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Optically driven Mott-Hubbard systems out of thermodynamic equilibrium

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We consider the Hubbard model at half filling, driven by an external, stationary laser field. This stationary, but periodic in time, electromagnetic field couples to the charge current, i. e. it induces an extra contribution to the hopping amplitude in the Hubbard Hamiltonian (photo-induced hopping). We generalize the dynamical mean-field theory (DMFT) for nonequilibrium with periodic-in-time external fields, using a Floquet mode representation and the Keldysh formalism. We calculate the non-equilibrium electron distribution function, the density of states and the optical DC conductivity in the presence of the external laser field for laser frequencies above and below the Mott-Hubbard gap. The results demonstrate that the system exhibits an insulator-metal transition as the frequency of the external field is increased and exceeds the Mott-Hubbard gap. This corresponds to photo-induced excitations into the upper Hubbard band.

1 Introduction

A major aim of optical technologies is the achievement of all-optical switching and computing devices. Mott-Hubbard insulating materials have the potential to be used for ultrafast electric switches, driven by an external laser field, because of the short relaxation times characteristic for strongly correlated systems. Recent pump-probe experiments on transition metal oxides display unusual effects, as e. g. a dramatic change in the optical conductivity or in the optical transmission [1–3]. These results suggest a strong potential of such materials for all-optical switching. The theoretical description of driven strongly correlated quantum systems is therefore highly desirable and is increasingly raising interest, e. g. [4–7].

Among the various lattice models for correlated electrons, e. g. the Falicov-Kimball model [8], the best suited approach to describe all main aspects of correlated electrons is certainly the Hubbard model [9]. An appropriate and powerful tool to analyze the Hubbard model is the dynamical mean-field theory (DMFT) [10] . The DMFT takes into account the strong local correlations of the electrons due to the Coulomb repulsion. Here we present a theory generalizing the equilibrium DMFT approach to describe non-equilibrium effects induced by periodic-in-time external fields, by adopting the Keldysh Green's function formalism [11] in conjunction with a Floquet mode expansion [12] of the electron density in terms of higher harmonics of the time-periodic field is employed. Numerical evaluations have been carried out for zero temperature, at half filling. Eventually, the theory combining those three methods is employed to study in detail the Hubbard model in the metallic as well as in the insulating regime by means of spectral functions, non-equilibrium electron distribution functions and also the DC-conductivity in the presence of an external driving laser field. As central results, we find (1) that the electronic occupation changes drastically for both large and small Hubbard U. For systems with large U density of states is generated within the energy gap

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and highly occupied. States in the upper Hubbard band are also occupied. (2) that the DC conductivity in the presence of an external field suddenly increases over several orders of magnitude as the frequency of external field is raised and the energy $\hbar\Omega_L$ becomes equal to width of the Mott Hubbard gap.

2 Model

As our starting point we choose a Hubbard model with nearestneighbor hopping, characterized by the hopping amplitude t and on-site repulsion U. Additionally, the vector potential $\vec{A(\tau)}$ of the external (laser) field couples to the current density of this correlated system. In the following we assume this periodic-in-time field to be independent of spatial coordinates. Hence, the Hamiltonian including the external field reads,

$$H = -\sum_{\langle ij\rangle,\sigma} t\; c^\dagger_{i,\sigma} c_{j,\sigma} + \frac{U}{2}\sum_{i,\sigma} c^\dagger_{i,\sigma} c_{i,\sigma} c^\dagger_{i,-\sigma} c_{i,-\sigma} + H_A(\tau) \tag{1}$$

where the first two terms constitute the bare Hubbard Hamiltonian, i. e. kinetic hopping and on-site Coulomb repulsion, whereas the last term represents the coupling to the external field via the elementary charge e,

$$H_A(\tau) = e\sum_{\vec{x}} \vec{j}(\vec{x})\cdot\vec{\mathcal{A}}(\tau) \tag{2}$$

where charge current in the system $j(\vec{x})$ is given by

$$j(\vec{x})_{\hat{a}} = \frac{t}{i}\sum_{\sigma}\left(c^\dagger_{\vec{x},\sigma} c_{\vec{x}-\hat{a},\sigma} - c^\dagger_{\vec{x}-\hat{a},\sigma} c_{\vec{x},\sigma}\right) \tag{3}$$

and $\hat{a} = \hat{x}, \hat{y}, \hat{z}$ characterizes spatial directions.

Due to the time dependence of the external field Green's functions truly depend on two separate time arguments. Therefore we use a double Fourier transformation similar to [13] from time to frequency space

$$\begin{aligned} G^{\alpha\beta}_{mn}(\omega) &= \int \mathrm{d}\tau_1^\alpha \int \mathrm{d}\tau_2^\beta e^{-i\Omega(m\tau_1^\alpha - n\tau_2^\beta)} e^{i\omega(\tau_1^\alpha - \tau_2^\beta)} G(\tau_1^\alpha, \tau_2^\beta) \\ &\equiv G^{\alpha\beta}(\omega - m\Omega, \omega - n\Omega), \end{aligned} \tag{4}$$

where (m, n) label the Floquet modes [12] and (α, β) specify on which branch of the Keldysh contour $(\pm)$ the respective time argument resides. A physical interpretation of such a Green's function $G^{\alpha\beta}_{mn}(\omega)$ is demonstrated in Fig. 1 (right panel).

The DMFT provides a mathematical scheme for mapping the originally lattice model onto an effective impurity model. In the limit of infinite dimensions (connectivity of the lattice) it was shown [10, 14] that the self-energy acquires a local form. In finite dimensions the local self-energy approach provides therefore a controlled approximation, which is widely used. This effective local impurity problem may then be characterized by the Green's function for effective Weiß-field (the "bath") $[\mathcal{G}(\omega)]^{\alpha\beta}_{mn}$. For the Hamiltonian, Eq. (1), a DMFT self-consistency scheme may be set up by following the general ideas outlined in [10]. Because of the periodicity in time and because of the non-equilibrium, the resulting self-consistency equation has now become a matrix equation in both Floquet and in Keldysh space and reads

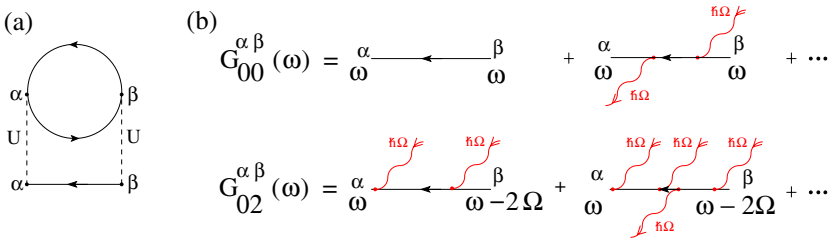

Fig. 1 (online colour at: www.ann-phys.org) The left panel (a) shows the second order in U diagram used within the IPT method to calculate the self-energy $\Sigma^{\alpha\,\beta}(\tau_1^{\alpha}, \tau_2^{\beta})$. Greek superscripts label the Keldysh contour, $\alpha = \pm$. Solid lines correspond to Weiß-field Green's functions, and the dashed line represents the local Coulomb interaction U. In panel (b) a physical picture in terms of absorption/emission of external energy quanta $\hbar\Omega$ of the Floquet mode Green's function, Eq. (4), is presented. $G_{00}^{\alpha\,\beta}(\omega)$, α, β being Keldysh labels, e. g. represents the sum of all balanced processes.

$$
\begin{aligned}
\left[\mathcal{G}^{-1}(\omega)\right]^{\alpha\beta}_{mn} = \left[g_0^{-1}(\omega)\right]^{\alpha\beta}_{mn} - \alpha\beta\ t\ [G(\omega)]^{\alpha\beta}_{mn}\ \ t \\
+ \alpha\beta\,\frac{T_0}{2}\ \left([G(\omega)]^{\alpha\beta}_{m-1,n} + [G(\omega)]^{\alpha\beta}_{m+1,n}\right)\ \ t \\
- \alpha\beta\ \ t\ \ \left([G(\omega)]^{\alpha\beta}_{m,n+1} + [G(\omega)]^{\alpha\beta}_{m,n-1}\right)\ \frac{T_0}{2} \\
+ \alpha\beta\,\frac{T_0}{2}\ \left(\ [G(\omega)]^{\alpha\beta}_{m+1,n+1} + [G(\omega)]^{\alpha\beta}_{m-1,n-1} + [G(\omega)]^{\alpha\beta}_{m+1,n-1} + [G(\omega)]^{\alpha\beta}_{m-1,n+1}\right)\ \frac{T_0}{2}
\end{aligned}
\tag{5}
$$

with t the bare hopping, T_0 the photo-induced hopping amplitude, (m, n) labeling the Floquet modes and (α, β) the Keldysh contour. In order to obtain Eq. (5), a semi-circular density of states has been assumed. The first line in Eq. (5) represents the physics of the bare Hubbard model, the second and third line describe how the electron enters or leaves the impurity site by one photo-assisted hopping event, whereas the last line is the fully photo-induced process.

In order to solve this matrix DMFT equation, we generalize the so-called iterated perturbation theory (IPT) method [14–16]. As a diagrammatic approach this method can be extended to nonequilibrium situations in a rather straightforward way and is known to provide qualitatively correct results.

3 Results and discussion

As a result of the above outlined concept, we obtain the various quantities characterizing this strongly correlated system out of equilibrium, such as e. g. electron distribution function, density of states, DC-conductivity. Among which we have chosen to discuss here as an example the density of states for a metallic ground state of the correlated system ($U/D = 2.5$ where D is the half bandwidth). The imaginary part of the total (advanced) Green's function $\mathrm{Im}\ G_{tot}(\omega) = \mathrm{Im}\ \sum_n G_{0n}(\omega)$ is displayed in Fig. 2 as a function of quasiparticle energy ω (x-axis) and of external laser frequency Ω_L along the y-axis. For $\Omega_L = 0$ the typical three-peak structure is visible ($\Omega_L = 0$ represents the equilibrium solution), which changes with increasing laser frequency, because the photo-induced excitations in the system (side-bands) evolve and are clearly observable, see Fig. 2. Several important features can be observed in this density of states.

At first, starting at the point (ω, Ω_L) = (0,0), the central many-body resonance evolves two (side-) features with increasing laser frequency. This transfer of spectral weight is caused by absorption/emission of field quanta $\hbar\Omega_L$ from/into the external field. The slope of $1/2$ as seen in Fig. 2 reflects the fact that only processes are allowed which involve an even number of field quanta. Therefore the lowest order process

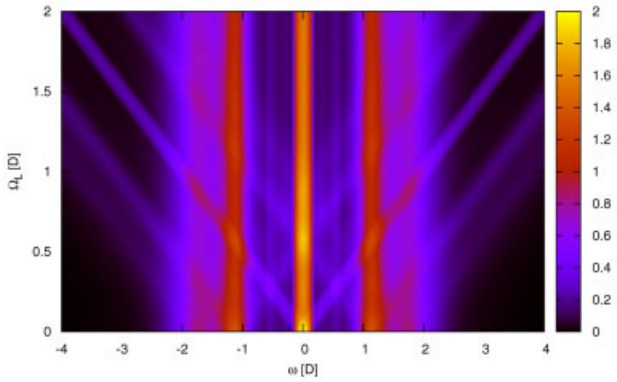

Fig. 2 (online colour at: www.ann-phys.org) Imaginary part of the Green's function as function of quasiparticle energy ω and of external laser frequency Ω_L. For a discussion see text.

is the emission/absorption of two field quanta. This effect can be shown to originate in the point inversion symmetry of the crystal we used.

The analogous behavior is found for the (preformed) Hubbard bands. Both peaks in the density of states evolve such side-bands on their own. Second and higher order processes (involving 4, 6 etc. photons) are not observed here, which is due to the small magnitude A of the external field.

In general, the external optical pumping will transfer spectral weight to $\omega = \pm 2\Omega_L, \pm 4\Omega_L, \ldots$ as explained above. This leads immediately to a decrease of the central many-body resonance, as can also seen in Fig. 2. However, as the increasing external frequency becomes large enough to excite into the (preformed) upper Hubbard band, the central resonance is (partially) re-established. This is observed at Ω_L/D 0.5 in Fig. 2. This represents the situation where the first side-band of the central resonance crosses the Hubbard bands and equally the first side bands of the Hubbard bands cross the central resonance. Further (Floquet) side bands, or bandstructure effects give rise to further collapse and revival events in the many-body resonance.

4 Conclusion

In conclusion, we studied an external field, being periodic in time and otherwise stationary, coupled to the charge current of a system of correlated electrons. We combined the DMFT approach with the Keldysh technique and a Floquet mode expansion to obtain, at zero temperature, a numerical solution of the Hubbard model in terms of the nonequilibrium electron distribution function and the density of states as well as the DC-conductivity. We discussed here in particular the metallic regime and found a collapse and revival effect in the central many-body resonance with increasing external frequency.

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