Nonequilibrium Physics of Correlated Electron Materials IV:

Nonequilibrium Phase Transitions

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College de France
Oct 12, 2015

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Two classes of nonequilibrium many-body phenomena

1. Steady state drive (current-driven) stabilization of metallic phase

   Maeno et al Ca_2RuO_4

2. Transient perturbation

   Long-lived response to carrier excitation

   Morrison et al VO_2

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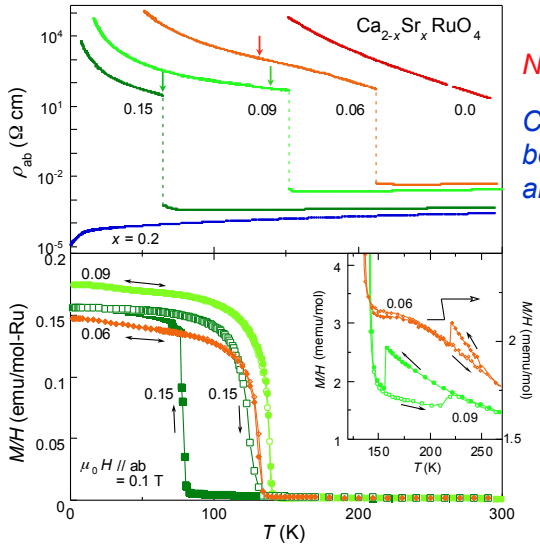
Steady State Drive

Y. Maeno

Mott transition in $Ca_{2-x}Sr_xRuO_4$

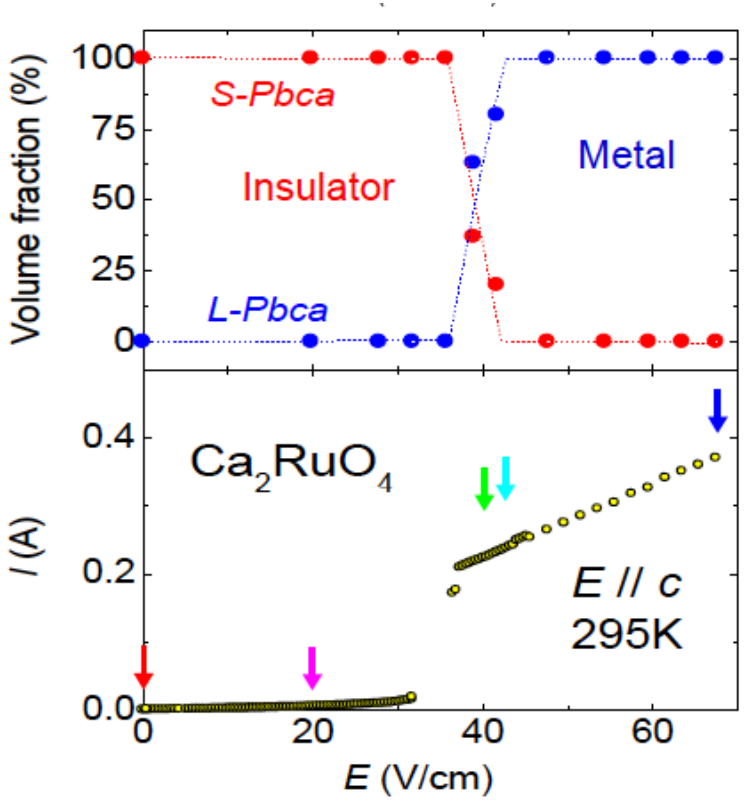


No anomaly at T_N

Clear separation between "charge gap" and "spin exitation".

Mott insulating ground state

Nakatsuji *et al.*, PRL.



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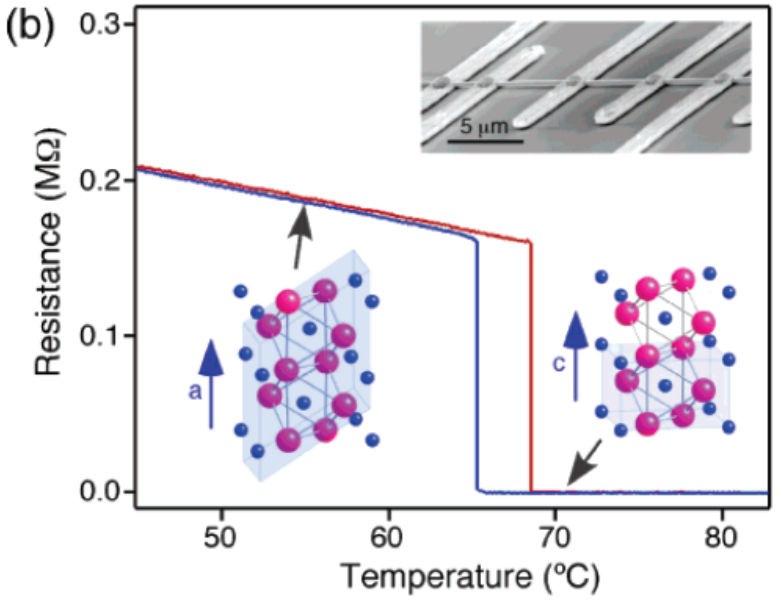
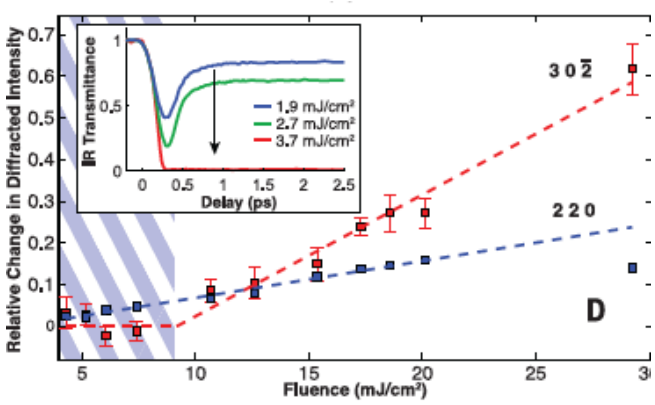
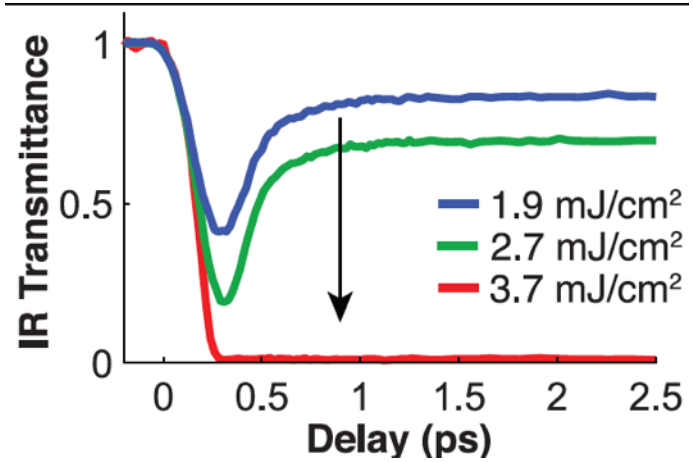
Response to pulse

Morrison et al VO_2







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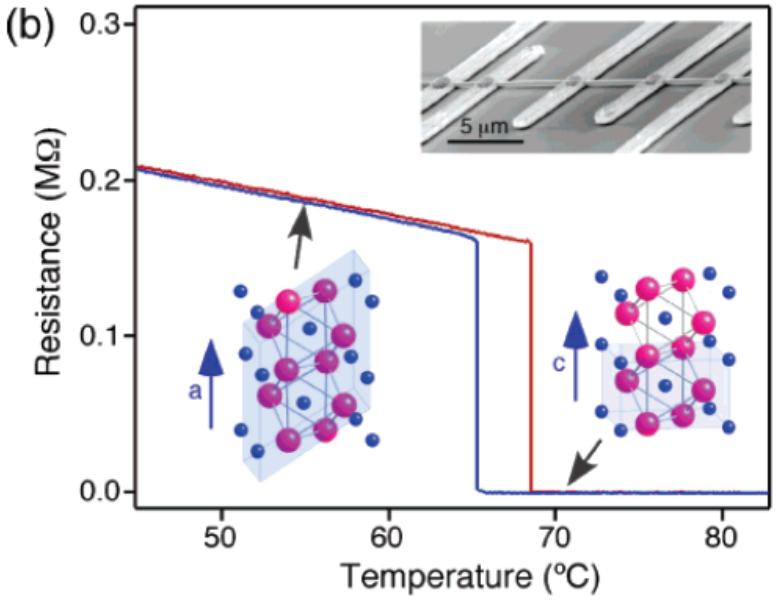
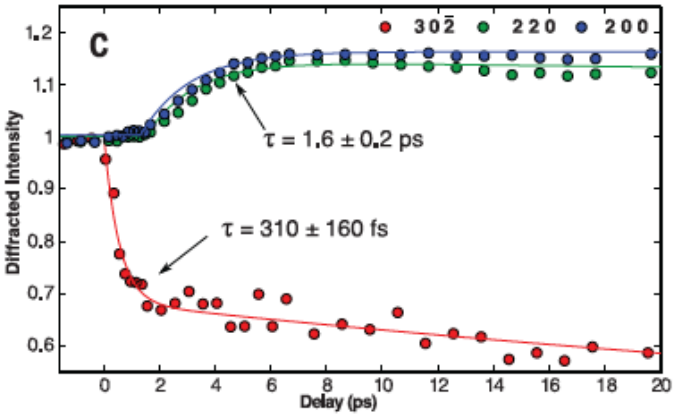
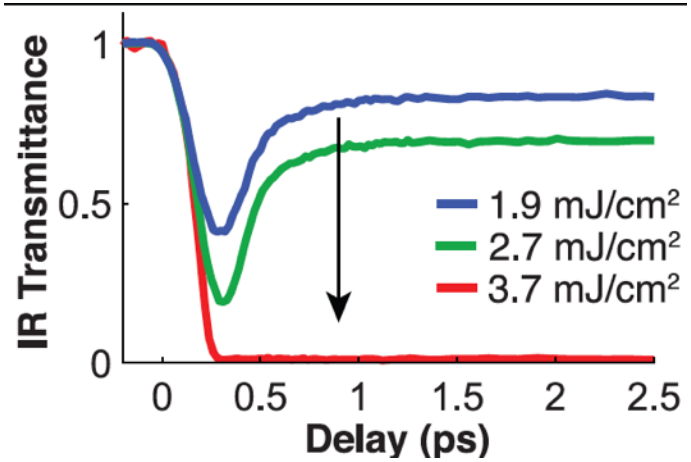
Response lasts >20ps

Morrison et al VO_2







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Outline

1. **Current driven metal-insulator transition**

2. **Can a response to a short pulse generate a new phase**

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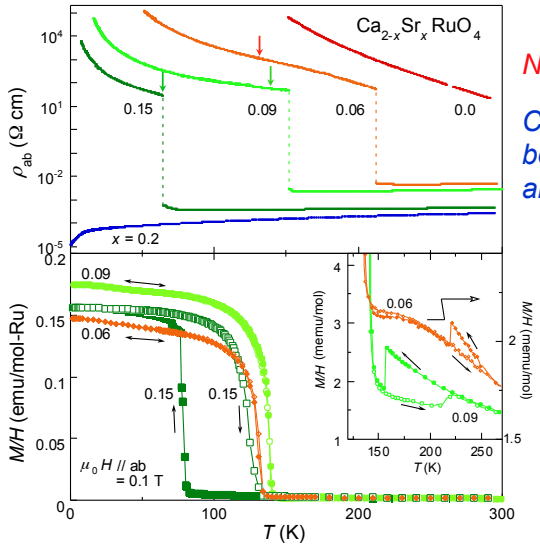
Steady State Drive

Y. Maeno

Mott transition in $Ca_{2-x}Sr_xRuO_4$



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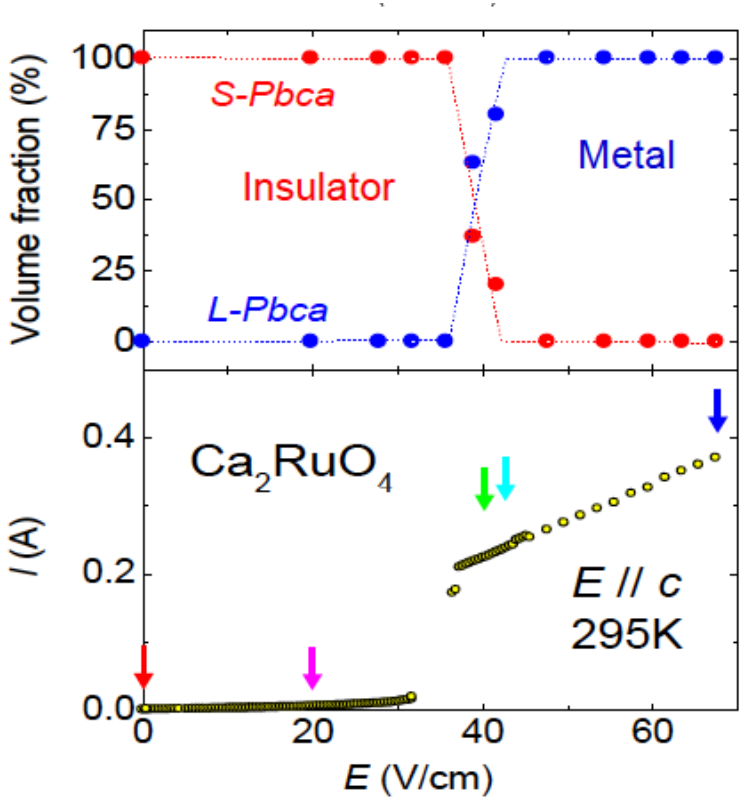

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Theory

Nonequilibrium dynamical mean field approx.

Perturbative solvers (IPT, NCA)
qualitative behavior seems reaonsable
quantitative accuracy is an open question

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Some concepts from single-particle physics

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Some concepts from single-particle physics

and how they show up in the many-body calculations

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Bloch Oscillations

(relevant to cold atom systems; unlikely to be important for actual condensed matter materials)

$$\text{gauge invariance} \implies \mathbf{k} \to \mathbf{k} + \frac{\mathbf{eEt}}{\hbar}$$

$$\mathbf{current} \propto ea \ \mathbf{sin}\ \mathbf{k(t)a}$$

$$\mathbf{period\ T} = \frac{\mathbf{h}}{\mathbf{eEa}}$$

Only relevant if scatt rate < voltage across a unit cell

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DMFT calculations:
no `thermostat'

=>possibility for energy to increase indefinitely

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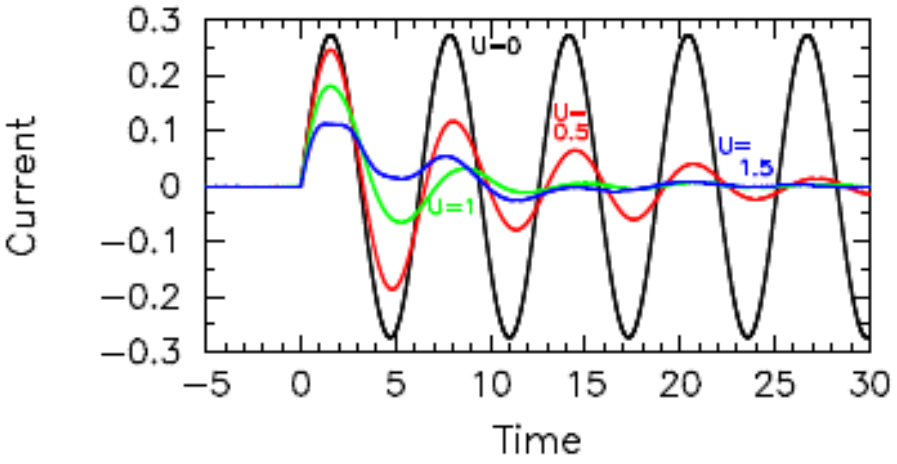
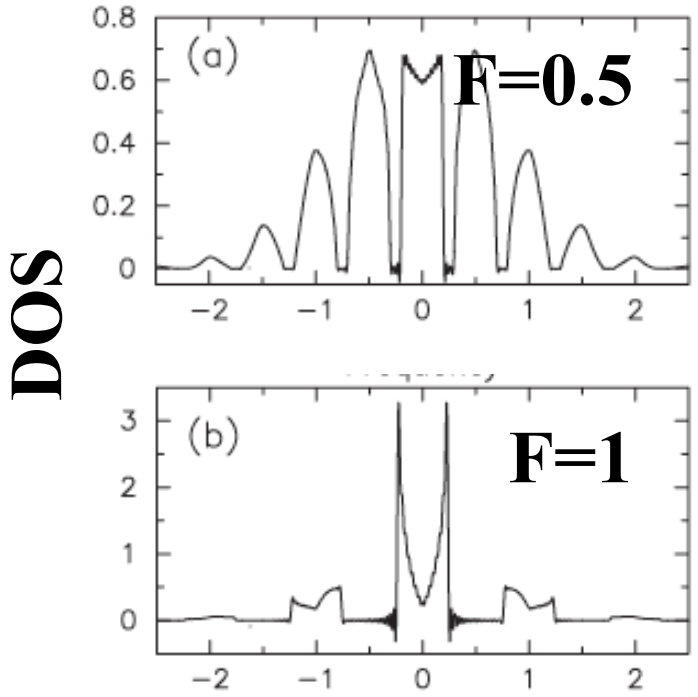
Freericks 2006
Falicov-Kimball Model

Stark Ladder

F=eEa=1





Oscillations damped by interactions
Long time behavior not quite established

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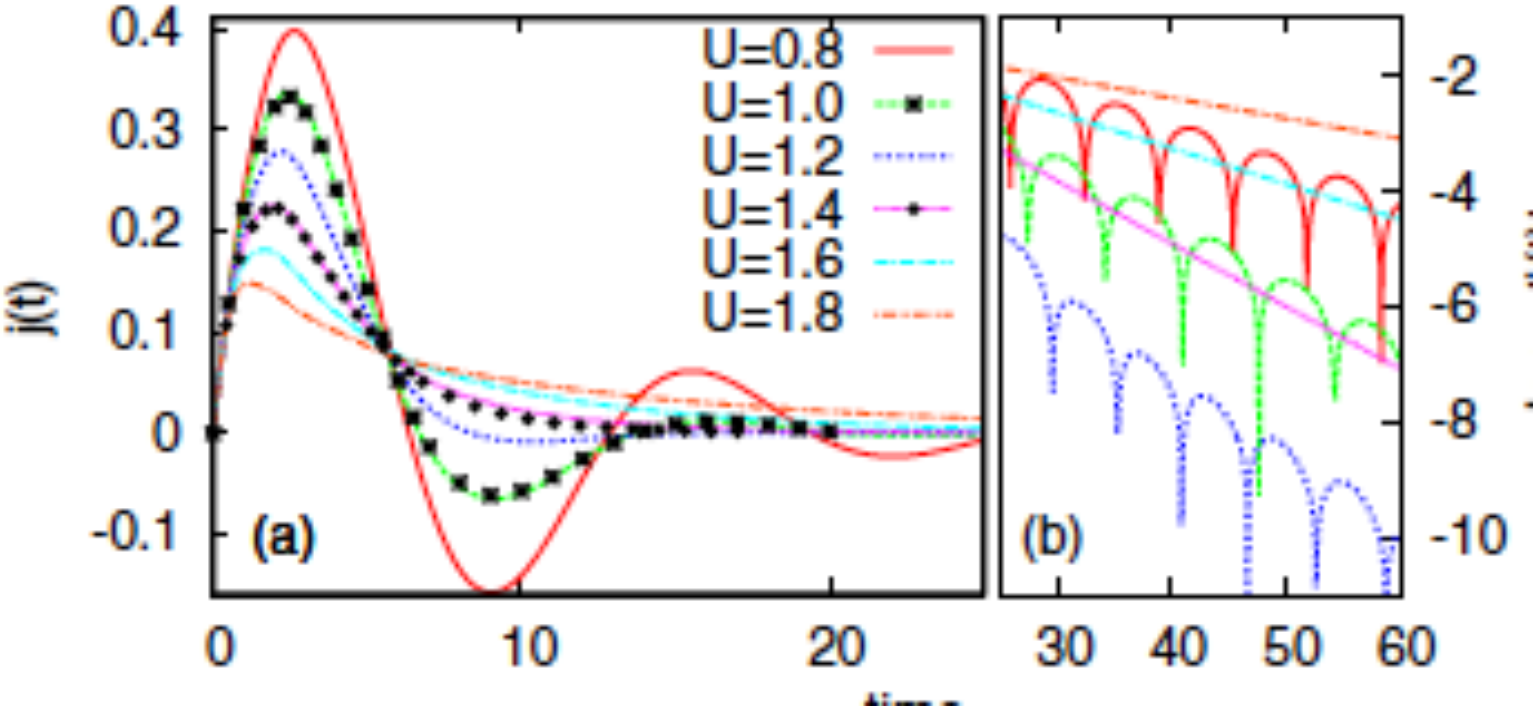
Werner-Eckstein
Hubbard mode; IPT solver



Bloch oscillations disappear above critical interaction strength (<U_{MIT})

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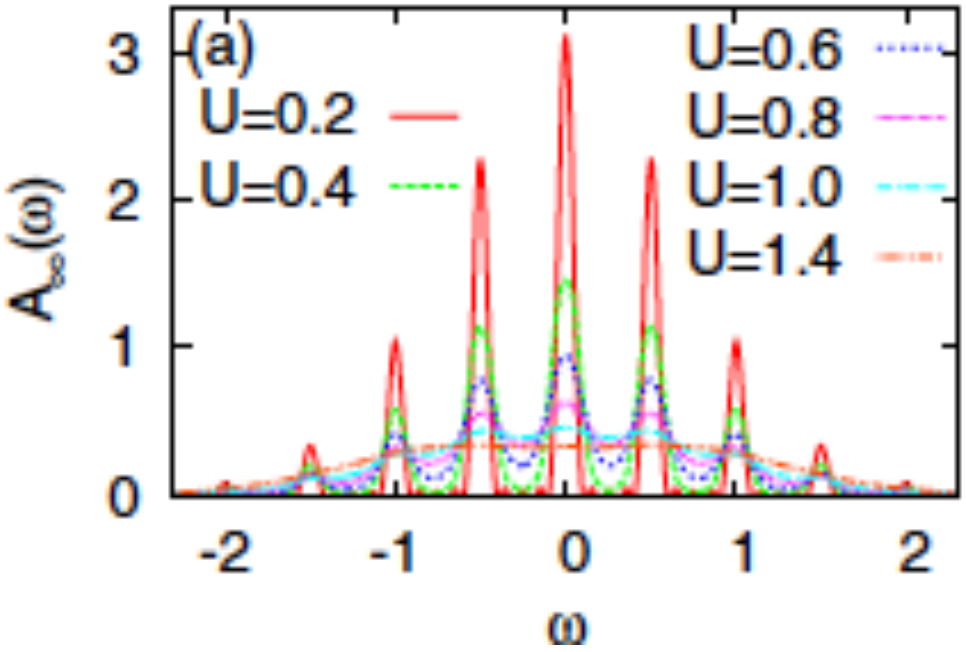
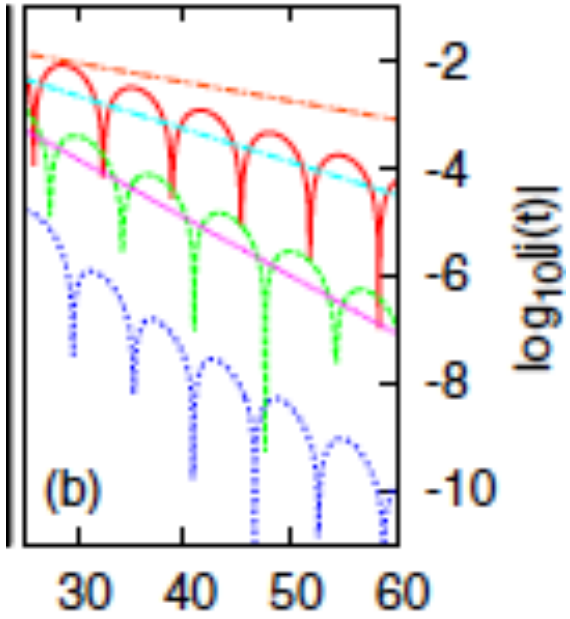
Werner-Eckstein
Bloch oscillations tied to Stark Ladder





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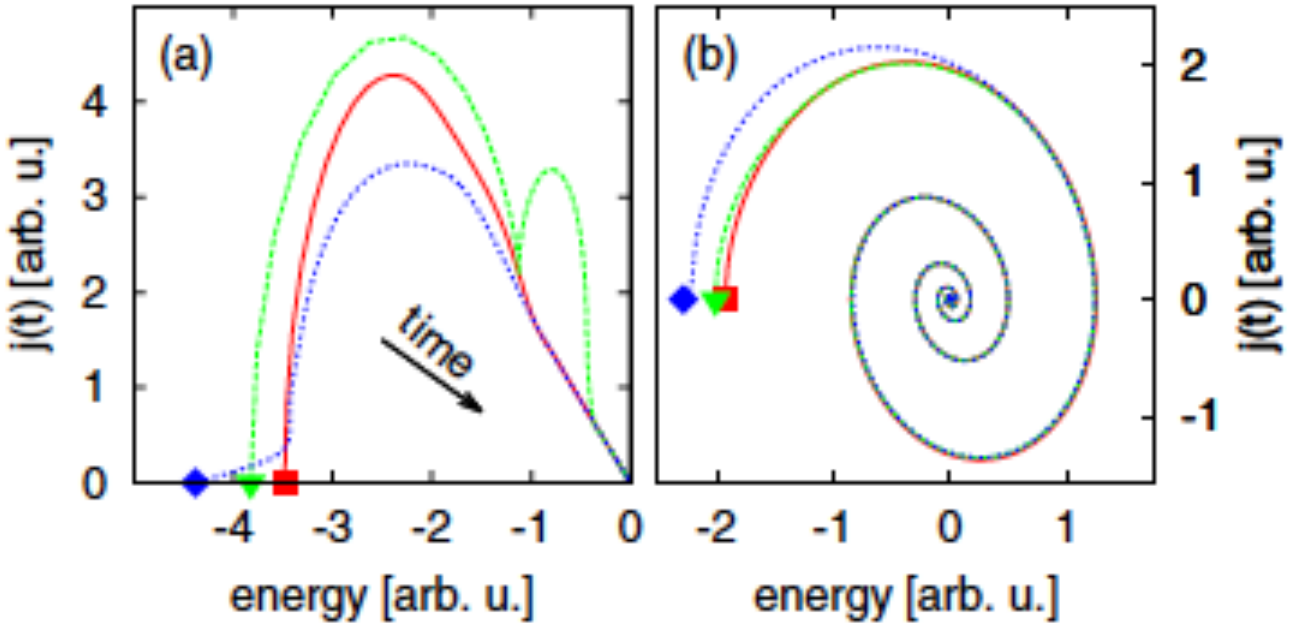
long time limit
infinite temperature (0 energy) state



$$\mathbf{E} = \left\langle \hat{H} \right\rangle = \left\langle \sum_{k\sigma} \varepsilon_k c^{\dagger}_{k\sigma} c_{k\sigma} \right\rangle + U \sum_i \left(n_{i,\uparrow} - \frac{1}{2} \right) \left(n_{i,\downarrow} - \frac{1}{2} \right)$$

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This transition appears similar to the transition from underdamped to overdamped behavior in the simple harmonic oscillator

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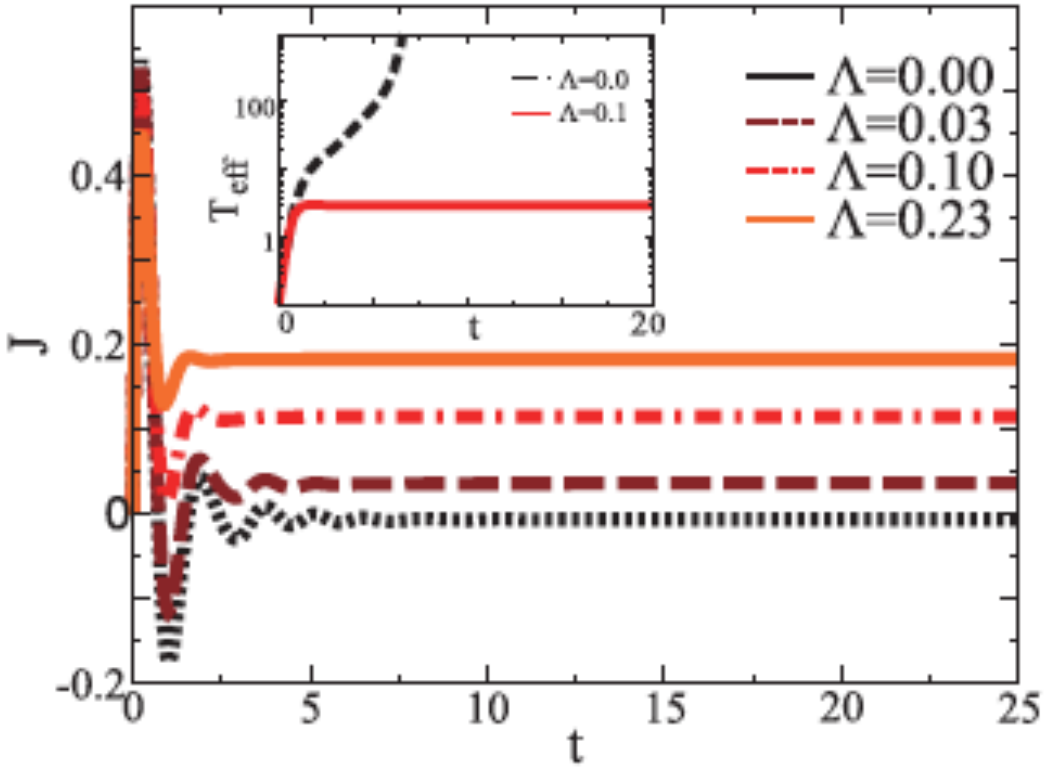
Add coupling to heat bath



F=eEa=4.7 /1.9

Properties crucially dependent on how you take the heat out

Amaricci, Kotliar et al. Hubbard model; metallic regime

Phys. Rev. B 86, 085110

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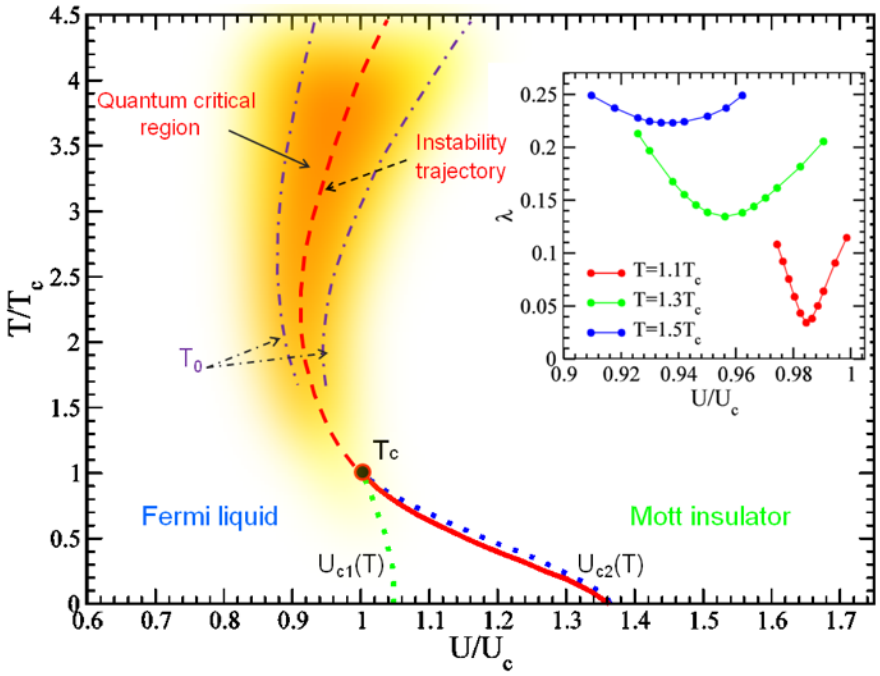
Aron et al: additional effect of Joule heating

In single-site DMFT



Raising T=> metal-insulator transition (at a quite low T)

Image from Dobrosavljevic et al PRL 107 026401

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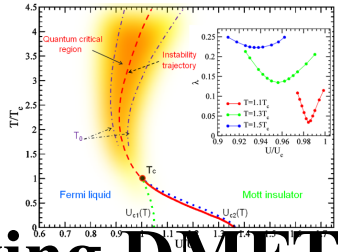
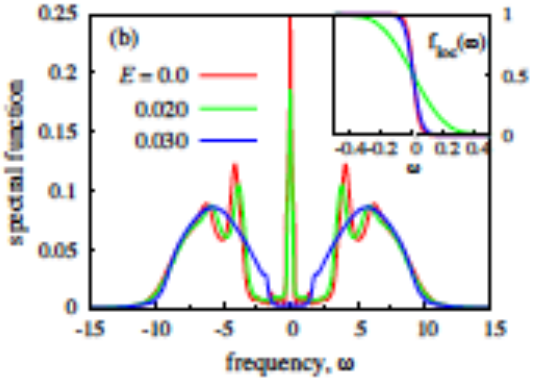
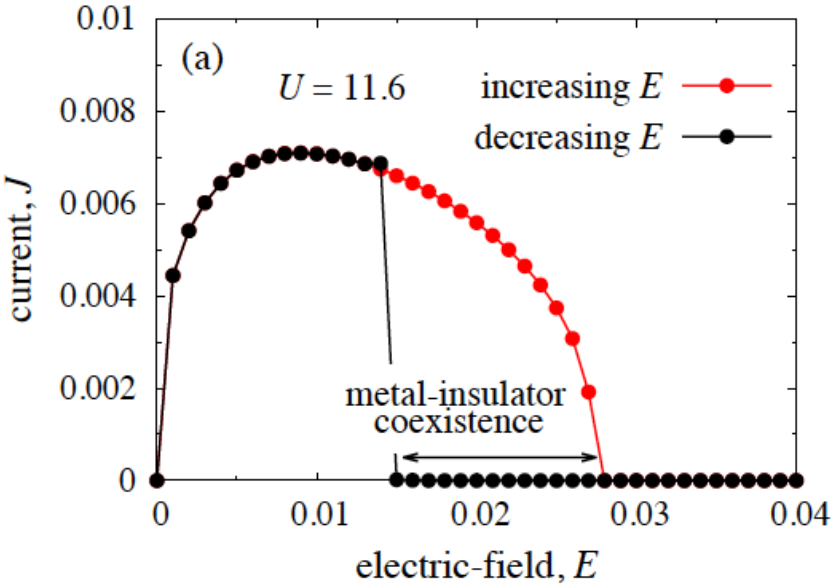
Arons et al



Clever method of solving DMFT eqns in steady state





Joule heating drives system across metal-insulator phase boundary

Proximity to MIT and weak coupling to reservoir=>small field scale

arXiv:1210.4926

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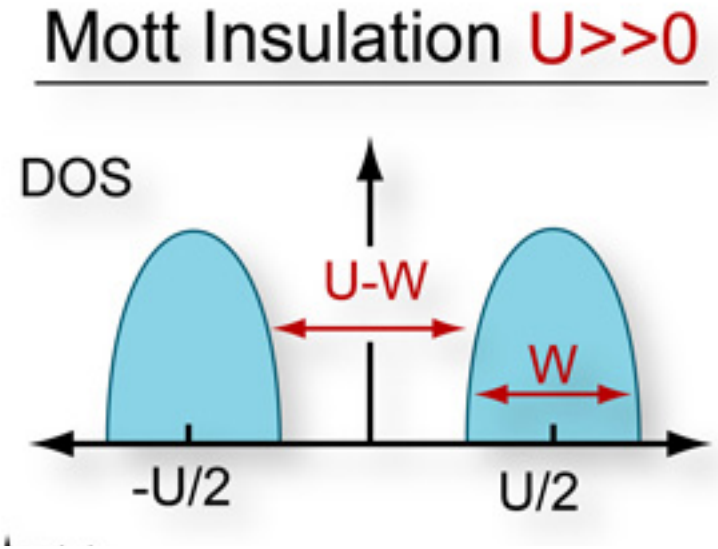
Zener Tunnelling



Real transitions allowed if tunnel a distance d such that $Ed = \Delta_{Mott}$

Tunnelling probability exponentialy small in d

$$j \sim E\, e^{-\frac{E_{th}}{E}}$$

If Mott gap is self consistent phenomenon: can a large enough current make it collapse? is there a transition at a critical current

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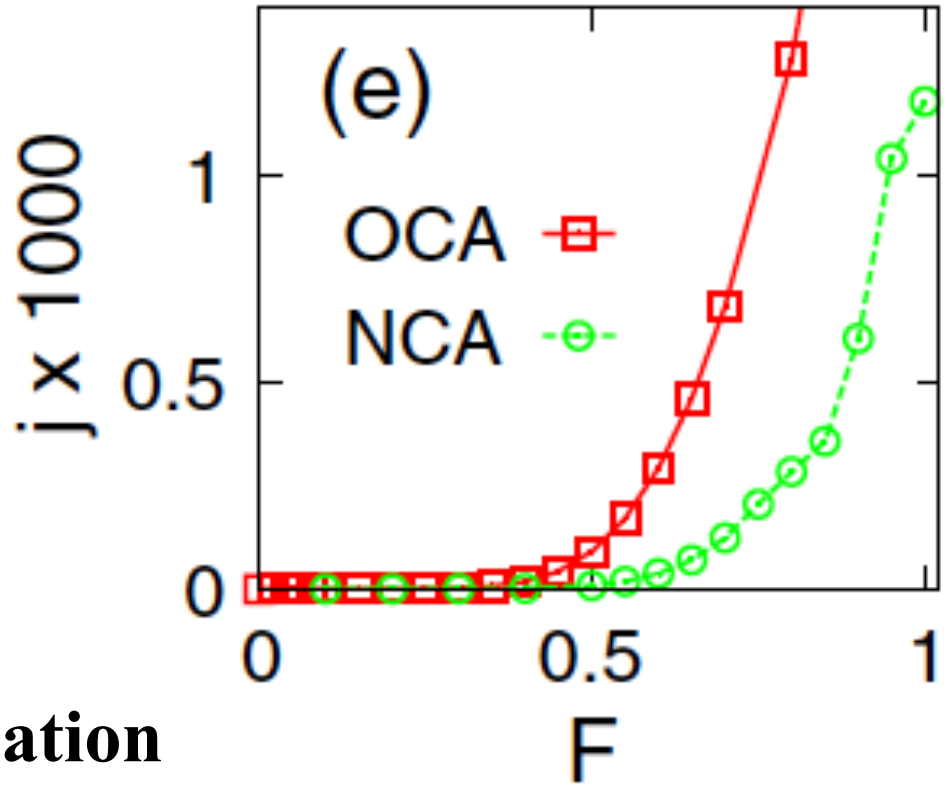
Eckstein and Werner
Steady state current

$$j = Fe^{-\frac{F_{th}}{F}}$$



Details dep on approximation
qualitative behavior similar

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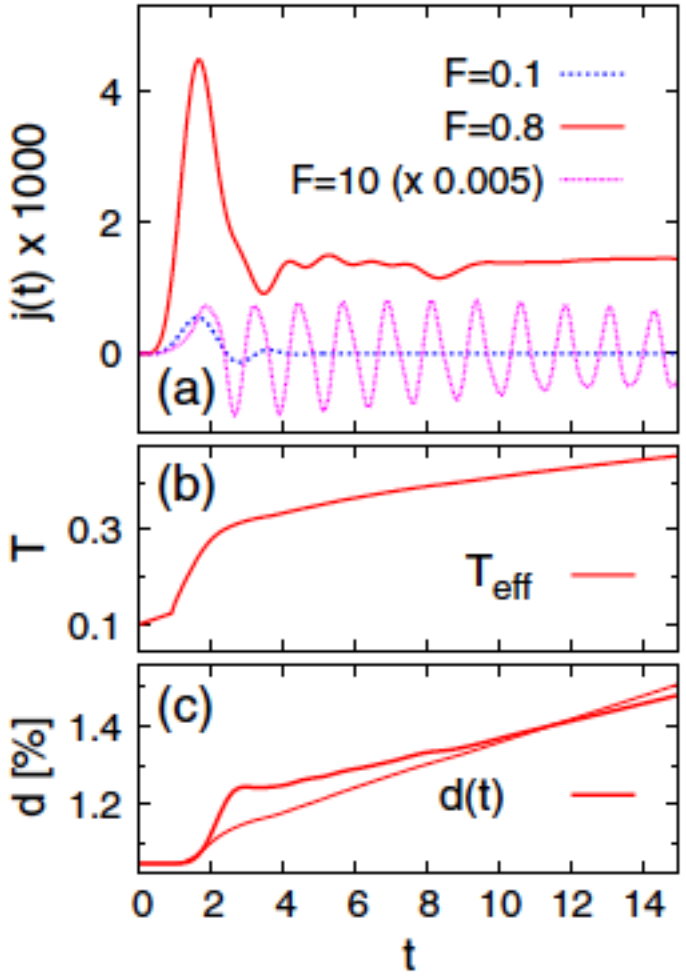
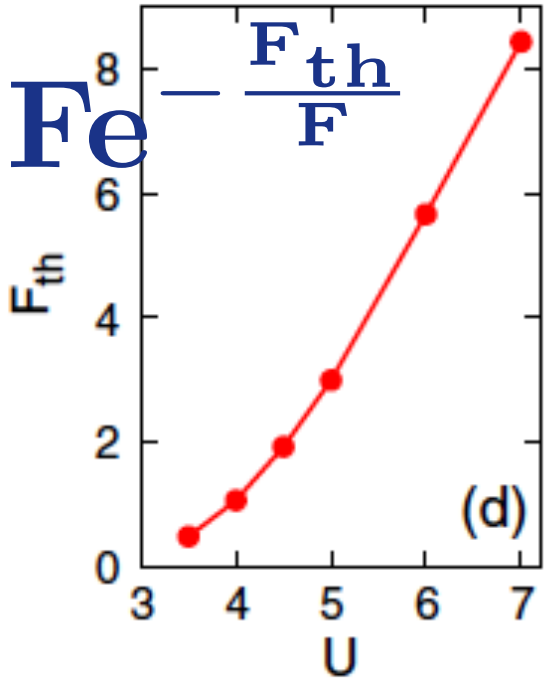
Nonequilibrium non-steady state



$$j = Fe^{-\frac{F_{th}}{F}}$$



Non-thermal state with approximately time-independent current

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Summary
DMFT on Hubbard and related models

Physics: Single particle physics + many-body scattering

Nonequilibrium many-body state with properties that depend on `thermostat'

Field scales: voltage drop over 1 unit cell ~ fraction of Mott gap=>~0.1eV or more over 1 unit cell

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An observation

from Y. Maeno.
Cambridge talk 2015

Bulk Materials	Breakdown Field (kV/cm)
$La_{2-x}Sr_xNiO_4$ [1)]	1~10
Sr_2CuO_3 [2)]	1~3
$SrCuO_2$ [2)]	0.3~1
(TTeC1TTF)-TCNQ [3)]	0.3~1.2
$GaTa_4Se_8$ [4)]	0.8~4
Ca_2RuO_4 [5)]	0.04
VO_2 (films in EDLT)	

One unit cell
$\sim 4 \times 10^{-8}$ **cm**

Voltage drop across one unit cell
$0.4 - 0.002$ **meV**

Tiny on electronic scales

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Implication

Bulk Materials	Breakdown Field (kV/cm)
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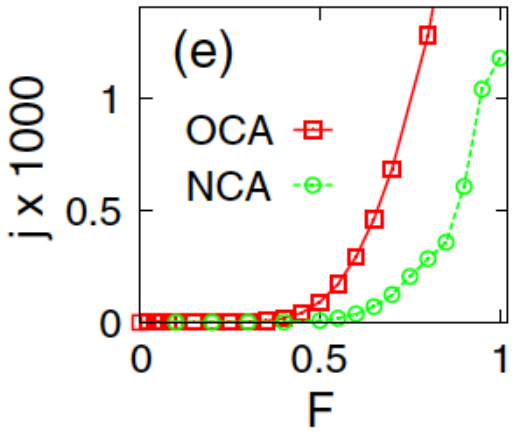

Current-driven transitions are not explicable in terms of the local physics accessible to DMFT. They are a collective phenomenon and the effect of current on longer length scale physics needs consideration.

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Observations
(not yet a theory)

- **The physics of the DMFT calculations is: voltage drive heats electrons: hot electrons drive physics**
- **We need: effective temperature for order parameter, even if electrons stay ``cold'' (equilibrated with reservoir)**

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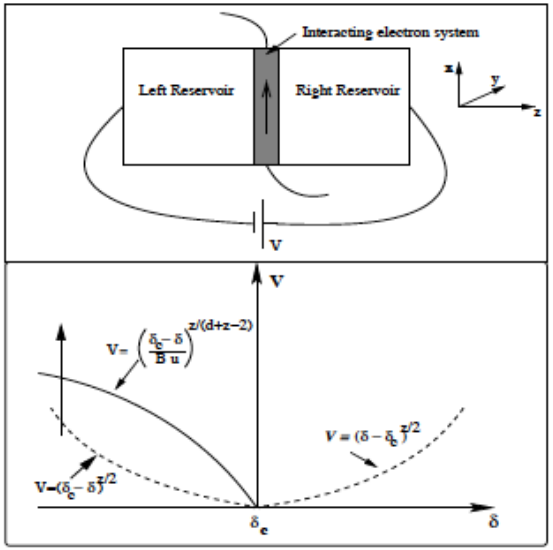
2 results from study of nonequilbrium magnetic quantum critical phenomena:



``Thermal" transition: Teff~V although electrons are in equilibrium with lattice

Phys. Rev. Lett. 97 236808 (2006)

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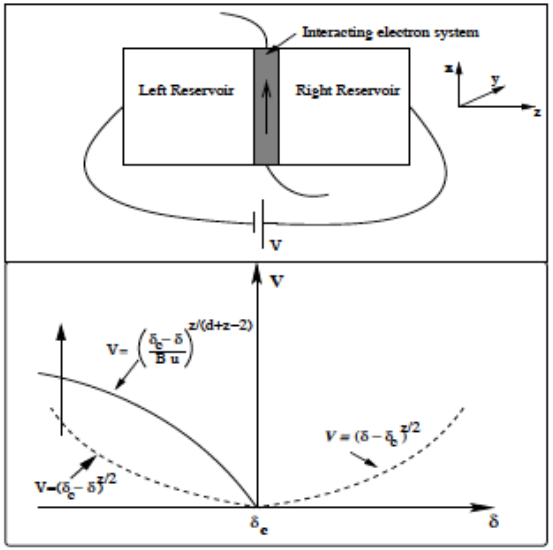

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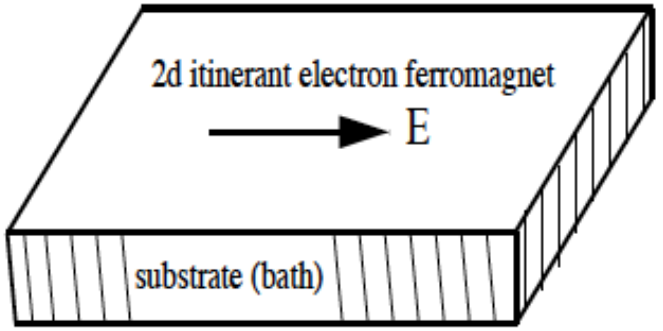
Current in-plane



$$T_{eff} = eEl_{sc}$$

Phys. Rev. B77, 220404 (2008)

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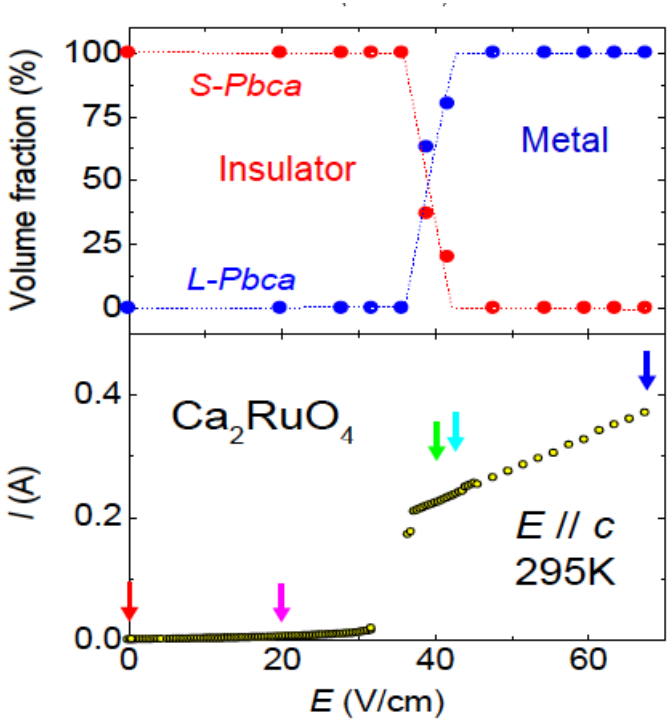
Idea: small gap=>long length scale Small electric field gives high order parameter temperature



$$T_{eff} = eEv_F/\Delta$$

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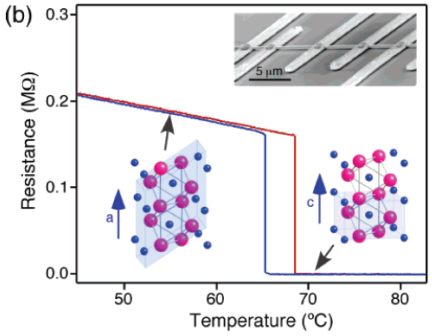
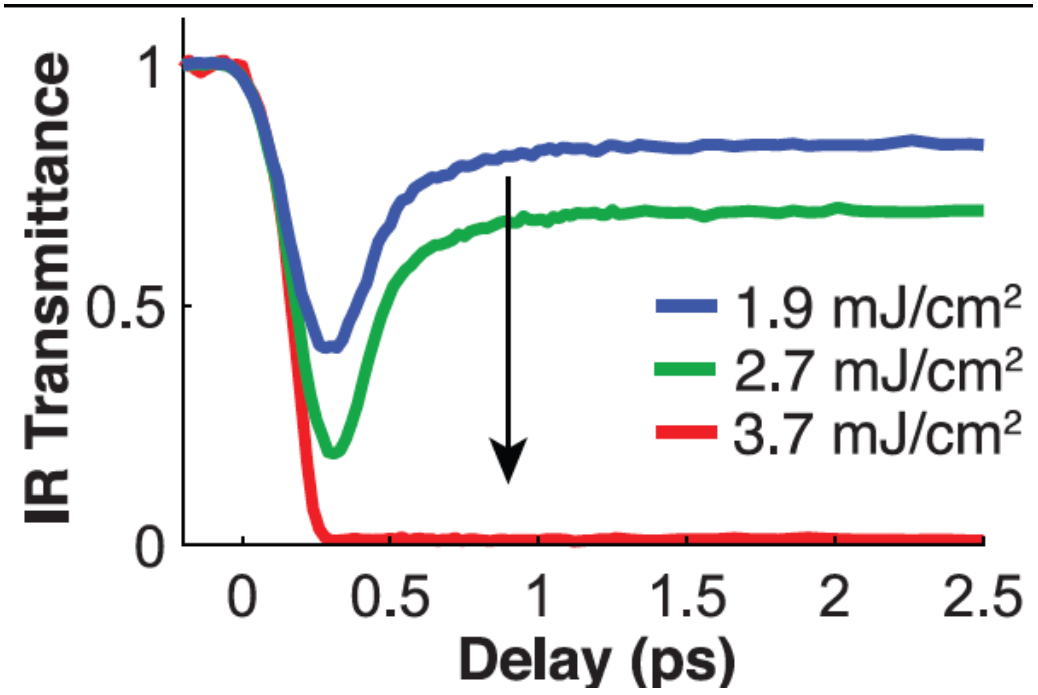
After a pulse

VO_2





lifetime of conducting state:
slow 1 electron kinetics
or new metastable electronic phase

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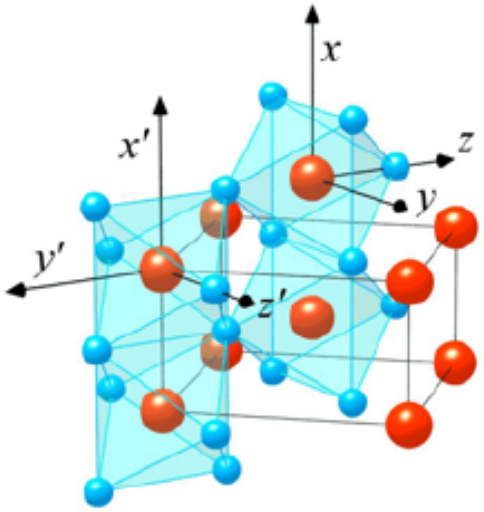
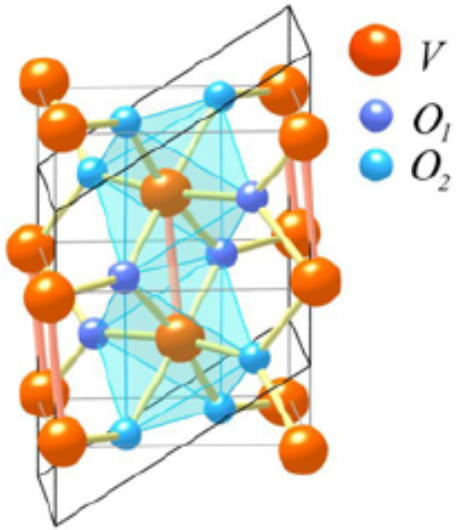
VO2: Structurally distorted insulator

High temperature : rutile phase (metallic)



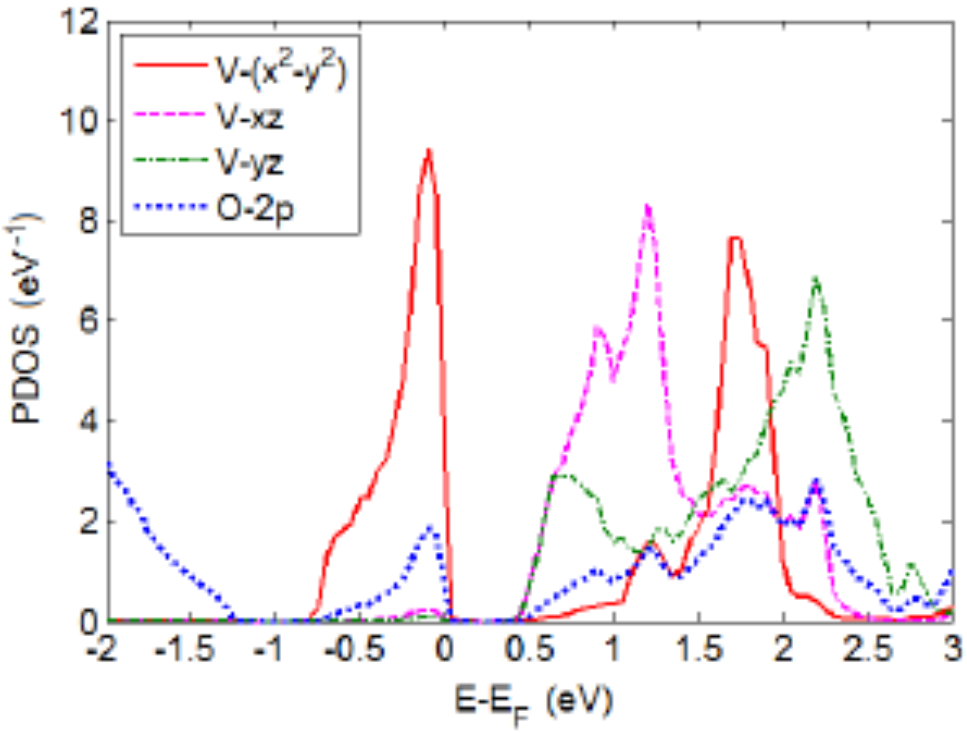
Low temperature : monoclinic phase (insulating)



The metal-insulator transition is accompanied by a structural transition with dimerization of the V atoms and tilting of the pairs out of the z axis.
(From V. Eyert, Ann. Phys. (Leipzig) 11, 650-702 (2002))

VO2: Insulating phase

Hartree-Fock band structure Z. He



Insulating phase
2 moving parts

Interaction-enhanced dimerization

Interaction-driven crystal field splitting

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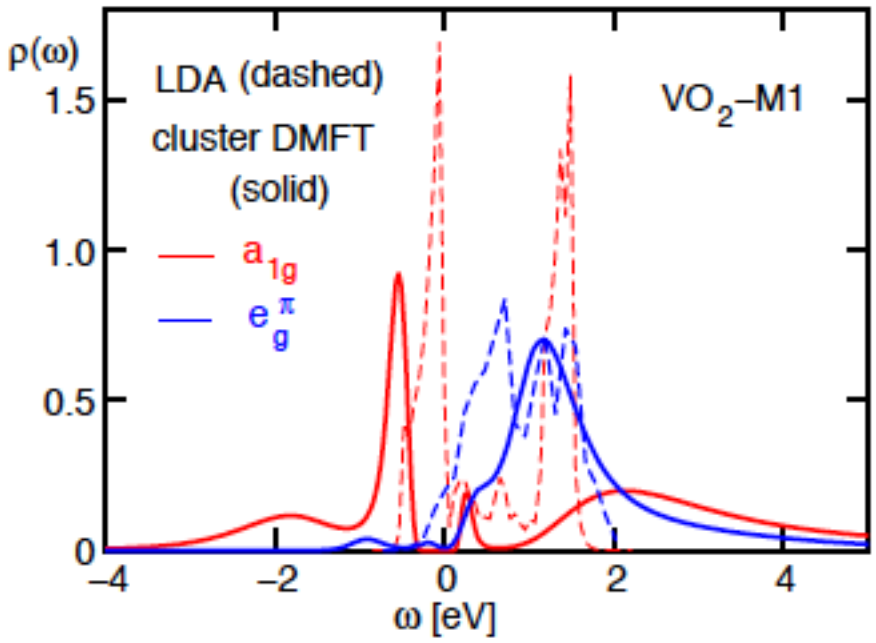
VO2: Insulating phase

DFt and DFT+DMFT



Biermann et al 2005

Insulating phase
2 moving parts

Interaction-enhanced dimerization

Interaction-driven crystal field splitting

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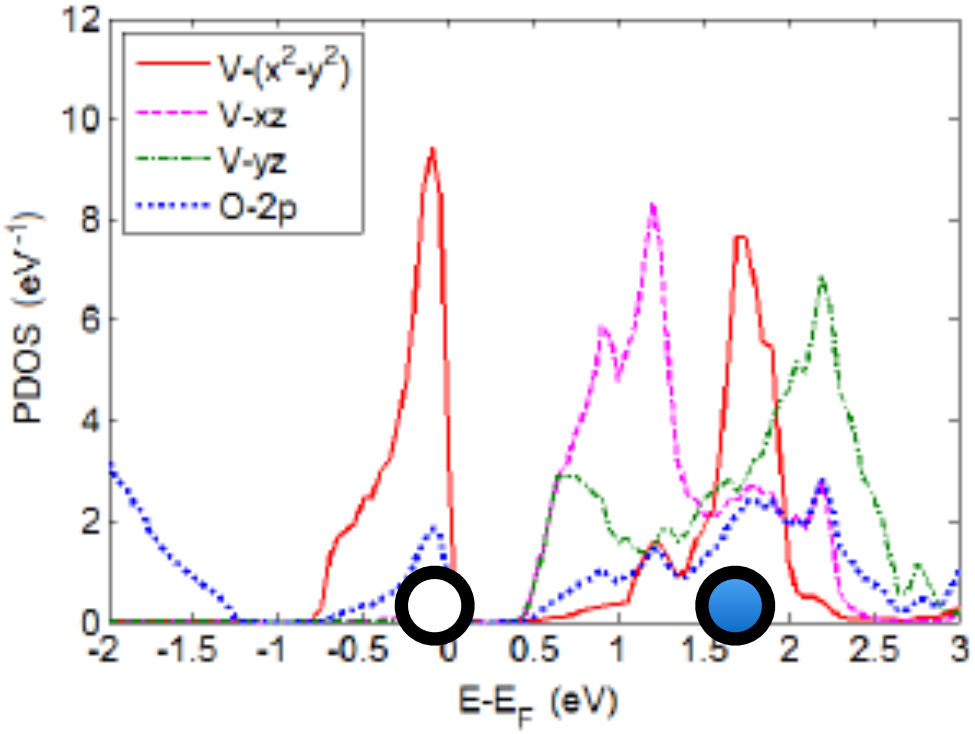
Experiment: excitation at 1.5eV (chosen for experimental convenience)



Excite carriers from bonding to antibonding orbitals

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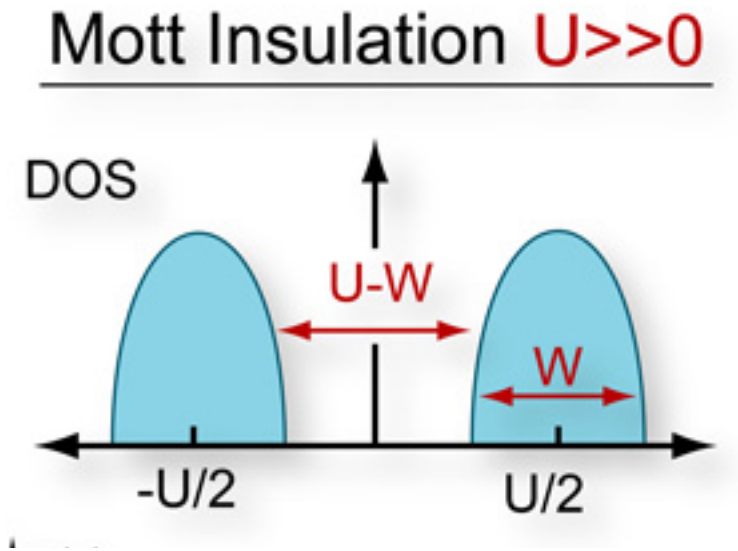
Interacting model



Photexcitation:

Add energy to system
Create doubly occupied and empty sites

$$\delta E \approx U \delta N_d$$

?Thermalization?

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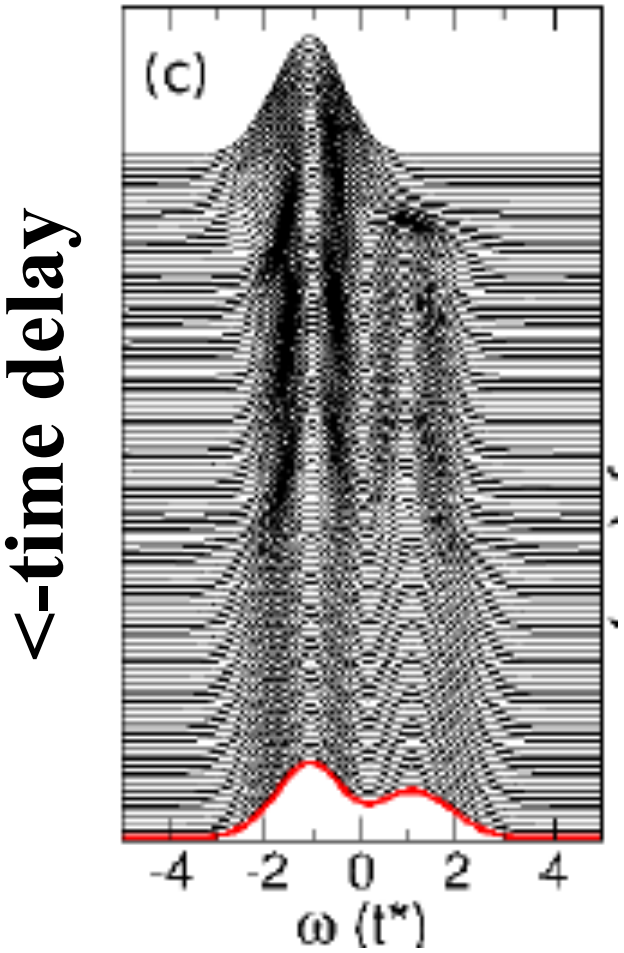
Falicov Kimball model



Moritz, B., A. F. Kemper, M. Sentef, T. P. Devereaux, and J. K. Freericks, 2013, Phys. Rev. Lett. **111**, 077401.

In Falicov-Kimball model, doubly occupied states live forever

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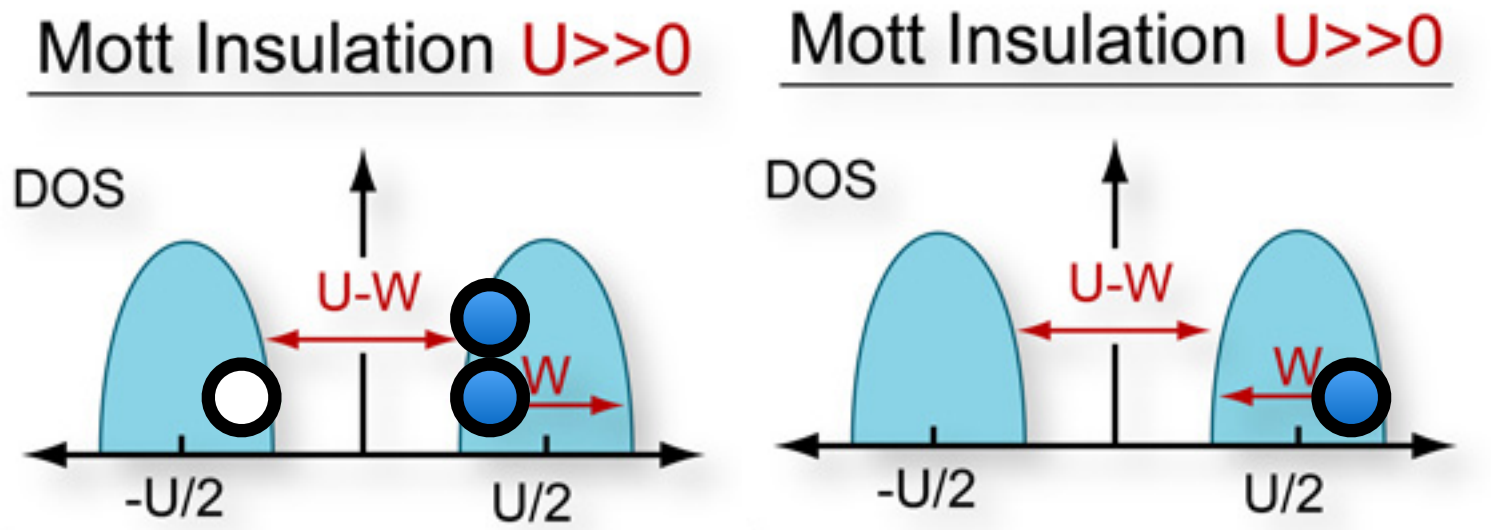
Hubbard model: thermalization



Recombination: : liberates energy U-W

=> if W<U-W, then possible final state

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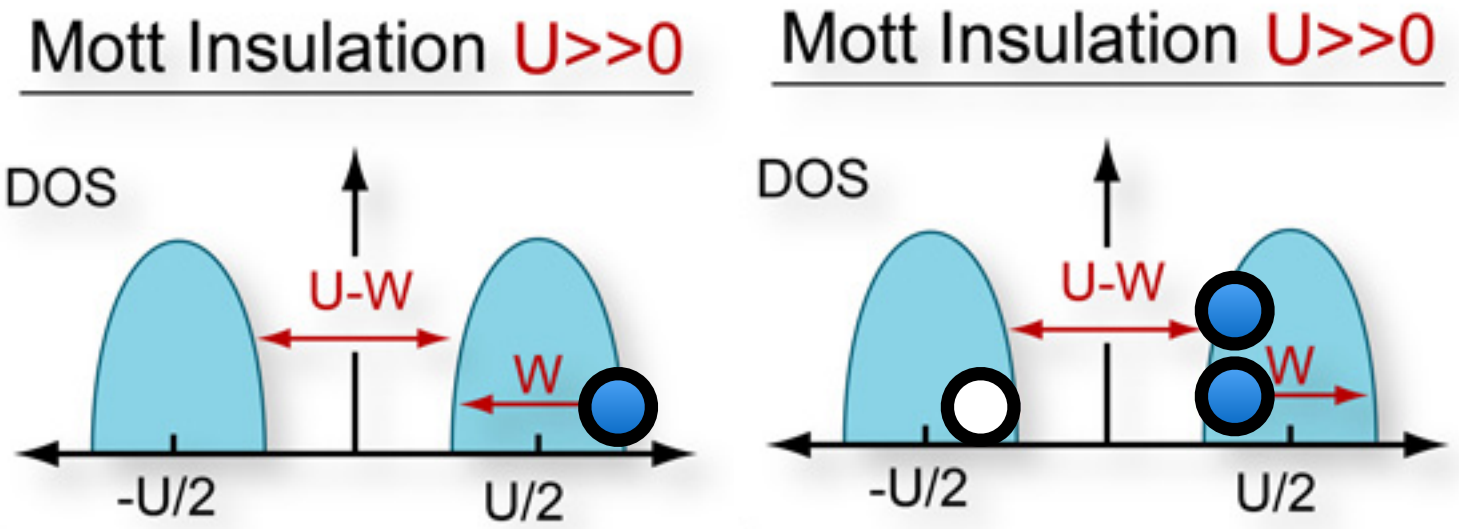
Hubbard model: thermalization



Auger: create more doublons

also if W<U-W, then possible final state

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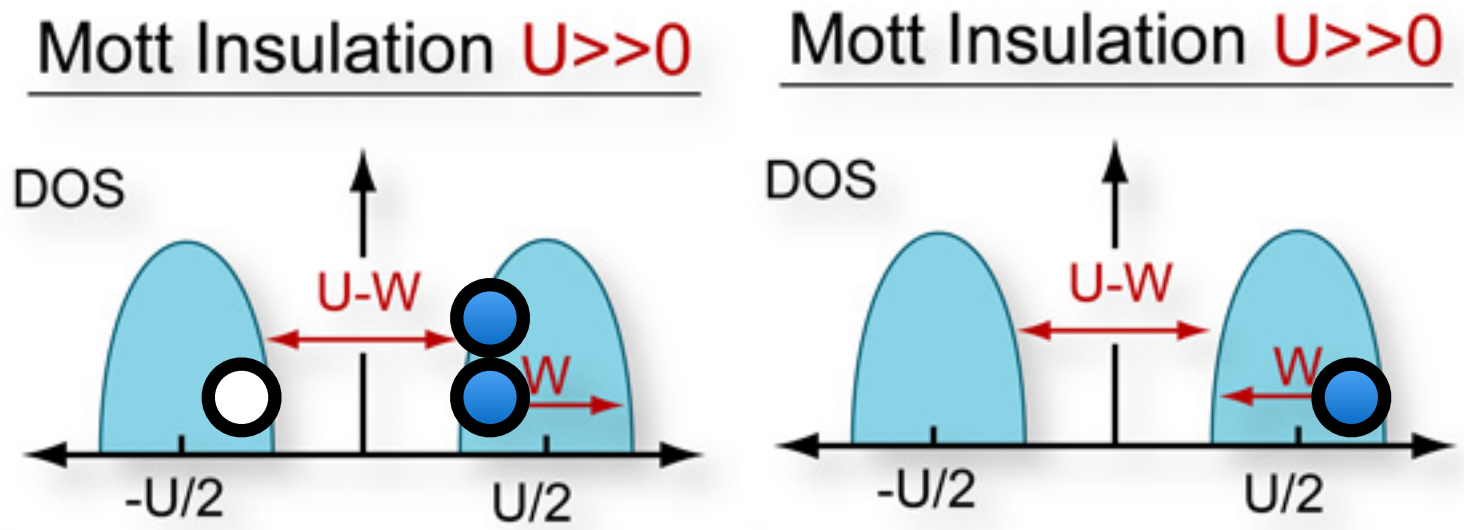
Hubbard model: thermalization



Recombination: : liberates energy U-W

=> if W<U-W, then possible final state

Spin and phonon degrees of freedom can also provide energy to relax dist (mainly within bands)

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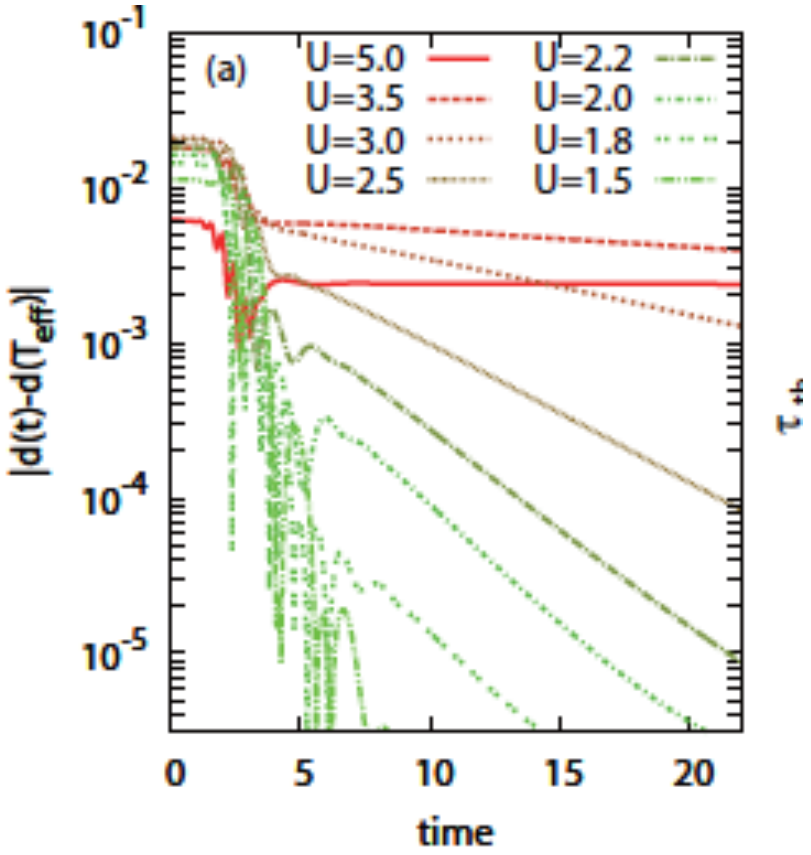
DMFT
Werner and Eckstein



Exponential relaxation to thermal equilibrium state

But no discernable relaxation for U-5: U-W>W

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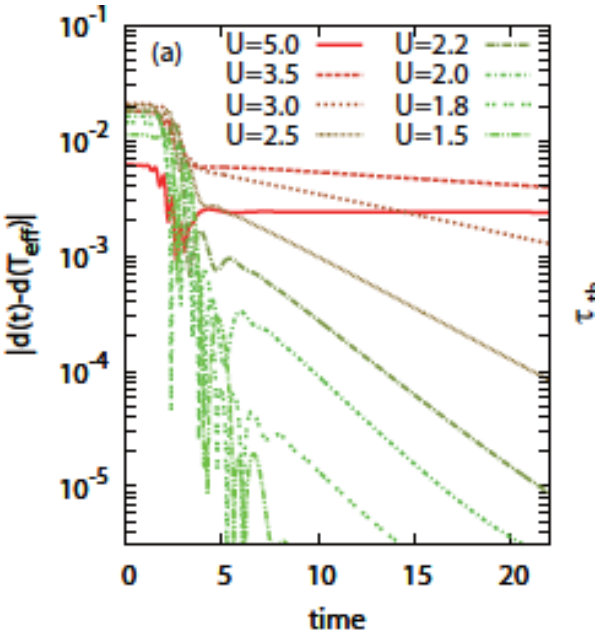
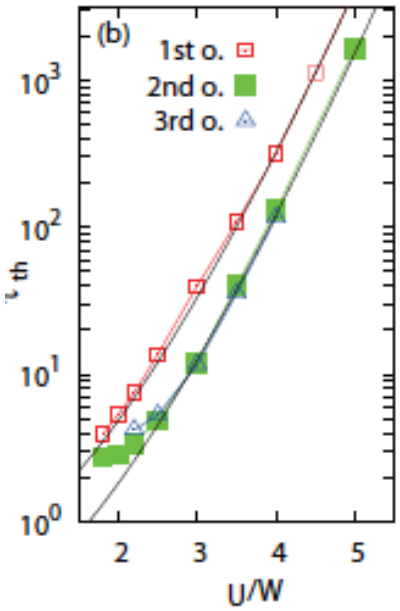
DMFT
Werner and Eckstein





Relaxation of number of `doublon' becomes arbitrarily slow at large gap

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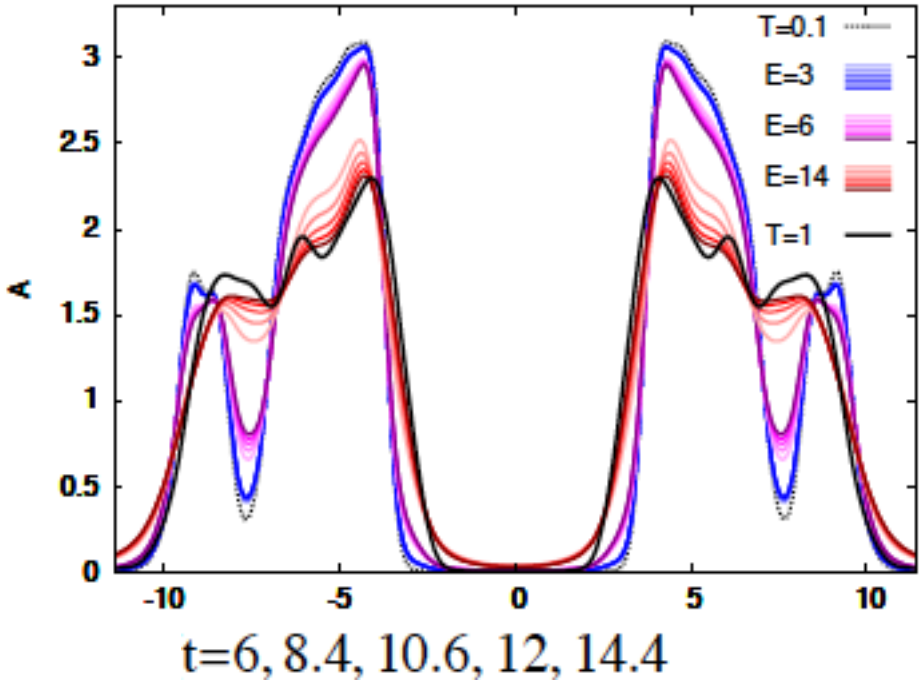
Werner/Eckstein arXiv:1410.3956

Details of relaxation within band depend on other DOF



E=3 d=0.002
E=6 d=0.009
E=14 d=0.045

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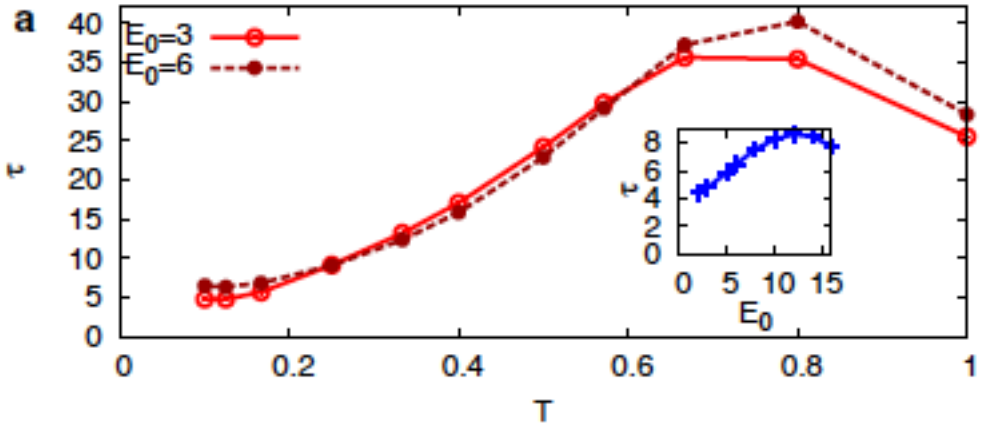
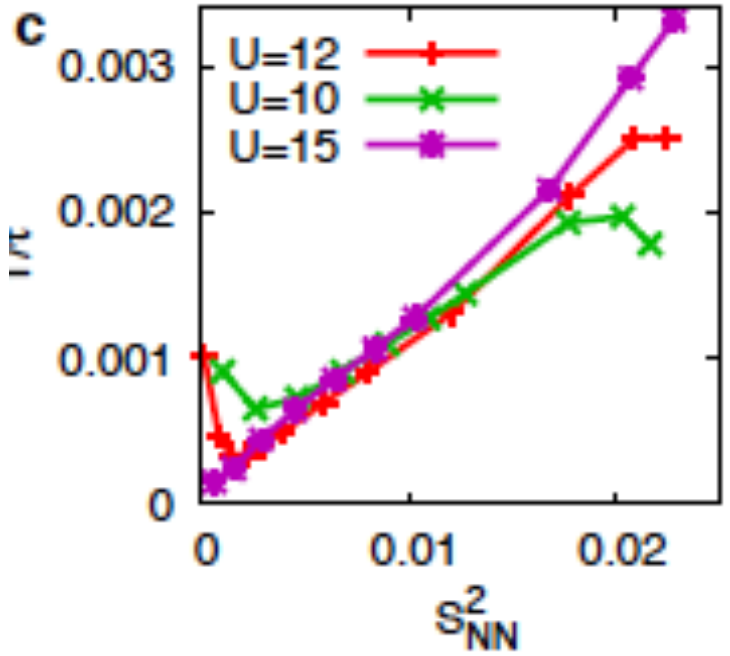
Magnon contribution to relaxation time





 # appropriate for oxides: ~10-100fs

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Summary: kinetics Hubbard model

- **General: thermalization on ~10-100fs scale to state with given energy.**
- **relaxation of doubly occupied sites via recombination**
- **Very significant kinetic barrier to recombination if Mott gap is larger than bandwidth; otherwise reasonably fast**

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Hartree-Fock Analysis of VO_2

Z. He

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

Recall Slater-Kanamori Interactions

$$
\begin{aligned}
H &= U\sum_{a} n_{a\uparrow}n_{a\downarrow} + (U-2J)\sum_{a>b,\sigma=\uparrow,\downarrow} n_{a\sigma}n_{b\sigma} \\
&\quad +(U-3J)\sum_{a\neq b\sigma} n_{a\sigma}n_{b\bar{\sigma}} - J\sum_{a\neq b} c^{\dagger}_{a\uparrow}c^{\dagger}_{a\downarrow}c_{b\uparrow}c_{b\downarrow} + c^{\dagger}_{a\uparrow}c^{\dagger}_{b\downarrow}c_{b\uparrow}c_{a\downarrow}
\end{aligned}
$$

Orbital number is preserved exc. by pair hopping ~J

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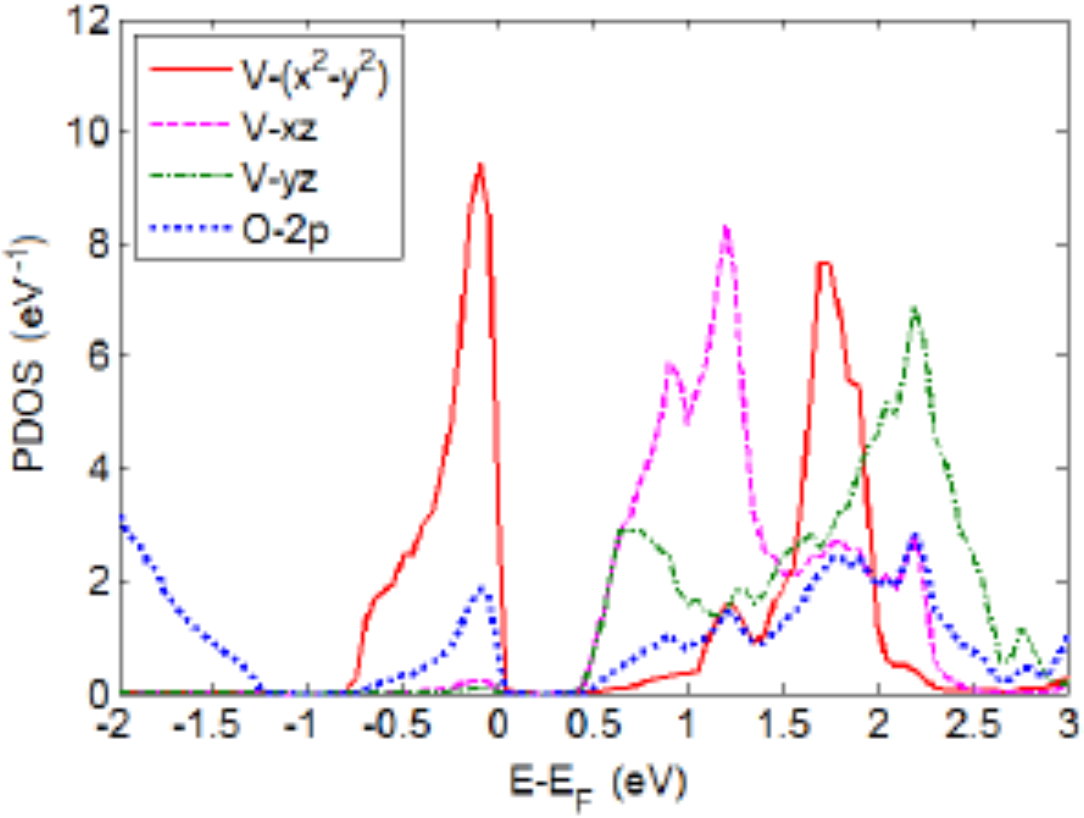
States at upper and lower gap edge different orbital character



?slower kinetics?

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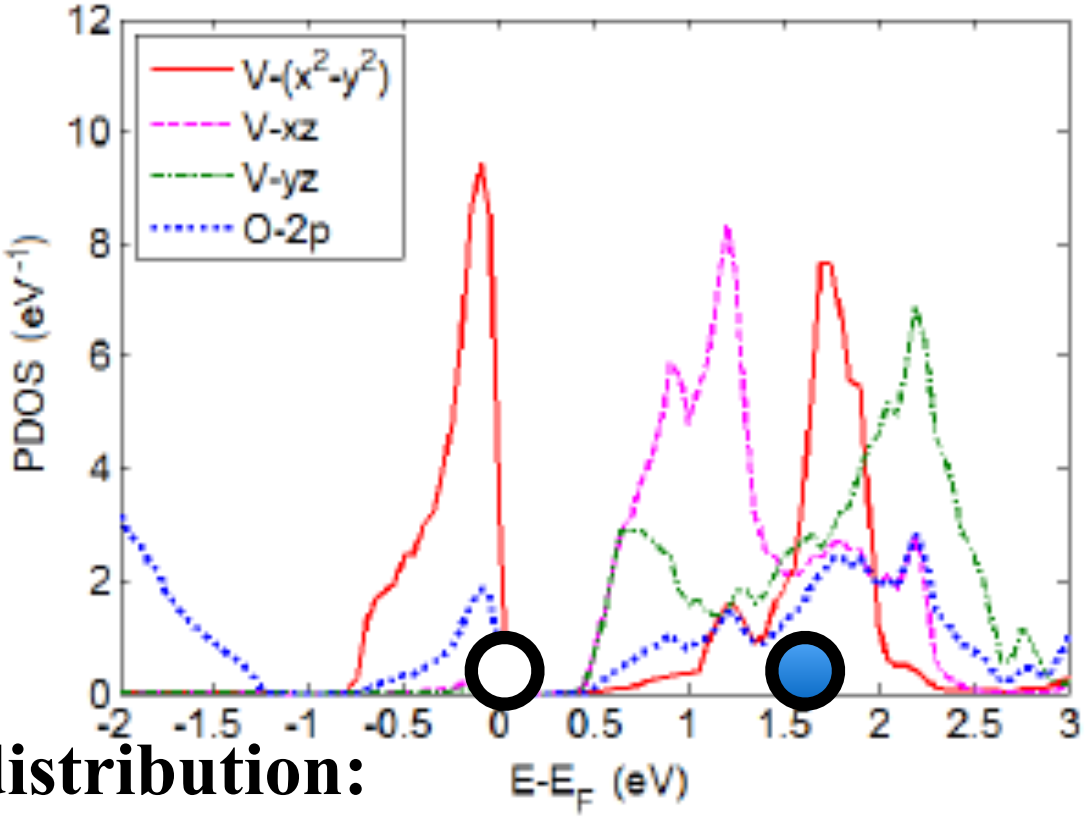
States at upper and lower gap edge different orbital character



Initial distribution:
each band thermalizes quickly
then Auger up-scattering (slow)=>final thermalization

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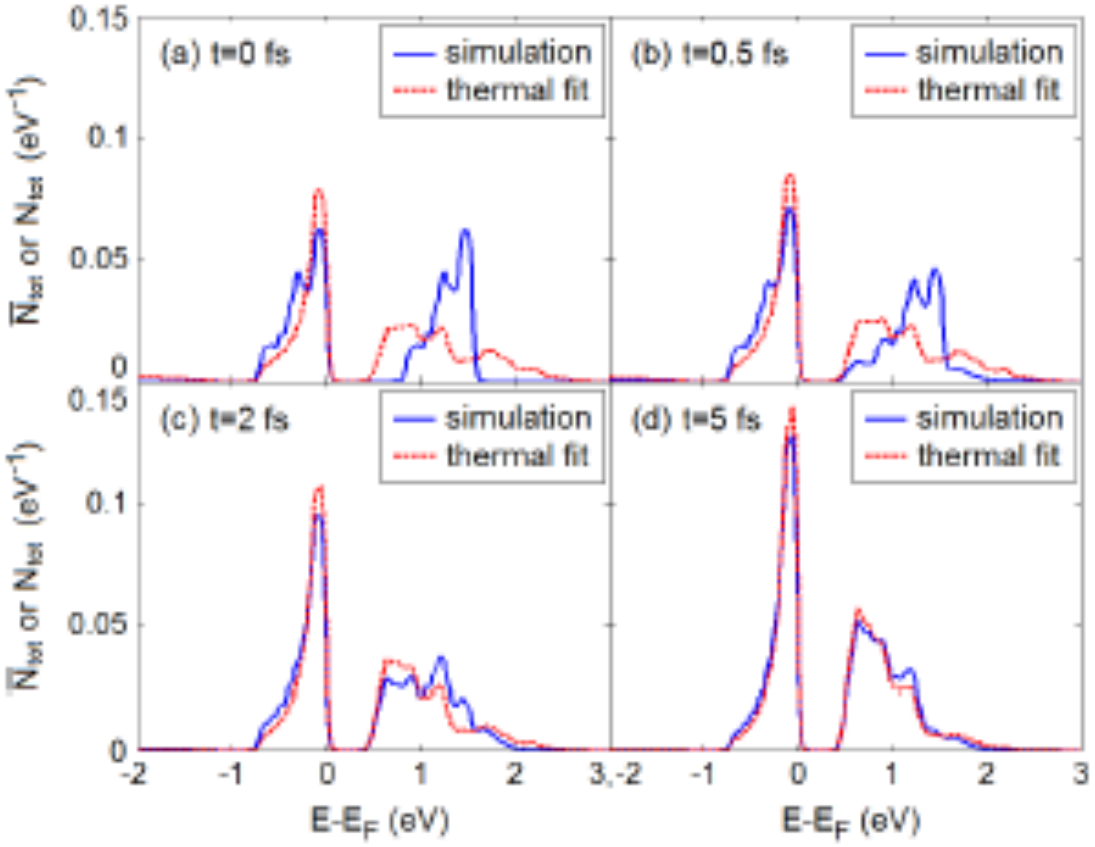
Boltzmann Kinetics
Initial thermalization, a few fs



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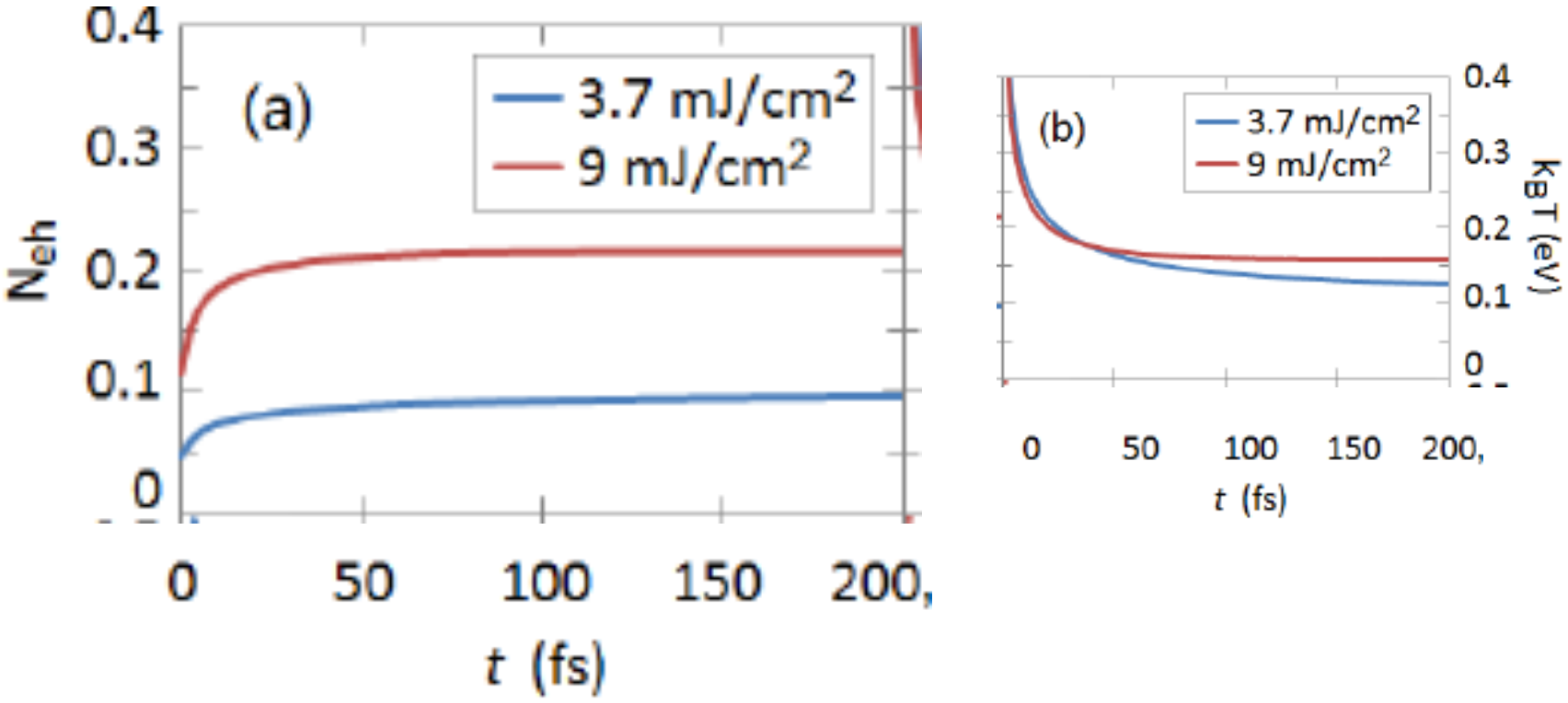
Boltzmann Kinetics: Auger upscattering final thermalization



Slower timescale for final relaxation: J

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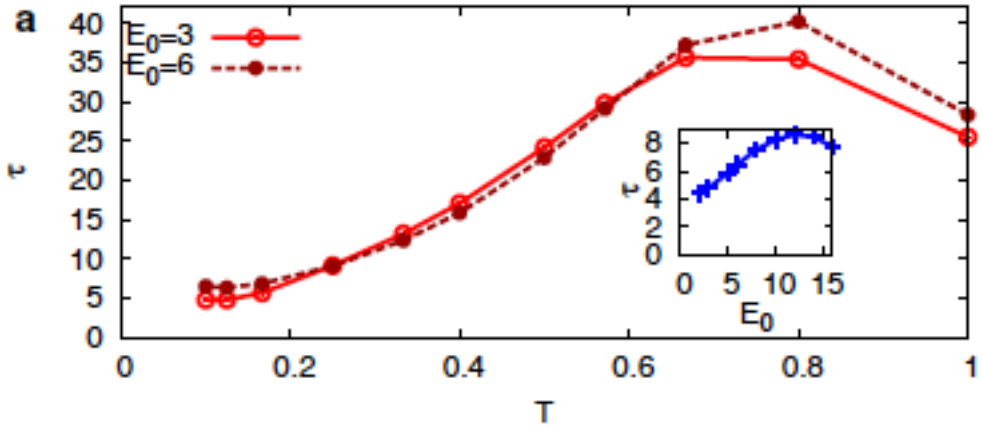
Key question:
how fast does energy go out of system?

Phonon

$$\frac{\hbar}{\Sigma''} \sim \frac{1}{\omega_D} \lesssim 100\text{fs}$$

magnons
same timescale



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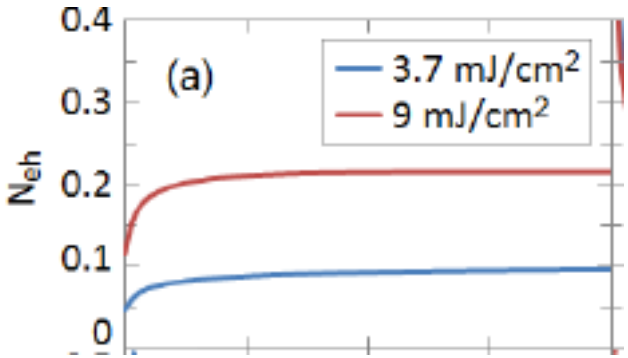
Key question:
how fast does energy go out of system?

Phonon $\frac{\hbar}{\Sigma''} \sim \frac{1}{\omega_D} \lesssim 100\text{fs}$

As upper and lower bands emit phonons and cool recombination should happen also on 10-100fs scale



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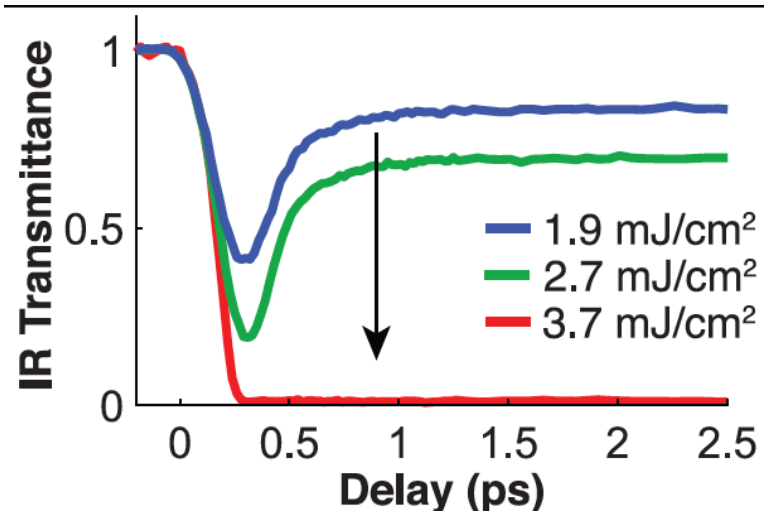
Kinetic estimate

Relaxation in at most few picoseconds

?How does the new metallic state live so long?



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Intriguing possibility
(not favored by current parameters)

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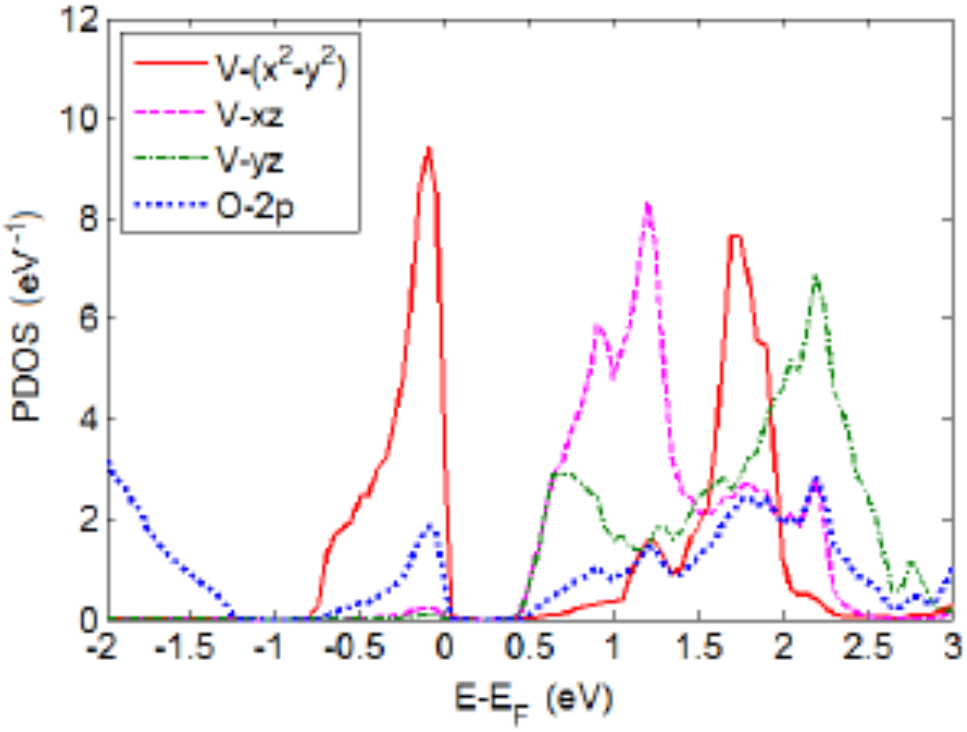
VO2: Insulating phase

Hartree-Fock band structure Z. He



Insulating phase
2 moving parts

Interaction-enhanced dimerization: V

Interaction-driven crystal field splitting on-site U

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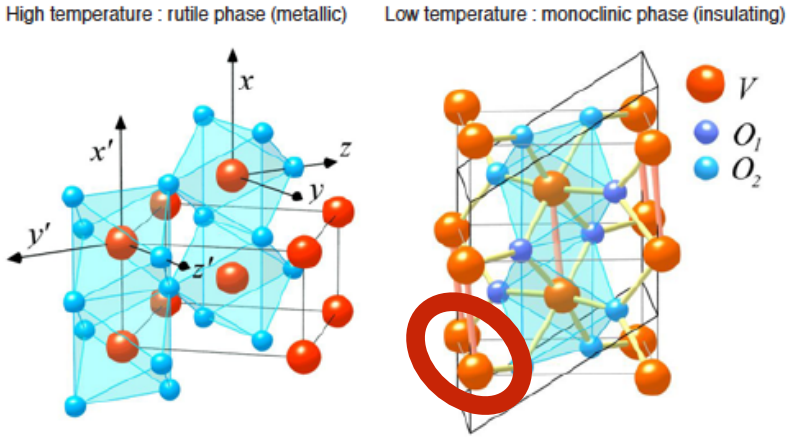
Hartree-Fock Hamiltonian

On-site U: Slater-Kanamori



The metal-insulator transition is accompanied by a structural transition with dimerization of the V atoms and tilting of the pairs out of the z axis.
(From V. Eyert, Ann. Phys. (Leipzig) 11, 650-702 (2002))

$$\mathrm{V} n_{1,x^2-y^2} n_{2,x^2-y^2}$$

$$\rightarrow \mathrm{V} c^{\dagger}_{1,x^2-y^2} c_{2,x^2-y^2} \left\langle c^{\dagger}_{2,x^2-y^2} c_{1,x^2-y^2} \right\rangle$$

Similar to J. Phys.: Condens. Matter 19 (2007) 365206

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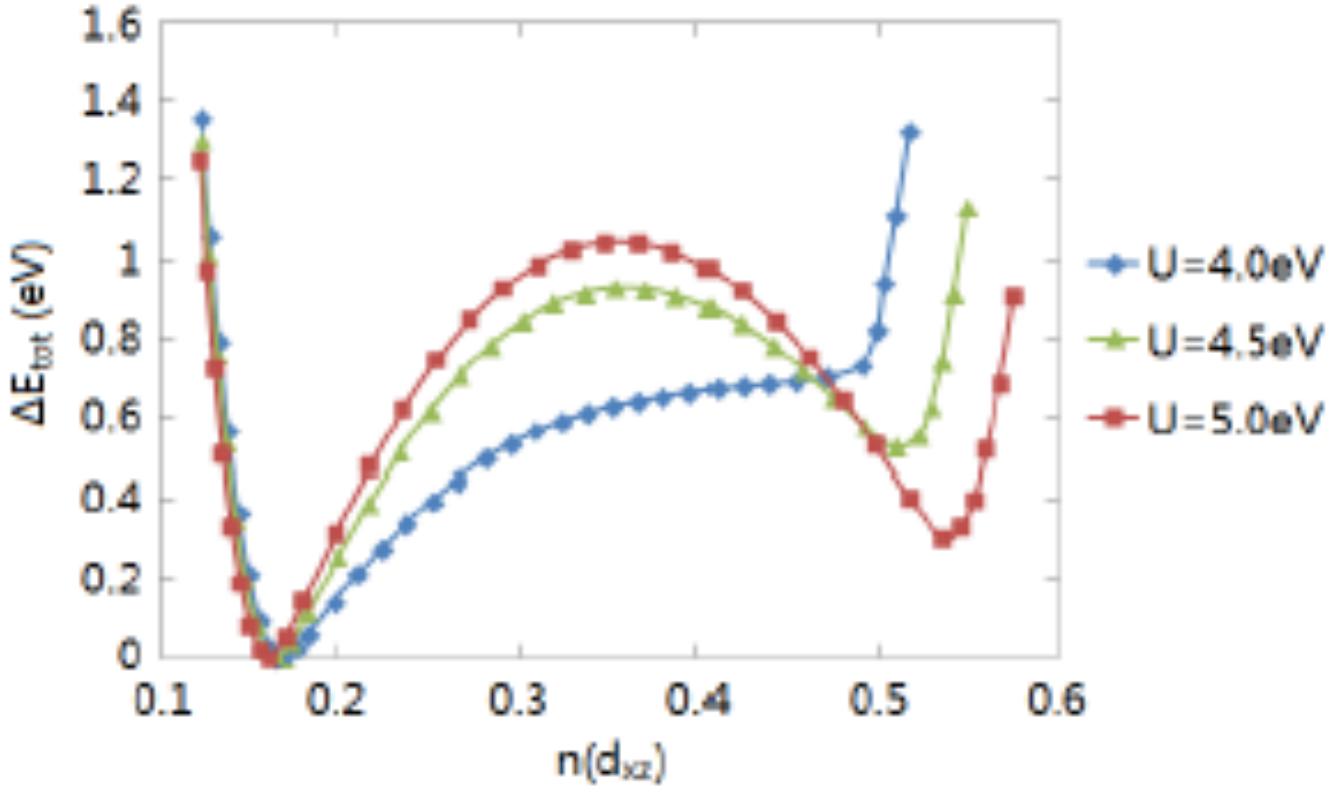
At T=0: Two extrema



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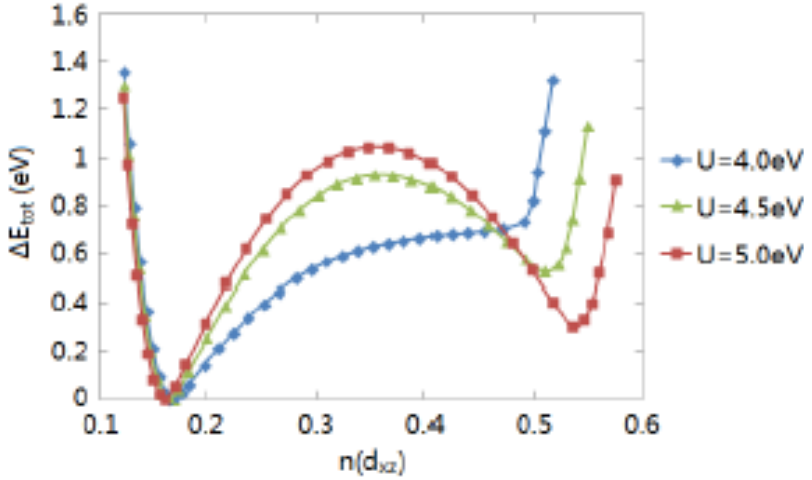
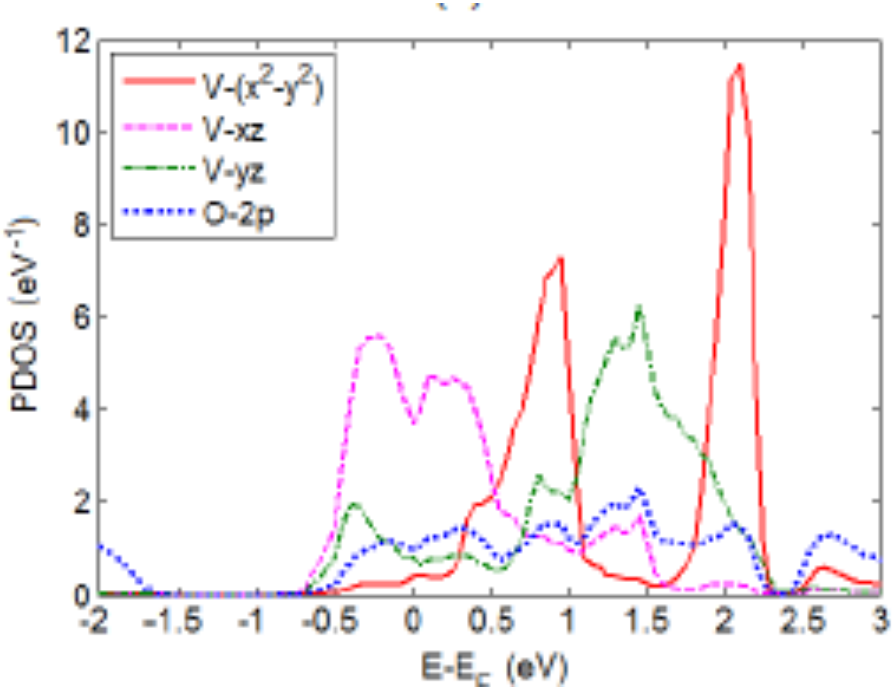
Higher extremum: metallic





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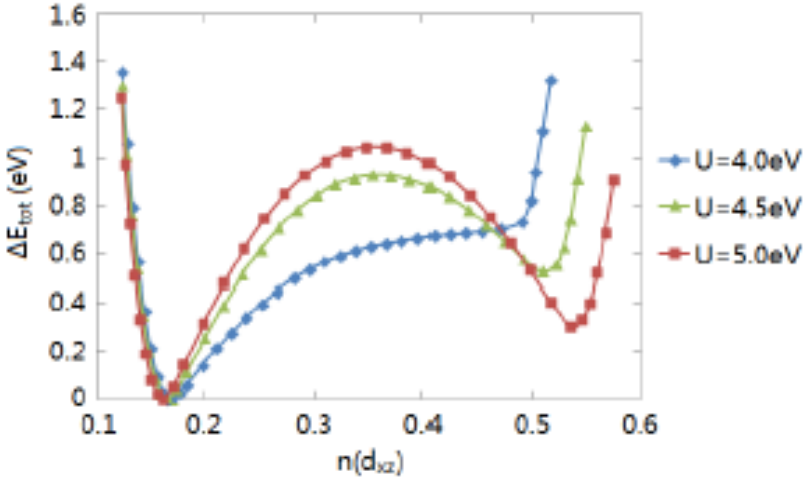
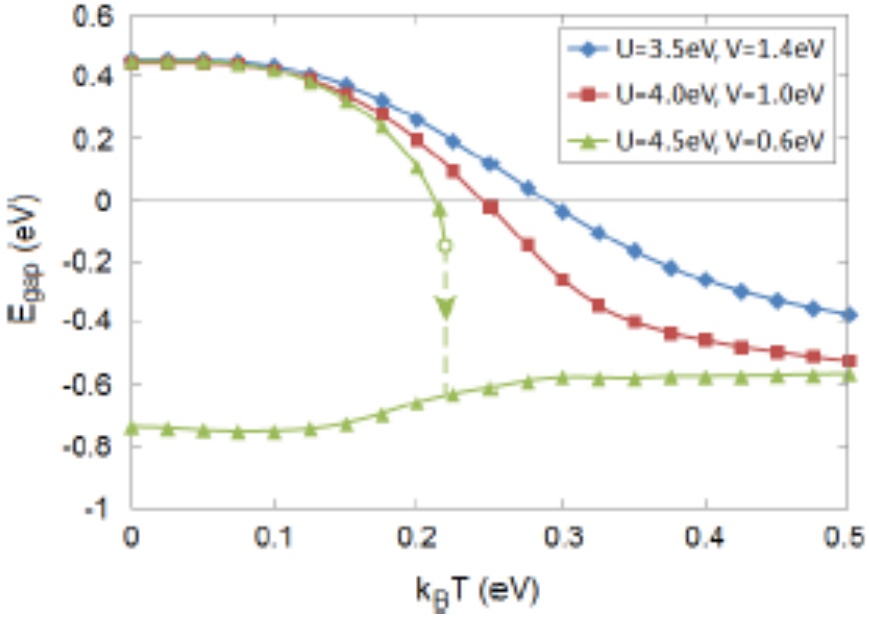
As raise electronic T, higher extremum may be favored





Note: lattice assumed to remain at low T

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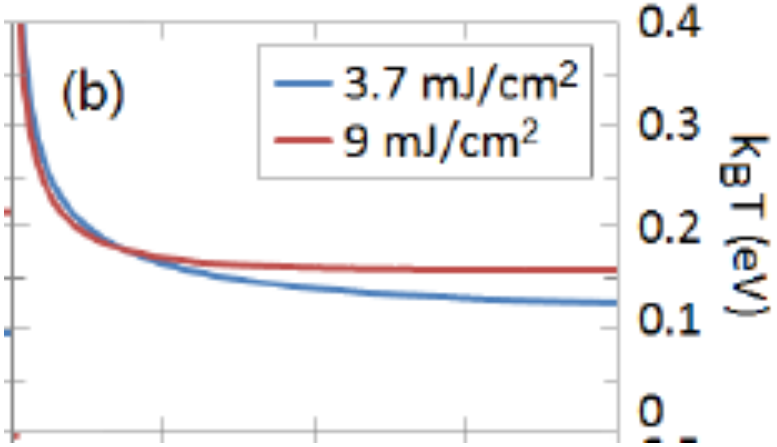
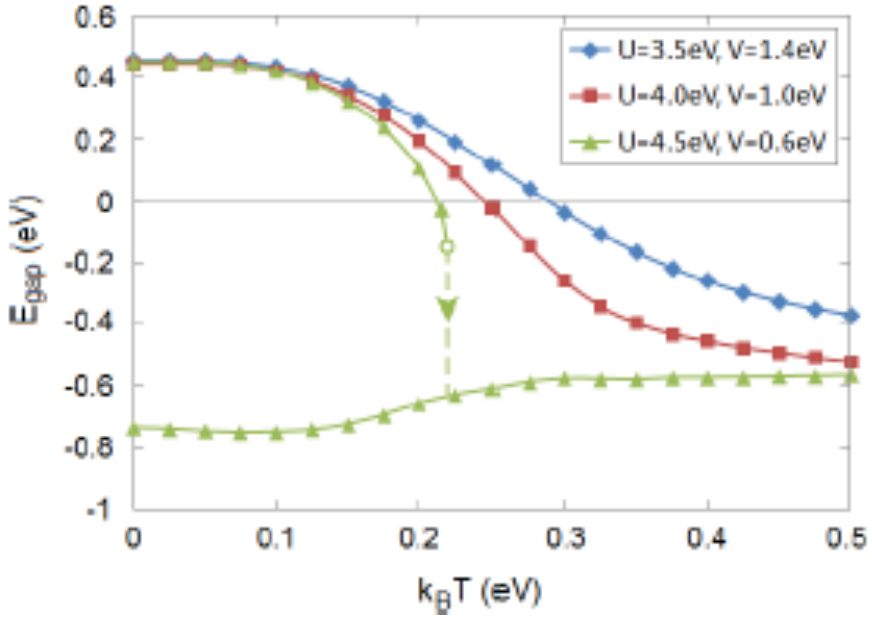
Our estimates:
fluence too low by factor of at least 2





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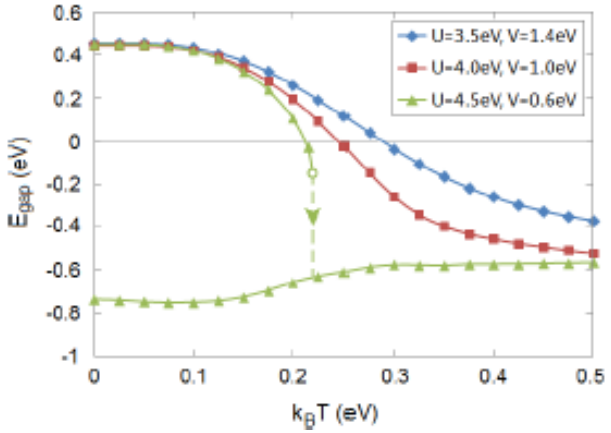
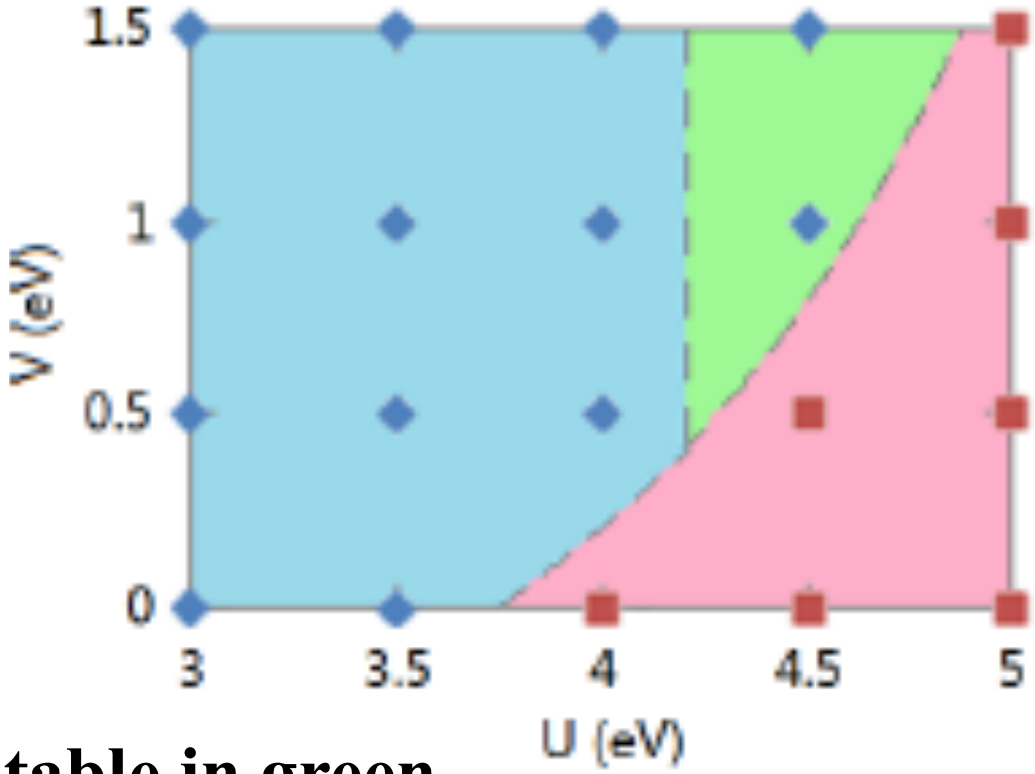
Stability of metallic phase





Metal phase metastable in green and red shaded regions

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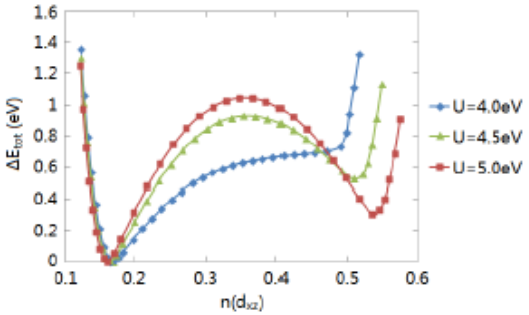
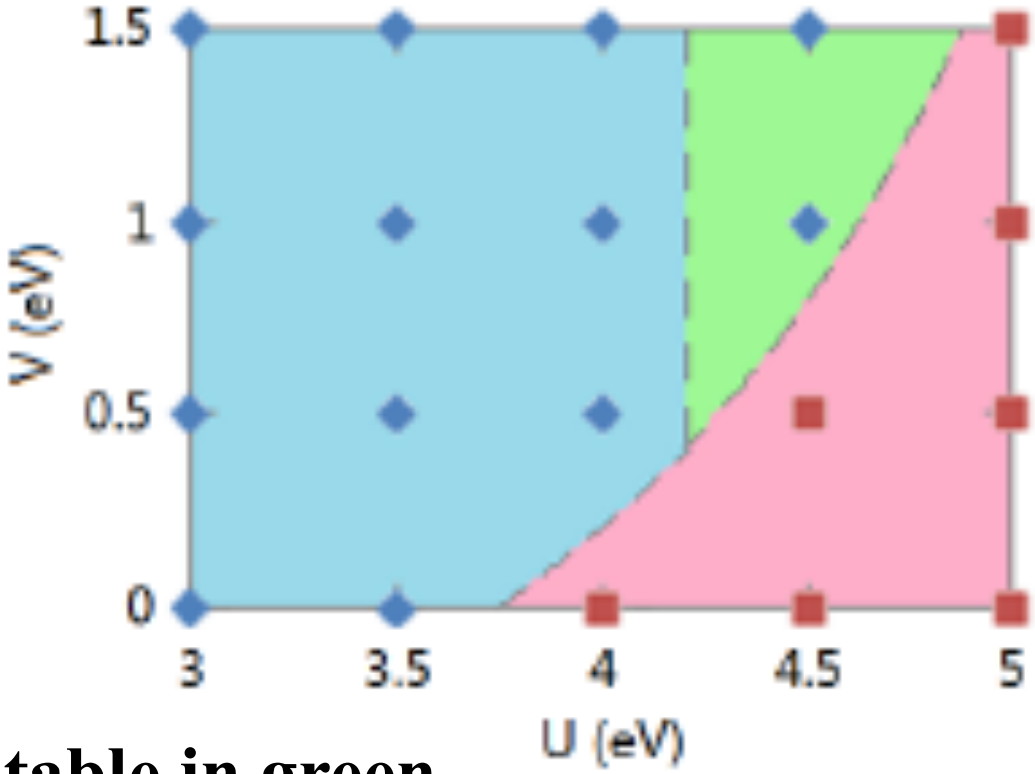
Stability of metallic phase





Metal phase metastable in green and red shaded regions

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Suggestion

Experiment has put the system into a metastable phase.

Open question: why would the lattice not relax

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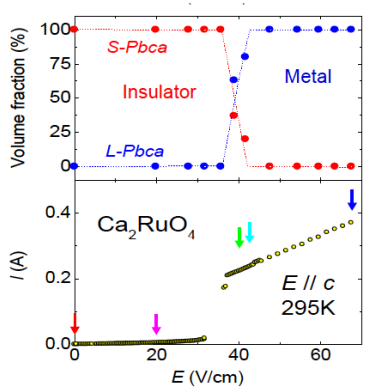
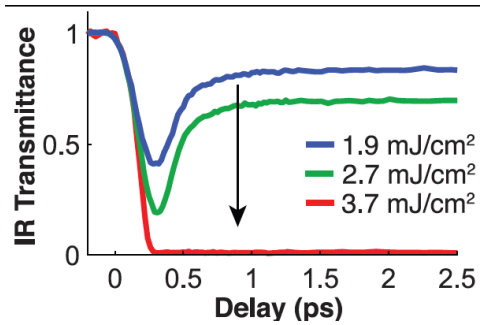
Summary





- **Current-driven and optical pump experiments imply new (at least metastable) nonequilibrium phases**
- **Present-day theory provides important insights but is inadequate to describe experiments**

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Key ideas

- **Nonequilibrium phases: effective temperature of order parameter decoupled from electronic and lattice temperature**

- **Metastable phases may be accessed by optical excitation**

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