Nonequilibrium dynamical mean field theory

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Ultrafast pump-probe spectroscopy

• Time resolution: ~10 fs \rightarrow measures excitation and relaxation processes on the intrinsic timescale of the electrons



- Pump pulse drives system out of equilibrium
- Time evolution measured by subsequent probe pulses
- "Disentangles" competing or cooperative effects on the time-axis

"Tuning" of material properties by external driving

Ultra-fast insulator-metal transition ("photo-doping")

Iwai et al. (2003)





"Tuning" of material properties by external driving

• Create long-lived transient states with novel properties e.g. light-induced high-temperature superconductivity Fausti et al. (2010), Kaiser et al. (2013)

temperature Mott insulator metal stripe order superconductor hole doping



THz pulse couples to phonons

"Tuning" of material properties by external driving

Create long-lived transient states with novel properties
 e. g. light-induced high-temperature superconductivity
 Mitrano et al. (2015)



equilibrium

driven phonons



"Tuning" of material properties by external driving

Switching into metastable, but long-lived "hidden states"
 e. g. Reversible switching of TaS₂ into / out of a metallic hidden state
 Stojchevska et al. (2015)





hidden state (conducting)

Challenge for theory/numerics



- Strongly interacting many-particle system
- Strong perturbations
- Different relevant time scales



- Dynamical mean field theory
- Nonequilibrium extension
- Physical observables
- Illustrations:
 - AC field quench tuning of the interaction strength by external driving
 - Nonequilibrium phase transition nonthermal fixed points

• Static mean field theory: mapping to a single-site problem Weiss (1903)



- Effective model: yields local observables (magnetization)
- Parameter of the effective model ("mean field"): optimized by requesting consistency between the lattice and single-site model

 Dynamical mean field theory DMFT: mapping to an impurity problem Georges & Kotliar (1992)



Impurity solver: computes the Green's function of the correlated site

Bath parameters = "mean field": optimized in such a way that the bath mimics the lattice environment

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• Single-site DMFT can treat two-sublattice order (e.g. AFM)

 $\operatorname{Bath}_{B,\sigma}[G_{A,\sigma}], \quad \operatorname{Bath}_{A,\sigma}[G_{B,\sigma}]$

• Pure Neel order: $\operatorname{Bath}_{B,\sigma} = \operatorname{Bath}_{A,\bar{\sigma}} \longrightarrow \operatorname{Bath}_{A,\bar{\sigma}}[G_{A,\sigma}]$

- Equilibrium DMFT phase diagram (half-filling)
- Paramagnetic calculation: Metal Mott insulator transition at low T
- Smooth crossover at high T



- Equilibrium DMFT phase diagram (half-filling)
- With 2-sublattice order: Antiferromagnetic insulator at low T
- Smooth crossover at high T



- Equilibrium DMFT phase diagram (half-filling)
- Transformation $c_{i\uparrow} \to c_{i\uparrow}^{\dagger}$ $(i \in A), c_{i\uparrow} \to -c_{i\uparrow}^{\dagger}$ $(i \in B)$

maps repulsive model onto attractive model



- Equilibrium DMFT phase diagram
- Half-filling: transformation $c_{i\uparrow} \rightarrow c_{i\uparrow}^{\dagger}$ $(i \in A)$, $c_{i\uparrow} \rightarrow -c_{i\uparrow}^{\dagger}$ $(i \in B)$ maps repulsive model onto attractive model



- Kadanoff-Baym contour
- Initial state described by the density matrix $\rho(0) = \frac{1}{Z}e^{-\beta H(0)}$
- State at time t described by $\rho(t) = \frac{U(t,0)}{\rho(0)} \rho(0) U(0,t)$

$$U(t,t') = \begin{cases} \mathcal{T} \exp\left(-i \int_{t'}^{t} d\bar{t} H(\bar{t})\right) & t > t' \\ \tilde{\mathcal{T}} \exp\left(-i \int_{t'}^{t} d\bar{t} H(\bar{t})\right) & t < t' \end{cases}$$

ullet Time dependent expectation value of observable ${\cal O}$

$$\langle \mathcal{O}(t) \rangle = \operatorname{Tr}\left[\rho(t)\mathcal{O}\right] = \operatorname{Tr}\left[\frac{U(t,0)}{\rho(0)U(0,t)}\mathcal{O}\right]$$

Kadanoff-Baym contour

• Express ho(0) as time-propagation along an imaginary time branch

 $\langle \mathcal{O} \rangle(t) = \operatorname{Tr} \left[\frac{1}{Z} e^{-\beta H(0)} U(0,t) \mathcal{O} U(t,0) \right]$ $= \operatorname{Tr}\left[\frac{1}{Z} \left(\mathcal{T}_{\tau} e^{-\int_{0}^{\beta} d\tau H(\tau)}\right) \left(\tilde{\mathcal{T}} e^{i\int_{0}^{t} ds H(s)}\right) \mathcal{O}\left(\mathcal{T} e^{-i\int_{0}^{t} ds H(s)}\right)\right]$ $\mathbf{0}$ inverse temperature

- Kadanoff-Baym contour
- Define contour ordering $\mathcal{T}_{\mathcal{C}}$ on the contour $\mathcal{C}: 0 \to t \to 0 \to -i\beta$

$$\langle \mathcal{O}(t) \rangle = \frac{1}{Z} \operatorname{Tr} \left[\mathcal{T}_{\mathcal{C}} e^{-i \int_{\mathcal{C}} ds \, H(s)} \mathcal{O}(t) \right]$$



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Contour-ordered formalism can also be applied to 2-point functions

$$\langle \mathcal{T}_{\mathcal{C}} \mathcal{A}(t) \mathcal{B}(t') \rangle \equiv \frac{1}{Z} \operatorname{Tr} \left[\mathcal{T}_{\mathcal{C}} e^{-i \int_{\mathcal{C}} ds \, H(s)} \mathcal{A}(t) \mathcal{B}(t') \right]$$

Particularly relevant: Green's function

$$G(t,t') \equiv -i \langle \mathcal{T}_{\mathcal{C}} d(t) d^{\dagger}(t') \rangle$$

- Kadanoff-Baym contour
- Due to the 3 branches, the Green's function has 9 components

$$G(t,t') \equiv G_{ij}(t,t'), \quad t \in \mathcal{C}_i, t' \in \mathcal{C}_j, \quad i,j = 1,2,3$$



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- Kadanoff-Baym contour
- Boundary conditions (cyclic invariance of the trace)

$$G(0_+, t') = -G(-i\beta, t')$$



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- Boundary conditions (cyclic invariance of the trace)

$$G(0_+, t') = -G(-i\beta, t')$$
$$G(t, 0_+) = -G(t, -i\beta)$$



• Nonequilibrium DMFT: Solve DMFT equations on the Kadanoff-Baym contour \mathcal{C} Freericks et al. (2006)



- \bullet Nonequilibrium DMFT: Solve DMFT equations on the Kadanoff-Baym contour ${\cal C}$
- Nonequilibrium Anderson impurity model

$$S_{\rm imp} = -i \int_{\mathcal{C}} dt \, H_{\rm loc}(t) - i \sum_{\sigma} \int_{\mathcal{C}} dt \, dt' \, d_{\sigma}^{\dagger}(t) \Delta(t, t') d_{\sigma}(t')$$

$$\bigwedge$$

interaction and chemical potential terms

contour hybridization function

Impurity Green's function

$$G_{\rm imp}(t,t') = -i \langle \mathcal{T}_{\mathcal{C}} d(t) d^{\dagger}(t') \rangle_{S_{\rm imp}}$$

$$\langle \cdots \rangle_{S_{imp}} = \frac{\operatorname{Tr}[\mathcal{T}_{\mathcal{C}} \exp(S_{imp}) \cdots]}{\operatorname{Tr}[\mathcal{T}_{\mathcal{C}} \exp(S_{imp})]}$$

- Calculation of the lattice Green's function
- Noninteracting lattice:

 $H_0(t) = \sum_k [\epsilon_k(t) - \mu(t)] d_k^{\dagger} d_k$ $G_{0,k}(t,t') = -i \langle \mathcal{T}_{\mathcal{C}} d_k(t) d_k^{\dagger}(t') \rangle_0$

• Green's function satisfies:

$$\begin{bmatrix} i\partial_t + \mu(t) - \epsilon_k(t) \end{bmatrix} G_{0,k}(t,t') = \delta_{\mathcal{C}}(t,t')$$

$$G_{0,k}(t,t') \begin{bmatrix} -i\overleftarrow{\partial_{t'}} + \mu(t') - \epsilon_k(t') \end{bmatrix} = \delta_{\mathcal{C}}(t,t')$$

Inverse lattice Green's function:

$$G_{0,k}^{-1}(t,t') = \left[i\partial_t + \mu(t) - \epsilon_k(t)\right]\delta_{\mathcal{C}}(t,t')$$

Calculation of the lattice Green's function

• Interacting lattice Green's function satisfies Dyson equation:

$$G_k(i\omega_n) = \frac{1}{G_{0,k}^{-1}(i\omega_n) - \Sigma(i\omega_n)} = \frac{1}{i\omega_n + \mu(0) - \epsilon_k(0) - \Sigma(i\omega_n)}$$

Usual equilibrium DMFT calculation for the initial equilibrium state





Real-time branches: initial-value problem

$$[i\partial_t + \mu(t) - \epsilon_k(t)]G_k(t, t') - \int_{\mathcal{C}} d\bar{t} \Sigma(t, \bar{t})G_k(\bar{t}, t') = \delta_{\mathcal{C}}(t, t')$$

 Defines time-propagation scheme for G in which the self-energy plays the role of a memory-kernel

- Electric fields
- Vector potential A(r,t), scalar potential $\Phi(r,t)$: $E = -\nabla \Phi \partial_t A$

$$\begin{aligned} v_{ij}(t) &= v_{ij} \exp\left(-ie \int_{R_i}^{R_j} dr A(r, t)\right) \\ & \swarrow \\ H(t) &= -\sum_{\langle ij \rangle \sigma} v_{ij}(t) \left(d_{i\sigma}^{\dagger} d_{j\sigma} + d_{j\sigma}^{\dagger} d_{i\sigma}\right) \\ &+ U(t) \sum_i n_{i\uparrow} n_{i\downarrow} - \sum_{i\sigma} \mu_i(t) n_{i\sigma} \\ & \swarrow \\ \mu_i(t) &= \mu + e \Phi(t)(R_i, t) \end{aligned}$$

Convenient choice: gauge with pure vector potential:

$$\Phi \equiv 0, E = -\partial_t A$$

• Electric fields

 Neglecting the r-dependence of A (assumption: field varies slowly on the atomic scale):

 Electric field enters in the lattice Dyson equation in the form of a time-dependent dispersion:

$$[i\partial_t + \mu(t) - \epsilon_k(t)]G_k(t, t') - \int_{\mathcal{C}} d\bar{t} \,\Sigma(t, \bar{t})G_k(\bar{t}, t') = \delta_{\mathcal{C}}(t, t')$$

- "Physical" Green's functions
- The 9 elements of the 3x3 Green's function matrix

$$\hat{G} = \begin{pmatrix} G_{11} & G_{12} & G_{13} \\ G_{21} & G_{22} & G_{23} \\ G_{31} & G_{32} & G_{33} \end{pmatrix}$$

are not independent:



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are not independent:



- "Physical" Green's functions
- We have the following redundancies

$$G_{11}(t,t') = G_{12}(t,t') \quad (\text{for } t \le t')$$

$$G_{11}(t,t') = G_{21}(t,t') \quad (\text{for } t > t')$$

$$G_{22}(t,t') = G_{21}(t,t') \quad (\text{for } t < t')$$

$$G_{22}(t,t') = G_{12}(t,t') \quad (\text{for } t \ge t')$$

$$G_{13}(t,\tau') = G_{23}(t,\tau')$$

$$G_{31}(\tau,t') = G_{32}(\tau,t')$$

which allow to eliminate 3 of the 9 components
 define 6 "physical" Green's functions

 G^R, G^A, G^K, \ldots

- "Physical" Green's functions
- Relevant for the following discussion: Retarded Green's function

$$G^{R}(t,t') = \frac{1}{2}(G_{11} - G_{12} + G_{21} - G_{22}) = -i\theta(t-t')\langle \{d(t), d^{\dagger}(t')\}\rangle$$

and lesser Green's functions

 $G^{<}(t,t') = G_{12} = i \langle d^{\dagger}(t')d(t) \rangle$

- In equilibrium:
 - Spectral function: $A(\omega) = -\frac{1}{\pi} \text{Im} \, G^R(\omega)$
 - Occupation: $N(\omega) = \frac{1}{2\pi} \operatorname{Im} G^{<}(\omega)$
 - Distribution function: $N(\omega)/A(\omega) = f(\omega)$ Fermi function

- "Physical" Green's functions
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- Out of equilibrium: $t_{\rm av} = (t + t')/2, t_{\rm rel} = t t'$
 - Spectral function: $A(\omega, t_{\rm av}) = -\frac{1}{\pi} \text{Im} \int dt_{\rm rel} e^{i\omega t_{\rm rel}} G^R(t, t')$
 - Occupation: $N(\omega, t_{\rm av}) = \frac{1}{2\pi} \text{Im} \int dt_{\rm rel} e^{i\omega t_{\rm rel}} G^{<}(t, t')$
 - "Distribution function": $N(\omega, t_{\rm av})/A(\omega, t_{\rm av})$

Time-resolved photoemission spectrum



$$I(k_f, E; t_p) \propto \sum_k \delta_{k_{\parallel} + q_{\parallel}, k_{f\parallel}} I_k(E - \hbar\Omega_q - W; t_p),$$

$$I_k(\omega; t_p) = -i \int dt dt' S(t) S(t') e^{i\omega(t'-t)} G_k^{<}(t + t_p, t' + t_p)$$

$$\bigoplus$$

probe time

Time-resolved photoemission spectrum



 $S(t) \sim \delta(t - t_p): \text{ measure occupation } n_k(t_p)$ $S(t) \sim \text{const}: \text{ measure spectral function } A_k(\omega, t_p)$ $I_k(\omega; t_p) = -i \int dt dt' S(t) S(t') e^{i\omega(t'-t)} G_k^{<}(t + t_p, t' + t_p)$

Formula contains time-energy uncertainty

I. Periodic electric fields

• AC-field quench in the Hubbard model (metal phase)

Tsuji, Oka, Werner & Aoki, PRL 106, 236401 (2011)



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I. Periodic electric fields

- AC-field quench in the Hubbard model (metal phase)
 - Sign inversion of the interaction: repulsive +> attractive
 - Dynamically generated high-Tc superconductivity?



Periodic E-field leads to a population inversion



Gauge with pure vector potential

$$E(t) = E\cos(\Omega t) = -\partial_t A(t)$$

$$\Rightarrow A(t) = -(E/\Omega)\sin(\Omega t)$$

- Peierls substitution $\epsilon_k \rightarrow \epsilon_{k-A(t)}$
- Renormalized dispersion

$$\overline{\epsilon_k} = \frac{\Omega}{2\pi} \int_0^{2\pi/\Omega} dt \epsilon_{k-A(t)} = \mathcal{J}_0(E/\Omega)\epsilon_k$$



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• Renormalized dispersion

$$\overline{\epsilon_k} = \frac{\Omega}{2\pi} \int_0^{2\pi/\Omega} dt \epsilon_{k-A(t)} = \mathcal{J}_0(E/\Omega)\epsilon_k$$

- Inverted population = negative temperature
- State with U > 0, T < 0 is equivalent to state with U < 0, T > 0

$$\tilde{T} < 0, \mathcal{J}_0 < 0 \qquad \rho \propto \exp\left(-\frac{1}{\tilde{T}}\left[\sum_{k\sigma} \mathcal{J}_0 \epsilon_k n_{k\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}\right]\right)$$
$$T_{\text{eff}} = \frac{\tilde{T}}{\mathcal{J}_0} > 0 \qquad = \exp\left(-\frac{1}{T_{\text{eff}}}\left[\sum_{k\sigma} \epsilon_k n_{k\sigma} + \frac{U}{\mathcal{J}_0} \sum_i n_{i\uparrow} n_{i\downarrow}\right]\right)$$

 \bullet Effective interaction of the $\,T_{\rm eff}>0\,$ state

$$U_{\rm eff} = \frac{U}{\mathcal{J}_0(E/\Omega)}$$

I. Effect on superconductivity

• AC-field quench from U = 1 to $U_{eff} = -2.5$ (NCA solver)



- Equilibrium DMFT phase diagram (half-filling)
- Half-filling: transformation $c_{i\uparrow} \rightarrow c_{i\uparrow}^{\dagger}$ $(i \in A), c_{i\uparrow} \rightarrow -c_{i\uparrow}^{\dagger}$ $(i \in B)$ maps repulsive model onto attractive model



• Weak-coupling regime

Tsuji, Eckstein & Werner, PRL 110, 136404 (2013)

Slow ramp from (Slater-)Antiferromagnet to Paramagnet



Weak-coupling regime

Tsuji, Eckstein & Werner, PRL 110, 136404 (2013)

• Time-evolution of the magnetization for different final U



• Weak-coupling regime

Tsuji, Eckstein & Werner, PRL 110, 136404 (2013)

• Time-evolution of the magnetization for different final U (Hartree)



Weak-coupling regime

Tsuji, Eckstein & Werner, PRL 110, 136404 (2013)

• Time-evolution of the magnetization for different final U



Weak-coupling regime

Tsuji, Eckstein & Werner, PRL 110, 136404 (2013)



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Summary

• Two examples



I. Floquet state

Dynamical band flipping and repulsion-toattraction conversion

II. Dynamical phase transition

Trapped antiferromagnetic order and nonthermal "Hartree" fixed point

References

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