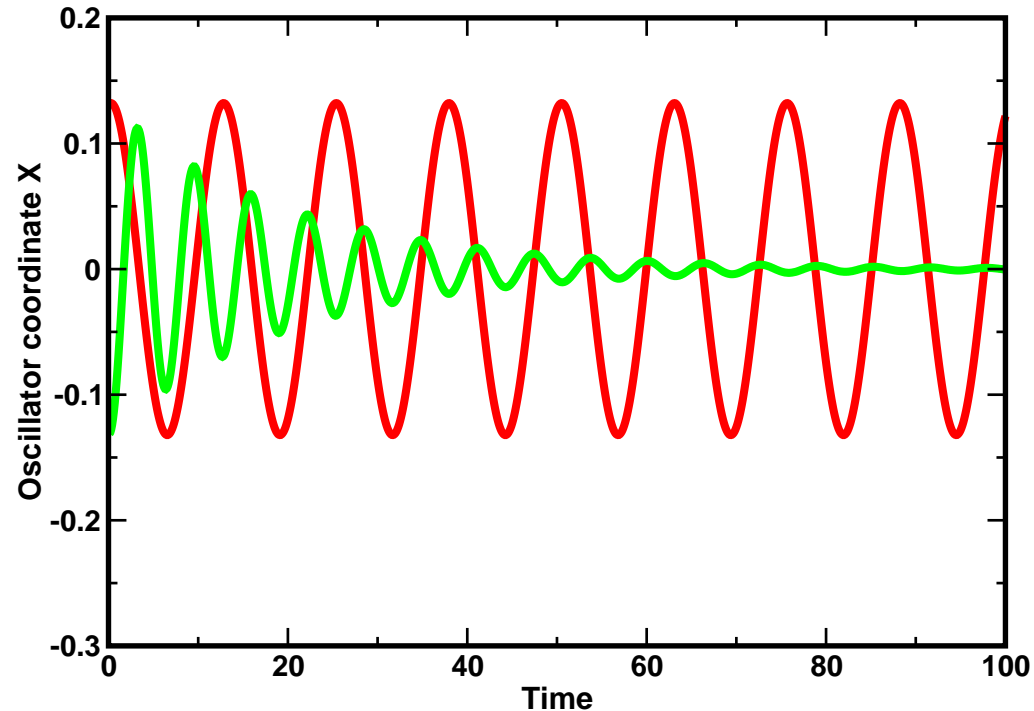
- A classical SHO that is driven by a sinusoidal force and damped has a complicated response due to the transients.



Driven damped simple harmonic oscillator

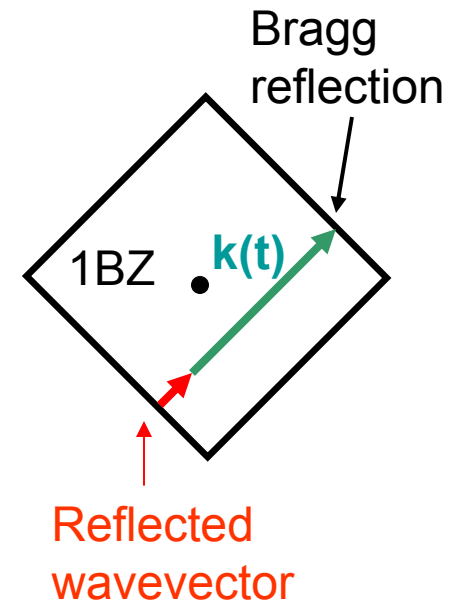


- Separating out the transient and steady-state response makes the picture clearer.



Electrons driven by an electric field

- In a **semiclassical** picture, the electron momentum, written as $\hbar\mathbf{k}=\mathbf{P}$, evolves with a linear time-dependence corresponding to the **acceleration** due to the field: $\mathbf{k}(t)=e\mathbf{E}t/\hbar$.
- **Scattering** modifies this picture: since the electrons are in a periodic lattice, the wavevector cannot increase **outside** of the first Brillouin zone; as it tries to move beyond the 1BZ it is **Bragg reflected** to the opposite side of the zone.
- Defects, impurities, lattice vibrations, and other electrons are other sources of **scattering**; they **interrupt the evolution** of the wavevector in the BZ.



Bloch Oscillations (Bloch 1928, Zener 1932)

Constant
potential
difference
(constant E
field)



Oscillating
current

- When on a periodic lattice, the electrons' motion is governed by their electronic bandstructure $\epsilon(\mathbf{k})$. In metals the last band is partially filled, so electrons can easily move. In insulators, the bands are completely filled, with a band-gap to the first unoccupied band.
- The electrons move with an effective velocity $\mathbf{v}(\mathbf{k}) = d\epsilon(\mathbf{k})/d\hbar\mathbf{k}$. So they carry a current equal to $e\mathbf{v}(\mathbf{k})$ summed over all wavevectors \mathbf{k} .
- As the **wavevector** evolves over the 1BZ, it changes **periodically**, and so does $\mathbf{v}(\mathbf{k})$.
- Hence, **Bragg reflection makes the current periodic in time!** *A dc electric field creates a periodic ac current in a perfect metal with electrons moving in a crystalline lattice.*

But this is never seen in any
conventional metal no matter
how clean it is.

Quenching Bloch oscillations

- **Tunneling** between bands makes the electrons move as if the lattice was not there. They continue to accelerate and do not undergo periodic motion. In this case there are no Bloch oscillations. Tunneling only occurs if the energy stored in the field is large enough to cross the energy barrier between bands. *This will not be considered in this talk.*
- If the **scattering** due to defects, impurities, lattice vibrations, or other electrons is **frequent enough**, the electrons won't have enough time to undergo the Bloch oscillation, as their wavevector becomes randomly changed with each scattering event, and they must restart their acceleration in the field again. *This is normally why Bloch oscillations are not commonly seen in metals.*

Drude-Sommerfeld picture

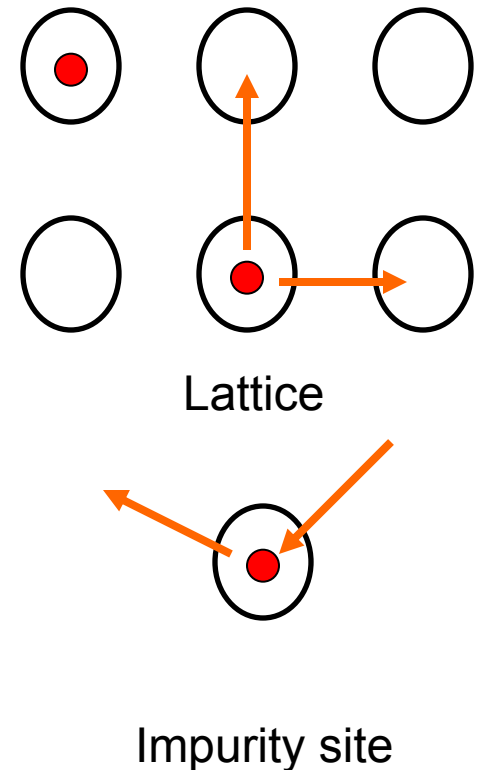
When the scattering time is **short enough**, the current starts to increase linearly with time, but is **randomized** before it can Bragg reflect.

In the **steady state**, the current density is **linearly proportional** to the electric field $\mathbf{j}=\sigma\mathbf{E}$, with σ being the dc conductivity. This is often referred to as **Ohm's law**.

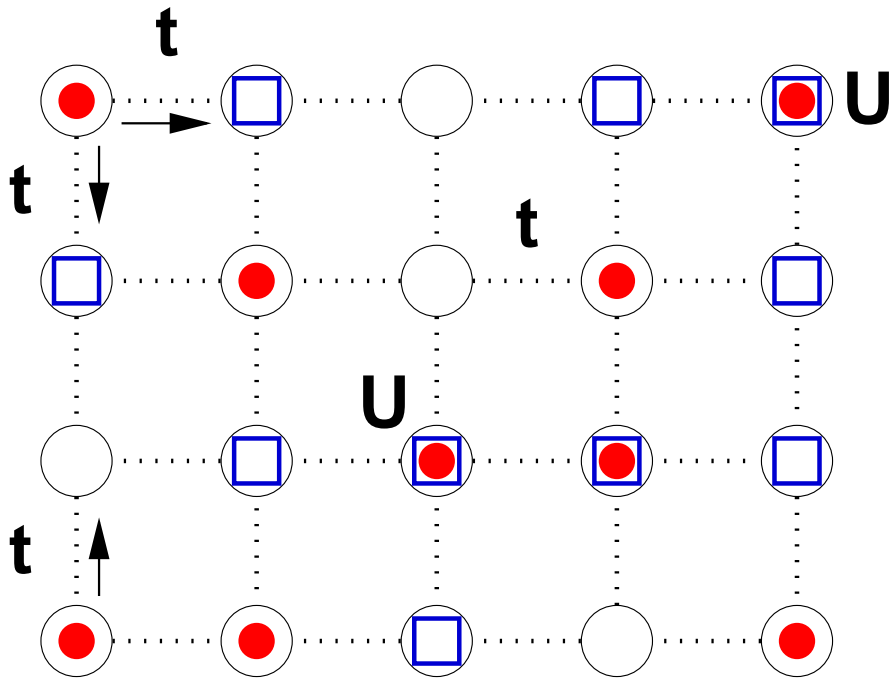
Using **quantum mechanics** and the so-called **Kubo formalism for linear response**, one can calculate the conductivity *using results obtained from an equilibrium calculation, which has no field!* Using a **nonequilibrium approach truncated to linear order** in the field, yields the identical result as the Kubo response.

Dynamical mean field theory

- Models of strongly correlated materials are difficult to solve.
- Significant progress has been made over the past 15 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**.

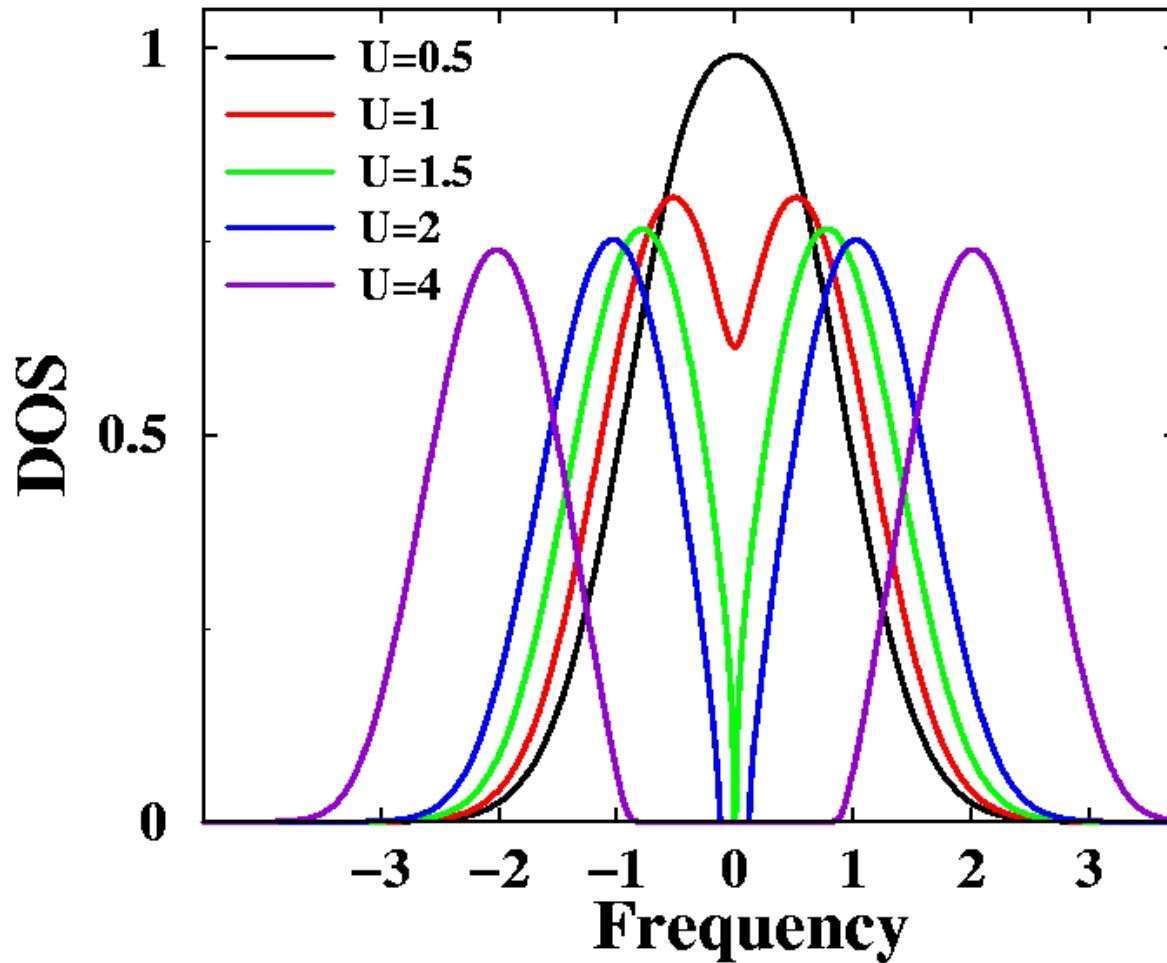


Falicov-Kimball Model



- 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**.

FK model Metal-Insulator transition (NFL)



- **Correlation-induced gap** drives the single-particle DOS to zero at $\omega=0$ for $U > \sqrt{2}$
- Interacting DOS is **independent of T** in DMFT (Van Dongen, PRB, 1992)
- *Examine the nonlinear current response through the Mott transition.*

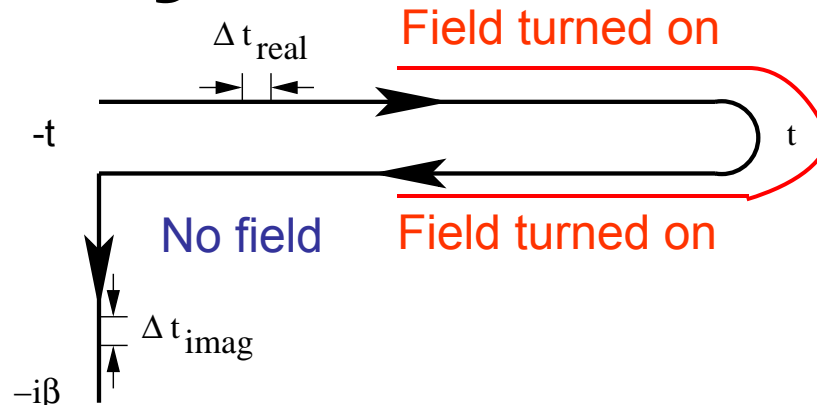
Peierl's substitution

- In the Landau gauge, the scalar potential **vanishes**, so we write $\mathbf{E}(t) = -\partial\mathbf{A}(t)/\partial t$, for the **electric field** in terms of the **vector potential**.
- We work with a **spatially uniform**, but **time-dependent** electric field only.
- The effect of the field is handled by substituting $\mathbf{k} \rightarrow \mathbf{k} - e\mathbf{A}(t)$ in the band structure: $\epsilon(\mathbf{k}) \rightarrow \epsilon[\mathbf{k} - e\mathbf{A}(t)]$.

Time-dependent evolution problem

- The Hamiltonian is now **time-dependent** due to the t -dependence of the vector potential in the band structure.
- Since we **turn the field on at $t=0$** , we do not have time-translation invariance.
- Examining the operator structure of the Green's function shows that we must evolve the fields **forward in time**, then we evolve them **backward in time** to determine G .

Keldysh formalism

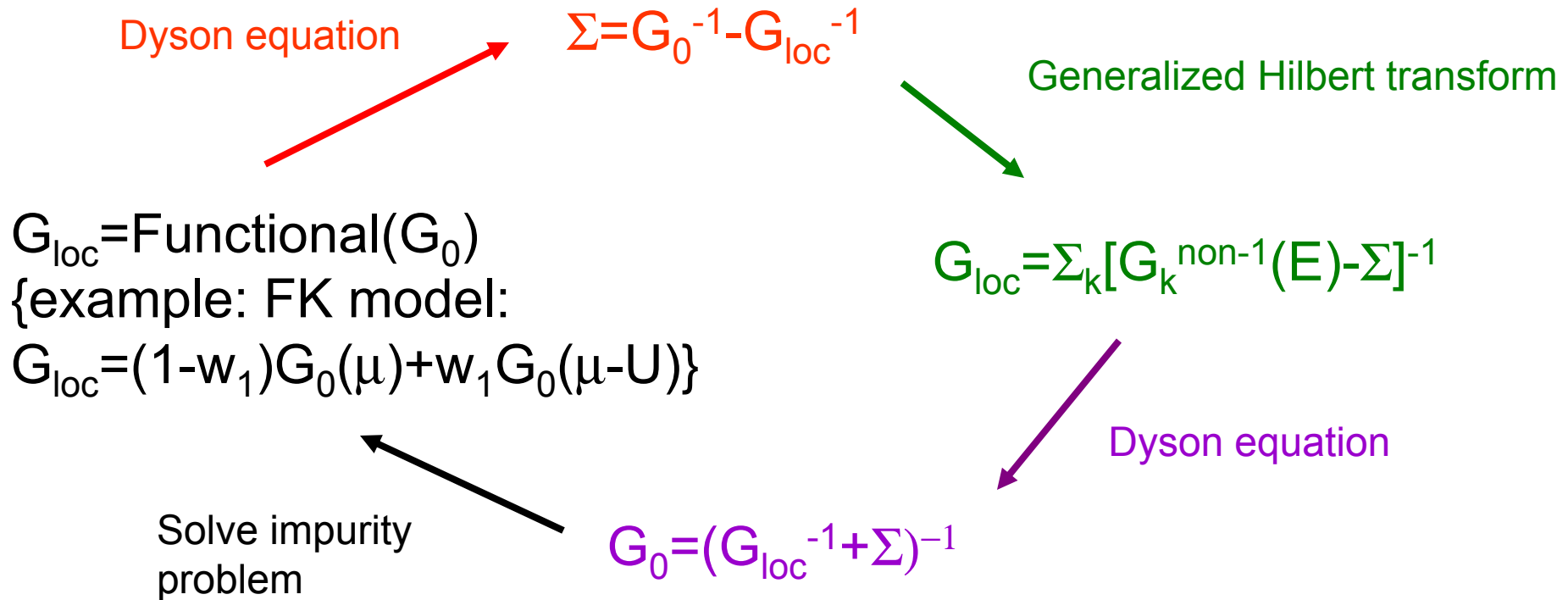


- Hence we solve the problem with a so-called **Keldysh formalism** for the contour-ordered Green's function.
- The contour-ordered Green's functions are defined with two time arguments that run over the **Kadanoff-Baym contour**.
- The electrons evolve in the fields **forwards** in time, then de-evolve in the fields **backwards** in time.
- **Functional derivatives** can be used to determine the Green's functions and other correlation functions of interest.

Transient plus steady-state response

- If we solve the Keldysh problem directly in real time, then we will see both the **steady-state** response and the **transient** response.
- One needs to wait **long enough** for the transient response to die off.
- This **may not be possible** given finite computational facilities.

Dynamical mean-field theory algorithm



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

Noninteracting Green's function in a field

- In a **uniform field**, we take $\mathbf{A} = -\mathbf{E}t$ ($c=1$). Then $\varepsilon(\mathbf{k}) \rightarrow \varepsilon(\mathbf{k} + e\mathbf{E}t) = -t^* \sum_i [\cos k_i \cos eE_i t - \sin k_i \sin eE_i t] / d$.
- Since the time-evolution of the noninteracting Green's functions can be determined **exactly**, we can directly solve for the Green's functions.
- The formalism simplifies greatly when the \mathbf{E} field is chosen to **lie along the diagonal**.

Generalized Hilbert Transform

- When the electric field lies along the **diagonal direction** $(1, 1, 1, \dots)$, the momentum dependence of all objects can be summarized in terms of two quantities: the conventional bandstructure $\varepsilon(\mathbf{k}) = -t \lim_{d \rightarrow \infty} \sum_{i=1}^d \cos \mathbf{k}_i / \sqrt{d}$ and an alternative bandstructure $\bar{E}(\mathbf{k}) = -t \lim_{d \rightarrow \infty} \sum_{i=1}^d \sin \mathbf{k}_i / \sqrt{d}$.
- The **joint DOS** can be found by using the techniques developed by Mueller-Hartmann and yield $\rho(\varepsilon, \bar{E}) = \exp(-\varepsilon^2 - \bar{E}^2) / \pi$ (Schmidt and Monien).
- *Hence the summation over momentum is generalized to a **two-dimensional integral** over the variables ε and \bar{E} .*

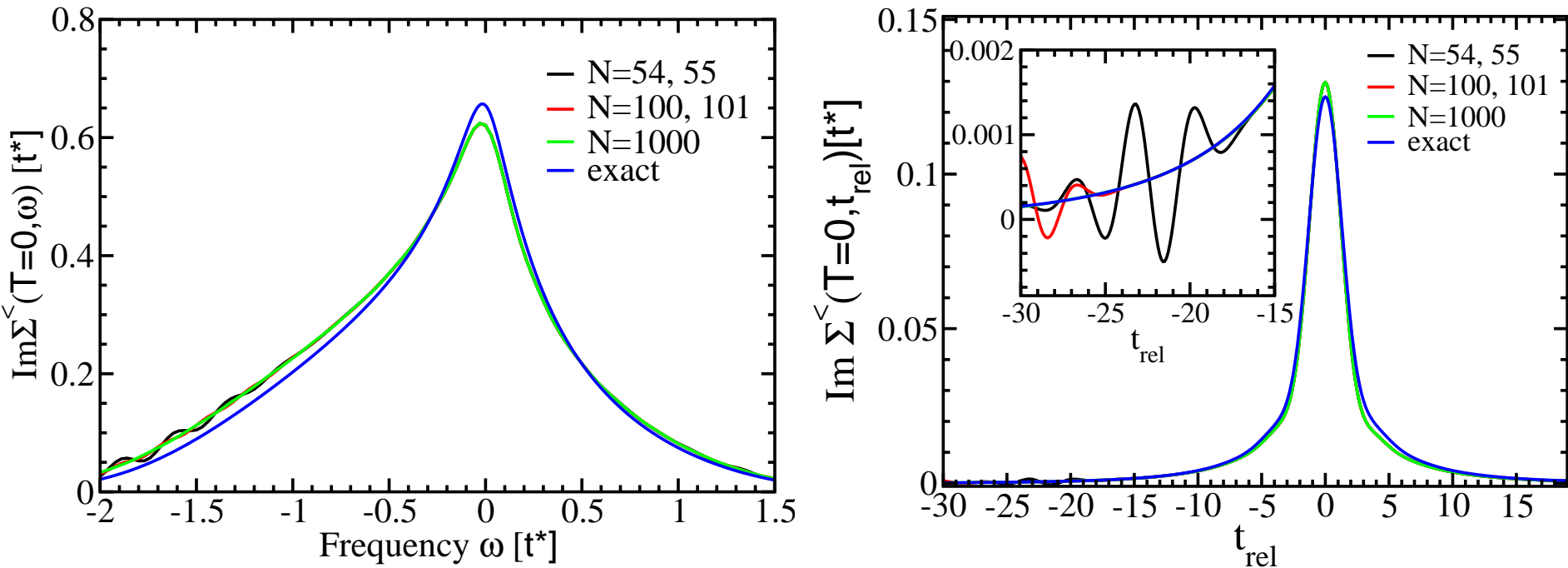
Computing the sum over k

- The local Green's function is found by using Dyson's equation with the **momentum-dependent noninteracting Green's function in a field** and the local self-energy.
- The self-energy and Green's function are each **two-time discrete matrices**; we need to compute *the double integral of every matrix element* (computed via taking a matrix inverse for the integrand) to get the local Green's function.
- *This approach is numerically intense, but **easily parallelizable** when we use a Gaussian integration for each energy integration.*

Numerical issues

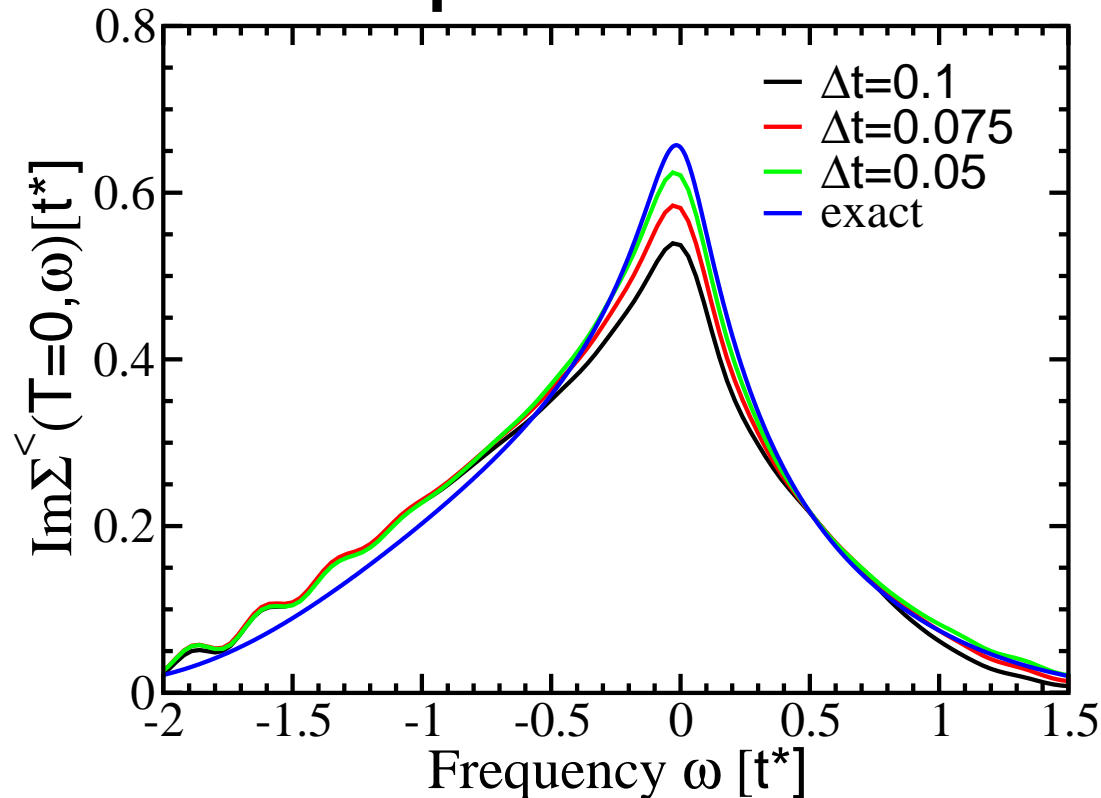
- There are three main numerical issues: (i) how large a **time cutoff** is used for the Kadanoff-Baym contour; (ii) how small a **step size** is taken for the discretization; and (iii) how small a **step size** is taken for the two-dimensional quadrature.
- We expect the **time cutoff** plays the biggest role in determining the interacting DOS, especially in the insulating regime, where the gap at low frequency induces a **long-time structure** to the Green's function and self-energies.
- The **discretization error** stems from the step size chosen. This is most likely creating similar issues as a **Trotter breakup** does in quantum Monte Carlo simulations.
- The discretization of the energy plays a role in determining when **spurious structure** can enter the frequency dependence. We illustrate this here.

Test versus equilibrium results (U=1)



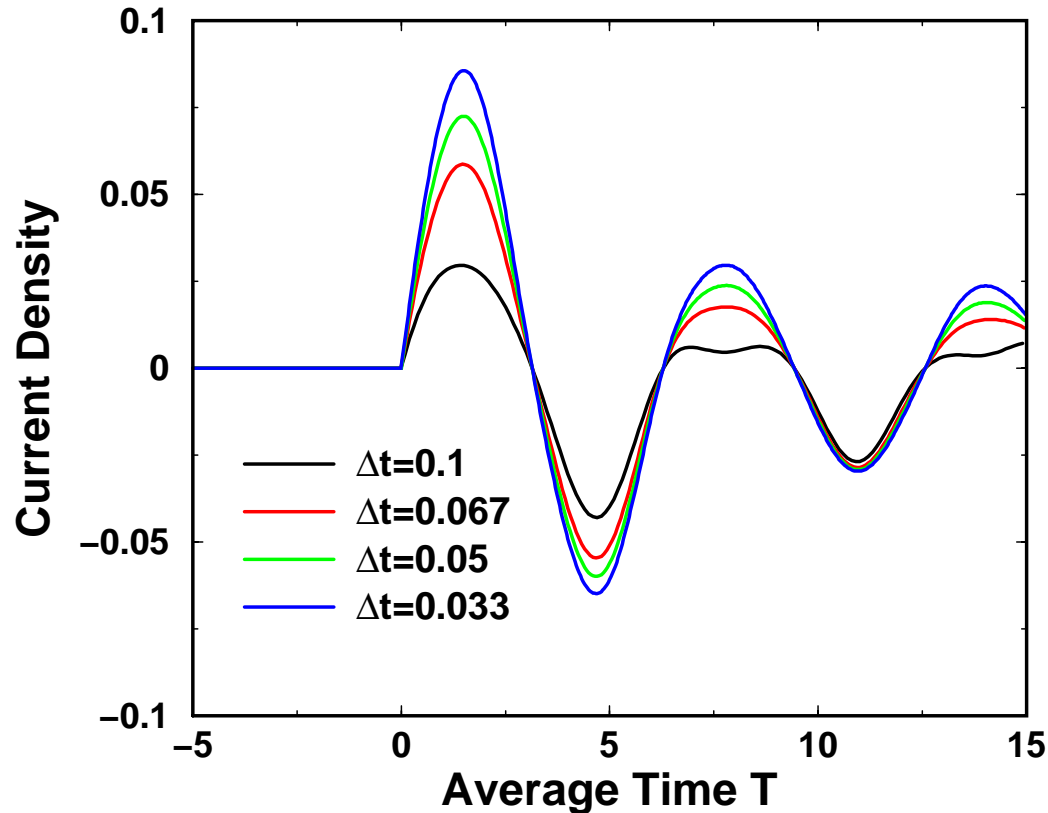
In a nonequilibrium formalism we use **Wigner coordinates** of the average and relative times (right panel). The relative time is Fourier transformed to a frequency (left panel). These results show the effect of the **discretization** of the two-dimensional **energy** quadrature.

Test versus equilibrium results (U=1)



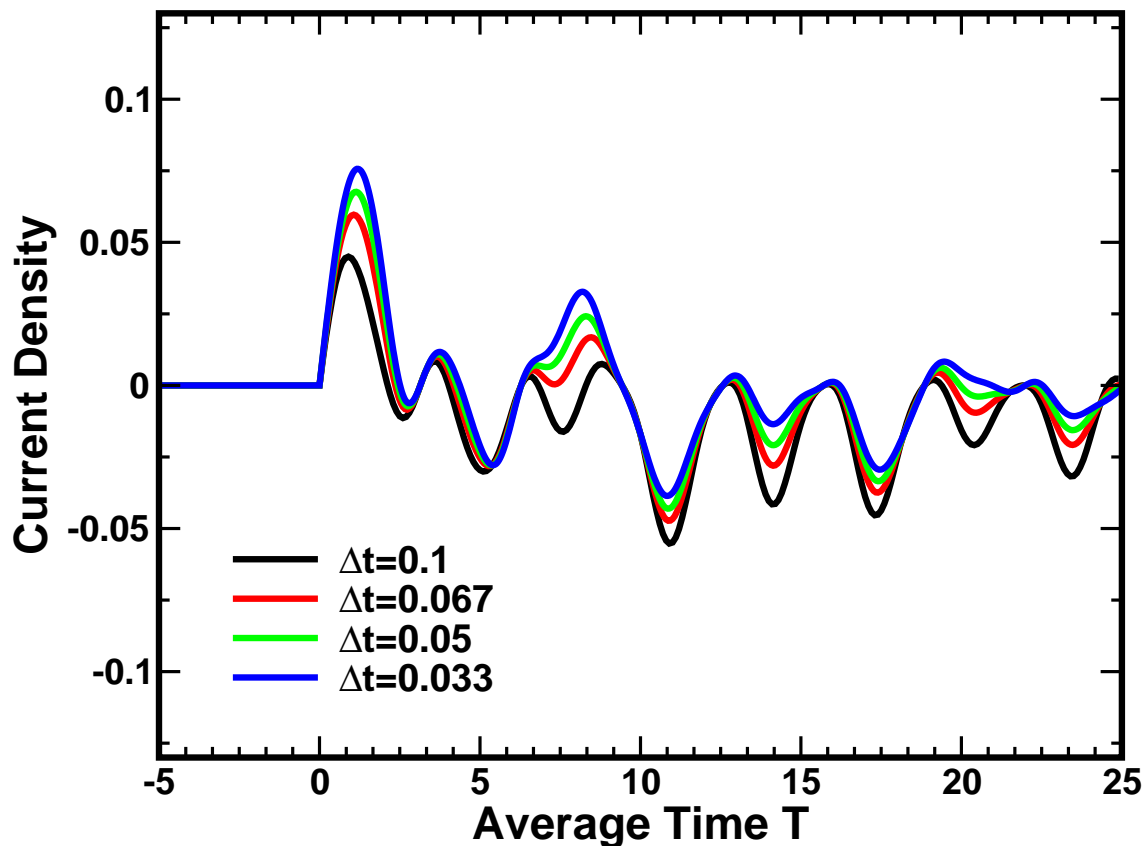
The error from the discretization of the Kadanoff-Baym contour has the **largest effect at small frequencies**. Systematically reducing the discretization gets **closer** to the exact result.

Bloch oscillations ($E=1$, $U=0.5$, $T=1$)



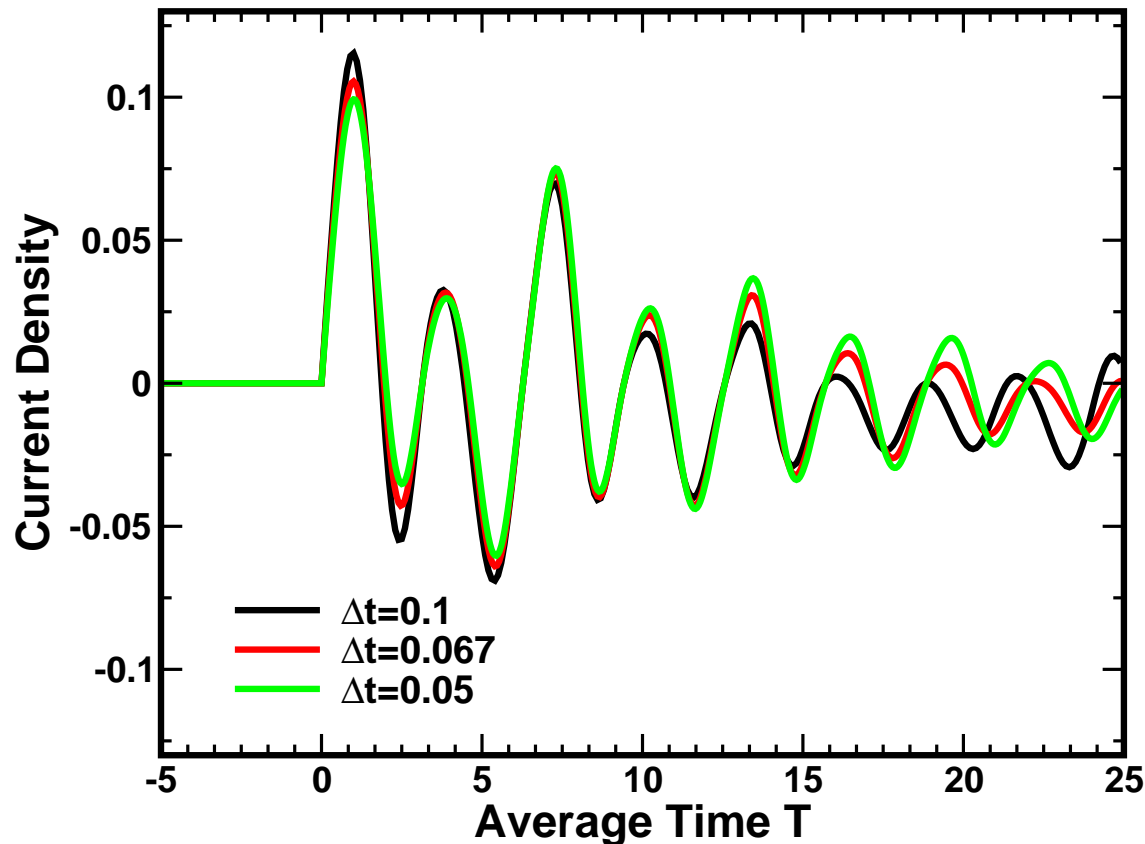
As we reduce the step size, the current appears to approach a limiting value, but the limit is **not approached uniformly**. Note how the amplitude **decreases** as the average time **increases** but we have not yet reached the steady state.

Bloch oscillations ($F=1$, $U=1.5$, $T=1$)



When we are in
insulating
state, the
convergence
appears slower,
requiring a
smaller step size
to be needed.
The results are
also quite
uniform.

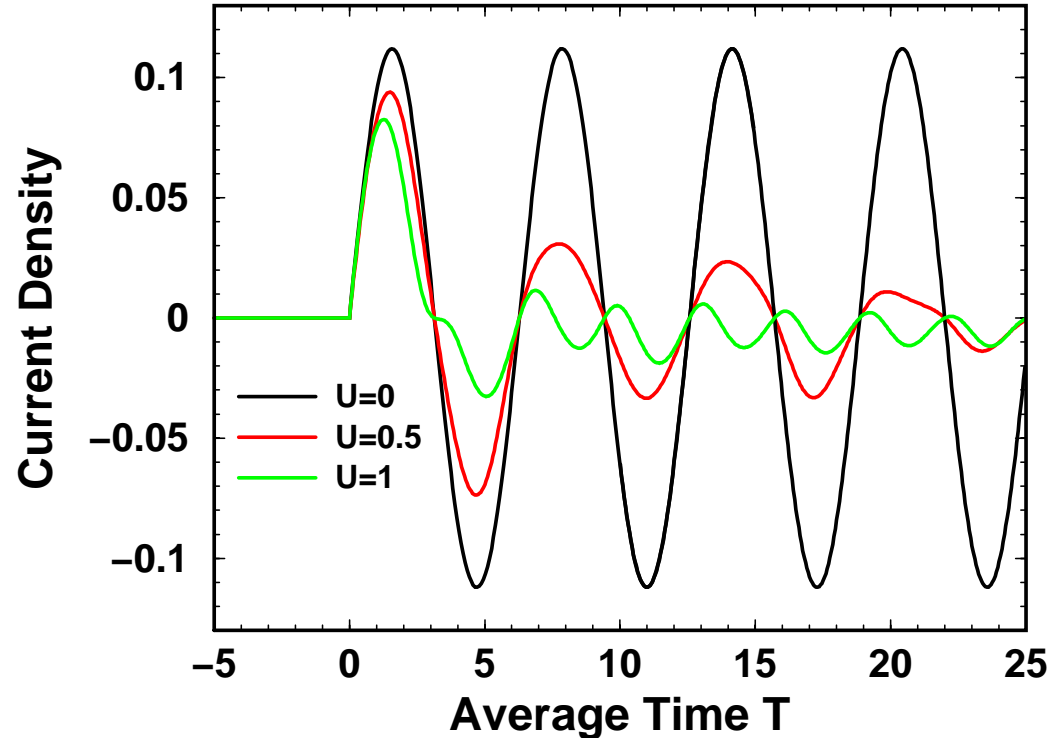
Bloch oscillations ($F=1$, $U=2$, $T=1$)



continues
is made
er. The data
are quite
iminary, as
smallest Δt
e has not yet
converged.

Bloch oscillations ($E=1$, $T=1$)

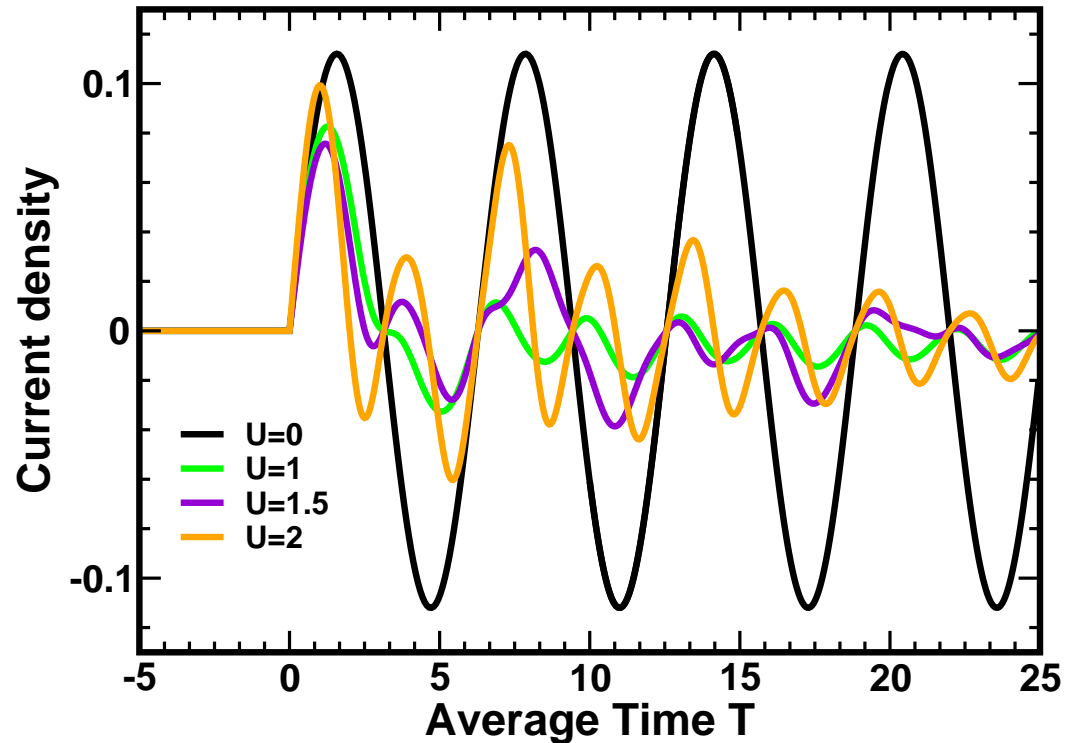
When $U=0$, we have the **conventional Bloch oscillations**. As U increases, so does the scattering, and the **period remains the same**, but the **amplitude is reduced**. As U is increased further it appears like the period changes, but this can be an **artifact of the transient response**; we have not reached the **steady state** yet.



Bloch oscillations ($E=1$, $T=1$)

Now when we move in the insulating regime the results seem strange. The $U=2$ curve lies **below** that of $U=1$ for small times, but then becomes **larger** at bigger times. Is this a real effect or a convergence issue? The results for $U=1.5$ seem even worse in this respect.

*Can the Mott insulator be driven to a better nonlinear metallic state if the gap is small enough?
Does the period change with U ?*



Conclusions

- Showed how to implement an **efficient parallel algorithm** to solve the **Keldysh problem** for strongly correlated electrons described by the Falicov-Kimball model.
- The procedure was applied to the **question of Bloch oscillations** and **how they disappear** as scattering is increased.
- We quantified the **accuracy** by comparing the results of our algorithm to **equilibrium results** obtained with other, direct methods.

Future work

- Solve the numerical issues associated with the **Mott insulating phase** to be able to produce accurate results in that regime.
- Systematically examine the phenomenon of **Bloch oscillations**, including using a different method to directly determine the **steady state**.
- Apply the nonequilibrium formalism to **nanostucture transport** and investigate both **electrical** and **thermal** transport within a **self-consistent framework**.