A. J. Millis

The Simons Foundation
Department of Physics, Columbia University

Workshop on Interacting Quantum Systems Driven Out of Equilibrium
Rice University, 2016

Support: NSF DMR 1306282
DOE SC-00123175 and SC-0012592

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Collaborators

- David Reichman (CU), Emanuel Gull (Michigan), Guy Cohen (Tel-Aviv)
- Zhuoran He, Eli Wilner, Dante Kennes (CU)
- Rainer Hartle (Gottingen)
Topics

• Nonequilibrium ``phases'' (Z. He)

• Numerical methods: (G. Cohen)
VO$_2$: high T metal, low T insulator.
Low T ‘M1’ phase: dimerized and monoclinic
a little bit of theory:

Vanadium on-site interaction

\[ H_{\text{int}} = U \sum_a n_{a,\uparrow} n_{a,\downarrow} + U' \sum_{a \neq b} n_{a,\uparrow} n_{b,\downarrow} + U'' \sum_{a > b, \sigma} n_{a\sigma} n_{b\sigma} + \ldots \]

Occupancy of one orbital pushes others up

Note: these terms conserve orbital. smaller (J) terms mix
Dimerization alone does not give insulator

see also Brito et al arXiv: 1509.02968

Biermann et al
PRL 94, 026404 (2005)
M1 phase of VO2: the electronic structure (Z. He, Hartree-Fock)

Dimerization enhanced by intersite interactions (Fock term)
M1 phase of VO2: the electronic structure (Z. He, Hartree-Fock)

Level splitting from on-site interactions

xz, yz pushed up by occupancy of \( x^2-y^2 \)
High degree of excitation

If the gap is of many-body origin, exciting carriers might lead to a new electronic state.
Apply $\sim 1.5\text{eV}$ pulse.
Measure time-resolved diffraction

Red (e.g. 302): peaks present only in M1 phase

Blue (e.g. 220): peaks present in both phases
Experiment
Morrison et al Science 346 445

After excitation: things change

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Experiment

fluence dependence.

- 220 (in both structures) linear in fluence
- 302 peak (labels M1): nonlinear fluence dependence not affected if fluence < 9 mJ/cm$^2$
Experiment

fluence dependence.

- 220 (in both structures) linear in fluence
- 302 peak (labels M1): nonlinear fluence dependence not affected if fluence < 9 mJ/cm²

Focus on low fluence range
Interpretation of low fluence regime: Lattice unchanged (302 stays) => change in 220 reflects change in electron distribution.
Physical properties of low fluence regime
Infra-Red transmission

Transparent (T high) => insulator
(gap means no absorption)

Non transparent => metal (all light absorbed)
Infra-Red transmission

Transparent (T high) => insulator (gap means no absorption)

Non transparent => metal (all light absorbed)

Very low fluence: insulating state recovered after brief transient

fluence > 3 mJ/cm²: long-lived metallic state (t > 20 ps)
Morrison et al interpretation of metallic phase

IR pulse $\Rightarrow$ drives long-lived orbital rearrangement without change in lattice
Experimental Summary

Morrison et al Science 346 445

For fluence above a modest threshold, lattice unchanged but new electronic properties:
long-lived M1 metal phase
Many questions

- How long does state "live"
- Why doesn’t lattice move
- How rapidly does the new state relax back to the equilibrium configuration

BUT

Key point: new phase (metallic M1) induced by excitation and not observed in equilibrium??

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If the gap is of many-body origin, excitation of carriers can lead to a new electronic state.
Theory: Z. He

- Evolution after initial excitation
  - Local boltzmann-type equation
  - Time evolution to hot electron dist.
- Possibility of new nonequilibrium phase
  - Hartree-Fock theory
- Questions
  - Why doesn't the lattice evolve
  - How much excitation needed to drive new state
Immediately after initial excitation

**DOS**

**Distribution of photoexcited electrons and holes (JDOS)**

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Initial Evolution of Photoexcited Holes

Boltzmann kinetic equation.
Hartree-Fock Band Structure
Local U, J interactions

Recall Slater-Kanamori Interactions

\[
H = U \sum_a n_{a\downarrow} n_{a\downarrow} + (U - 2J) \sum_{a>b, \sigma=1, \downarrow} n_{a\sigma} n_{b\sigma} \\
+ (U - 3J) \sum_{a\neq b\sigma} n_{a\sigma} n_{b\sigma} - J \sum_{a\neq b} c_{a\downarrow}^\dagger c_{a\downarrow}^\dagger c_{b\uparrow} c_{b\uparrow} + c_{a\downarrow}^\dagger c_{b\downarrow}^\dagger c_{b\uparrow} c_{a\uparrow}
\]

U interactions conserve orbital index
Non-conservation of orbital index: pair hopping \(\sim J\) and intersite hybridization

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Initial relaxation: fast (fs) thermalization

U-terms allow for exchange of energy between electrons and holes => rapid thermalization (number of electrons and holes remains constant). $T$ determined by energy put in by pump.
Auger up-scattering: from J terms and band effects create a particle-hole pair: pay for energy by down-scattering from thermal tail. Slow kinetics but factor of $\sim$ increase in number of excitations
Population inversion: M1 metal phase

Level splitting from on-site interactions

xz, yz pushed up by occupancy of $x^2-y^2$

Excitation can lead to inversion of levels

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T=0 Hartree Fock theory for large U: two minima

Question: can the optical pulse invert the population enough to put the system into the other minimum
Equilibrium Hartree Fock: Phase transition as function of electron temperature
Recast as function of energy transferred by pump
Recast as function of energy transferred by pump

Energy transferred in experiment
What we learned

- 100fs initial relaxation to pseudoequilibrium electronic state
- Long-lived M1 metal for fluence above critical value
- Related to metastable HF extremum but theoretically needed fluence too high
- Related to theoretically existing high T phase

Questions:
- Time for lattice motion
- How large a population inversion is required
- How long does the M1 metal live?
- Physics beyond Hartree-Fock
Beyond Hartree Fock: Dynamical Mean Field Theory

The quantum many-body problem is NP-hard

Path forward: obtain a restricted (approximate) class of information by solving a simpler model.

ex: Density functional band theory: ground state energy and charge density from solution of 1-particle Schroedinger equation + self consistency condition
Beyond Hartree Fock: 
Dynamical Mean Field Theory

The quantum many-body problem is NP-hard

Path forward: obtain a restricted (approximate) class of information by solving a simpler model.

ex: Dynamical Mean Field Theory: momentum averaged self energy from quantum impurity model + self consistency condition
Equilibrium DFT+DMFT

Impurity model with self-consistently chosen hybridization functional
Nonequilibrium DMFT

Original formulation:

- Schmidt and Monien, arXiv:cond-mat/0202046
- Freericks, Turkowski, and Zlatic, PRL 97, 266408

Nice description of modern understanding

Key issue: solving impurity model
Continuous time Monte Carlo:

\[ H = H_a + H_b \]

- interaction representation with respect to \( H_b \)

\[
Z = \text{Tr} \, T_\tau e^{-\beta H_a} \exp \left[ - \int_0^\beta d\tau H_b(\tau) \right]
\]

- formal expansion in \( \tilde{H}_b \)

\[
= \sum_k (-1)^k \int_0^\beta d\tau_1 \ldots \int_{\tau_{k-1}}^\beta d\tau_k \\
\quad \times \text{Tr} \left[ e^{-\beta H_a} H_b(\tau_k) H_b(\tau_{k-1}) \ldots H_b(\tau_1) \right]
\]

- sample series stochastically
Equilibrium: Continuous-Time Quantum Monte Carlo

Works very well because the method is estimating a real exponential

$$Z = \text{Tr} \left[ e^{-\beta H} \right]$$

Number of vertices needed set by $1/T$

Method is now very widely used.
Out of equilibrium

\[ \langle \hat{O} \rangle_t = Tr \left[ \hat{O} \hat{\rho}(t) \right] = Tr \left[ \hat{O} e^{-i\hat{H}t} \hat{\rho}_0 e^{i\hat{H}t} \right] \]

• Real time evolution forward from initial condition
• Number of vertices \( \sim \) time interval

• Two time contours required (twice as many vertices)
• Convergence by cancellation of oscillations

Cost: exponential in number of vertices
CT-QMC in real time

Results for Anderson model
(single level quantum dot with 2 leads)

Real time, equilibrium

Current at non-zero voltage

At interesting coupling strengths, method is limited to brutally short times.

Werner-Millis
Rabani-Muhlbacher
Schiro-Fabrizio

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First improvement: analytic resummation of diagrams

`NCA' (non-crossing approximation): resummation of all diagrams with no crossing hybridization lines or `OCA” (one crossing approx)
Stochastically sample corrections (give up wick’s theorem, but have to evaluate fewer diagrams)

In real time, convergence with respect to diagram order occurs because sum of all diagrams of a given order decreases rapidly with order.

This cancellation of diagrams (``sign blessing”―N. Prokof’ev) is problematic for Monte Carlo

Solution: put wall in calculation (no diagrams of order \( n > N_{\text{max}} \)). Then systematically increase \( N_{\text{max}} \) until convergence occurs.
Stochastically sample corrections 
(give up wick’s theorem, but have to evaluate fewer diagrams)

Method works if calculations converges at a low enough order that the sign does not kill you. Low T and strong interactions are hard to do.

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Magnetization relaxation

NCA good for elements of density matrix; bad (~factor of 2) for relaxation times

nb: density is easy to get right. Spin dynamics is real test of method
Bold expansion: accessible region $\sim 5 \times \Gamma$ not long enough!

Refinement (Cohen/Rabani): long timescale comes from small value of relaxation time. But relaxation time is determined by short time physics accessible to bold CT-QMC.

Compute relaxation kernel using diagrams up to time $t$. Then use computed kernel to evaluate physical properties to longer times.

Convergence tests again crucial.
convergence tests
state of the art summer 2015
Fall 2015: 
`Inchworm’ method. G Cohen

In standard QMC the number of vertices needed scales with the time interval.

<sign>~Exp[-# vertices]

Guy’s insight: interaction lines are finite range in time =>can compute G at longer times in terms of G for shorter times.
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\[ G((n+1)\Delta t, t_i) = \text{Diagrams}[\Delta t] \ast G(n\delta t, t_f) \]

to add a time interval \( dt \), need only a finite set of diagrams:

iteratively construct \( G \) starting from short times. \( \Rightarrow \) total cost \( (t/dt)^2 \)
the Kondo regime is now accessible

\[ U = 8\Gamma \quad T = \frac{\Gamma}{50} \]

NB: need to go to at least 2CA for decent accuracy
Quantum dot in ac field

\[ f(t) = A + B e^{-\gamma t} + C \sin(\omega_0 t + \phi). \]
Summary

1. Are there new nonequilibrium "phases" (long-lived metastable states). Yes, I think
2. Reasonable understanding of initial stages of excited carrier 'pseudo-thermalization'
3. Open questions:
   • Existence of metastable phase beyond HF
   • Dynamics of relaxation to equilibrium
   • Motion of lattice
4. Beyond HF: situation is improving