Theoretical simulations of pump-probe spectroscopies in solids

Michael A. Sentef

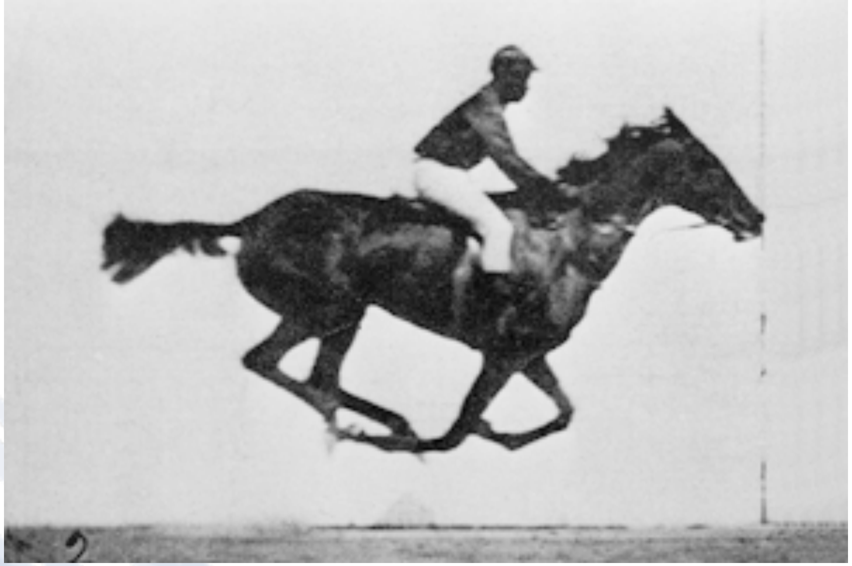
lab.sentef.org

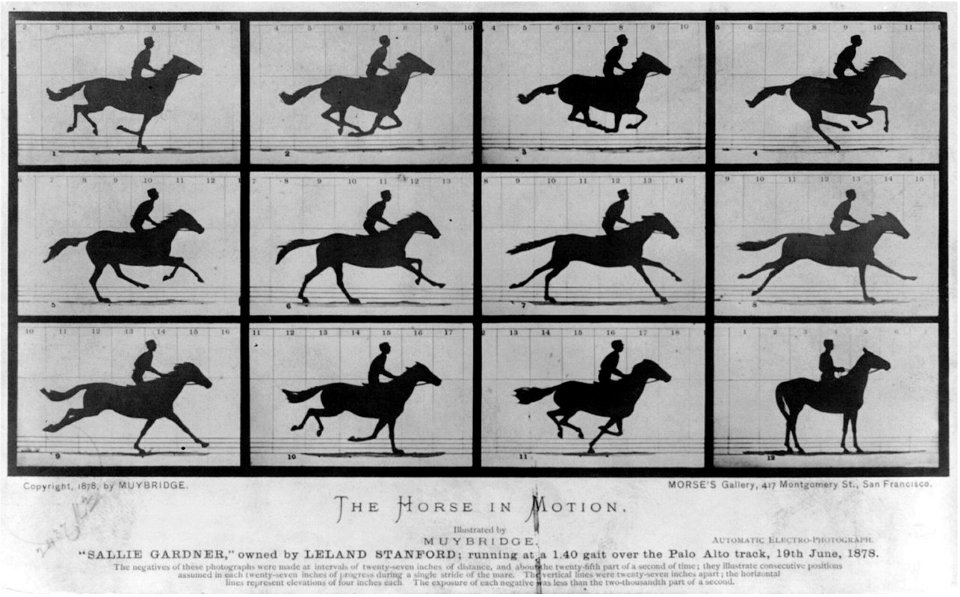
Max-Planck Institute for the Structure and Dynamics of Matter, Hamburg

DPG Spring Meeting, Dresden, March 2017

Pump-probe spectroscopy (1887)

- stroboscopic investigations of dynamic phenomena





Copyright, 1878, by MUYBRIDGE.

MORSE'S Gallery, 417 Montgomery St., San Francisco.

THE HORSE IN MOTION.

Illustrated by
MUYBRIDGE.

AUTOMATIC ELECTRO-PHOTOGRAPH.

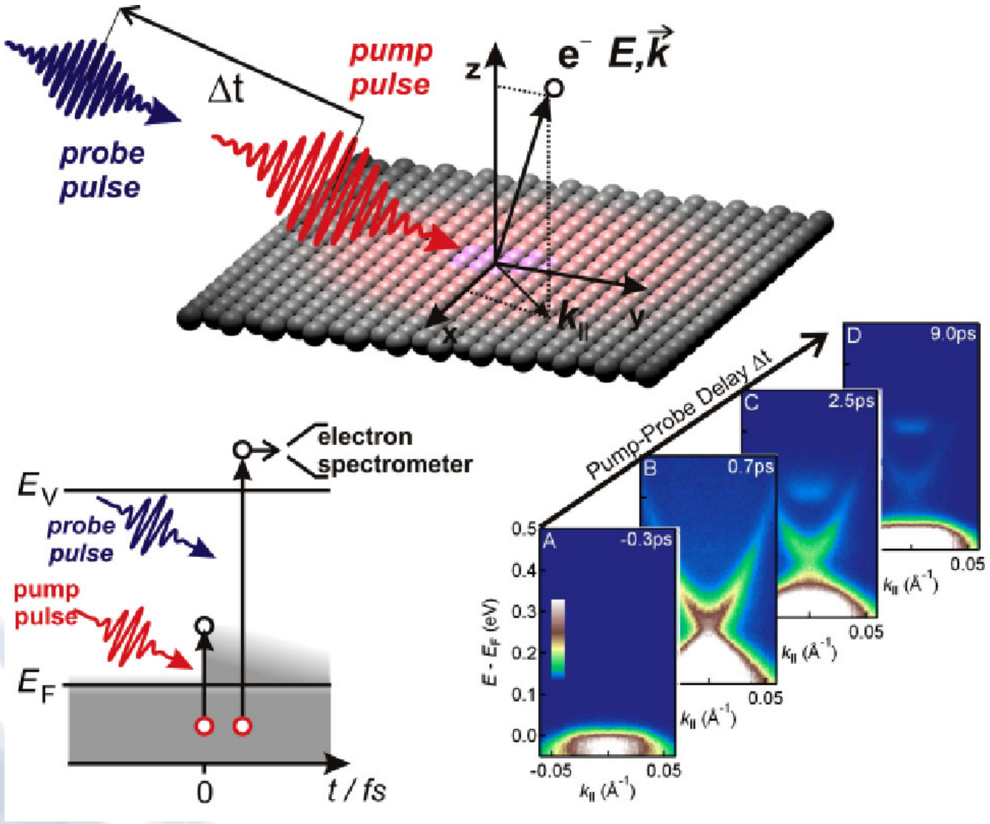
"SALLIE GARDNER," owned by LELAND STANFORD; running at a 1.40 gait over the Palo Alto track, 19th June, 1878.

The negatives of these photographs were made at intervals of twenty-seven inches of distance, and about the twenty-fifth part of a second of time; they illustrate consecutive positions assumed in each twenty-seven inches of progress during a single stride of the mare. The vertical lines were twenty-seven inches apart; the horizontal lines represent elevations of four inches each. The exposure of each negative was less than the two-thousandth part of a second.

Muybridge 1887

Pump-probe spectroscopy (today)

- stroboscopic investigations of dynamic phenomena



TbTe3 CDW metal

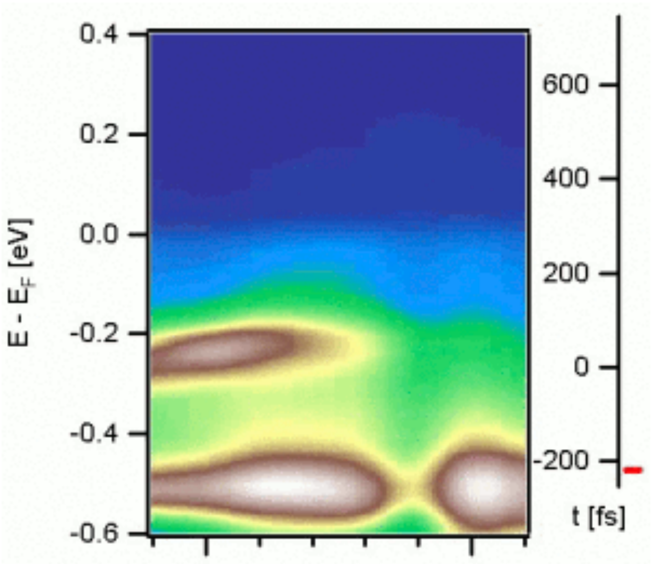
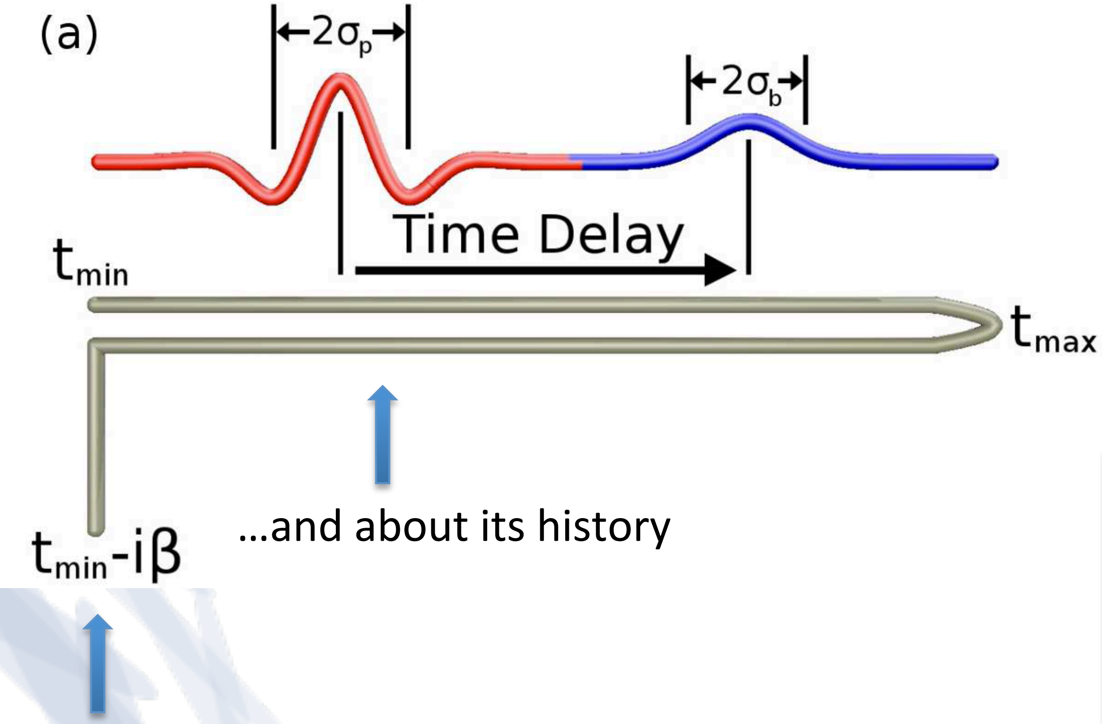


Image courtesy:
J. Sobota / F. Schmitt

Non-Equilibrium Keldysh Formalism

$$G_k(\omega) = G_k^0(\omega) + G_k^0(\omega)\underline{\Sigma(\omega)}G_k(\omega)$$

$$G_{\mathbf{k}}(t,t') = G^0_{\mathbf{k}}(t,t') + \oint dt_1\, dt_2\, G^0_{\mathbf{k}}(t,t_1)\Sigma(t_1,t_2)G_{\mathbf{k}}(t_2,t')$$



…and about its history

System knows about its thermal initial state…

self-energy Σ:
electron-electron scattering
electron-phonon scattering
...

same problem as in equilibrium (but worse):
use your favorite self-energy approximation, e.g. perturbation theory, nonequilibrium DMFT, ...

Include the effects of driving field through time-dependent electronic dispersion

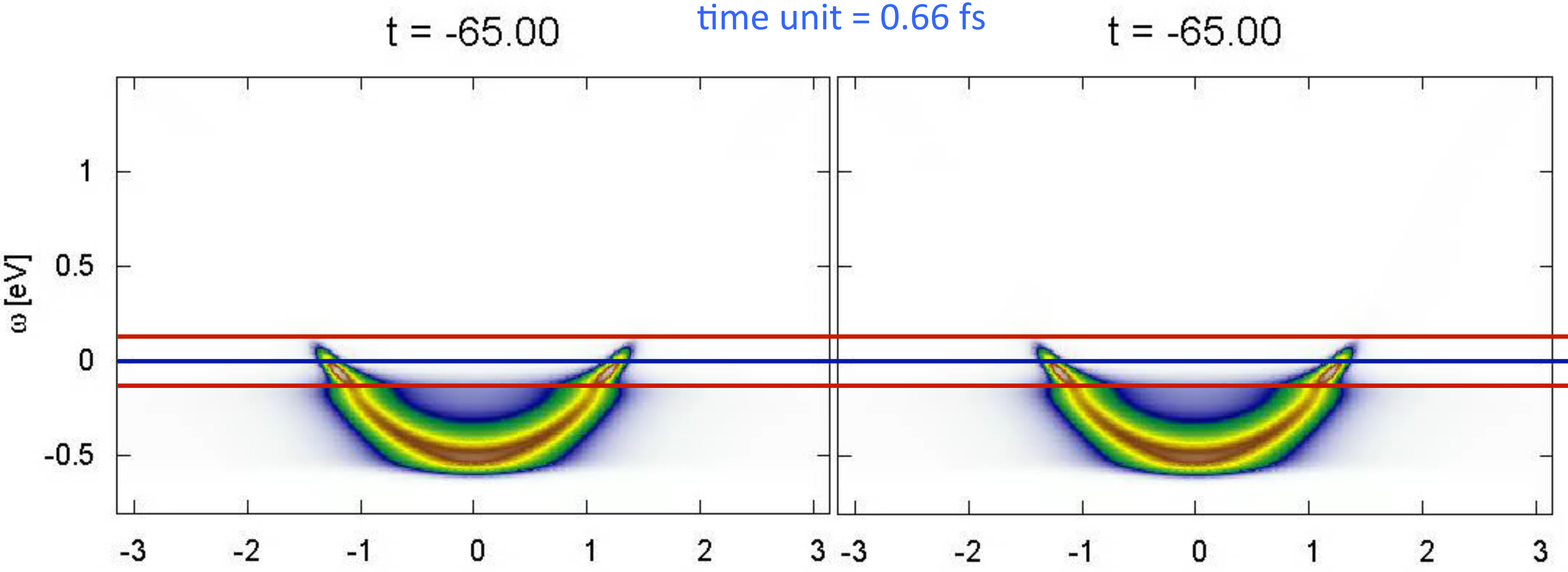
$$\varepsilon(k) \rightarrow \varepsilon(k,t)$$

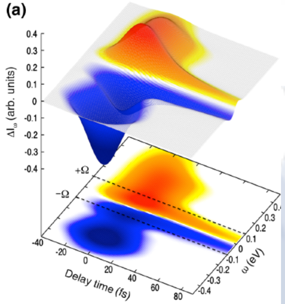
Electron-boson coupling

PRX 3, 041033 (2013)

Weak pump

Strong pump





boson window effect for fast versus slow relaxation

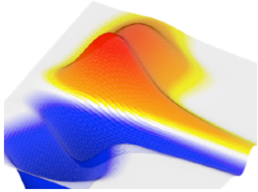
nonlinear response for strong pump

Ultrafast Materials Science today

Understanding the nature of quasiparticles

- Relaxation dynamics
- Control of couplings

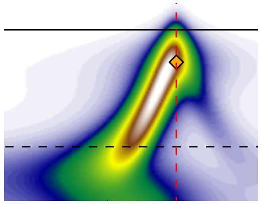
PRL 111, 077401 (2013)
PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
Nature Commun. 7, 13761 (2016)
PRB 95, 024304 (2017)
arXiv:1702.00952



Understanding ordered phases

- Collective oscillations
- Competing order parameters

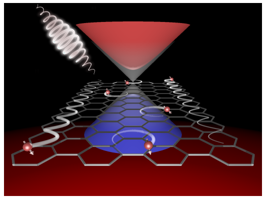
PRB 92, 224517 (2015)
PRB 93, 144506 (2016)
PRL 118, 087002 (2017)



Creating new states of matter

- Photo-induced phase transitions
- Floquet topological states

Nature Commun. 6, 7047 (2015)
Nature Commun. 8, 13940 (2017)



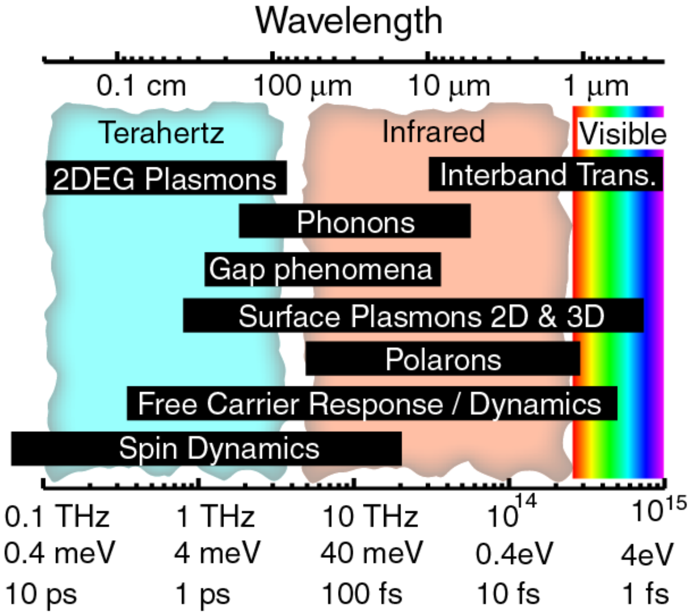


Image courtesy:
D. Basov

Outline

- Part I: Floquet engineering of topological solids
  - Floquet Chern insulator in graphene

    Nature Commun. 6, 7047 (2015)
  - Floquet-Weyl semimetal in Na_3Bi

    Nature Commun. 8, 13940 (2017)
- Part II: Light-enhanced electron-phonon coupling

  PRB 95, 024304 (2017)
  arXiv:1702.00952
- Part III: Laser-controlled competing orders

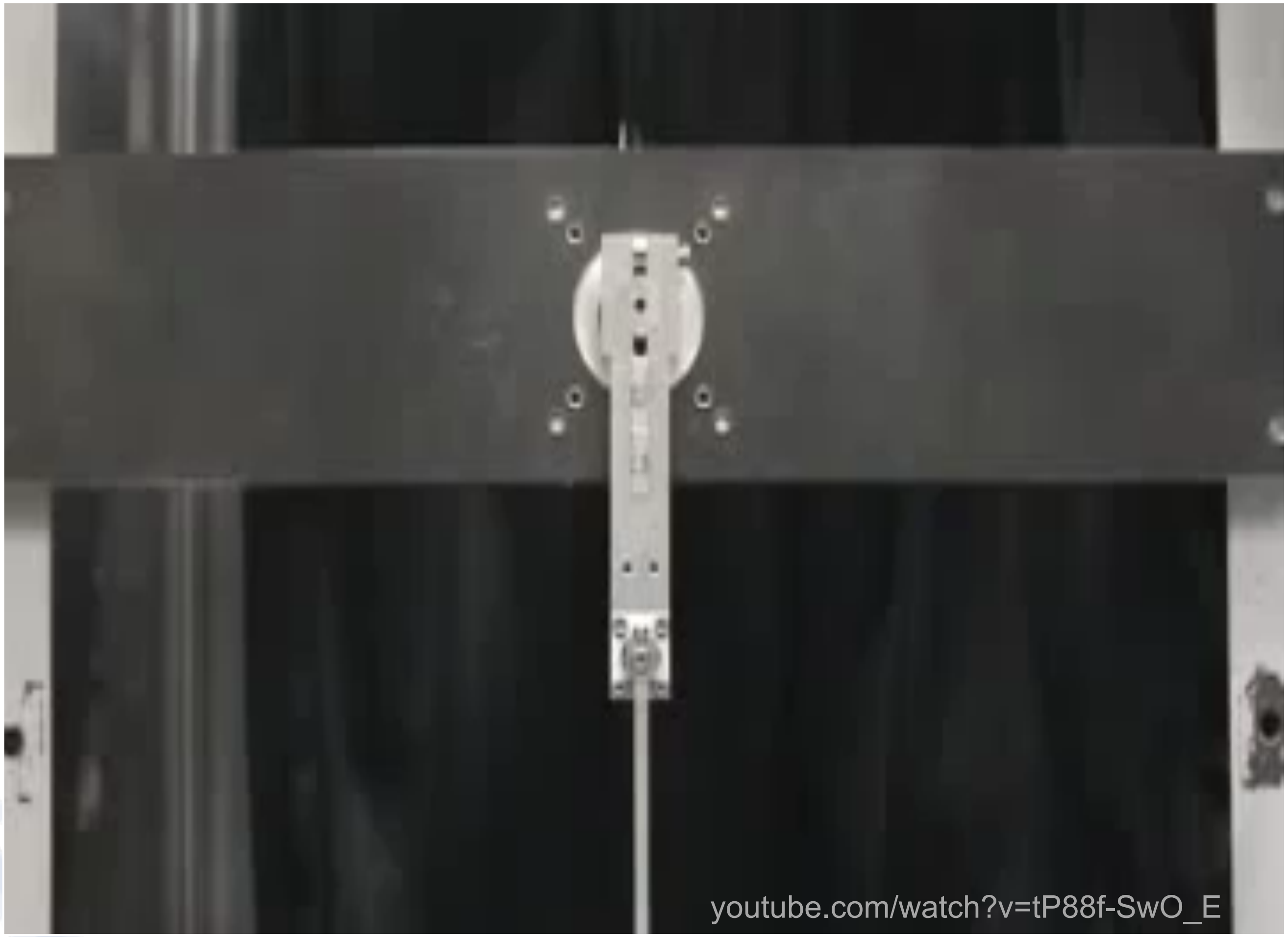
  PRL 118, 087002 (2017)

„as time permits“

I. Floquet engineering in solids

Driven is different

Kapitza pendulum



youtube.com/watch?v=tP88f-SwO_E

dynamical stabilization of a metastable state

Driven is interesting

ω -> infinity

Kapitza class, dynamical stabilization

Bukov, d'Alessio, Polkovnikov, Adv. Phys. 64, 139-226 (2015)

ω -> finite but largest scale

Floquet engineering

ω -> resonances

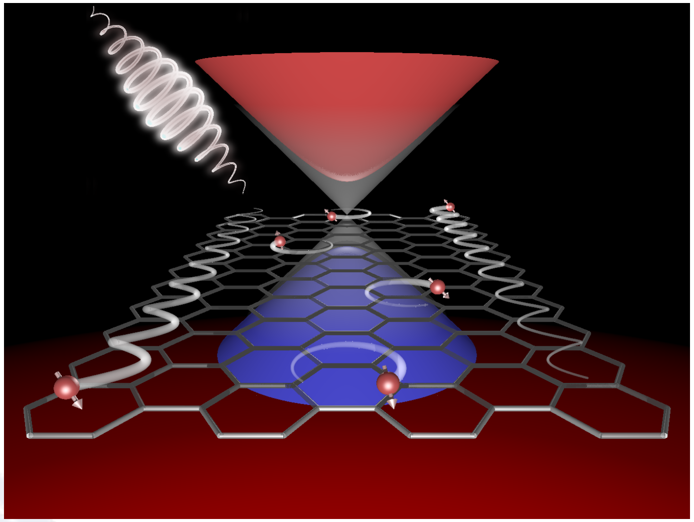
sidebands, huge effects, detuning

ω -> smallest

dc physics, adiabatic evolution

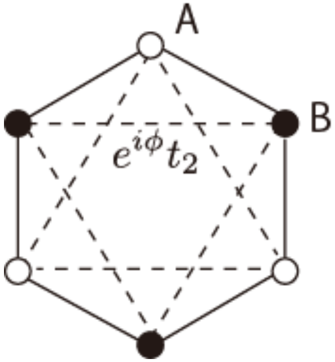
Floquet topological states

Graphene + circularly polarized light (breaks trs)

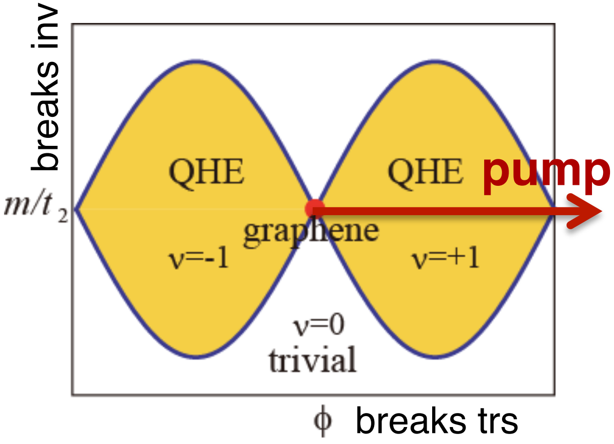


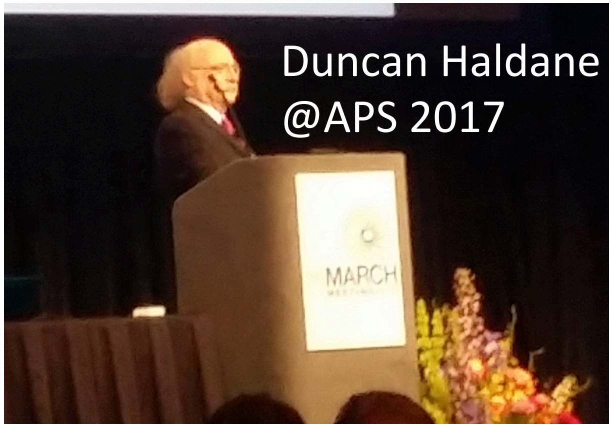
?

Haldane model (PRL 61, 2015 (1988))



Local flux ϕ
Staggered field *m*
Fictitious fields!





Duncan Haldane
@APS 2017

Floquet engineering in a nutshell

time periodic system

$$i\partial_t\psi = H(t)\psi \qquad H(t) = H(t+T) \qquad \Omega = 2\pi/T$$

“Floquet mapping”
= Bloch state in time

$$\Psi(t) = e^{-i\varepsilon t}\sum_m \phi^m e^{-im\Omega t}$$

Floquet Hamiltonian (static eigenvalue problem)

$$\sum_{m=-\infty}^{\infty} \mathcal{H}^{mn}\phi_\alpha^m = \varepsilon_\alpha \phi_\alpha^n$$

ε: Floquet quasi-energy

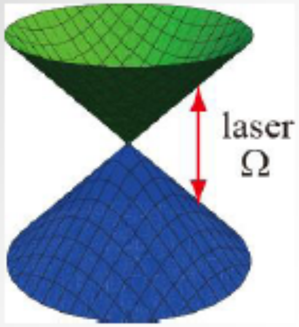
$$(\mathcal{H})^{mn} = \frac{1}{T}\int_0^T dt H(t) e^{i(m-n)\Omega t} \boxed{+ m\delta_{mn}\Omega I}$$

~ absorption of *m* “photons”

slides courtesy of Takashi Oka

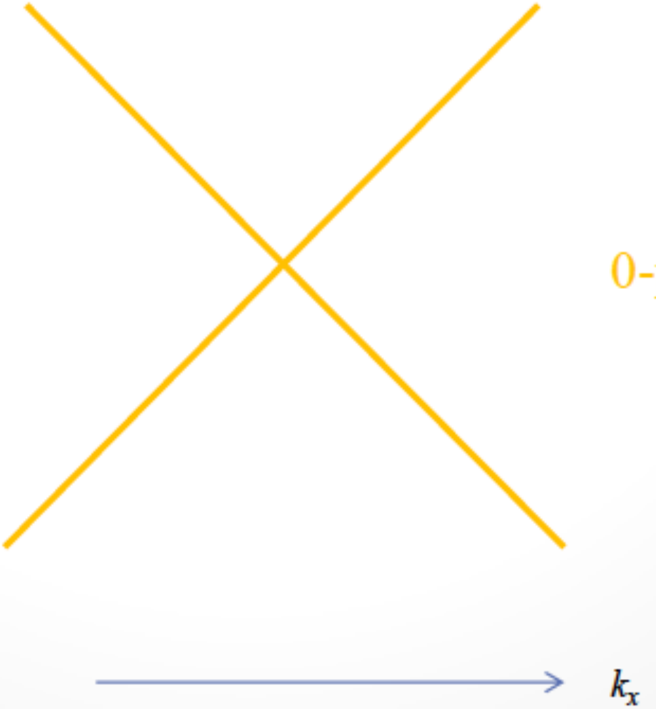
Floquet spectrum: Dirac model + circularly polarized laser

TO, Aoki 2009



$$
H^{\text{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}
$$

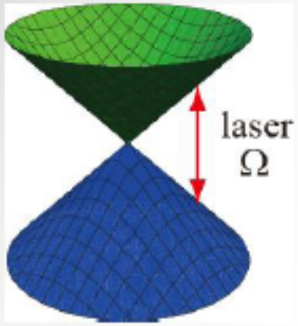
0-photon absorbed state



0-photon absorbed state

Floquet spectrum: Dirac model + circularly polarized laser

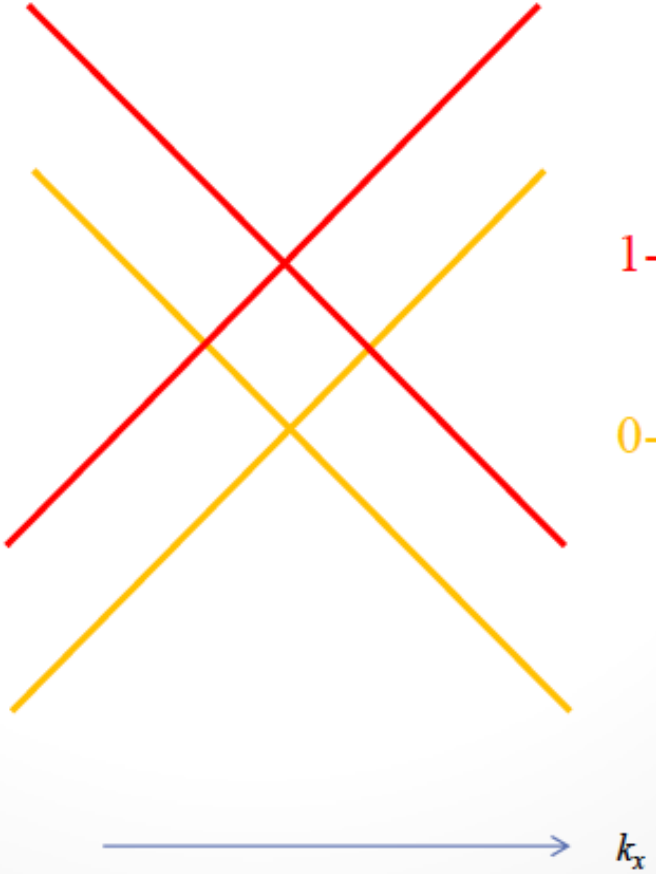
TO, Aoki 2009



$$H^{\mathrm{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

1-photon absorbed state

0-photon absorbed state

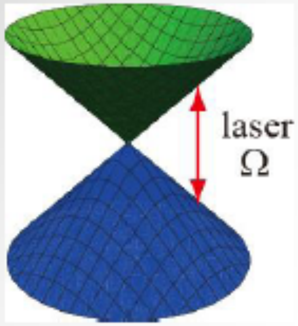


1-photon absorbed state

0-photon absorbed state

Floquet spectrum: Dirac model + circularly polarized laser

TO, Aoki 2009

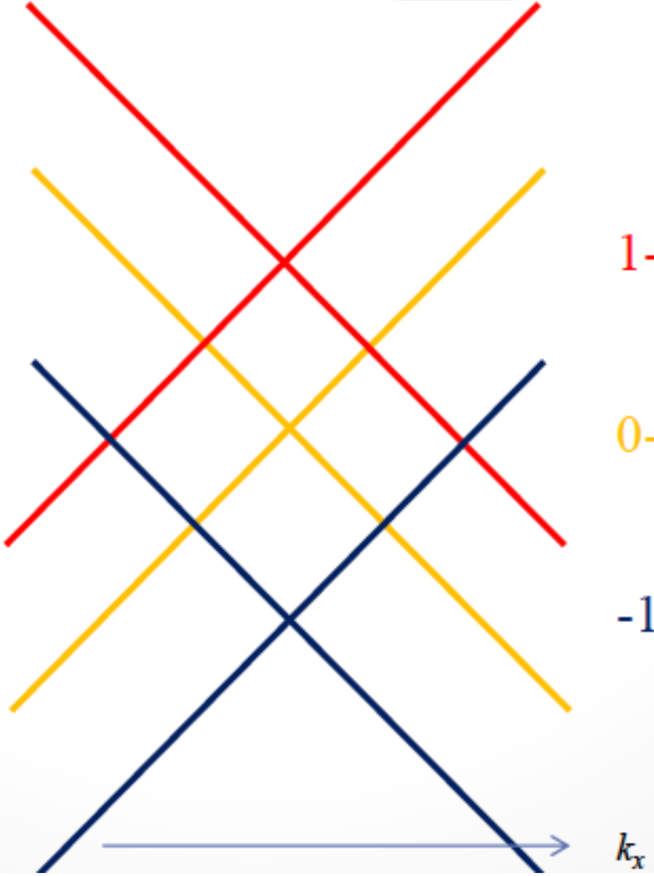


$$H^{\text{Floquet}} = \begin{pmatrix} \boxed{\begin{matrix} \Omega & k \\ \bar{k} & \Omega \end{matrix}} & \begin{matrix} 0 & A \\ 0 & 0 \end{matrix} & \begin{matrix} 0 & 0 \\ 0 & 0 \end{matrix} \\ \begin{matrix} 0 & 0 \\ A & 0 \end{matrix} & \boxed{\begin{matrix} 0 & k \\ \bar{k} & 0 \end{matrix}} & \begin{matrix} 0 & A \\ 0 & 0 \end{matrix} \\ \begin{matrix} 0 & 0 \\ 0 & 0 \end{matrix} & \begin{matrix} 0 & 0 \\ A & 0 \end{matrix} & \boxed{\begin{matrix} -\Omega & k \\ \bar{k} & -\Omega \end{matrix}} \end{pmatrix}$$

1-photon absorbed state

0-photon absorbed state

-1-photon absorbed state



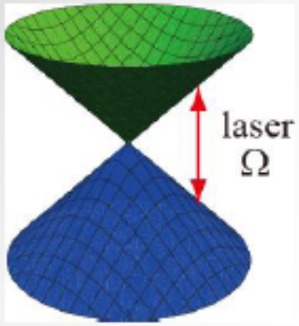
1-photon absorbed state

0-photon absorbed state

-1-photon absorbed state

Floquet spectrum: Dirac model + circularly polarized laser

TO, Aoki 2009

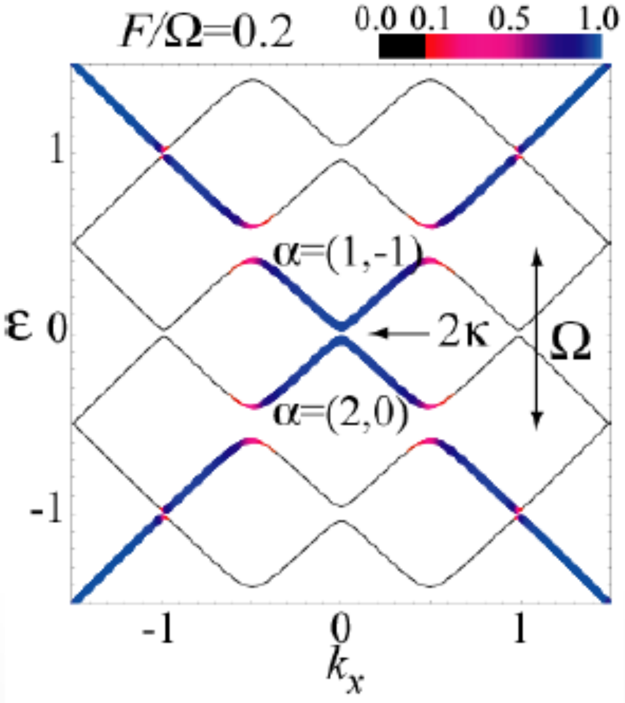


$$H^{\text{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

1-photon absorbed state

0-photon absorbed state

-1-photon absorbed state



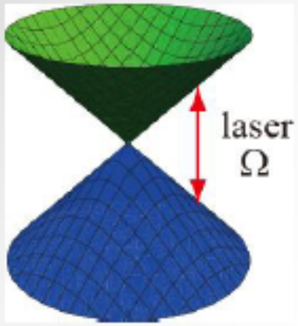
1-photon absorbed state

0-photon absorbed state

-1-photon absorbed state

Floquet spectrum: Dirac model + circularly polarized laser

TO, Aoki 2009



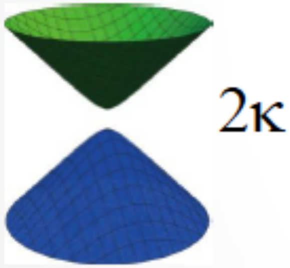
$$H^{\text{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

1-photon absorbed state

0-photon absorbed state

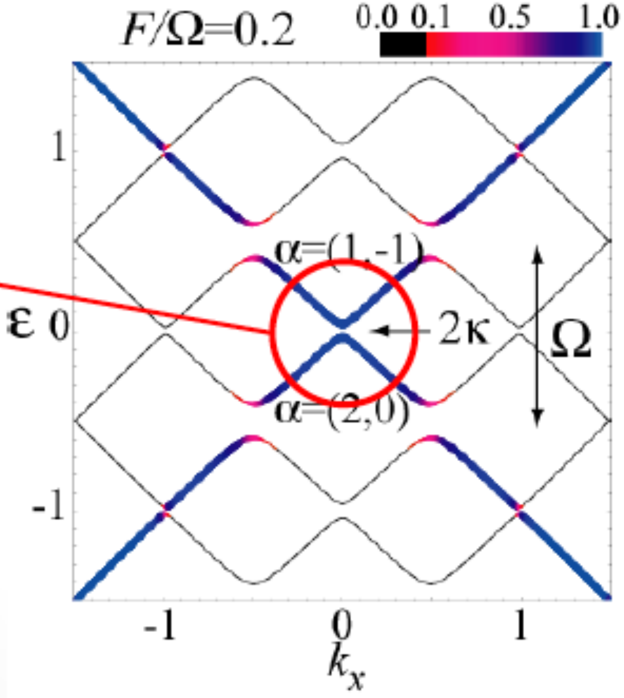
-1-photon absorbed state

near Dirac point



Dirac gap

$$\kappa = \frac{\sqrt{4A^2+\Omega^2}-\Omega}{2} \sim A^2/\Omega$$



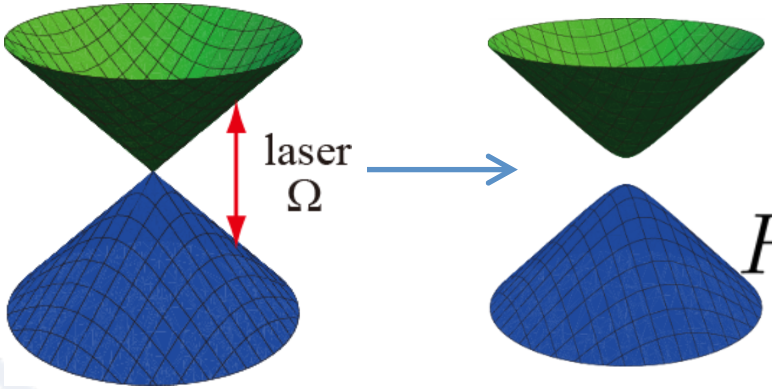
1-photon absorbed state

0-photon absorbed state

-1-photon absorbed state

Dirac fermion + circularly polarized laser

Mass term = energy gap =
synthetic field stemming from a real time-dependent field A(t)



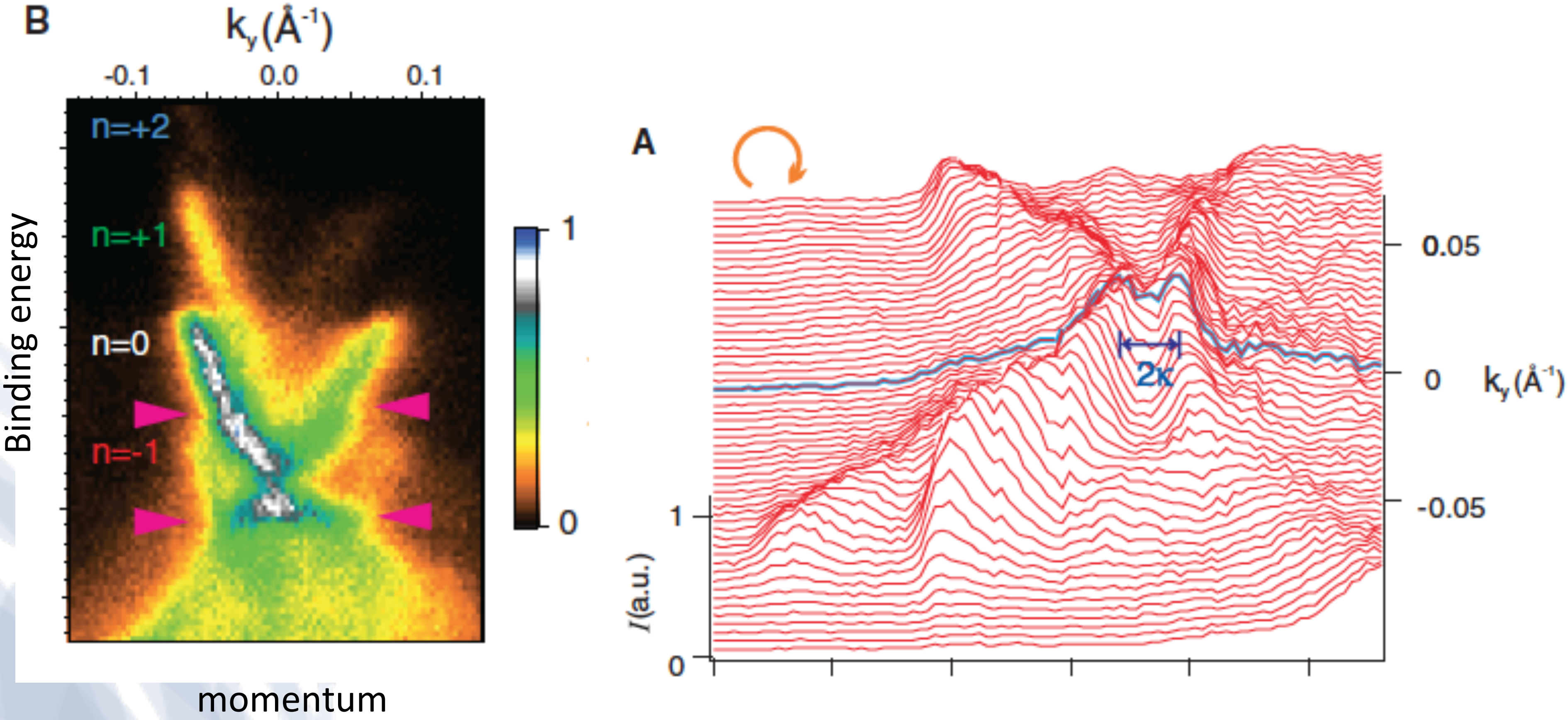
$$H_{\mathrm{eff}} = \underbrace{H_0}_{\text{Kapitza}} + \overbrace{\boxed{\frac{[\overset{\sim A\sigma_-}{\boxed{H_{-1}}}, \overset{\sim A\sigma_+}{\boxed{H_1}}]}{\Omega}}}^{\text{Floquet engineering}} + \mathcal{O}(A^4)$$

Oka and Aoki,
PRB 79, 081406 (2009)

Observation of Floquet-Bloch States on the Surface of a Topological Insulator

Y. H. Wang,* H. Steinberg, P. Jarillo-Herrero, N. Gedik†

Bi_2Se_3
Science ***342****, 453 (2013)*

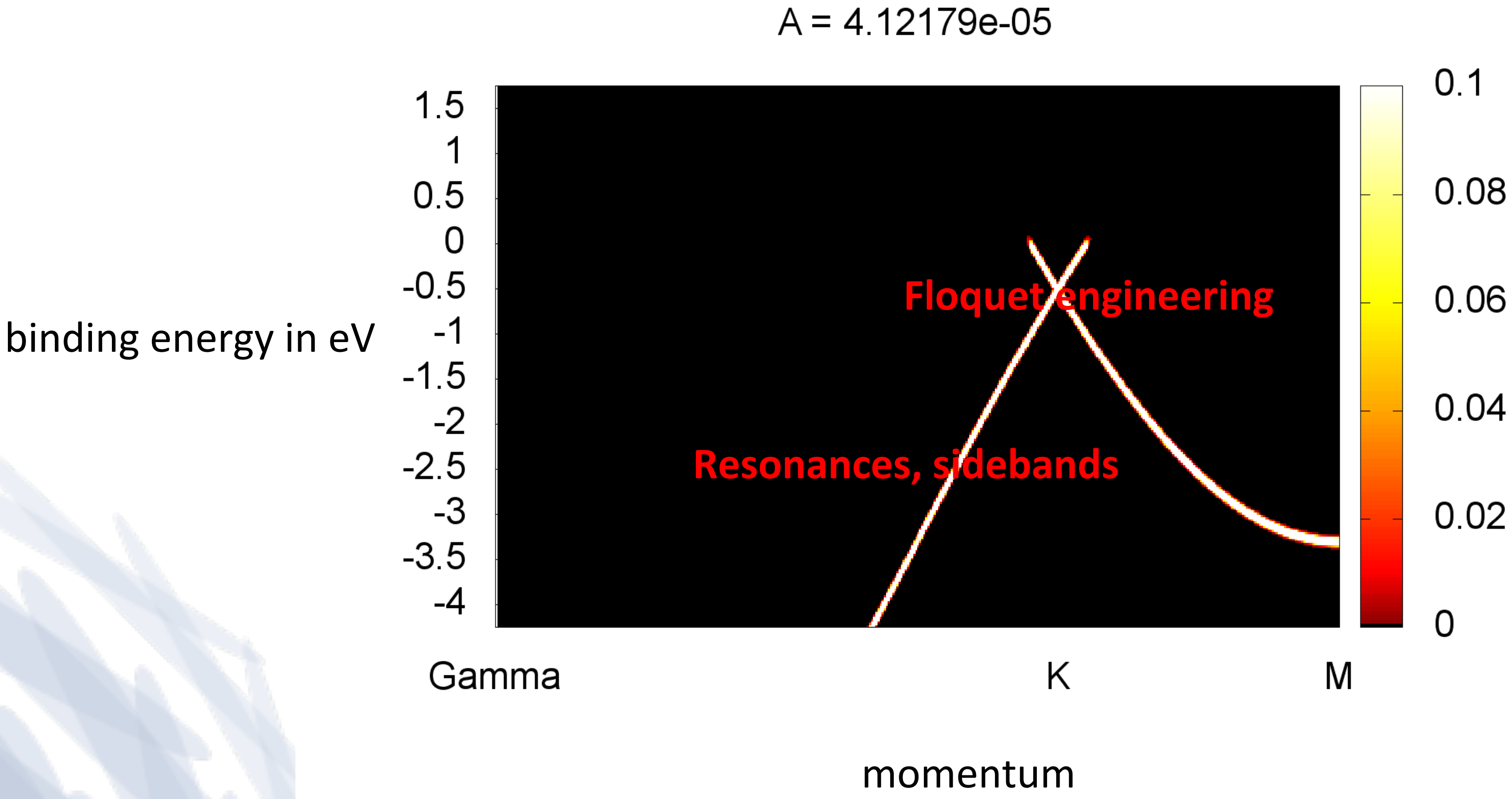


Binding energy

Floquet-Bloch states in graphene

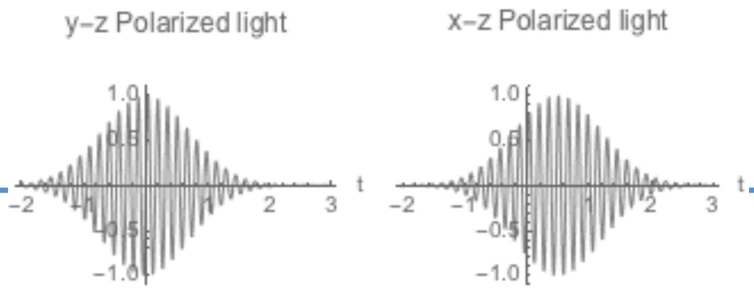
Tight-binding model + nonequilibrium Keldysh formalism:
Time-resolved ARPES during 1.5 eV circularly polarized laser pulse

Turning graphene into a Chern insulator

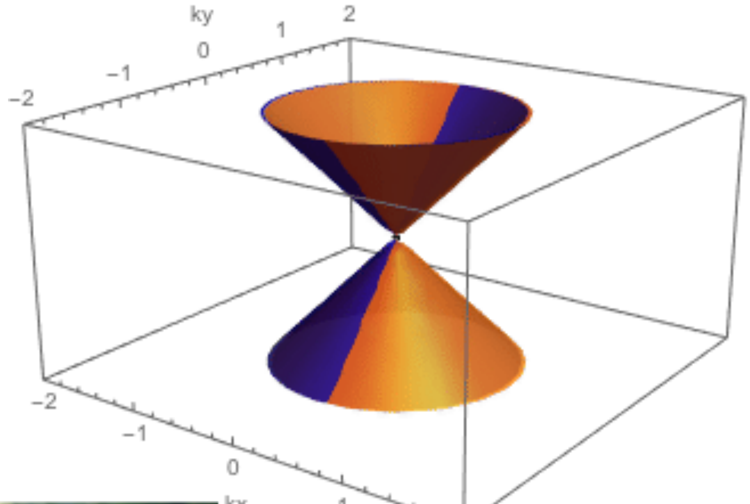


M. A. Sentef, M. Claassen, A. F. Kemper, B. Moritz, T. Oka, J. K. Freericks, and T. P. Devereaux, Nature Commun. 6, 7047 (2015)

Floquet-Weyl semimetal



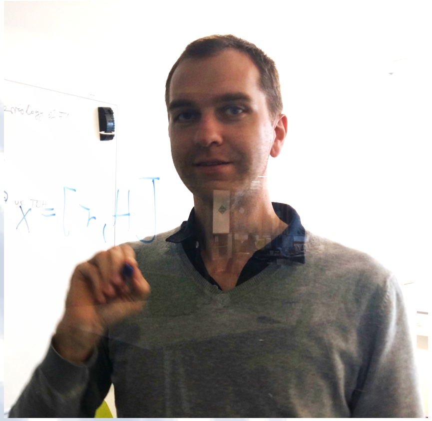
Ab initio: TDDFT + Floquet theory



Creating stable Floquet–Weyl semimetals by laser-driving of 3D Dirac materials

Hannes Hübener ✉, Michael A. Sentef, Umberto De Giovannini, Alexander F. Kemper & Angel Rubio ✉

Nature Commun. 8, 13940 (2017)



Hannes Hübener

Umberto de Giovannini

Alexander Kemper (NC State)

Angel Rubio

Summary I

- Floquet engineering: tuning effective parameters and changing materials properties by laser driving

Nature Commun. 6, 7047 (2015)

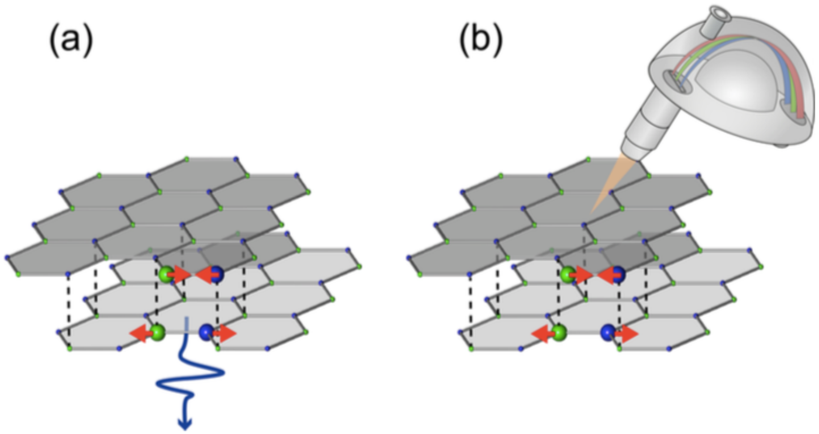
Nature Commun. 8, 13940 (2017)

II Dynamically enhanced coupling

Enhanced electron-phonon coupling in graphene with periodically distorted lattice

E. Pomarico, M. Mitrano, H. Bromberger, M. A. Sentef, A. Al-Temimy, C. Coletti, A. Stöhr, S. Link, U. Starke, C. Cacho, R. Chapman, E. Springate, A. Cavalleri, and I. Gierz
Phys. Rev. B **95**, 024304 – Published 13 January 2017

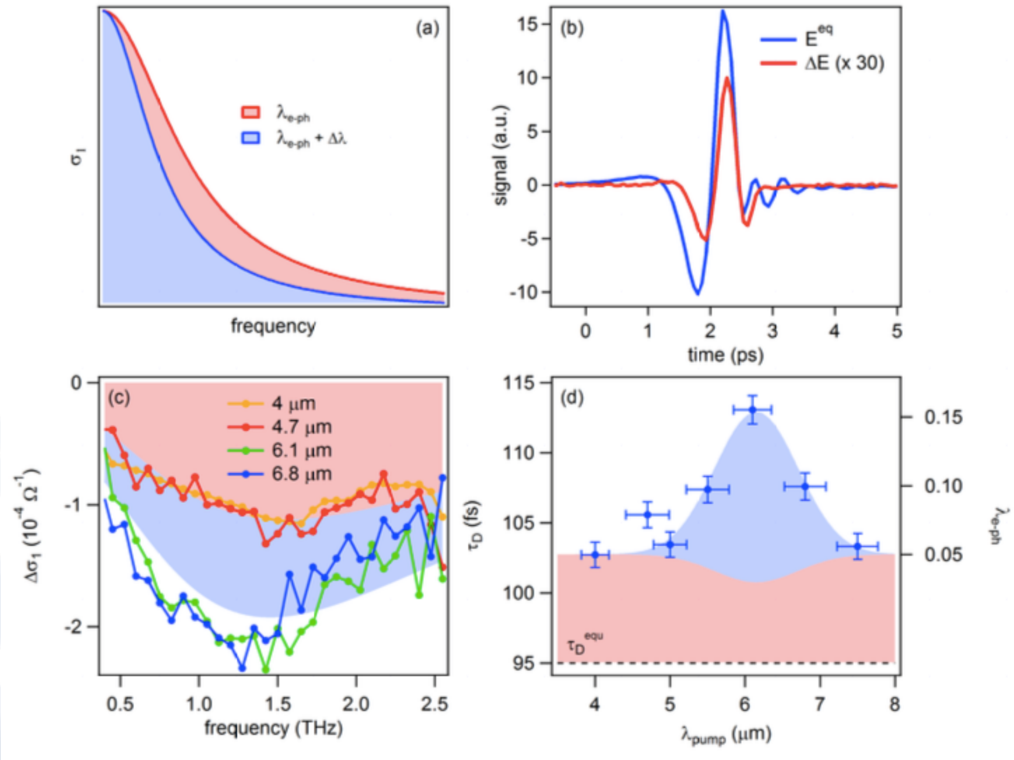
PRB 95, 024304 (2017)
enhanced electron-phonon for pump on resonance with IR phonon



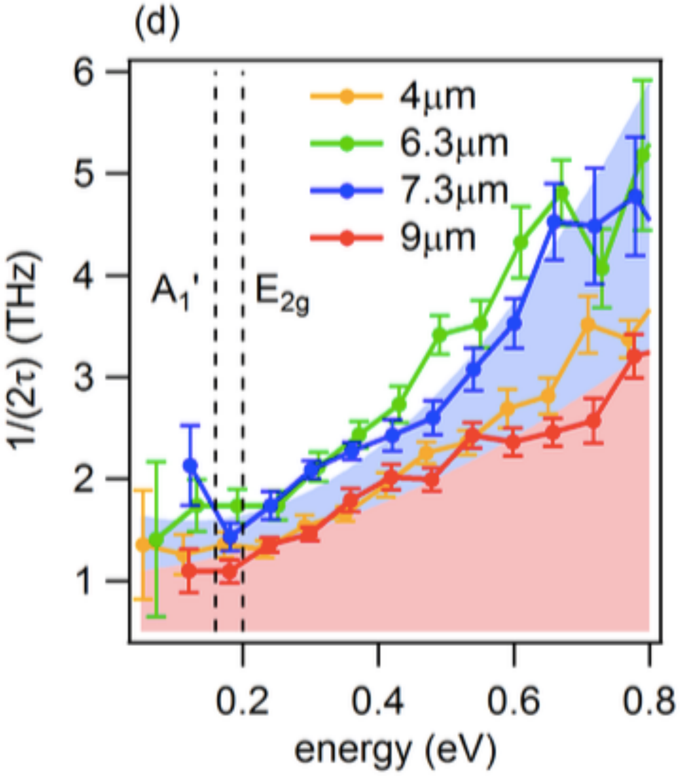
Dynamically enhanced coupling?

Enhanced electron-phonon coupling in graphene with periodically distorted lattice *PRB 95, 024304 (2017)*

transient reduction of Drude weight



enhanced tr-ARPES relaxation



3-fold enhancement of effective $\lambda_{el\text{-}ph}$! Why?

2-site model with nonlinear coupling

arXiv:1702.00952

$$\hat{H}(t) = -J \sum_{\sigma} (c^{\dagger}_{1,\sigma} c_{2,\sigma} + c^{\dagger}_{2,\sigma} c_{1,\sigma})$$
$$+ g_2 \sum_{\sigma, l=1,2} \hat{n}_{l,\sigma} (b_l + b^{\dagger}_l)^2$$
$$+ \Omega \sum_{l=1,2} b^{\dagger}_l b_l + F(t) \sum_{l=1,2} (b_l + b^{\dagger}_l),$$

also cf.
Kennes et al.,
Nature Physics (2017),
1609.03802

Idea: Drive nonlinearly coupled phonon and look at electronic response

Drive: $$F(t) = F \sin(\omega t),$$

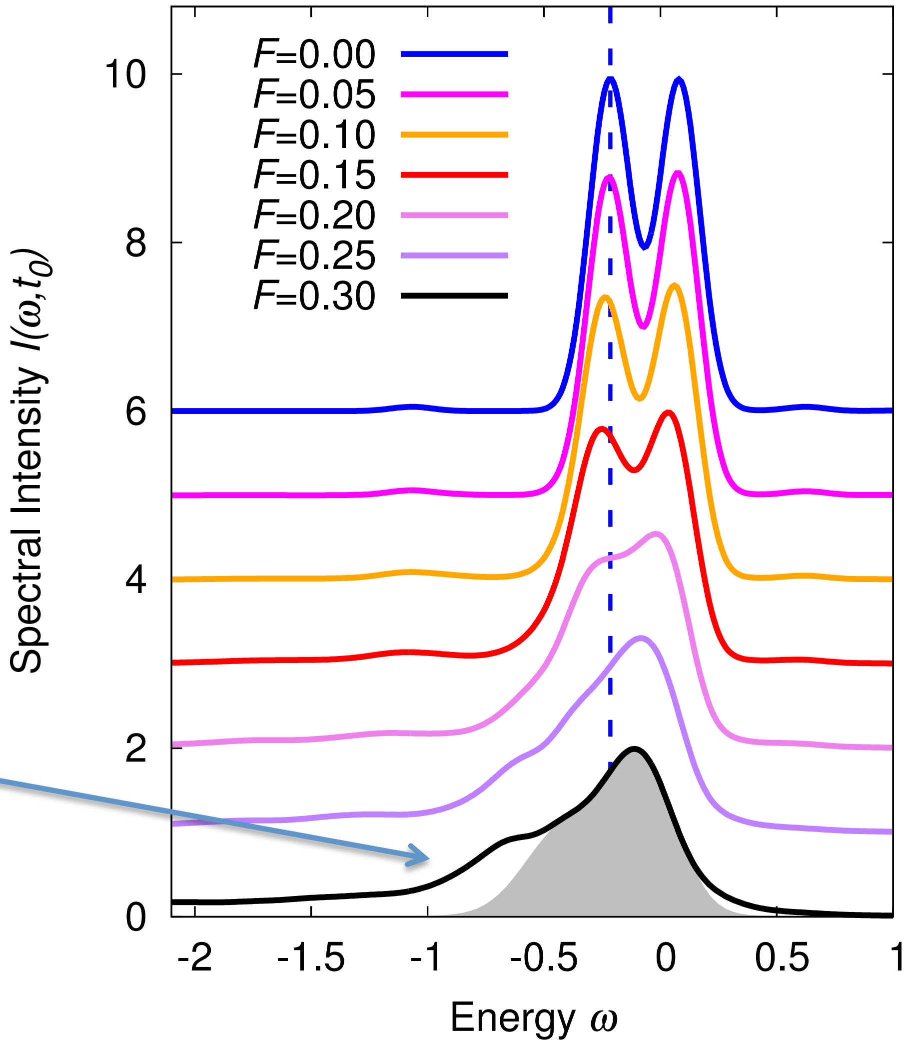
Response: $$I(\omega, t_0) = \mathrm{Re} \int dt_1 \, dt_2 \, e^{i\omega(t_1 - t_2)} s_{t_1, t_2, \tau}(t_0)$$
$$\times \Big[\langle \psi(t_2) | c^{\dagger}_{1,\uparrow} \mathcal{T} e^{-i \int_{t_1}^{t_2} H(t) dt} c_{1,\uparrow} | \psi(t_1) \rangle +$$
$$+ \langle \psi(t_1) | c_{1,\uparrow} \mathcal{T} e^{-i \int_{t_2}^{t_1} H(t) dt} c^{\dagger}_{1,\uparrow} | \psi(t_2) \rangle \Big],$$

Time-resolved el spectrum

arXiv:1702.00952

Reduced coherence peaks with stronger driving

Looks like enhanced el-ph coupling

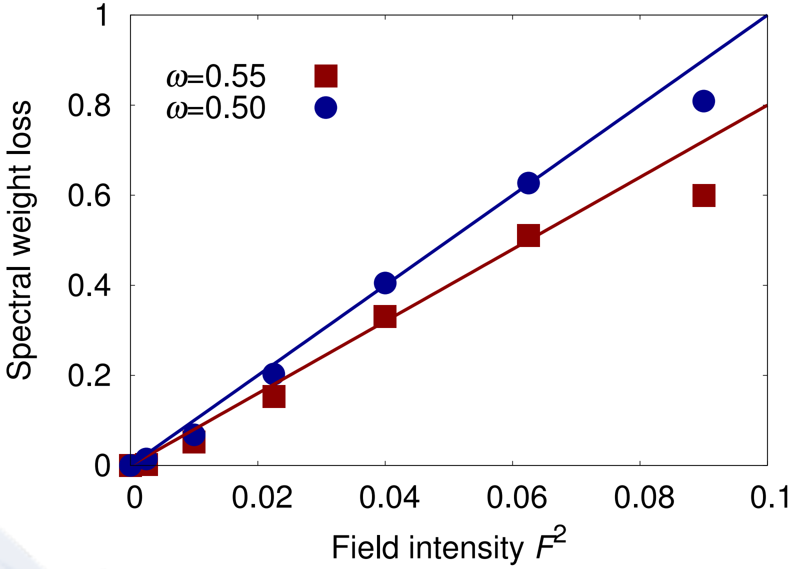


light-induced polaron formation

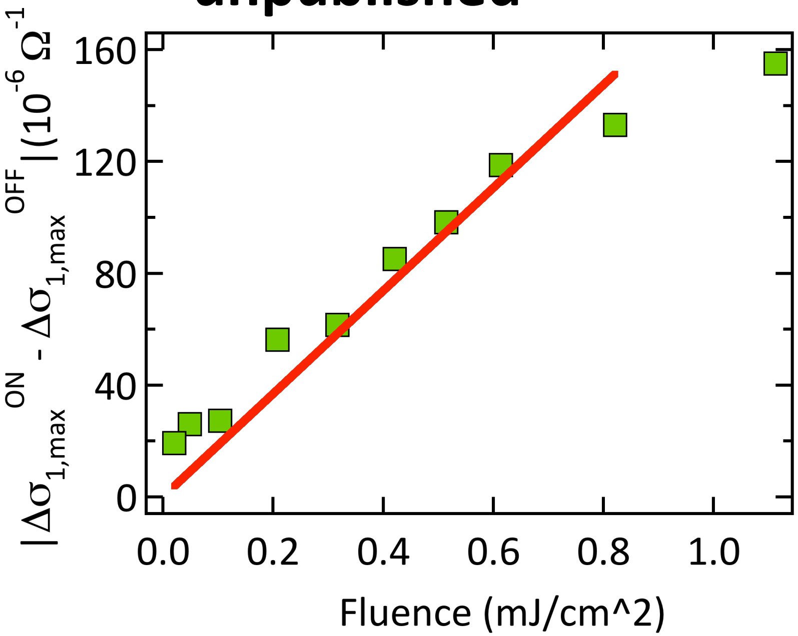
Field scaling

arXiv:1702.00952

Theory



Data by E. Pomarico, unpublished



Scaling of coherent spectral weight loss: proportional to field intensity consistent with experiments

Summary II

- enhanced electron-phonon coupling in phononically driven bilayer graphene

PRB 95, 024304 (2017)

E. Pomarico

I. Gierz

A. Cavalleri

Exact solution of (small) electron-phonon model system:

- theoretical proposal: nonlinear el-ph coupling as mechanism behind this enhancement

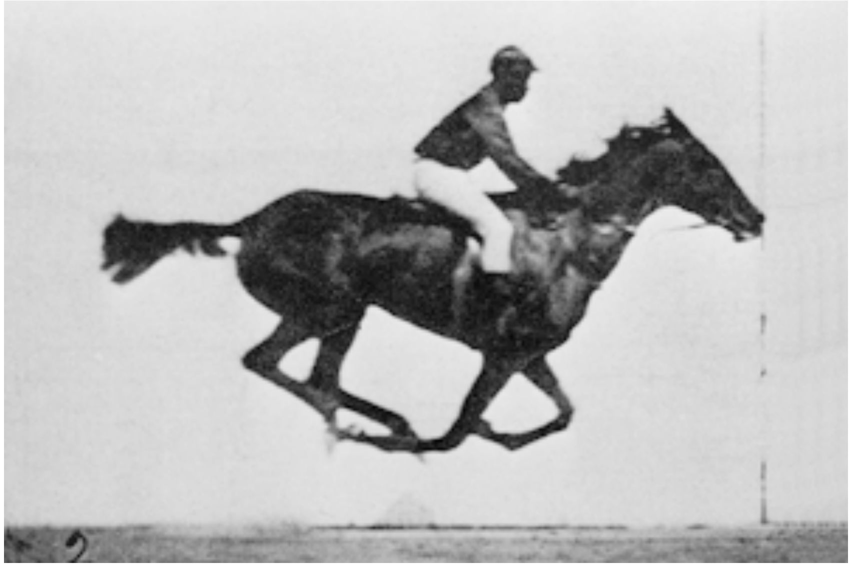
arXiv:1702.00952

Summary

From models to materials:

- theoretical simulations enable us to reveal mechanisms behind ultrafast dynamics in solids

THANK YOU!



Emmy Noether-Programm
Deutsche Forschungsgemeinschaft
DFG

III Theory of laser-controlled competing orders

Phys. Rev. Lett. 118, 087002 (2017)

Nonequilibrium superconductivity

Why?

- understand ordering mechanisms
- control ordered states: ultrafast switching
- induce new states of matter

How?

- laser near resonance with collective modes

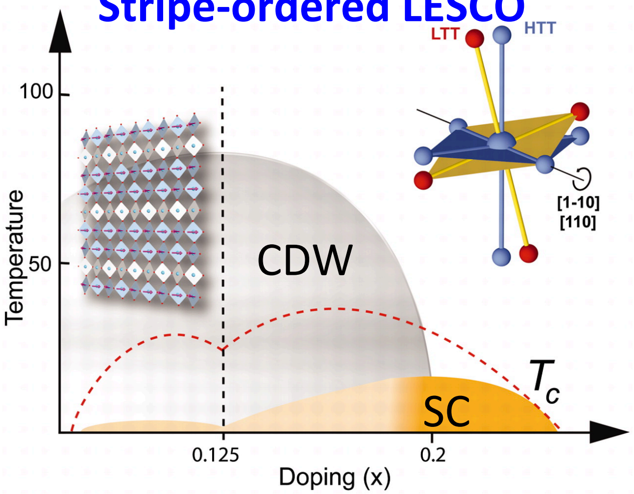
Generic mechanism to control competing orders with light?

Recent theories on laser-controlled couplings and competing orders:

Akbari et al., EPL 101, 17003 (2013); Moor et al., PRB 90, 024511 (2014); Fu et al., PRB 90, 024506 (2014); Dzero et al., PRB 91, 214505 (2015); Tsuji&Aoki, PRB 92, 064508 (2015); Cea et al., PRB 93, 180507 (2016); Kemper et al., PRB 92, 224517 (2015); Sentef et al., PRB 93, 144506 (2016); Krull et al., Nat. Commun. 7, 11921 (2016); Patel&Eberlein, PRB 93, 195139 (2016); Knap et al., PRB 94, 214504 (2016); Komnik&Thorwart EPJB 89, 244 (2016); Coulthard et al., 1608.03964; Kennes et al., Nat. Physics (2017), doi:10.1038/nphys4024; Sentef, 1702.00952; Babadi et al. 1702.02531; Murakami et al.,1702.02942; Mazza&Georges, 1702.04675; Dehghani&Mitra, 1703.01621

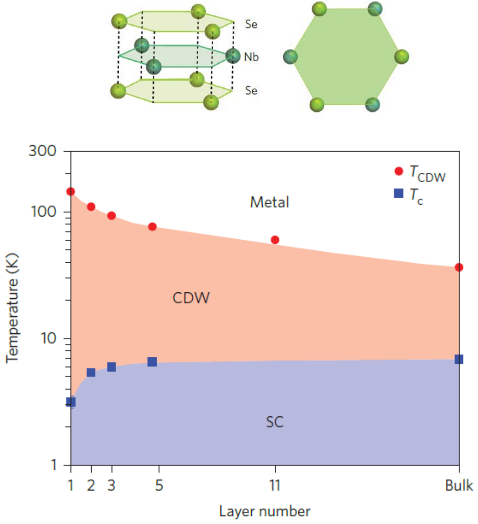
Experimental motivation: competing orders

Stripe-ordered LESCO



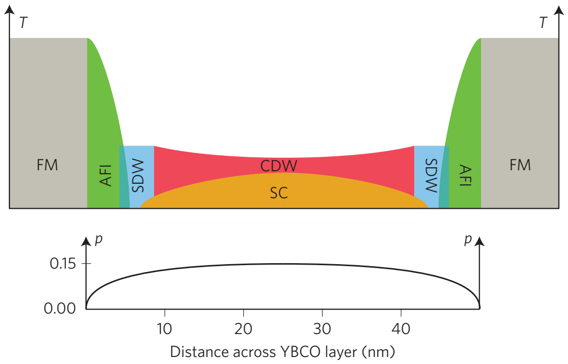
D. Fausti et al., Science, 331, 189 (2011)

NbSe2



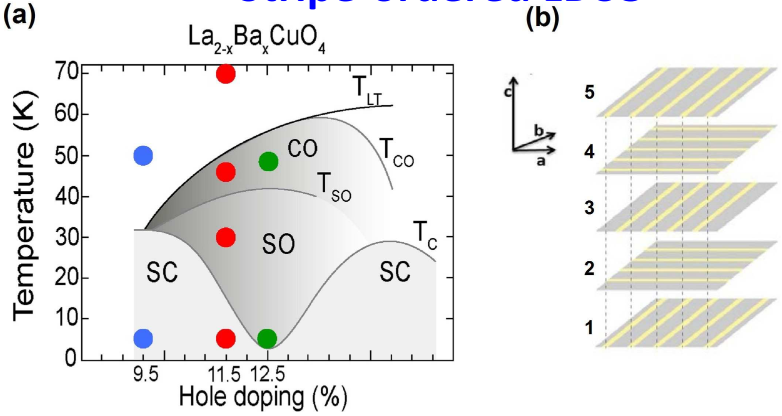
X. Xi et al., Nat. Nanotechnol. 10, 765 (2015)

YBCO-LCMO heterostructure



A. Frano et al., Nat. Mater. 15, 831 (2016)

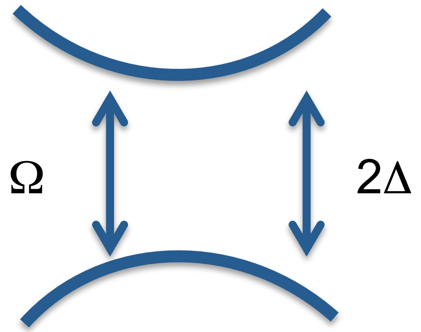
Stripe-ordered LBCO



D. Nicoletti et al., PRB 90, 100503 (2014)

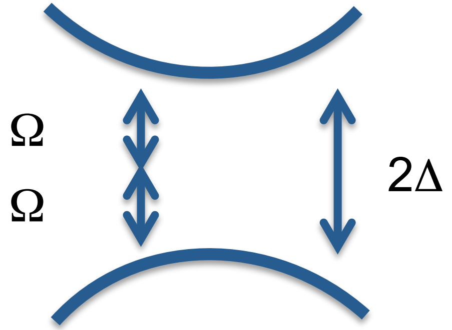
Driven SC/CDW

CDW ~ A
1-photon resonance

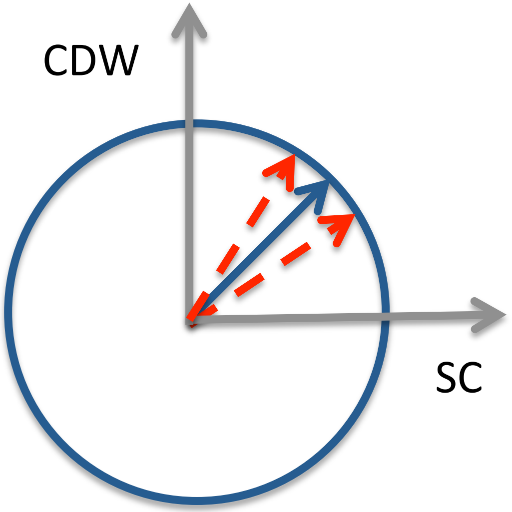


SC ~ A^2
2-photon resonance

Tsuji&Aoki, PRB 92, 064508 (2015)
Cea et al., PRB 93, 180507 (2016)

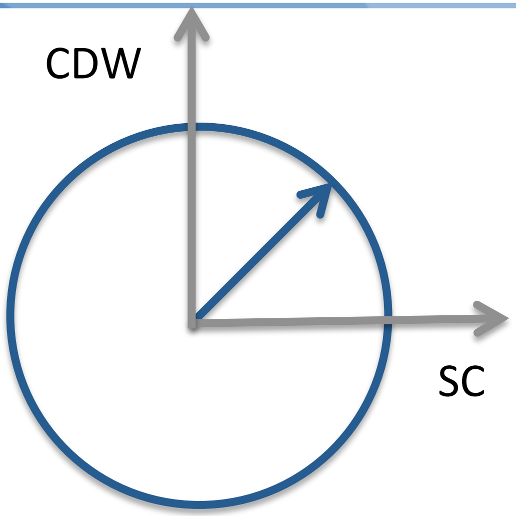


... laser lifts SC/CDW degeneracy
... Goldstone-like collective mode?



Competing orders

- attractive -U Hubbard model
- degeneracy of SC and CDW at perfect nesting
- SO(4) symmetry (SC, CDW, eta pairing)



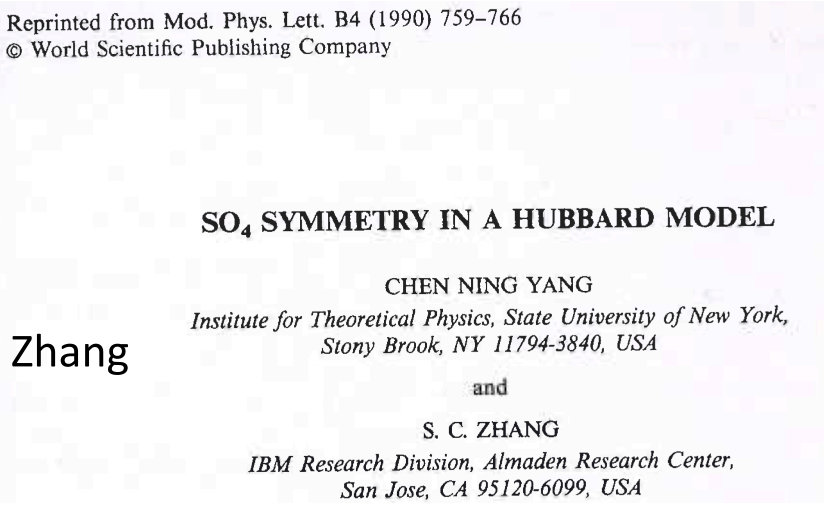

VOLUME 63, NUMBER 19 PHYSICAL REVIEW LETTERS 6 NOVEMBER 1989

η Pairing and Off-Diagonal Long-Range Order in a Hubbard Model

Chen Ning Yang

C. N. Yang

S.-C. Zhang


Reprinted from Mod. Phys. Lett. B4 (1990) 759–766
© World Scientific Publishing Company

SO_4 SYMMETRY IN A HUBBARD MODEL

CHEN NING YANG
Institute for Theoretical Physics, State University of New York, Stony Brook, NY 11794-3840, USA

and

S. C. ZHANG
IBM Research Division, Almaden Research Center, San Jose, CA 95120-6099, USA

Minimal Model

$$H = \sum_{k\sigma} \epsilon(k) n_{k\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} = H_J + H_U,$$
$$\epsilon(k) = -2J(\cos(k_x) + \cos(k_y)),$$

2D square lattice + attractive U + mean-field decoupling

$$\Delta_{SC} = U \sum_k f_k, \qquad f_k \equiv \langle c_{-k\downarrow} c_{k\uparrow} \rangle \qquad \text{(SC)},$$
$$\Delta_{CDW} = U \sum_k g_k, \qquad g_k \equiv \frac{1}{2} \sum_\sigma \langle c^\dagger_{k\sigma} c_{k+Q\sigma} \rangle \quad \text{(CDW)},$$
$$\Delta_\eta = U \sum_k \eta_k. \qquad \eta_k \equiv \langle c_{-(k+Q)\downarrow} c_{k\uparrow} \rangle \quad (\eta \text{ pairing}).$$

Mean-field equations

Equations of motion for electronic driving:

eta pairing provides coupling

$$i\partial_t n_k = -\Delta_{SC}(f_k - f_k^*) + \Delta_{CDW}(g_k - g_k^*) - \Delta_\eta^* \eta_k + \Delta_\eta \eta_k^*,$$

$$i\partial_t f_k = \Delta_{SC}(1 - (n_k + n_{-k})) + (\epsilon(k-A) + \epsilon(k+A)) f_k + \Delta_{CDW}(\eta_k + \eta_{k+Q}) - \Delta_\eta (g_k^* + g_{-k}^*),$$

$$i\partial_t g_k = \Delta_{CDW}(n_k - n_{k+Q}) - 2\epsilon(k-A) g_k + \Delta_{SC}(\eta_k^* - \eta_{k+Q}) + \Delta_\eta f_k^* - \Delta_\eta^* f_{k+Q},$$

$$i\partial_t \eta_k = \eta_k(\epsilon(k-A) - \epsilon(k+A)) + \Delta_{CDW}(f_k + f_{k+Q}) - \Delta_{SC}(g_{-k} + g_k^*) - \Delta_\eta (n_k + n_{-(k+Q)} - 1).$$

nonlinear equations:
self-consistency in real time

$$\Delta_{SC} = U \sum_k f_k,$$

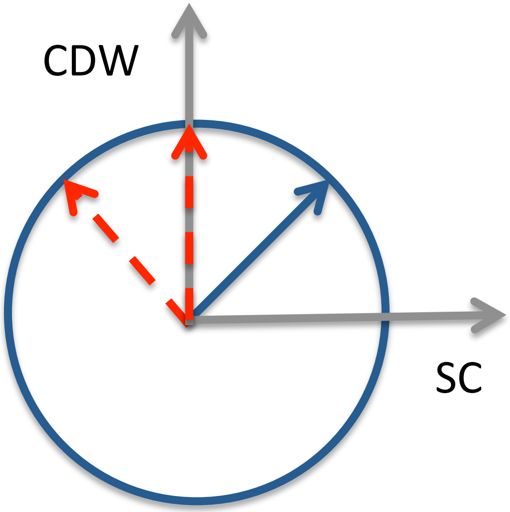
$$\Delta_{CDW} = U \sum_k g_k,$$

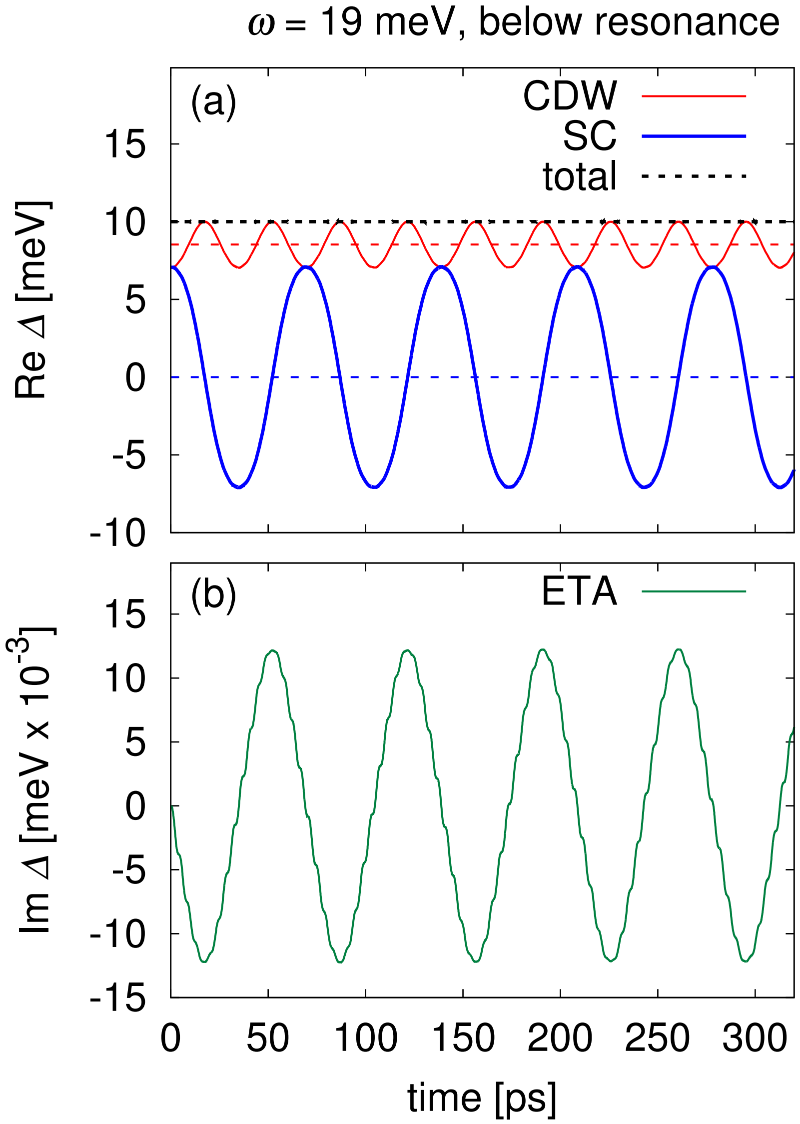
$$\Delta_\eta = U \sum_k \eta_k.$$

Nonequilibrium:
Periodic driving field: $A(t) = A_{max} \sin(\omega t)\,(e_x + e_y)$
$A_{max} = 5 \times 10^{-5}$, $E_{max} \sim 10\text{-}100$ V/cm – weak fields!

Gap resonance – coexisting initial state

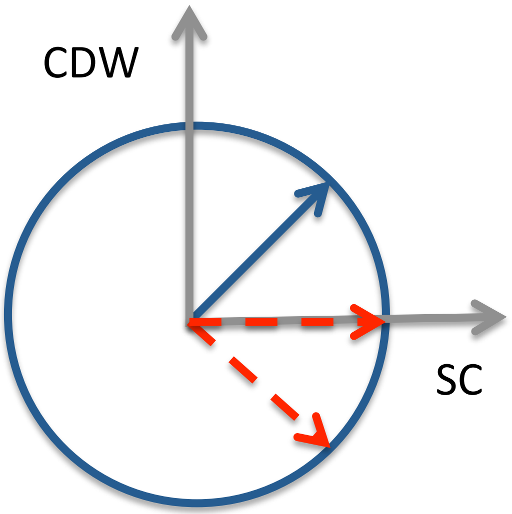
Below resonance:
SC down, CDW up

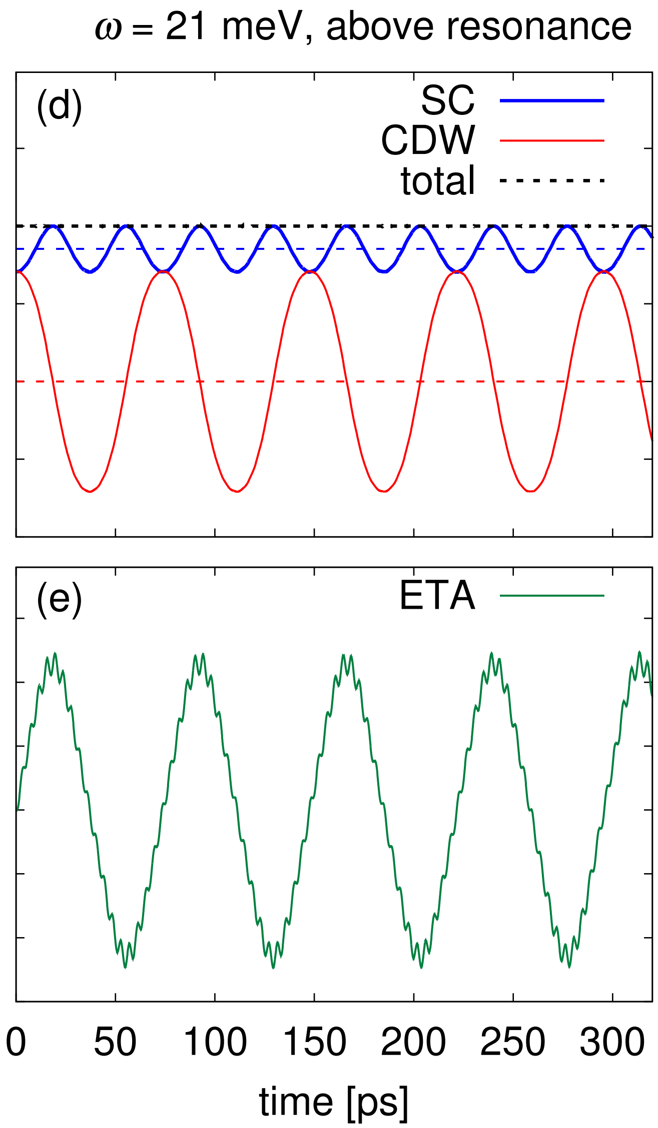




Gap resonance – coexisting initial state

Above resonance:
SC up, CDW down





Gap resonance – coexisting initial state

nature

THE INTERNATIONAL WEEKLY JOURNAL OF SCIENCE

Time crystals

First observations of exotic new state of matter PAGES 164, 185, 217 & 221

BEHAVIOUR
COLLECTIVE AMNESIA
How social media and fake news are rewriting history
PAGE 168

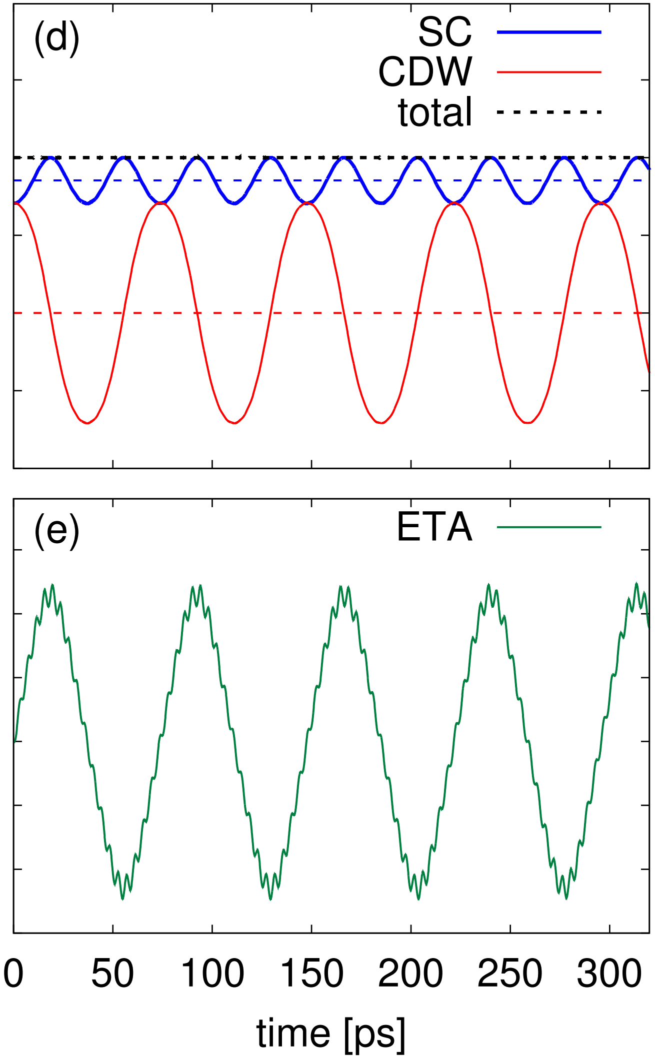
APPLIED PHYSICS
COHERENT STRATEGY
Ways to commercialize quantum computers
PAGE 171

ARCHAEOLOGY
TRACING THE SILK ROAD
Iconic trade route arose from nomadic herding network
PAGES 188 & 193

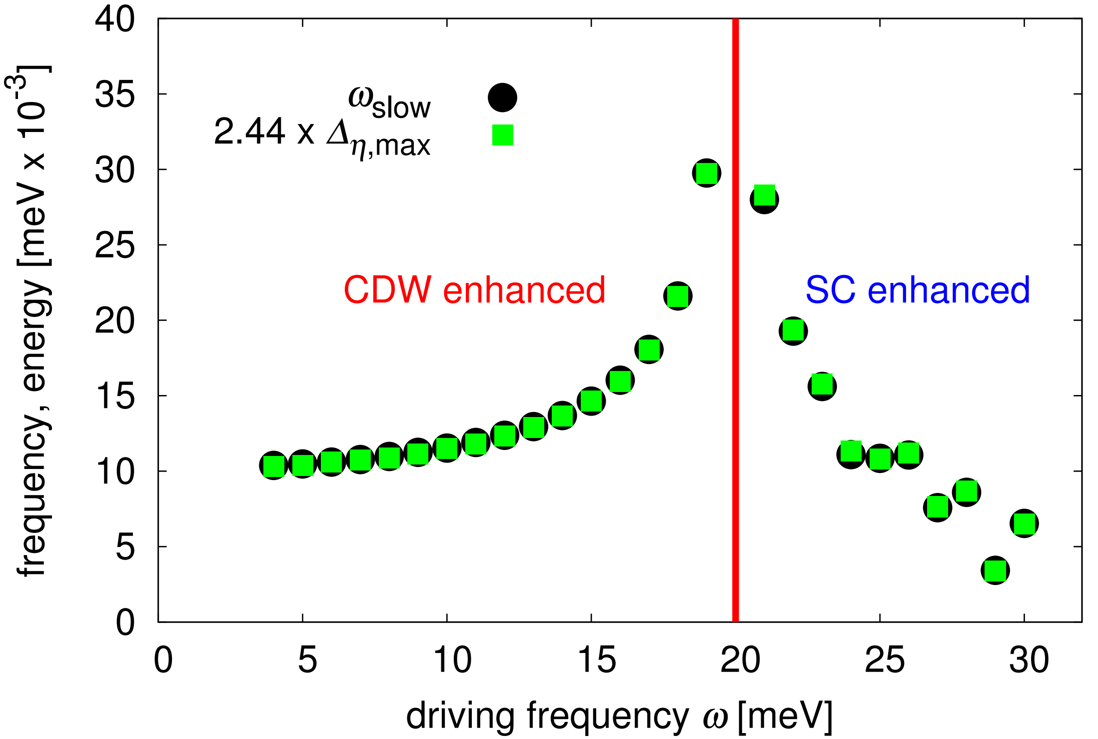
NATURE.COM/NATURE
9 March 2017 £10
Vol. 543, No. 7644

„Floquet time crystal“??

ω = 21 meV, above resonance



Gap resonance



oscillation frequency set by light-induced eta pairing amplitude, which gives „mass" to collective mode

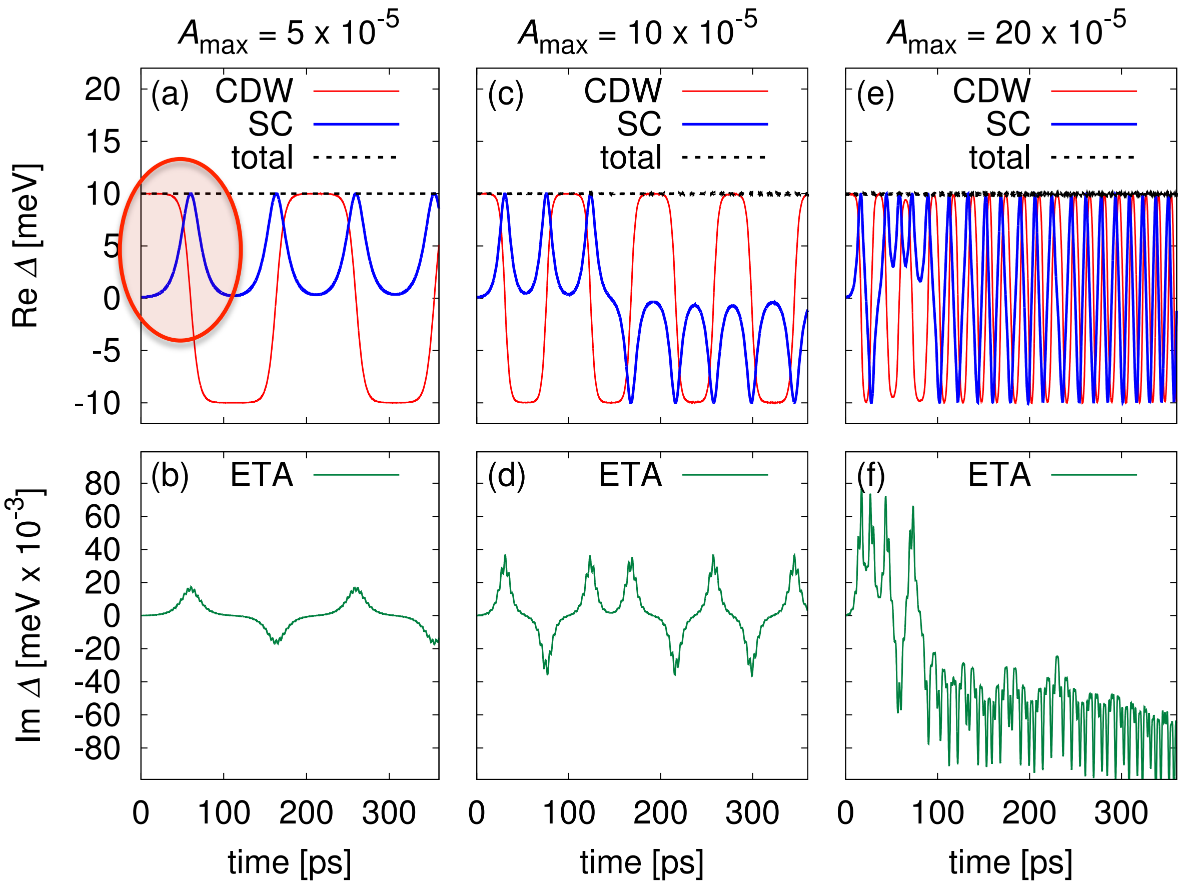
resonant behavior at $\Omega=2\Delta$ = single-particle gap

Inducing superconductivity

99% CDW initial state

Drive slightly above gap

Irregular behavior for stronger driving

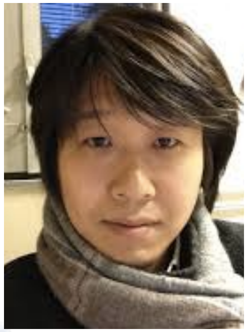


Summary III

Tight-binding model + time-dependent mean-field theory:

- laser-controlled switching between SC/CDW
- path to understanding of light-induced superconductivity in systems with competing orders?

Phys. Rev. Lett. 118, 087002 (2017)



Akiyuki Tokuno

Antoine Georges

Palaiseau/Paris/Geneva

Corinna Kollath

University of Bonn