Nonequilibrium dynamical mean-field theory

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(Dated: March 23, 2022)

The many-body formalism for dynamical mean-field theory is extended to treat nonequilibrium problems. We illustrate how the formalism works by examining the transient decay of the oscillating current that is driven by a large electric field turned on at time $t = 0$. We show how the Bloch oscillations are quenched by the electron-electron interactions, and how their character changes dramatically for a Mott insulator.

PACS numbers: 71.27.+a, 71.10.Fd, 71.45.Gm, 72.20.Ht

Introduction. Dynamical mean-field theory (DMFT) was introduced in 1989 as a technique to solve the quantum many-body problem by taking the limit where the number of spatial dimensions goes to infinity $\mathbf{1}$. In this limit, with a proper scaling of the hopping matrix elements, the electron-electron correlations are described by a local self-energy. Hence the many-body problem on a lattice is mapped onto an effective many-body problem for a single-site impurity (in a time-dependent field), with a self-consistency condition that fixes the time-dependent field so that the Green’s function for the impurity is identical with the local Green’s function for the lattice. Since then, DMFT has been employed to solve virtually all many-body problems described by model Hamiltonians $\mathbf{2}$, has been generalized to describe strong electron correlations in real materials $\mathbf{3}$ and to describe inhomogeneous systems $\mathbf{4, 5}$. All of this work has focused on the equilibrium case. In this contribution, we illustrate how to generalize DMFT to nonequilibrium situations, and we present results for how the Bloch oscillations of a strongly correlated material are quenched by electron-electron interactions, and how their character changes after the Mott metal-insulator transition.

Bloch $\mathbf{6}$ and Zener $\mathbf{7}$ theorized that electrons on a lattice undergo an oscillatory motion when placed in a uniform static electric field, because the electron wavevector, which evolves under the electric field, is Bragg reflected whenever it reaches a Brillouin zone boundary. But in metals, Bloch oscillations have never been seen, because the electron relaxation time is so short, the electrons are scattered before they reach the zone boundary and Bragg reflect. Bloch oscillations have been observed in semiconducting heterostructures $\mathbf{8}$, Josephson junctions $\mathbf{9}$, and cold-atom systems $\mathbf{10}$.

Formalism. The many-body formalism for nonequilibrium dynamical mean-field theory is straightforward to develop within the Kadanoff-Baym-Keldysh approach $\mathbf{11, 12}$. Because nonequilibrium problems are not time-translation invariant, we need to work with Green’s functions that depend on two times. There are two independent Green’s functions that need to be determined—the retarded Green’s function $G^R$, which describes the density of available quantum-mechanical states, and the lesser Green’s function $G^<$, which determines how electrons occupy those quantum states. Both Green’s functions can be extracted from the contour-ordered Green’s function, which is defined for any two time values that lie on the Kadanoff-Baym-Keldysh contour shown in Fig. $\mathbf{13}$. We imagine our system to be in equilibrium until time $t = 0$ where a field is turned on. The contour starts at some time before the field is turned on, runs out to a maximal time, then returns to the original time, and finally moves parallel to the negative imaginary axis a distance $\beta$ (equal to the inverse of the temperature of the original equilibrium distribution).

Since the many-body perturbation theory diagrams are identical in structure for equilibrium and nonequilibrium perturbation theories $\mathbf{13}$, the perturbative analysis of Metzner $\mathbf{14}$ guarantees that the nonequilibrium self-energy is also local in DMFT. Hence, the nonequilibrium DMFT problem can be mapped onto an impurity problem in time-dependent fields, just like the equilibrium problem, except now the fields have two time arguments. The basic structure of the iterative approach to solving the DMFT equations $\mathbf{15}$ continues to hold. We start with a guess for the self-energy (which is usually

FIG. 1: Kadanoff-Baym-Keldysh contour for the two-time Green’s functions in the nonequilibrium case. We take the contour to run from $-5$ to $t_{\text{max}}$ and back, and then extends downward parallel to the imaginary axis for a distance of $\beta$. The field is turned on at $t = 0$; i.e., the vector potential is nonzero only for positive times.
chosen to be equal to the equilibrium self-energy), then we sum the momentum-dependent Green’s function over the Brillouin zone to produce the local Green’s function. Next the dynamical mean-field for the impurity problem is extracted by using Dyson’s equation for the local Green’s function and self-energy, the impurity problem is solved in the dynamical mean-field to produce the impurity Green’s function, and Dyson’s equation is used again to extract the impurity self-energy. In the self-consistent solution of the DMFT equations, the impurity self-energy will be equal to the lattice self-energy. If they are different, then the new lattice self-energy is taken to be equal to the new impurity self-energy, and the loop is iterated until it converges. The nonequilibrium algorithm is modified as follows: (i) the summation over the Brillouin zone now requires at least a double integral over two band energies; (ii) the Green’s functions are described by discrete matrices with time indices that run over the contour; and (iii) the impurity problem solver must be generalized to the nonequilibrium case.

For concreteness, we assume the electric field \( \mathbf{E}(t) \) is spatially uniform, but can depend on time (we assume that the magnetic field, however, is small, and neglect all magnetic-field effects). We work in the Hamiltonian gauge, where the scalar potential vanishes, and the electric field is determined by a time derivative of the vector potential \( \mathbf{E}(t) = -d\mathbf{A}(t)/dt \), in units where \( c = 1 \). The noninteracting problem of Bloch electrons in an electric field can be solved exactly by using the Peierls substitution \[16, 17\], and if we take the electric field to lie along the diagonal direction, then the noninteracting momentum-dependent Green’s functions on the lattice depend only on two explicit functions of momentum

\[
\epsilon_k = -\frac{t^*}{2\sqrt{d}} \sum_{i=1}^{d} \cos k_i, \quad \tilde{\epsilon}_k = -\frac{t^*}{2\sqrt{d}} \sum_{i=1}^{d} \sin k_i,
\]

rather than all components of the momentum. Here we set the lattice constant \( a \) equal to 1, and we consider the case of nearest-neighbor hopping on a hypercubic lattice in \( d \)-dimensions with a hopping parameter \( t = t^*/2\sqrt{d}; \) \( t^* \) will be taken as the energy unit. In the limit \( d \to \infty \), the two “band energies” are distributed with a joint Gaussian density of states \[18\]

\[
\rho(\epsilon_k, \tilde{\epsilon}_k) = \frac{1}{\pi} e^{-\epsilon_k^2 - \tilde{\epsilon}_k^2}.
\]

In the interacting case, the dressed contour-ordered Green’s function satisfies Dyson’s equation, with a local self-energy, so that

\[
G_\text{imp}(\mathbf{k}, t, t') = \left[ G^{-1}_0(\mathbf{k}, t, t') - \Sigma(t, t') \right]^{-1},
\]

where the Green’s functions and self-energy are continuous matrix operators defined on the contour \( (i. e., \) the time indices of the matrices run along the contour), and

\[
G_0(\mathbf{k}, t, t') = i[f(\epsilon_k - \mu) - \theta_c(t, t')e^{-i\mu(t-t')}]
\times e^{-i\epsilon_k \sin \theta c \sin E t / c E}
\times e^{-i\epsilon_k \cos \theta c \cos E t' / c E},
\]

where \( e \) is the electron charge, \( E \) is one component of the electric field along a Cartesian axis (all components are equal for a field directed along the diagonal), and \( f(x) = 1/[1 + \exp(x)] \) is the Fermi-Dirac distribution (we set \( \hbar = 1 \)). The symbol \( \theta_c(t, t') \) is equal to one if \( t \) is ahead of \( t' \) on the contour and is zero otherwise. Calculating the local Green’s function requires evaluating a two-dimensional integral over \( \rho(\epsilon_k, \tilde{\epsilon}_k) \) of a matrix-valued integrand, which requires a matrix inversion to determine it. Once the local Green’s function \( G \) has been found, we use Dyson’s equation to extract the dynamical mean-field, denoted \( \lambda(t, t') \), which satisfies

\[
\lambda(t, t') = (i\partial_t^c + \mu)\delta_c(t, t') - G^{-1}(t, t') - \Sigma(t, t'),
\]

where the derivative with respect to time is taken along the contour, and the delta function is defined on the contour such that \( \int d\delta_c(t, t')F(t') = F(t') \).

Next, the impurity problem must be solved for electrons evolving in the dynamical mean field. In general, algorithms have not yet been developed to solve this problem for all Hamiltonians, but the impurity problem can be immediately solved for the spinless Falicov-Kimball model \[19\], which involves single-band conduction electrons hopping on a lattice, and localized electrons which do not move but do interact with the conduction electrons when they are in the same unit cell via a screened Coulomb interaction \( U \). The Hamiltonian (in the absence of a field) is then

\[
\mathcal{H} = -\frac{t^*}{2\sqrt{d}} \sum_{(ij)} (c_i^\dagger c_j + c_j^\dagger c_i) + U \sum_i w_i c_i^\dagger c_i.
\]

Here, we have \( c_i^\dagger \) (\( c_i \)) create (annihilate) a spinless conduction electron at site \( i \) and \( w_i = 0 \) or 1 is the localized electron number operator at site \( i \). Although the Falicov-Kimball model is a simple many-body physics model, it does have a Mott metal-insulator transition (but the model does not include Zener tunneling because there are no higher energy bands). The solution to the impurity problem can be found by solving the equations of motion for the contour-ordered Green’s function resulting in

\[
G_\text{imp}(t, t') = (1 - w_i) \left[ (i\partial_t^c + \mu)\delta_c(t, t') - \lambda(t, t') \right]^{-1}
\times w_i \left[ (i\partial_t^c + \mu - U)\delta_c(t, t') - \lambda(t, t') \right]^{-1}.
\]
with \( n_1 \) the average localized electron filling. The Dyson equation in Eq. (3) is then employed to extract the impurity self-energy, and the algorithm is iterated until it converges.

There are a number of important technical details in these calculations. First, we discretize the contour (we choose a real-time spacing \( \Delta t \) which varies from 0.1 to 0.0333, and we fix the spacing along the imaginary axis to \( \Delta \tau = 0.1i \)) and evaluate integrals over the contour by discrete summations using the midpoint rectangular integration rule. The matrix operators are general complex matrices, which are manipulated using standard linear algebra packages (the largest matrices used are about \( 2200 \times 2200 \)). In addition, the delta function changes sign along the negative real time branch, and is imaginary along the last branch of the contour while the derivative of the delta function is evaluated by a two-point discretization involving the diagonal and the first subdiagonal, but one also needs to include one matrix element at the upper right corner to preserve the proper boundary conditions.

We perform the two-dimensional energy integration by Gaussian quadrature with both 100 and 101 points in each dimension, and we average the two results. Since the calculation of each matrix in the integrand of the integral is independent of every other quadrature point, this part of the code is easily parallelized. We require 20,201 matrix inversions for each DMFT iteration. The impurity solver is a serial code, that cannot be parallelized, because the matrix operations need to all be performed in turn. We typically require between ten and fifty iterations to reach convergence of the results (the total computer time for the calculations presented here was about 600,000 cpu-hours on a Cray XT3). Once converged, we calculate the current by evaluating the operator average

\[
\langle j(t) \rangle = -ei \sum_k \mathbf{v}(k - eEt)G^{<}(k, t, t).
\]

The velocity component is \( v_i(k) = t^* \sin(k_i) / \sqrt{d} \), and all components of the current are equal when the field lies along the diagonal. We also calculate the equal time retarded and lesser Green’s functions and their first two derivatives and compare the results to the exact values \( \langle 21 \rangle \). In general, these “moments” are quite accurate as the step size is made smaller, and we find that if we use a Lagrange interpolation formula to extrapolate the results to \( \Delta t = 0 \), we can achieve even higher accuracy for most values of \( U \). Details of these numerical issues and of the accuracies will be presented elsewhere \( \langle 21 \rangle \).

Numerical results. We produce numerical calculations of the nonequilibrium current as a function of time for the case of half-filling, where the conduction electron and the localized electron fillings are each equal to 0.5. This system has a metal-insulator transition at \( U = \sqrt{2} \). In the case where there is no scattering \((U = 0)\), the Bloch oscillations continue forever. In the presence of scattering, the Bloch oscillations maintain the same approximate “periodicity”, but the amplitude decays. In Fig. 2 we plot the current for the noninteracting case, the case of a strongly scattering metal \((U = 0.5, \text{red})\), the case of an anomalous metal \((U = 1, \text{green})\), and of a near critical insulator \((U = 1.5, \text{blue})\). The initial temperature of the system satisfied \( \beta = 10 \), and the field is turned on at \( t = 0 \). The electric field is set equal to one in magnitude, \( E = 1 \). Note how the Bloch oscillations are damped as the scattering increases. Although a Boltmann equation approach always predicts that the oscillations are damped and disappear on a time-scale on the order of the relaxation time, and approach a constant steady state, we do not see this full evolution within the time-window that we performed these calculations. Most of the data given here involve a scaling of the data with \( \Delta t = 0.1, 0.067, 0.05, \) and 0.04 to the \( \Delta t \to 0 \) limit. Note that the data for a fixed step size in time always shows a small current for \( t < 0 \), but when scaled, the current is completely flat and vanishes for negative times (we estimate the scaled data has a relative error of less than 1%). The Bloch oscillations are always nearly as large as in the noninteracting case, and then they begin to decay. We are unable to determine whether they decay to a constant value as predicted by the Boltzmann equation, or whether there are oscillations present in the steady state. In the quantum-mechanical system, there are two relevant time scales, the average time, and the relative time. The relative time governs the decay of the quasiparticle-like excitations, and this decay becomes rapid as the scattering increases. The average time governs the Bloch oscillations, and it is not obvious from either the formalism or our results whether the steady-state current must be a constant, or whether it can oscillate if the electric field is large enough (of course, with a period as short as these Bloch oscillations would have, they could not be measured in an experiment).

In Fig. 2 we show results for the current with \( U = 0.5 \) and \( U = 1 \). We take the time window to be larger here.
to see if we can shed any further light on how the data evolves to the steady state but the time window is still too short. In Fig. 4, we plot the current as a function of time for the small-gap Mott insulator with $U = 2$. It is much harder to achieve convergence for these results, and the scaling approach does not seem to work well, as the calculated moments are less accurate for the scaled data, than for the data at the smallest $\Delta t$ value (which is 0.0333 for the $U = 2$ data; relative errors here are probably at the 10% level). Note how there is nonzero current at negative times, indicating that this data is not quite as accurate as the data with smaller $U$ values. Note further, that the oscillatory behavior is quite irregular here, and it is not an exponentially decaying Bloch oscillation anymore. The Bloch oscillations rapidly change their character as the metal-insulator transition is crossed.

Summary. We have developed the formalism for nonequilibrium dynamical mean-field theory. The basic method is similar to that of the equilibrium case, except we need to work in a real-time representation for all Green’s functions, self-energies, and dynamical mean fields. The summation over momentum to yield the local Green’s function now involves a two-dimensional integration of a matrix-valued integrand. We presented numerical results for the Bloch oscillations in the presence of a large electric field, and showed how they decay as a function of time when there is electron-electron scattering. In the transient response calculations presented here, we cannot determine whether oscillations are present in the steady state. We are currently working on a steady-state formalism should be able to shed light on that issue.

Acknowledgments. We acknowledge useful conversations with J. Serene and A. Joura. This work is supported by the N. S. F. under grant number DMR-0210717 and by the O. N. R. under grant number N000140510078. Supercomputer time was provided by the ERDC XT3 under a CAP phase II project in the winter of 2006.

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