Nonlinear Response of Strongly Correlated Materials to a Large Electric Field

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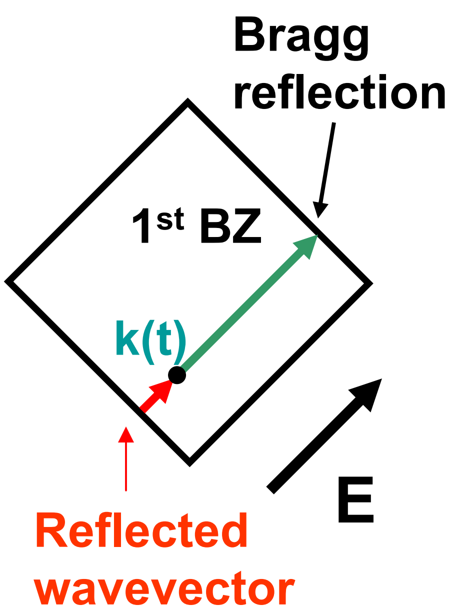
Electrons driven by a constant electric field

- In a **semiclassical** picture, the electron momentum, written as **ħk=P**, evolves with a linear time-dependence corresponding to the **acceleration** due to the field: **k(t)=eEt/ħ**.
- **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 sources of **scattering**, which also 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 **ε(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 **$v(k)=d\varepsilon(k)/d\hbar k$**. So they carry a current equal to **$ev(k)$** summed over all wavevectors **k**.
- As the **wavevector** evolves over the 1BZ, it changes **periodically**, and so does **v(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. It only occurs if the energy stored in the field is large enough to induce a tunneling between bands. ***This will not be considered in this work*.**
- 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 start their acceleration in the field again. ***This is why Bloch oscillations are not commonly seen in metals*.**

Drude-Sommerfeld picture

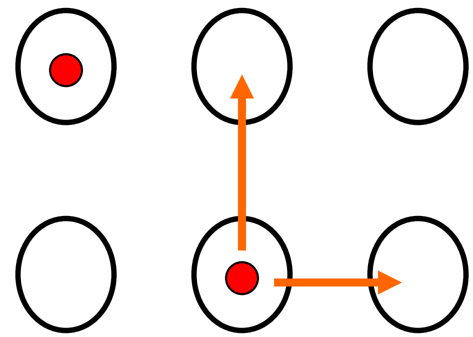
- When the scattering time is **short enough**, the current starts to increase linearly with time, but is **randomized** before it can Bragg reflect.
- In the **steady state**, the current is **linearly proportional** to the electric field $j=\sigma E$, with σ being the dc conductivity. This is often 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!*

Military Relevance

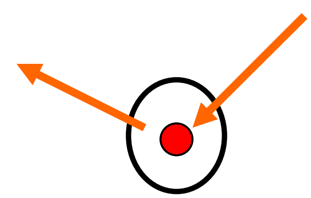
- Strongly correlated electron materials have properties (conductivity, magnetism, superconductivity) that can be **tuned** by varying pressure, temperature, or doping.
- They have potential for use in so-called **smart materials technologies**.
- Many military applications involve subjecting materials and devices to extreme conditions with **strong electromagnetic fields**, where **nonequilibrium** and **nonlinear** effects are important (*examples include lightning strikes or electronic warfare attacks on devices, or high-power applications*)

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**.

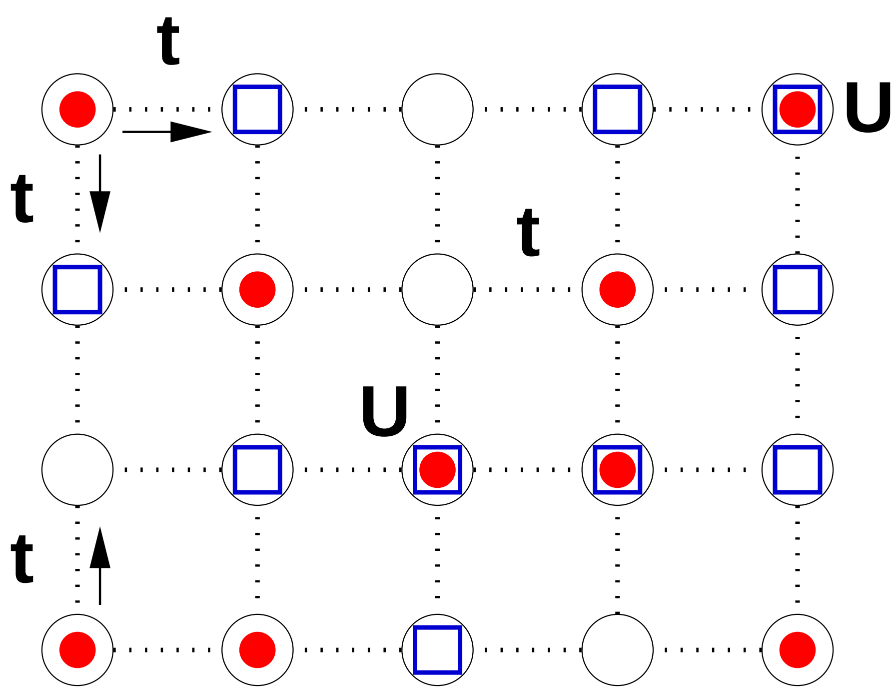


Lattice



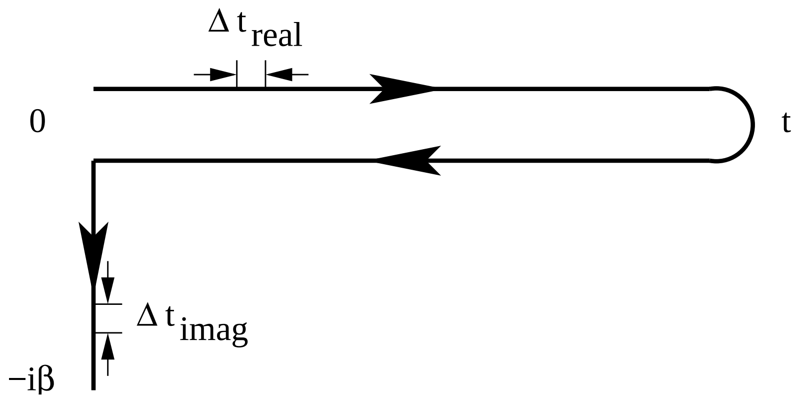
Impurity site

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

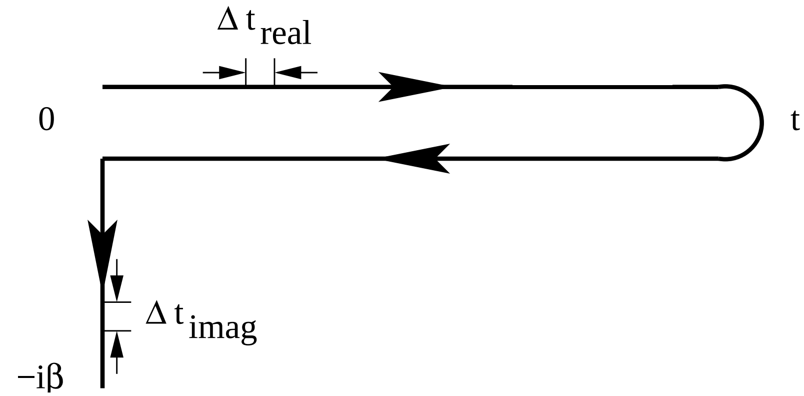
$G_0 = (G_{loc}^{-1} + \Sigma)^{-1}$

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.

Computational elements



The key issue in calculating the real-time Green's function is to evaluate the **Dyson equation of a continuous integral operator** defined on the Kadanoff-Baym-Keldysh contour.

This operator is first **discretized** on a grid to be represented by **finite-dimensional** matrices.

We need to integrate the dependence of the **matrix elements** over a **two-dimensional** energy space.

Each matrix element is constructed from **matrix inverses** and **matrix multiplications**. We typically work with (approximately 20,000) **general complex matrices** of size up to about 2200X2200.

Since the only information needed to generate the matrices is the local self-energy matrix Σ, the electric field **E**, and the temperature T, **this procedure is easily parallelized**.

Parallel implementation

(1) **Solve** for the local self-energy using Dyson's equation on the master node.

(2) **Broadcast** the self-energy Σ(t,t'), the field E, and the temperature T to **all slave nodes**.

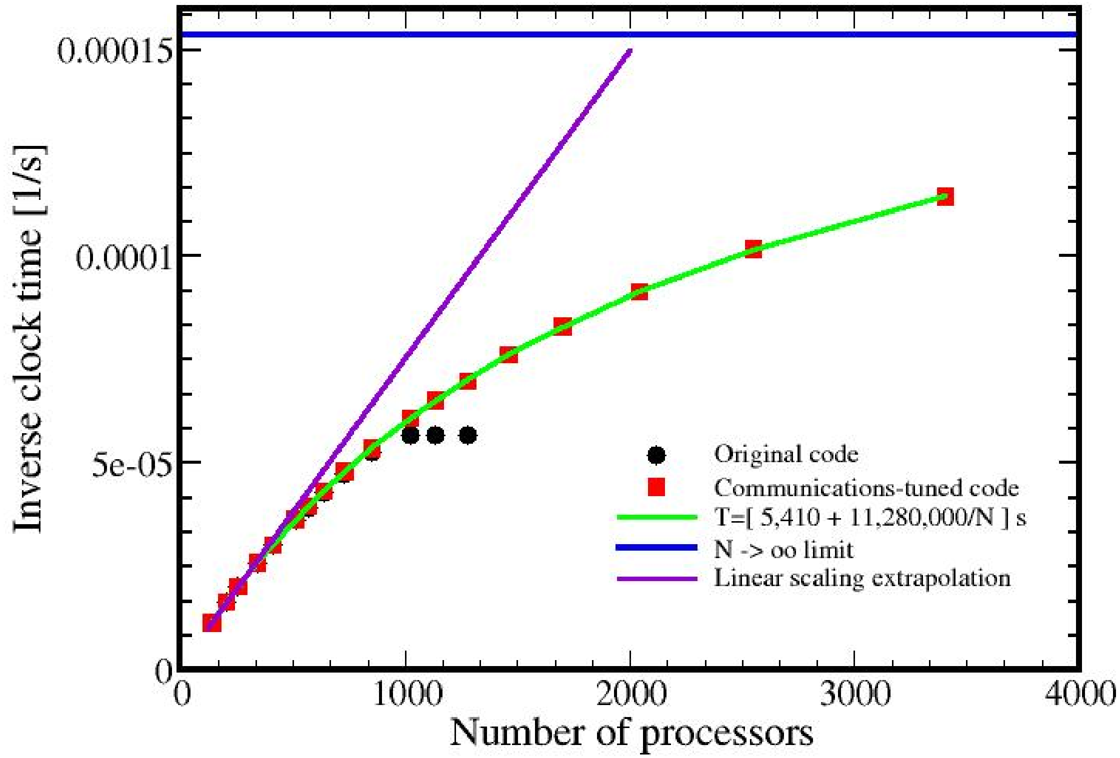
(3) **Send** each **slave node** a value of energies for the momentum dependence of the Green's function and compute the **matrix** that enters the two-dimensional quadrature. LAPACK routines are used for efficiency.

(4) **Store** data on the **slave nodes** for accumulation; use **recursive binary gather** to accumulate for the **master**.

(5) **Solve** the impurity problem on the **master** node to determine the new self energy.

(6) **Repeat until converged**. Then extract the interesting time-dependent quantities like the current as a function of time, or the distribution of the electrons.

Scaling



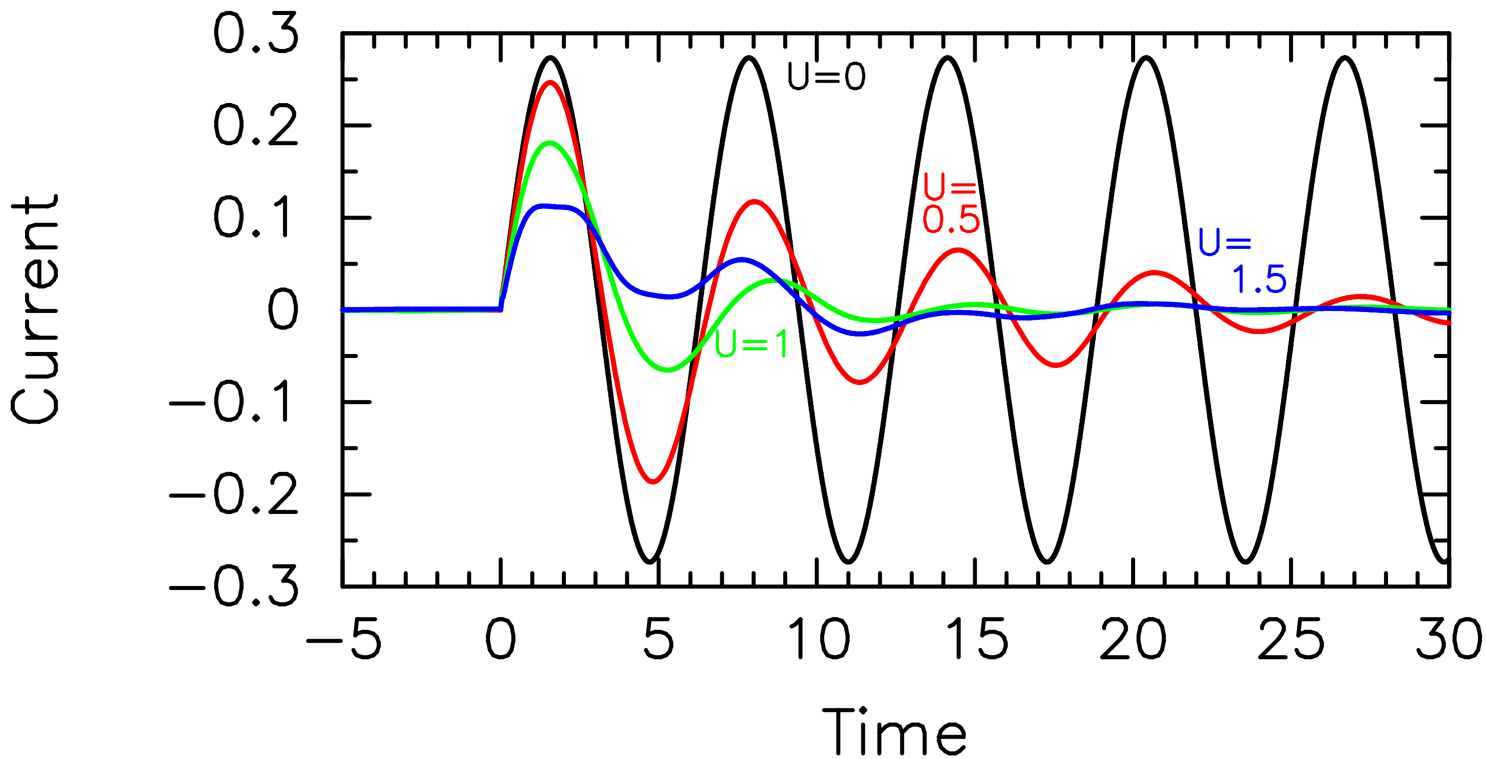
The algorithm has a natural parallelizable piece and a serial piece, so it can never achieve pure linear scaling. The green curve is the scaling prediction, red squares the actual data.

When originally scaled, the data showed a bottleneck when increased beyond about 900 processors. This was a communications issue, resolved by using a recursive binary gather operation. Scaling is sublinear, but achieves about 70% of linear efficiency on 1500 procs.

Recursive Binary Gather

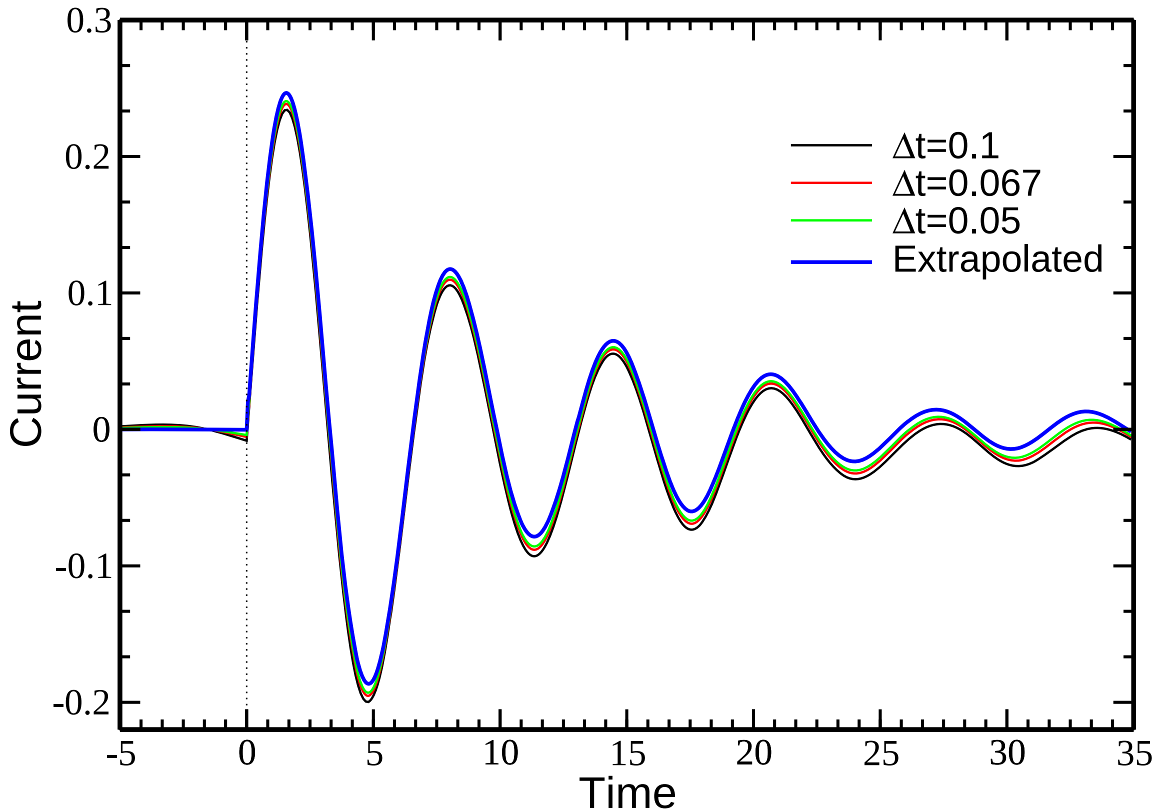
- In computing the two-dimensional matrix-valued integration, we originally used a **many-to-one** communication, sending results to the master node immediately after being completed.
- But all nodes finished at **about the same time**, creating a data bottleneck with the master node.
- The **recursive binary gather** operation has each node store their results until all computation is finished, then the slave nodes are divided in two, and one half sends their data to the other half.
- The sending of data and “collapsing” of the nodes is repeated until all data is on one node, which is then sent to the master node.

Bloch oscillations in metals



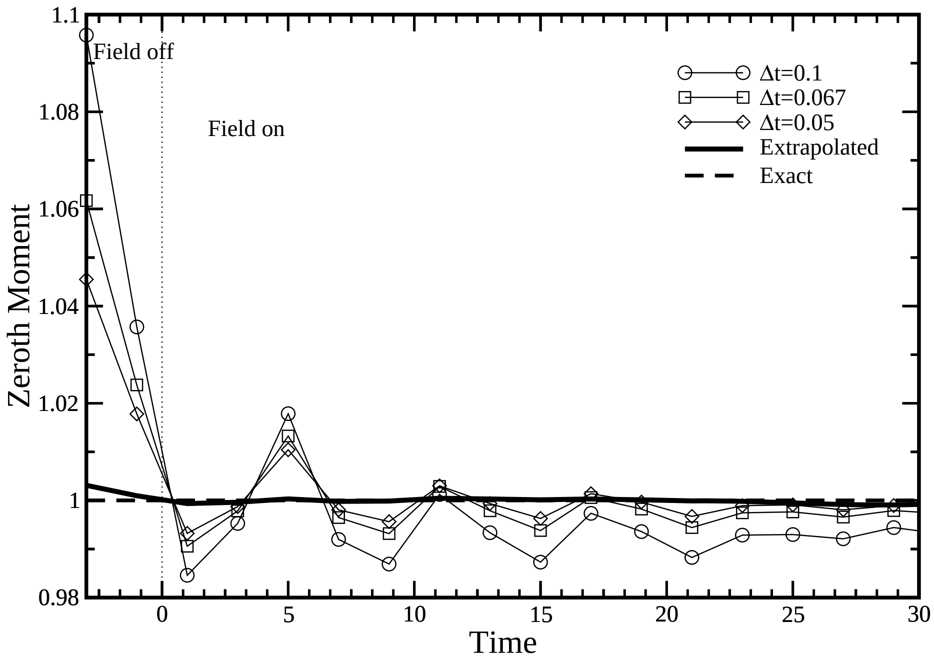
As the scattering increases, the amplitude of the current decays faster, but we cannot tell whether the oscillations survive at long time, or are completely damped.

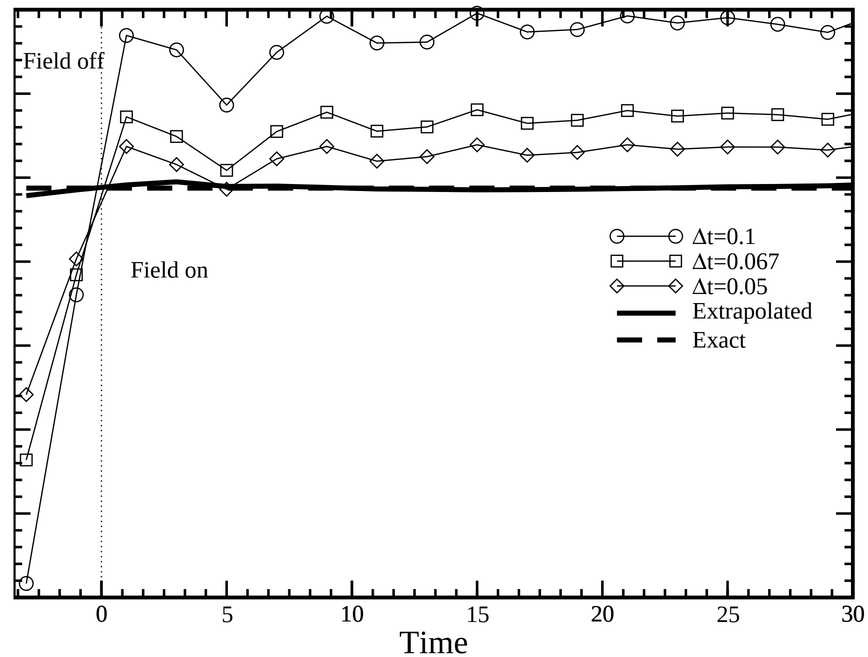
Accuracy of results—scaling of the current



The accuracy of the current is illustrated here with a plot showing results for different discretizations and the extrapolated current.

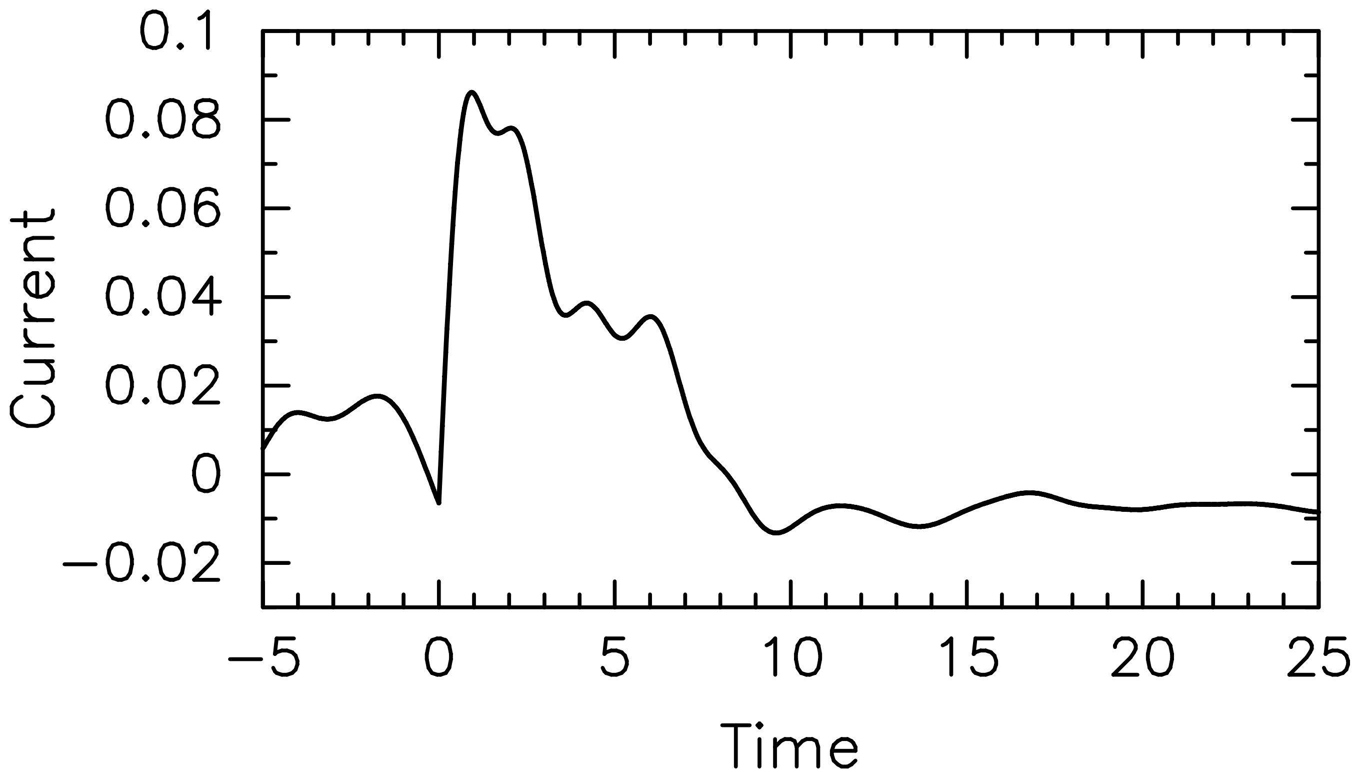
Accuracy of the results---scaling of moments





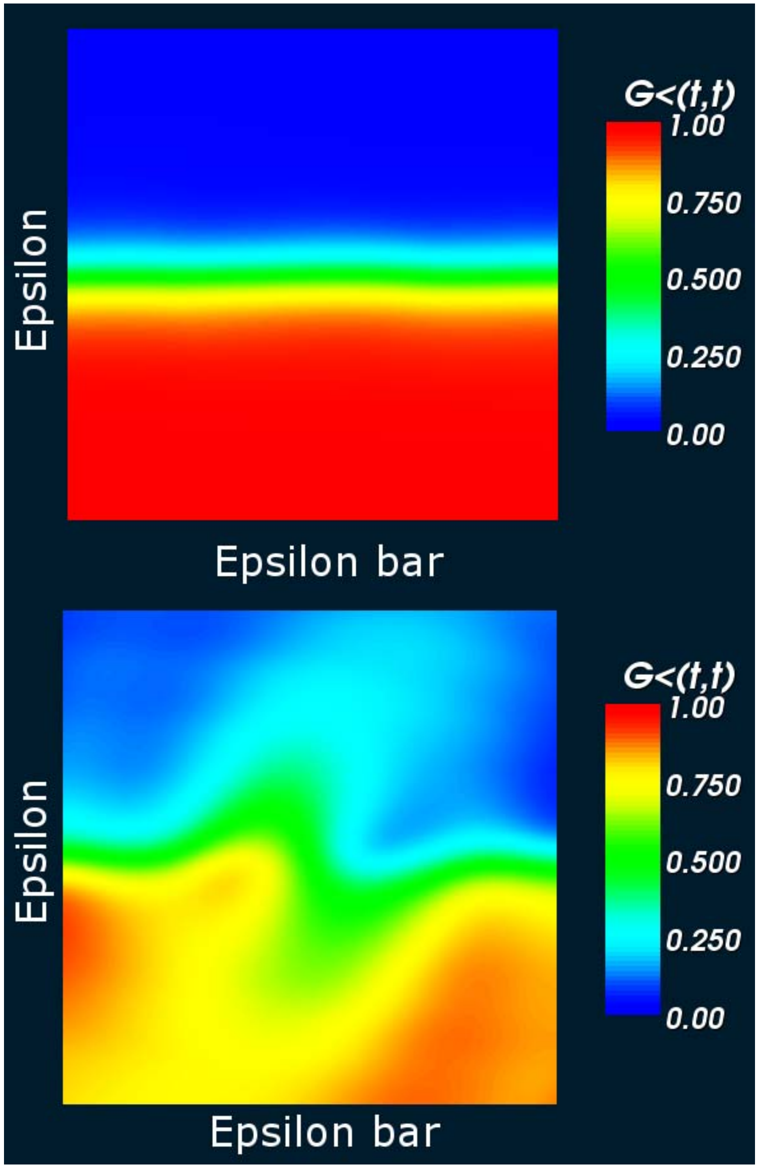
Exact results are known for the equal time Green's functions and their first two derivatives.
Extrapolating the results to zero discretization size yields **excellent agreement** with the exact results.

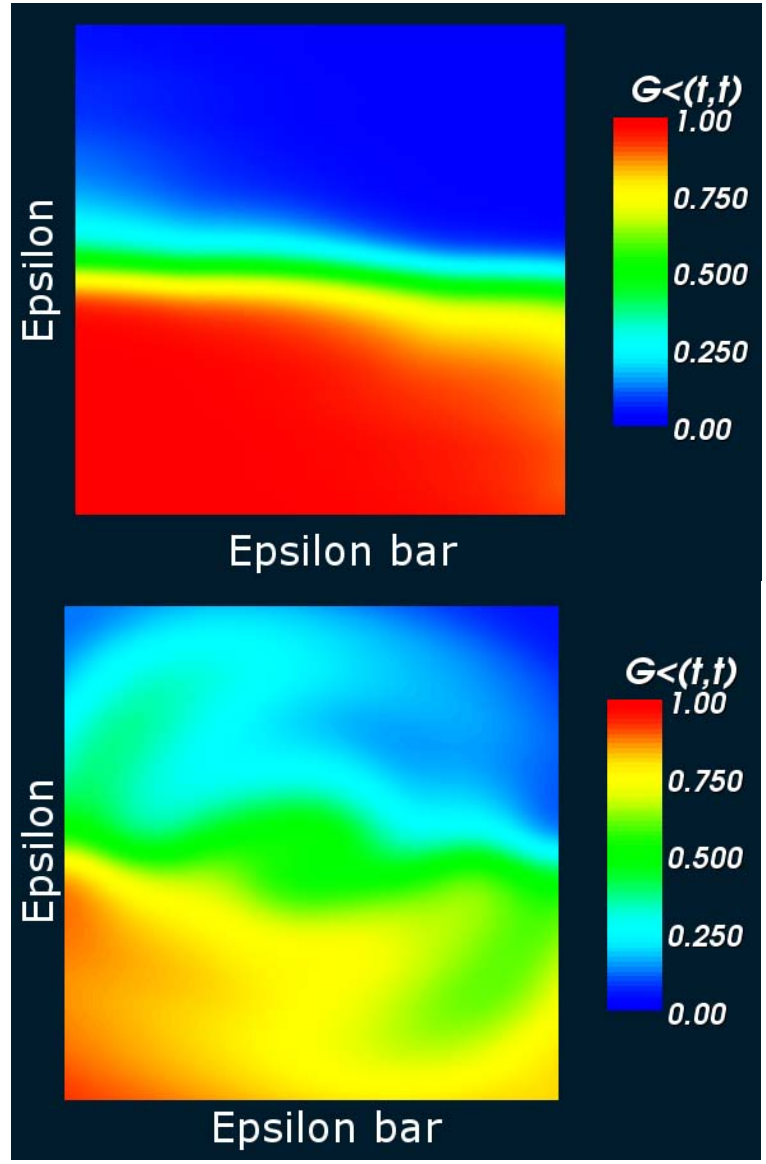
Current in the Mott Insulator



In the Mott insulator, scaling does not work well for the discretization size we can get down to. The current looks irregular with no clear Bloch oscillations.

Distribution function of the electrons





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**.**
- Our algorithm showed **efficient usage and good scaling** to thousands of processors on the **Cray-XT3** (we used a total of about **600,000 cpu-hours** on the project).