Ultrafast laser control of the magnetic exchange interaction

ISSP Kashiwa, June 26, 2014

Martin Eckstein

Collaborations:


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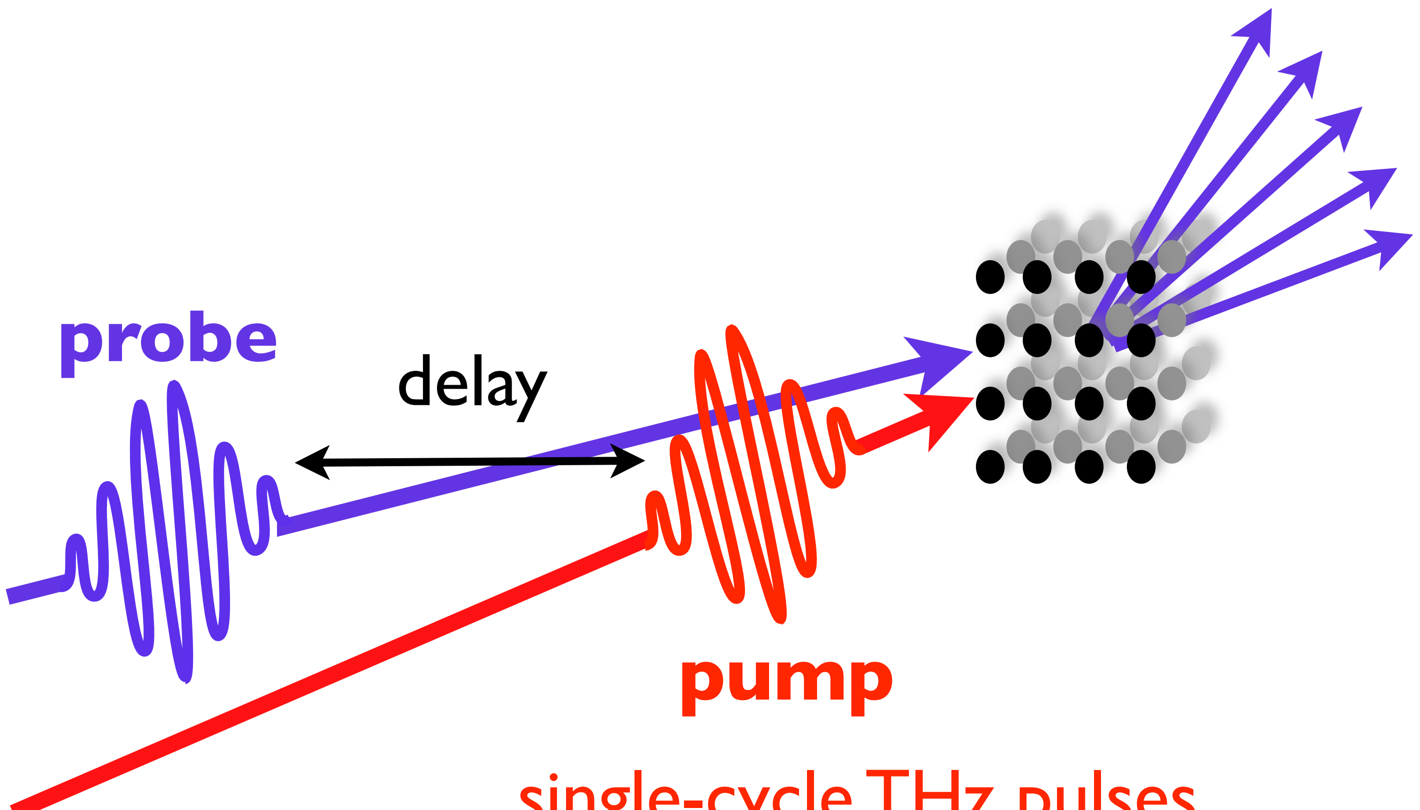
CFEL Hamburg

Philipp Werner Uni. Fribourg
Naoto Tsuji Tokyo

Pump-probe experiments



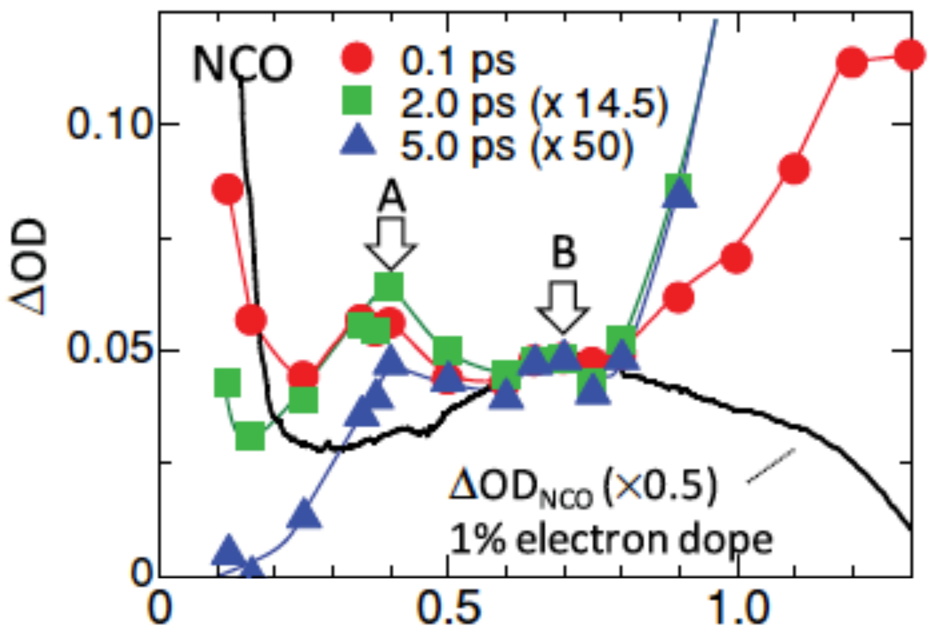
single-cycle THz pulses,
fs optical pulses, ...

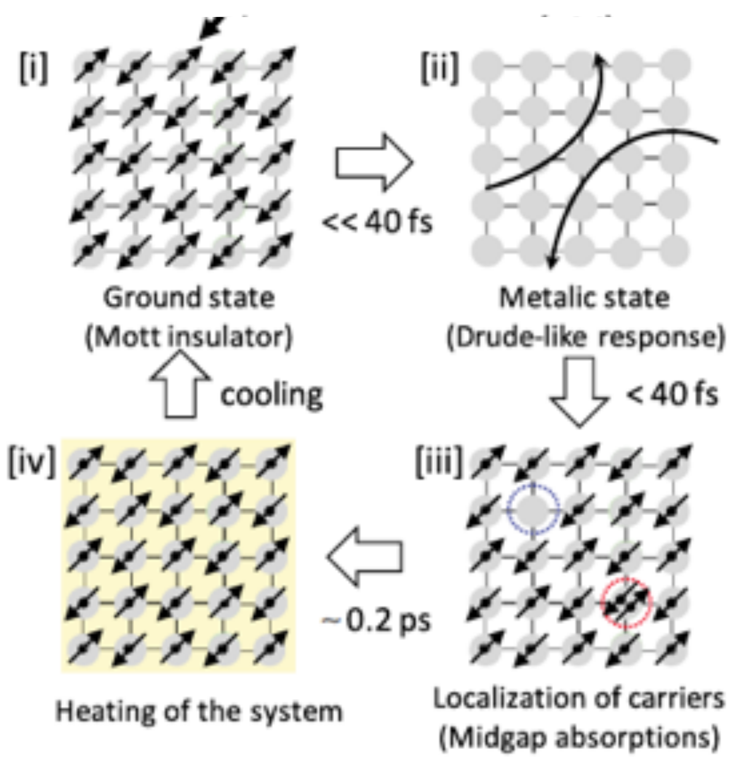
time-resolved ...

- photo-emission
- optical spectroscopy
- electron difraction
- X-ray scattering
- RIXS

Many-body systems out of equilibrium

Relaxation/Thermalization



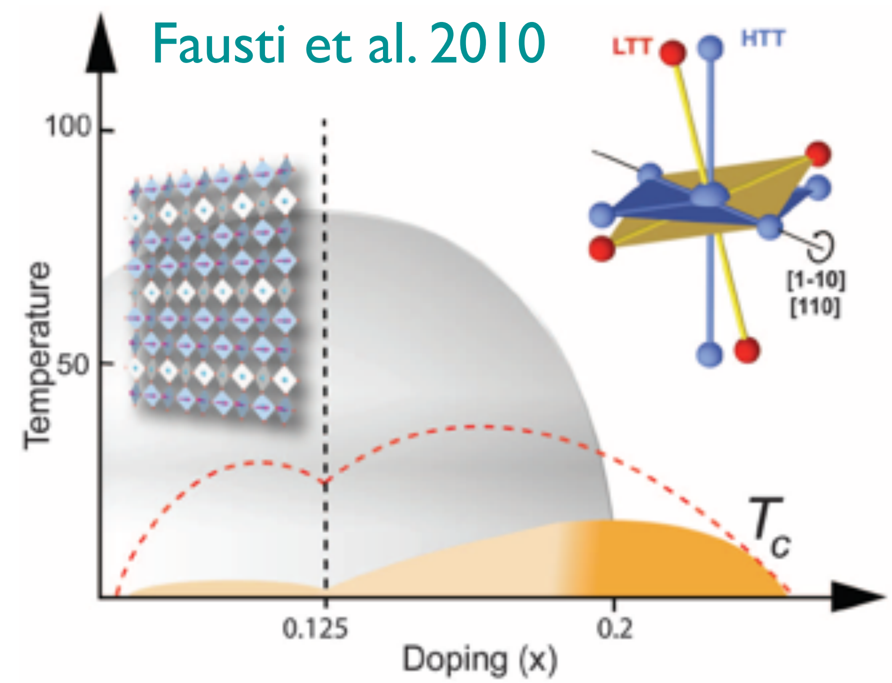


Okamoto et al, 2011

Relaxation:
→ Probing interactions?
→ Nonequilibium quasi-particles?

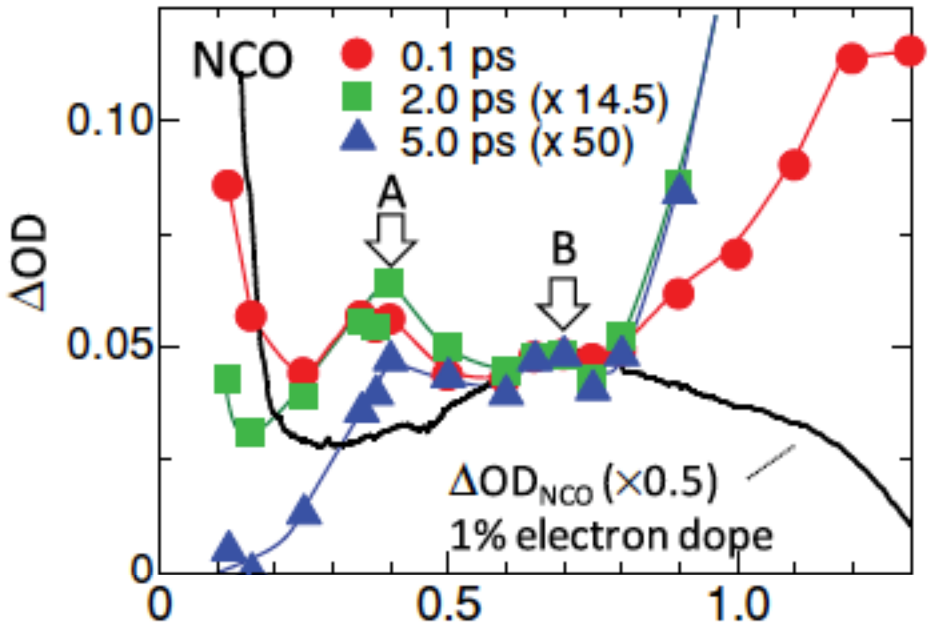
„Control“

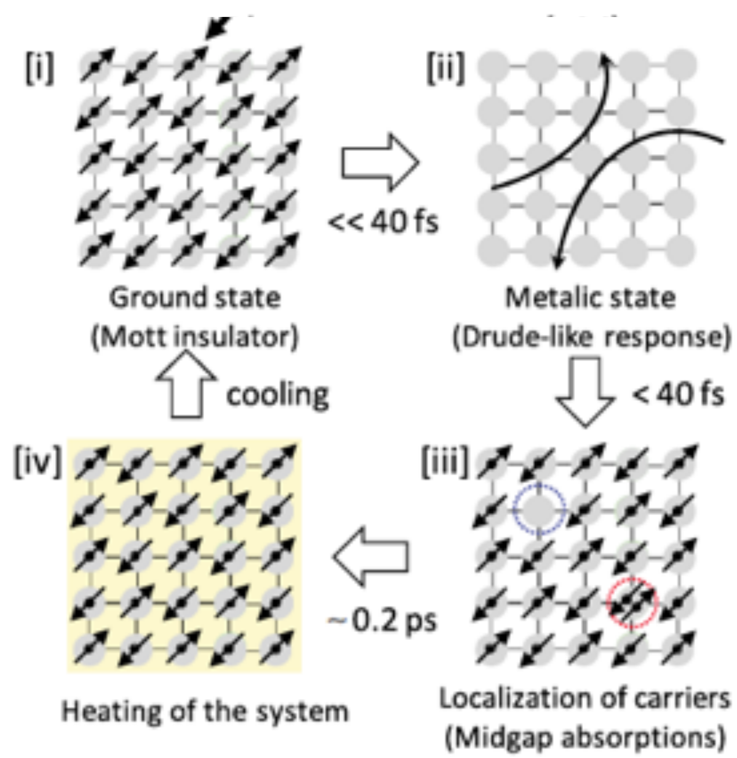
→ Nonthermal transition pathways?
→ Metastable states? Hidden states?
→ Driven states?



Many-body systems out of equilibrium

Relaxation/Thermalization





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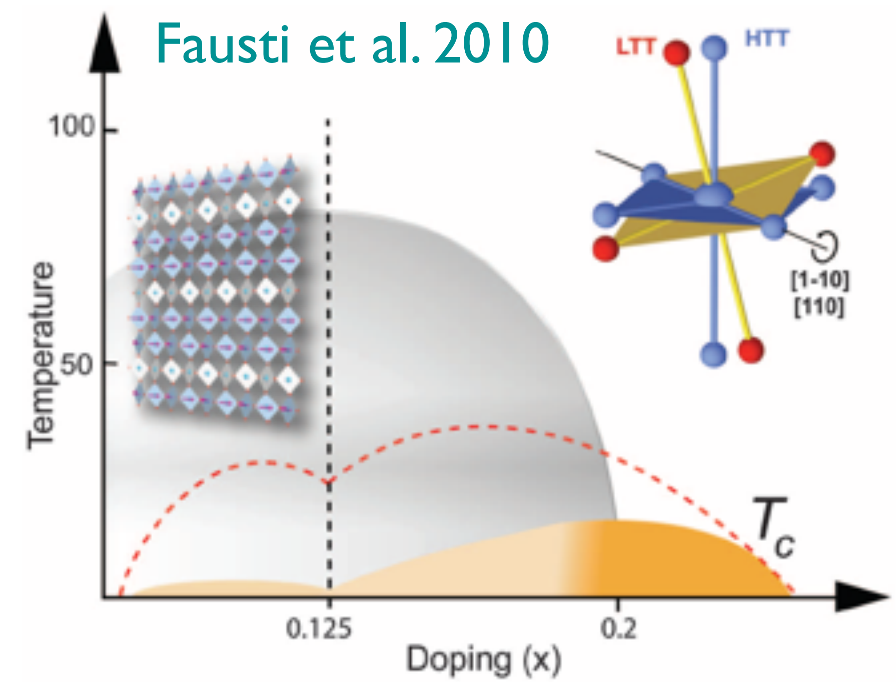


Optical control of magnetism?

Kirilyuk, Kimel & Rasing, RMP 2010

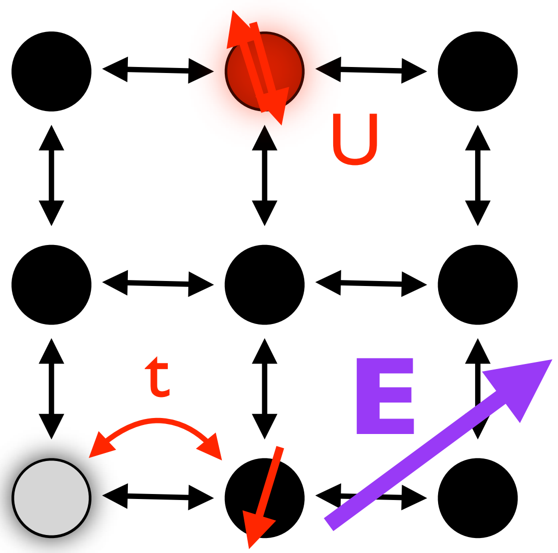
➙ Controlling exchange interactions?

$LaMnO_3$ Wall, Prabhakaran, Boothroyd & Cavalleri, 2009

$Eu_{1-x}Gd_xO$ Matsubara et al. arXiv 1304:2509

#Nonequilibrium DMFT

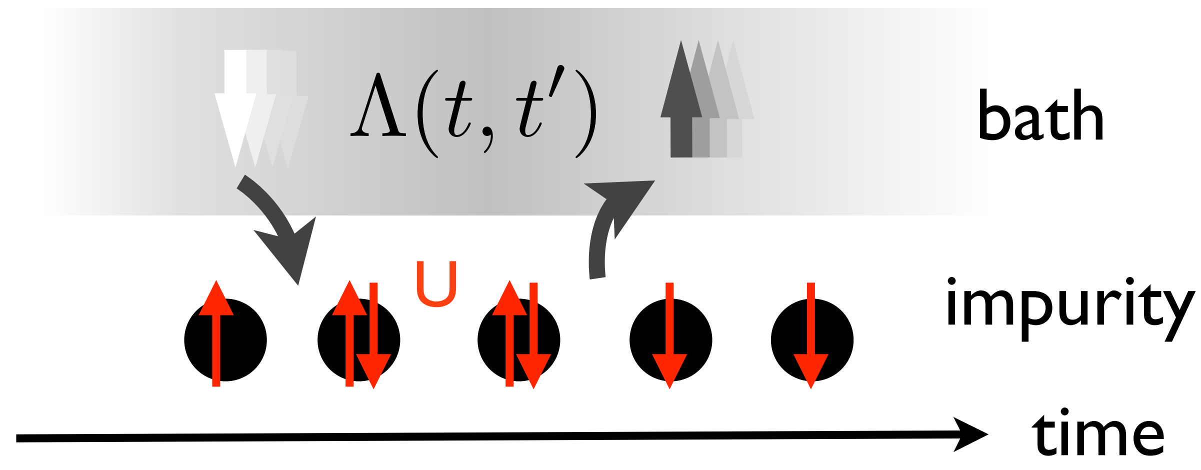
$$H = -t \sum_{\langle ij \rangle \sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$ +External field E(t)



Metzner & Vollhardt 1989
Georges et al, RMP 1996

DMFT

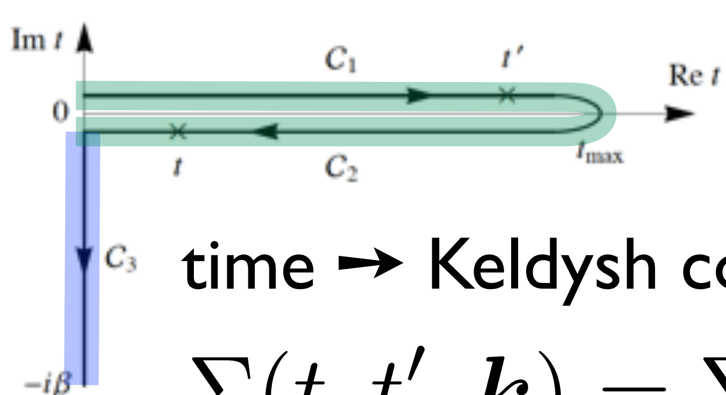
$$\Sigma(\omega, \boldsymbol{k}) = \Sigma(\omega)$$



one „impurity problem“ per site,
coupled by self-consistent bath

→ Nonequilibrium DMFT

Schmidt & Monien 2002
Freericks, Turkowski & Zlatic 2006



time → Keldysh contour

$$\Sigma(t, t', \boldsymbol{k}) = \Sigma(t, t')$$

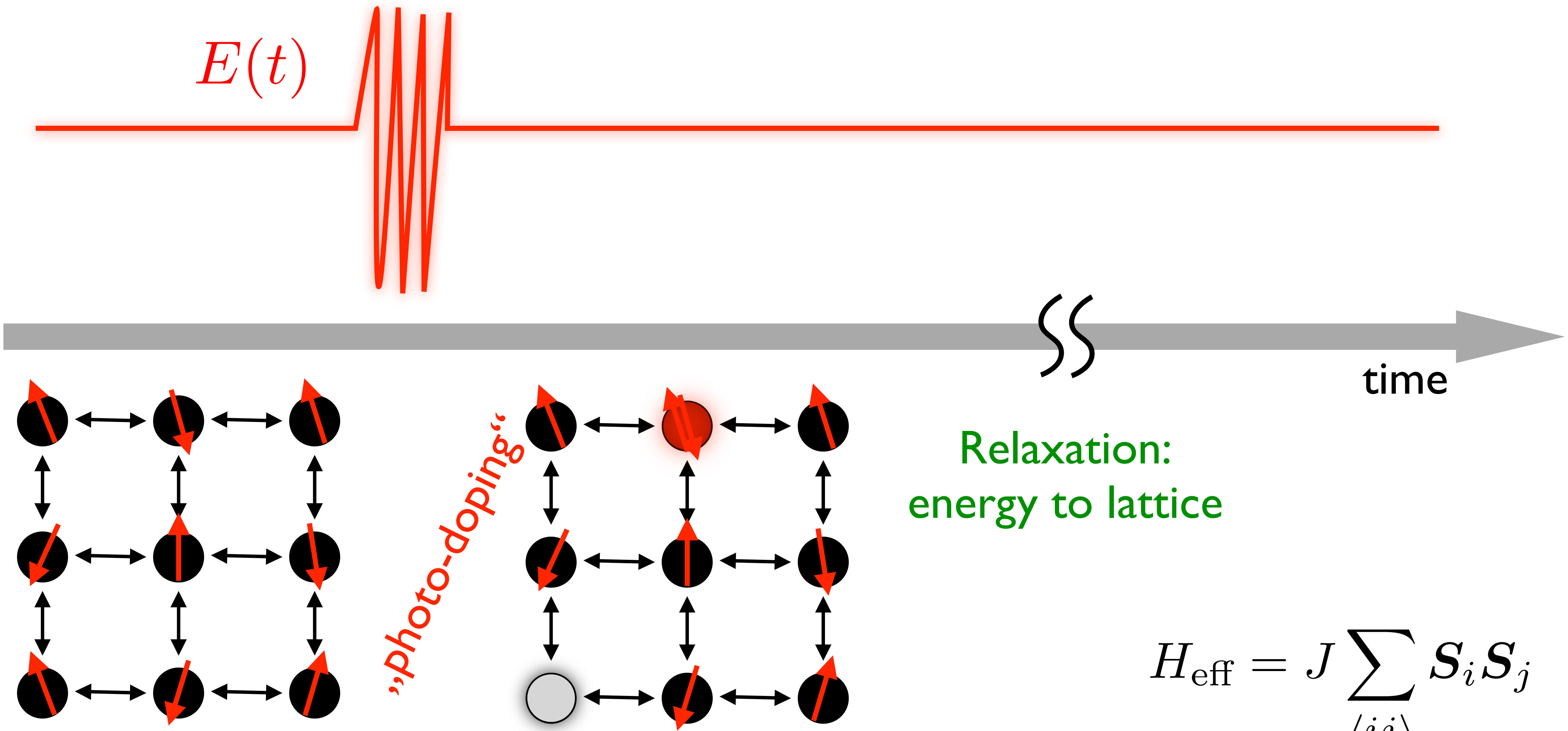
Impurity solver:

CTQMC,	Werner, Oka Millis 2009
Krylov,	Gramsch, Balzer, Eckstein, Kollar 2013
strong-coupling expansion (NCA)	Eckstein & Werner 2010
weak-coupling theory (IPT etc.)	Tsuji & Werner 2013

⇒ talk by E. Gull

Review: Aoki, Tsuji, Eckstein, Kollar, Oka & Werner, RMP 2014

Modification of J_{ex} by photo-doping



$$H_{\text{eff}} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j$$

$$J_{\text{eff}} = ?$$

$$H_{\text{eff}} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j$$

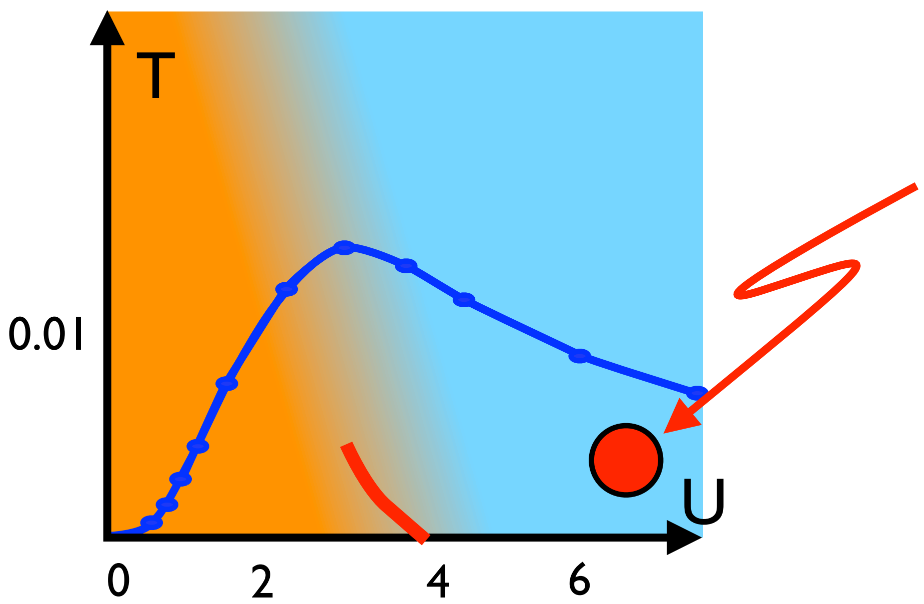
$$\boxed{J_{\text{ex}} = t^2/U}$$

What is J_{ex} in a nonequilibrium state?

How fast can it be changed ? (When is J_{ex} a useful concept at all?)

Photo-induced melting of the antiferromagnetic phase

see also Tsuji, Eckstein & Werner, 2013 (U<<t); Werner, Tsuji, Eckstein, 2012 (U>>t)

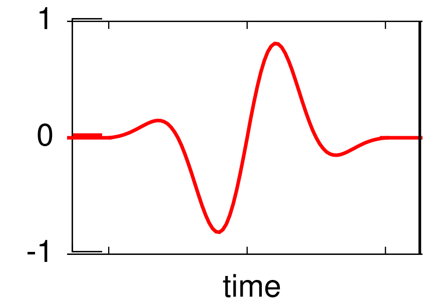


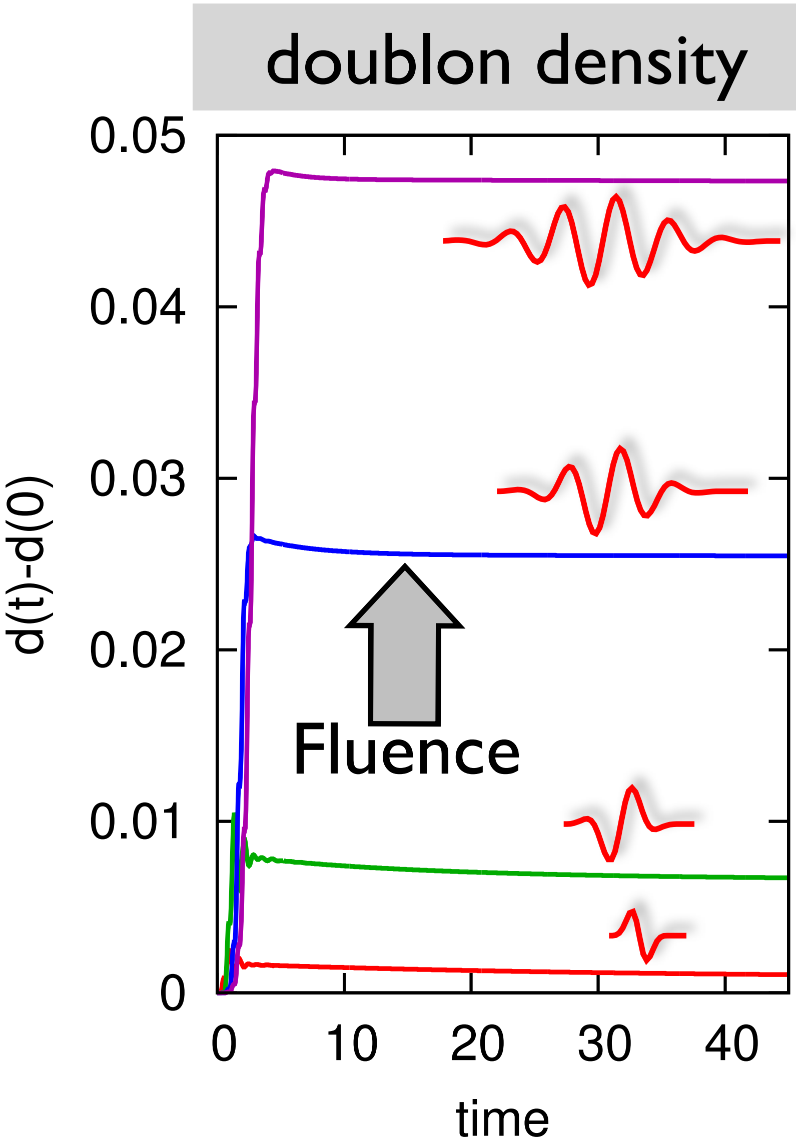
Laser excitation of *antiferromagnetic* Mott insulator

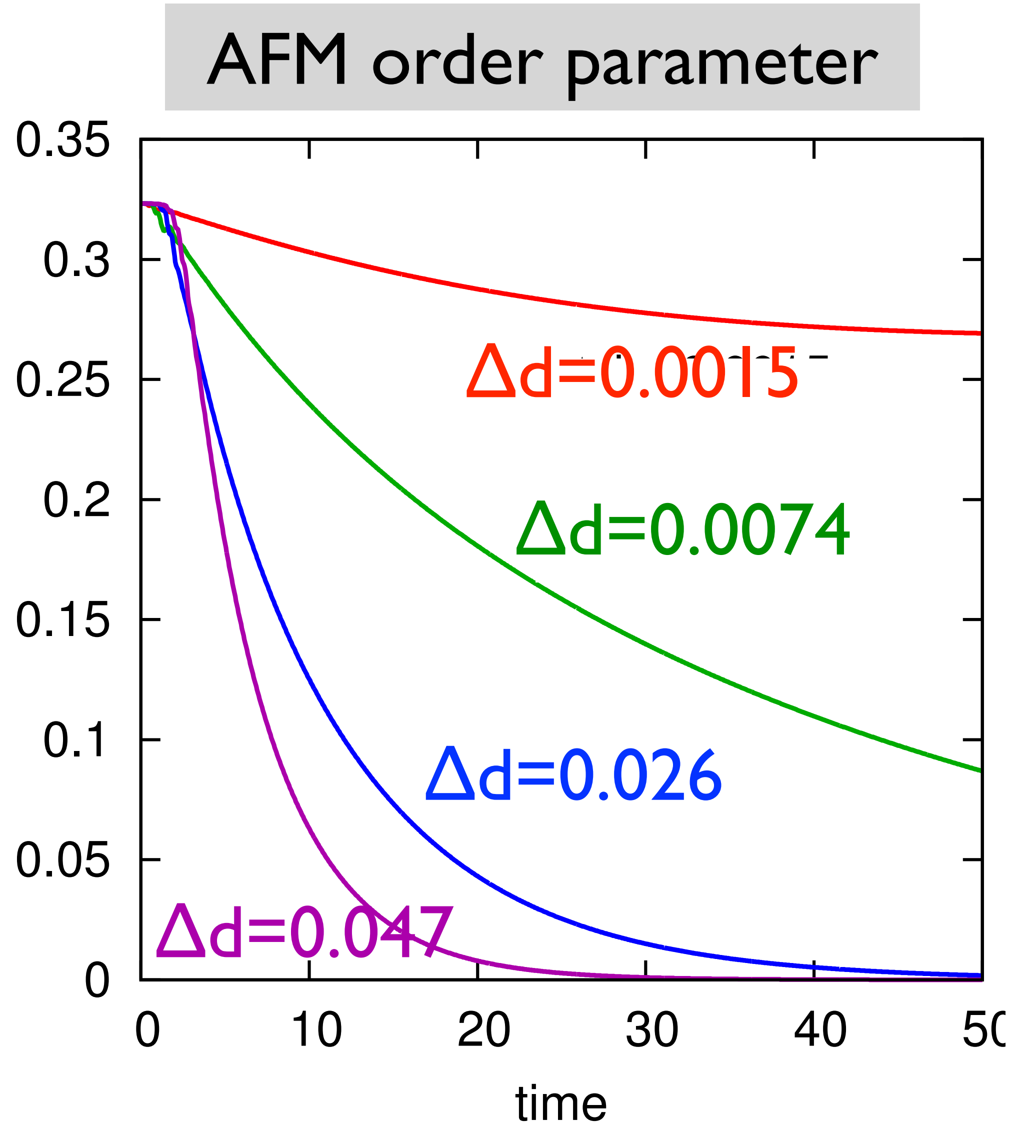
Pulse: Ω = U $\approx$ Mott gap

(hypercubic lattice, U=8)

E(t): frequency $\approx$ U







non-thermal trapped state

Fluence

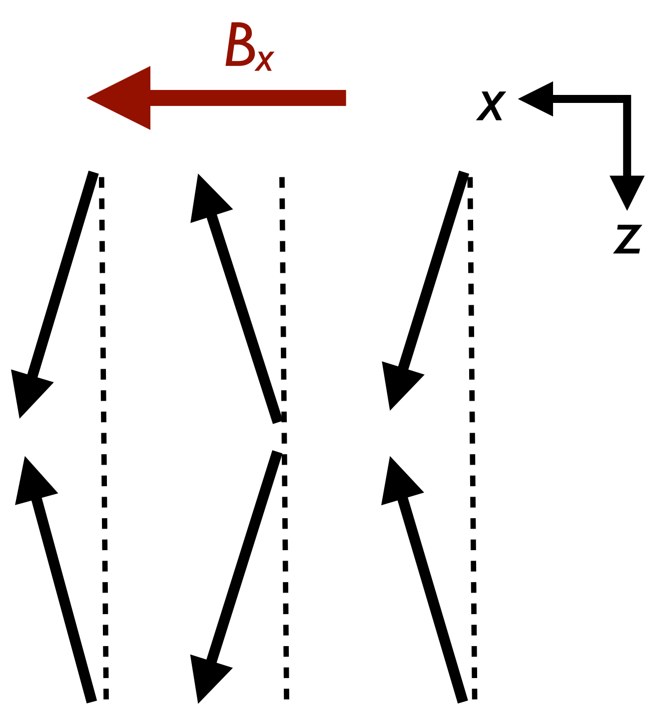
ultrafast melting:

Rigid spin dynamics

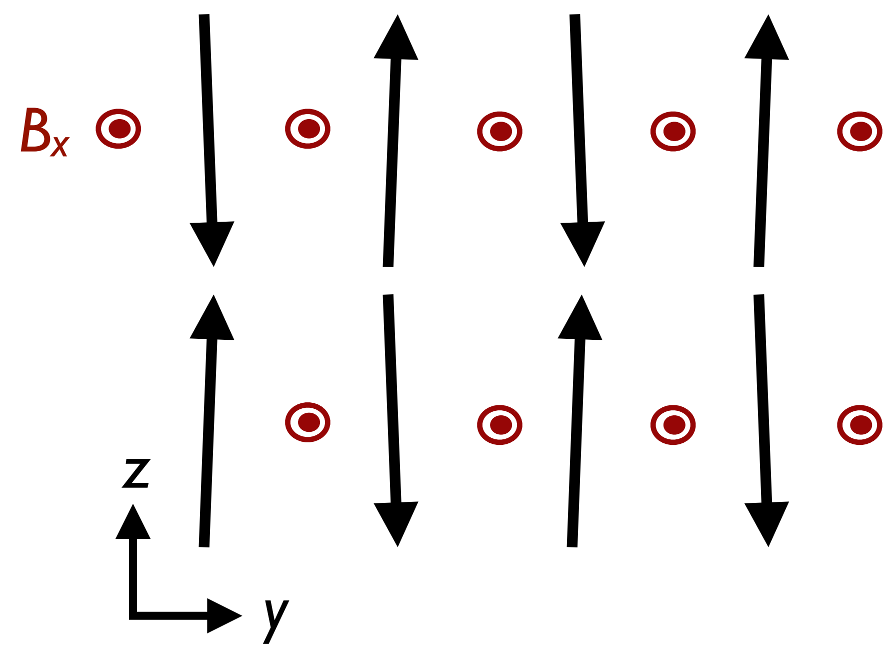
Direct „measurement“ of the exchange:

$$\frac{d\boldsymbol{S}}{dt} = -\gamma \boldsymbol{S} \times \Big(J_{\rm ex} \sum_{\text{neighbors } j} \boldsymbol{S}_j + \boldsymbol{B} \Big)$$ classical mean-field dynamics

AFM in transverse B field: equilibrium



AFM in transverse B field: dynamics



$$S_x = -\frac{B_x}{2\tilde{J}_{\rm ex}}$$

change of $J_{ex} \Rightarrow$ precession around B_x

$$\frac{dS_x}{dt} = 0 \qquad \frac{dS_y}{dt} = S_z(t) B_x \frac{\Delta J_{\rm ex}}{J_{\rm ex}}$$

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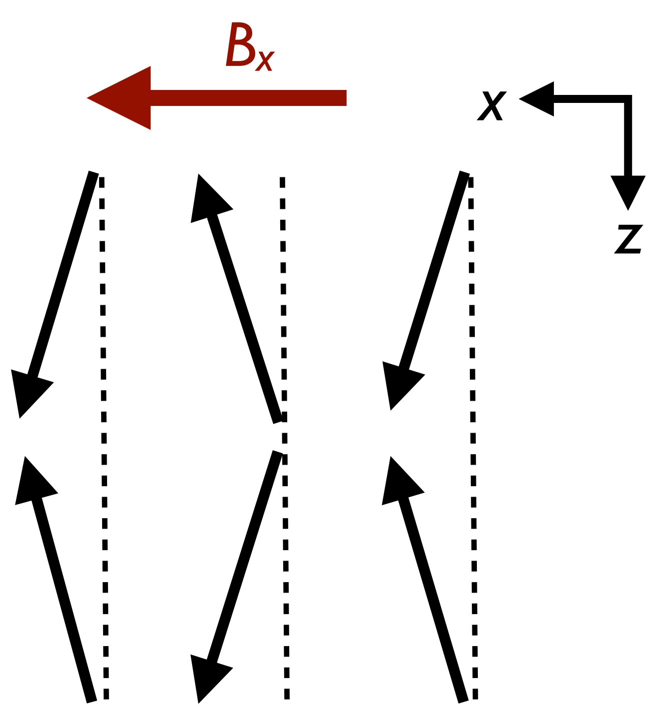

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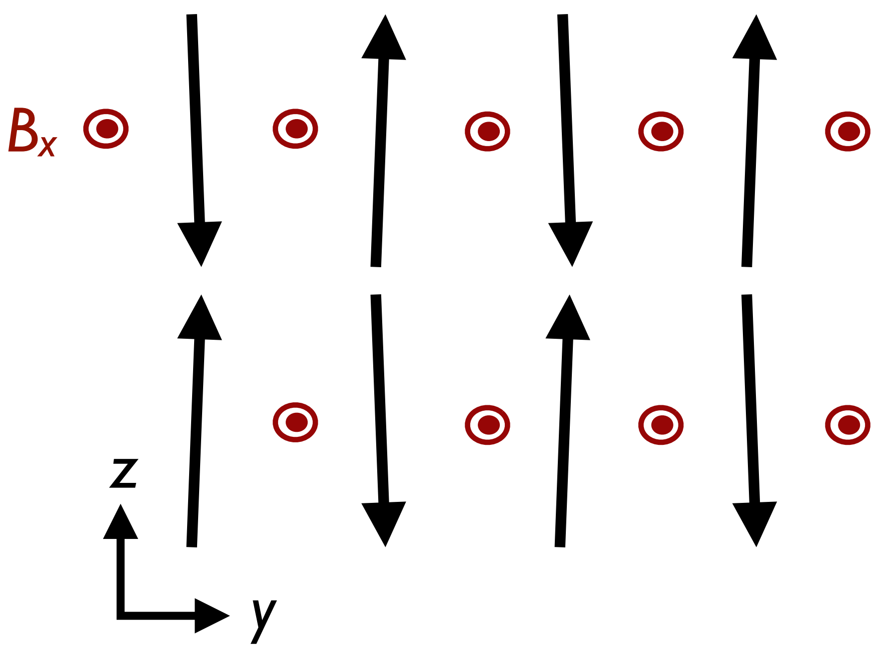

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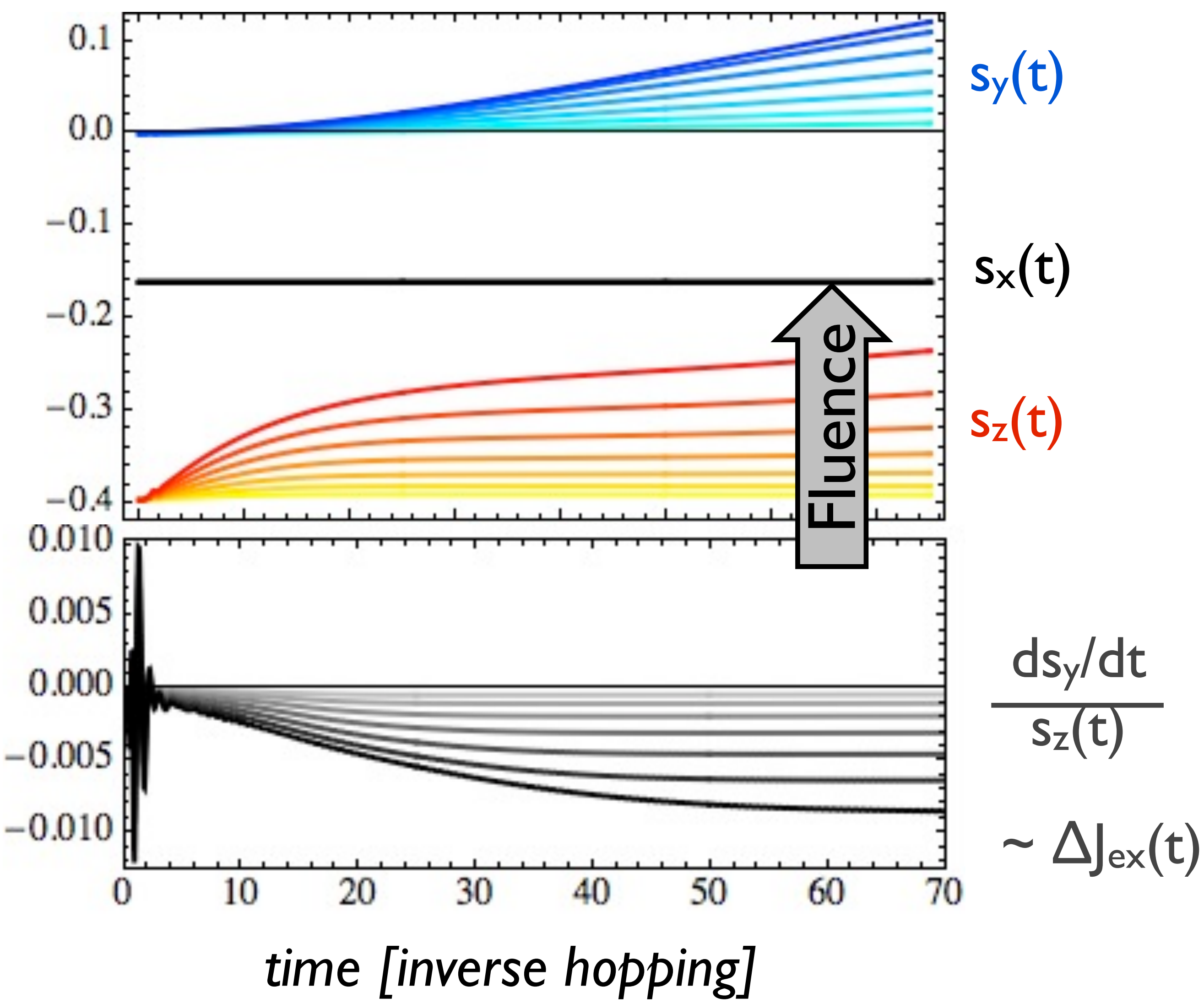
Spin dynamics in DMFT

Mentink & Eckstein, arXiv 1401.5308

DMFT: *AFM in transverse B field*

Laser Pulse: $\Omega = U \approx$ Mott gap

(hypercubic lattice, U=8)



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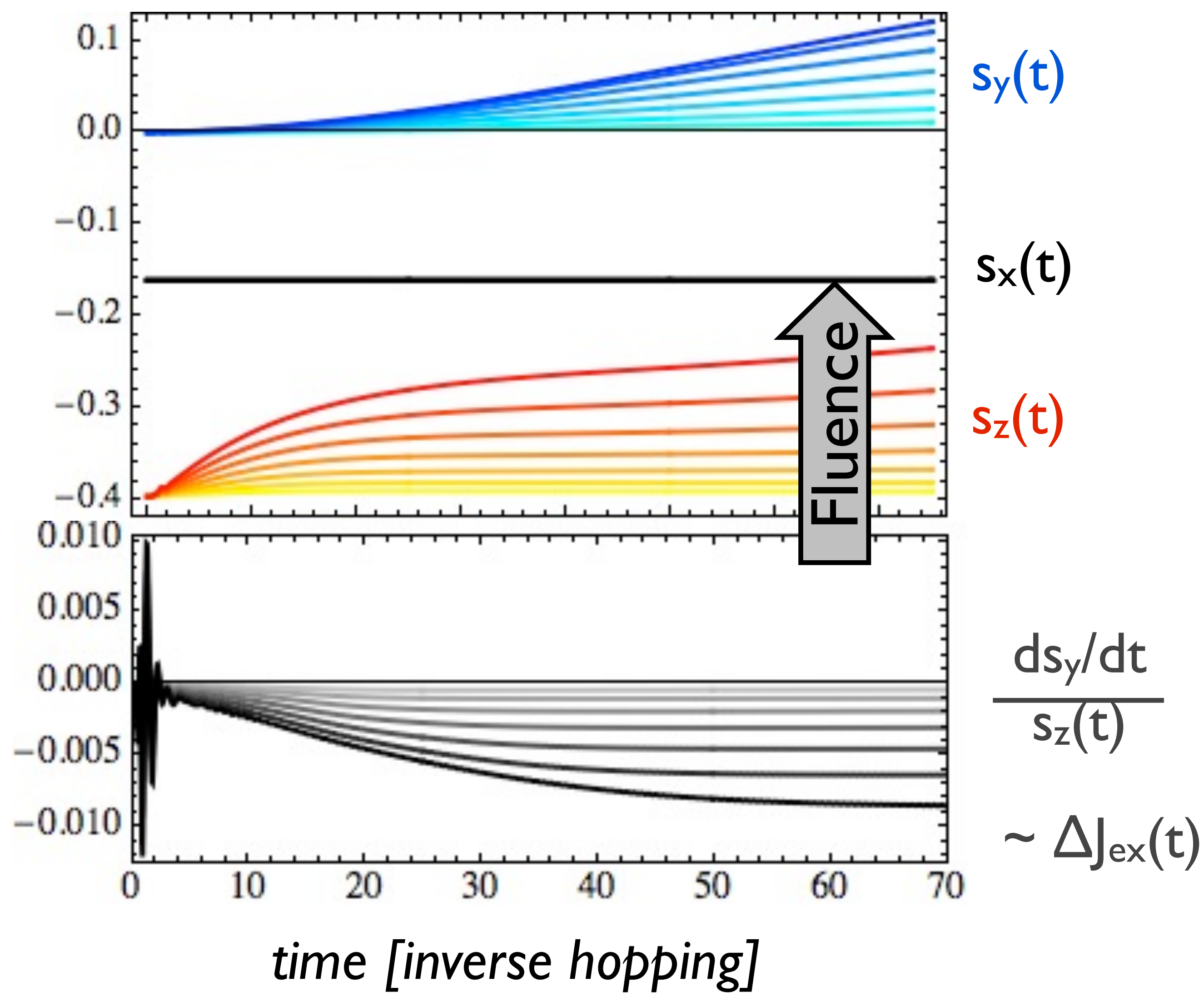
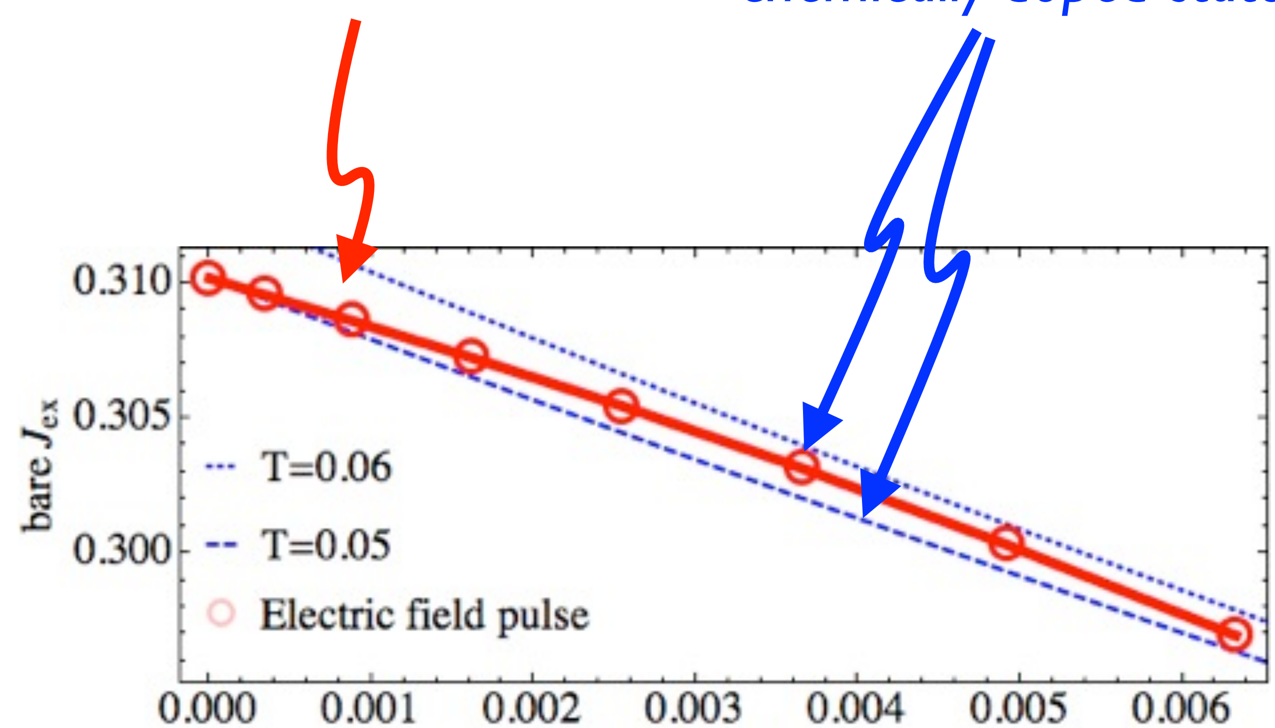


Exchange J_{ex} :



chemical doping $n_\uparrow + n_\downarrow - 1$

photo-doping $2[\langle n_\uparrow n_\downarrow \rangle - d(0)]$

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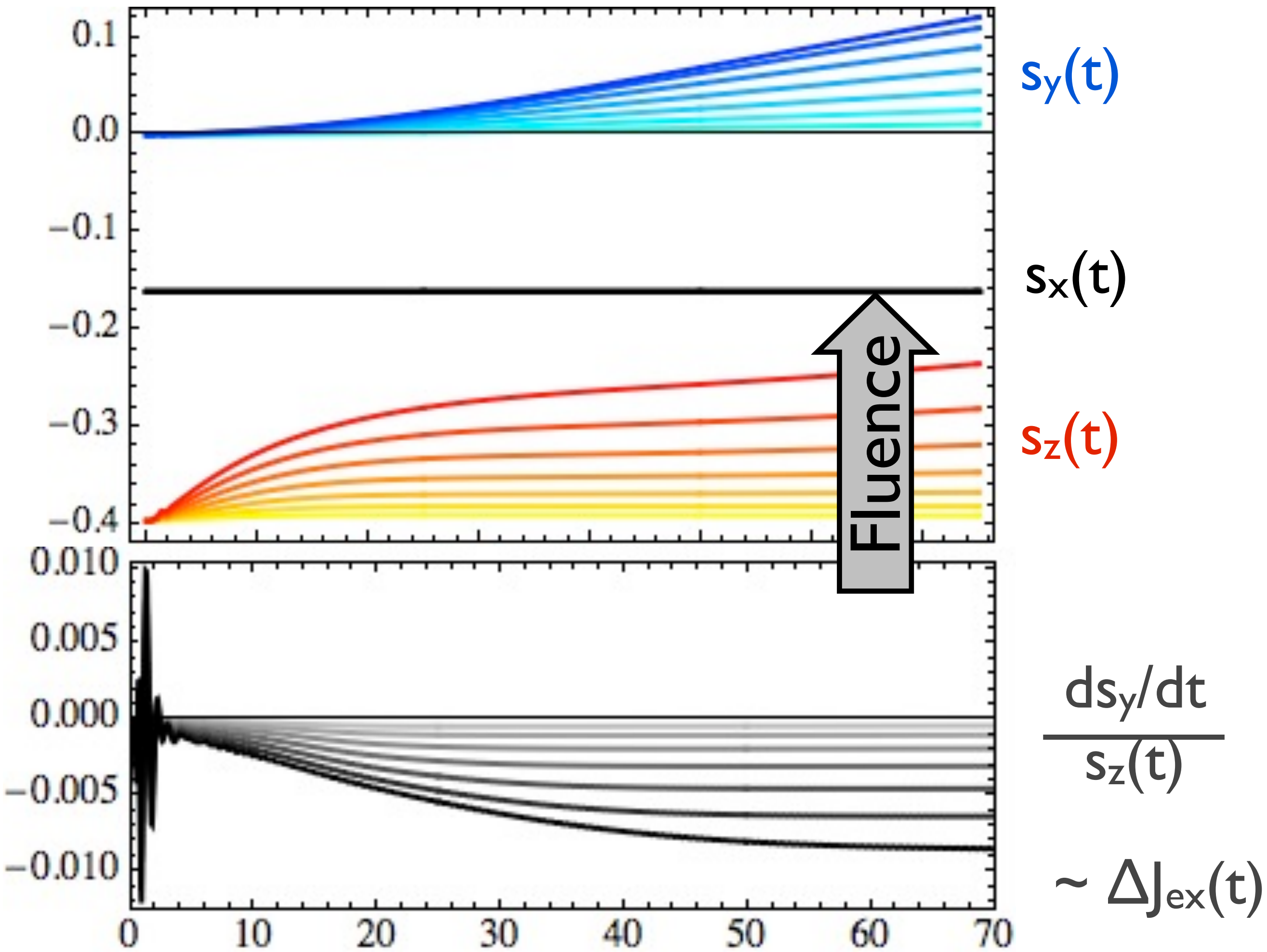
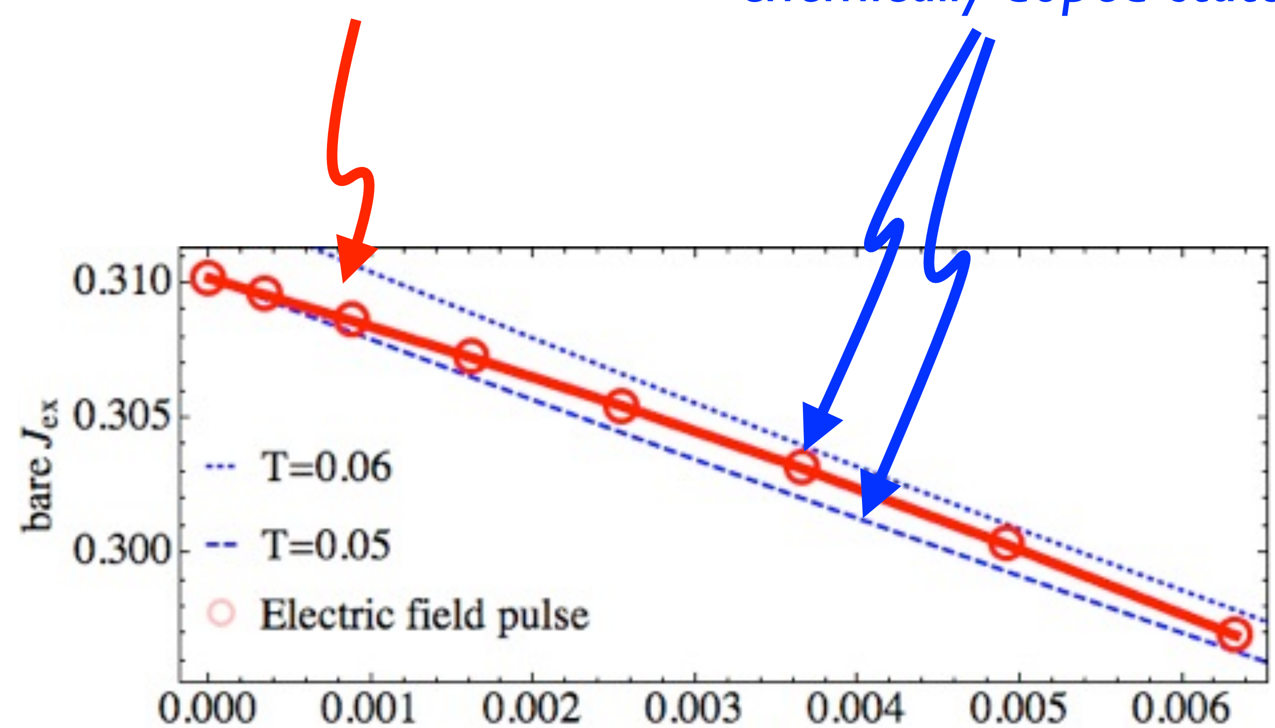


time [inverse hopping]

Exchange J_{ex} :

photo-doped state

chemically-doped state



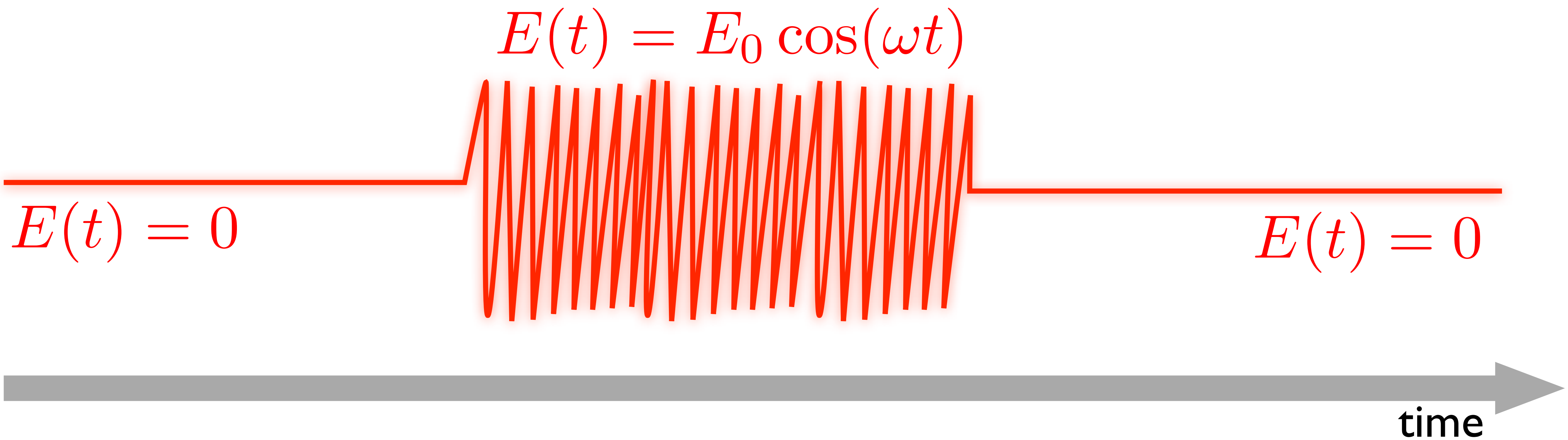
chemical doping $n_\uparrow + n_\downarrow - 1$

photo-doping $2[\langle n_\uparrow n_\downarrow \rangle - d(0)]$

Modification of J_{ex} like for chemical doping

Timescale=hopping < $1/J_{ex}$

Modification of J_{ex} by in a driven state ??

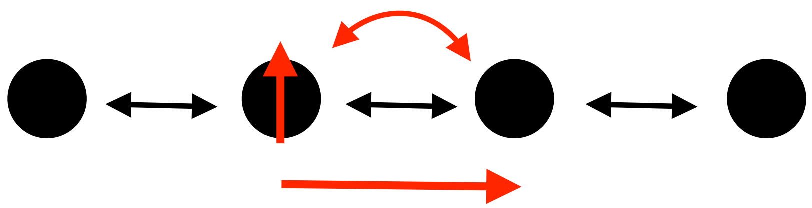


???

$$H_{\text{eff}} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j \qquad H_{\text{eff}} = -J \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j \qquad H_{\text{eff}} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j$$

Reversible control of J_{ex} under the off-resonant action of laser?

Coherent desctruction of tunneling



$$E(t) = E_0 \cos(\omega t)$$

$$\epsilon(\boldsymbol{k}) \to \epsilon(\boldsymbol{k} + \boldsymbol{A}(t))$$

$$A(t) = \frac{E_0}{\omega} \cos(\omega t - \pi/2)$$

⇒ *time-averaged dispersion:*

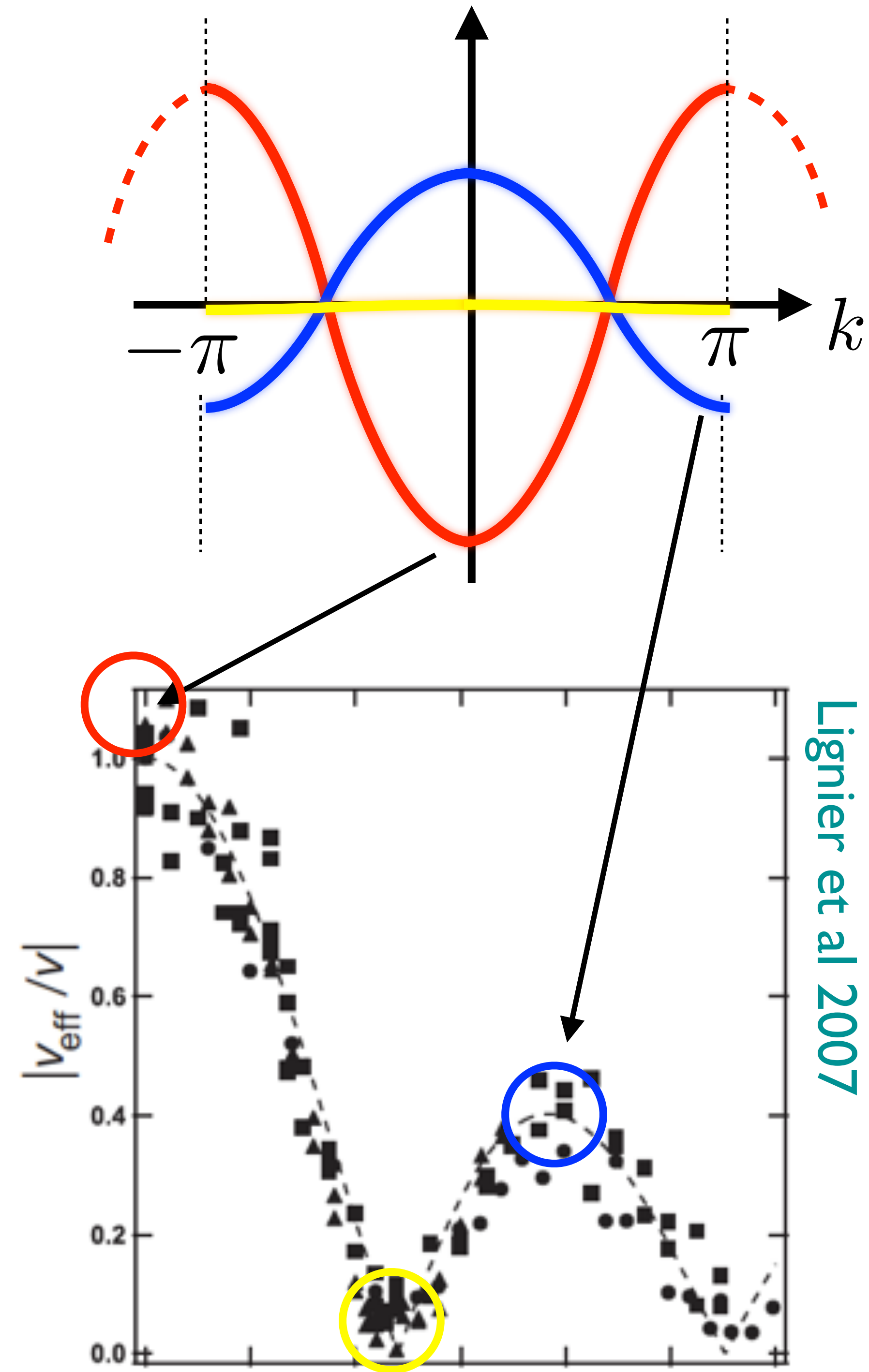
$$\bar{\epsilon}(k) \equiv \frac{1}{T} \int_0^T dt\, \epsilon(k + A(t))$$

$$= J_0(\mathcal{E}) \epsilon(k)$$

Grossmann & Haenggi 1992
Struck et al. 2011
Tsuji et al. 2011

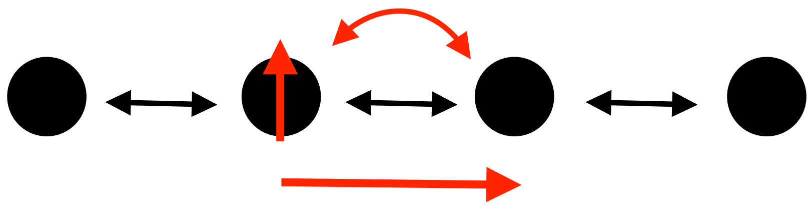
⇒ talk by T. Oka

$$\epsilon(k) = -2t_* \cos(k)$$



$$\mathcal{E} = \frac{eaE_0}{\hbar\omega}$$

Coherent desctruction of tunneling



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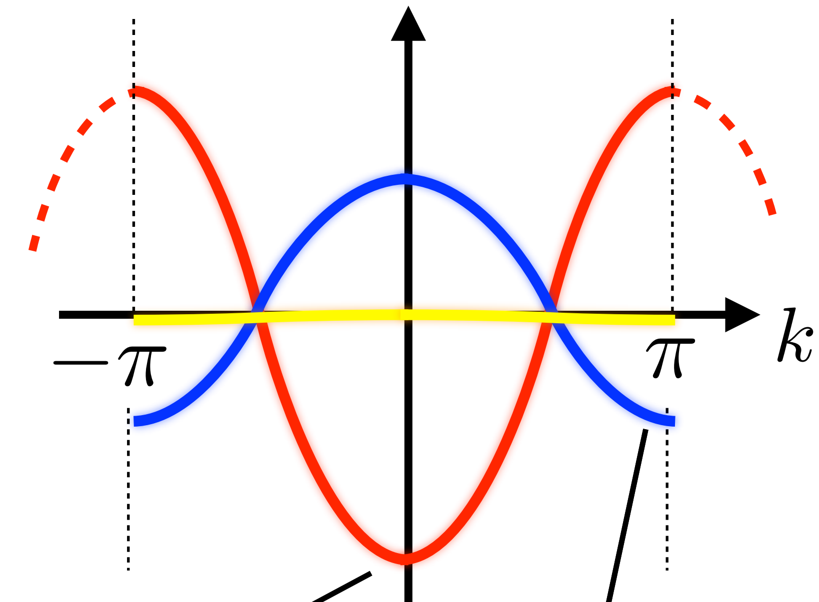
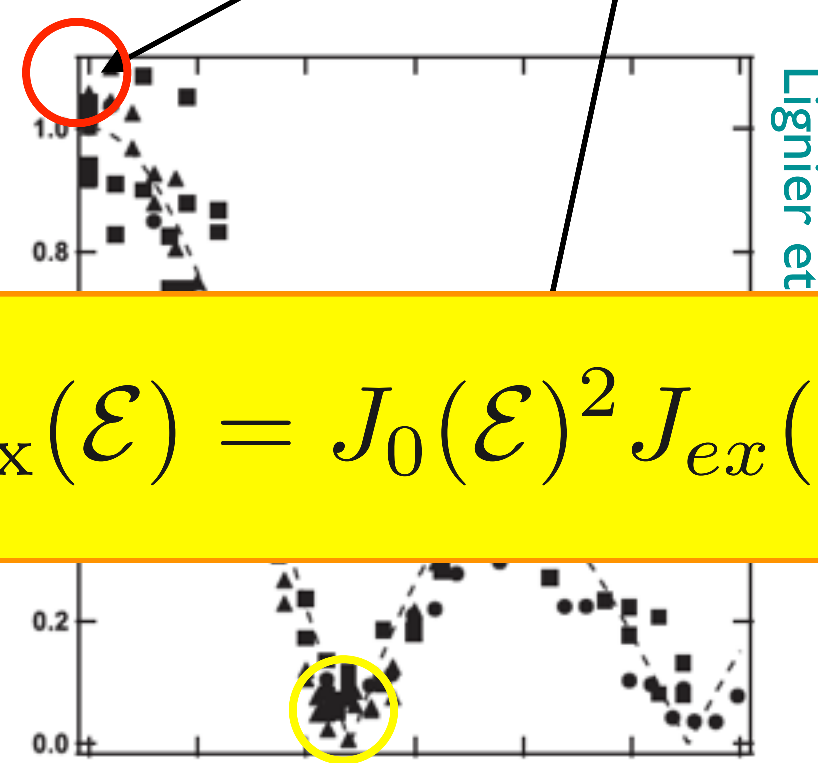




$$J_{\text{ex}}(\mathcal{E}) = J_0(\mathcal{E})^2 J_{ex}(0)$$

$$\mathcal{E} = \frac{eaE_0}{\hbar\omega}$$

Floquet theory

Schrödinger equation for time-periodic Hamiltonian ⇒ Solution via Floque's theorem „Bloch theorem in time"

$$i\partial_t|\Psi(t)\rangle = H(t)|\Psi(t)\rangle$$

$$H(t+T) = H(T)$$

$$|\Psi(t)\rangle = e^{-i\epsilon_\alpha t}|\psi_\alpha(t)\rangle$$

$$|\psi_\alpha(t+T)\rangle = |\psi_\alpha(t)\rangle$$

„adiabatic principle" Change amplitude/frequency slowly: $|\dot{A}/A| \ll \omega$

adiabatic evolution into Floquet state

Floquet matrix: $$|\psi_\alpha(t)\rangle = \sum_n e^{i\omega n t}|\psi_{\alpha,n}\rangle$$

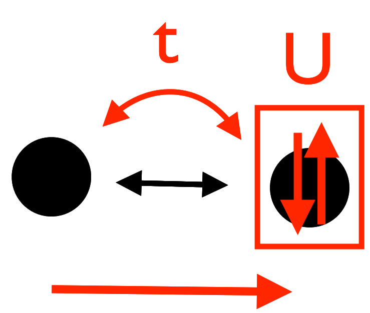
„component in nth Floquet sector"
„n-photon absorbed state"

$$\begin{pmatrix} \ddots & \ddots & & & & \\ \ddots & \hat{h}_{-1} & \hat{h}_0+\omega & \hat{h}_1 & \hat{h}_2 & \\ & \hat{h}_{-2} & \hat{h}_{-1} & \hat{h}_0 & \hat{h}_1 & \\ & & \hat{h}_{-2} & \hat{h}_{-1} & \hat{h}_0-\omega & \ddots \\ & & & & \ddots & \ddots \end{pmatrix} \begin{pmatrix} \vdots \\ \psi_1 \\ \psi_0 \\ \psi_1 \\ \vdots \end{pmatrix} = \epsilon \begin{pmatrix} \vdots \\ \psi_1 \\ \psi_0 \\ \psi_1 \\ \vdots \end{pmatrix}$$

$$\hat{h}_n = \frac{1}{T}\int_0^T dt\, H(t) e^{in\omega t}$$

#Application to two-site Hubbard model

two-site Hubbard model:

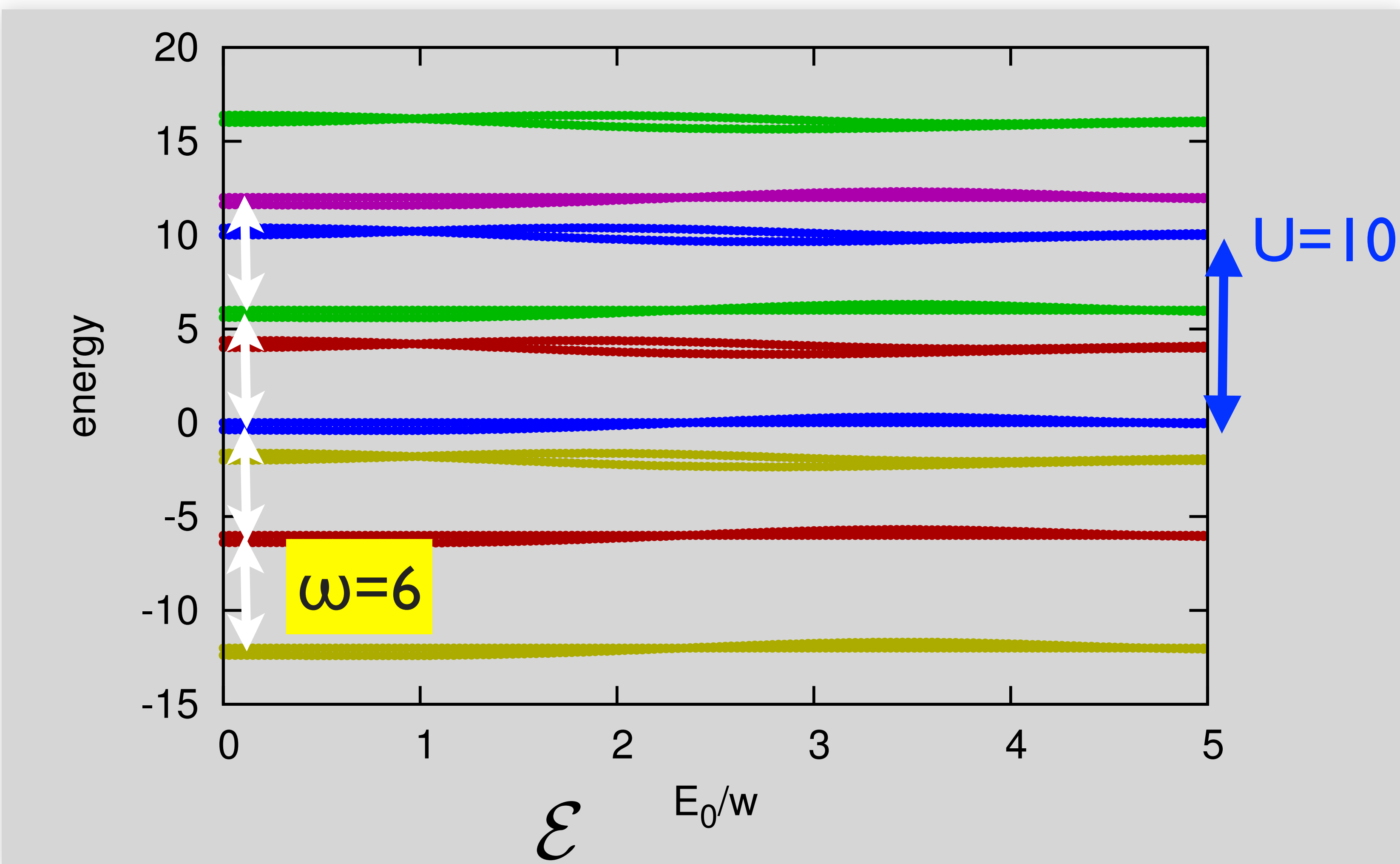


$E(t) = E_0 \cos(\omega t)$

$|0\rangle|\uparrow\downarrow\rangle$, $|\uparrow\downarrow\rangle|0\rangle$ $\qquad E \approx U$

$(|\uparrow\rangle|\downarrow\rangle - |\downarrow\rangle|\uparrow\rangle)/\sqrt{2}$ $\qquad E_T = 0$

$(|\uparrow\rangle|\downarrow\rangle + |\downarrow\rangle|\uparrow\rangle)/\sqrt{2}$ $\qquad E_S \approx -4t_*^2/U$



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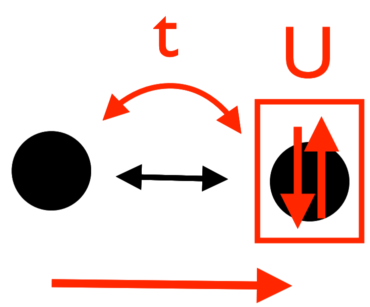

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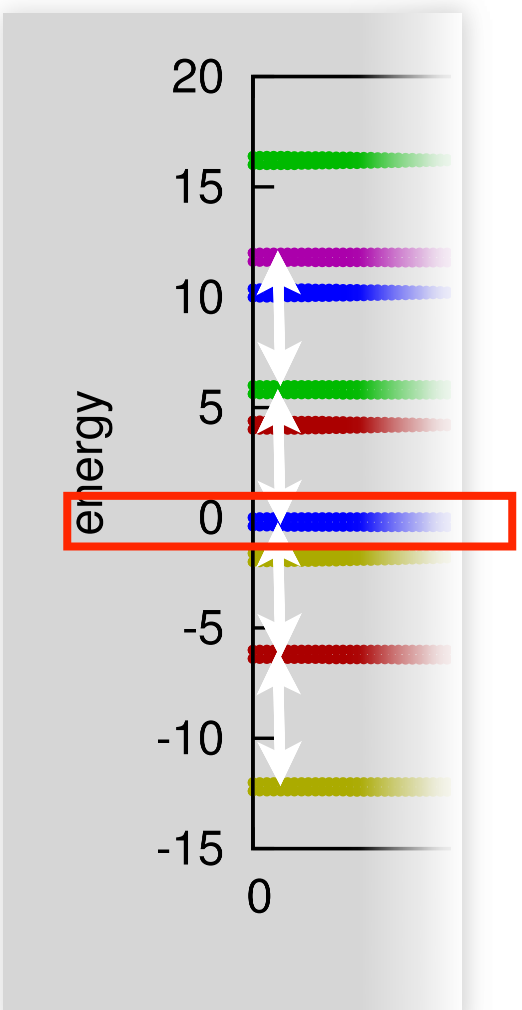
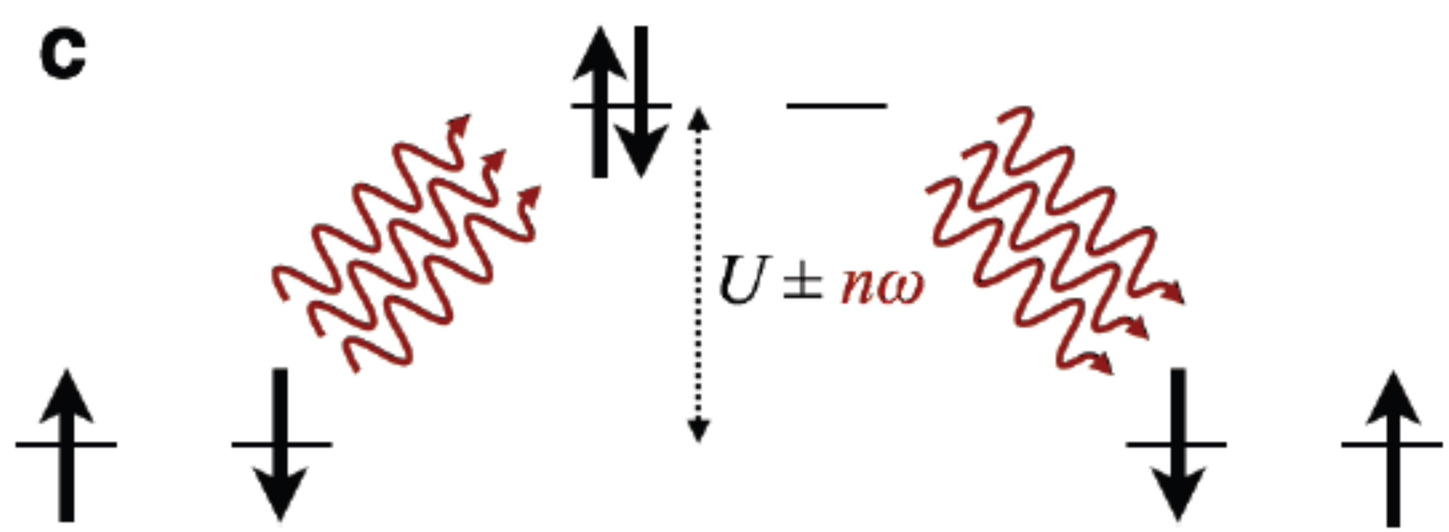


perturbative result $\mathcal{E} \ll 1$

$$J_{ex} = \frac{2t_0^2}{U} + \Delta J_{ex} \quad \Delta J_{ex} = \frac{\mathcal{E}^2 t_0^2}{2}\left(\frac{1}{U+\omega} + \frac{1}{U-\omega} - \frac{2}{U}\right)$$



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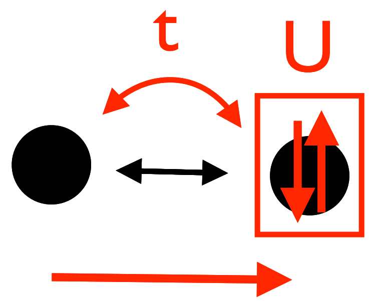

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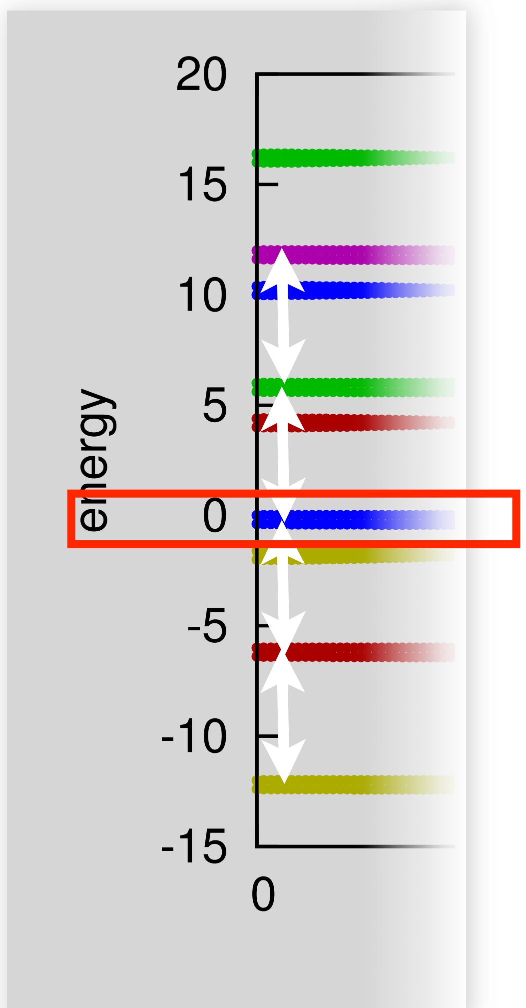
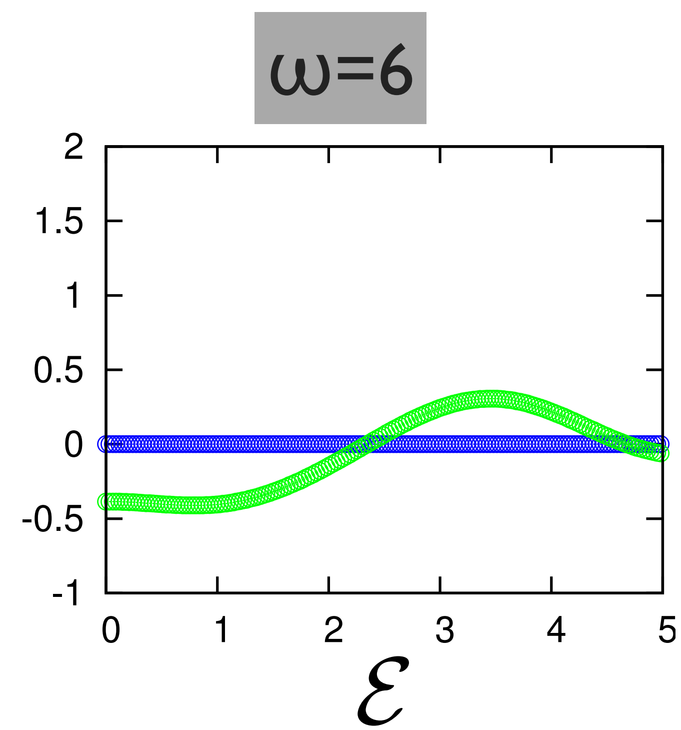
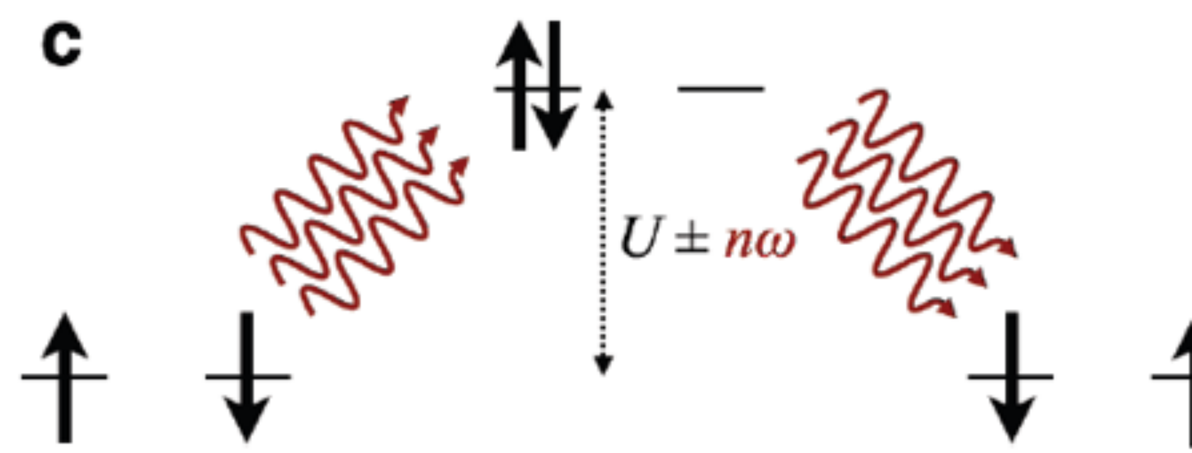
$(\lvert\uparrow\rangle\lvert\downarrow\rangle - \lvert\downarrow\rangle\lvert\uparrow\rangle)/\sqrt{2}$	$E_S \approx -4t_*^2/U$



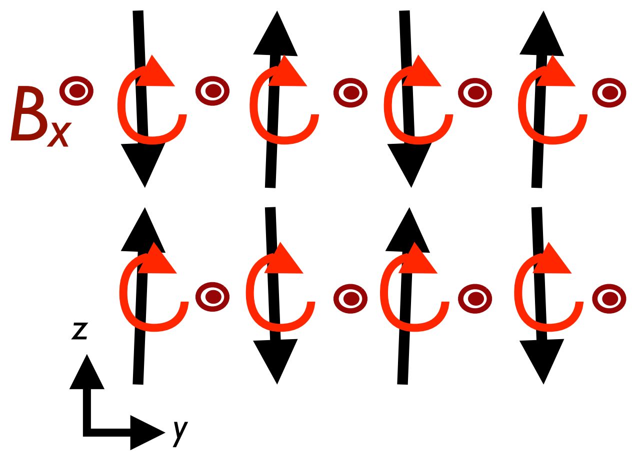




DMFT ...

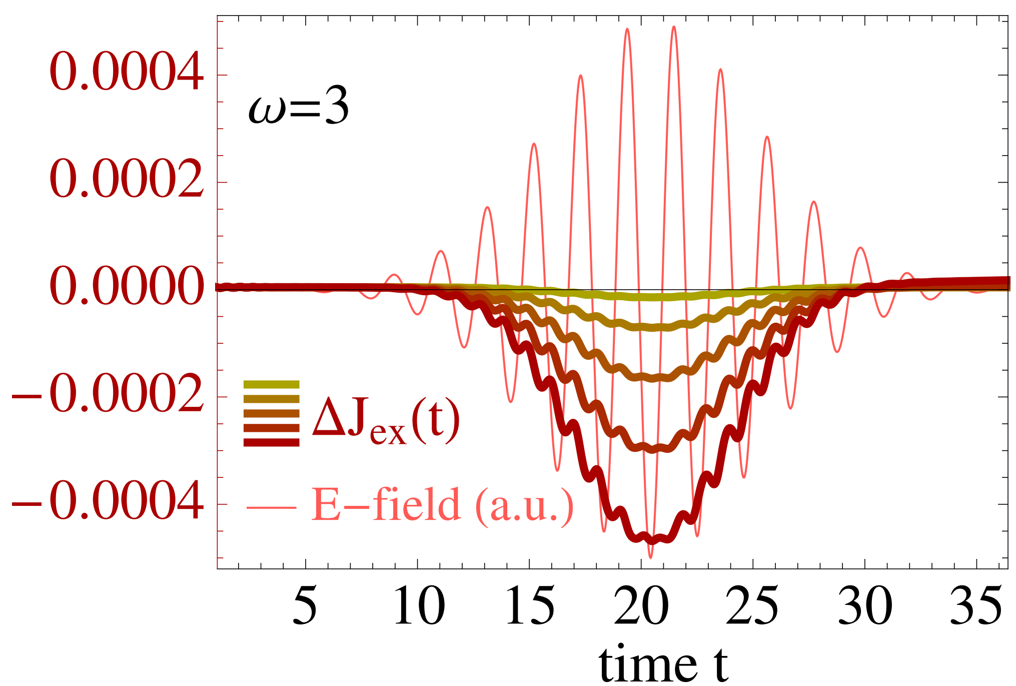
DMFT: *AFM in transverse B field + electric field* $$E(t) = E_0 \cos(\omega t) \exp - \left[\frac{(t-t_0)^2}{2t_0^2} \right]$$

(hypercubic lattice, U=10)



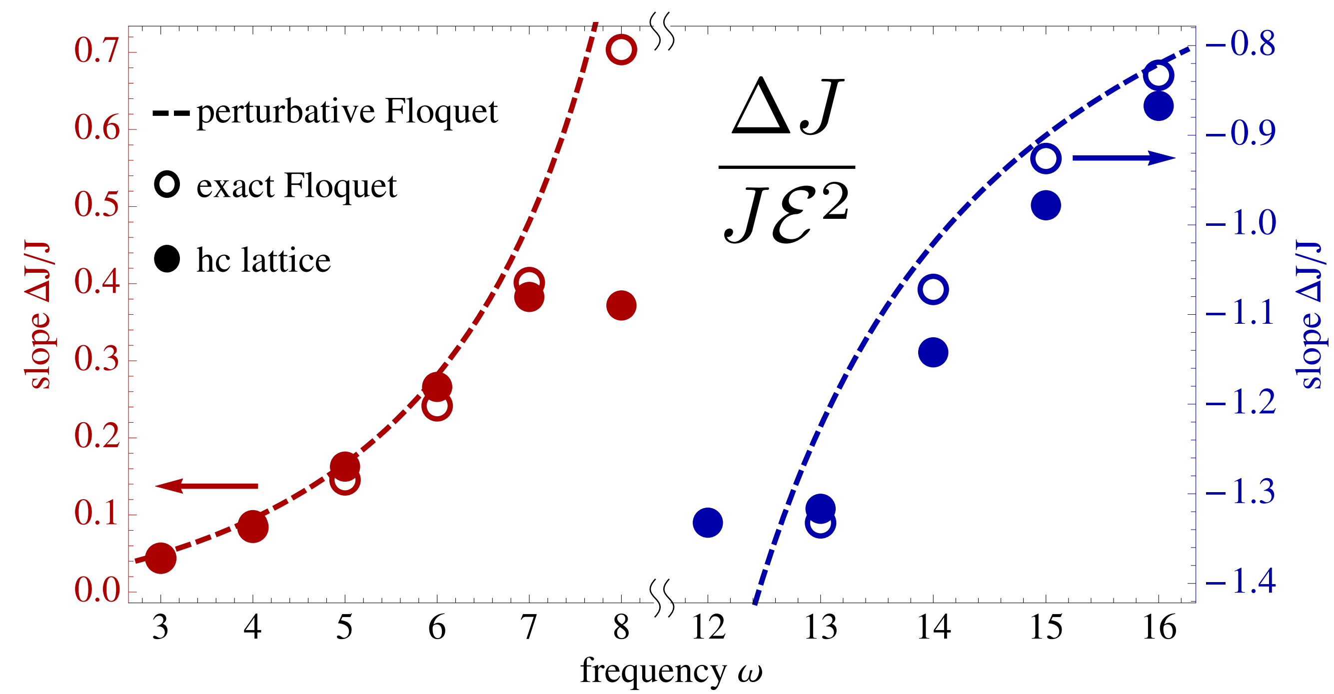
$\Rightarrow$ precession during action of laser:

$$\frac{dS_y}{dt} = S_z(t) B_x \frac{\Delta J_{ex}}{J_{ex}}$$



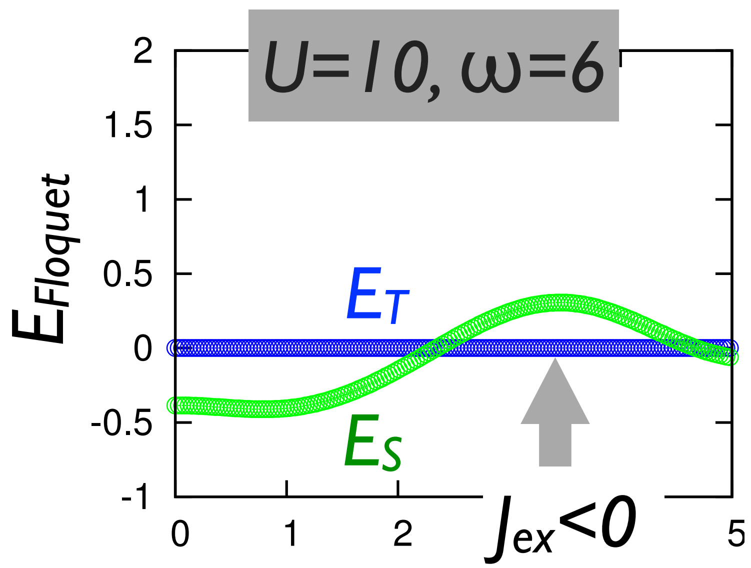
below gap

above gap



ΔJ follows laser intensity

Strong driving: reversing sign of J_{ex}



⇒ transition to ferromgnetic state under driving ?

Angular momentum conserved,
need at least spin-orbit coupling

⇒ evolution of *AFM* with *FM* spin-Hamiltonian?

$$\mathcal{U}_{\mathrm{AFM}}(t) = \exp\Big(- it J_{\mathrm{ex}} \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j \Big)$$

$$\mathcal{U}_{\mathrm{FM}}(t) = \exp\Big(- it(-J_{\mathrm{ex}}) \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j \Big) = \mathcal{U}_{\mathrm{AFM}}(t)^{-1}$$

⇒ „propagation backwards in time“

Strong driving: reversing sign of J_{ex}

10-site Hubbard chain U=20

exact time-propagation

initial state = perfect Neel State

$$|\Psi(0)\rangle = c^\dagger_{0\uparrow}c^\dagger_{1\downarrow}\cdots c^\dagger_{9\uparrow}c^\dagger_{10\downarrow}|0\rangle$$

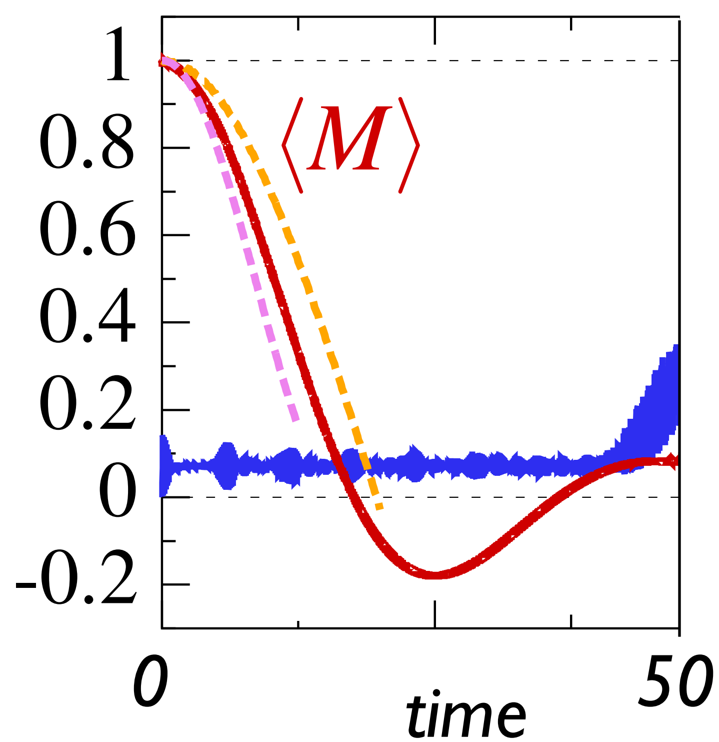
$$= |\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle$$

Free time-evolution

$$H_{\rm Hubbard} \approx J_{\rm ex}\sum_{\langle ij\rangle}\boldsymbol{S}_i\boldsymbol{S}_j = J_{\rm ex}\sum_{\langle ij\rangle}S_i^zS_j^z + \frac{1}{2}\left(S_i^+S_j^- + S_i^-S_j^+\right)$$

quantum spin-flip terms

decay of *AFM*-order



Strong driving: reversing sign of J_{ex}

10-site Hubbard chain U=20

exact time-propagation

initial state = perfect Neel State

$$|\Psi(0)\rangle = c^\dagger_{0\uparrow} c^\dagger_{1\downarrow} \cdots c^\dagger_{9\uparrow} c^\dagger_{10\downarrow} |0\rangle$$

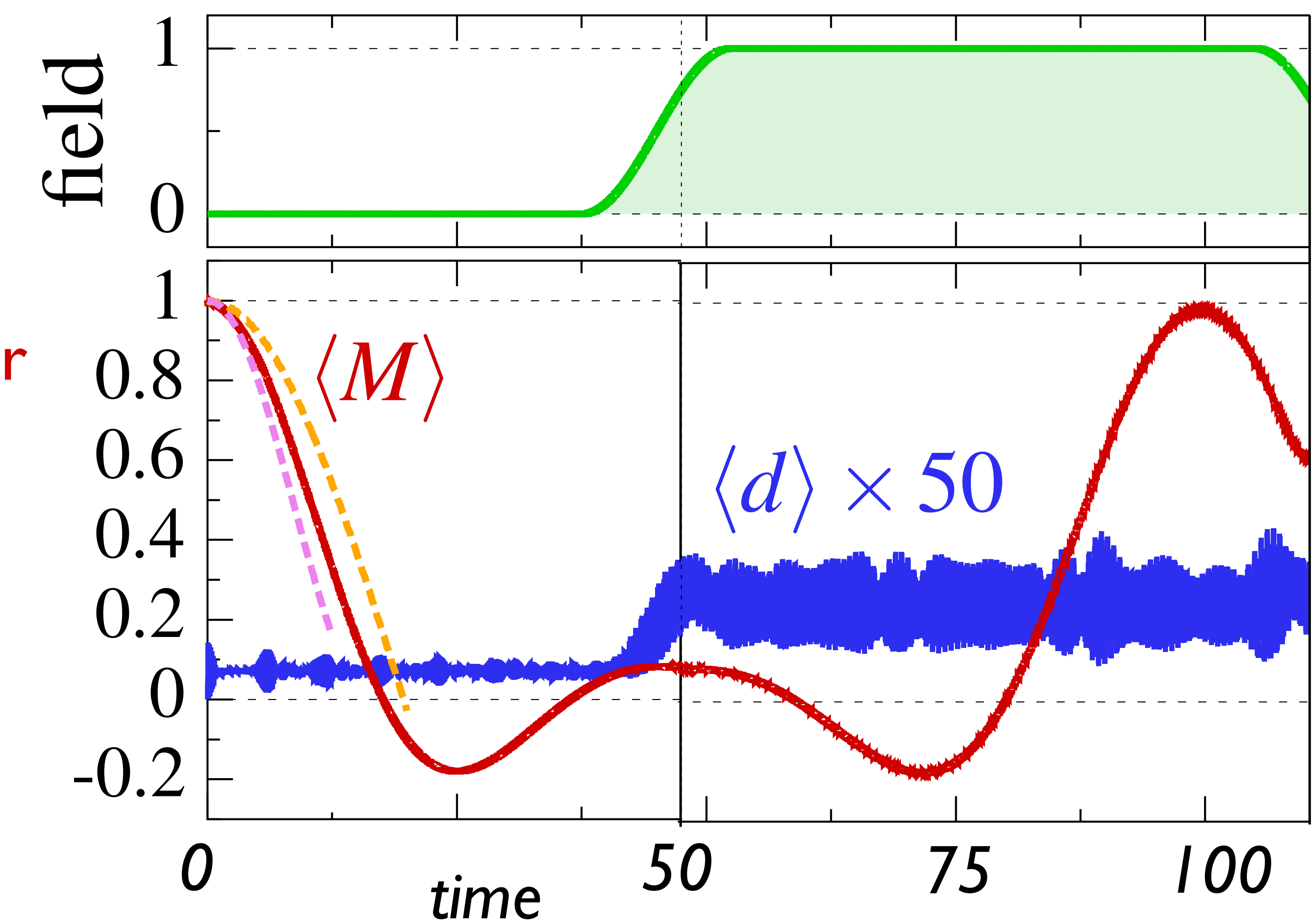
$$= |\uparrow\rangle |\downarrow\rangle |\uparrow\rangle |\downarrow\rangle |\uparrow\rangle |\downarrow\rangle |\uparrow\rangle |\downarrow\rangle |\uparrow\rangle |\downarrow\rangle$$

Free time-evolution

$$H_{\text{Hubbard}} \approx J_{\text{ex}} \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j = J_{\text{ex}} \sum_{\langle ij \rangle} S^z_i S^z_j + \frac{1}{2}\left(S^+_i S^-_j + S^-_i S^+_j\right)$$

quantum spin-flip terms

decay of *AFM*-order



driving field envelope

backward evolution

Strong driving: reversing sign of J_{ex}

10-site Hubbard chain U=20

exact time-propagation

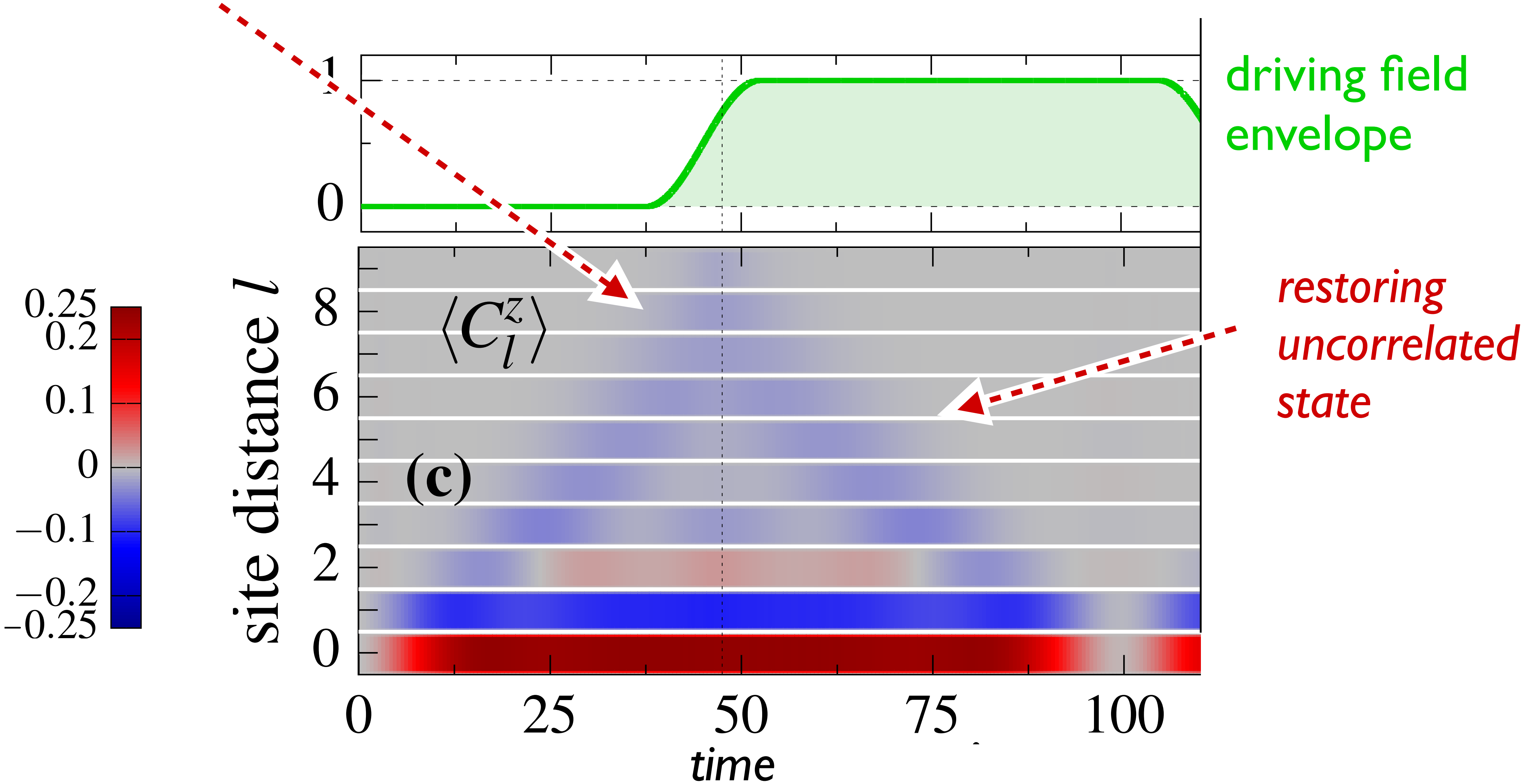
initial state = perfect Neel State

$$|\Psi(0)\rangle = c^\dagger_{0\uparrow}c^\dagger_{1\downarrow}\cdots c^\dagger_{9\uparrow}c^\dagger_{10\downarrow}|0\rangle$$

$$= |\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle$$

buildup of short-range correlations

$$C(l) = \langle S^z_0 S^z_l\rangle - \langle S^z_0\rangle\langle S^z_l\rangle$$



Conclusion

The exchange interaction can be controlled with electric field of the laser

→ Photo-excited states with modified exchange:
Effect on exchange interaction (~2%) similar to chemical doping

Mentink & Eckstein, arXiv 1401.5308

- exchange in laser-driven state

→ anything is possible (J_{ex} → $-J_{ex}$)

→ driven superexchange: $\Delta J/J \sim 0.1\%$

Mentink, Balzer, Eckstein, to be submitted