Dynamical phase transitions in the photodriven charge-ordered Dirac-electron system

Yasuhiro Tanaka

Math. and Science Education Research Center, Kanazawa Institute of Technology

Masahito Mochizuki

Department of Applied Physics, Waseda University

YT and M. Mochizuki, Phys. Rev. B 104, 085123 (2021)
YT and M. Mochizuki, Phys. Rev. Lett. 129, 047402 (2022)

Outline

1. Introduction

    Photoinduced phase transitions

    Physical properties of α-(BEDT-TTF)$_2$I$_3$

2. Model and Method

    Interacting Dirac electron system

    Time dependent Schödinger eq. + Floquet theory

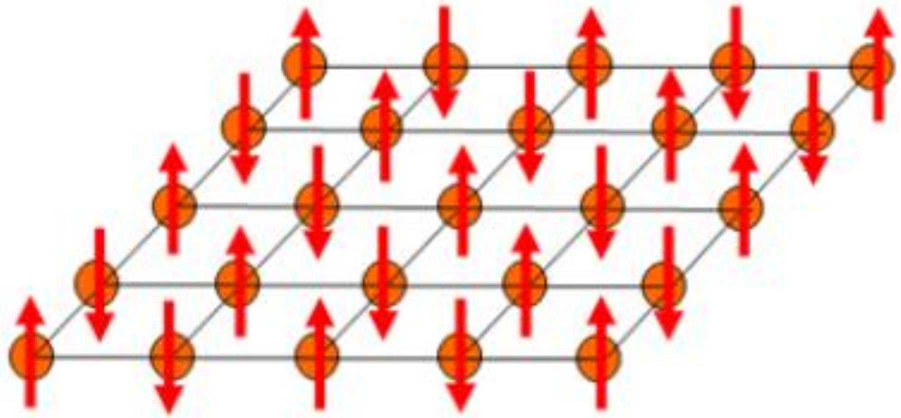
3. Results

    Successive dynamical phase transitions

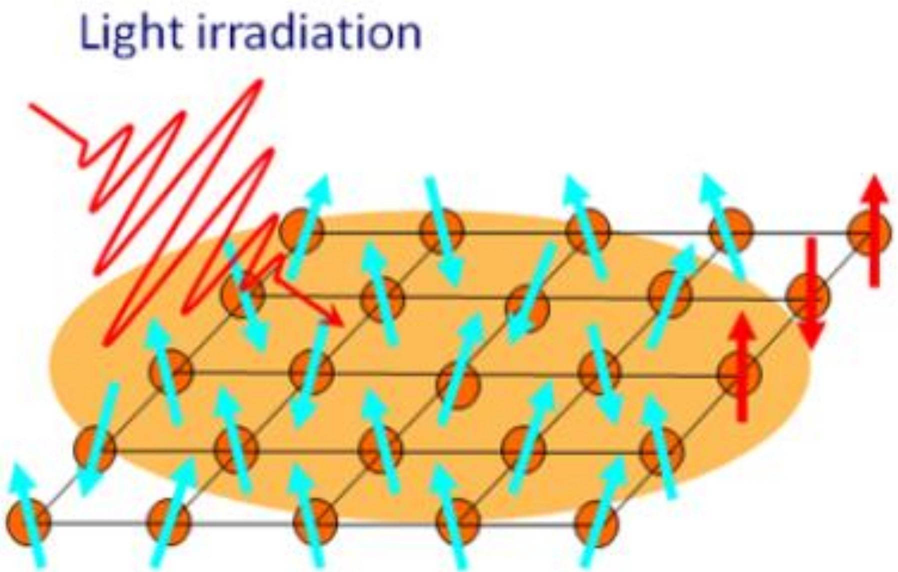
4. Summary

Photoinduced Insulator to Metal transition

Correlated electron systems



Mott insulator
(Charge Ordered Insulator)

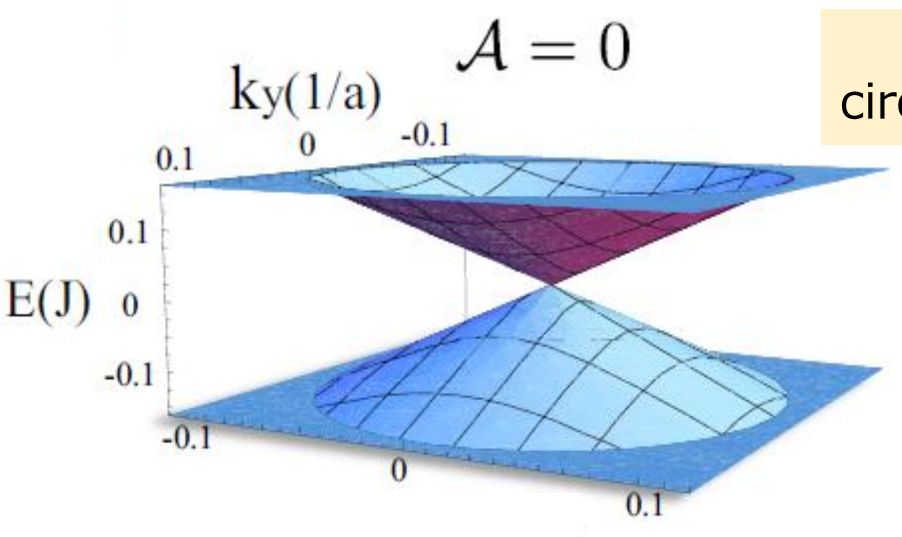


Photoinduced Metallic State

S. Iwai, Crystals 2, 590 (2012)
K. Yonemitsu and K. Nasu, Phys. Rep. 465, 1 (2008)
H. Aoki *et al.*, Rev. Mod. Phys. 86, 779 (2014)

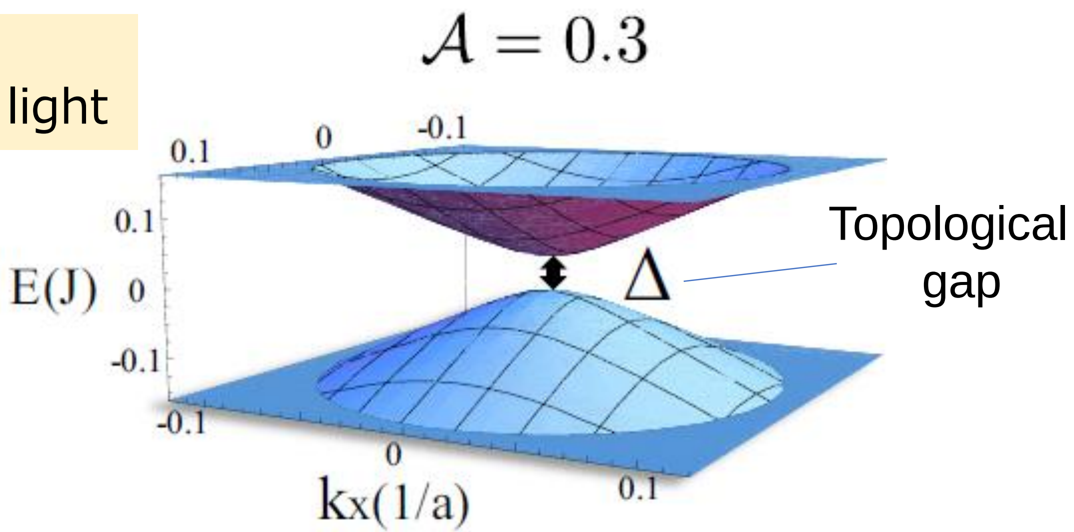
Photoinduced topological phase transition

Dirac fermion system



Irradiation with
circularly polarized light

Floquet Chern insulator



Floquet theory

T. Oka and H. Aoki, Phys. Rev. B 79, 081406(R) (2009)

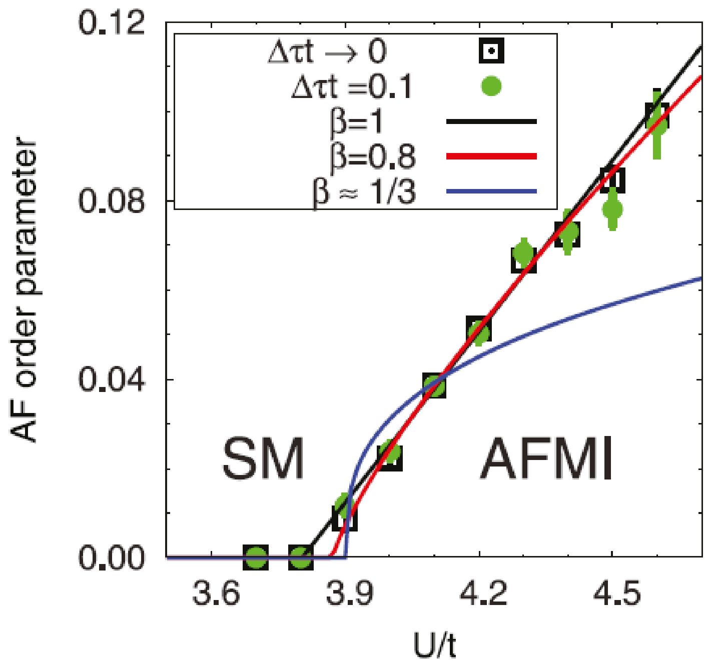
T. Kitagawa *et al.,* Phys. Rev. B 84, 235108 (2011)

Light-induced anomalous Hall effect in graphene

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

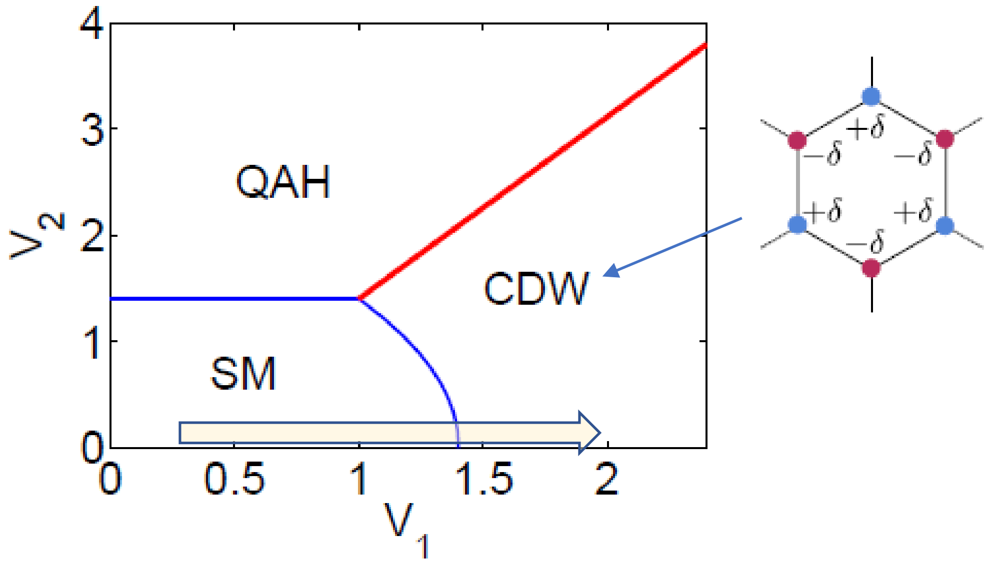
S. A. Sato *et al*., Phys. Rev. B 99, 214302 (2019)

Interacting Dirac fermion systems (honeycomb lattice)



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

S. Sorella, Y. Otsuka, and S. Yunoki, Sci. Rep. 2, 992 (2012)



$$H = -\sum_{\langle ij \rangle} t \left(c^\dagger_i c_j + h.c. \right) + \underline{V_1} \sum_{\langle i,j \rangle} (n_i - 1)(n_j - 1)$$

$$+ V_2 \sum_{\langle\langle i,j \rangle\rangle} (n_i - 1)(n_j - 1) - \mu \left(\sum_i n_i - N \right)$$

S. Raghu *et al.,* Phys. Rev. Lett. 100, 156401 (2008)

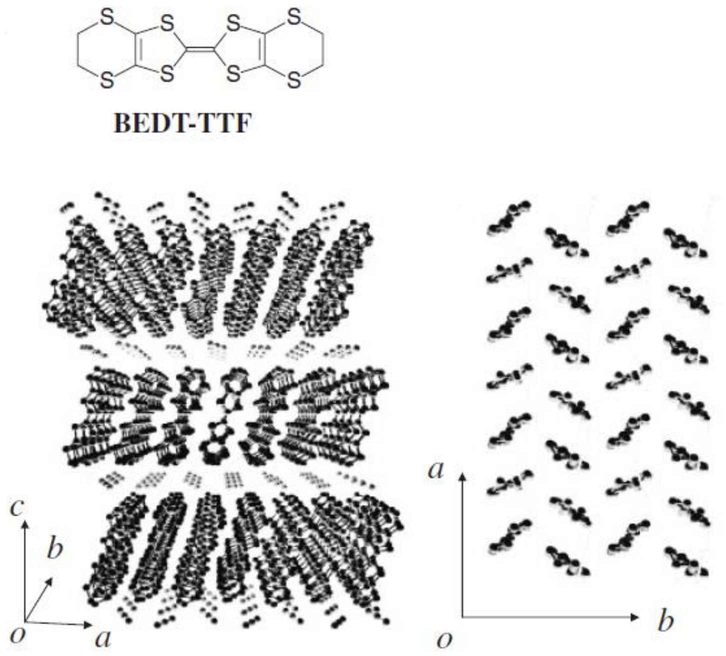
Photoirradaition to interacting Dirac fermion system ?

α-(BEDT-TTF)$_2$I$_3$: Correlated Dirac fermion system with charge order

Physical properties of α-(BEDT-TTF)$_2$I$_3$

Crystal structure

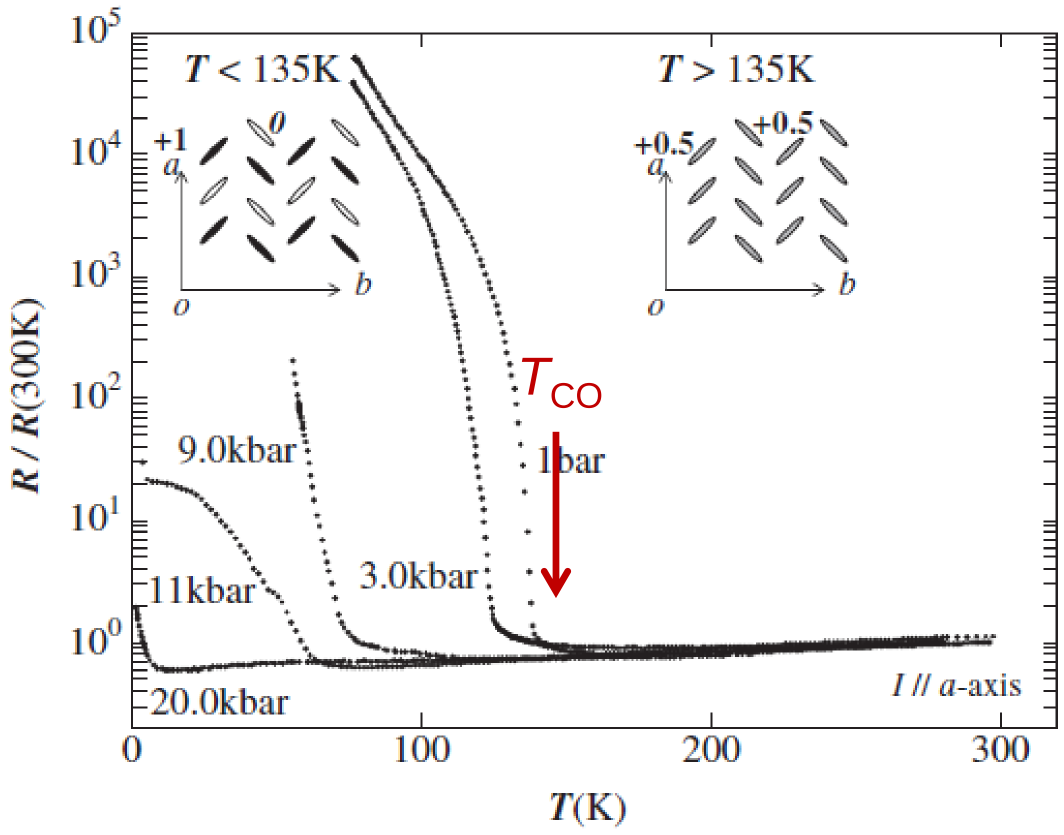
- quasi-2d structure
- 3/4-filling
  (1 hole/2 molecules)



N. Tajima, *et al*.
J. Phys Soc. Jpn. 75, 051010 (2006)

Charge order

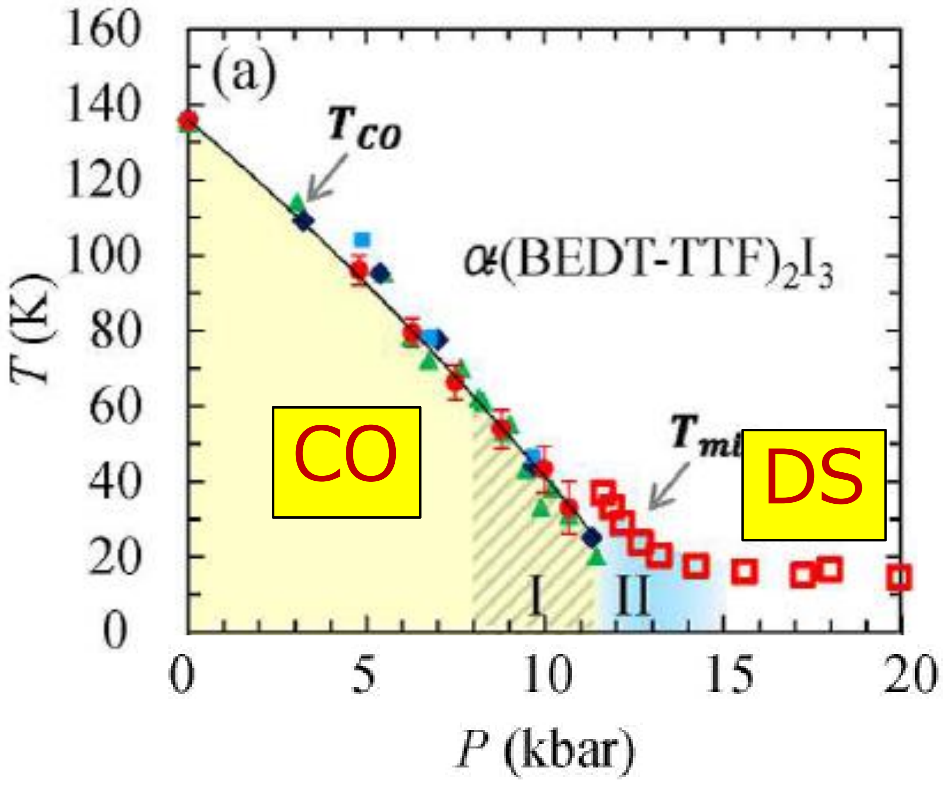
- 1st order transition T_{CO}=135 K
- Spin gap (nonmagnetic) below T_{CO}



Dirac semimetal phase under pressure

CO → Dirac semimetal (DS)

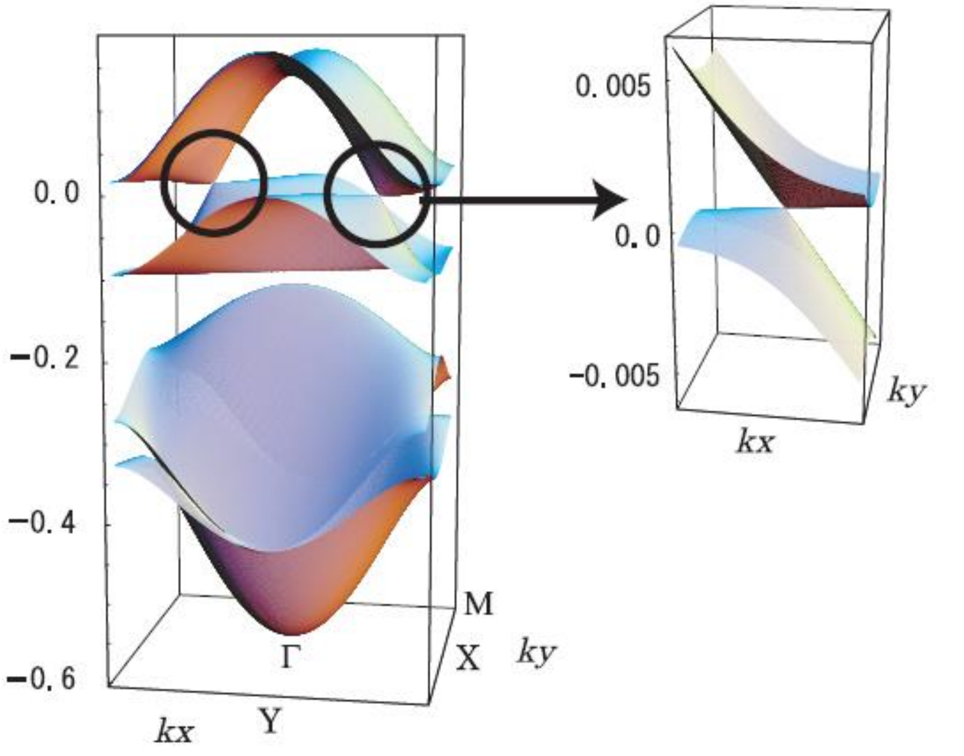
- *P*-*T* phase diagram



D. Liu *et al.*, Phys. Rev. Lett. 116, 226401 (2016)

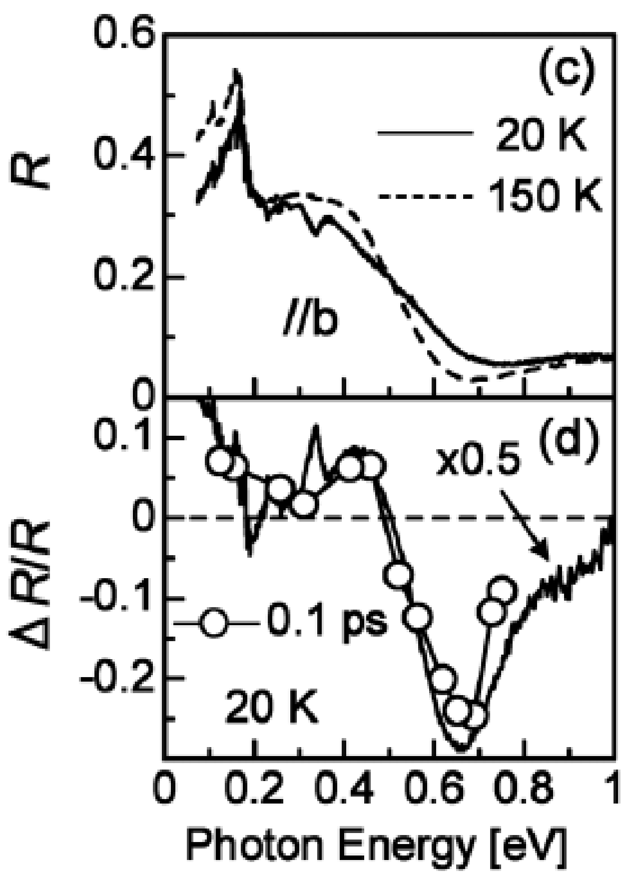
N. Tajima, *et al.*, J. Phys Soc. Jpn. 75, 051010 (2006)

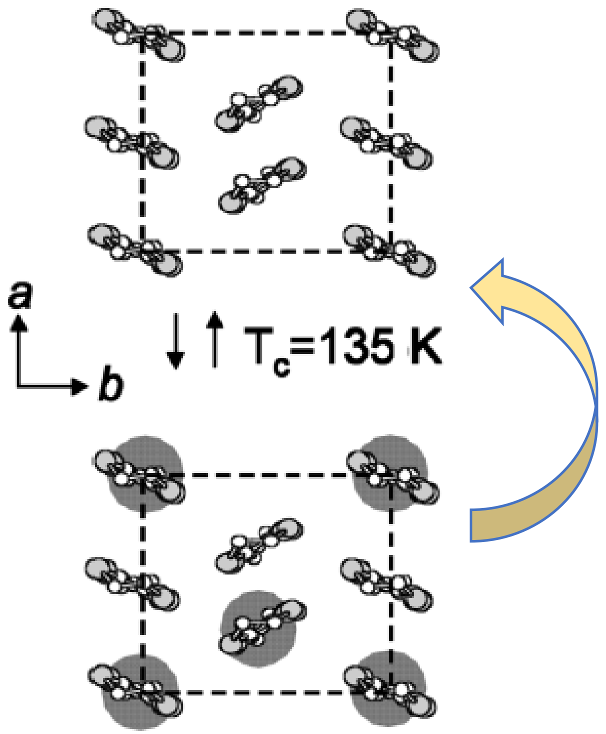
DS



S. Katayama, *et al.*, J. Phys. Soc. Jpn. 75, 054705 (2006)

Photoinduced melting of charge order





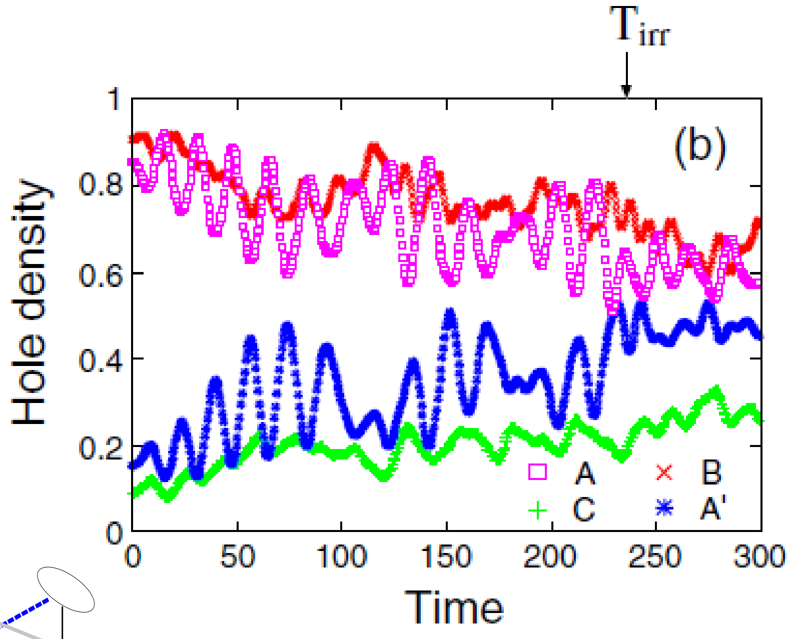
Femtosecond Pump-probe spectroscopy

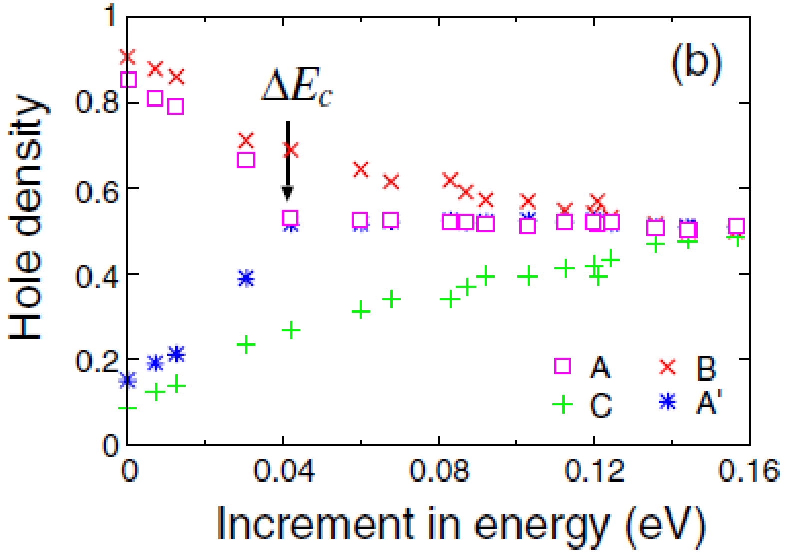
S. Iwai, *et al.*, Phys. Rev. Lett. 98, 097402 (2007)

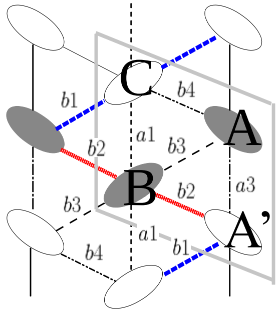
Theories for photodriven α-(BEDT-TTF)$_2$I$_3$: CO melting

- Interacting model (Extended Hubbard model)
- Linearly polarized light
- Time-dependent Schrödinger eq.

⇒ melting of CO by photoexcitation







Hartree-Fock appr.

YT and K. Yonemitsu, J. Phys. Soc. Jpn. 79, 024712 (2010)

Exact diagonalization

S. Miyashita, YT, S. Iwai, and K. Yonemitsu, J. Phys. Soc. Jpn. 79, 034708 (2010)

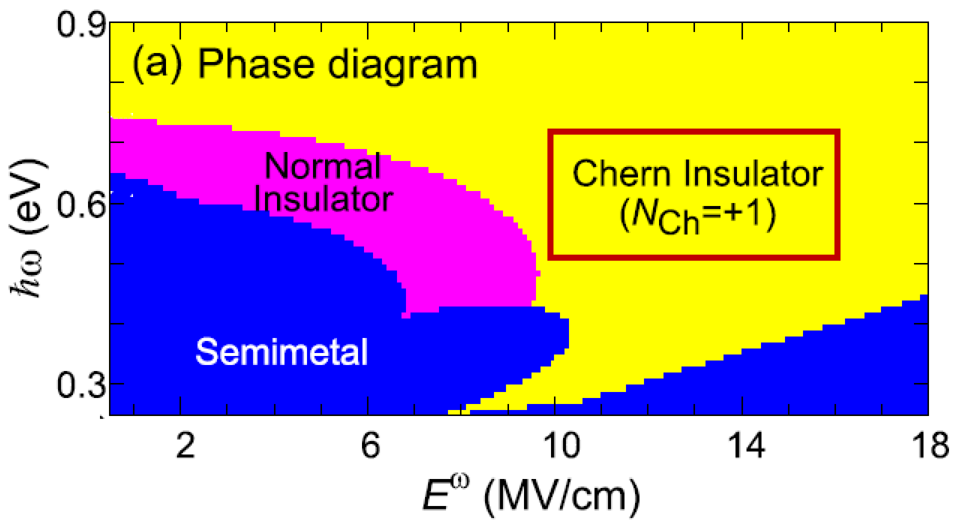
Theories for photodriven α-(BEDT-TTF)$_2$I$_3$: DS phase

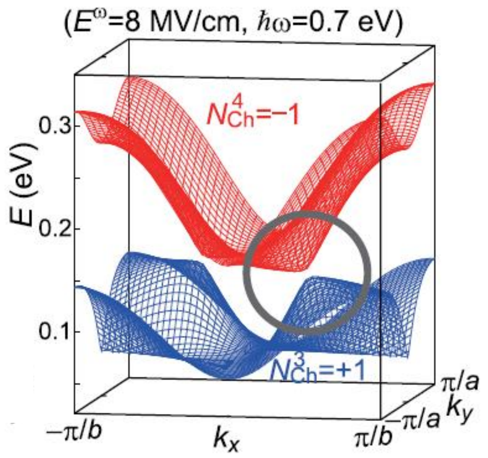
- Noninteracting model + Floquet theory

  - Circularly polarized light

    K. Kitayama and M. Mochizuki, Phys. Rev. Res. 2, 023229 (2020)
    K. Kitayama, YT, M. Ogata, and M. Mochizuki, J. Phys. Soc. Jpn. 90, 104705 (2021)

    Photoinduced topological phase transition





  - Linearly polarized light

    K. Kitayama, M. Mochizuki, YT, and M. Ogata, Phys. Rev. B 104, 075127 (2021)

    Photoinduced pair annihilation of magnetic charges

  - Elliptically polarized light

    K. Kitayama, M. Ogata, M. Mochizuki, and YT, J. Phys. Soc. Jpn. 91, 104704 (2022)

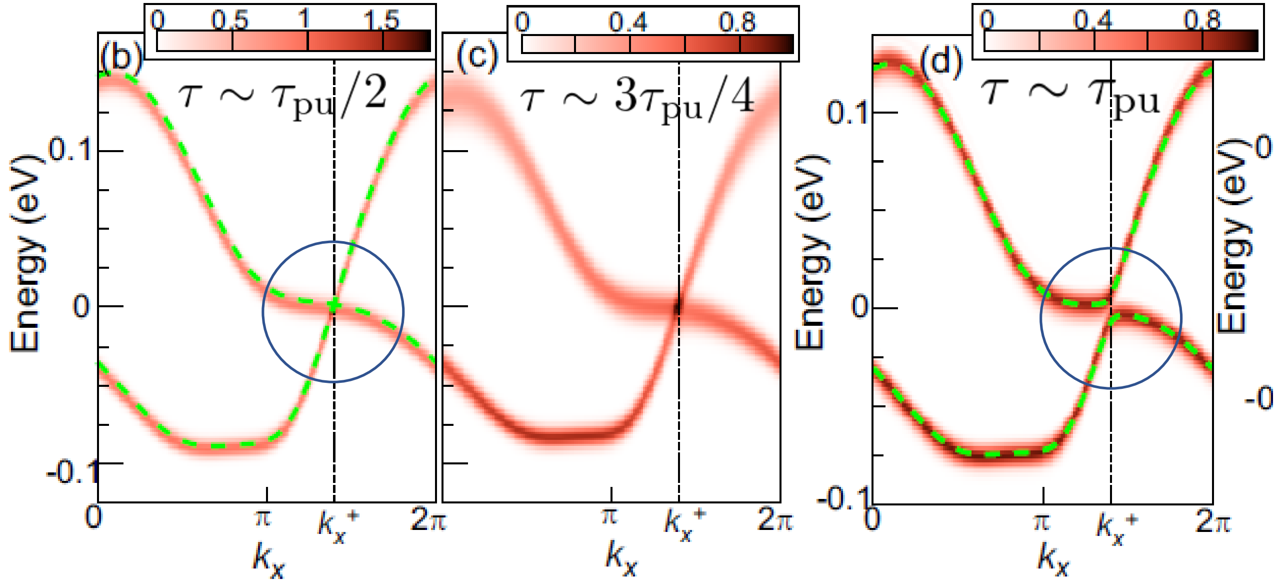
    Collision and Collapse of the Dirac cones

Theories for photodriven α-(BEDT-TTF)$_2$I$_3$: DS phase

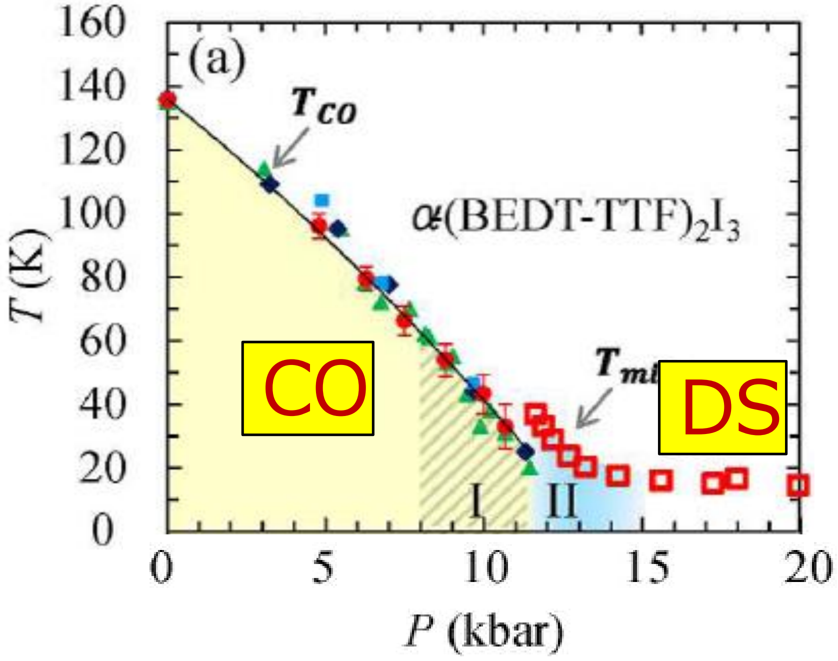
- Noninteracting model + time dependent Schrödinger eq.

➢ DS ➡ Floquet Chern ins. (Circularly polarized light)

Transient spectra, Hall conductivity



YT and M. Mochizuki, Phys. Rev. B 104, 085123 (2021)



At ambient pressure, CO appears

CO ➡ DS ?

CO ➡ Floquet Chern insulator ?

Interacting model + CPL

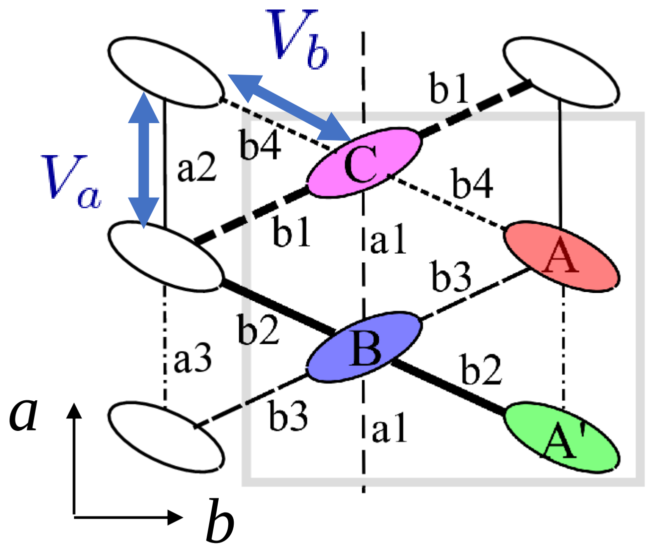
Model and Method

Hamiltonian

H. Kino and H. Fukuyama, J. Phys. Soc. Jpn. 64, 1877 (1995)
H. Seo, J. Phys. Soc. Jpn. 69, 805 (2000)

$$H = \sum_{\langle ij \rangle \sigma} t_{i,j}(c^{\dagger}_{i\sigma} c_{j\sigma} + h.c.) + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \sum_{\langle ij \rangle} V_{i,j} n_i n_j$$

Interaction terms ➡ Hartree-Fock approximation



Transfer integrals

b1=0.127(eV), b2=0.145, b3=0.062, b4=0.025
a1=-0.035, a2=-0.046, a3 =0.018

A. Kobayashi *et al*., J. Phys. Soc. Jpn. 73, 3135 (2004)

Time evolution (Time-dependent Schrodinger eq.)

Photoexcitation : Peierls phase

$$t_{i,j} \rightarrow t_{i,j} e^{i\boldsymbol{\delta}_{i,j} \cdot \boldsymbol{A}(\tau)}$$

$$\boldsymbol{\delta}_{i,j} = \boldsymbol{r}_j - \boldsymbol{r}_i$$

$\boldsymbol{A}(\tau)$: Vector potential for circularly polarized light

$$|\psi_{\boldsymbol{k},\nu}(\tau + d\tau)\rangle = T\exp\left[-i \int_{\tau}^{\tau + d\tau} d\tau' H^{\mathrm{HF}}_{\boldsymbol{k}}(\tau')\right] |\psi_{\boldsymbol{k},\nu}(\tau)\rangle$$

$$\simeq \exp\left[-id\tau H^{\mathrm{HF}}_{\boldsymbol{k}}(\tau + d\tau/2)\right] |\psi_{\boldsymbol{k},\nu}(\tau)\rangle$$

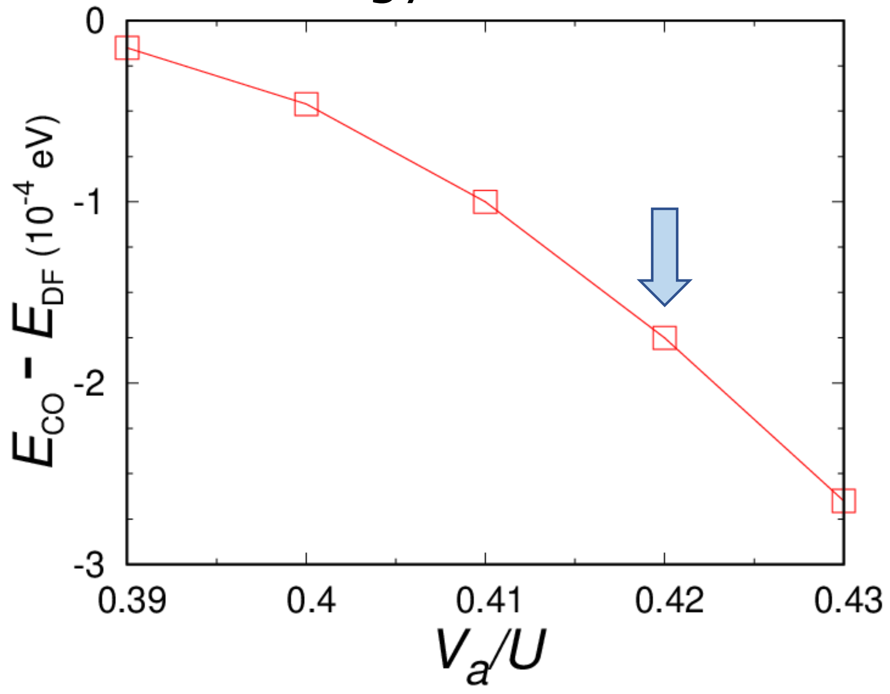
A. Terai and Y. Ono, Prog. Theor. Phys. Suppl. 113, 177 (1993)
M. Kuwabara and Y. Ono, J. Phys. Soc. Jpn. 64, 2106 (1995)

Parameters

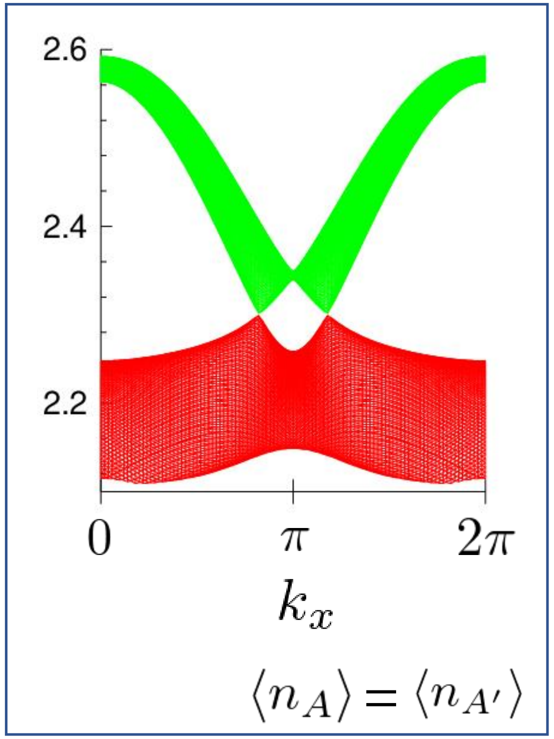
- HF solutions
  - horizontal CO with no spin order
  - DS (no CO)

$$U = 0.6,\ V_a = 0.42U,\ V_b = 0.6V_a$$

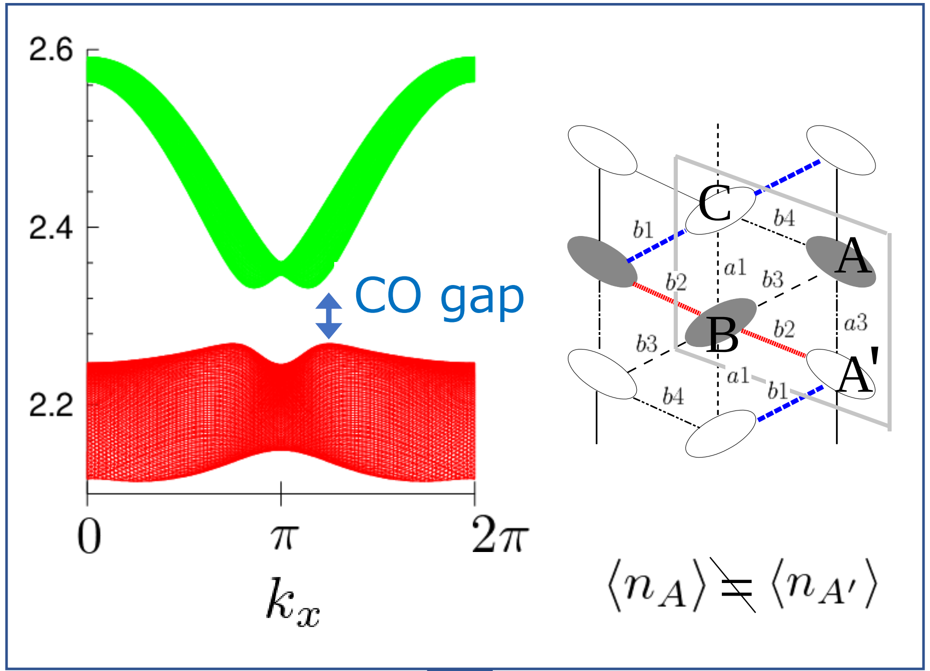
Energy difference



DS



CO



Initial state before the photoexcitation

Parameters

- Pump pulse

$$\boldsymbol{A}(\tau) = A_0 \exp\left[-\frac{(\tau-\tau_{\rm pu})^2}{2\sigma_{\rm pu}^2}\right](\cos\omega\tau, \sin\omega\tau)$$

Pulse width : $\sigma_{\rm pu} = 75T$

Pulse center : $\tau_{\rm pu} = 250T$

- Spectral function : $A_{\boldsymbol{k}}(\varepsilon, \tau_{\rm pr})$

J. K. Freericks *et al*., Phys. Rev. Lett. 102, 136401 (2009)
M. A. Sentef *et al*., Nat. Commun. 6, 7047 (2015)

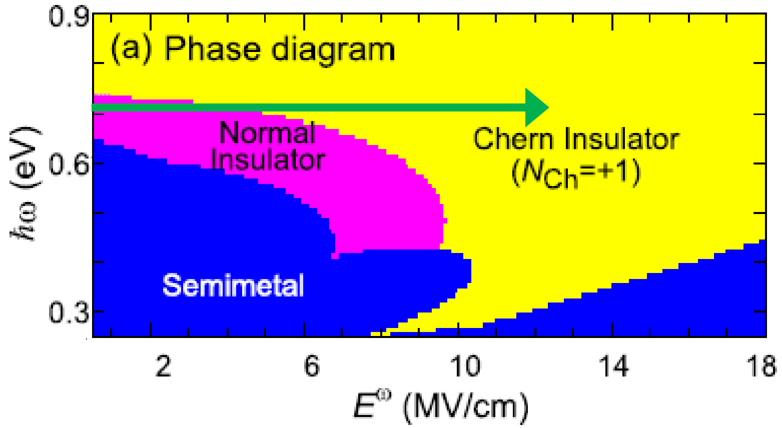
$$A_{\boldsymbol{k}}(\varepsilon, \tau_{\rm pr}) = {\rm Im}\sum_{\alpha}\int d\tau_1 d\tau_2 s(\tau_1-\tau_{\rm pr})s(\tau_2-\tau_{\rm pr}) e^{i\varepsilon(\tau_1-\tau_2)}\left[G^{<}_{\boldsymbol{k},\alpha\alpha}(\tau_1,\tau_2) - G^{>}_{\boldsymbol{k},\alpha\alpha}(\tau_1,\tau_2)\right]$$

Probe pulse: $s_{\sigma_{\rm pr}}(\tau-\tau_{\rm pr}) = \frac{1}{\sigma_{\rm pr}\sqrt{2\pi}}\exp\left[-\frac{(\tau-\tau_{\rm pr})^2}{2\sigma_{\rm pr}^2}\right]$

Pulse width : $\sigma_{\rm pr} = 25T$

Pulse center : $\tau_{\rm pr} = \tau_{\rm pu}$

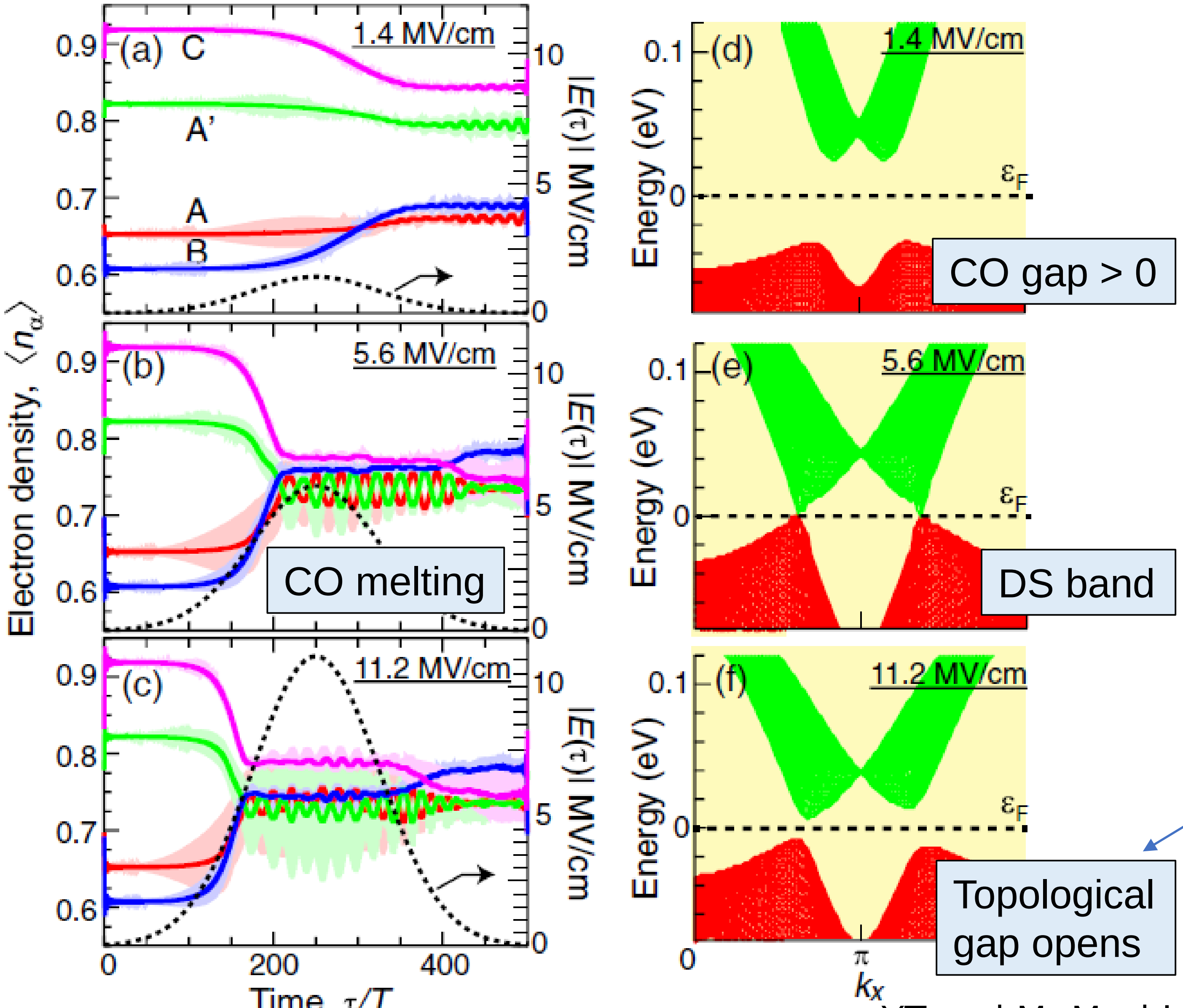
- Light frequency : $\omega = 0.7$



Floquet theory (noninteracting model)

K. Kitayama and M. Mochizuki, Phys. Rev. Res. 2, 023229 (2020)

Transient band and time evolution of electron density



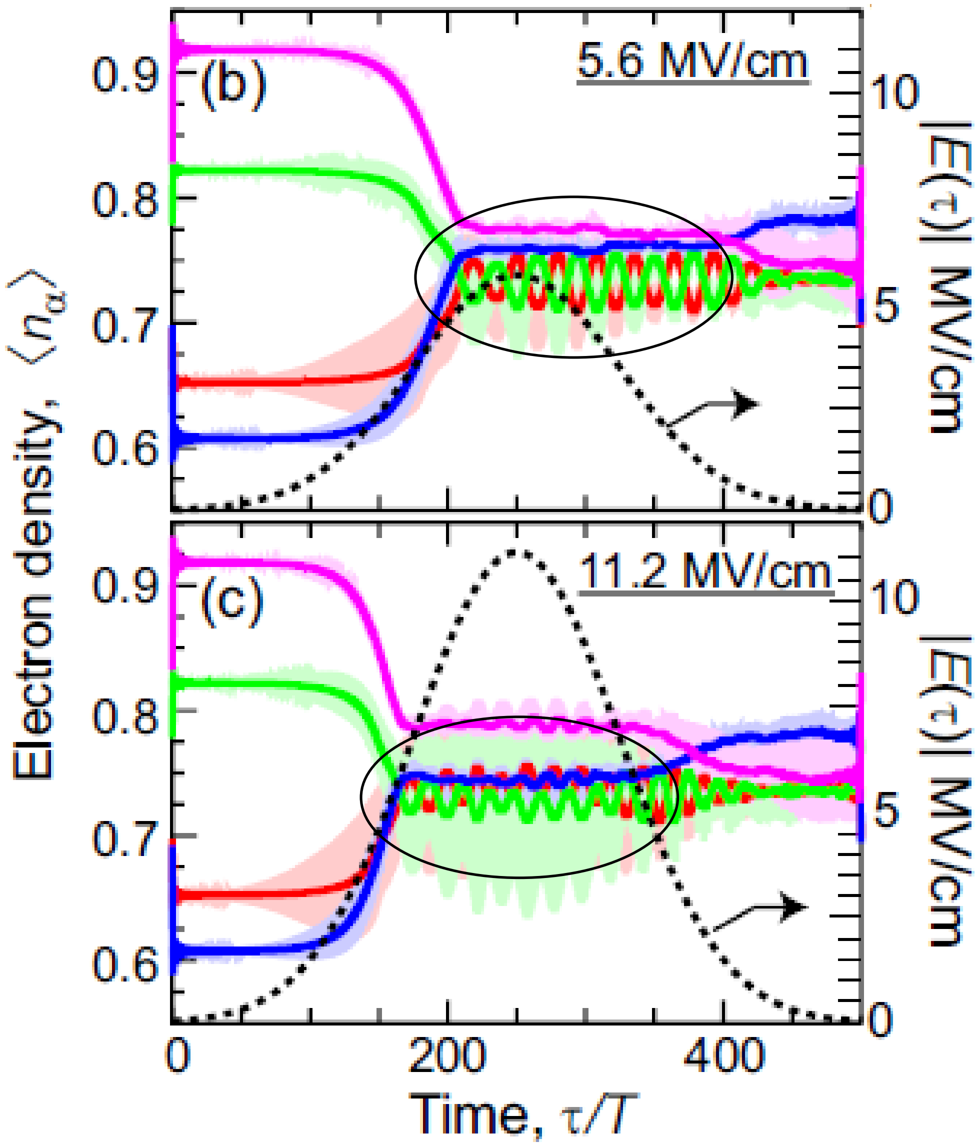
- Floquet theory

  K. Kitayama and M. Mochizuki, Phys. Rev. Res. (2020)
- Real-time dynamics

  YT and M. Mochizuki, Phys. Rev. B (2021)

YT and M. Mochizuki, Phys. Rev. Lett. 129, 047402 (2022)

Slow oscillation of charge densities



slow-oscillation components of $\langle n_A \rangle$ and $\langle n_{A'} \rangle$

→ the same oscillation centers

A and A′ sites are equivalent

→ oscillation centers slightly differ between $\langle n_A \rangle$ and $\langle n_{A'} \rangle$

A and A′ sites are NOT equivalent

Floquet theory

- Floquet theorem

K. Kitagawa et al., Phys. Rev. B 82, 235114 (2010)
M. Burkov *et al.*, Adv. Phys. 64, 139 (2015)

time-periodic Hamiltonian

$$\mathcal{H}(\tau) = \mathcal{H}(\tau + T)$$

$$i\frac{\partial}{\partial t}|\Psi(\tau)\rangle = \mathcal{H}(\tau)|\Psi(\tau)\rangle$$

$$|\Psi(t)\rangle = e^{-i\varepsilon t}|\Phi(t)\rangle$$

$$|\Phi(t)\rangle = |\Phi(t+T)\rangle$$: Floquet state

FFT with respect to time

$$\sum_{m=-\infty}^{\infty} \mathcal{H}_{nm}|\Phi_{m,\lambda}\rangle = \varepsilon_{n,\lambda}|\Phi_{n,\lambda}\rangle$$

Static eigenvalue equation

$$\mathcal{H}_{nm} = H_{n-m} - m\omega\delta_{n,m}$$

$$H_n = (1/T)\int_0^T \mathcal{H}(\tau)e^{in\omega\tau}d\tau$$

$$|\Phi_{n,\lambda}\rangle = (1/T)\int_0^T |\Phi(\tau)\rangle e^{in\omega\tau}$$

Floquet analysis with mean-field order parameters

- time profiles of mean-field order parameters

$$\rho_{ij}(\tau) \equiv \sum_\sigma \langle c^\dagger_{i\sigma} c_{j\sigma} \rangle \Rightarrow \rho_{ij,n} = \sum_{\tau=\tau_{\rm pu}-N_w T}^{\tau_{\rm pu}+N_w T} \rho_{ij}(\tau) e^{in\omega\tau}$$

FFT within a time domain around the pulse center

$N_w = 10$

H_n in our mean-field treatment

$$A(\tau) = (E^\omega/\omega)(\cos\omega\tau, \sin\omega\tau)$$

$$H_n^{\rm MF} = \sum_{\langle i,j \rangle} t_{ij} e^{-in\theta_{ij}} [J_n(-\mathcal{A}_{ij}) c^\dagger_{i\sigma} c_{j\sigma} + J_n(\mathcal{A}_{ij}) c^\dagger_{j\sigma} c_{i\sigma}] + \sum_{i,\sigma} \left(\frac{U}{2} \rho_{ii,n} + \sum_j V_{ij} \rho_{jj,n} \right) n_{i\sigma} - \frac{1}{2} \sum_{i,j,\sigma} V_{ij} \rho_{ij,n} c^\dagger_{j\sigma} c_{i\sigma}$$

$\mathcal{A}_{ij} \equiv (E^\omega/\omega)|\boldsymbol{\delta}_{ij}|$

$\theta_{ij} \equiv \tan^{-1}(\delta^x_{ij}/\delta^y_{ij})$

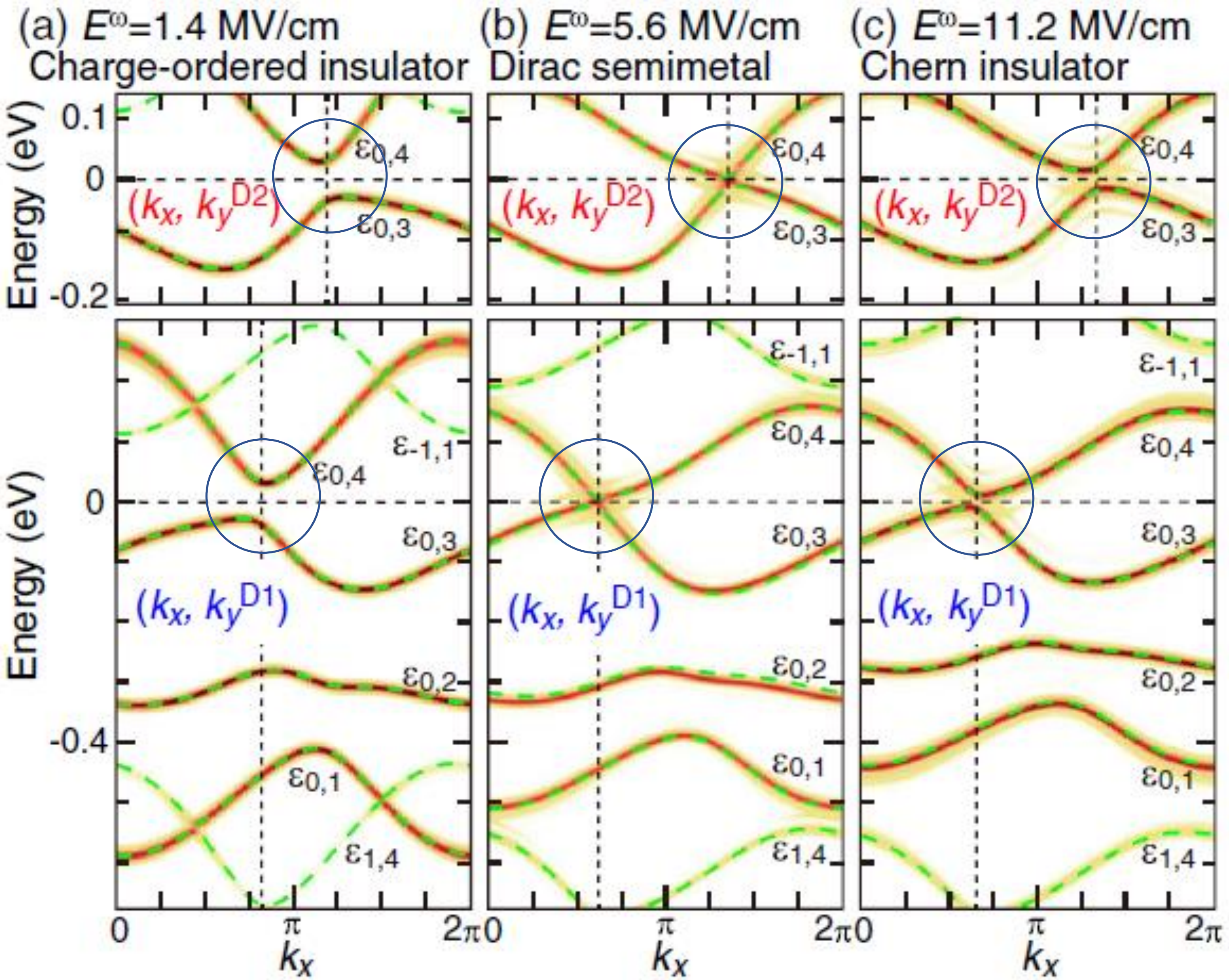
J_n : n-th Bessel's function

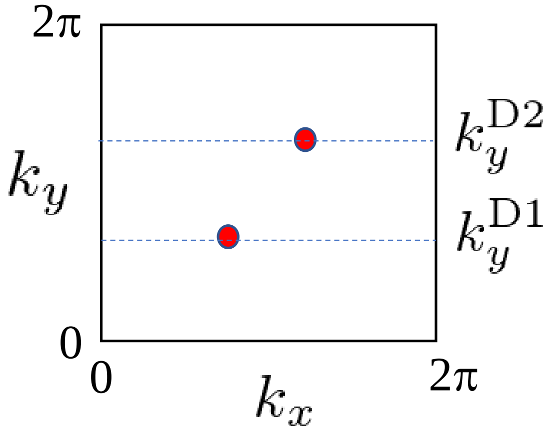
$$|n|, |m| \le 10 \Rightarrow \varepsilon_{n,\lambda}(\boldsymbol{k}), |\Phi_{n,\lambda}(\boldsymbol{k})\rangle$$

Chern number : $N_{\rm Ch} = \sum_{\lambda=1}^{3} N^\lambda_{\rm Ch}$

T. Fukui *et al.,* J. Phys. Soc. Jpn. 74, 1674 (2005)

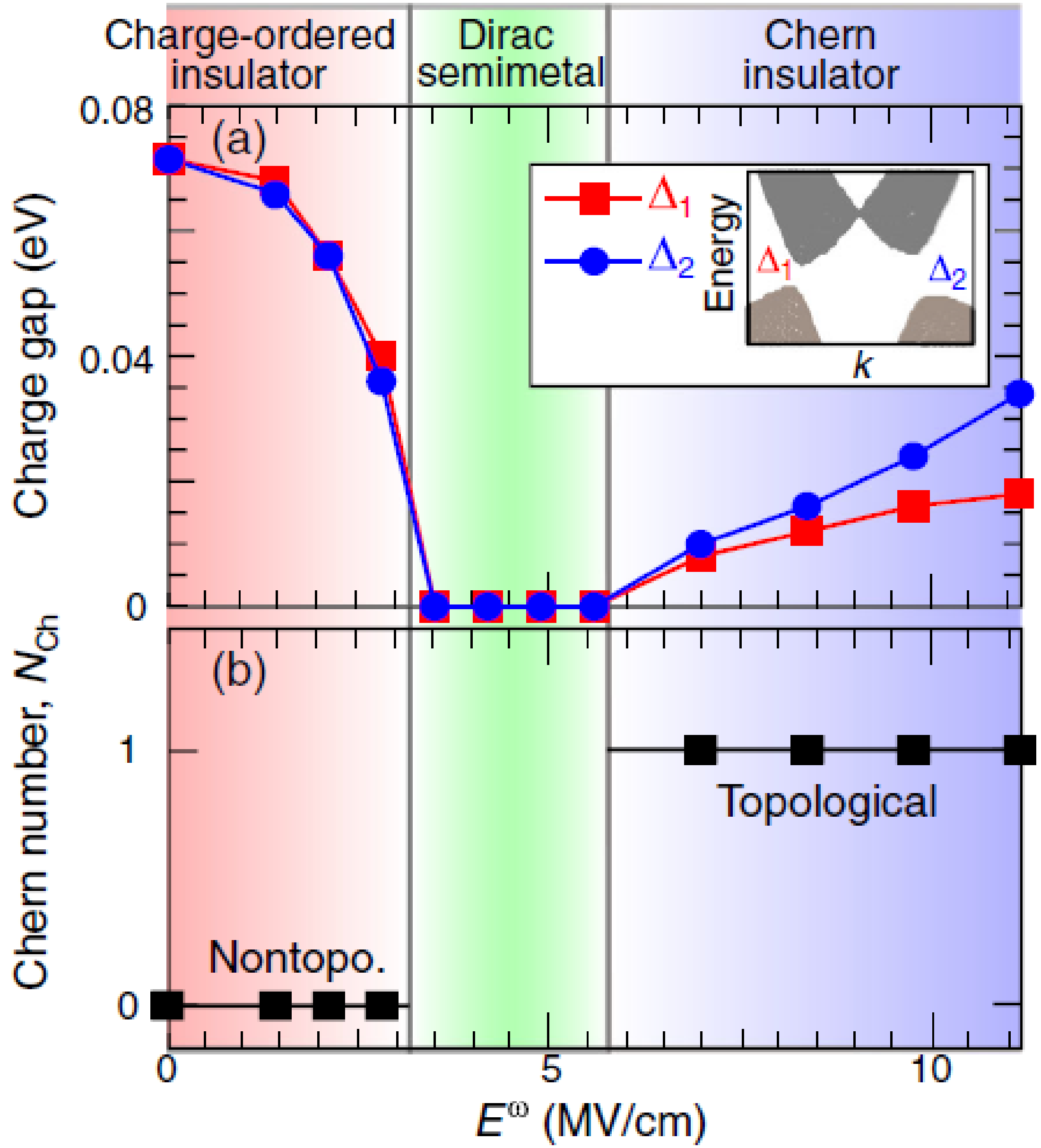
Floquet bands and transient bands





Floquet bands $\varepsilon_{n,\lambda}(\boldsymbol{k})$ coincide with $A_{\boldsymbol{k}}(\varepsilon, \tau_{\mathrm{pr}})$

E^ω dependence of charge gap and Chern number



$E^\omega > 5.6\ \mathrm{MV/cm}$

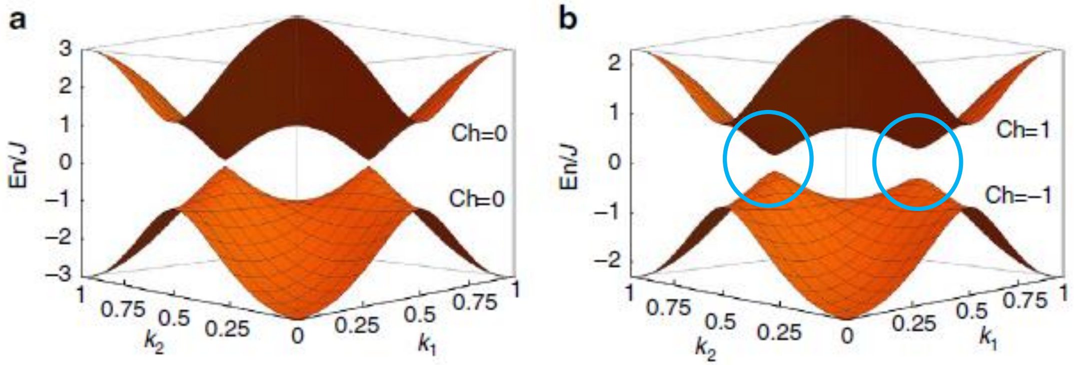
staggered site potential ϕ_A and $\phi_{A'}$

➡ $\Delta_1 \neq \Delta_2$

honeycomb lattice

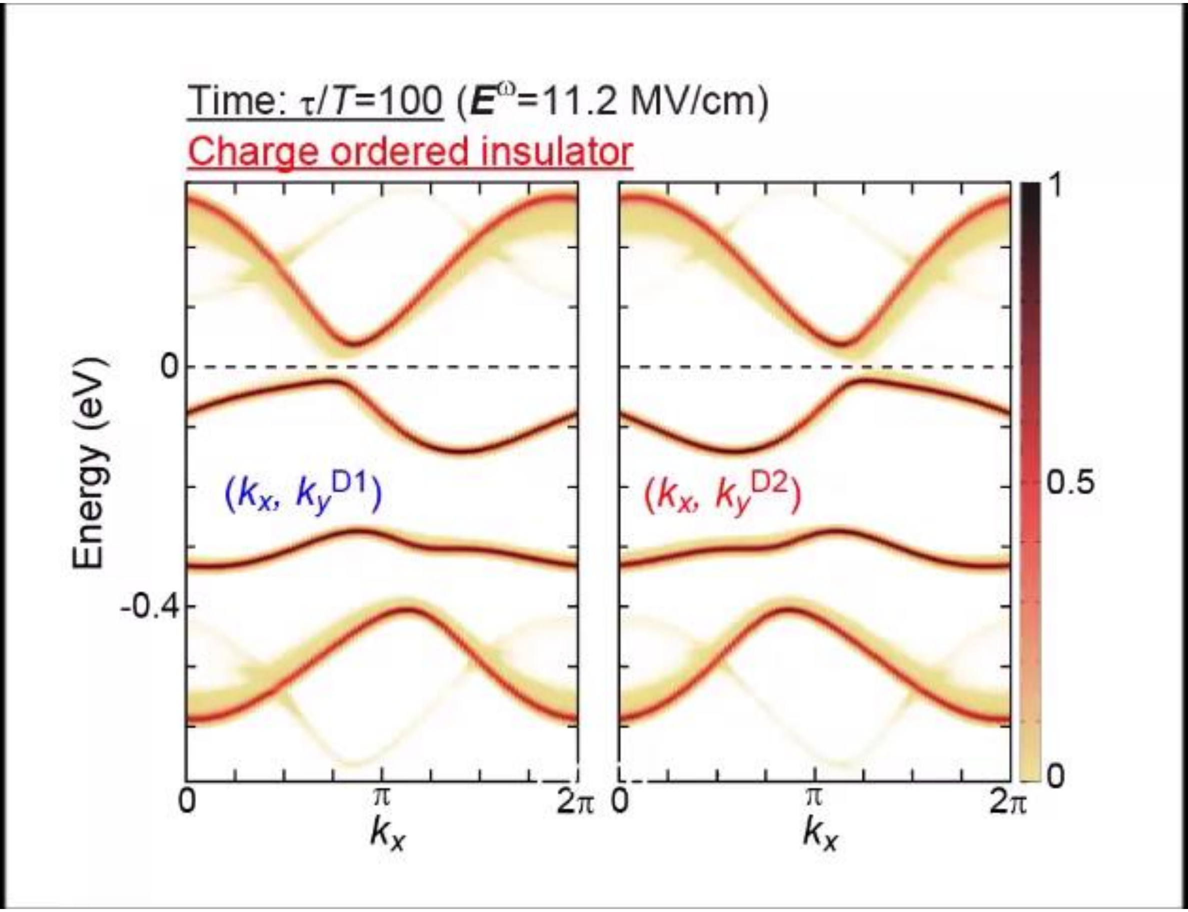
+ staggerd sublattice potential Δ + CPL

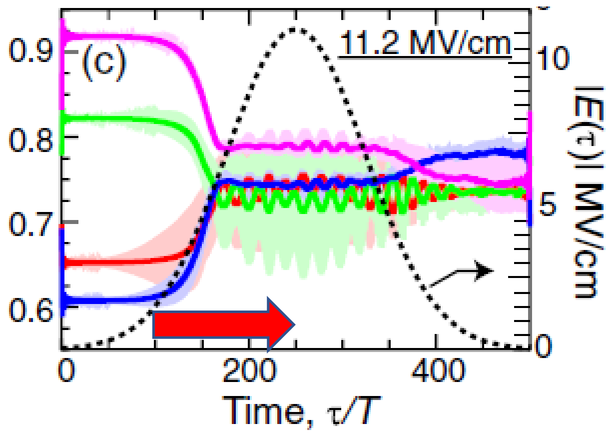
L D'Alessio and M. Rigol, Nat. Commun. 6. 8336 (2014)



Floquet theory

Time evolution of transient bands





YT and M. Mochizuki, Phys. Rev. Lett. 129, 047402 (2022)

Summary

Melting of CO and topological phase transition induced by circularly polarized light in organic conductor α-(BEDT-TTF)$_2$I$_3$

- Combined method of numerical simulation based on time-dependent Schrodinger eq. and Floquet theory
- Transient spectral function and time evolution of electron densities

Effects of CPL

Closing the charge gap through melting the CO

Opening the topological gap at the Dirac point

⇨ Successive dynamical phase transitions

CO melting ➡ gapless DF band ➡ Floquet Chern ins.

YT and M. Mochizuki, Phys. Rev. Lett. 129, 047402 (2022)