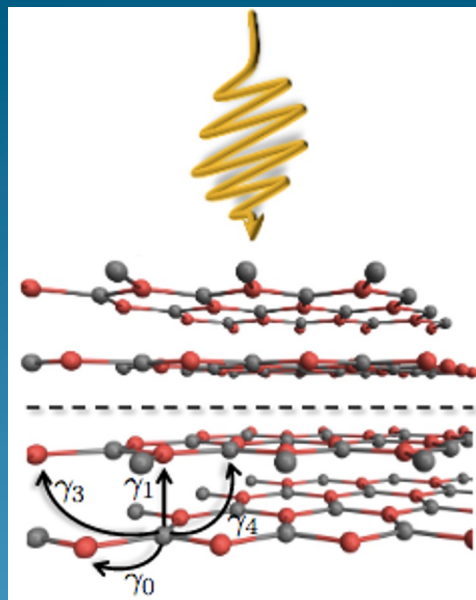


Inducing Quantum States in Materials using Light



Gregory A. Fiete
Northeastern University

Outline



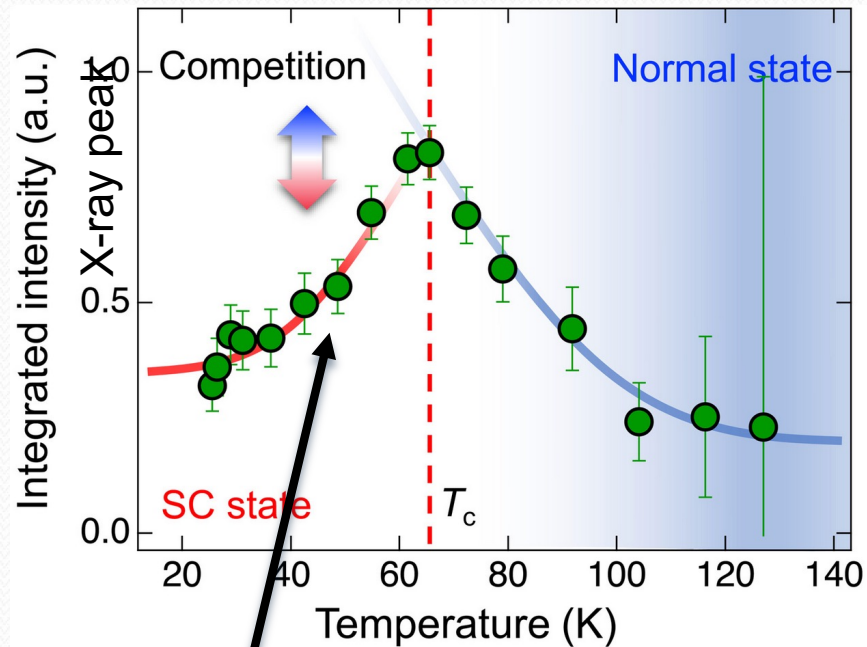
- Introduction
 - General considerations for laser-driven systems
- Time-dependent Ginzburg-Landau theory for competing orders
 - Essential formalism and results
- Creating pseudo-magnetic fields with light
 - Signatures in optical responses
- Reversible opto-electric control of lattice structure w/light
 - Modifying crystal chirality & nonlinear signatures



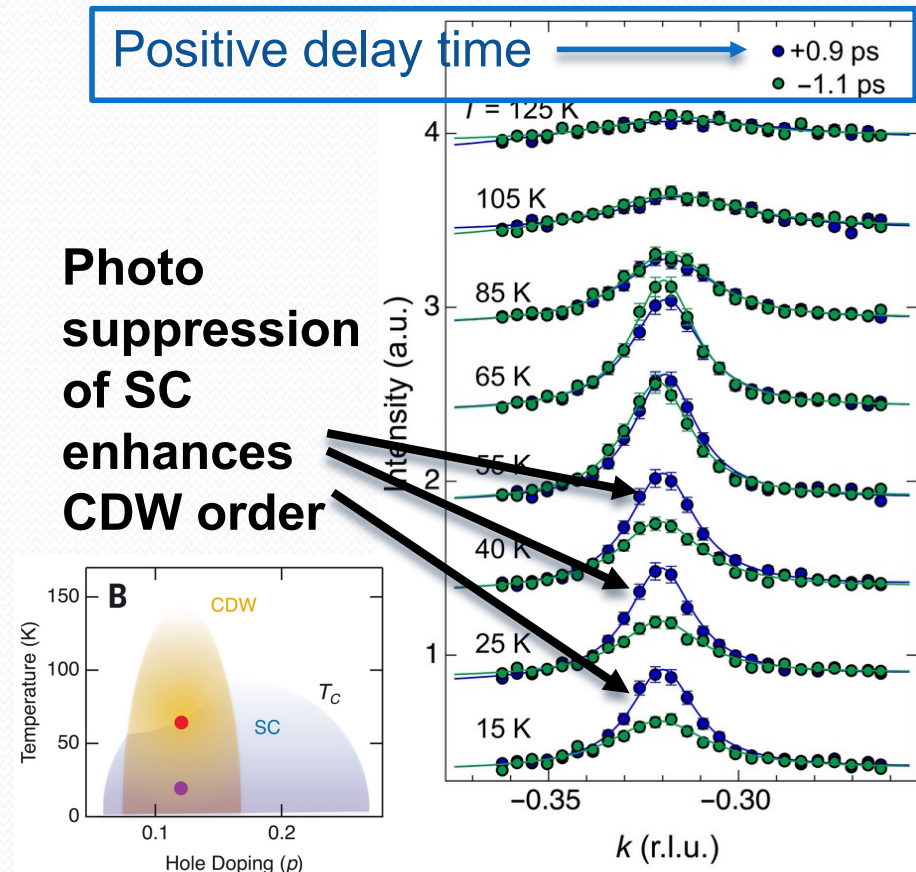
Laser Parameters for Driven Materials

- Parameters that can be controlled
 - Polarization (linear vs. circular)
 - Frequency (selectively couple to electrons or phonons)
 - Intensity (fluence, time integrated flux of laser)
 - Angle of incidence relative to material orientation
 - Pulse shaping (used in quantum chemistry)
 - Multiple drive lasers (could produce a “response on top of a response”)

Enhanced CDW order with a photo suppressed SC state in $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$

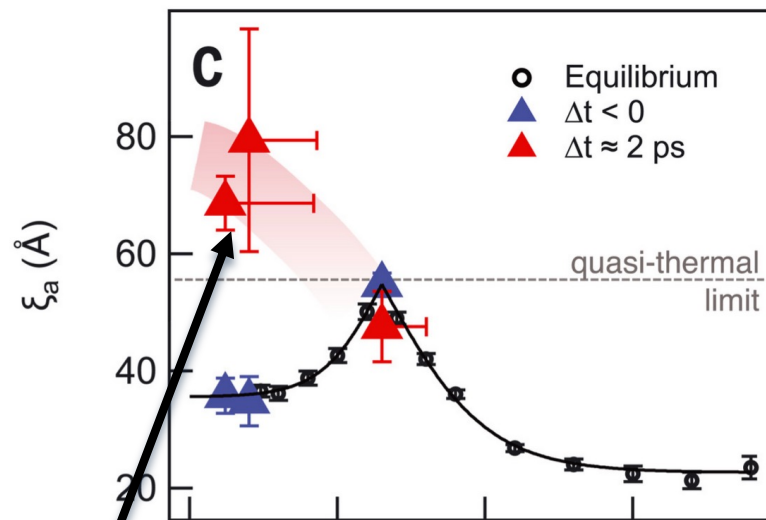


Suppression of CDW order below T_c suggests competition with SC order

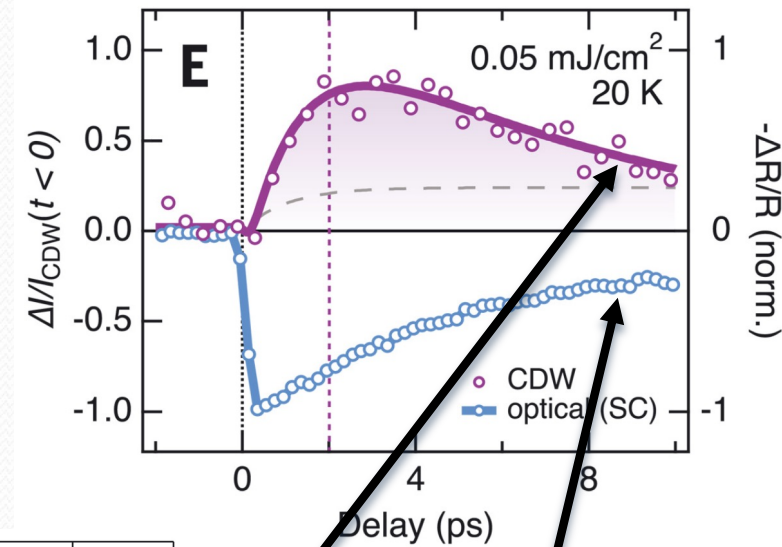
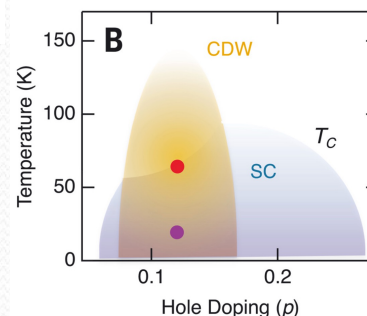


G. Ghiringhelli, G. A. Sawatsky, E. Weschke, B. Keimer, and L. Braicovich, *Science* **337**, 821 (2012).
 H. Jang ... B. Keimer, C.-C. Kao, and J.-S. Lee, *Sci. Adv.* **8** (2022).

Enhanced CDW order with a photo suppressed SC state in $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$



Enhancement of CDW order following pump



CDW and SC relax on similar time scales indicating coupling

G. Ghiringhelli, G. A. Sawatsky, E. Weschke, B. Keimer, and L. Braicovich, *Science* **337**, 821 (2012)
 H. Jang ... B. Keimer, C.-C. Kao, and J.-S. Lee, *Sci. Adv.* **8** (2022).

Non-thermal states: A. de la Torre, D. M. Kennes, M. Claassen, S. Gerber, J.W. McIver, M. A. Sentef, *RMP* **93**, 041002 (2021).

Theory for metastability in a pumped material with coexisting competing orders

Order parameter coupling

$$F = \int d^2\mathbf{r} \left(f_1 + f_2 + c|\psi_1|^2|\psi_2|^2 + \frac{L}{2}(\nabla \times \mathbf{A})^2 \right),$$

$$f_i = \frac{1}{2}\alpha_i(t)|\psi_i|^2 + \frac{1}{2}b_i|D_\mu\psi_i|^2 + u_i|\psi_i|^4.$$

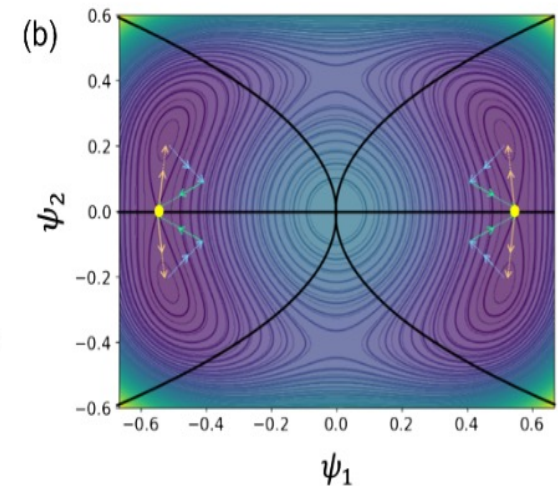
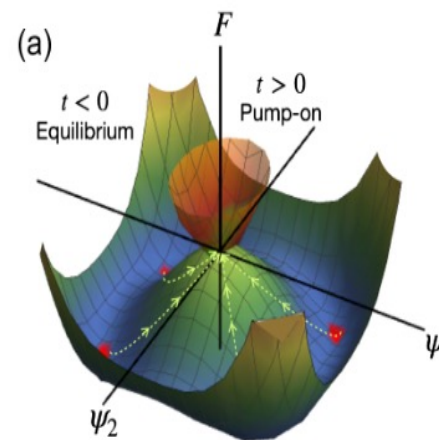
$$\alpha_2(t) = \begin{cases} -\alpha_2^{\text{eq}} < 0 & \text{if } t < 0 \\ \alpha'_2 > 0 & \text{if } 0 < t < \tau_0 \\ (\alpha'_2 + \alpha_2^{\text{eq}})e^{-\frac{t-\tau_0}{t_c}} - \alpha_2^{\text{eq}} & \text{if } t > \tau_0 \end{cases}$$

Dominant coupling to one order

$$\frac{\partial\psi_i(\mathbf{r}, t)}{\partial t} = -\Gamma_i \frac{\delta F}{\delta\psi_i(\mathbf{r}, t)} + \eta_i(\mathbf{r}, t),$$

Order parameter dynamics w/noise

$$\langle \eta_i(\mathbf{r}, t) \eta_j(\mathbf{r}', t') \rangle = \Gamma_i T \delta_{i,j} \delta(t - t') \delta(\mathbf{r} - \mathbf{r}')$$



Theory for metastability in a pumped material with coexisting competing orders

$$F = \int d^2\mathbf{r} \left(f_1 + f_2 + c|\psi_1|^2|\psi_2|^2 + \frac{L}{2}(\nabla \times \mathbf{A})^2 \right),$$

$$f_i = \frac{1}{2}\alpha_i(t)|\psi_i|^2 + \frac{1}{2}b_i|D_\mu\psi_i|^2 + u_i|\psi_i|^4.$$

Laser intensity $\longrightarrow I(t) = I_0 \left(1 + 4 \left(\frac{t}{k_0 c \tau_0^2} \right)^2 \right) e^{-2\left(\frac{t}{\tau_0}\right)^2}$

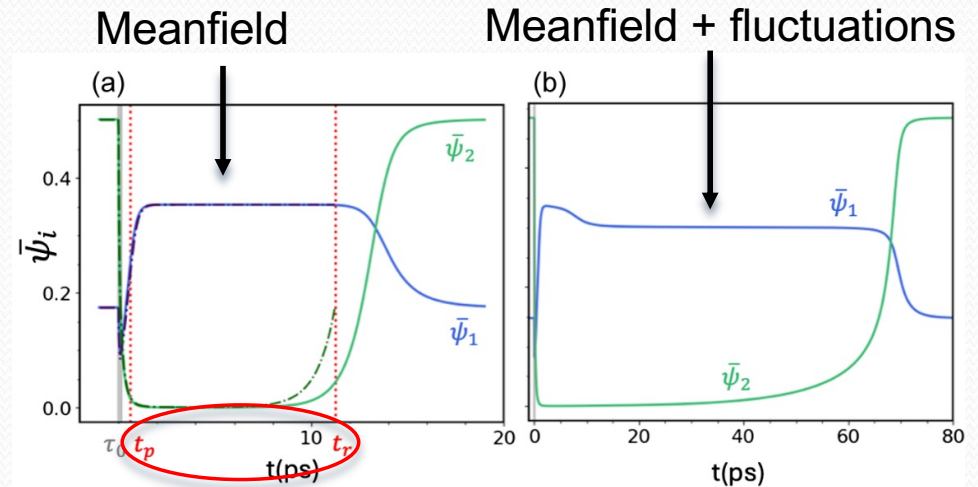
Closed-form expression for metastability time

$$\frac{2u_1}{\Gamma_2 c} \ln \frac{\bar{\psi}_{2,p}}{\bar{\psi}_1^0} = \left(\alpha_1^{\text{eq}} - 2 \frac{u_1}{c} \alpha_2^{\text{eq}} \right) (t_r - t_p) - \frac{2u_1}{c} t_c (\alpha_2' + \alpha_2^{\text{eq}}) e^{-\frac{t_p - \tau_0}{t_c}} \left(e^{\frac{t_r - t_p}{t_c}} - 1 \right)$$

Model A dynamics:

$$\frac{\partial \psi_i(\mathbf{r}, t)}{\partial t} = -\Gamma_i \frac{\delta F}{\delta \psi_i(\mathbf{r}, t)} + \eta_i(\mathbf{r}, t),$$

$$\alpha_2(t) = \begin{cases} -\alpha_2^{\text{eq}} < 0 & \text{if } t < 0 \\ \alpha_2' > 0 & \text{if } 0 < t < \tau_0 \\ (\alpha_2' + \alpha_2^{\text{eq}}) e^{-\frac{t-\tau_0}{t_c}} - \alpha_2^{\text{eq}} & \text{if } t > \tau_0 \end{cases}$$



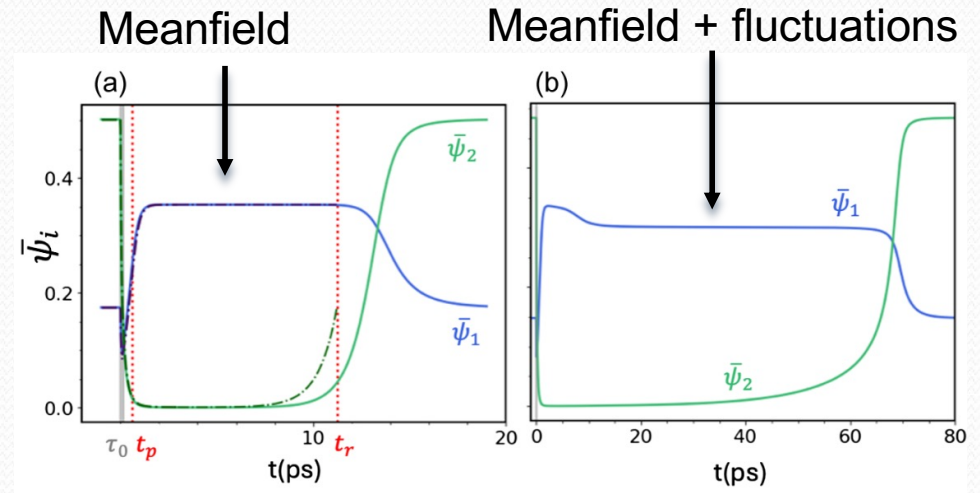
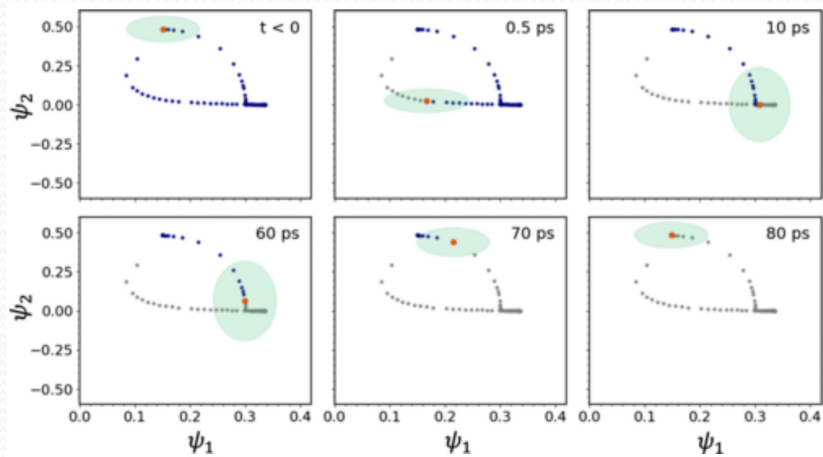
Y. Masoumi, A. de la Torre, *GAF Phys. Rev. Lett.* **135**, 066551 (2025)

Theory for metastability in a pumped material with coexisting competing orders

Spatial fluctuations $\longrightarrow \psi_j(\mathbf{r}) = \bar{\psi}_j + \sum_{\mathbf{k} \neq 0} \delta\psi_{\mathbf{k}}^j e^{i\mathbf{k} \cdot \mathbf{r}} \quad C_{\mathbf{q}}^{ij} = \langle \delta\psi_{\mathbf{q}}^i \delta\psi_{-\mathbf{q}}^j \rangle_c \quad S^{ij} = [1/(2\pi)^2] \int_0^\Lambda d^2\mathbf{q} C_{\mathbf{q}}^{ij}$

Order parameter dynamics $\longrightarrow -\frac{1}{\Gamma_i} \partial_t \bar{\psi}_i = r_i \bar{\psi}_i + 4c \bar{\psi}_j S^{ij} \quad r_i = \alpha_i(t) + b_i e_i^2 I(t) + 4u_i(\bar{\psi}_i^2 + 3S^{ii}) + 2c(\bar{\psi}_j^2 + S^{jj})$

Correlation dynamics $\longrightarrow -\frac{1}{2\Gamma_i} \partial_t C_{\mathbf{q}}^{ii} = \beta_i C_{\mathbf{q}}^{ii} + 4c(\bar{\psi}_i \bar{\psi}_j + S^{ij}) C_{\mathbf{q}}^{ij} - T_v \quad \beta_i = r_i + 8u_i \bar{\psi}_i^2 + b_i \mathbf{q}^2$
 $\partial_t C_{\mathbf{q}}^{ij} = -\Gamma_i(\beta_i C_{\mathbf{q}}^{ij} + 4c(\bar{\psi}_i \bar{\psi}_j + S^{ij}) C_{\mathbf{q}}^{jj}) + (i \leftrightarrow j)$

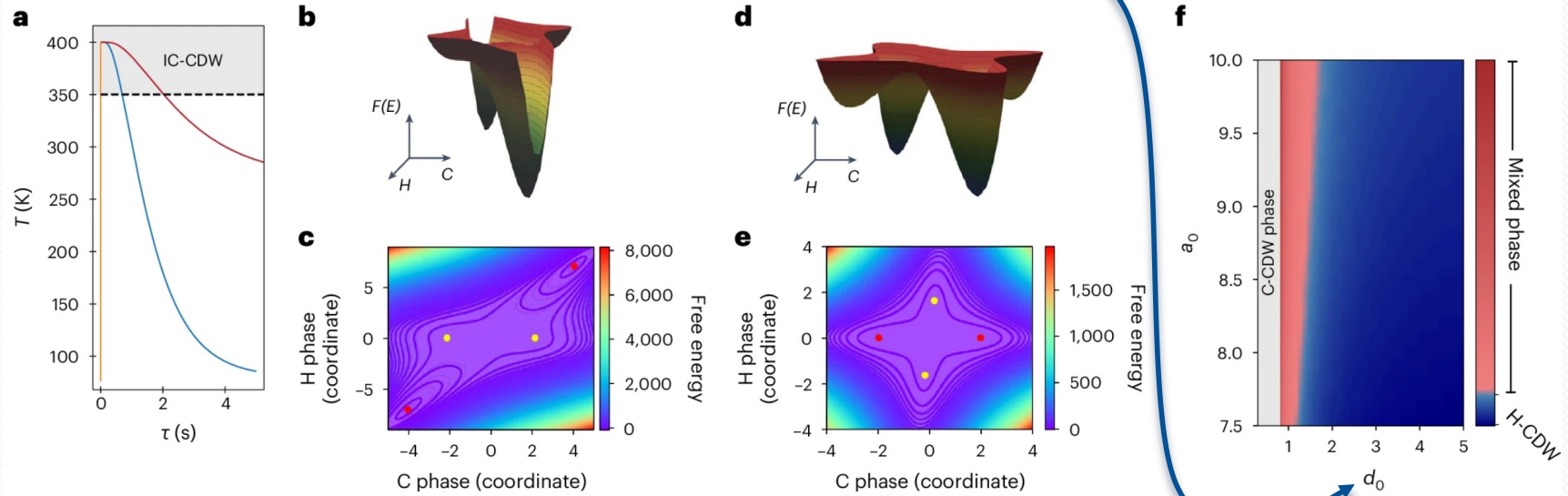
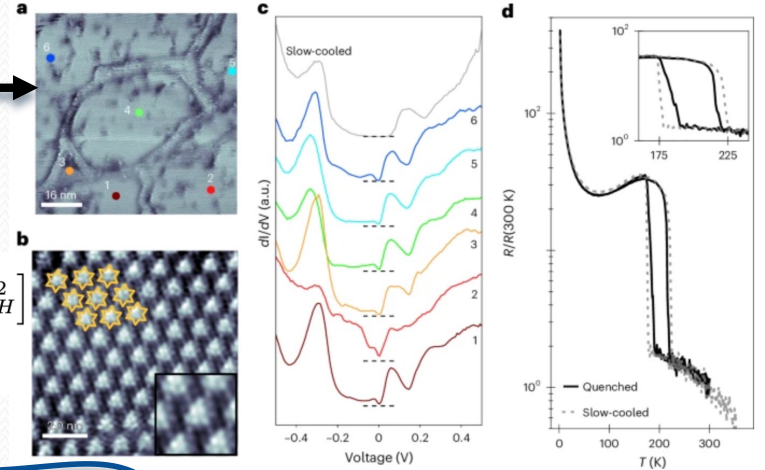


Y. Masoumi, A. de la Torre, GAF *Phys. Rev. Lett.* **135**, 066551 (2025)

Application to metastable CDW state in 1T-TaS₂

Metastable state is mixed CDW with domains for sample cooling at a specified rate; **persists up to 210K while warming and 180K while cooling**

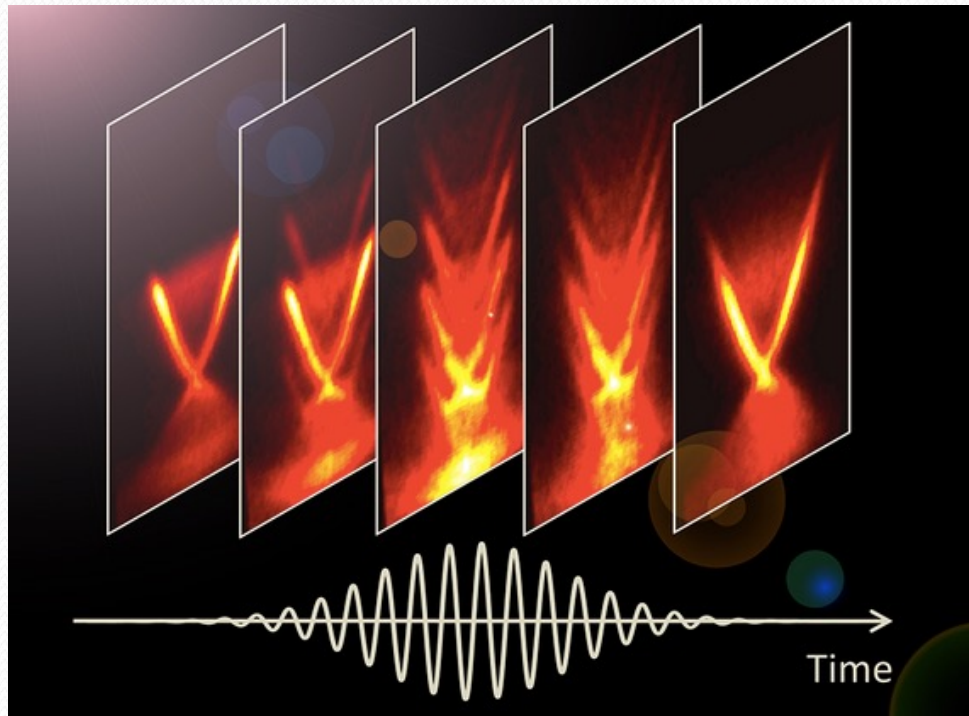
$$F_0 = -\frac{a_0}{2}(\bar{\phi}_C^2 + \bar{\phi}_H^2) + \frac{3c_0}{8}(\bar{\phi}_C^4 + \bar{\phi}_H^4) - \frac{3c_1}{4}\bar{\phi}_C\bar{\phi}_H^3 + e_0[(\vec{p}_1 \cdot \vec{Q}_C - p_1^2)\bar{\phi}_C^2 + (\vec{p}_2 \cdot \vec{Q}_H - p_2^2)\bar{\phi}_H^2] + f_0[(\vec{p}_1 \times \vec{Q}_C)^2\bar{\phi}_C^2 + (\vec{p}_2 \times \vec{Q}_H)^2\bar{\phi}_H^2] + d_0\bar{\phi}_C^2\bar{\phi}_H^2.$$



A. de la Torre, Q. Wang, Y. Masoumi, B. Campbell, J. V. Riffle, D. Balasundaram, P. M. Vora, J. P. C. Ruff, GAF, S. M. Hollen, and K. W. Plumb, *Nat. Phys.* **21**, 1267 (2025).

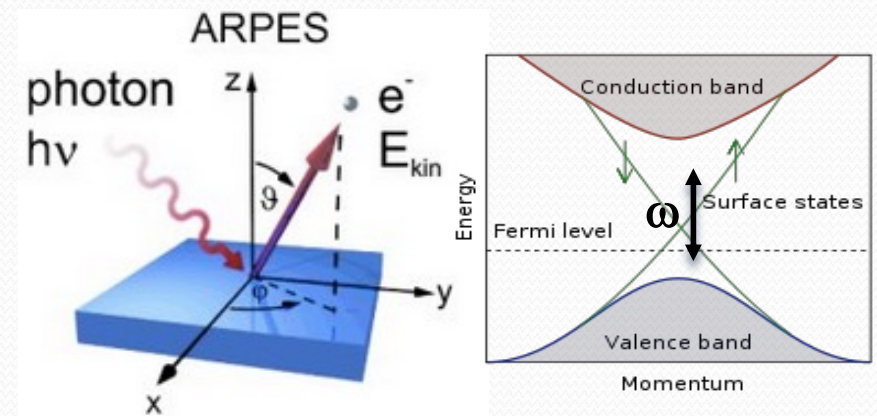
Experimental Realization of Floquet (time-periodic) Systems

- Time-resolved ARPES: Nuh Gedik Group, MIT



Surface states of Bi_2Se_3

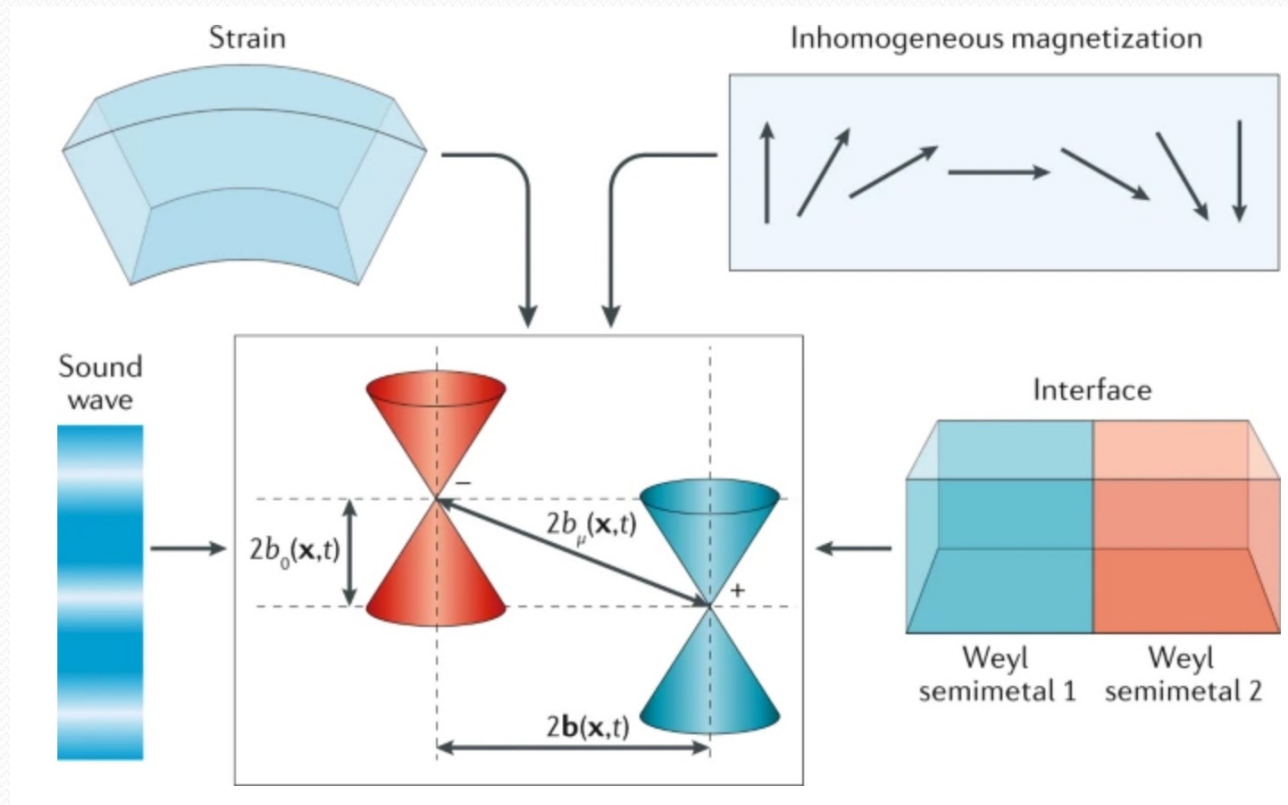
Floquet states appear and go as periodic drive is present and then absent. (Subgap drive.)



Wang, Steinberg, Jarillo-Herrero, and Gedik *Science* (2013)

Mahmood, Chan, Alpichshev, Gardner, Lee, Lee, and Gedik *Nat. Phys.* (2016)

Pseudo-magnetic fields from space-dependent Weyl node separation (e.g., bending a material)



Pseudo-magnetic fields act on electrons just like real magnetic fields and lead to Landau levels, etc.

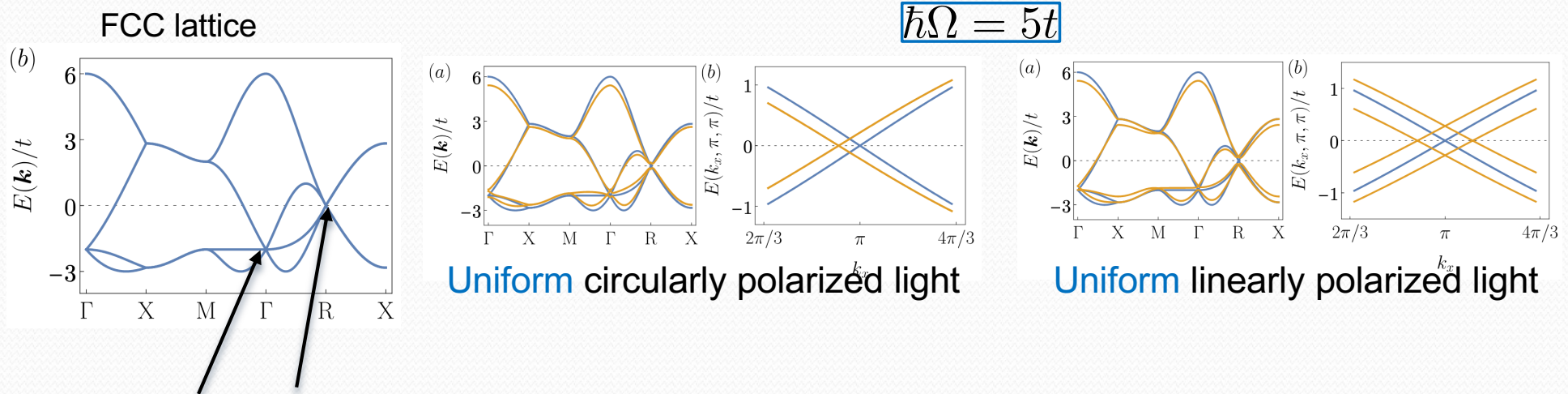
A. G. Grushin, J. W. F. Venderbose, A. Vishwanath, and R. Ilan, *Phys. Rev. X* **6**, 041046 (2016)

R. Ilan, A. G. Grushin, and D. I. Pikulin, *Nat. Rev. Phys.* **2**, 29 (2020)

Creating pseudo-magnetic fields mimicking strain with spatially inhomogeneous light

- We consider a Hamiltonian:

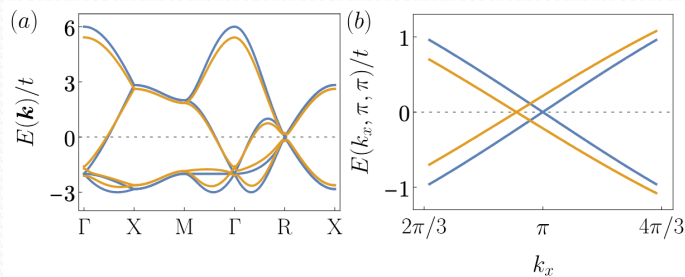
$$\mathcal{H}(\mathbf{k}) = 2t \left[\cos \frac{k_x}{2} \tau_0 \left(\cos \frac{k_y}{2} \sigma_x - \sin \frac{k_y}{2} \sigma_y \right) + \cos \frac{k_y}{2} \left(\cos \frac{k_z}{2} \tau_x \sigma_0 - \sin \frac{k_z}{2} \tau_y \sigma_z \right) + \cos \frac{k_z}{2} \left(\cos \frac{k_x}{2} \tau_x \sigma_x - \sin \frac{k_x}{2} \tau_y \sigma_y \right) \right]$$



$$\mathbf{E}_5 = -\partial_t \mathbf{A}_5$$

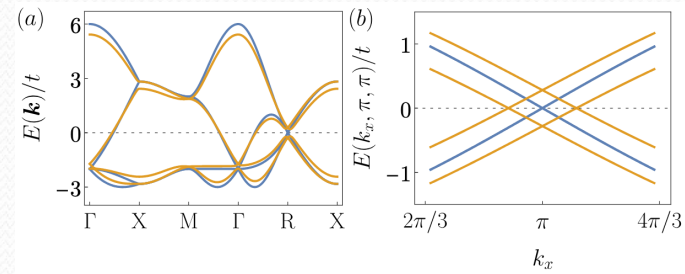
Shifting Weyl points in $\mathbf{k}$ -space can create axial vector potentials $\mathbf{A}_5(\mathbf{r})$ and **chiral anomaly**

Creating pseudo-magnetic fields mimicking strain with spatially inhomogeneous light



Uniform circularly polarized light

$$\hbar\Omega = 5t$$



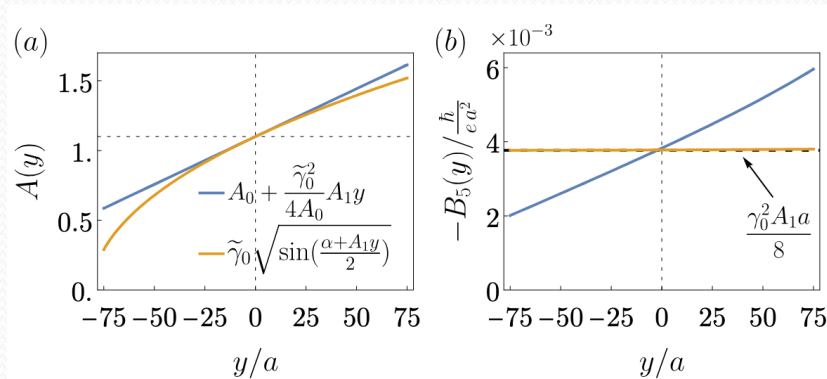
Uniform linearly polarized light

Nonuniform linearly polarized light:

$$\mathbf{A}(y, t) = A(y) \sin(\Omega t)(\hat{\mathbf{y}} + \hat{\mathbf{z}})$$

$$\mathbf{B}_5(y) = \nabla \times \mathbf{A}_5(y)$$

$$\mathbf{A}_5(\mathbf{r}) = \frac{2\hbar}{e a} \cos^{-1} \left(\frac{J_0 \left(\frac{a A(y)}{\hbar/e} \right) - 1}{2J_0 \left(\frac{a A(y)}{2\hbar/e} \right)} \right) \hat{\mathbf{x}}$$

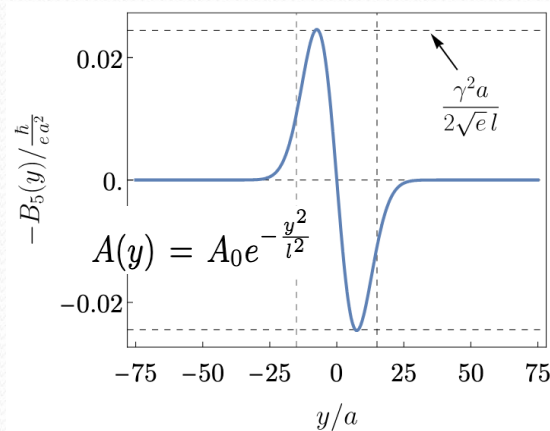


Internal magnetic fields $\sim 100\text{T}$

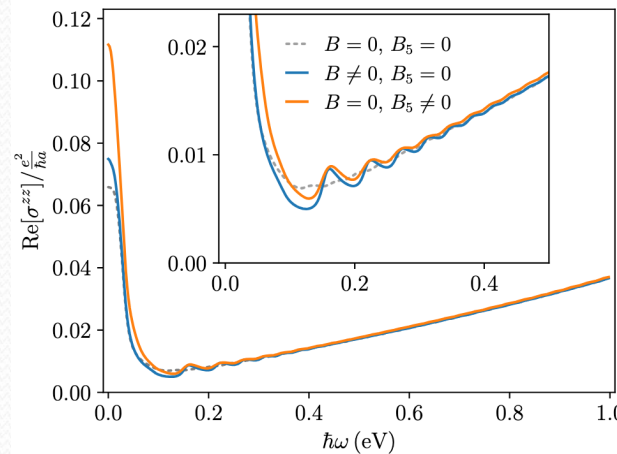
How can one distinguish pseudo-magnetic fields from real magnetic fields in physical responses?

Advantages: dynamic control, complete reversibility, spatial selectivity and ultra-fast switching timescales with no material deformation required.

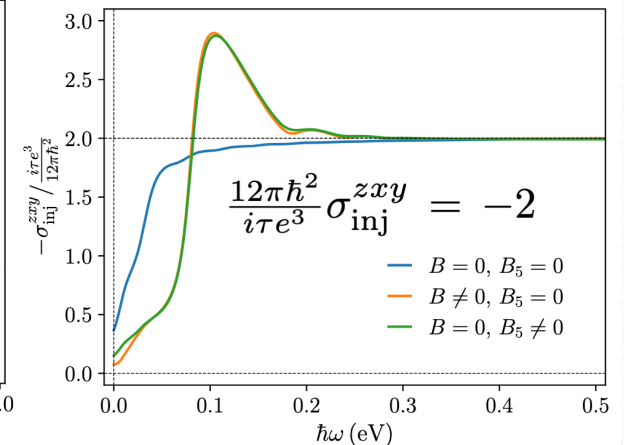
Create pseudo-magnetic domain walls:



Can be dynamically created, moved and erased by adjusting laser profile.

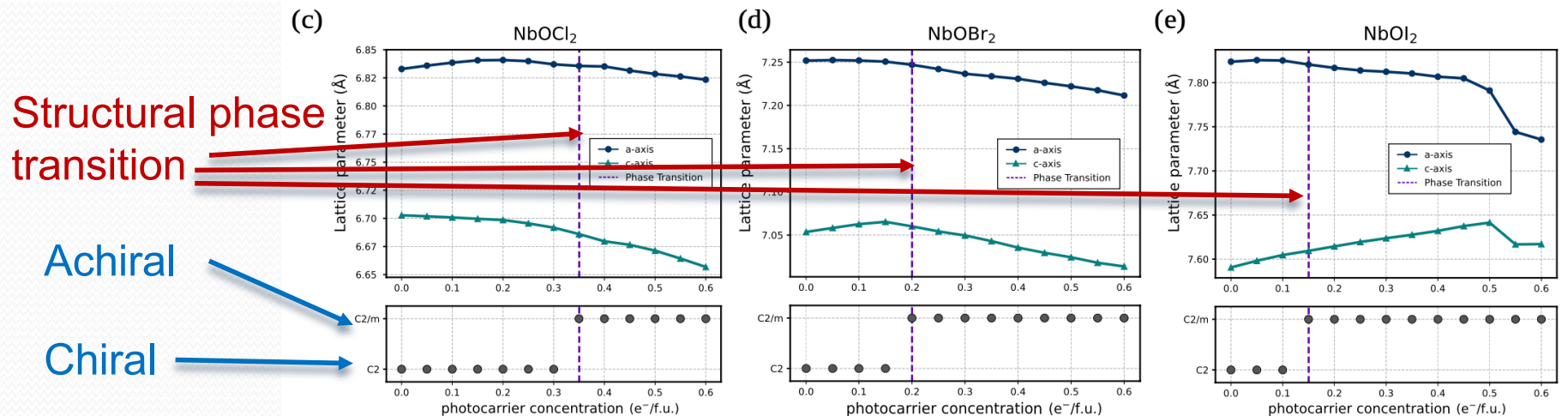
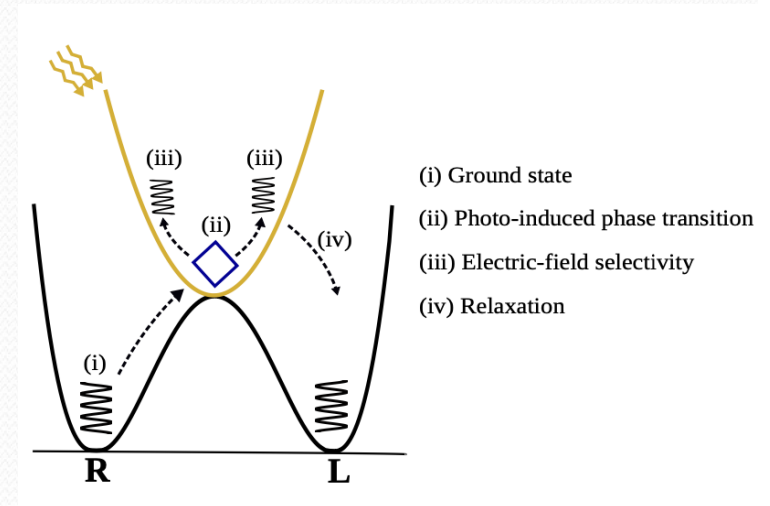
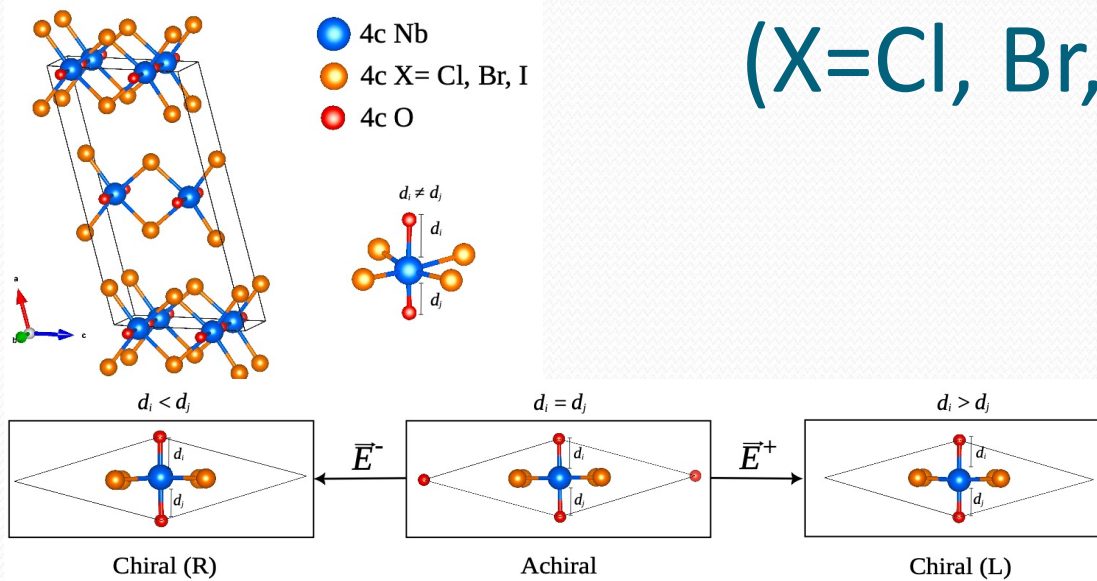


Longitudinal linear conductivity



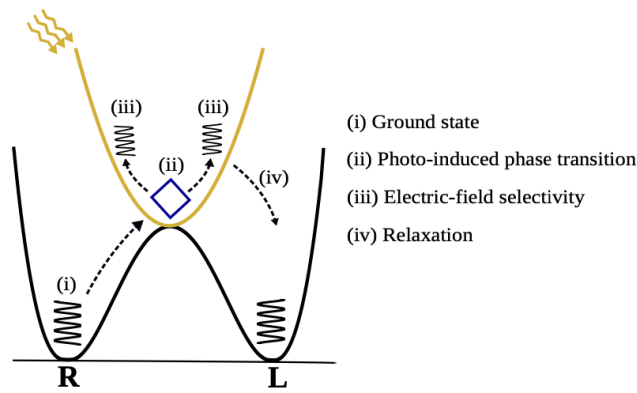
Second order injection conductivity

Enantiomer Selectivity in Chiral NbOX₂ (X=Cl, Br, I)

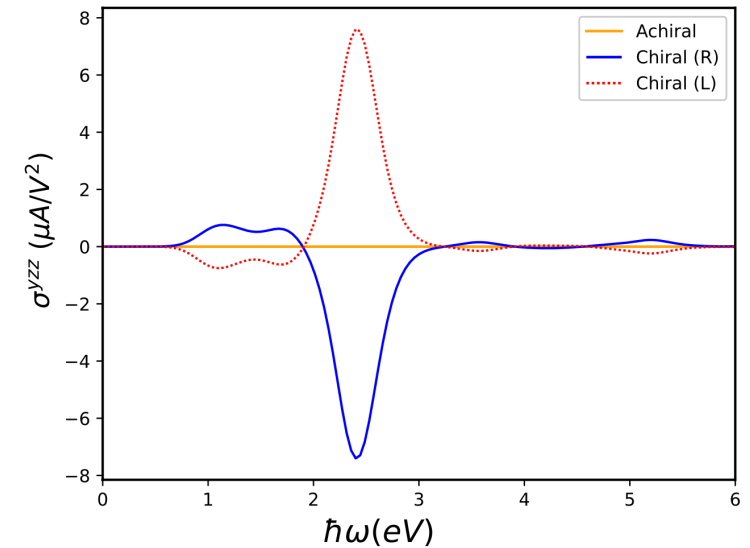


J. Cardenas-Gamboa, M. Gutierrez-Amigo, A. Leonardo, GAF, J. L. Manes, J. van den Brink, C. Felser, and M. G. Vergniory, *npj Quantum Materials* **11**, 56 (2026).

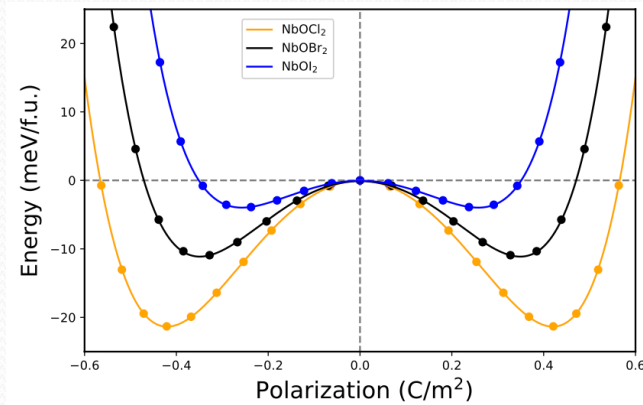
Enantiomer Selectivity in Chiral NbOX₂ (X=Cl, Br, I)



Shift current



Small energy barrier



Estimate fluence for transition

$$n_e = \frac{I_0(1 - R)\alpha\Omega_0}{\hbar\omega}$$

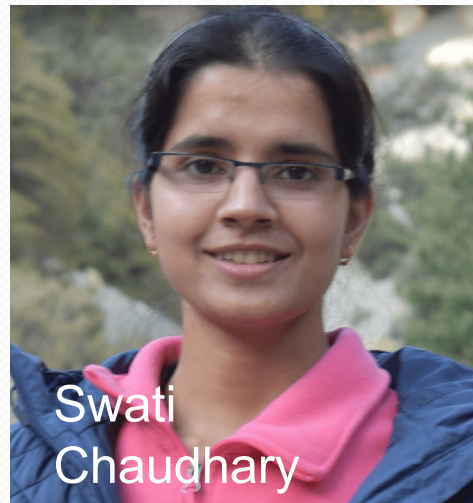
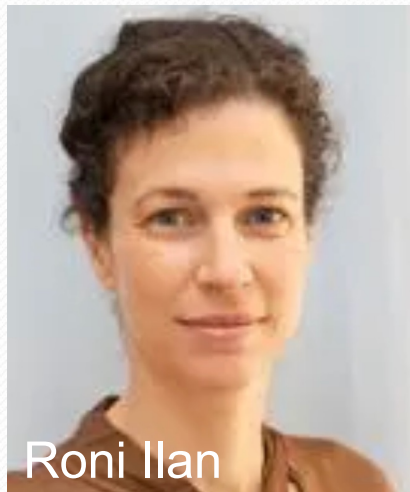
$$1.55 - 25.3 \text{ mJ/cm}^2$$

J. Cardenas-Gamboa, M. Gutierrez-Amigo, A. Leonardo, GAF, J. L. Manes, J. van den Brink, C. Felser, and M. G. Vergniory, *npj Quantum Materials* **11**, 56 (2026).

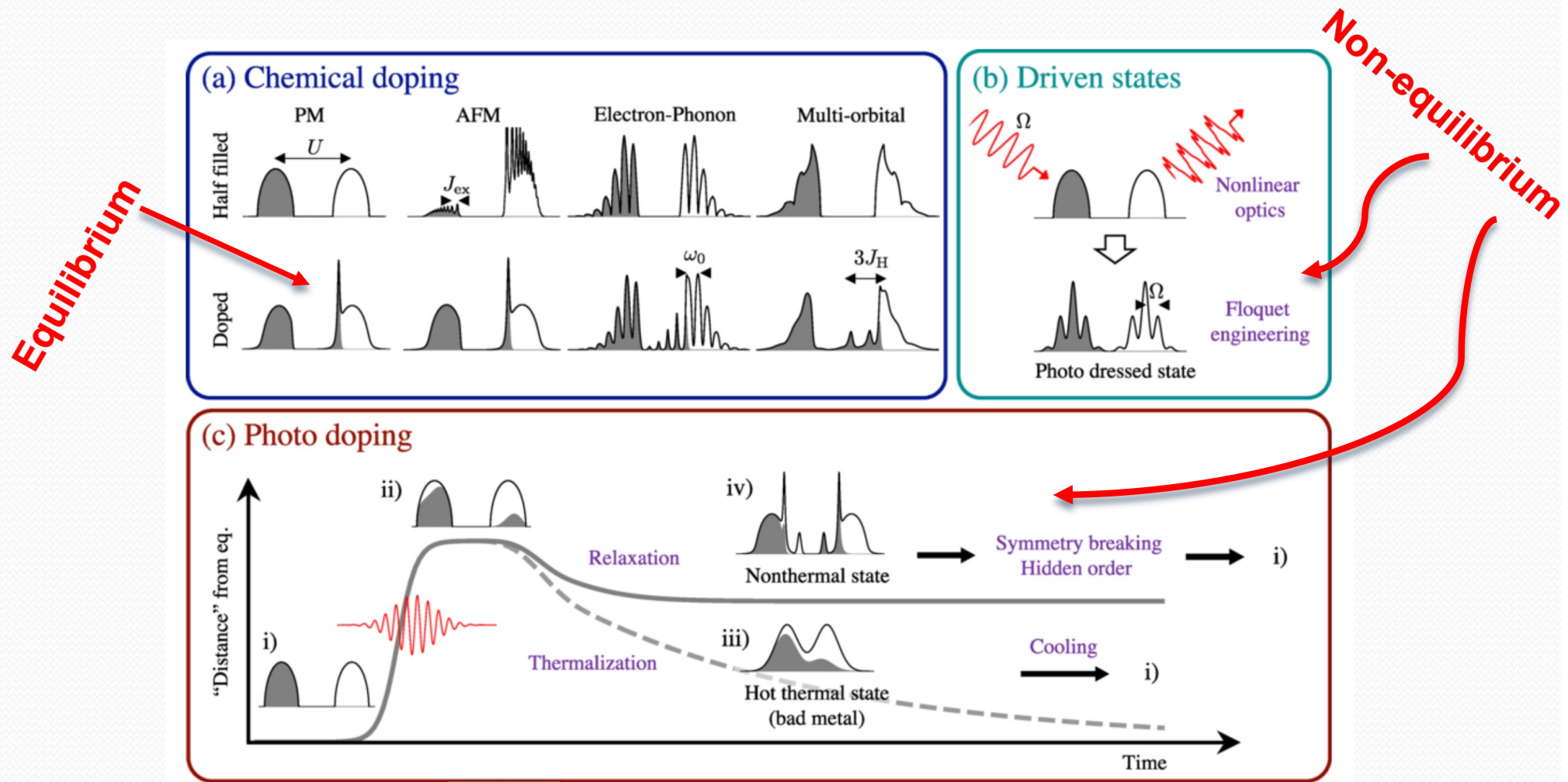
Summary

- We have presented a theory of metastable phases for competing orders.
- We have shown how to generate pseudomagnetic fields with spatially inhomogeneous light.
- We have shown how light can be used for enantiomer selectivity in the chiral materials NbOX_2 ($\text{X}=\text{Cl}, \text{Br}, \text{I}$).
- An exciting frontier is to use AI to develop drive protocols for desired quantum states.

Wonderful Collaborators—Thank you!

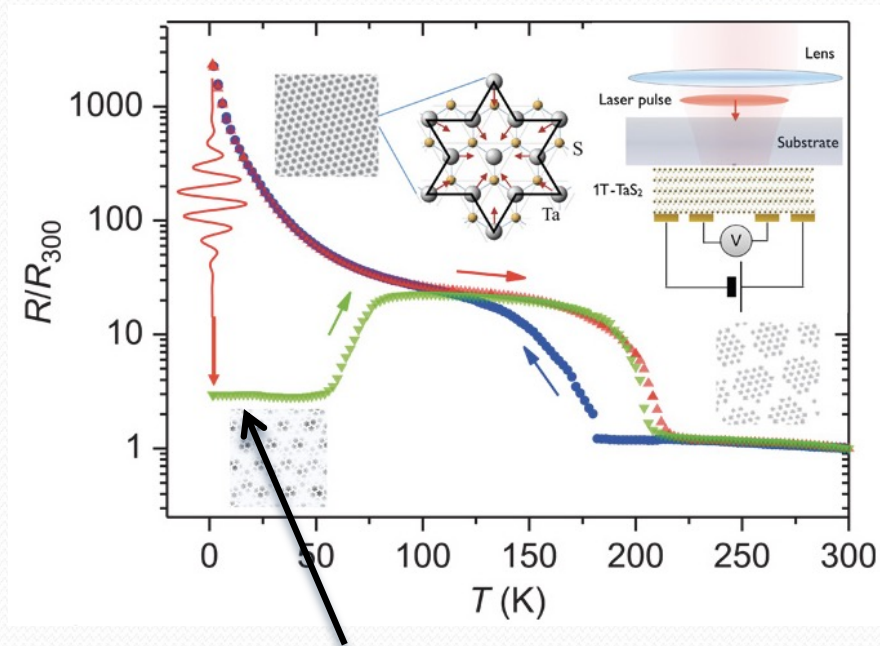


What is the light doing microscopically?

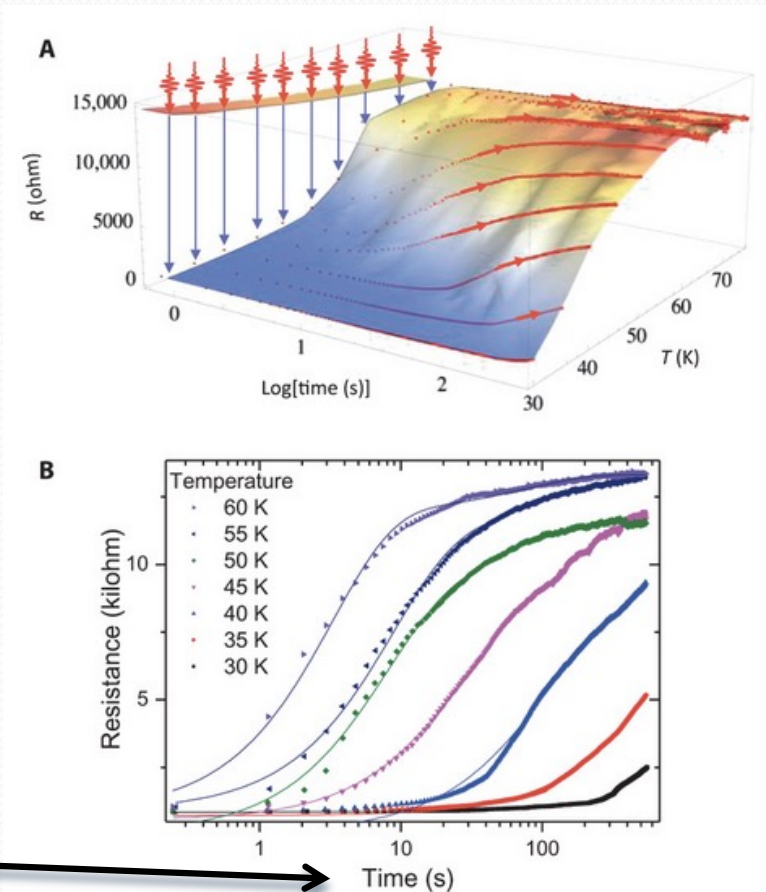


Y. Murakami, D. Golez, M. Eckstein, and P. Werner, *RMP* **97**, 035001 (2025).

Experimental Realization of Metastable States: 1T-TaS₂

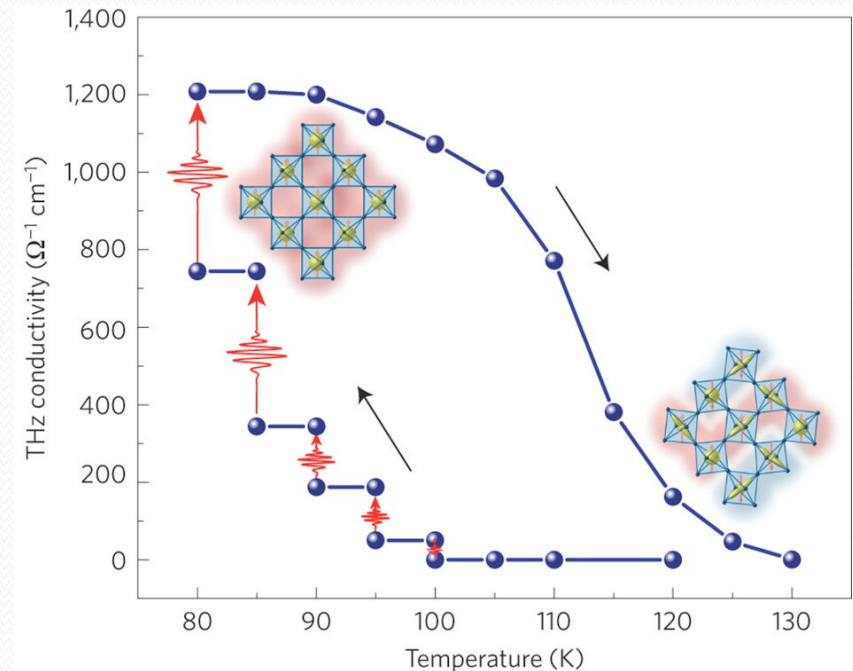
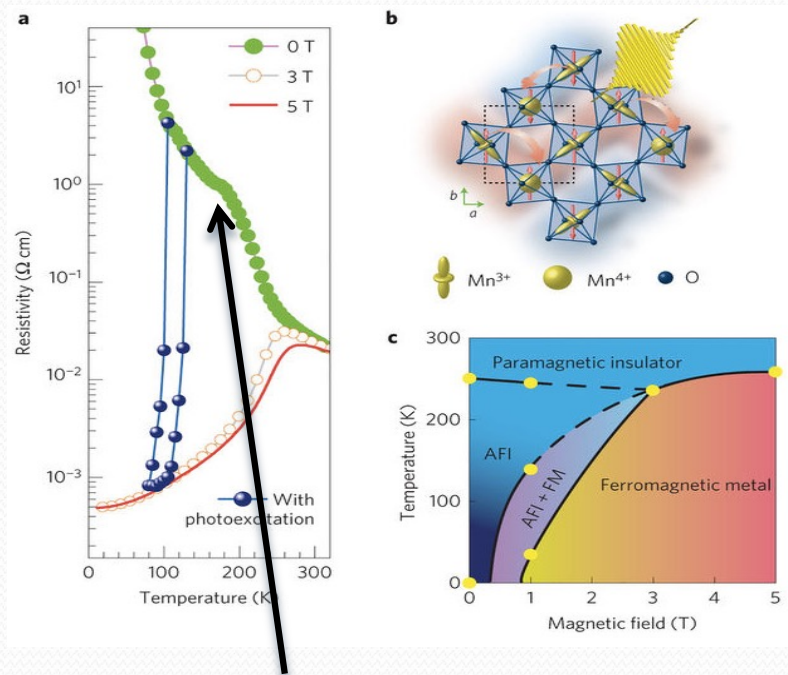


Single 35 fs pulse drives system to a “hidden” metallic CDW state that survives for 100s of seconds.



I. Vaskivskyi, .., D. Mihailovic, *Science Advances* **1**, 1500168 (2015)

Experimental Realization of Metastable States: $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$

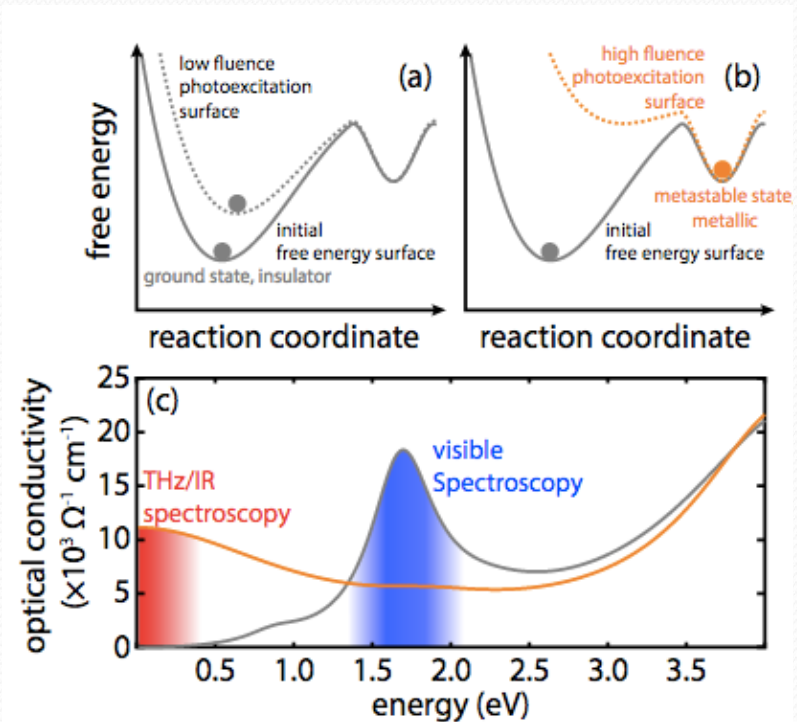
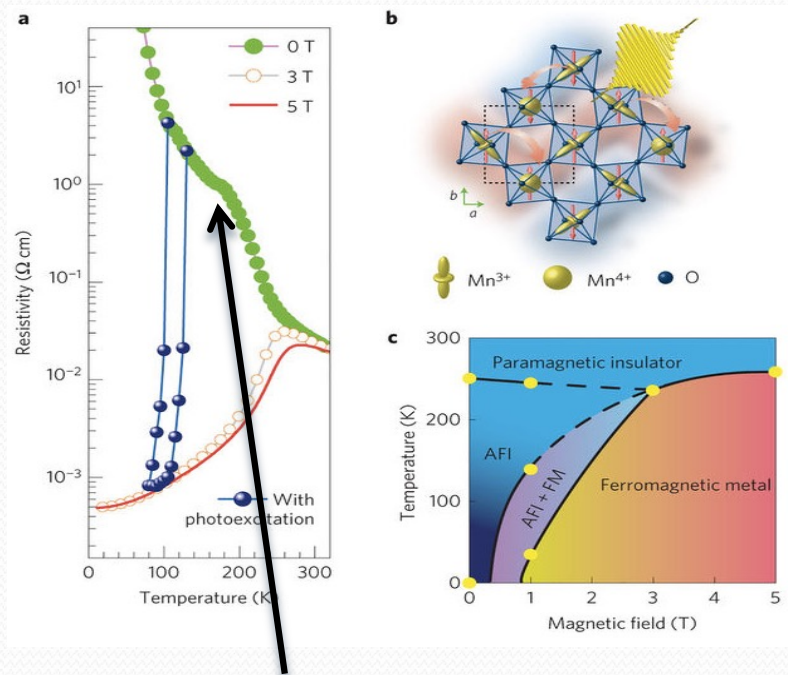


Single 35 fs pulse drives system to a “hidden” metallic state **not thermally accessible** in a strain-induced AFI state. **Metallic state stable as long as low-T maintained.**

J. Zhang, .., R. Averitt, *Nature Materials* **15**, 956 (2016)

S. W. Teitelbaum, ..., R. Averitt, K. A. Nelson, *Phys. Rev. Lett.* **123**, 267201 (2019)

Experimental Realization of Metastable States: $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$

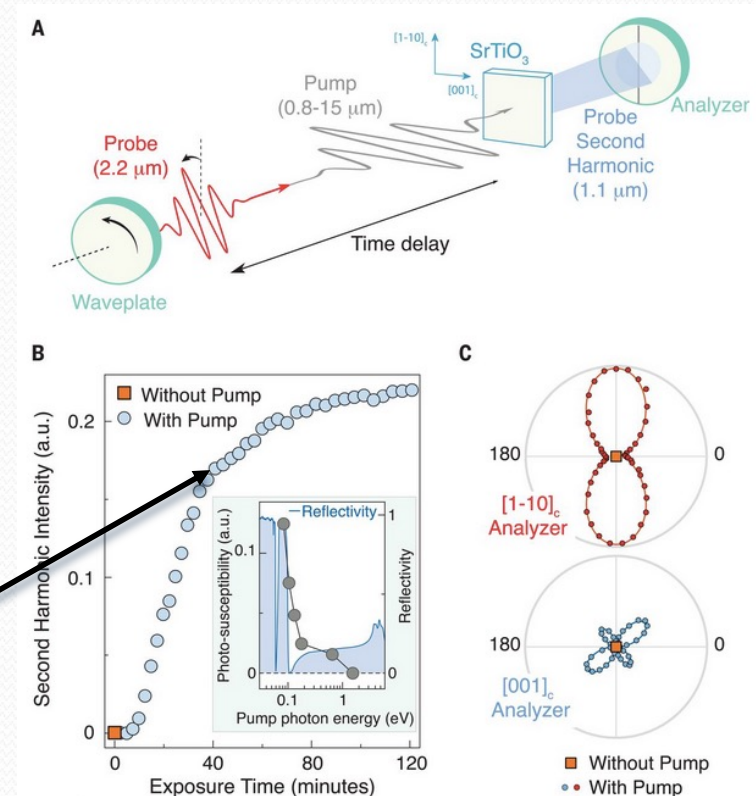
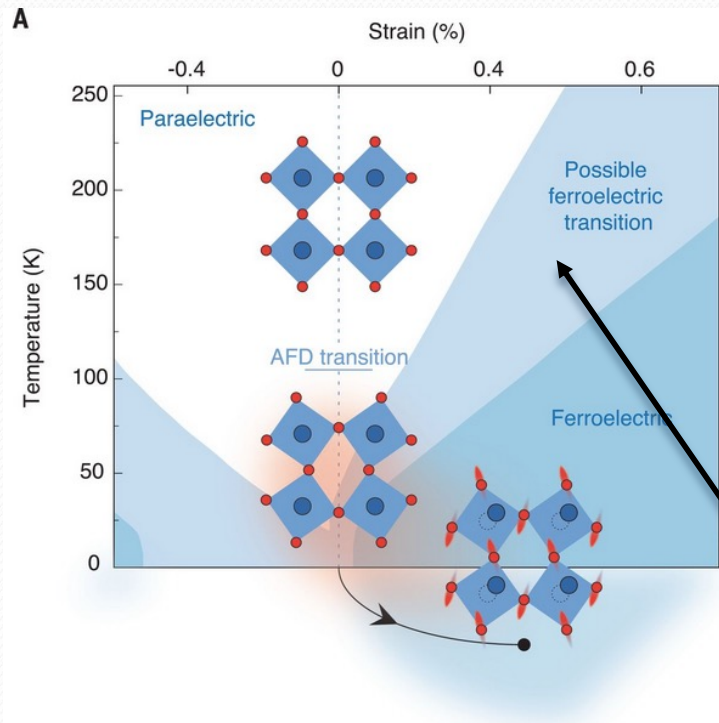


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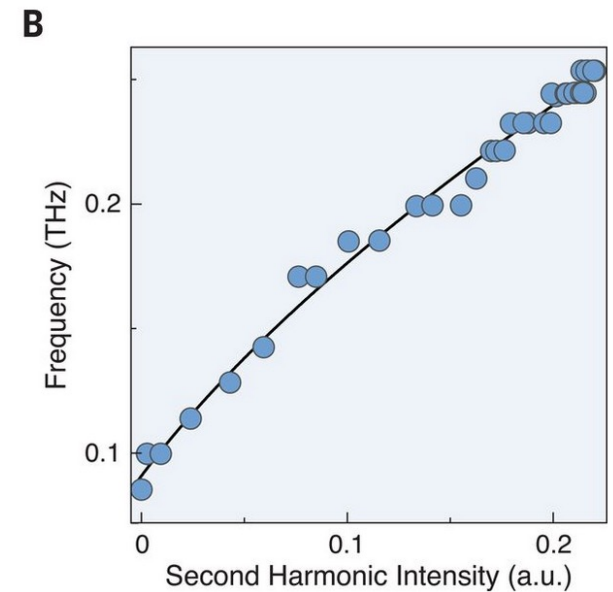
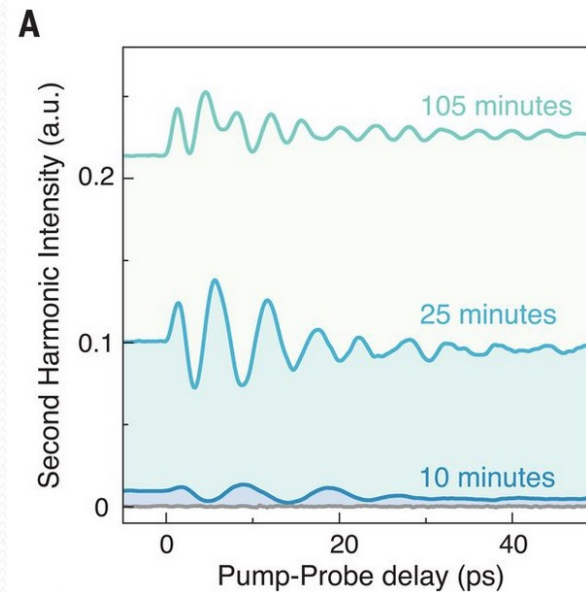
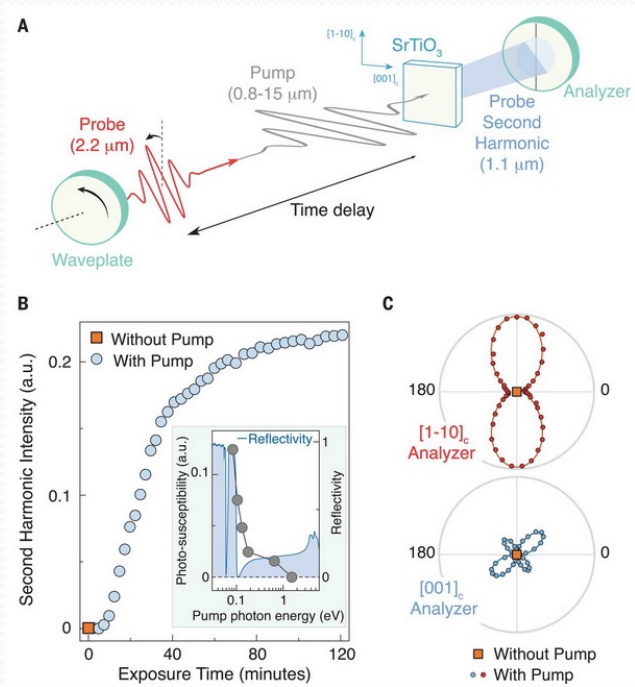
Experimental Realization of Metastable Ferroelectricity: SrTiO_3



Exposure time-dependent “hidden” ferroelectric phase **not thermally accessible in some cases**.
Ferroelectric phase visible in second harmonic.

T. F. Nova, A. S. Disa, M. Fechner, and A. Cavalleri, *Science* **364**, 1075 (2019)

Experimental Realization of Metastable Ferroelectricity: SrTiO_3

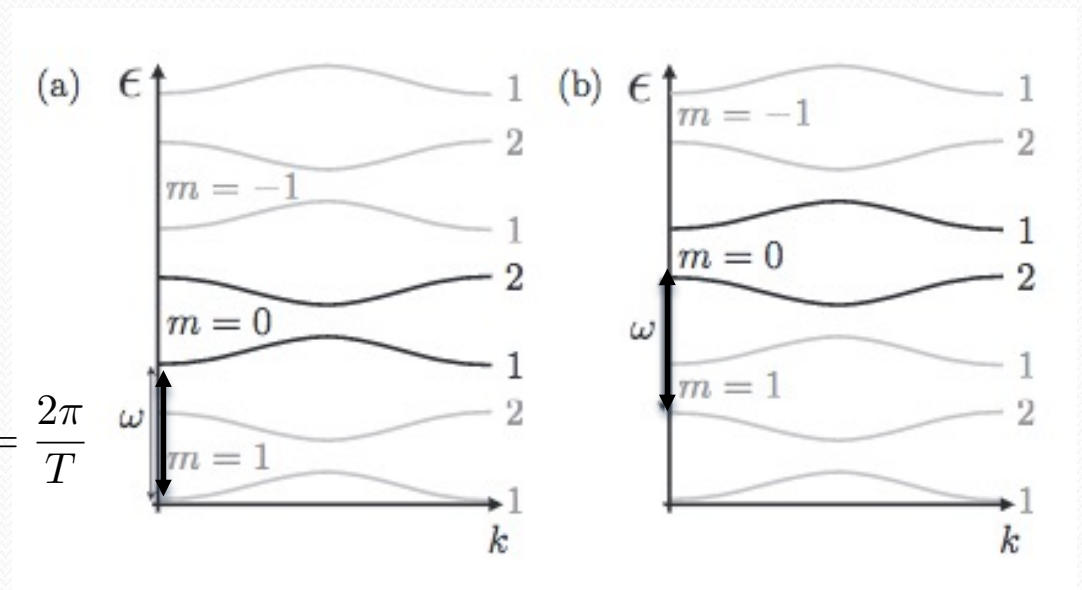
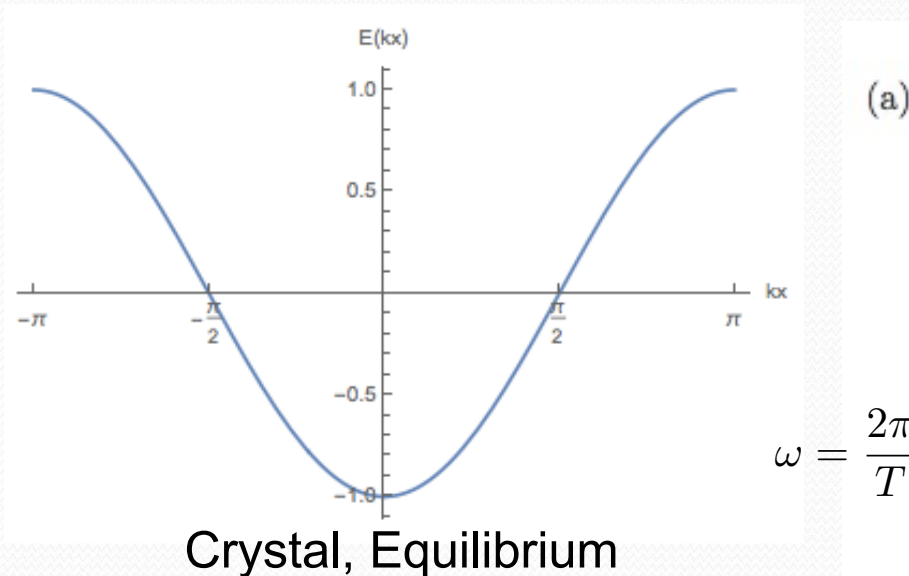


Ferroelectric phase visible in second harmonic and lasts for hours!

T. F. Nova, A. S. Disa, M. Fechner, and A. Cavalleri, *Science* **364**, 1075 (2019)

Floquet systems

- Hamiltonian is periodic in time: $\mathcal{H}(t + T) = \mathcal{H}(t)$
- Time periodicity leads to a “quasi-energy” zone, analogous to the Brillouin zone in momentum space.



Review: Bukov, D'Alessio, Polkovnikov
Adv. Phys.(2015)

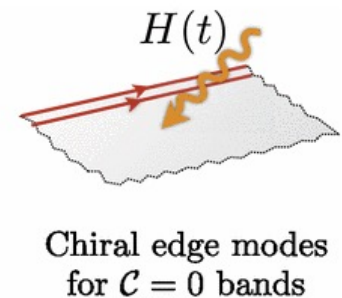
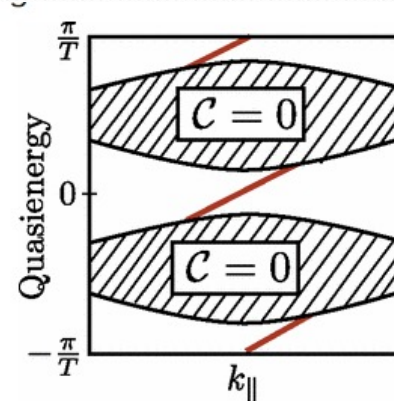
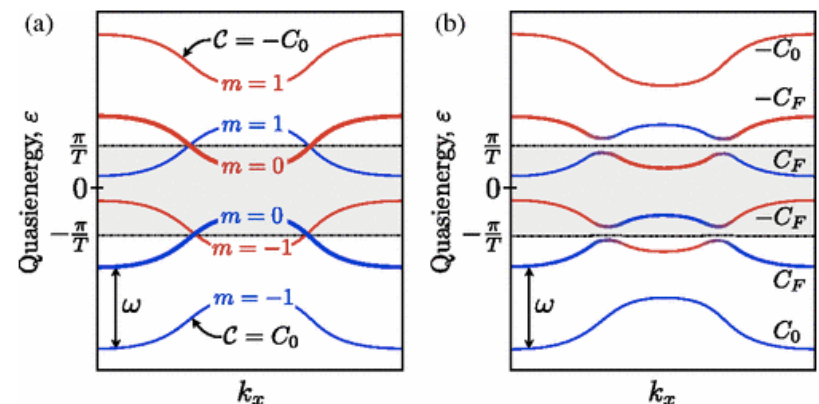
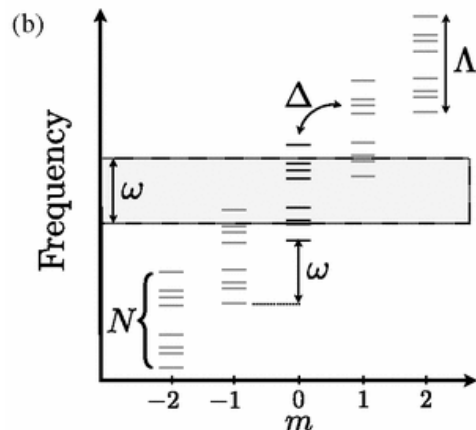
Which band is “lower” in energy?
Iadecola, Neupert, Chamon *PRB* (2015)

Topological Transitions in Floquet systems

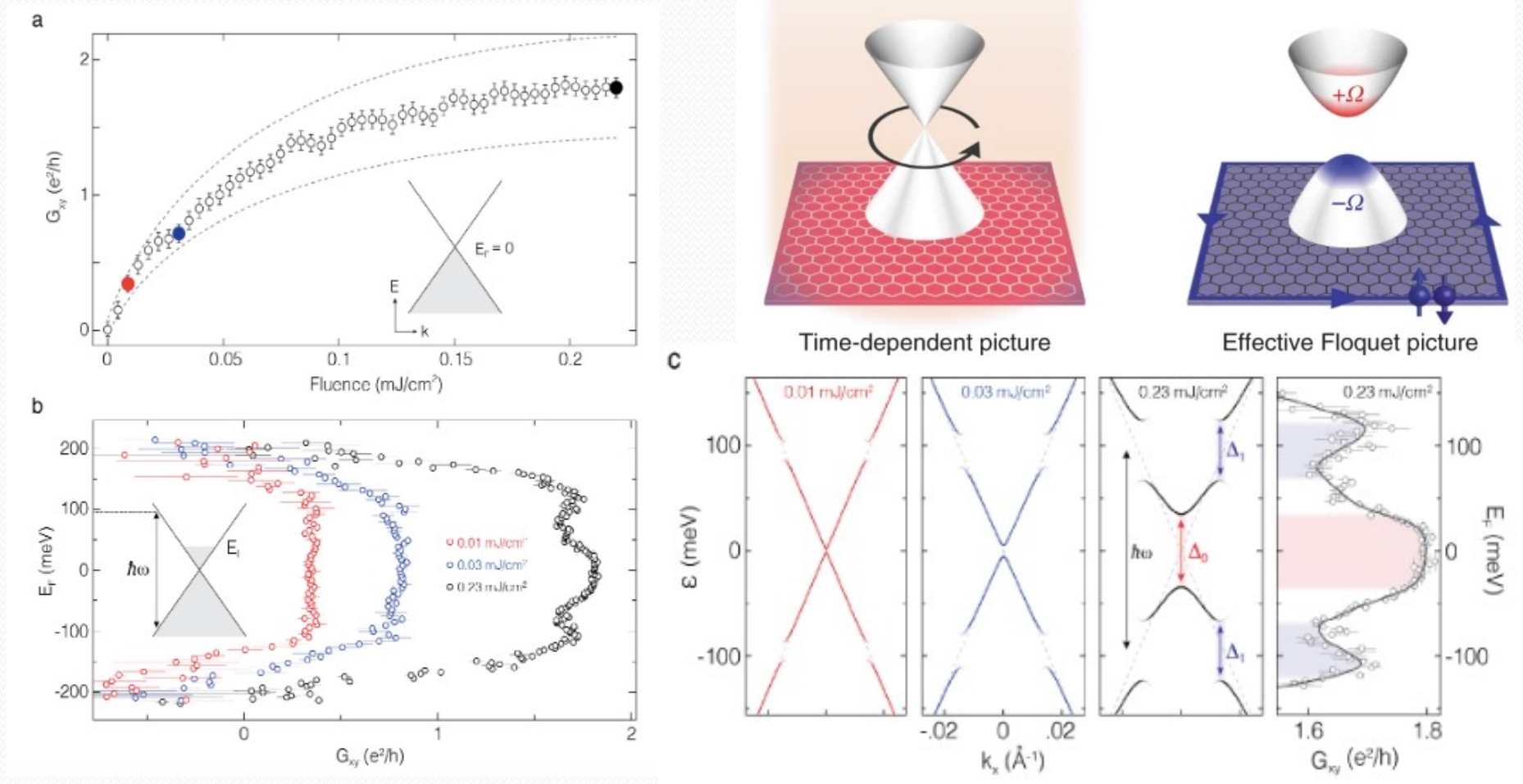
- Band inversions from drive, similar to spin-orbit coupling induced band inversions.

(a)

$$\mathcal{H}(\mathbf{k}) = \begin{pmatrix} \dots & 1 & m'_0 & -1 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ H_0 + \omega & \Delta & & & \\ \Delta^\dagger & H_0 & \Delta & & \\ & & \Delta^\dagger & H_0 - \omega & \\ & & & & \vdots \\ & & & & 1 \\ & & & & 0 \\ & & & & m \\ & & & & -1 \\ & & & & \vdots \end{pmatrix}$$



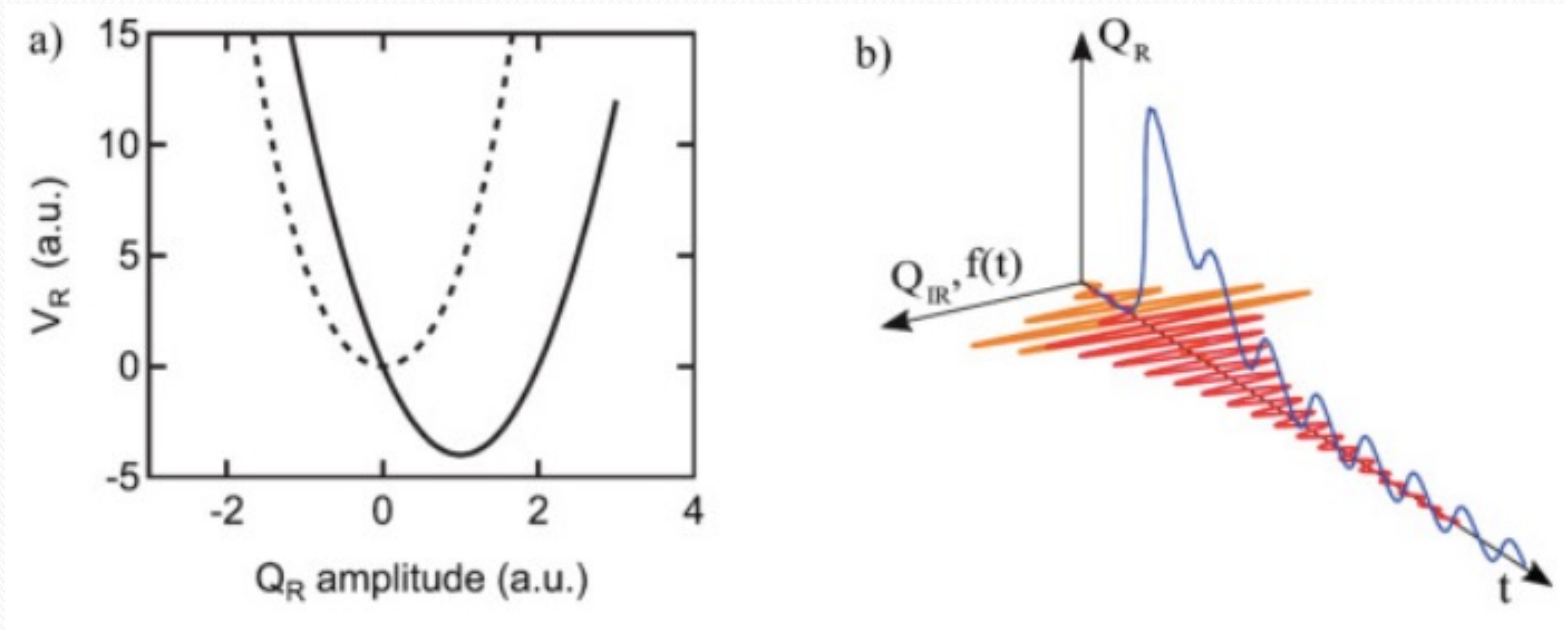
Light induced anomalous Hall effect in graphene



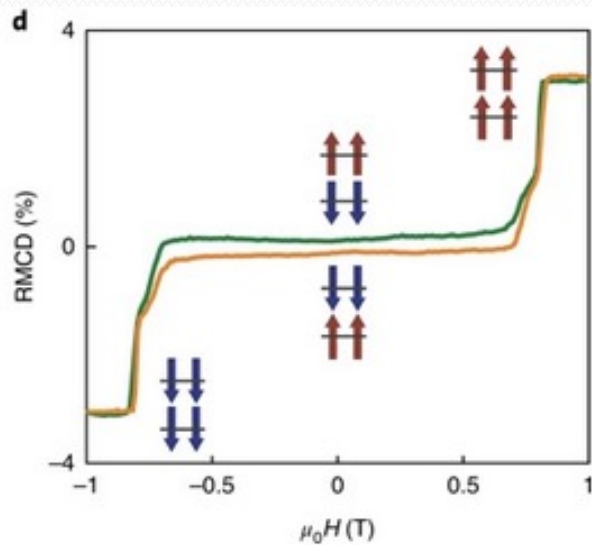
J. W. McIver, B. Schulte, F.-U. Stein, T. Matsuyama, G. Jotzu, G. Meier and A. Cavalleri, *Nature Physics* **16**, 38 (2020).

Non-linear phononics

- Selectively excite **infrared active modes** to create transient lattice distortion through nonlinearly coupled **Raman modes** $\rightarrow$ temporarily shifts equilibrium ion positions which modifies electronic properties.

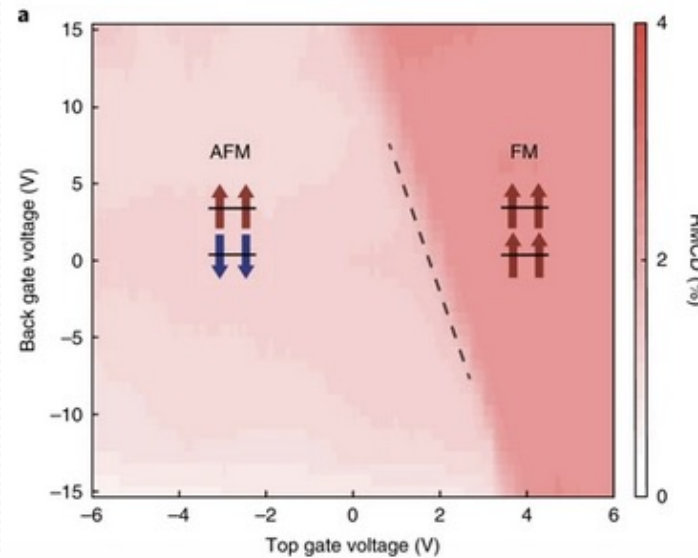


Experimental results for CrI_3



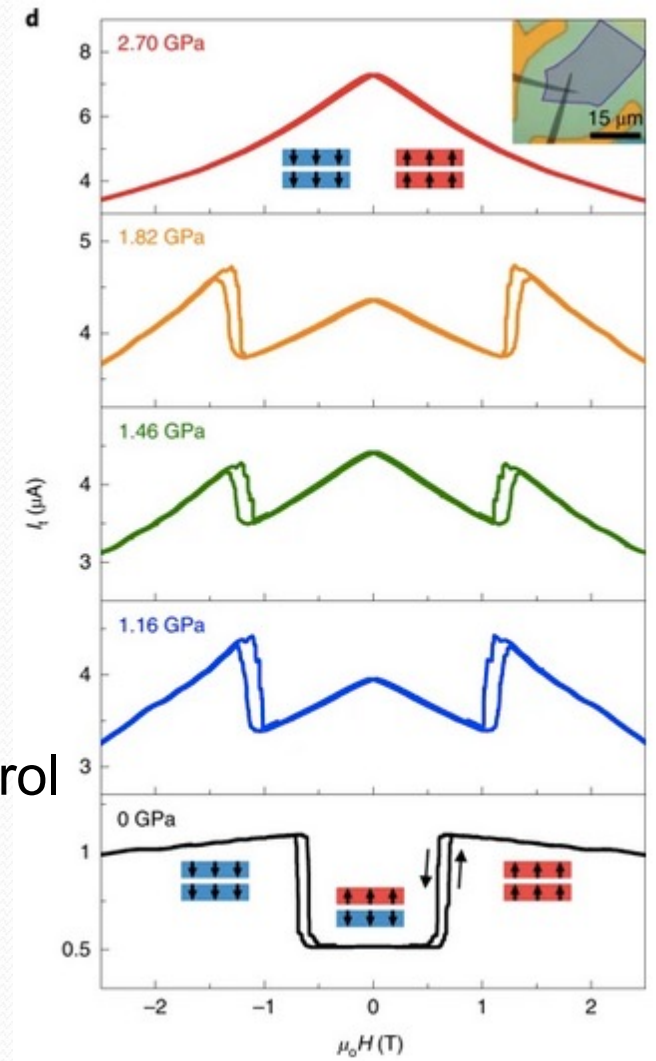
Magnetic field control
of magnetism

Bevin Huang, ..., Xiaodong Xu,
Nat. Nano. **13**, 544 (2018)



Electric field control
of magnetism

Pressure control
of magnetism

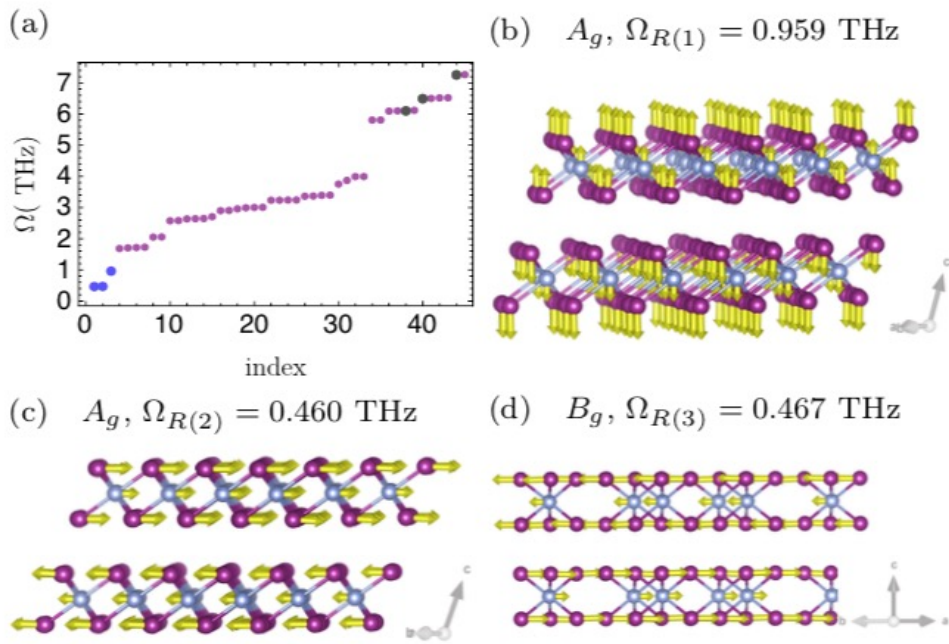


- Can we create an analogous response with a laser drive?

Tiancheng Song, ..., Xiaodong Xu, *Nat. Mat.* **18**, 1298 (2019)

Phonon mediated dimensional crossover in CrI_3

- Idea: modulate interlayer exchange coupling via nonlinear phonon effects.
- 2D interlayer AF, bulk interlayer FM



$$V[Q_{\text{IR}}, Q_{\text{R}(i)}] = \frac{1}{2}\Omega_{\text{IR}}^2 Q_{\text{IR}}^2 + \sum_{i=1}^3 \frac{1}{2}\Omega_{\text{R}(i)}^2 Q_{\text{R}(i)}^2$$

$$+ \sum_{i=1}^2 \frac{\beta_i}{3} Q_{\text{R}(i)}^3 + Q_{\text{IR}}^2 \sum_{i=1}^2 \gamma_i Q_{\text{R}(i)} + \delta Q_{\text{R}(1)}^2 Q_{\text{R}(2)}$$

$$+ \epsilon Q_{\text{R}(1)} Q_{\text{R}(2)}^2 + Q_{\text{R}(3)}^2 \sum_{i=1}^2 \zeta_i Q_{\text{R}(i)}.$$

$$Q_{\text{R}}(t) = \frac{\gamma\pi(Z^*E_0\tau^3)^2}{(4\Omega_{\text{IR}}^2 - \Omega_{\text{R}}^2)} \frac{\Omega_{\text{IR}}^2}{\Omega_{\text{R}}^2} (\Omega_{\text{R}}^2 \cos(2\Omega_{\text{IR}}t) +$$

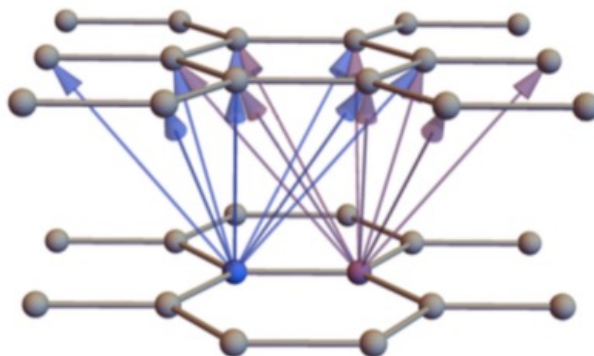
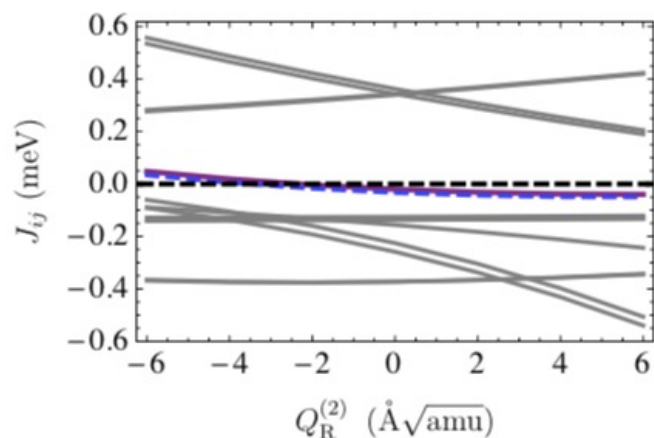
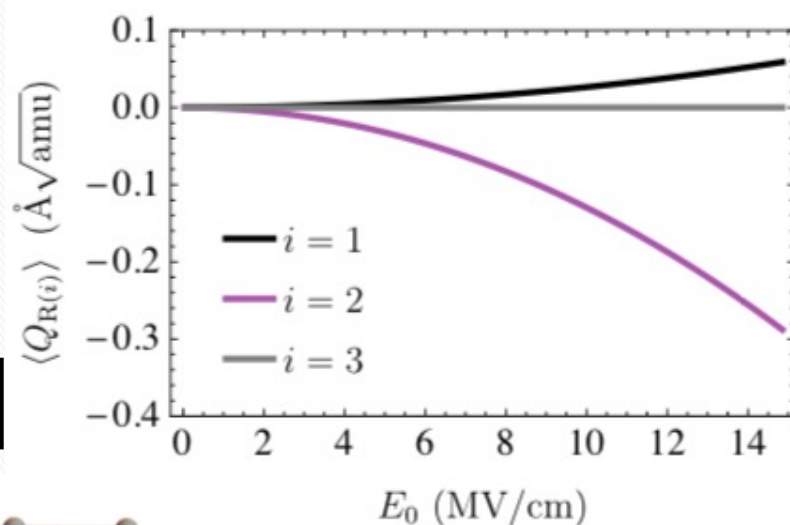
$$2(\Omega_{\text{IR}}^2 - \Omega_{\text{R}}^2) \cos(\Omega_{\text{R}}t) + \Omega_{\text{R}}^2 - 4\Omega_{\text{IR}}^2).$$

Phonon mediated dimensional crossover in CrI_3

- Idea: modulate interlayer exchange coupling via nonlinear phonon effects.
- 2D interlayer AF, bulk FM

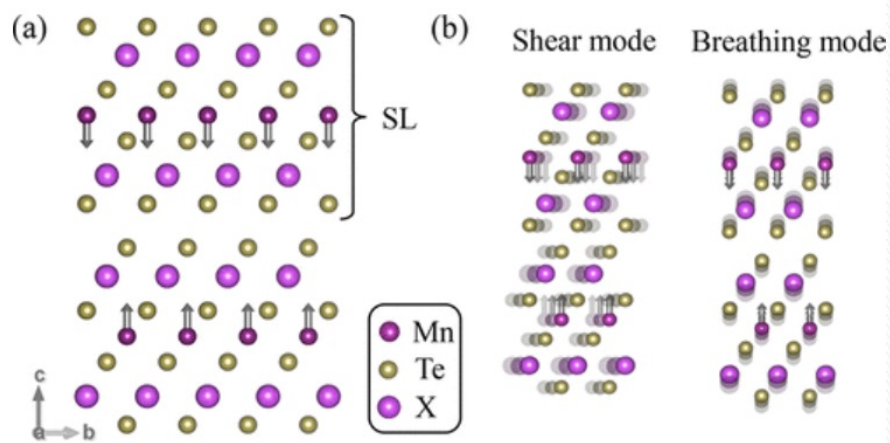
$$\mathcal{H}_{\text{intra}} = \sum_{\langle ij \rangle \in \lambda\mu(\nu)} \mathcal{J} \mathbf{s}_i \cdot \mathbf{s}_j + K s_i^\nu s_j^\nu + \Gamma (s_i^\lambda s_j^\mu + s_i^\mu s_j^\lambda)$$

$$\mathcal{H}_{\text{inter}} = \frac{1}{2} \sum_{ij \in \text{int.}} J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j \quad \boxed{J^{\text{eff}} = J^0 + \delta J \hat{\delta} \cdot \langle \mathbf{u}_R \rangle}$$



Change sign of interlayer
Coupling: AFM $\rightarrow$ FM

Phonon mediated magnetic transition and topological band transition in MnX_2Te_4 , $\text{X}=\text{Bi, Sb}$ bilayers $T_c \sim 20$



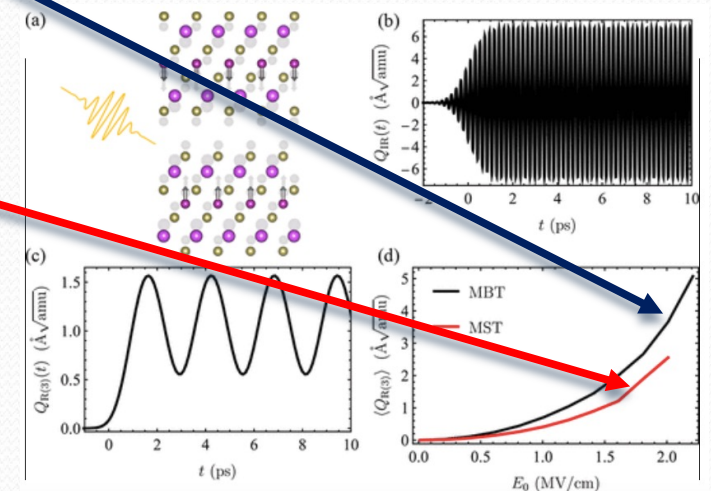
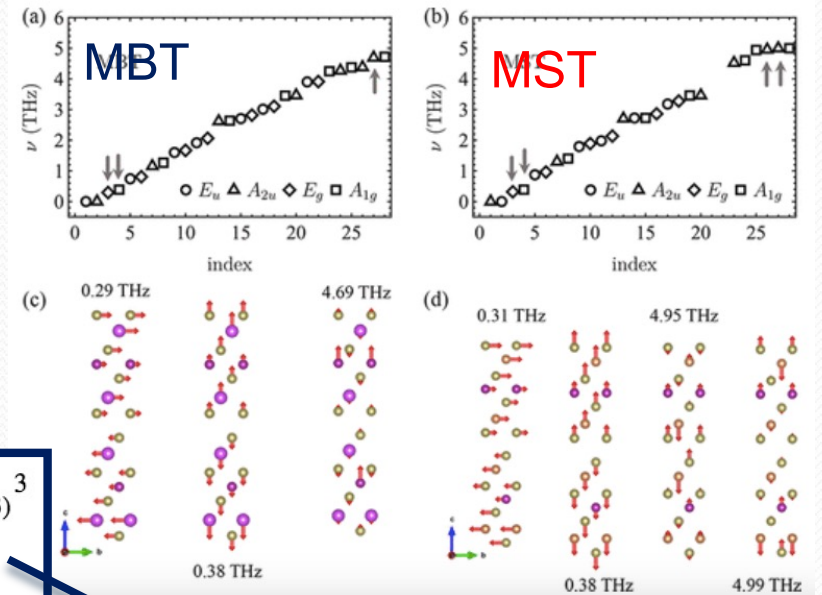
$$V[Q_{\text{IR}}, Q_{\text{R}(3)}, t] = \frac{1}{2} \Omega_{\text{IR}}^2 Q_{\text{IR}}^2 + \frac{1}{2} \Omega_{\text{R}(3)}^2 Q_{\text{R}(3)}^2 + \gamma_3 Q_{\text{IR}}^2 Q_{\text{R}(3)} + \frac{1}{3} \beta_3 Q_{\text{R}(3)}^3 + \mathbf{Z} \cdot \mathbf{E}_0 \sin(\Omega t) F(t) Q_{\text{IR}}$$

MBT

$$V[\{Q_{\text{IR}(i)}\}, Q_{\text{R}(3)}, t] = \sum_{i=1}^3 \frac{1}{2} \Omega_{\text{IR}(i)}^2 Q_{\text{IR}(i)}^2 + \frac{1}{2} \Omega_{\text{R}(3)}^2 Q_{\text{R}(3)}^2 + \gamma_{1,3} Q_{\text{IR}(1)}^2 Q_{\text{R}(3)} + \gamma_{2,3} Q_{\text{IR}(2)}^2 Q_{\text{R}(3)} + \gamma_{1,2,3} Q_{\text{IR}(1)} Q_{\text{IR}(2)} Q_{\text{R}(3)} + \frac{1}{3} \beta_3 Q_{\text{R}(3)}^3 + [\mathbf{Z}_1^* Q_{\text{IR}(1)} + \mathbf{Z}_2^* Q_{\text{IR}(2)}] \cdot \mathbf{E}_0 \sin(\Omega t) F(t)$$

MST

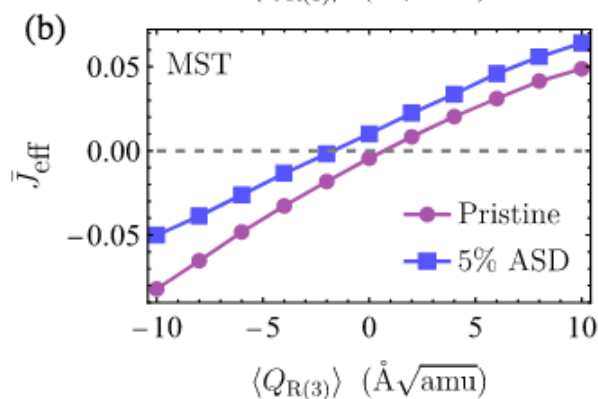
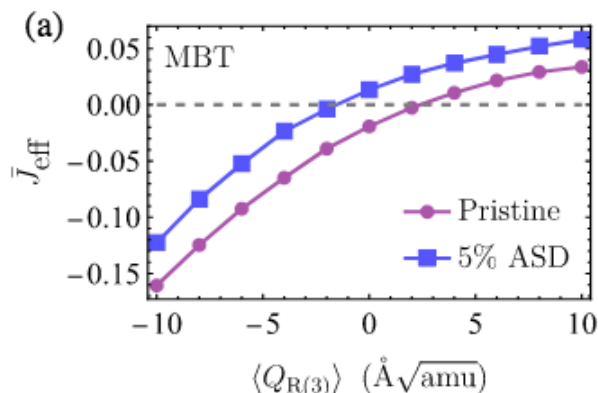
M. Rodriguez-Vega, Z. Lin, A Leonardo, A. Ernst, M. G. Vergniory, and GAF, JPCL (2022).



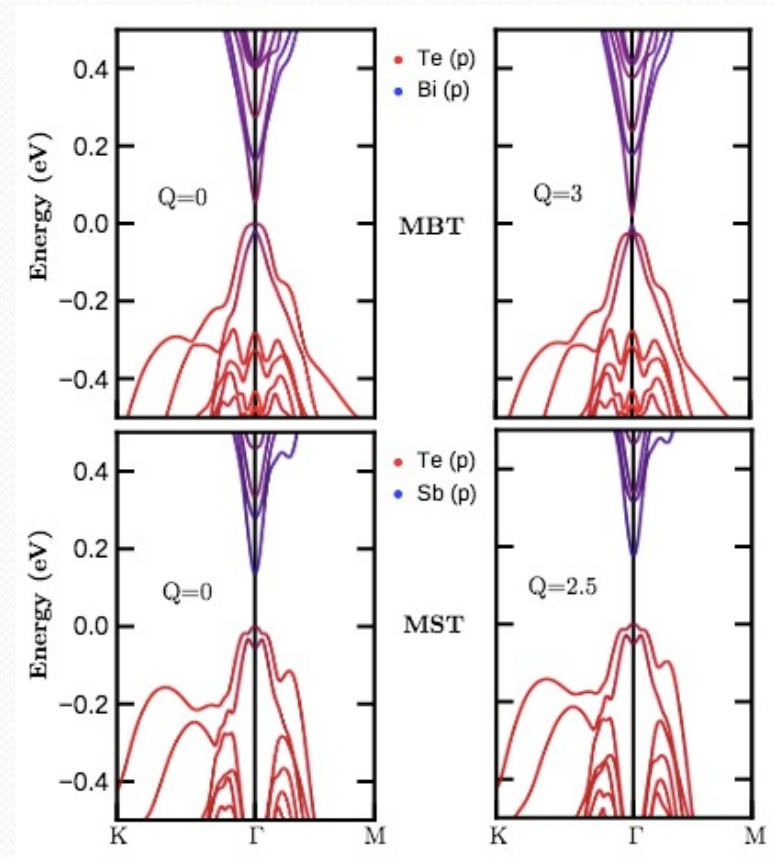
Phonon mediated magnetic transition and topological band transition in MnX_2Te_4 , $\text{X}=\text{Bi, Sb}$ bilayers

$$\mathcal{H} = \mathcal{H}_{\text{intra}} + \mathcal{H}_{\text{inter}} \quad \mathcal{H}_{\text{intra}} = -\sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_i (S_i^z)^2 \quad \mathcal{H}_{\text{inter}} = -J_c \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$$J^{\text{eff}} = J^0 + \delta J \hat{\delta} \cdot \langle \mathbf{u}_R \rangle$$

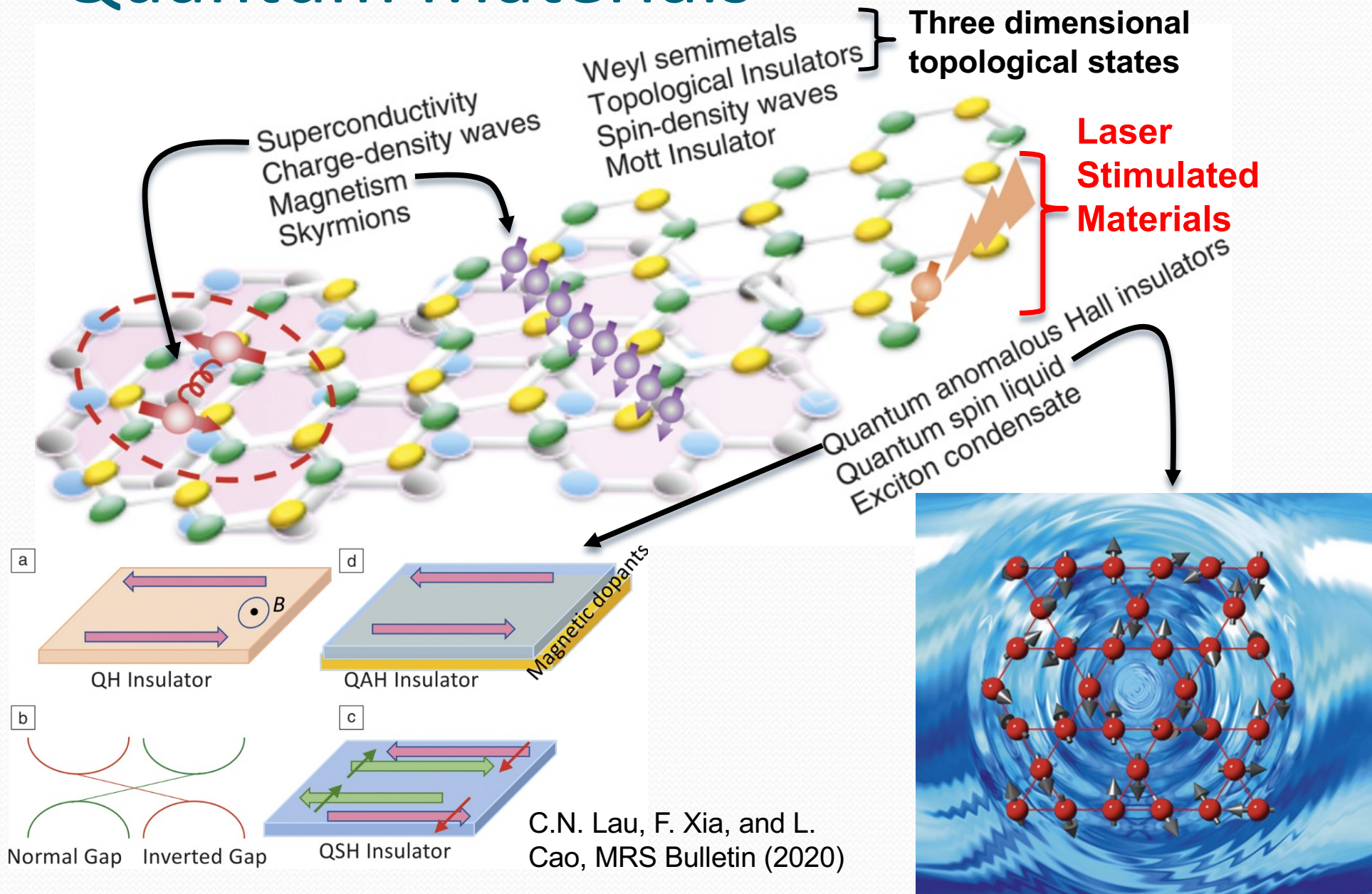


$T_c \sim 20\text{K}$



M. Rodriguez-Vega, Z. Lin, A Leonardo, A. Ernst, M. G. Vergniory, and GAF, JPCL (2022).

Quantum Materials

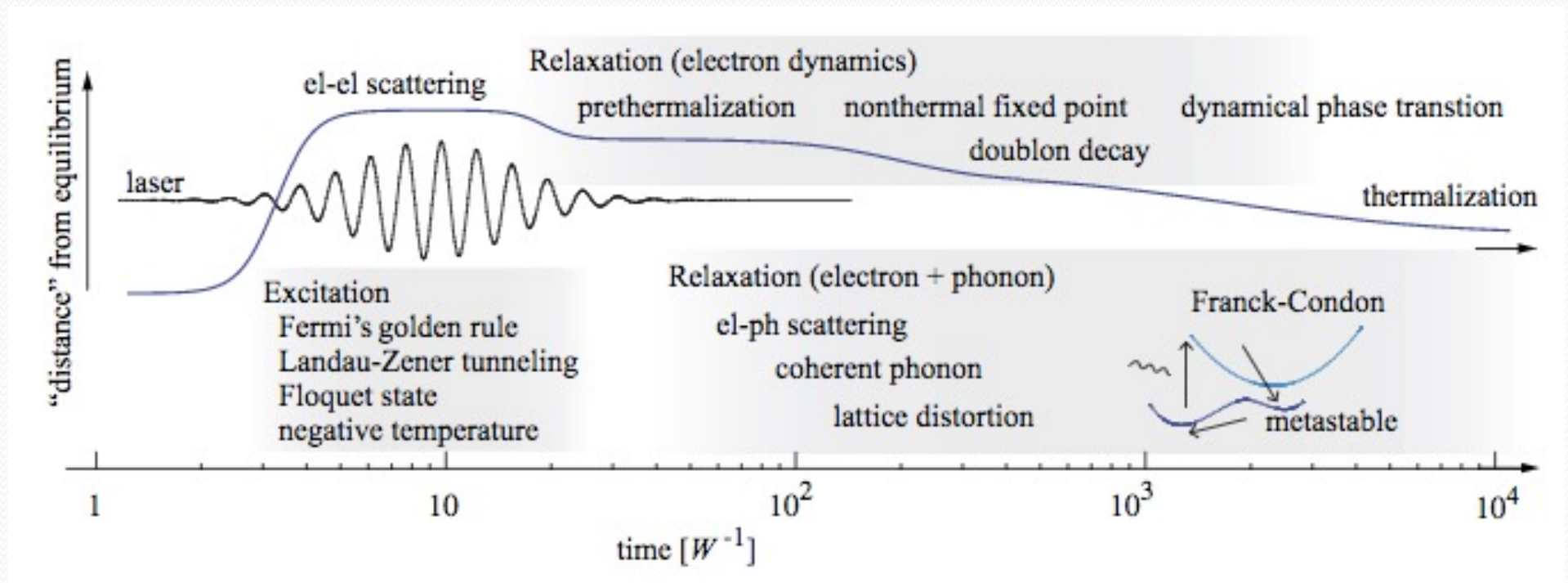




Laser-stimulated systems (periodically driven systems)

- Chief goal is to access states not achievable in equilibrium: “**Hidden Phases/States**”
- Two limits:
 - Short pulse limit (tip state into some metastable regime)
 - Long pulse limit (Floquet, time-periodic Hamiltonian)
- Focus on “pre-heating” regime in later part of this talk.
 - Negligible energy absorbed from time-dependent Hamiltonian (Abanin, De Roeck, Ho, Huvaneers, 2015).

Time scales in the solid state



Aoki, Tsuji, Eckstein, Kollar, Oka, and Werner, *RMP* **86**, 779 (2014)