Optical control of topological memory in Chern insulators realized in moiré multilayers

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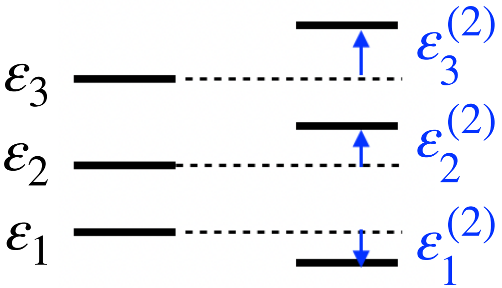
- Energy shift in atoms and solids in an oscillating electric field: the dynamical Stark shift
- Application to transition-metal dichalcogenides
- Optical control of the sign of magnetization in a Chern insulator by circularly polarized light, *PRB* **105**, 064423 (2022) Erratum *PRB* **108**, 059904(E) (2023)
- Optical control of the order parameters in other materials

Dynamical Stark shift in atoms and solids

Energy levels of an atom



An oscillating electric field $\mathbf{E}(t) = \frac{1}{2}\left[\mathbf{E}(\omega)\, e^{-i\omega t} + \mathbf{E}^*(\omega)\, e^{i\omega t}\right]$,

$H' = -e\,\mathbf{r}\cdot\mathbf{E}(t)$, induces a dipole $d_n^{\alpha}(\omega) = \langle e r^{\alpha}\rangle_n = \chi_n^{\alpha\beta}(\omega)\, E_\beta(\omega)$

and the Stark shift of the n-th energy level to the 2nd order

$$\varepsilon_n^{(2)} = -\frac{1}{4}\chi_n^{\alpha\beta}(\omega)\, E_\alpha^*(\omega)\, E_\beta(\omega)$$

The electric polarizability tensor $\chi^{\alpha\beta}(\omega) = \chi_s^{\alpha\beta}(\omega) + \chi_a^{\alpha\beta}(\omega)$ (symmetric + antisymmetric)

In a crystal, Bloch wavefunctions $\psi_{n,\mathbf{k}} = e^{i\mathbf{kr}}\, u_{n,\mathbf{k}}(\mathbf{r})$ have energies $\varepsilon_n(\mathbf{k})$, denote $\varepsilon_{nm}(\mathbf{k}) = \varepsilon_n(\mathbf{k}) - \varepsilon_m(\mathbf{k})$

Shift of the energy bands in an oscillating electric field (Pershoguba-Yakovenko 2022):

$$\varepsilon_n^{(2)}(\mathbf{k}) = \frac{e^2}{4(\hbar\omega)^2}\frac{\partial^2\varepsilon_n(\mathbf{k})}{\partial k_\alpha \partial k_\beta}\,\mathrm{Re}\left[E_\alpha^*(\omega)E_\beta(\omega)\right] - \frac{e^2\Omega_{n,\gamma}(\mathbf{k})}{4\,\hbar\omega}\epsilon^{\alpha\beta\gamma}\,\mathrm{Im}\left[E_\alpha^*(\omega)E_\beta(\omega)\right]$$

intraband symmetric — intraband antisymmetric

$$-\frac{e^2}{4}\mathrm{Re}\sum_{m\neq n}\frac{r_{nm}^{\alpha}(\mathbf{k})r_{mn}^{\beta}(\mathbf{k})}{\varepsilon_{mn}(\mathbf{k}) - \hbar\omega}E_\alpha^*(\omega)E_\beta(\omega) - \frac{e^2}{4}\mathrm{Re}\sum_{m\neq n}\frac{\left[r_{nm}^{\alpha}(\mathbf{k})r_{mn}^{\beta}(\mathbf{k})\right]^*}{\varepsilon_{mn}(\mathbf{k}) + \hbar\omega}E_\alpha^*(\omega)E_\beta(\omega)$$

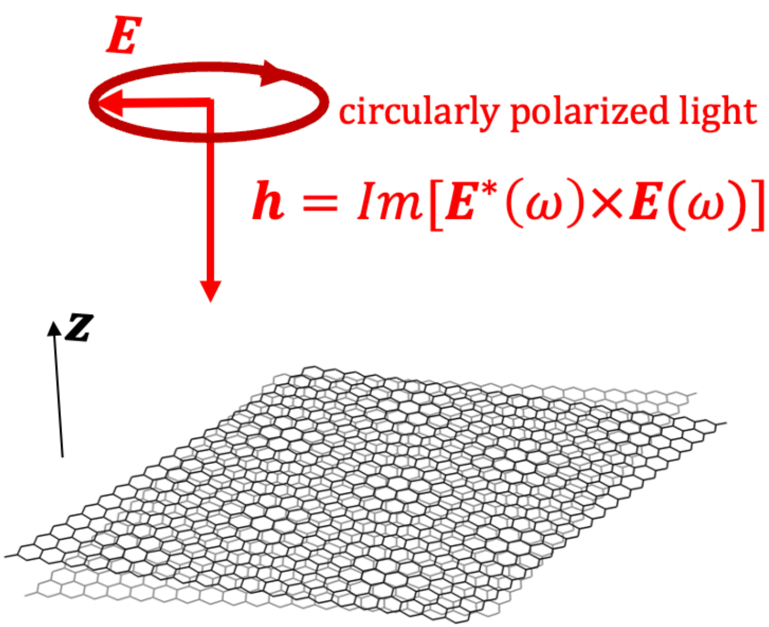
interband Stark shift — interband Bloch-Siegert shift

Berry connection $\mathbf{r}_{nm}(\mathbf{k}) = \langle u_{n,\mathbf{k}}\,|\, i\frac{\partial}{\partial\mathbf{k}}\,|\, u_{n,\mathbf{k}}\rangle$ Berry curvature $\boldsymbol{\Omega}_n(\mathbf{k}) = \frac{\partial}{\partial\mathbf{k}}\times\mathbf{r}_{nn}(\mathbf{k})$

Symmetric and antisymmetric, time-reversal even and odd terms

	Permutation of indices	Time reversal
$\mathrm{Re}\left[E_\alpha^*(\omega)E_\beta(\omega)\right] = \mathrm{Re}\left[E_\beta^*(\omega)E_\alpha(\omega)\right]$	symmetric	even
$\mathrm{Im}\left[E_\alpha^*(\omega)E_\beta(\omega)\right] = \frac{1}{2}\epsilon_{\alpha\beta\gamma}h^\gamma$	antisymmetric	odd

The helicity **h** of the incident light represents its angular momentum



Renormalized energy:

$$\tilde{\varepsilon}_n(\mathbf{k}) = \varepsilon_n(\mathbf{k}) + \varepsilon_n^{(2)}(\mathbf{k})$$

bare — second-order correction

Energy shift splits in two terms

$$\varepsilon_n^{(2)}(\mathbf{k}) = \varepsilon_n^{(s)}(\mathbf{k}) + \varepsilon_n^{(a)}(\mathbf{k})$$

symmetric — antisymmetric

The symmetric energy shift

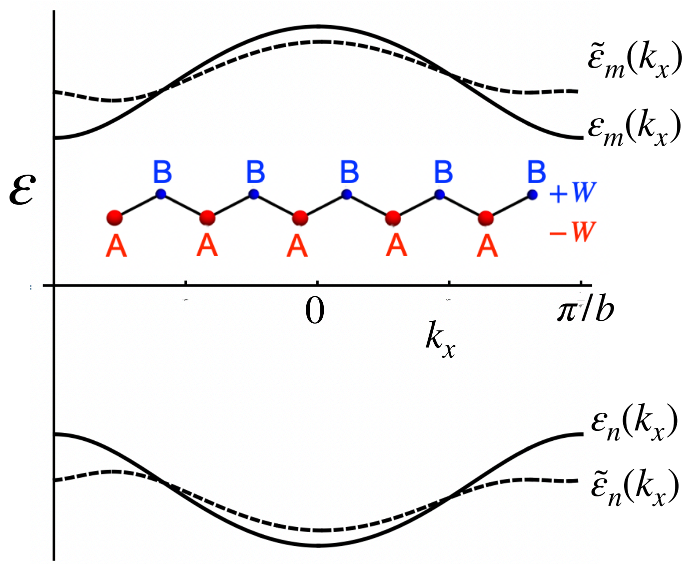
Newton's law for the electrons $\dfrac{d\tilde{\mathbf{k}}(t)}{dt} = \dfrac{e}{\hbar}\mathbf{E}(t)$

The momentum $\tilde{\mathbf{k}}(t) = \mathbf{k} + \delta\mathbf{k}(t)$ oscillates around its average value $\mathbf{k} = \langle \tilde{\mathbf{k}}(t) \rangle_t$

Expanding in small $\delta k \sim eE(\omega)/\omega$ and averaging over time

$$\langle \varepsilon_n[\mathbf{k} + \delta\mathbf{k}(t)] \rangle_t \approx \left\langle \varepsilon_n(\mathbf{k}) + \delta k_\alpha(t) \frac{\partial \varepsilon_n(\mathbf{k})}{\partial k_\alpha} + \frac{1}{2}\delta k_\alpha(t)\delta k_\beta(t) \frac{\partial^2 \varepsilon_n(\mathbf{k})}{\partial k_\alpha \partial k_\beta} \right\rangle_t = \varepsilon_n(\mathbf{k}) + e^2 \mathrm{Re}[E^*_\alpha(\omega) E_\beta(\omega)] \frac{1}{4(\hbar\omega)^2} \frac{\partial^2 \varepsilon_n(\mathbf{k})}{\partial k_\alpha \partial k_\beta}$$

The bare $\varepsilon(k_x)$ and renormalized $\tilde{\varepsilon}(k_x)$ energy dispersions for a two-band model



The full symmetric term for a two-band model

$$\varepsilon_n^{(s)}(\mathbf{k}) = e^2 \mathrm{Re}\left[E^*_\alpha(\omega) E_\beta(\omega)\right] \left\{ \frac{1}{4(\hbar\omega)^2} \frac{\partial^2 \varepsilon_n(\mathbf{k})}{\partial k_\alpha \partial k_\beta} - \frac{\varepsilon_{mn}(\mathbf{k})\,\mathrm{Re}\left[\mathbf{r}^\alpha_{\mathbf{nm}}(\mathbf{k})\mathbf{r}^\beta_{\mathbf{mn}}(\mathbf{k})\right]}{2\left[\varepsilon^2_{mn}(\mathbf{k}) - (\hbar\omega)^2\right]} \right\}$$

intraband

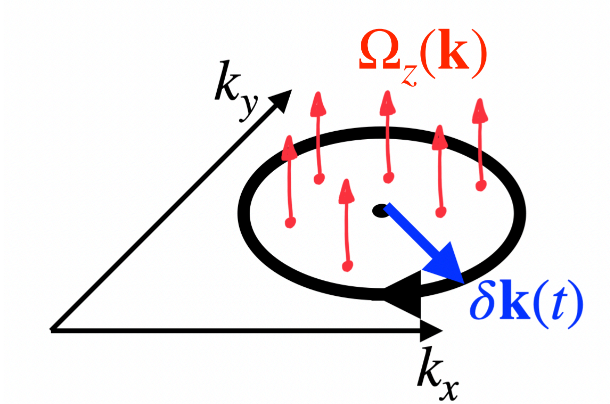
interband

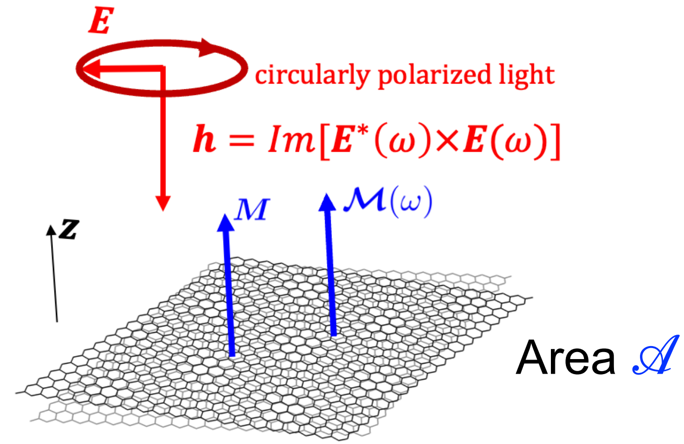
Oscillating electric field leads to flattening of energy spectrum.

Flatness controls emergence of strongly-correlated phases in moire' materials.

The antisymmetric energy shift in the presence of circularly polarized light

Circularly polarized light causes the momentum $\tilde{\mathbf{k}}(t) = \mathbf{k} + \delta\mathbf{k}(t)$ to move on a circular orbit with $|\delta\mathbf{k}| = eE/\hbar\omega$





The Berry phase accumulation over one loop per time period T

$$\phi_{loop} \approx \Omega_{n,z}(\mathbf{k})\,\pi\,(\delta k)^2 = \Omega_{n,z}(\mathbf{k})\,\pi \left(\frac{eE}{\hbar\omega}\right)^2$$

results in the energy shift

$$\varepsilon_n^{(a)}(\mathbf{k}) = \frac{\hbar\,\phi_{\text{loop}}}{T} = \frac{(eE)^2\,\Omega_{n,z}(\mathbf{k})}{2\,\hbar\omega}$$

Antisymmetric energy shift for 2-band model

$$\varepsilon_n^{(a)}(\mathbf{k}) = -\frac{\varepsilon_{mn}^2(\mathbf{k})}{\varepsilon_{mn}^2(\mathbf{k}) - (\hbar\omega)^2}\,\frac{e^2\,\mathbf{h}\cdot\mathbf{\Omega}_n(\mathbf{k})}{4\,\hbar\omega}$$

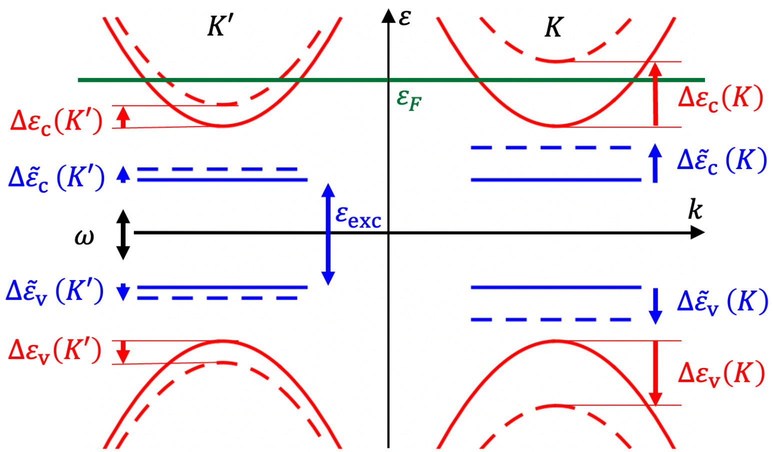
Total energy shift due to helicity ***h*** of light

$$U^{(a)} = -\frac{1}{4}\mathbf{h}\cdot\mathscr{M}(\omega), \quad \mathscr{M}_\gamma(\omega) = -\frac{1}{2}\epsilon_{\alpha\beta\gamma}\mathrm{Im}[\chi^{\alpha\beta}(\omega)]$$

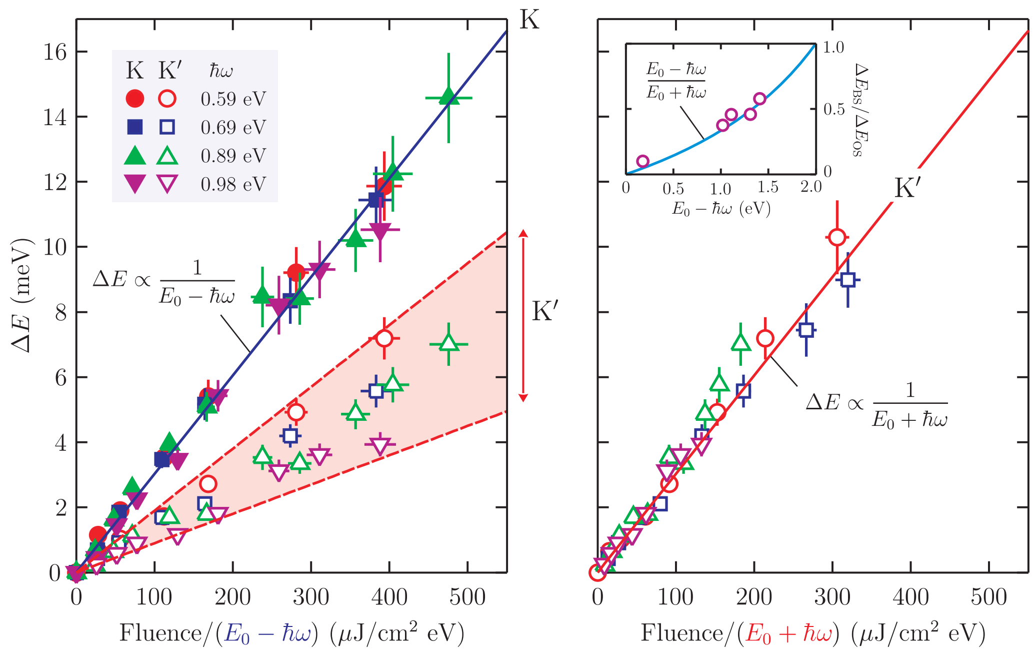
$$\mathscr{M}(\omega) = \mathscr{A}e^2 \int \frac{d^2k}{(2\pi)^2}\,\frac{\varepsilon_{mn}^2(\mathbf{k})}{\varepsilon_{mn}^2(\mathbf{k}) - (\hbar\omega)^2}\,\frac{\mathbf{\Omega}_n(\mathbf{k})}{\hbar\omega}$$

$\mathscr{M}(\omega) \neq 0$ requires (spontaneous) time-reversal symmetry breaking by orbital (ferro)magnetism

Energy shift in transition-metal dichalcogenides



Energy shifts near the massive Dirac points K and K' ($s = \pm 1$), induced by circularly polarized light ($\sigma = \pm 1$) of a subgap frequency $\omega < 2\Delta$.



Sie, …, Gedik, *Science* **355**, 1066 (2017)

Our prediction for free electrons

$$\Delta\varepsilon_{\rm c,v} = \pm \frac{1}{\omega^2} \frac{|evE_\sigma(\omega)|^2}{2\Delta - s\sigma\hbar\omega}$$

Both intra and interband terms

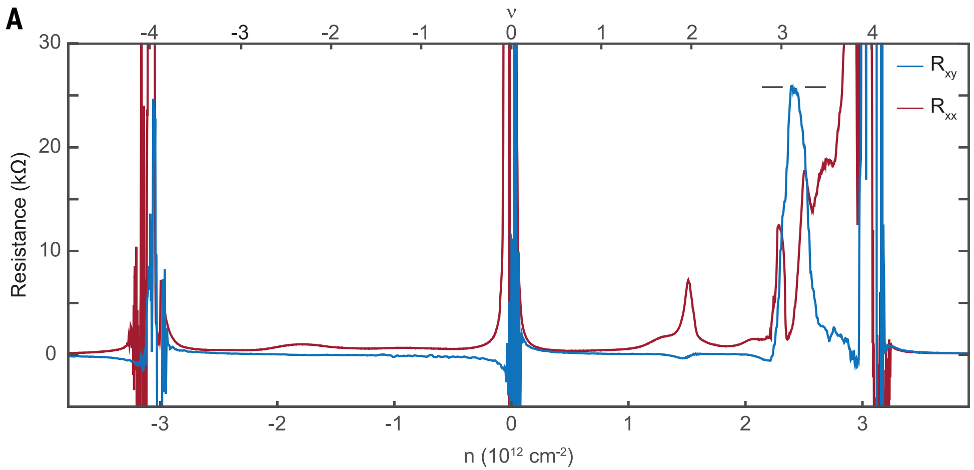
The MIT experiment for excitons in WS_2

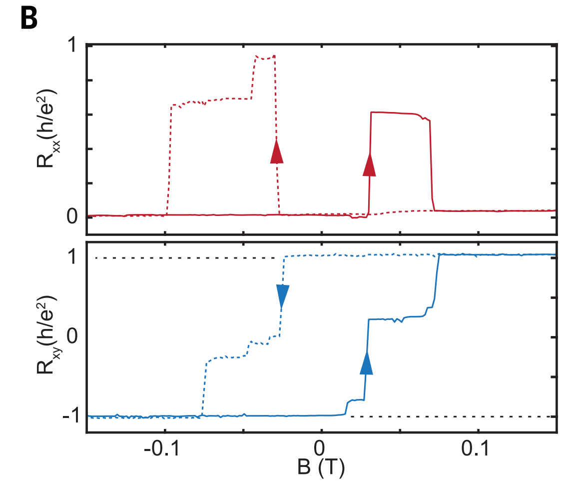
$$\Delta\varepsilon_{\rm exc} = 2\left(\frac{\hbar}{2\Delta}\right)^2 \frac{|evE_\sigma(\omega)|^2}{2\Delta - s\sigma\hbar\omega}$$

Only interband, but no intraband terms

The band edge shift can be measured by tuning the Fermi energy ε_F by an electrostatic gate.

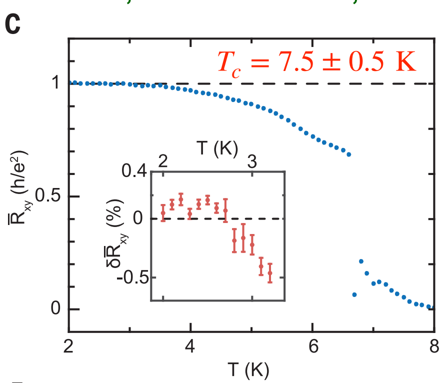
Chern insulator in twisted bilayer graphene on BN

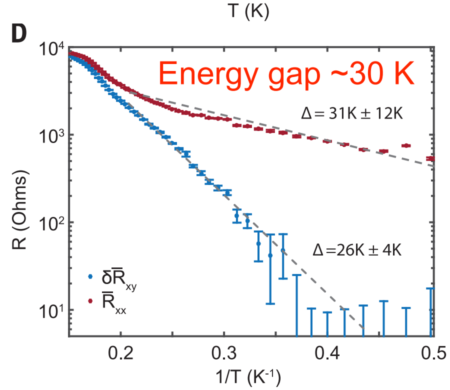


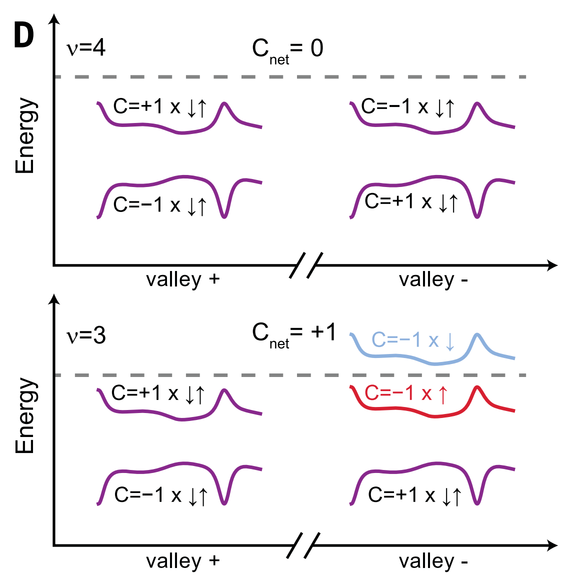


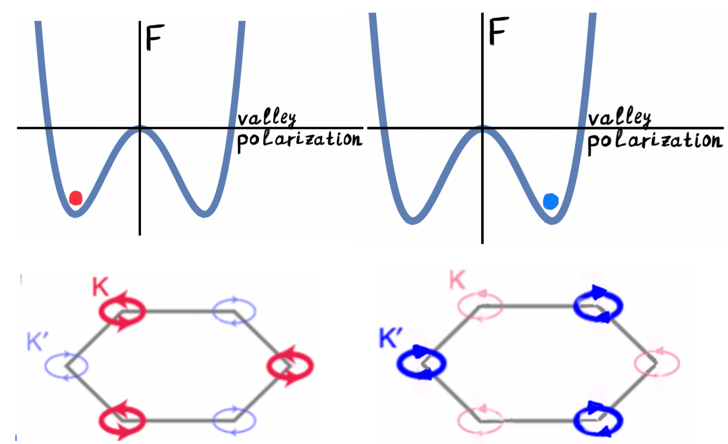
Serlin *et al.*, *Science* **367**, 900 (2020) UCSB, also MIT, Stanford, Barcelona, …

Anomalous quantum Hall effect
= Topological Memory
= Orbital Ferromagnetism





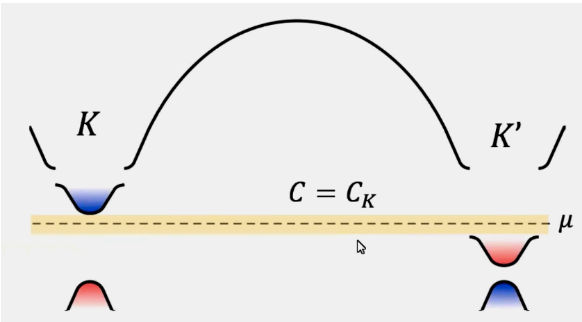




Spontaneous valley imbalance at ν=3 produces Chern number C=+1 or -1

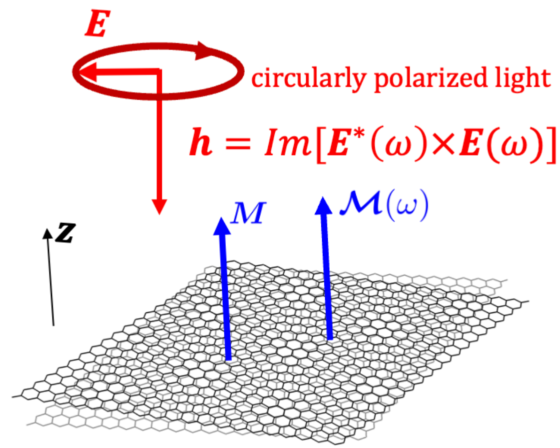
Optical control of orbital magnetism in Chern insulators

Spontaneous valley polarization induced by electron interactions

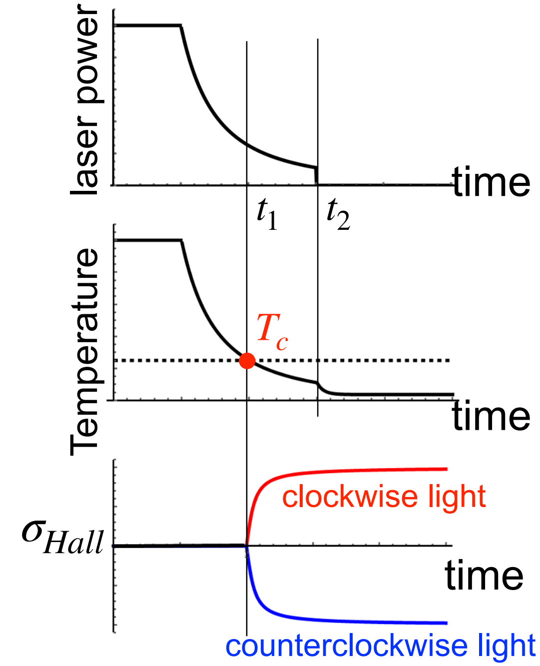


Graphene "Chernburgers", Song *et al*., PNAS **112**, 35 (2015)

Circularly polarized light incident on Moire superlattice



Cool through T_c in the presence of circular light



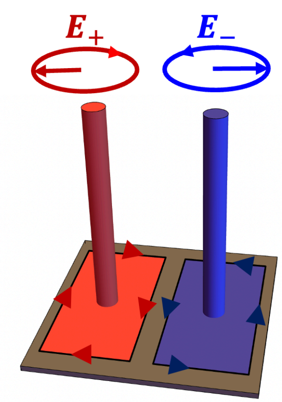
For a Chern insulator with two bands, occupied *n* and empty *m*:

Quantum Hall conductivity $\sigma_H = -\frac{e^2}{\hbar}\int \frac{d^2k}{(2\pi)^2}\Omega_n(\mathbf{k})$

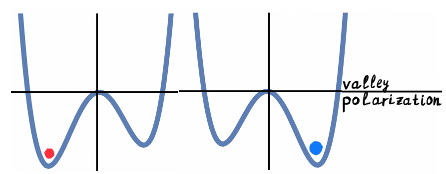
Orbital magnetization $\mathbf{M} = -\frac{e}{2\hbar}\int \frac{d^2k}{(2\pi)^2}[\varepsilon_m(\mathbf{k}) + \varepsilon_n(\mathbf{k}) - 2\varepsilon_F]\,\mathbf{\Omega}_n(\mathbf{k})$, Energy $U = -\mathbf{B}\cdot\mathbf{M}$

Energy shift where light helicity ***h*** couples to "optical magnetization" $\mathscr{M}(\omega)$

$$U^{(a)} = -\frac{1}{4}\mathbf{h}\cdot\mathscr{M}(\omega), \quad \mathscr{M}(\omega) = \mathscr{A}e^2\int\frac{d^2k}{(2\pi)^2}\frac{\varepsilon_{mn}^2(\mathbf{k})}{\varepsilon_{mn}^2(\mathbf{k}) - (\hbar\omega)^2}\frac{\mathbf{\Omega}_n(\mathbf{k})}{\hbar\omega}$$



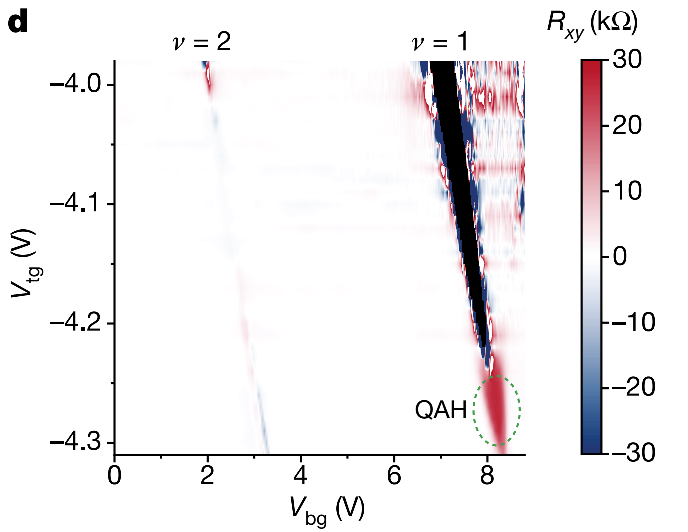
The coupling to the helicity *h* is linear in *M* and dominates over the Landau energy $U \sim (T - T_c)\,M^2 + M^4$ near T_c

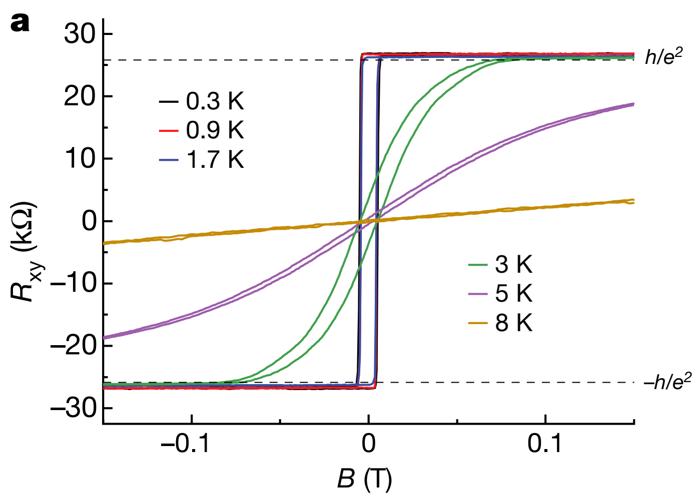


Optical writing of topological domains

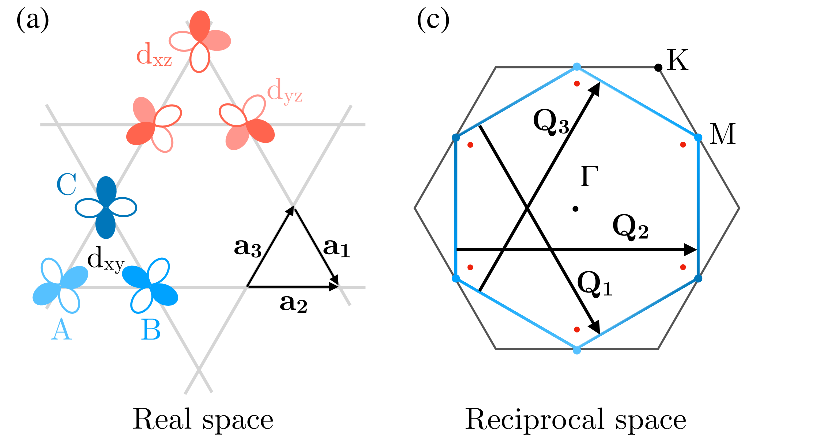
Chern insulator in other materials

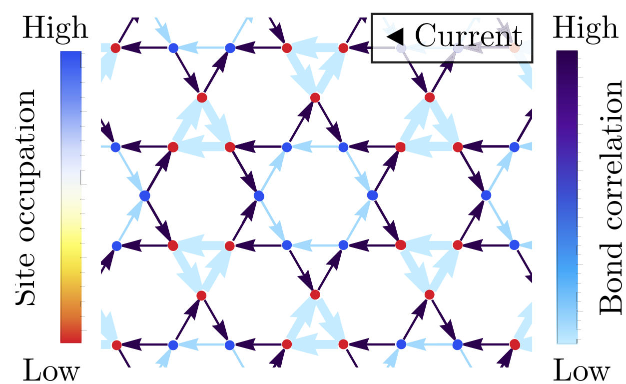
AB-stacked $MoTe_2/WSe_2$ bilayers, experiment
Tingxin Li, …, Kin Fai Mak, *Nature* **600**, 641 (2021)





Kagome metals AV_3Sb_5 (A=K,Rb,Cs), theory
Denner, Thomale, Neupert, *PRL* **127**, 217601 (2021)





- Currents due to three charge-density/bond-density waves with relative phases: a Chern insulator?
- Some experimental evidence for time-reversal symmetry breaking at T^*~80 K, but controversial

Optical control in other materials

Yu, Claassen, Kennes, Sentef (Hamburg), *PRR* **3**, 013253 (2021)
Optical manipulation of domains in chiral topological superconductors (theory)

Qiu, … , Vishwanath, Ni Ni, Su-Yang Xu (Harvard), *Nat Mat* **22**, 583 (2023)
Axion optical induction of antiferromagnetic order (even-layered $MnBi_2Te_4$)

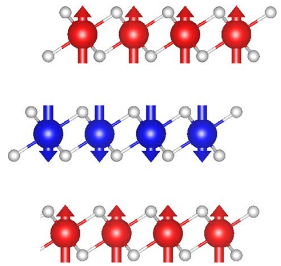
Xu, … , Jarillo-Herrero, Gedik (MIT), *Nature* **578**, 545 (2020)
Spontaneous gyrotropic electronic order in a transition-metal dichalcogenide
(1T-$TiSe_2$, spiral CDW proposed by van Wezel)

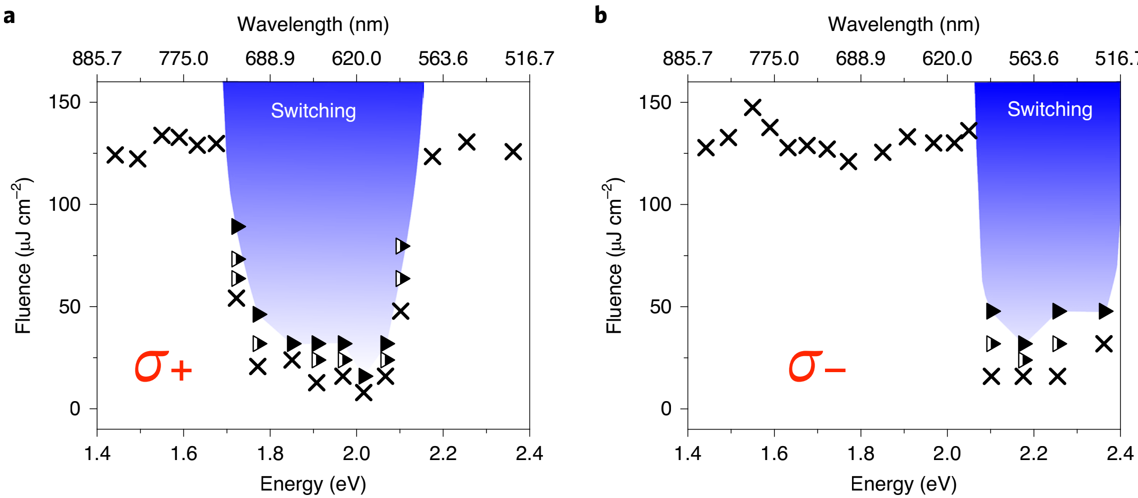
The group of Cheng Gong (UMD), submitted to *Nat Phys* (2024)
High-efficiency optical training of itinerant 2D magnets
(spin Ising ferromagnet Fe_3GeTe_2)

Peiyao Zhang, … , Xiang Zhang (Berkeley), *Nat Mat* **21**, 1373 (2022)
All-optical switching of magnetization in atomically thin CrI_3
(odd-layered 2D Ising ferromagnet, optical pulses)

Magnetization switching by strong optical pulses

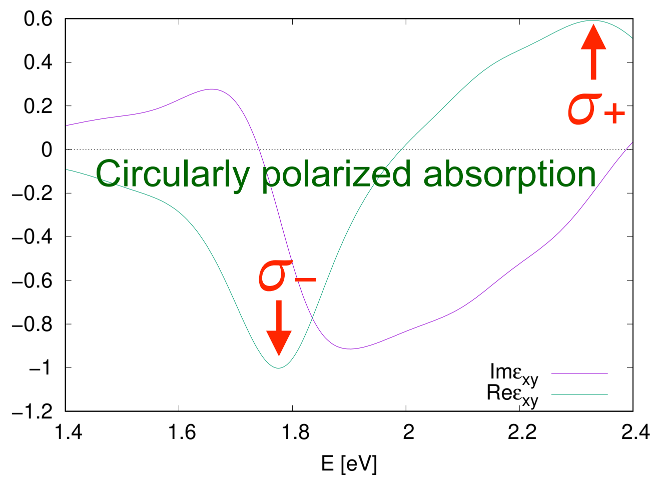
Zhang, … , Zhang (Berkeley), *Nat Mat* **21**, 1373 (2022)
All-optical switching of magnetization in atomically thin CrI_3
(odd-layered 2D Ising ferromagnet, optical pulses)





The 2D ferromagnet flips magnetization in order to maximize absorption of circularly polarized light.

Calculated spectrum courtesy of Igor Mazin (GMU)



This behavior agrees with the principle of maximal entropy production, i.e. maximal dissipation, for systems driven far from equilibrium.

Conclusions

Phys Rev B **105**, 064423 (2022) and **108**, 059904(E) (2023)
Annals of Physics **447**, 169075 (2022)

- We derived the Stark energy shift for solids, where the wave functions are delocalized with the momentum $\mathbf{k}(t)$. The prediction can be experimentally verified for transition-metal dichalcogenides.
- The new intraband contributions result in (i) flattening of energy spectrum (ii) coupling to the helicity of light.
- Topological memory based on orbital magnetization in Chern insulators can be controlled by circularly polarized light.
- Optical control also applies to order parameters in other materials.
- Optical switching of magnetization in 2D ferromagnet CrI_3 can be explained using the principle of maximal entropy production.