Nonequilibrium Dynamical Mean-Field Theory

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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.

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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 [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 [2], has been generalized to describe strong electron correlations in real materials [3] and to describe inhomogeneous systems [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 [6] and Zener [7] theorized that electrons on a lattice undergo an oscillatory motion when placed in a uniform static electric field, because the electron wave vector, 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 [8], Josephson junctions [9], and cold-atom systems [10].

Formalism.—The many-body formalism for nonequilibrium dynamical mean-field theory is straightforward to develop within the Kadanoff-Baym-Keldysh approach [11,12]. Because nonequilibrium problems are not time-translation invariant, we need to work with Green's functions that depend on 2 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. 1. 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 β (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 [13], the perturbative analysis of Metzner [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 [15] continues to hold. We start with a guess for the self-energy (which is usually chosen to be equal to the equilibrium self-energy), then we sum the momentum-dependent Green's function over the Brillouin zone to

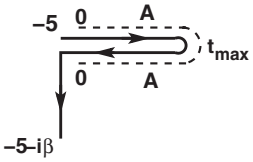

FIG. 1. Kadanoff-Baym-Keldysh contour for the two-time Green's functions in the nonequilibrium case. We take the contour to run from -5 to $t_{\max}$ and back, and then extends downward parallel to the imaginary axis for a distance of β. The field is turned on at $t = 0$; i.e., the vector potential is nonzero only for positive times.

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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

$$\epsilon_{\mathbf{k}} = -\frac{t^*}{2\sqrt{d}}\sum_{i=1}^{d}\cos\mathbf{k}_i, \qquad \bar{\epsilon}_{\mathbf{k}} = -\frac{t^*}{2\sqrt{d}}\sum_{i=1}^{d}\sin\mathbf{k}_i, \quad (1)$$

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]

$$\rho(\epsilon_{\mathbf{k}}, \bar{\epsilon}_{\mathbf{k}}) = \frac{1}{\pi} e^{-\epsilon_{\mathbf{k}}^2} e^{-\bar{\epsilon}_{\mathbf{k}}^2}. \quad (2)$$

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

$$G(\mathbf{k}, t, t') = [G_0^{-1}(\mathbf{k}, t, t') - \Sigma(t, t')]^{-1}, \quad (3)$$

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 the -1 superscripts denote the matrix inverse of the respective operators. The noninteracting Green's function in a field is (for both times larger than 0, one can easily work out the generalizations for cases when the field has not been turned on, or for more complicated time dependence of the field [17])

$$G_0(\mathbf{k}, t, t') = i[f(\epsilon_{\mathbf{k}} - \mu) - \theta_c(t, t')]e^{+i\mu(t-t')} \times e^{-i\epsilon_{\mathbf{k}}(\sin eEt - \sin eEt')/eE} e^{-i\bar{\epsilon}_{\mathbf{k}}(\cos eEt - \cos eEt')/eE}, \quad (4)$$

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 $f(x) = 1/[1 + \exp(\beta x)]$ 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 $\rho(\epsilon_{\mathbf{k}}, \bar{\epsilon}_{\mathbf{k}})$ 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

$$\lambda(t, t') = (i\partial_t^c + \mu)\delta_c(t, t') - G^{-1}(t, t') - \Sigma(t, t'), \quad (5)$$

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

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

$$\mathcal{H} = -\frac{t^*}{2\sqrt{d}}\sum_{\langle ij\rangle}(c_i^\dagger c_j + c_j^\dagger c_i) + U\sum_i w_i c_i^\dagger c_i. \quad (6)$$

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

$$G_{\mathrm{imp}}(t, t') = (1 - \langle w_1\rangle)[(i\partial_t^c + \mu)\delta_c(t, t') - \lambda(t, t')]^{-1} + \langle w_1\rangle[(i\partial_t^c + \mu - U)\delta_c(t, t') - \lambda(t, t')]^{-1} \quad (7)$$

with $\langle w_1\rangle = \sum_i \langle w_i\rangle/N$ the average localized electron filling. The Dyson equation in Eq. (5) 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 Δt which varies from 0.1 to 0.02, and we fix the spacing along the imaginary axis to $\Delta\tau = 0.1i$ so our largest matrix is 4100×4100) and evaluate integrals over the contour by discrete summations using the midpoint rectangular integration rule. 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 (some large matrix calculations use 54 and 55 points). 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 10 and 90 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 and 100 000 cpu hours on a SGI ALTIX). Once converged, we calculate the current by evaluating the operator average

$$\langle \mathbf{j}(t) \rangle = -ei \sum_{\mathbf{k}} \mathbf{v}(\mathbf{k} - e\mathbf{E}t) G^{<}(\mathbf{k}, t, t). \tag{8}$$

The velocity component is $v_i(\mathbf{k}) = t^* \sin(\mathbf{k}_i)/2\sqrt{d}$, 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 [20]. 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 [21].

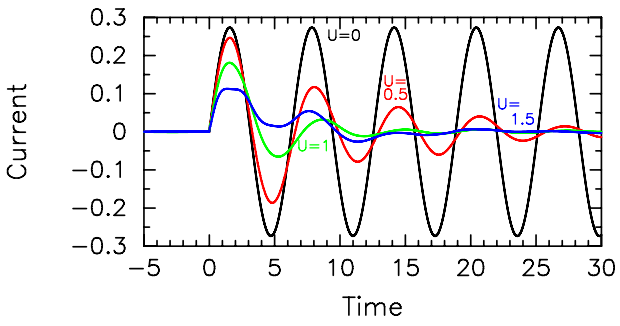

FIG. 2 (color online). Scaled nonequilibrium current for different values of U. Note how the Bloch oscillations are rapidly reduced in amplitude as the scattering increases.

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 ($U = 0.5$, red), the case of an anomalous metal ($U = 1$, green), and of a near critical insulator ($U = 1.5$, 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 Boltzmann 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 initial Bloch oscillations are always nearly as large as in the noninteracting case, and then they begin to decay. For

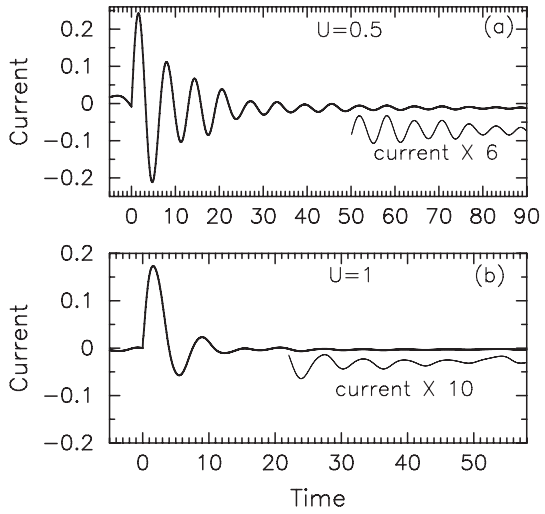

FIG. 3. Nonequilibrium current for (a) $U = 0.5$ ($\Delta t = 0.1$) and (b) $U = 1$ (scaled from $\Delta t = 0.1$ and 0.0667) and longer times. Note how the Bloch oscillations are still present but become more erratic at large times. The current is multiplied by either 6 or 10 to enhance it at large times.

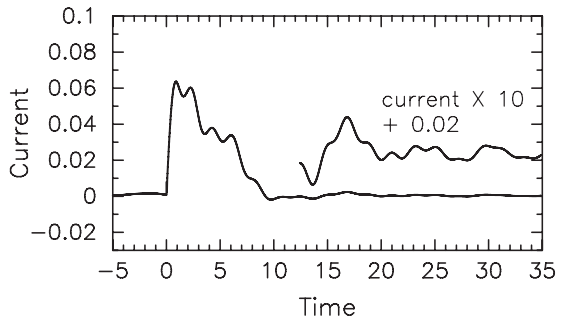

FIG. 4. Nonequilibrium current for $U = 2$ (the tail is multiplied by 10 and shifted by 0.02). Note how the "Bloch oscillations" are now quite irregular.

short times, one can fit to a Bloch oscillation with an exponentially decaying amplitude and a relaxation time that decreases as U increases (our fits are $\tau = 9.6$ for $U = 0.5$, 3.6 for $U = 1$ and 2.2 for $U = 1.5$), but the amplitude in the data is higher than predicted at longer times, and the fits become poor for large U or long time. We are also unable to determine whether the currents 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 quasiparticlelike 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.

In Fig. 3, 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 $U = 2$. Here we need to include data with $\Delta t = 0.0333$, 0.025, and 0.02 to get scaling. Note 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. Perhaps these extra oscillations occur due to a beating effect with the energy gap.

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 Bloch oscillations in a large electric field and how they decay due to electron correlations. In our transient response calculations, we cannot determine whether oscillations are present in the steady state; we are developing a steady-state formalism to resolve this.

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