Nonequilibrium steady states in correlated electron systems — Photoinduced insulator-metal transition and optical response

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Abstract. To reveal the nature of the photoinduced insulator-metal transition, we show that an exact analysis of the Falicov-Kimball model subject to external ac electric fields becomes possible with Floquet’s method combined with the nonequilibrium dynamical mean-field theory. The nonequilibrium steady state that appears during irradiation of a pump light is shown to be determined if the dissipation in a certain heat-bath model is introduced. This has enabled us to predict that novel features characteristic of the photoexcited steady states, i.e., negative weight (gain) in the low-energy region and dip structures around the photon energy of the pump light, should be observed in the optical conductivity. Special emphasis is put on the role of dissipation, for which we elaborate the dependence of the steady state on the strength of dissipation and the temperature of the heat bath.

1. Introduction
Photoinduced insulator-metal transitions are interesting, because they are nonequilibrium transitions that can be controlled by photons. This is heightened by recent advances in the pump-probe spectroscopy that has made studies of the dynamics of correlated electron systems in the femtosecond time scale a reality [1, 2, 3]. The experiments show that irradiation of a pump light on a strongly correlated insulator generates a Drude-like weight in a time-resolved optical conductivity spectrum \( \sigma(\nu) \) (\( \nu \): the frequency of the probe light), which implies that the created photo-carriers turn the system into a metallic state. As opposed the conventional chemical doping, the “photo-doping” is envisaged to induce new excited states with novel physical properties.

Here we focus on photoinduced nonequilibrium steady state (NESS) that emerge when the balance between pumping and relaxation is achieved during irradiation of the pump light. Identifying NESS is indeed a long-standing problem in nonequilibrium statistical mechanics, where one important issue is: how NESS appearing in a system isolated from other environments differs from the one in a system having extrinsic dissipations. In real experiments, there are many sources of extrinsic dissipation that can be relevant to the dynamics of the electrons, but how they affect NESS is yet to be fully clarified.

Since both of the nonequilibrium and electron correlation, notoriously difficult ingredients, are involved, an exact analysis will be valuable. Here we demonstrate an exact analysis is in fact possible for the dissipative Falicov-Kimball model driven by an ac electric field [4]. The model is solved with Floquet method [5] combined with the dynamical mean-field theory [6, 7]
(Floquet + DMFT [8, 9]) to understand the nature of photoinduced NESS and reveal the role of dissipation in the system. Part of the results given here (Fig. 1) has already been published in Ref. [4].

2. Falicov-Kimball model with dissipation in intense laser fields

We start from the Falicov-Kimball (FK) model [10, 7], which is a correlated electron model known, in DMFT, to exhibit a metal-insulator transition as the strength of the interaction $U$ is increased. The model has a simple optical-excitation structure (with $\sigma(\nu)$ having a single charge-transfer (CT) peak around $\nu = \Omega_{CT} \sim U$), which is why we have adopted the model as a benchmark. To include the effect of extrinsic dissipation, we employ a solvable “Büttiker probe” reservoir [11]. The total Hamiltonian then reads $H_{tot} = H_{sys} + H_{mix} + H_{bath}$ with $H_{sys} = \sum_k c_k - eA(t) \hat{c}_k$ and $H_{bath} = \sum_i \sum_p \varepsilon_p b_{i,p}^\dagger b_{i,p} + \text{h.c.} + \sum_i \sum_p (V_p^2/\varepsilon_p) c_i$, and $H_{mix} = \sum_i \sum_p \varepsilon_p b_{i,p}^\dagger b_{i,p}$, where $c_i$, $f_i$, and $b_{i,p}$ create, respectively, an itinerant electron, a localized electron, and bath’s fermionic degrees of freedom at site $i$, $\{k\}$, the band dispersion, $A(t)$ the vector potential of the ac field, and $\varepsilon_p$ and $V_p$ the kinetic energy and the coupling to the mode $p$ of the bath, respectively. In $H_{mix}$ we have included a counter term that cancels a potential shift due to the coupling to the bath. For the DMFT study, we take the standard hypercubic lattice $\{k\} = -t^* \sum_{i=1}^d \cos k_i/\sqrt{d}$ in $d (\to \infty \text{ in DMFT})$ dimensions ($t^*$: the unit of energy), and assume a field direction $A(t) = A(t)(1,1, \ldots, 1)$ with $A(t) = -(E/\Omega) \sin \Omega t$ ($E, \Omega$: the amplitude and frequency of the pump light, respectively).

The bath is assumed to be in equilibrium with temperature $T$, and its chemical potential is determined so that the current does not flow between the bath and the system. Let us define a hybridization function $\Gamma(\omega) = \sum_p \pi V_p^2 \delta(\omega - \varepsilon_p)$, and ignore the usually unimportant $\omega$-dependence of $\Gamma$. Then the extrinsic dissipation is characterized by $T$, and $\Gamma^{-1}$ which can be interpreted as a relaxation time.

3. Floquet + DMFT

To determine NESS in the photoexcited system, we put two assumptions: (i) NESS exists, and (ii) NESS should not depend on the initial condition and correlations between the initial state and NESS (“initial correlations”), since we expect that these effects will be wiped out due to the dissipation if we wait for long enough ($\sim \Gamma^{-1}$) for the system to relax to NESS.

These assumptions allow us to use the Keldysh Green’s function formalism for nonequilibrium systems. Since in an ac electric field NESS is periodic in time, we can further employ Floquet’s method [5], which is based on Floquet’s theorem, a temporal analog of Bloch’s theorem for spatially periodic potentials. One consequence of the theorem is that one can map the time-dependent problem into a time-independent problem with internal degrees of freedom, $n$, which labels the Floquet subbands. Accordingly, each Green’s function turns into a matrix $G(\omega) = (G_{mn}(\omega))$ (Floquet matrix) [8].

After integrating out bath’s degrees of freedom, we derive the Dyson equation in the Floquet matrix form,

$$
\begin{pmatrix}
\hat{G}_k^R(\omega) & \hat{G}_k^K(\omega) \\
0 & \hat{G}_k^A(\omega)
\end{pmatrix}^{-1} =
\begin{pmatrix}
\hat{G}_k^{R0}(\omega) & \hat{G}_k^{-1K0}(\omega) \\
\hat{G}_k^{A0}(\omega) & \hat{G}_k^{A0}(\omega)
\end{pmatrix}^{-1} +
\begin{pmatrix}
i\Gamma \hat{1} & 2i \Gamma \hat{F}(\omega) \\
0 & -i\Gamma \hat{1}
\end{pmatrix} - \hat{\Sigma}^R(\omega) \hat{\Sigma}^K(\omega),
$$

where $\hat{G}$ ($\hat{G}_0^0$) is the full (noninteracting) Green’s function, $\hat{\Sigma}$ the self-energy, $\hat{1}$ the identity matrix, and $(\hat{F})_{mn}(\omega) = \delta_{mn} \tanh[(\omega + n\Omega)/2T]$. We can neglect the Keldysh component $(\hat{G}^{-1})^{K0}$ due to the absence of initial correlation. Similarly we can rewrite all the other self-consistent equations in nonequilibrium DMFT in Floquet matrix forms, which constructs Floquet + DMFT. We use the analytic solution for the DMFT impurity problem for the FK model [12].
4. Results and discussion

We have calculated the optical conductivity in the presence of the pump light, which is defined in such a way that the current change due to an infinitesimal probe light $\delta \mathcal{E} e^{-i\omega t}$ (which we assume to be parallel to the pump light) be $\delta j(t) = \sigma(\nu) \delta \mathcal{E} e^{-i\omega t} + \sum_{n \neq 0} (\cdots) \delta \mathcal{E} e^{-i(\nu + mf)t}$. Here we define a photoduced insulator-metal transition as a change of a low-energy spectral weight in $\sigma(\nu)$ from zero to a finite value.

Figure 1 shows the results for $\sigma(\nu)$ for two cases: (a) $\Omega = 2.7 \lesssim \Omega_{\text{CT}} \sim U(= 3.0$ here), and (b) $\Omega = 3.3 \gtrsim \Omega_{\text{CT}}$. We can first notice that the CT peak around the CT frequency $\nu \sim \Omega_{\text{CT}} \sim U$ collapses with the laser field intensity $E$, while curious dip structures emerge around $\nu \sim \Omega$ in both cases.

The low-energy behavior, on the other hand, strongly depends on $\Omega$: in Fig. 1(a), a Drude-like peak with a finite width appears in the optical gap region, whereas a negative weight increases continuously as a function of the intensity $\Omega$. We can first notice that the CT peak around the CT frequency $\nu \sim \Omega_{\text{CT}} \sim U$ collapses with the laser field intensity $E$, while curious dip structures emerge around $\nu \sim \Omega$ in both cases.

To study the role of dissipation in NESS, we illustrate the dependence of $\sigma_{\text{bubble}}(\nu)$ (Fig. 2(b)) and $\sigma_{\text{vertex}}(\nu)$ (3(b)), the temperature dependence is quite weak for sufficiently low temperatures ($\lesssim 0.1t^*$). At higher temperatures, both $\sigma_{\text{bubble}}(\nu)$ and $\sigma_{\text{vertex}}(\nu)$ decay over the whole frequency range.

By contrast, the $\Gamma$-dependence is rather intriguing. We can see that the $\Gamma$ dependence for $\sigma_{\text{bubble}}(\nu)$ (Figures 2(a)) and $\sigma_{\text{vertex}}(\nu)$ (3(a)) is quite significant even for small $\Gamma$, where the optical conductivity is amplified as $\Gamma$ approaches zero. If we look at Fig. 3(a) more closely, we can see the width of the dip shrinks and its depth grows with $\Gamma \rightarrow 0$. In the limit of $\Gamma = 0$, the Dyson equation in fact becomes ambiguous (i.e., singular) due to the lack of boundary conditions, which
Figure 2. The dependence of $\delta \sigma_{\text{bubble}}(\nu) = \sigma_{\text{bubble}}(\nu)|_{E=0.4} - \sigma(\nu)|_{E=0}$ on $\Gamma$ with a fixed $T = 0.05$(a), or on $T$ with a fixed $\Gamma = 0.05$(b) for $U = 3$, $\Omega = 2.7$. 

Figure 3. The dependence of $\sigma_{\text{vertex}}(\nu)$ on $\Gamma$ with a fixed $T = 0.05$(a), or on $T$ with a fixed $\Gamma = 0.05$(b) around the dip structure (note the horizontal scale different from the previous figures) for $U = 3.0$, $\Omega = 2.7$, $E = 0.4$.

is reflected by the $\Gamma$-dependence of $\sigma(\nu)$. Our message here is that the extrinsic dissipation can strongly affect NESS even if the coupling to the bath is small, and that the dip structure may be a key quantity that characterizes extrinsic dissipation in photoinduced insulator-metal transitions. We expect that our approach can be extended to the Hubbard model, where magnetism will be of particular interest in photoinduced phenomena.

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