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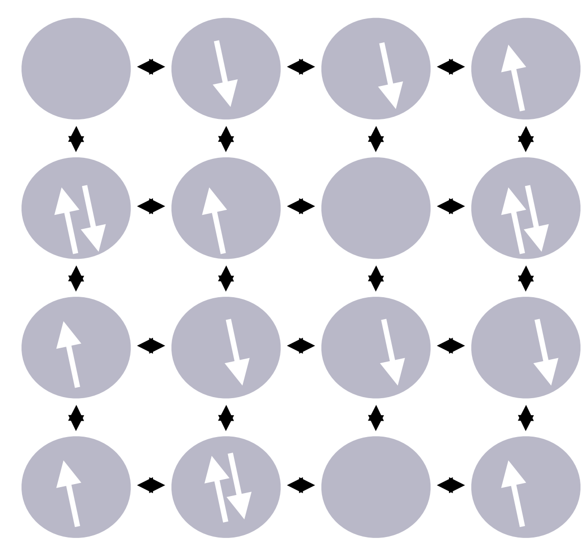
Probing and controlling the ultrafast dynamics in complex materials

Martin Eckstein (University of Erlangen)

Cargese, June 2022



Ultrafast dynamics in solids

Femtosecond pump-probe experiments: Selective probe of the dynamics of various degrees of freedom on very different timescales

- Electronic structure (tr-ARPES, XAS)
- Lattice (Xray, electron diffraction)
- Collective spin/orbital excitations (RIXS)
- Phonons (THz - midIR)

Major goals:

⇨ Learn from the dynamics about the relevant degrees of freedom and their interactions?

⇨ Reach novel states out of equilibrium?

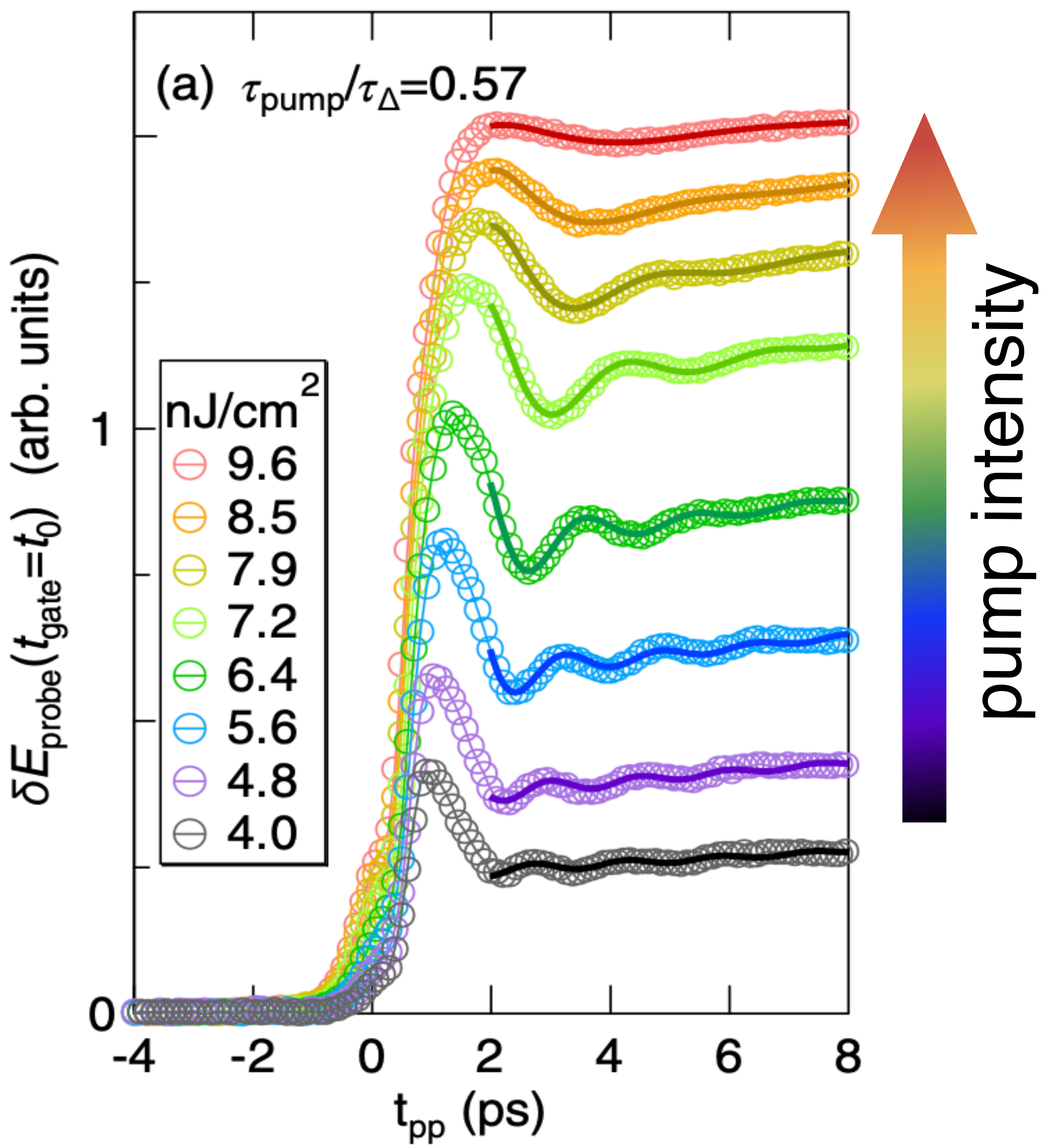
some reviews:

- Aoki et al. Rev. Mod. Phys. 86, 779 (2014)
- Giannetti et al. Advances in Physics, **65**, 58 (2016)
- Basov, Hsieh, Averitt, Nature Materials **16**, 1077 (2017)
- de la Torre et al., Rev. Mod. Phys. **93**, 041002 (2021)

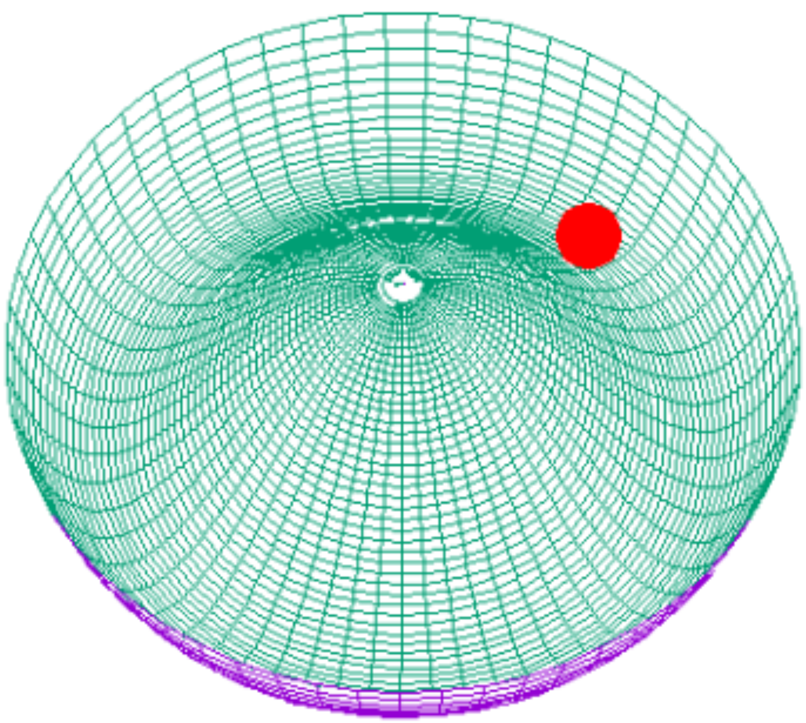
Some examples (towards superconductivity) for motivation

Transient optical transmission on THz-pumped $Nb_{1-x}Ti_xNi$

Matsunaga et al., PRL **111**, 057002 (2013)



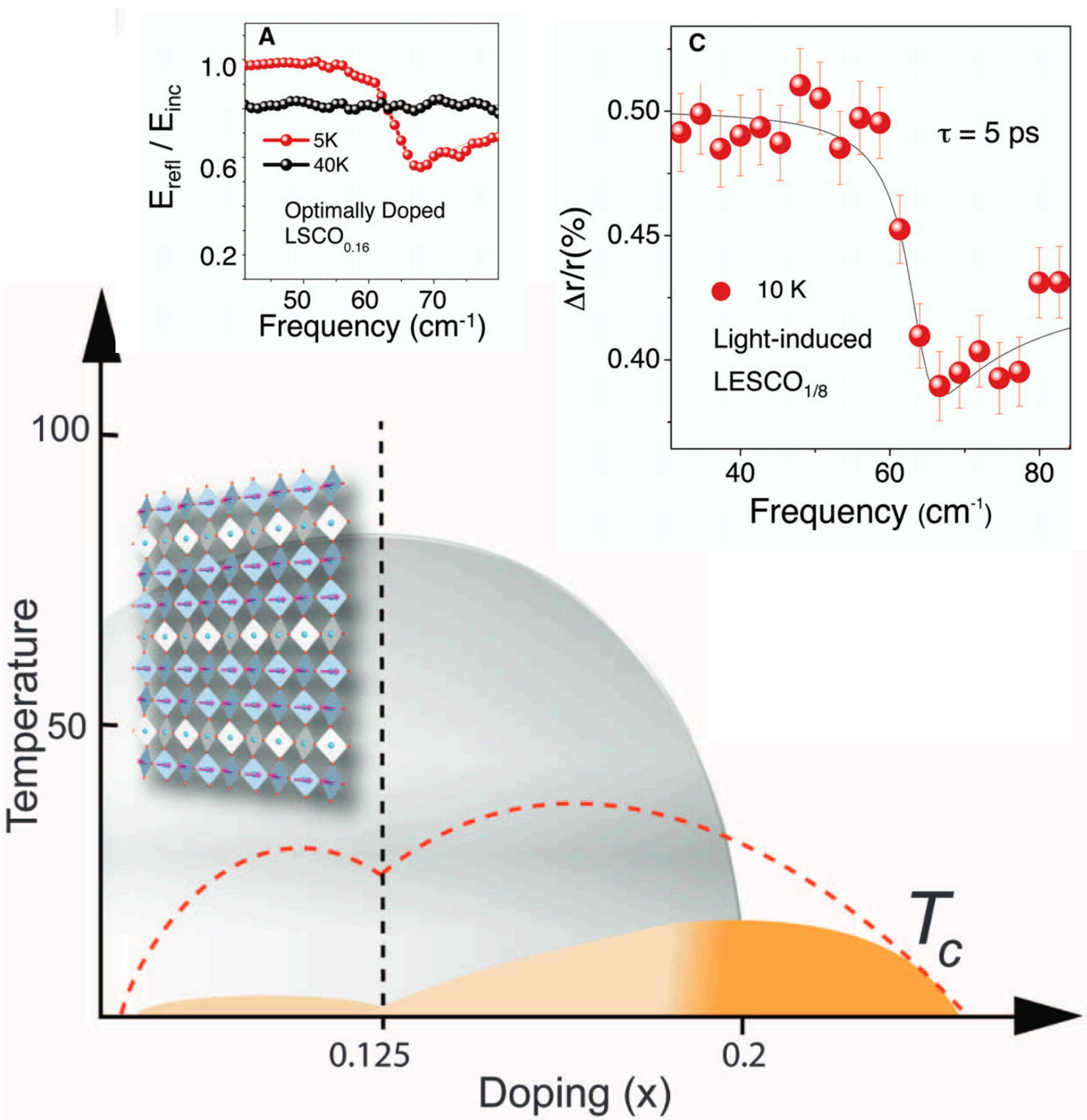
Non-equilibrium excitation and real-time observation of collective modes of the superconductor (Higgs mode)



Some examples (towards superconductivity) for motivation

Light-Induced Superconductivity in a Stripe-Ordered Cuprate

Fausti et al., Science **331**, 189 (2011)



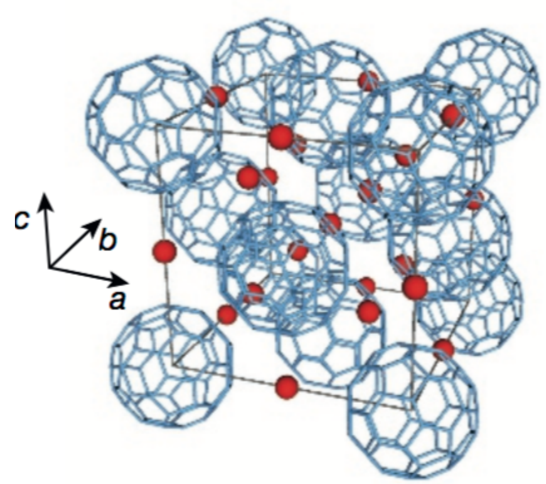
Revealing SC response (Josephson plasmon) at 1/8 doping after mid-IR excitation

⇨ Non-equilibrium suppression of competing (stripe) phase?

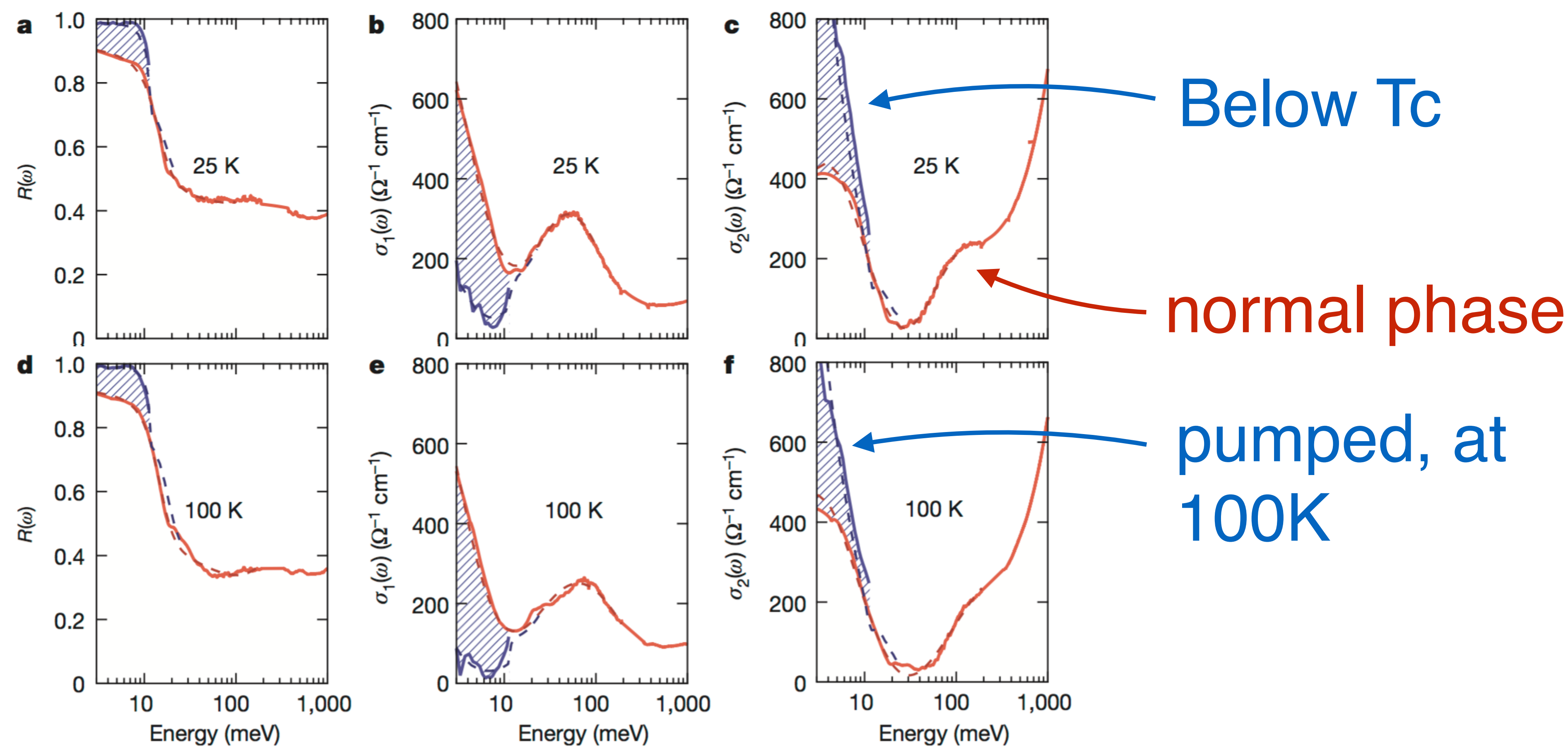
Some examples (towards superconductivity) for motivation

Possible light-induced superconductivity in K_3C_{60} at high T

Mitrano et al., Nature **530**, 461 (2016)



Optical signatures of superconductivity



pump: mid-IR excitation, close to resonance to C_{60} vibrations (?)

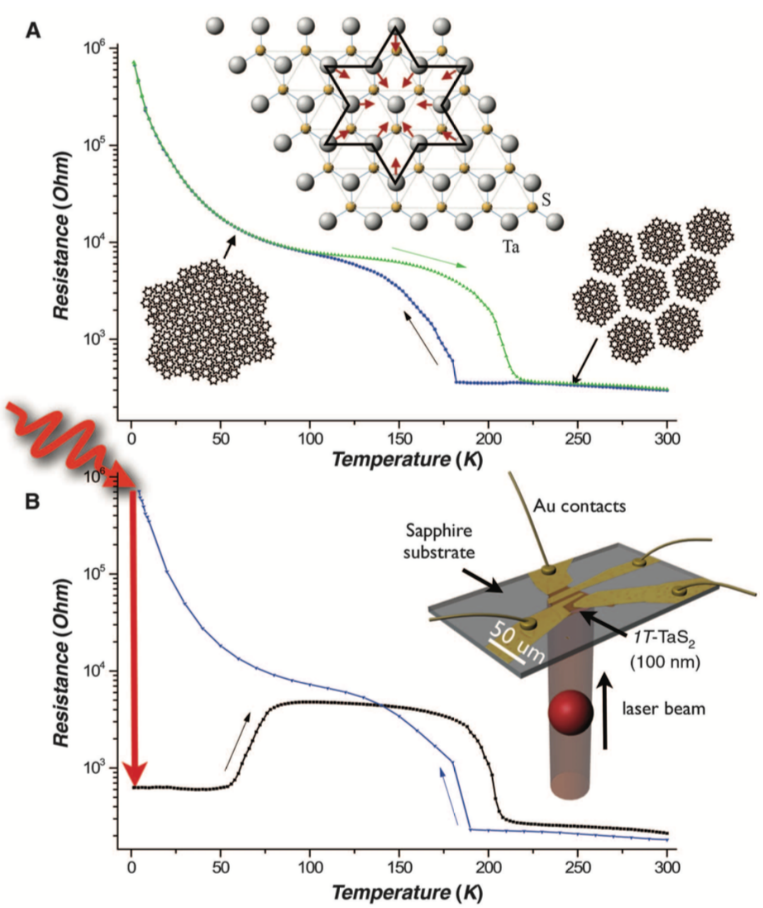
⇨ Stabilize transient states, which do not exist under equilibrium conditions, trough external driving?

Some examples for motivation

”Hidden states”: Long-lived or truly metastable states, reachable only along “non-thermal” pathways

Ultrafast Switching to a Stable Hidden Quantum State in an Electronic Crystal

L. Stojchevska,[1,2] I. Vaskivskyi,[1] T. Mertelj,[1] P. Kusar,[1] D. Svetin,[1] S. Brazovskii,[3,4] D. Mihailovic[1,2,5]*

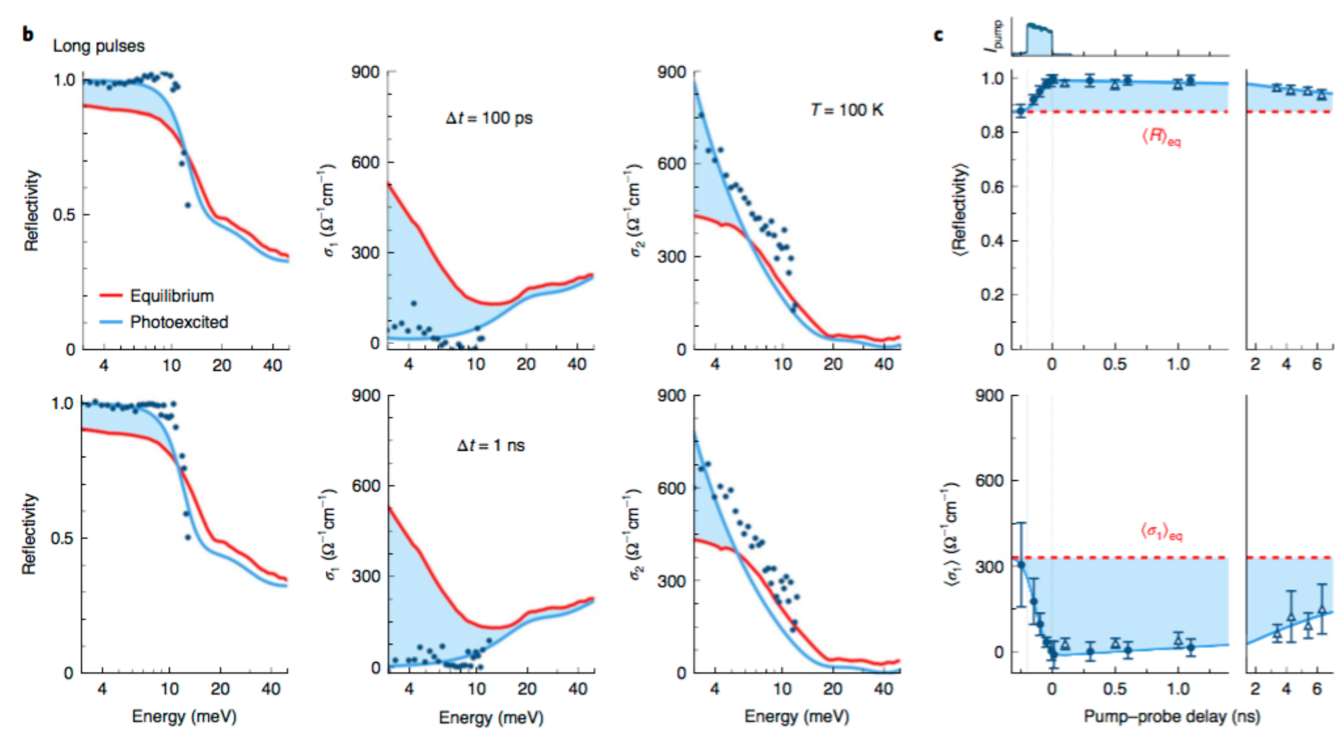


nature physics
ARTICLES
https://doi.org/10.1038/s41567-020-01148-1

OPEN

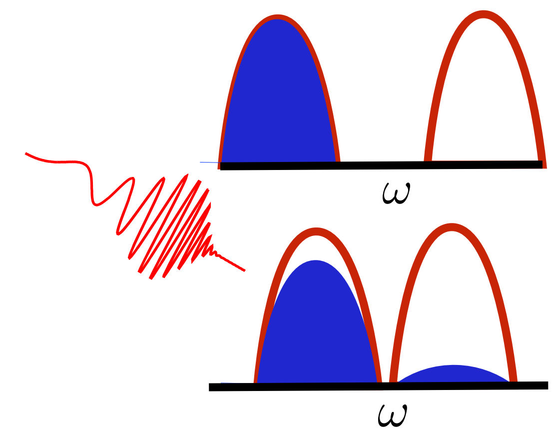
Evidence for metastable photo-induced superconductivity in K_3C_{60}

M. Budden[1]✉, T. Gebert[1], M. Buzzi[1], G. Jotzu[1], E. Wang[1], T. Matsuyama[1], G. Meier[1], Y. Laplace[1], D. Pontiroli[2], M. Riccò[2], F. Schlawin[3], D. Jaksch[3] and A. Cavalleri[1,3]✉



Pathways to control states out of equilibrium

Impulsive generation of non-equilibrium states:



- ⇨ Transient modification of the electronic structure
- ⇨ Non-thermal free energy potentials, hidden states

Control during application of external fields:

- ⇨ Floquet engineering: Change microscopic parameters through external driving
- ⇨ Non-linear phononics: Stabilize novel states through coherent excitation of phonons
- ⇨ Coherent ultrafast processes: Light-induced currents, HHG, …

Outline

This lecture:

More broad (theoretical) view on concepts, in particular pathways to reach (novel) states under non-equilibrium conditions

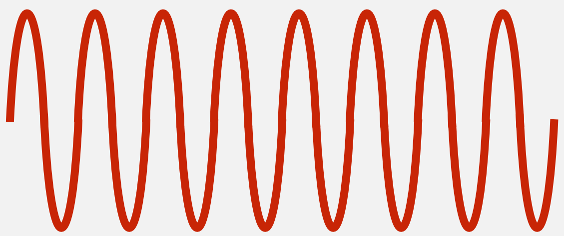
- Part 1: Floquet engineering: (new states by periodic driving)
- Part 2: Population dynamics: (new states by photo-doping)
- Part 3: Understanding dynamics of symmetry broken states

This will not only be focused on superconductivity (… actually, mostly not …)
I guess, the lecture by C. Giannetti will be more specific on non-equilibrium superconductivity

Part 1: Floquet engineering

General idea:

time-periodic Hamiltonian $H(t+T) = H(t)$ due to external fields:



$f(t) = S(t)\cos(\Omega t)$

Projecting out fast oscillations (suitably time-averaged dynamics) leads to effective Hamiltonian $H_{eff} = H_F[S,\Omega]$

H_F can be very different from undriven system:

new band structure, artificial magnetic fields, modified many body interactions (magnetic exchange, superconducting pairing)

⇨ Stabilize new equilibrium states?

⇨ for slowly varying envelope: $H_{eff}(t) = H_F[S(t),\Omega]$: nontrivial time-dependent forces

Classical example(s)

Effective potential due to rotating magnetic field ⇨ new stable minimum

Mathematical description: Stroboscopic time evolution

Time evolution operator over one period written in terms of Hermitian H_F

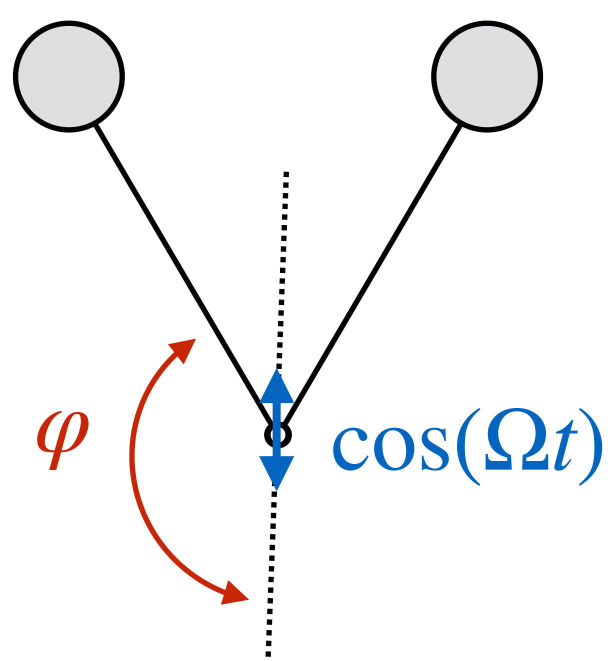
$$U(t+T,t) = T_t e^{-i\int_t^{t+T} ds H(s)} \equiv e^{-iTH_F}$$

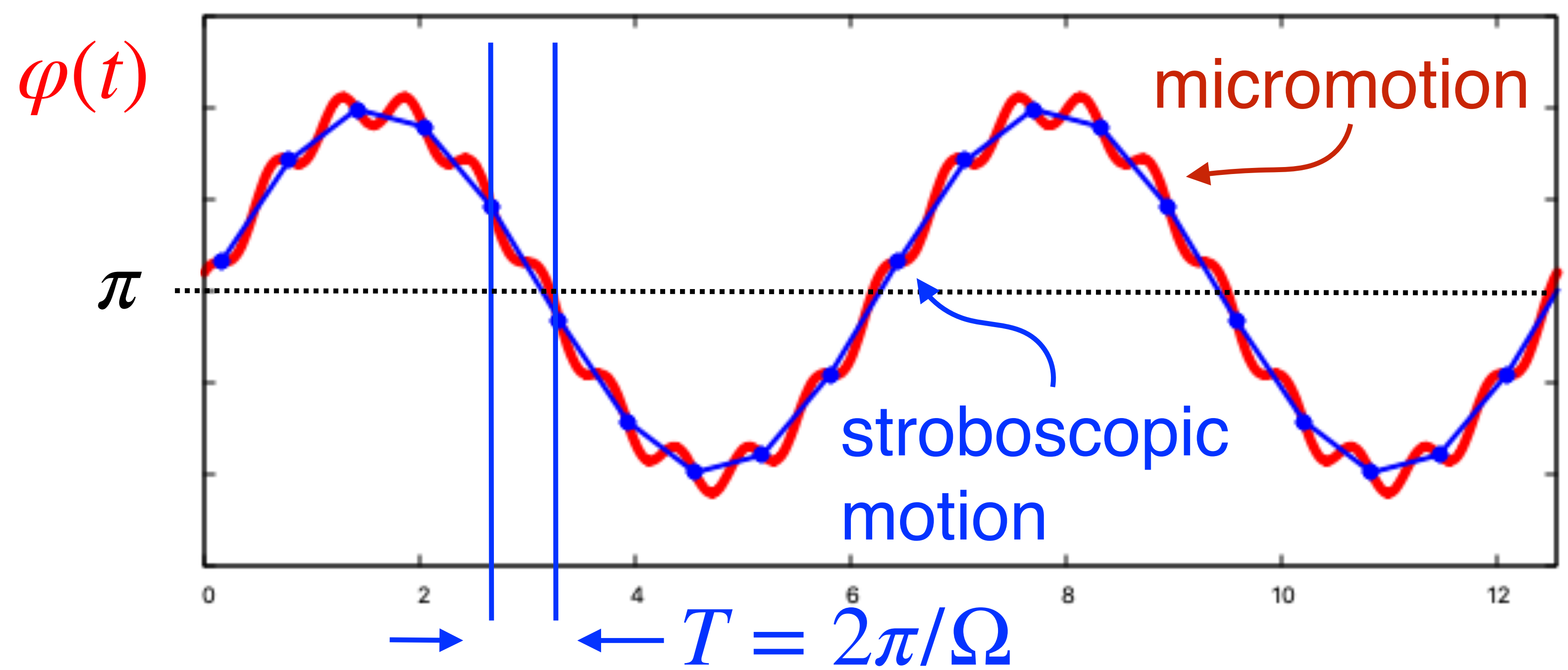
⇨ Dynamics at integer multiples of T $\equiv$ evolution with time-independent H_F

Review: Bukov, D'Alessio, Polkovnikov, Adv. in Phys, **64**, 139 (2015)

Leading approximation for fast driving $H_F = \dfrac{1}{T}\displaystyle\int_t^{t+T} ds\, H(s)$

Illustration: Kapitza pendulum $H_F \approx$ potential $V(\varphi)$ with new stable minimum





Mathematical description: Photon picture

"Bloch Ansatz in time": (Floquet 1886)

$$i\partial_t |\psi(t)\rangle = H(t)|\psi(t)\rangle \;\Rightarrow\; \text{Floquet ansatz:} \quad |\psi(t)\rangle = \underbrace{|u(t)\rangle}_{\#} e^{-i\epsilon t}$$

is periodic: $|u(t)\rangle = \sum_n |u_n\rangle e^{-i\Omega n t}$

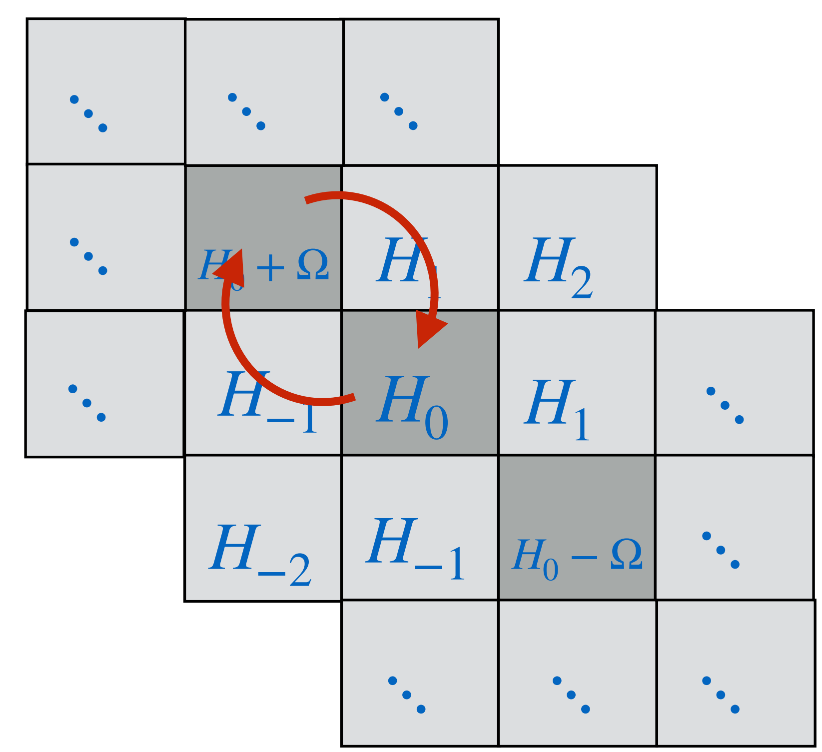
$$(\epsilon + n\Omega)|u_n\rangle = \sum_{n'} H_{n-n'}|u_{n'}\rangle \qquad H_{n-n'} = \frac{1}{T}\int_0^T ds\, e^{i\Omega(n-n')s} H(s)$$

Time-independent Schrödinger equation $\mathscr{H} u = \epsilon u$

in extended Hilbertspace:

$|\alpha, n\rangle$ $\begin{cases} \alpha\text{: Basis for matter} \\ n\text{: "Photon index"} \end{cases}$

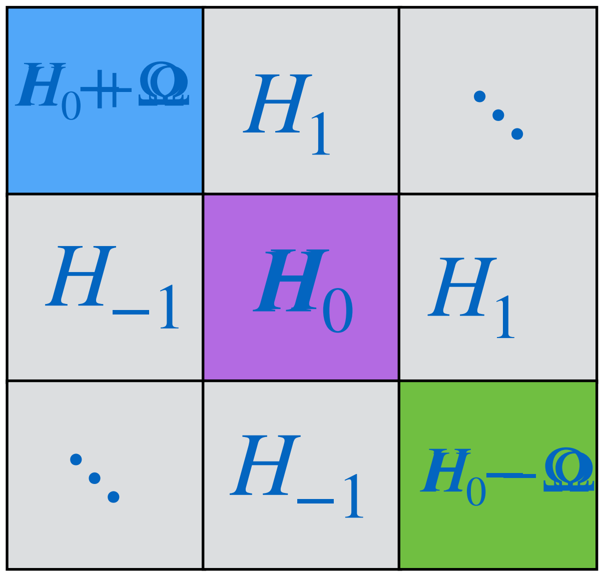
$\mathscr{H} =$

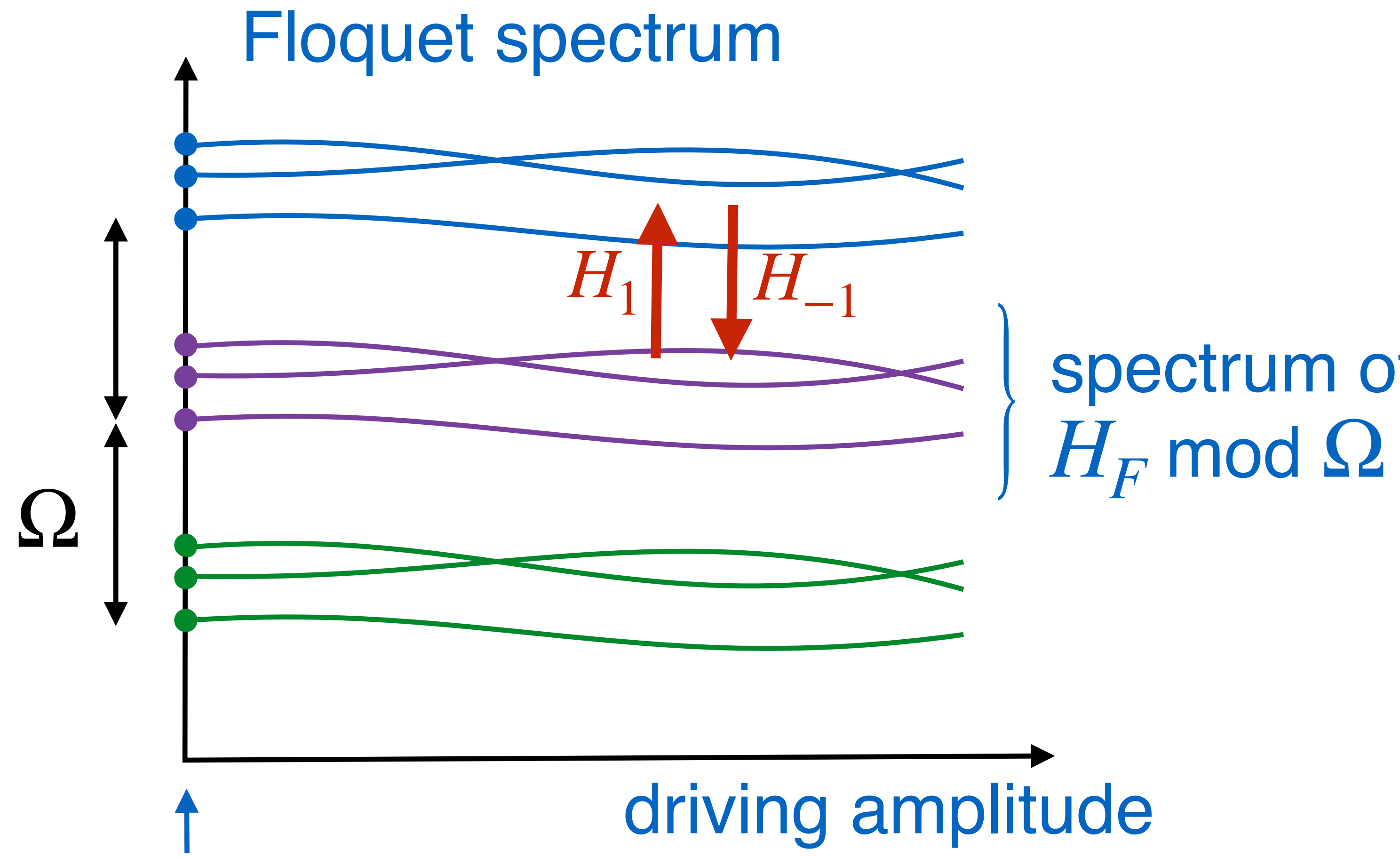


$H_{\pm n}$: n-Photon absorption/emission

High-frequency expansion

$\mathscr{H} =$





undriven: $\epsilon = E_\alpha + n\Omega$

As long as Ω is the largest relevant scale, we can do regular perturbation theory to get spectrum of H_F

$$H_F = H_0 - \sum_{n=-\infty}^{\infty} H_{-n} \frac{1}{n\Omega} H_n$$

$\equiv$ time-average $\bar{H}$

"Virtual photon emission and absorption"

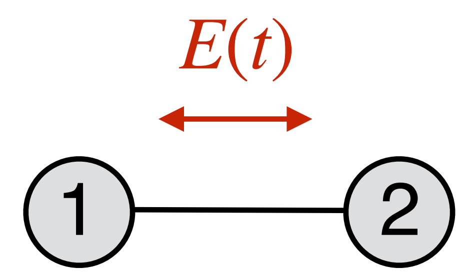
Mikami et al. PRB **93**, 144307 (2016)
Bukov, et al. Adv. Phys, **64**, 139 (2015)
Eckhard & Anisimovas, NJP **17**, 093039 (2015)

Dynamical localization and band flipping

Tight-binding model with external electric field

- $H = -t_0\left(c_1^\dagger c_2 + h.c.\right) + gE(t)\left(c_1^\dagger c_1 - c_2^\dagger c_2\right)$

  $\underbrace{gE(t)\left(c_1^\dagger c_1 - c_2^\dagger c_2\right)}_{\text{scalar potential}}$



- Vector-potential representation: $E(t) = -\partial_t A$

  Time-dependent unitary transformation $W(t)$ ("rotating frame"):

$$H_{rot} = W^\dagger H W + W^\dagger i\partial_t W = -t_0\left(c_1^\dagger c_2 e^{i2gA(t)} + h.c.\right)$$

$$W(t) = e^{-igA(t)(n_1 - n_2)}$$

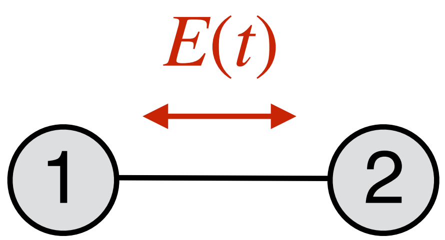
- general light matter coupling in TB models:

  Luttinger, Phys. Rev. **84**, 814 (1951).
  Li et al., PRB **101**, 205140 (2020)

  Peierls phase: $t_{ab} \to t_{ab} e^{ig\vec{A}(t)\cdot(\vec{R}_a - \vec{R}_b)}$
  (+ dipolar matrix elements)

  "$\epsilon(\vec{k}) \to \epsilon(\vec{k} - g\vec{A})$"

Dynamical localization



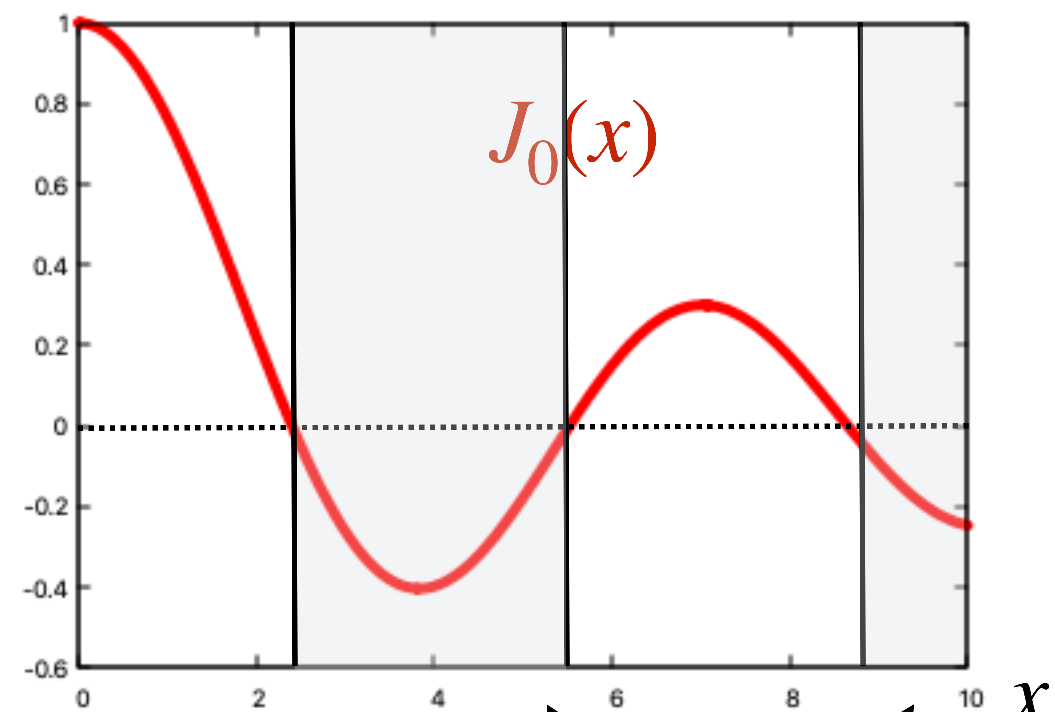
$$H(t) = -t_0\left(c_1^\dagger c_2 e^{igA_0\cos(\Omega t)(t)} + h.c.\right)$$

Time average $\overline{e^{igA_0\cos(\Omega t)}} = J_0(gA_0)$

H_F in high frequency limit:

tight-binding model with
renormalized hopping

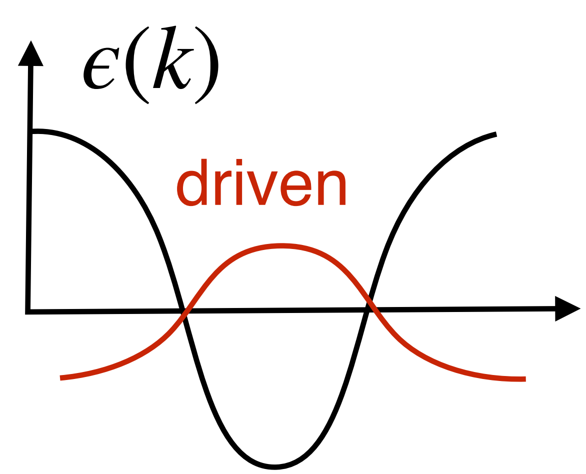
Dunlap & Krenke, PRB **34**, 3625 (1986)



$x \approx 2.4$

dynamical localization

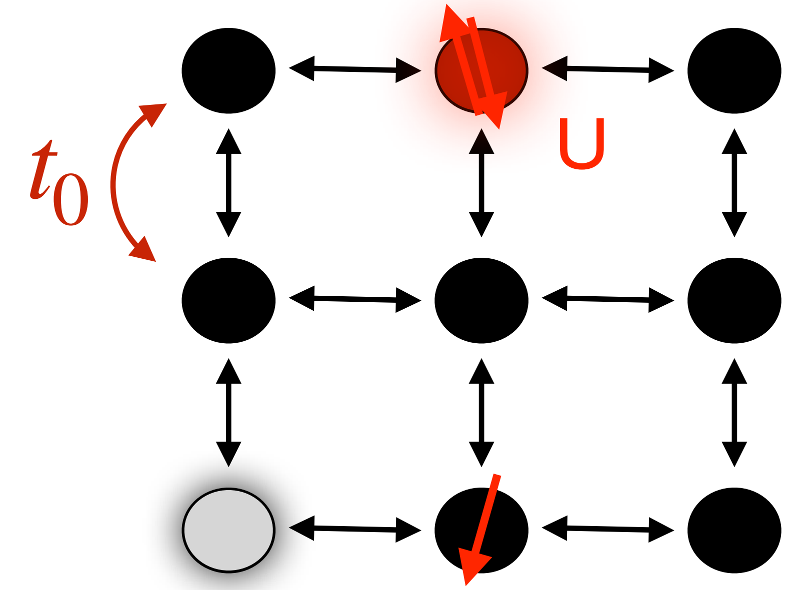
band flipping



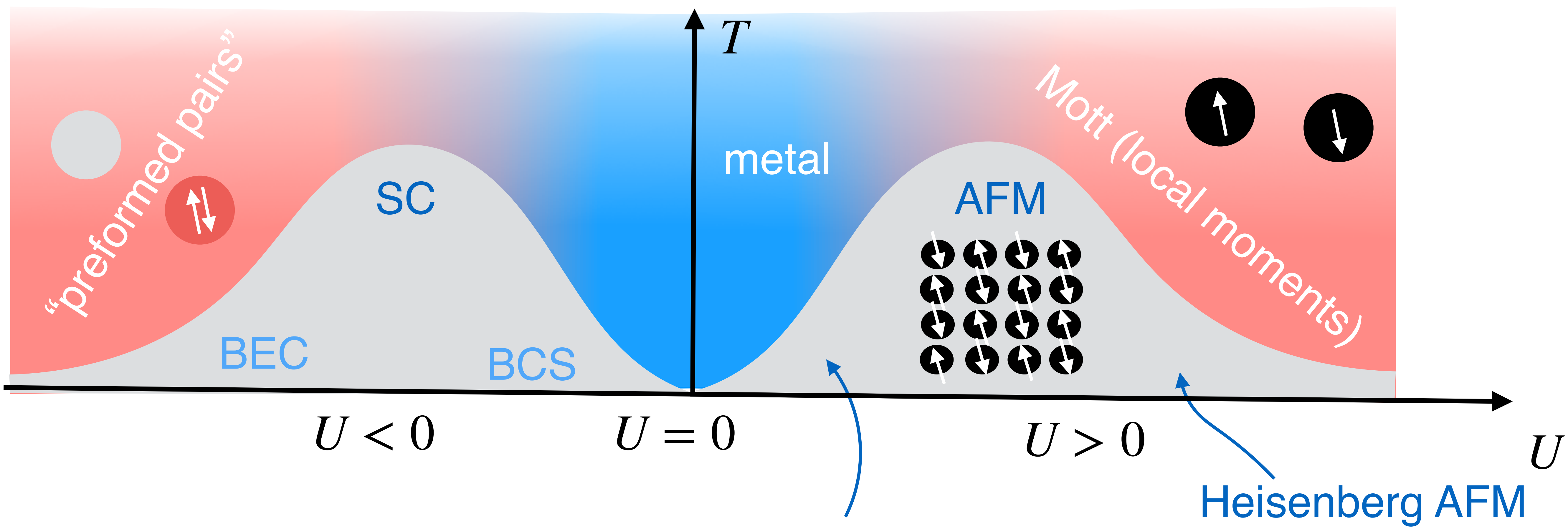
Dynamical band flipping in Hubbard model

Hubbard model:

$$H = -t_0 \sum_{\langle i,j \rangle} c^\dagger_{i,\sigma} c_{j,\sigma} + U \sum_i \left(n_{i,\downarrow} - \tfrac{1}{2}\right)\left(n_{i,\uparrow} - \tfrac{1}{2}\right)$$



Phase diagram of half filled Hubbard model:



$$H = \frac{4t_0^2}{U} \sum_{\langle i,j \rangle} \vec{S}_i \vec{S}_j$$

Dynamical band flipping in Hubbard model

Relation attractive / repulsive Hubbard model? … particle-hole transformation

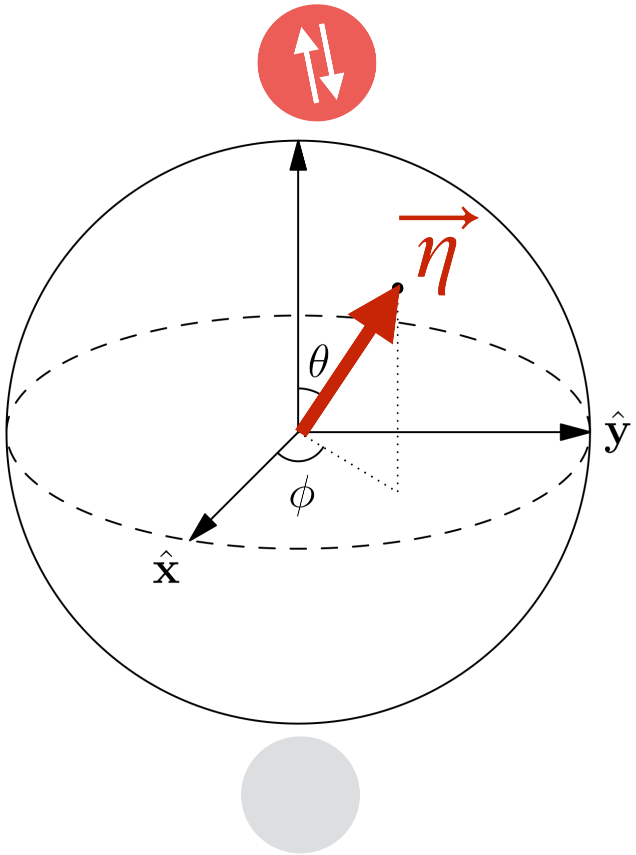
$$c_{i,\downarrow} \to (-1)^i \, c^\dagger_{i,\downarrow}: \quad \Rightarrow \quad \begin{cases} \left(n_{i,\downarrow} - \frac{1}{2}\right) \to -\left(n_{i,\downarrow} - \frac{1}{2}\right) \\ H \to -t_0 \sum_{\langle i,j \rangle} c^\dagger_{i,\sigma} c_{j,\sigma} - U \sum_i \left(n_{i,\downarrow} - \frac{1}{2}\right)\left(n_{i,\uparrow} - \frac{1}{2}\right) \end{cases}$$

check-board sign

Spin → Charge pseudospin

$$S_i^z = \frac{1}{2}(n_{i\uparrow} - n_{i\downarrow}) \to \frac{1}{2}(n_{i\uparrow} + n_{i\downarrow} - 1) \equiv \eta_i^z$$

$$S_i^+ = c^\dagger_{i\uparrow} c_{i\downarrow} \to (-1)^i c^\dagger_{i\uparrow} c^\dagger_{i\downarrow} \equiv \eta_i^+$$



AFM at $U > 0$ → SC or CDW at $U < 0$:

$$\langle \eta_i^\pm \rangle \sim (-1)^i \Rightarrow \langle c^\dagger_{i\uparrow} c^\dagger_{i\downarrow} \rangle \neq 0 \text{ (SC)}$$

Dynamical band flipping in Hubbard model

Use Floquet drive to flip from attractive to repulsive model?

- Periodic driving ⇨ hopping renormalization $t_0 \to t_0 J_0(gA_0)$

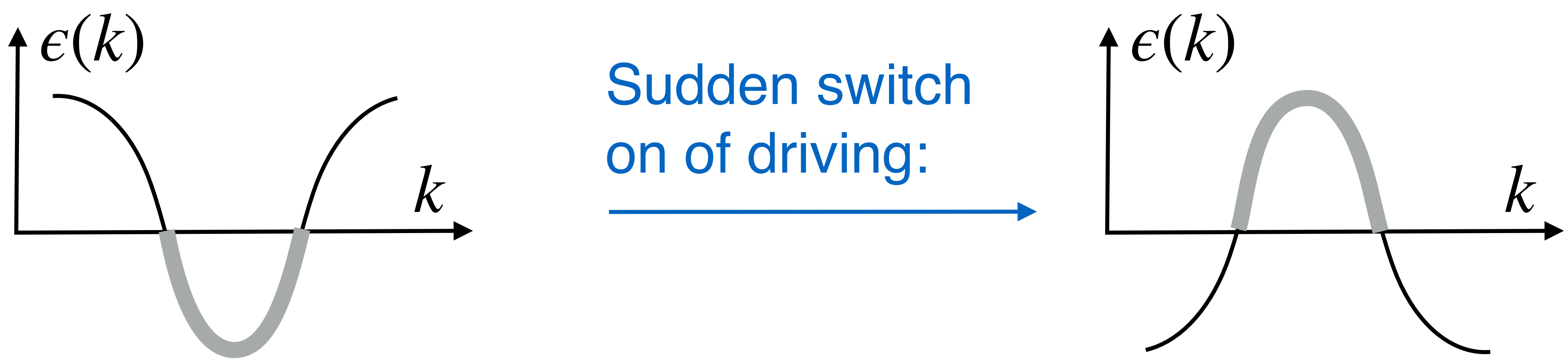
  ⇨ sign reversal for $gA_0 \gtrsim 2.4$

- Equilibrium:

  $\hat{H} = -t_0\hat{K} + U\hat{D} \quad \leftrightarrow \quad \hat{H} = +t_0\hat{K} + U\hat{D}$ related by $c_{i\sigma} \to (-1)^i c_{i\sigma}$

  ⇨ same equilibrium phases (AFM)

- But here we are not talking about equilibrium states!



population inversion, neg. T

Dynamical band flipping in Hubbard model

Thermalization in driven Hamiltonian after switch on of driving?

Temperature $T = -T_F$

($T_F > 0$, value depends on interaction, switch-on, …)

Hamiltonian $H_F = +t_0\hat{K} + U\hat{D}$

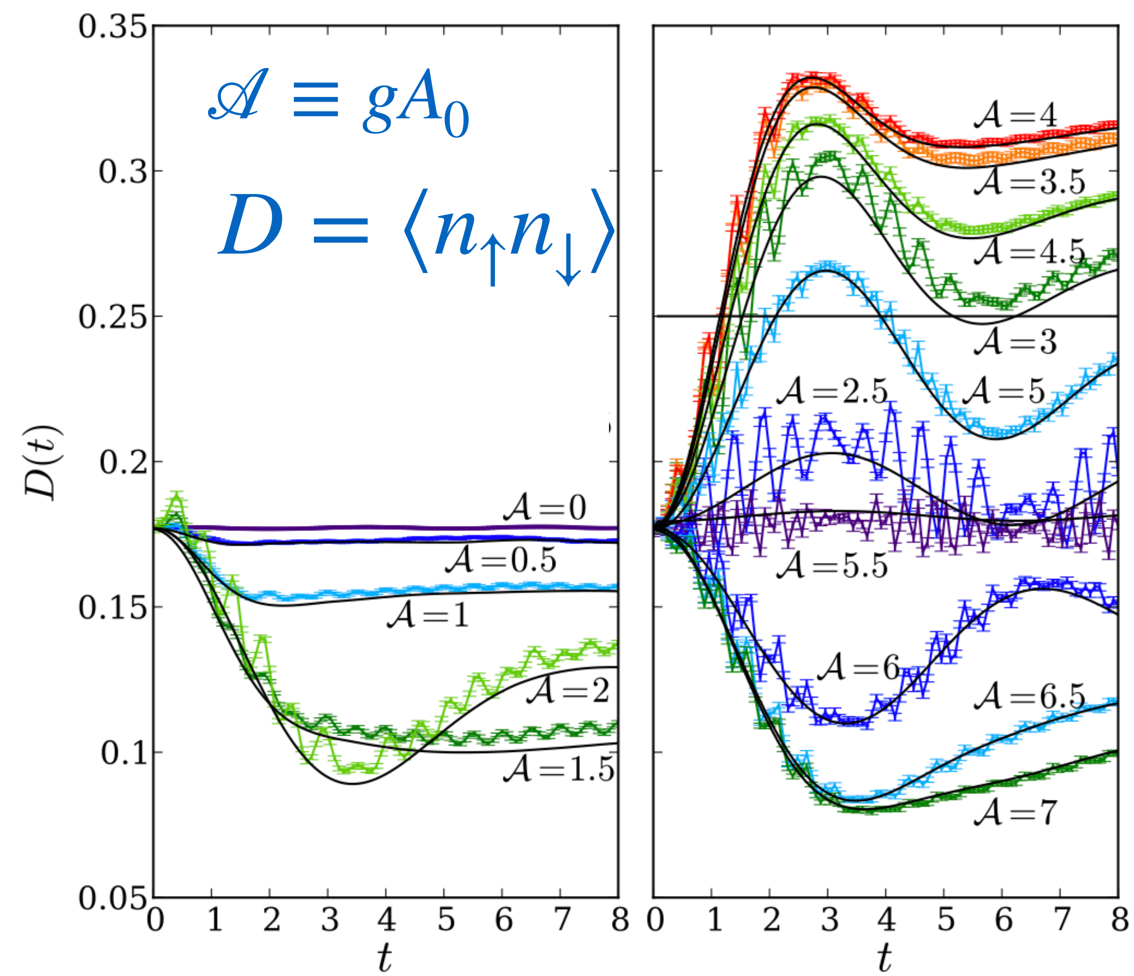
$\Downarrow$

$$\rho_F \propto e^{+\beta_f(t_0\hat{K}+U\hat{D})} = e^{-\beta_f(-t_0\hat{K}-U\hat{D})}$$

driven state thermalizes to same state as attractive model at positive temperature T_f

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

Time evolution under periodic driving:



$D(t) > \frac{1}{4}$ … thermalization at U<0

Dynamical band flipping in Hubbard model

Use this to realize superconductivity at high T? (Tc like for AFM!)

⇨ hard to find switch on protocol for driving to that T_f is low (even within effective Hamiltonian picture)

⇨ Effective Hamiltonian works for $\Omega \gg U, t_0$ … otherwise absorption leads to additional heating (see below)

⇨ Negative Temperature state unstable when coupled to other degrees of freedom

Nevertheless, this examples shows how driving a subset of degrees of freedom to a highly excited state, plus engineering of the Hamiltonian parameters, can be a route to reach new phases

Floquet Bloch bands

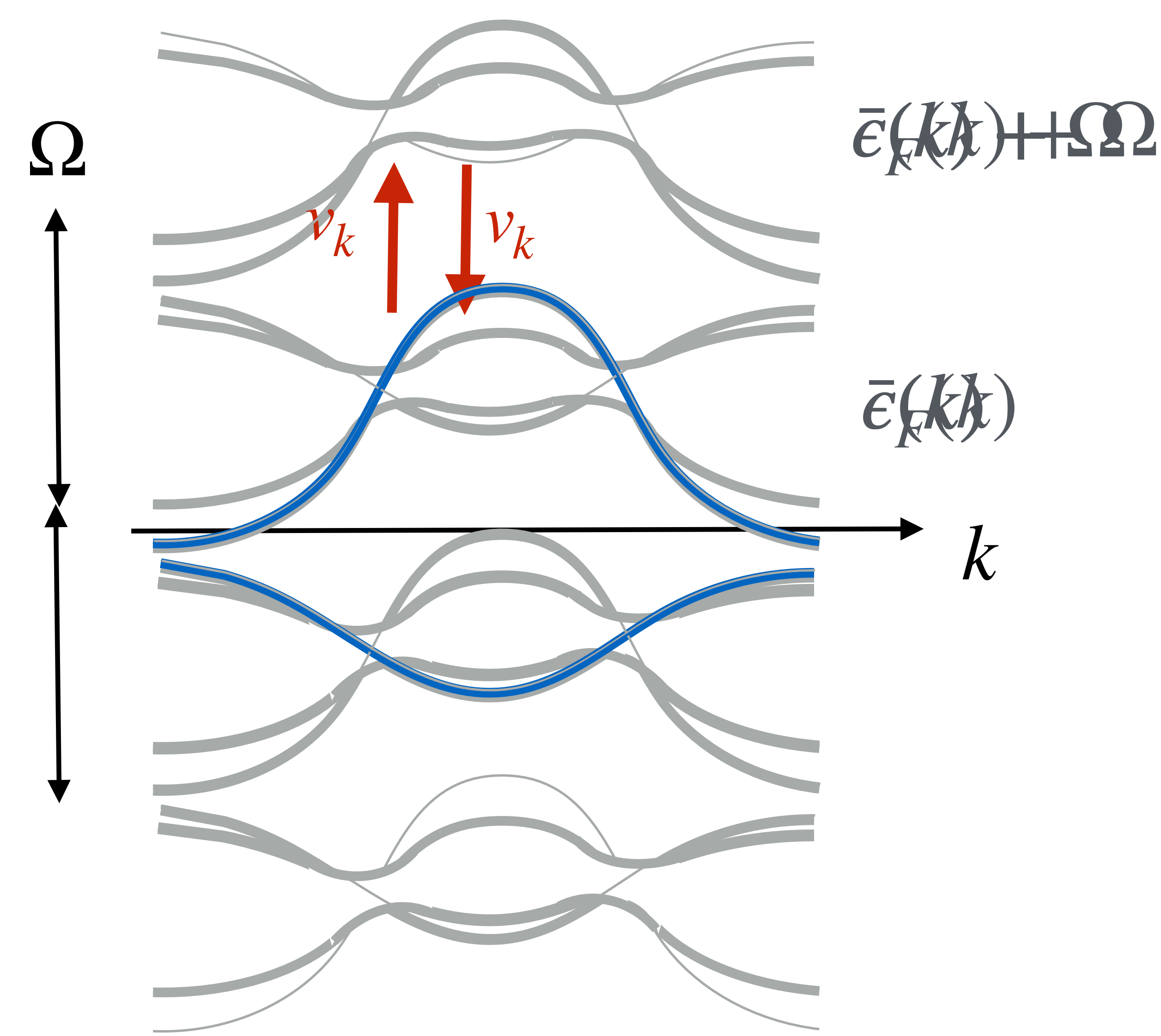
Floquet Bloch bands

Tight-binding model with external force $E(t) = -\partial_t A(t)$

$$H = \sum_k \underbrace{\epsilon_{ab}(k - eA(t))}_{\bar{\epsilon}_{ab}(k) + v^{(1)}_{ab}(k) e^{-i\Omega t} + \cdots} c^\dagger_{k,a} c_{k,b}$$

Floquet Blockmatrix:

$$\begin{pmatrix} \ddots & \vdots & & & \\ \cdots & \bar{\epsilon}_k + \Omega & v_k^{(1)} & v_k^{(2)} & \\ & v_k^{(1)} & \bar{\epsilon}_k & v_k^{(1)} & \\ & v_k^{(2)} & v_k^{(1)} & \bar{\epsilon}_k - \Omega & \cdots \\ & & & \vdots & \ddots \end{pmatrix} \begin{pmatrix} \vdots \\ u_k^{(1)} \\ u_k^{(0)} \\ u_k^{(-1)} \\ \vdots \end{pmatrix} = \epsilon_k^F \begin{pmatrix} \vdots \\ u_k^{(1)} \\ u_k^{(0)} \\ u_k^{(-1)} \\ \vdots \end{pmatrix}$$

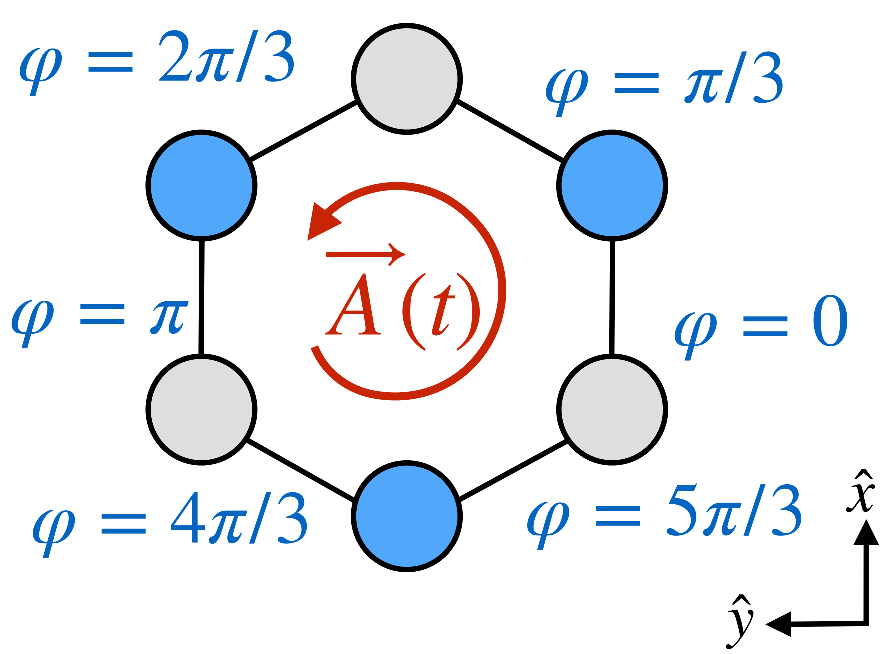


Mixing of Floquet bands due to virtual photon absorption emission

Nontrivial example: Kitaev model on Honeycomb lattice

Oka and Aoki, Phys. Rev. B **79**, 081406(R) (2009); Lindner, Refael, Galitski, Nature Physics **7**, 490 (2011)

Honeycomb lattice driven by circularly polarized light:



$$t_{ab} = t_0\, e^{i\phi_{ab}(t)} \qquad \phi_{ab} = e\vec{A}(t)\cdot(\vec{R}_a - \vec{R}_b)$$

circular polarized light:

$$\vec{A}(t) = \hat{x}A_0\cos(\Omega t) + \hat{y}A_0\sin(\Omega t)$$

$$\Rightarrow\ \phi_{ab} = A_0\cos(\Omega t + \varphi_{ab})$$

Fourier transform:

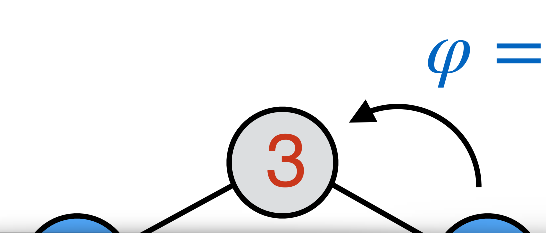
$$t_{ab}^{(n)} = \frac{1}{T}\int_0^T dt\, e^{in\Omega t + i\phi_{ab}(t)} = e^{-in\varphi_{ab}} \underbrace{\frac{1}{T}\int_0^T dt\, e^{in\Omega t + iA_0\cos(\Omega t)}}_{=J_n(A_0)}$$

Nontrivial example: Kitaev model on Honeycomb lattice

Oka and Aoki, Phys. Rev. B **79**, 081406(R) (2009); Lindner, Refael, Galitski, Nature Physics **7**, 490 (2011)

$$\Rightarrow \quad \sum_{n=-\infty}^{\infty} H_{-n}\frac{1}{n\Omega}H_n = \ ??$$

(1st oder HF correction to time averaged H)

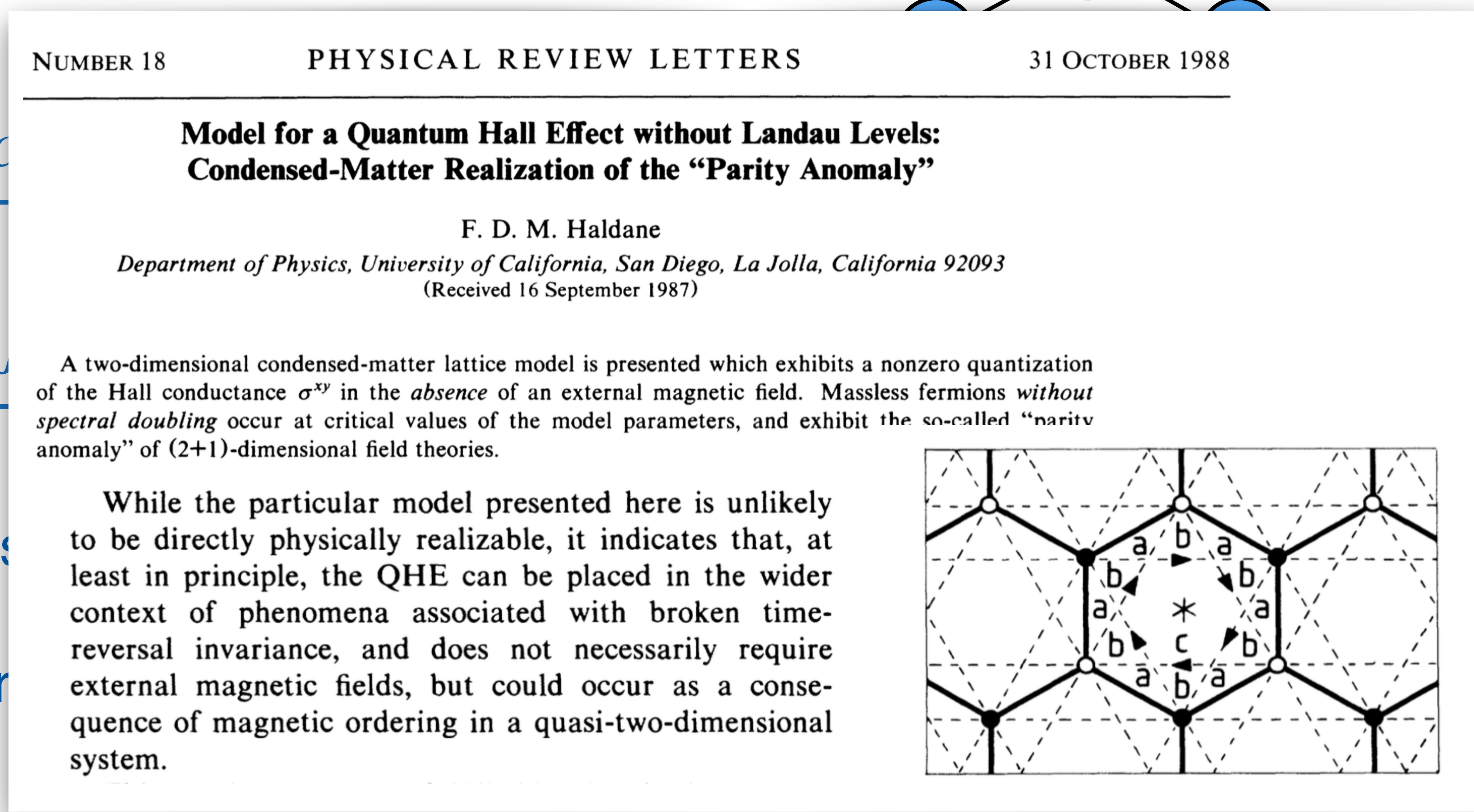


$$\frac{[t_{32}^{(-1)} c_3^\dagger c_2, c_2^\dagger c \ldots}{n\Omega}$$

$$= c_3^\dagger c_1 \frac{|J \ldots}{\ldots}$$

complex s…

Floquet-r…



NUMBER 18 PHYSICAL REVIEW LETTERS 31 OCTOBER 1988

Model for a Quantum Hall Effect without Landau Levels: Condensed-Matter Realization of the "Parity Anomaly"

F. D. M. Haldane

Department of Physics, University of California, San Diego, La Jolla, California 92093

(Received 16 September 1987)

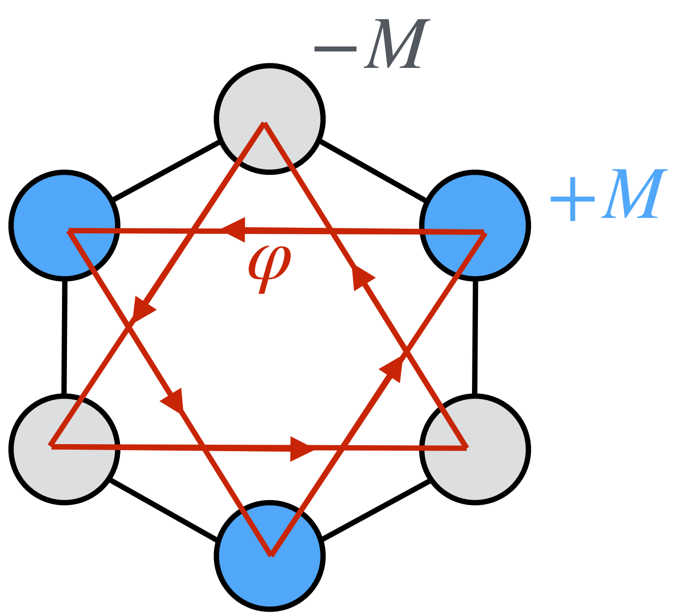
A two-dimensional condensed-matter lattice model is presented which exhibits a nonzero quantization of the Hall conductance σ^{xy} in the *absence* of an external magnetic field. Massless fermions *without spectral doubling* occur at critical values of the model parameters, and exhibit the so-called "parity anomaly" of (2+1)-dimensional field theories.

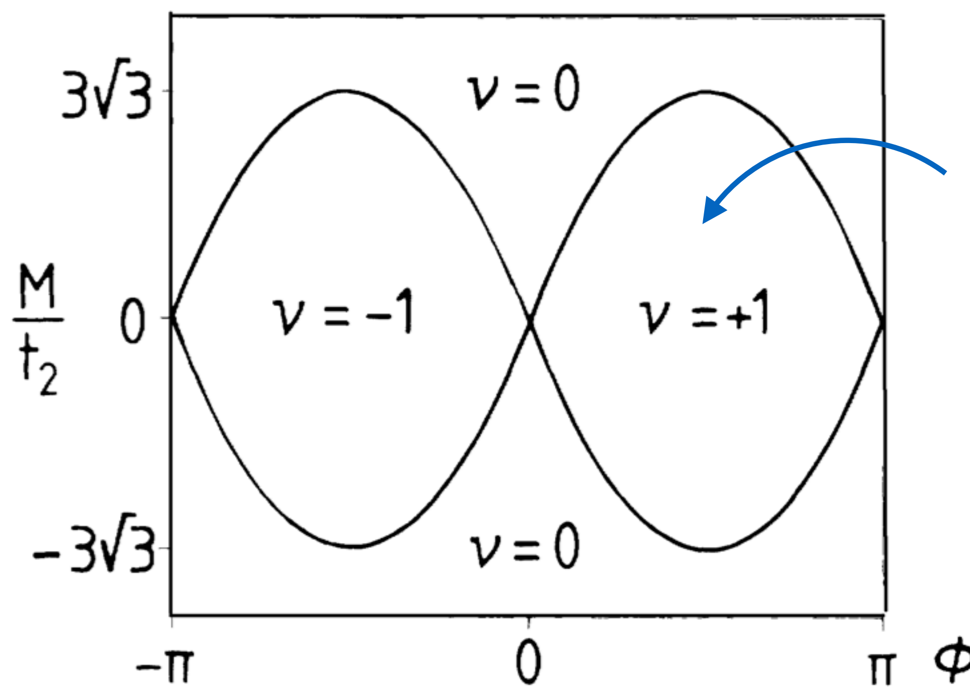
While the particular model presented here is unlikely to be directly physically realizable, it indicates that, at least in principle, the QHE can be placed in the wider context of phenomena associated with broken time-reversal invariance, and does not necessarily require external magnetic fields, but could occur as a consequence of magnetic ordering in a quasi-two-dimensional system.

Experimental realization

Haldane model

Haldane (1988)





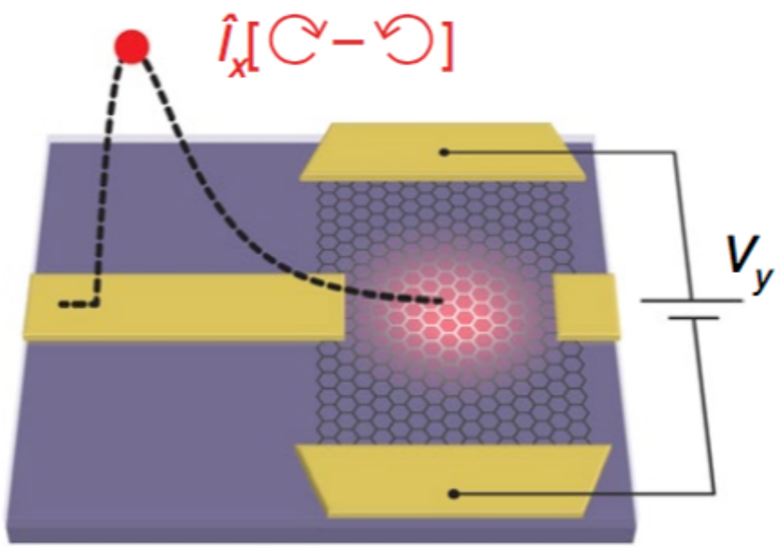
zero field Hall effect

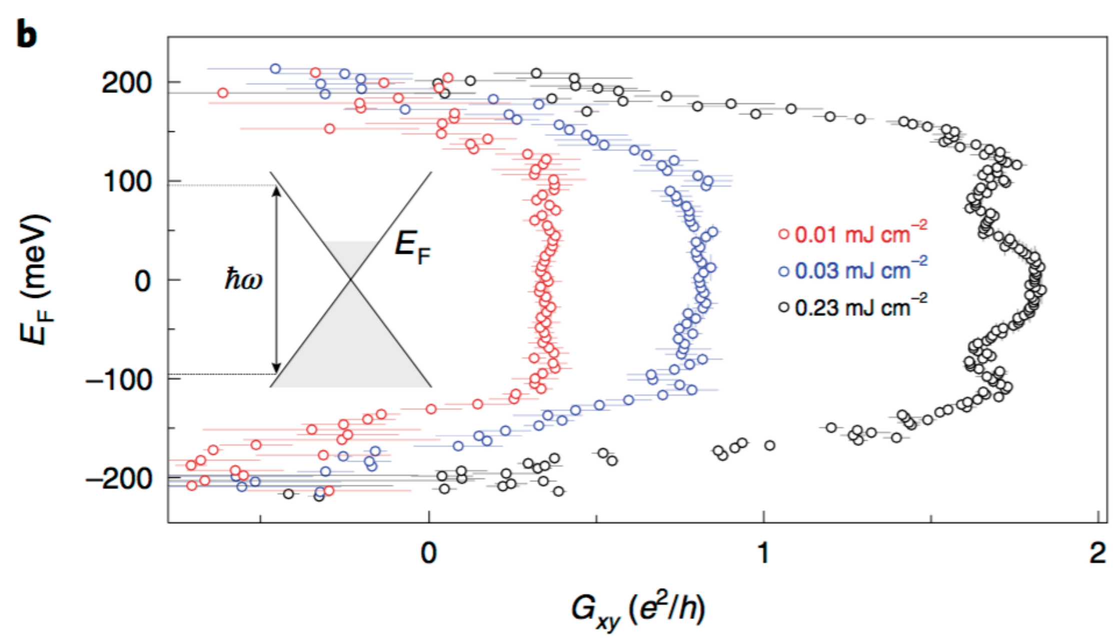
$$\sigma_{xy} = \nu \frac{e^2}{h}$$

Experiment:

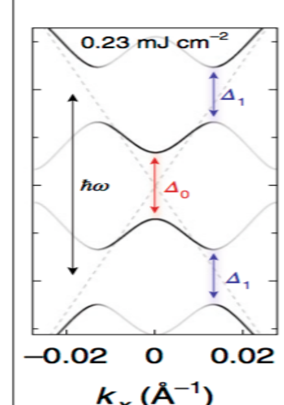
Graphene: “Light-induced anomalous Hall effect”

McIver et al., Nat Phys **16**, 38 (2020)





Floquet bands



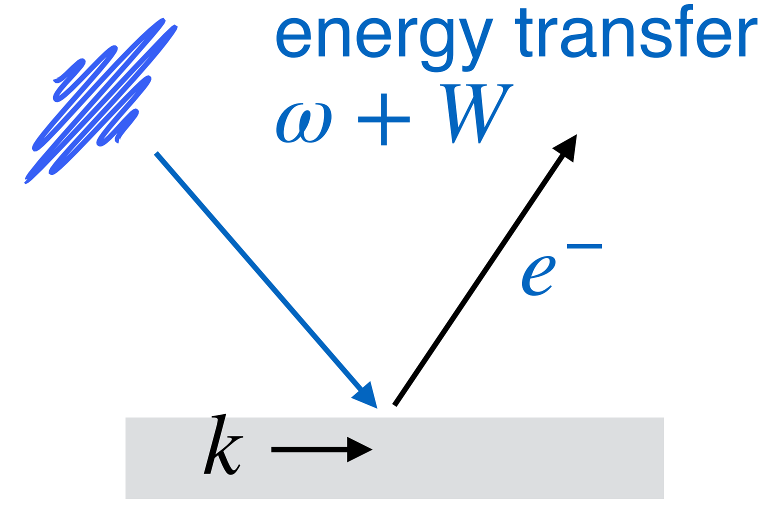
Cold atoms:

Jotzu et al., Nature **515**, 237 (2014)

Seeing Floquet Bloch bands in trARPES?

Simple theory of ARPES: (for a long probe)

$$A^{<}(\omega) = \lim_{T\to\infty} \frac{1}{T}\int_0^T dt \int ds \underbrace{\langle c_k^\dagger(t+s)c_k(t)\rangle}_{\text{hole propagator}} e^{i\omega s}$$



Driven (noninteracting!) system

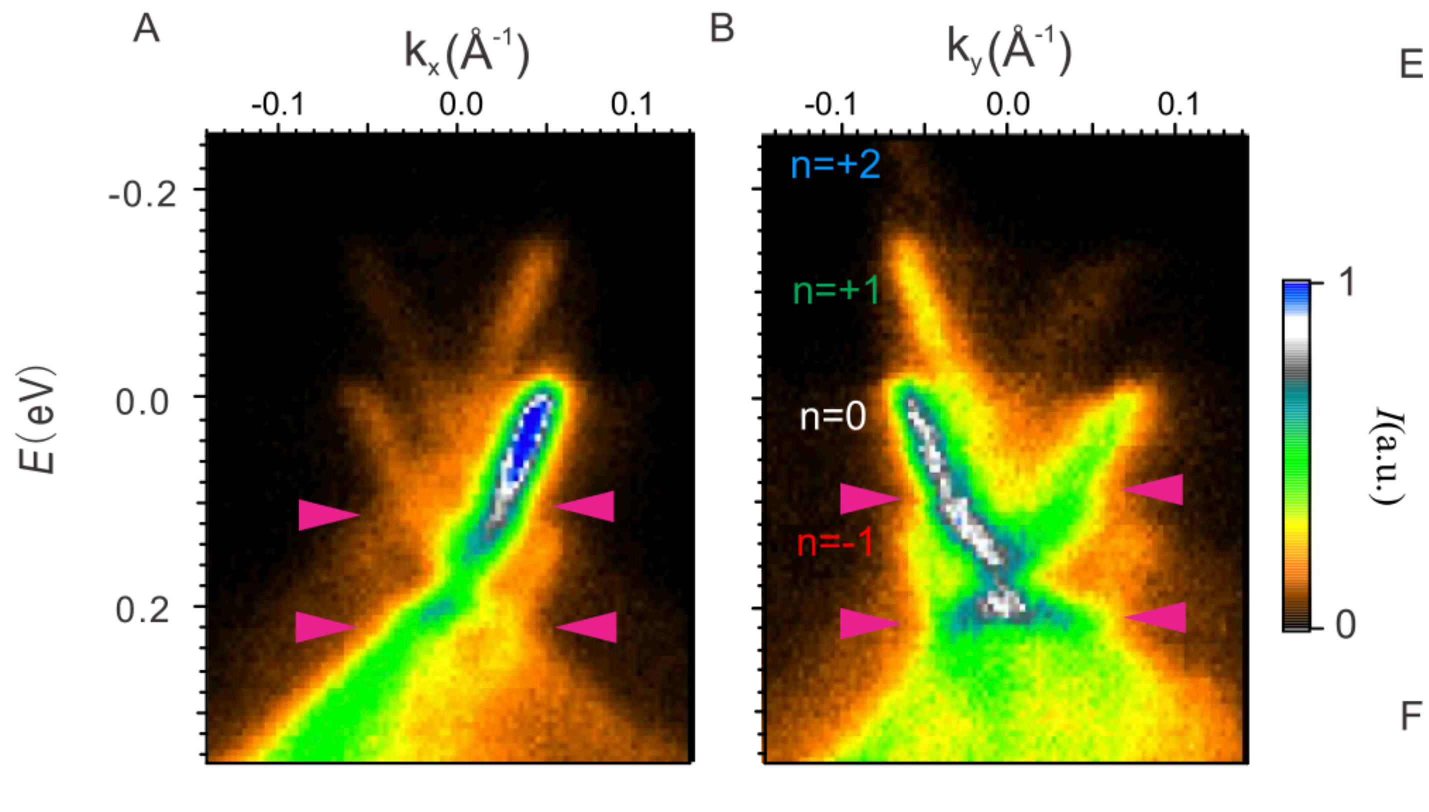
$$i\partial_t c_k(t) = \epsilon(k - gA(t))c_k(t) \quad \Rightarrow \quad c_k(t) = e^{-i\epsilon_k^F t}\sum_n u_{k,n} e^{-in\Omega t} c_k$$

$$A(\omega) \propto \sum_n |u_{k,n}|^2 \delta(\omega - \epsilon_F - n\Omega)\langle c_k^\dagger c_k\rangle$$

Sidebands, weight $\propto |u_{k,n}|^2$

Seeing Floquet Bloch bands in trARPES?

Floquet Bloch bands in BiSe₃ Wang et al, Science (2013)



proper theory of trARPES, using NEGF needed

Problems:

- Disentangling from LAPE
- Decoherence

  see also Aeschlimann et al. (2021)

$$A(\omega) = \lim_{T\to\infty} \frac{1}{T} \int_0^T dt \int ds \, \underbrace{\langle c_k^\dagger(t+s) c_k(t) \rangle}_{\sim\, e^{-\gamma|s|}} e^{i\omega s}$$

- Heating (or non-thermal steady distribution under driving)