

Optical Readout of Reconfigurable Layered Magnetic Domain Structure in CrSBr

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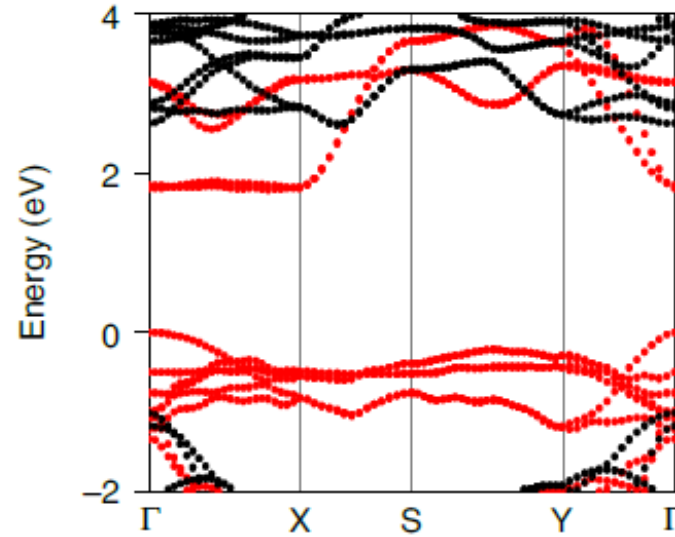
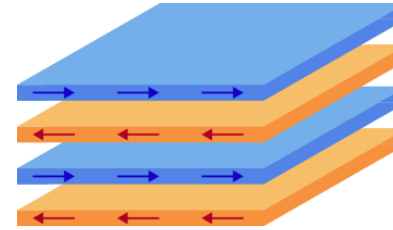
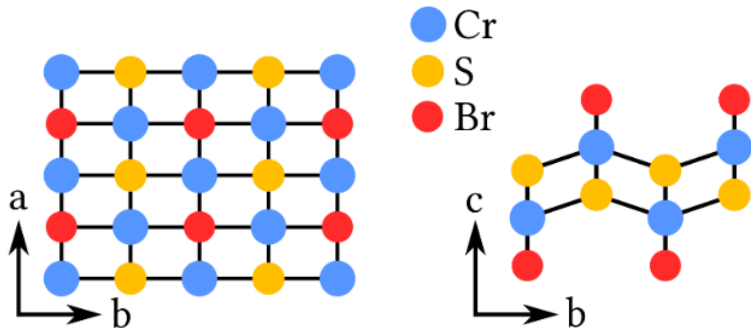
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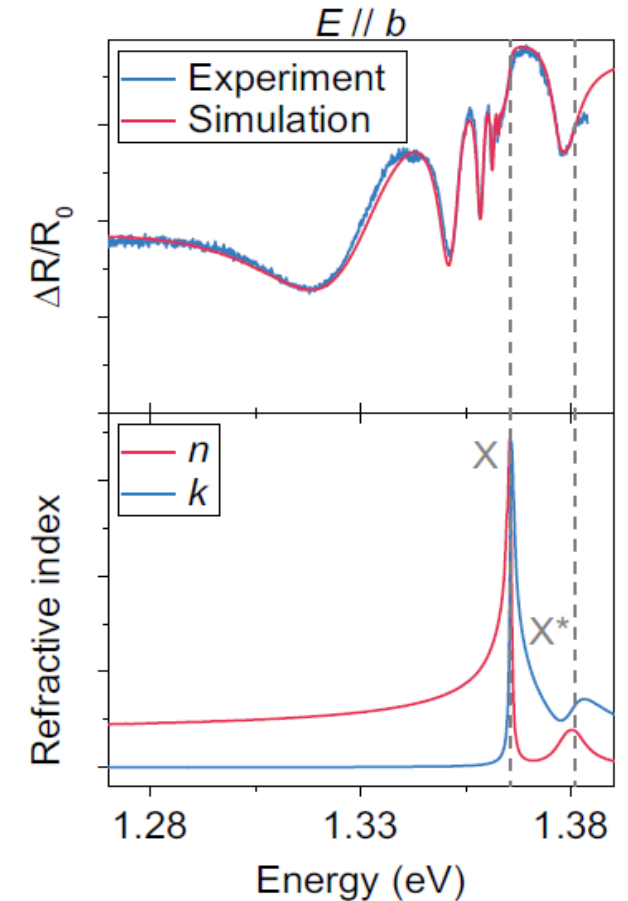


Chromium sulfide bromide (CrSBr)

- layered **magnetic** semiconductor
- high anisotropy (optical, magnetic...)
- direct bandgap – $E_A \approx 1.3\text{eV}$
- strong exciton** → dispersive $\epsilon(\omega)$



N. Wilson, et al. *Nature Materials* 20.12 (2021): 1657-1662



Magnetically-dressed CrSBr excitonpolaritons in ultrastrong coupling regime, *Nat. Comm.* | (2023) 14:5966

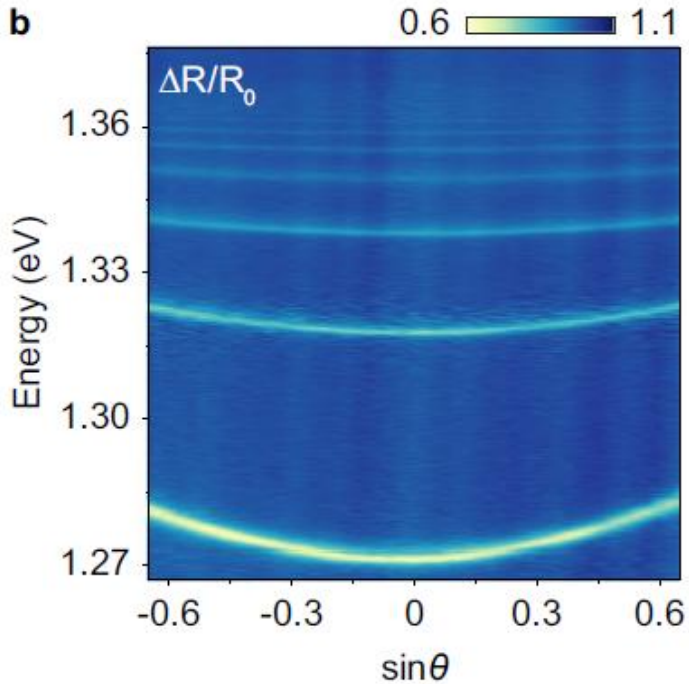
Strong excitonic resonance in CrSBr

A-exciton (≈ 1.34 eV):

high oscillator amplitude $\hbar^2 f \approx 1.7-2$ eV² and small linewidth: $0.68 \hbar \gamma \approx 0.68$ meV [1]

high refractive index of CrSBr in the range of the excitonic resonance

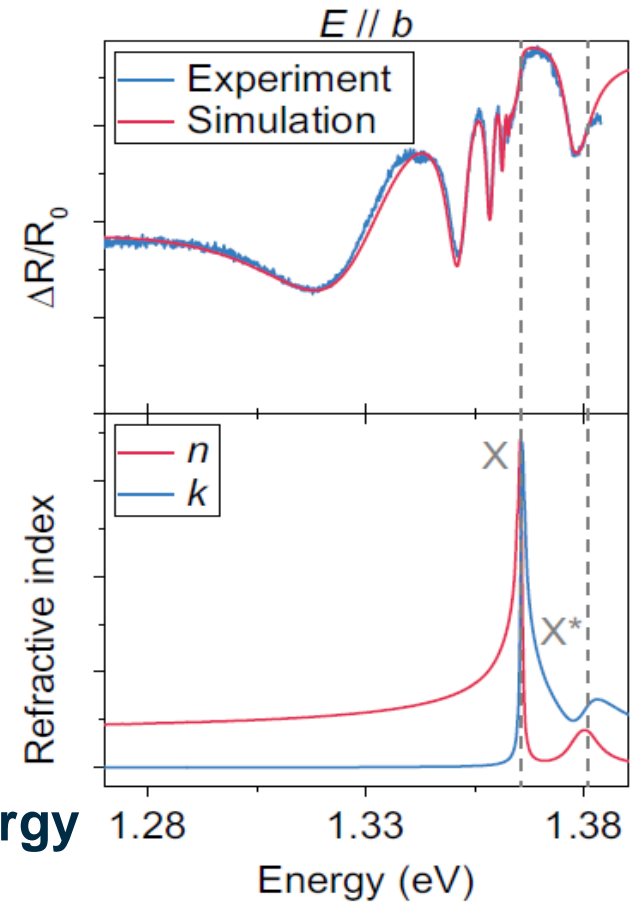
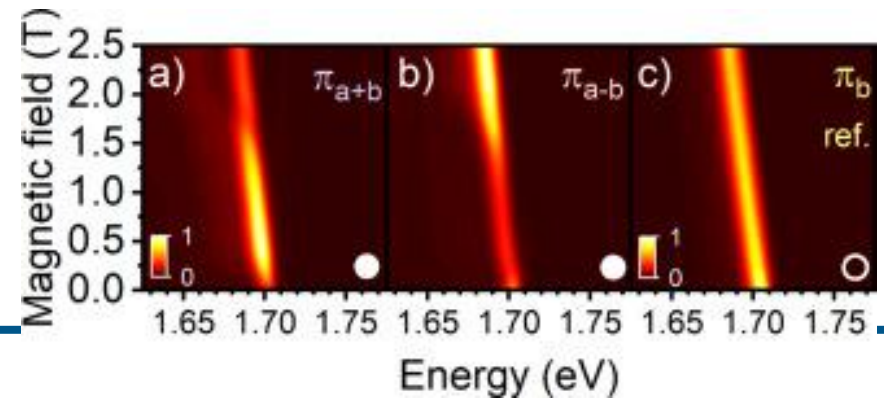
bulk CrSBr crystals create self-sufficient optical microcavities with self-hybridized exciton-polaritons [2]



Magnetically-dressed CrSBr exciton-polaritons in ultrastrong coupling regime, Nat. Comm. | (2023) 14:5966

+ magnetic field dependence of exciton energy
(different for AFM and FM ordering)

we can think about magnetic switching of the optical properties



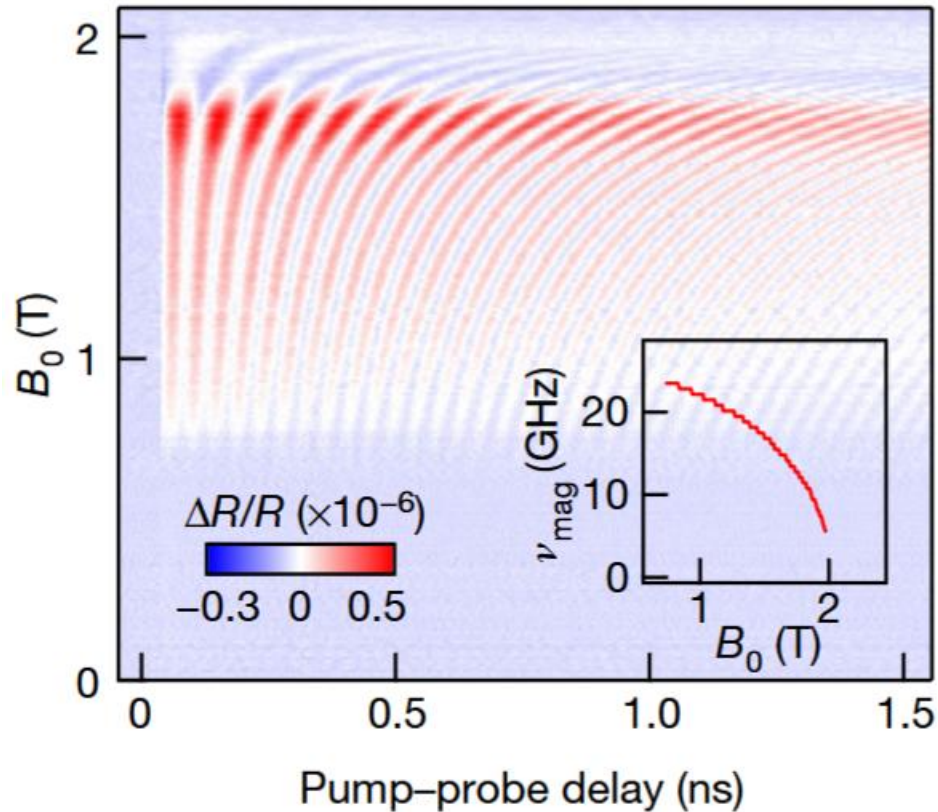
[1] [3] T. Wang et al., Nature Communications, 14(1), 2023.

[2] F. Dirnberger, et al., Nature, 620(7974):533–537, 2023.

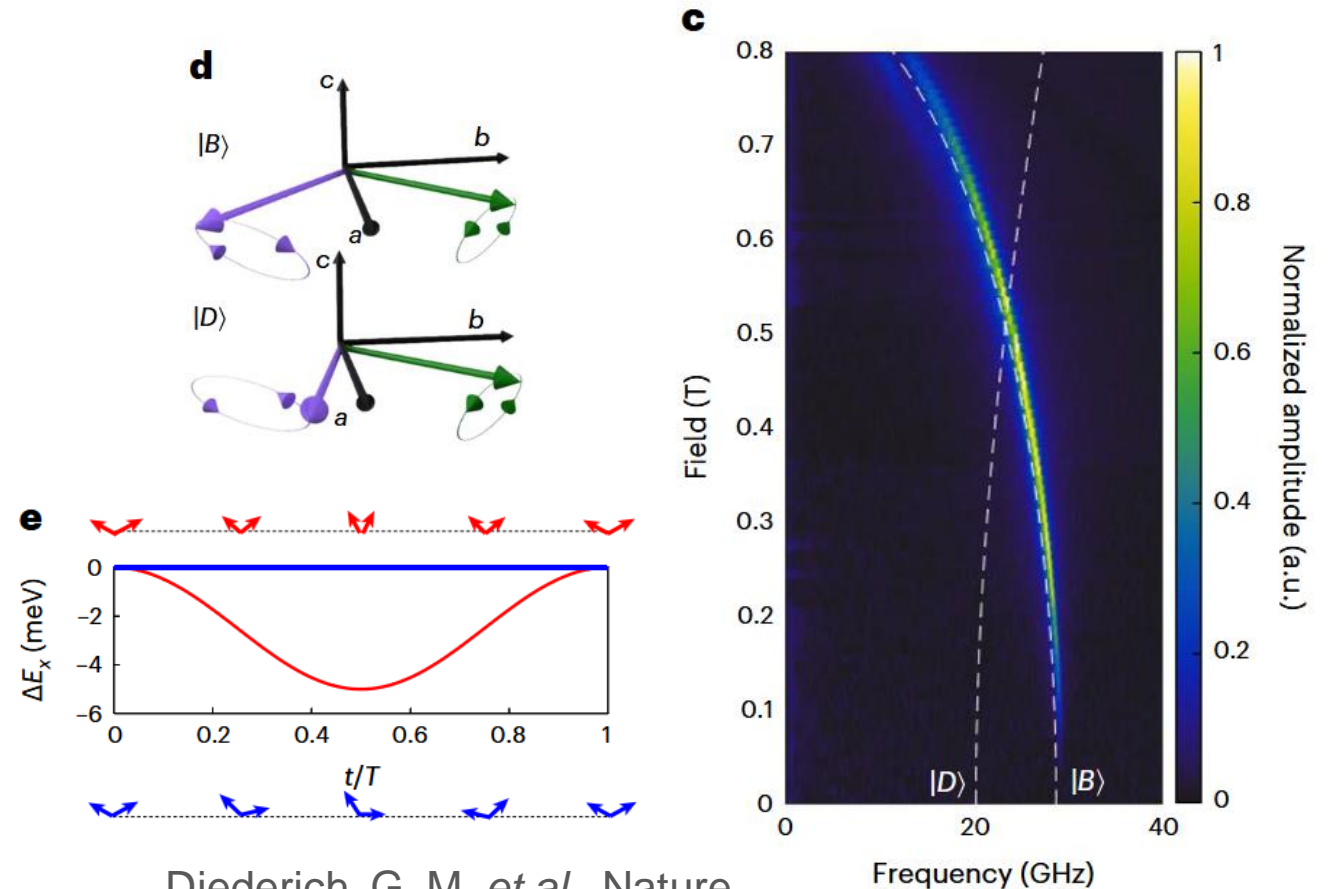
Why excitons are useful in CrSBr: they sense spin order

exciton–magnon coupling tunable by field direction / strain

coherent magnons modulate exciton energy



Bae, Y. J. *et al.*, Nature 609, 282–286 (2022)

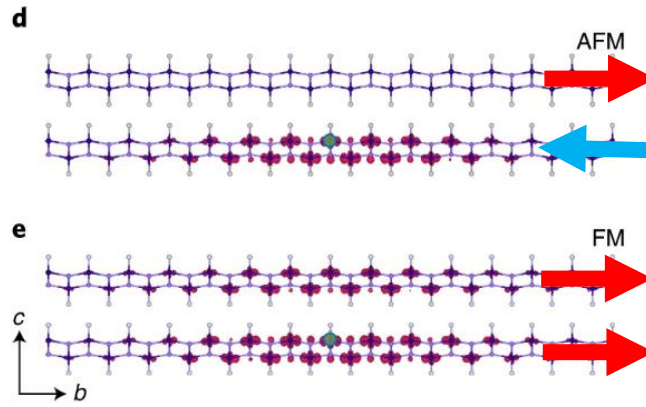
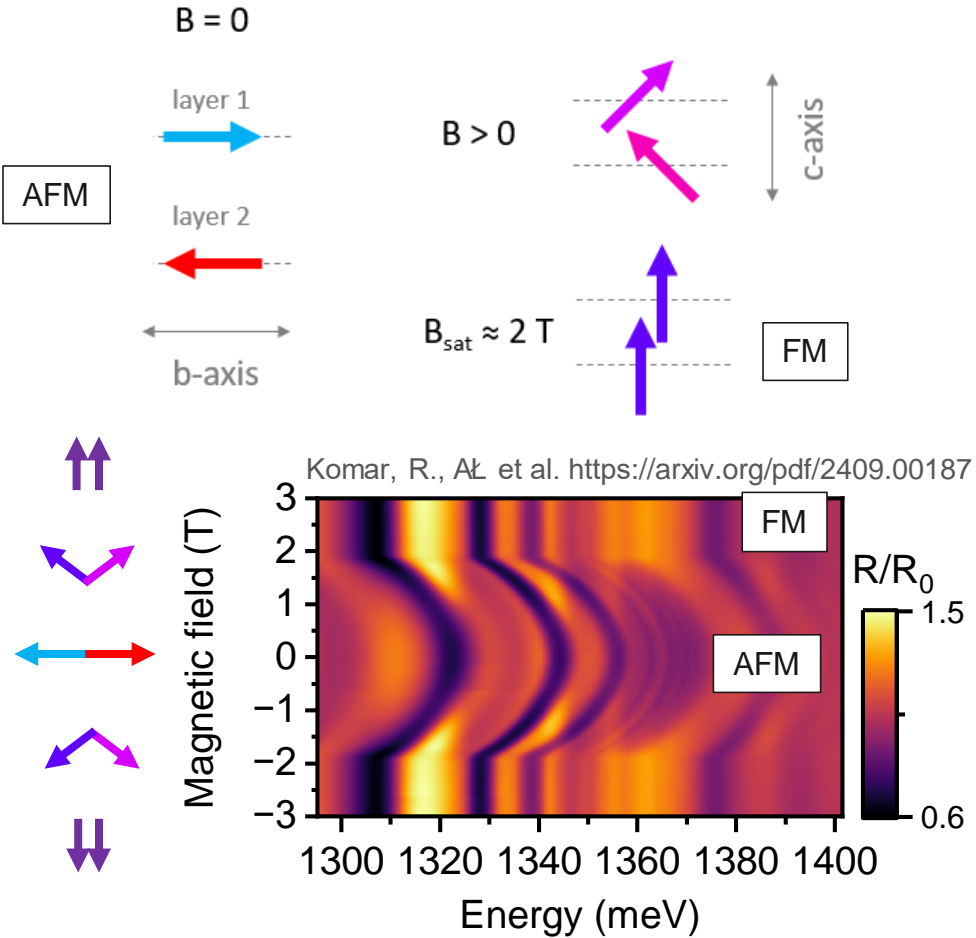


Diederich, G. M. *et al.*, Nature
Nanotechnology 18, 23–28 (2023)

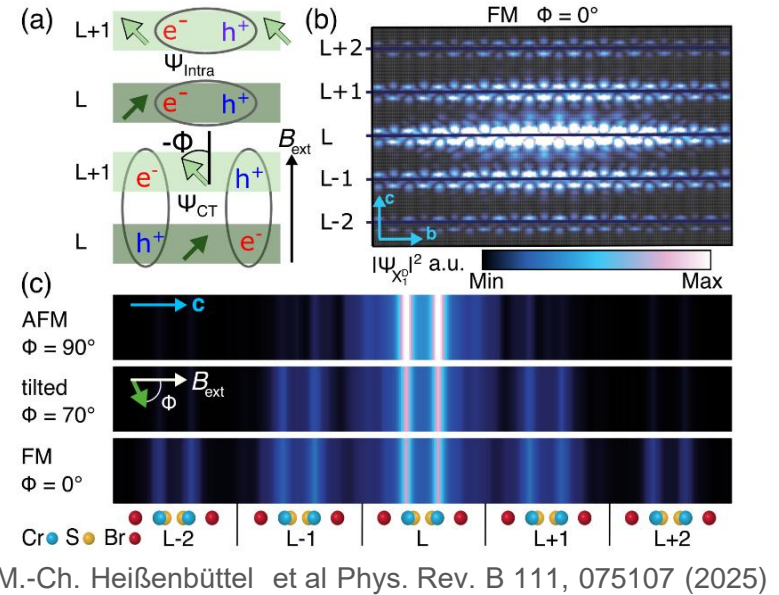


Why optics is sensitive to magnetism in CrSBr

magnetic alignment tunes interlayer hybridization → excitonic dielectric response

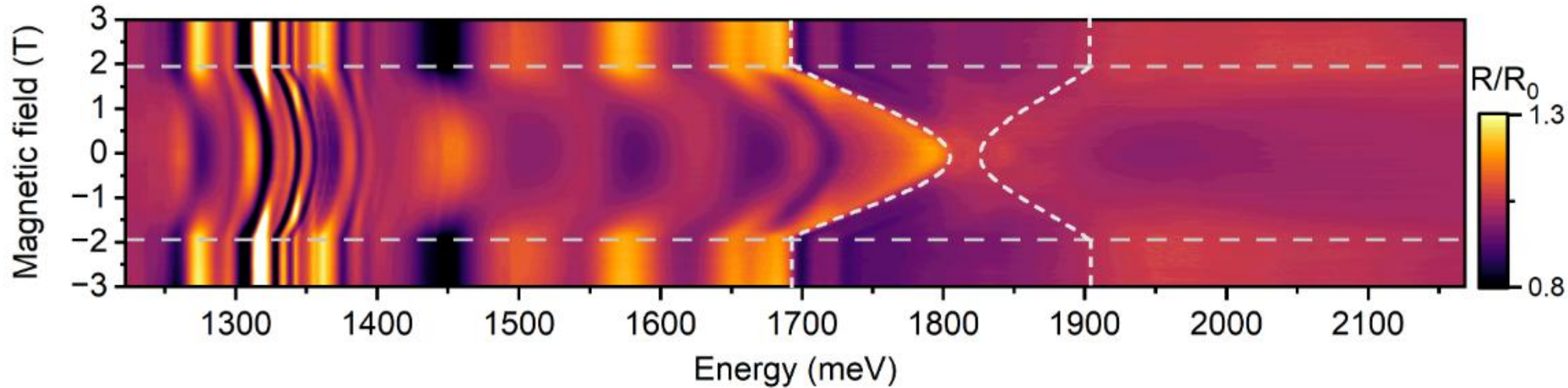


N. Wilson, et al. *Nature Materials* 20.12 (2021): 1657-1662



strong coupling between optical and magnetic properties

Higher energy excitons



A-exciton band

Beyond the A-exciton:

higher-energy excitons
different spin–exciton coupling channels
→ giant magneto-optical shifts

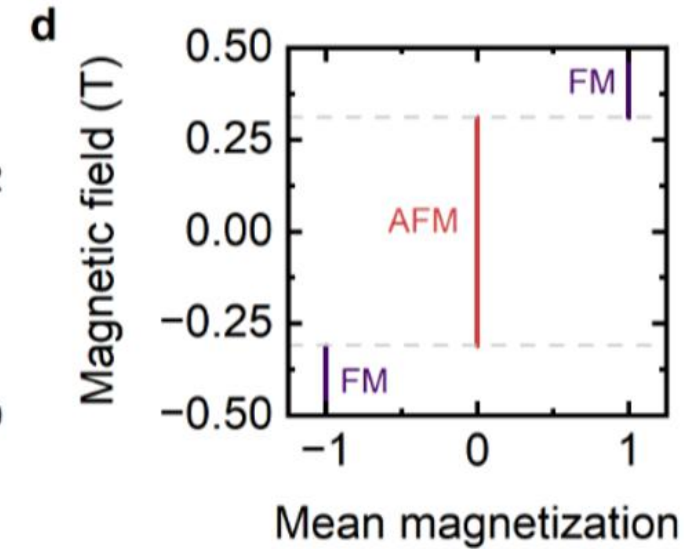
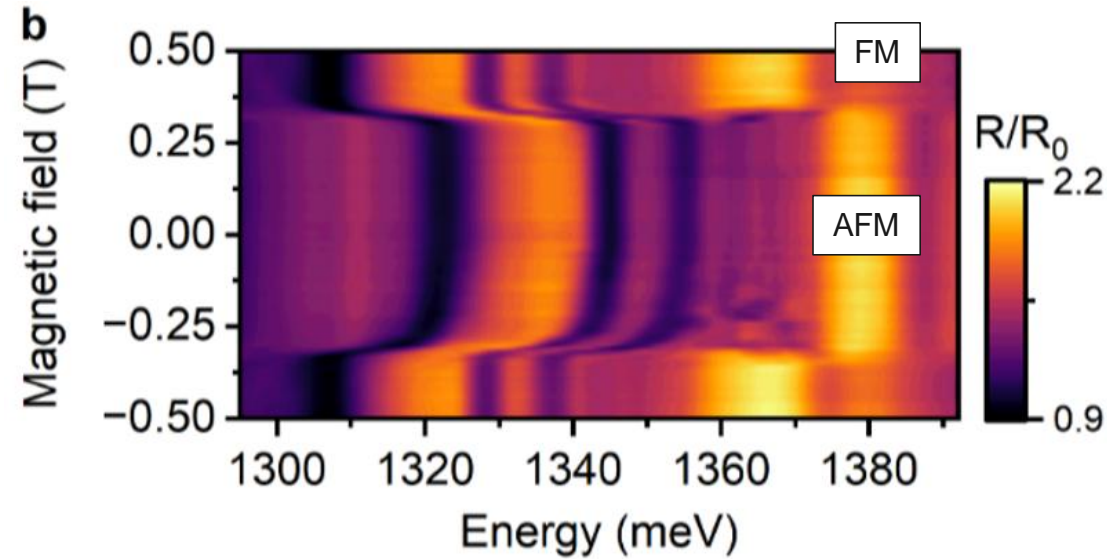
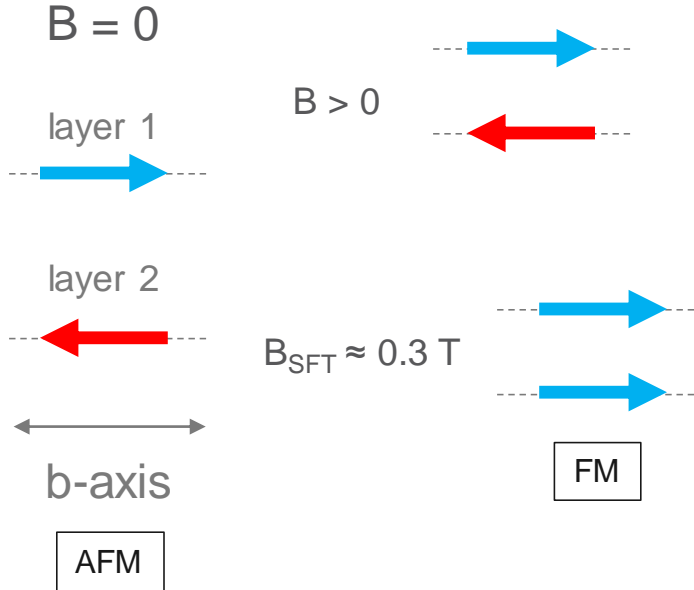


Komar, R., AŁ. et al.
accepted in Nat. Commun.
arXiv:2409.00187

See also: Śmirtka, M. et al., Nat. Commun. 17, 1777 (2026)

Easy-axis field is special: switching vs canting

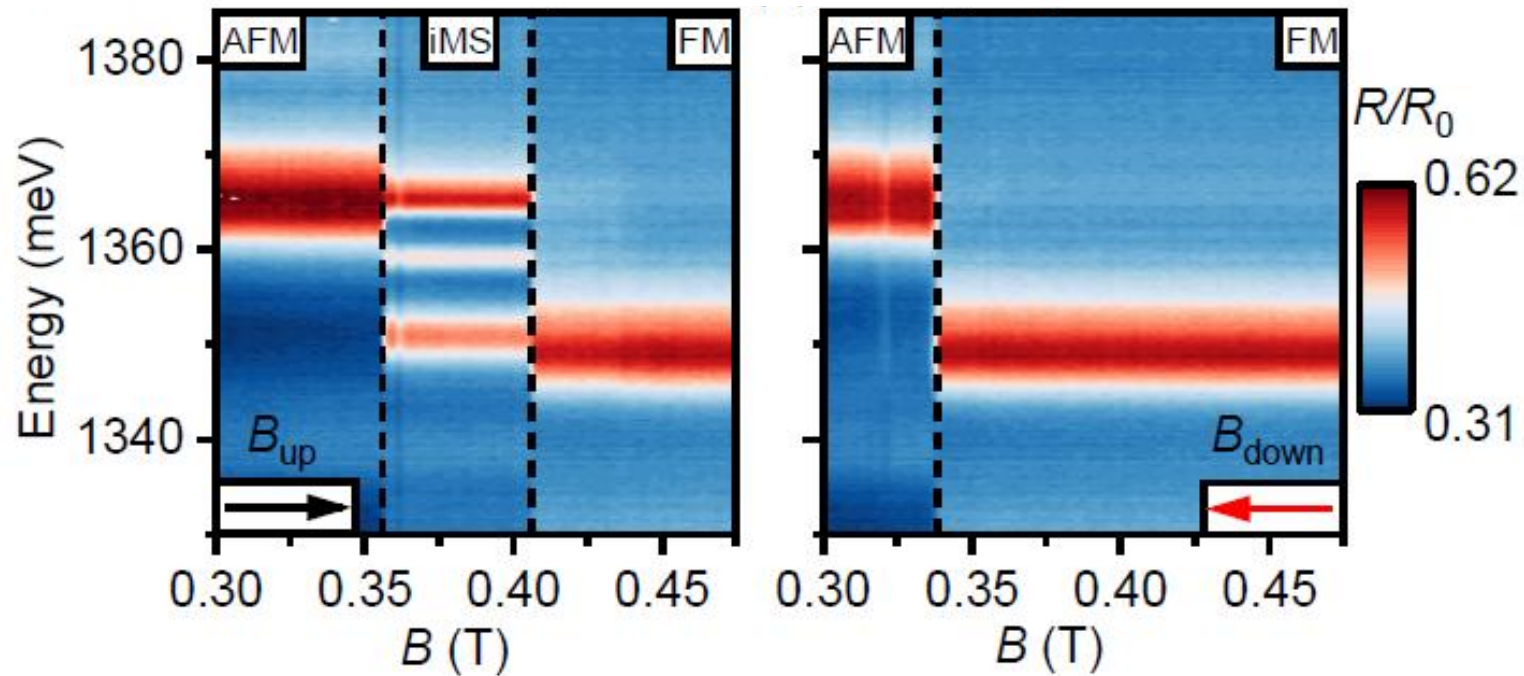
- $B \perp b$ (hard/intermediate axes): canting \rightarrow smooth spectral shifts
- $B \parallel b$ (easy axis): discrete switching between configurations



Komar, R, AŁ., et al. <https://arxiv.org/pdf/2409.00187>

Next: intermediate configurations and sweep-history dependence for $B \parallel b$

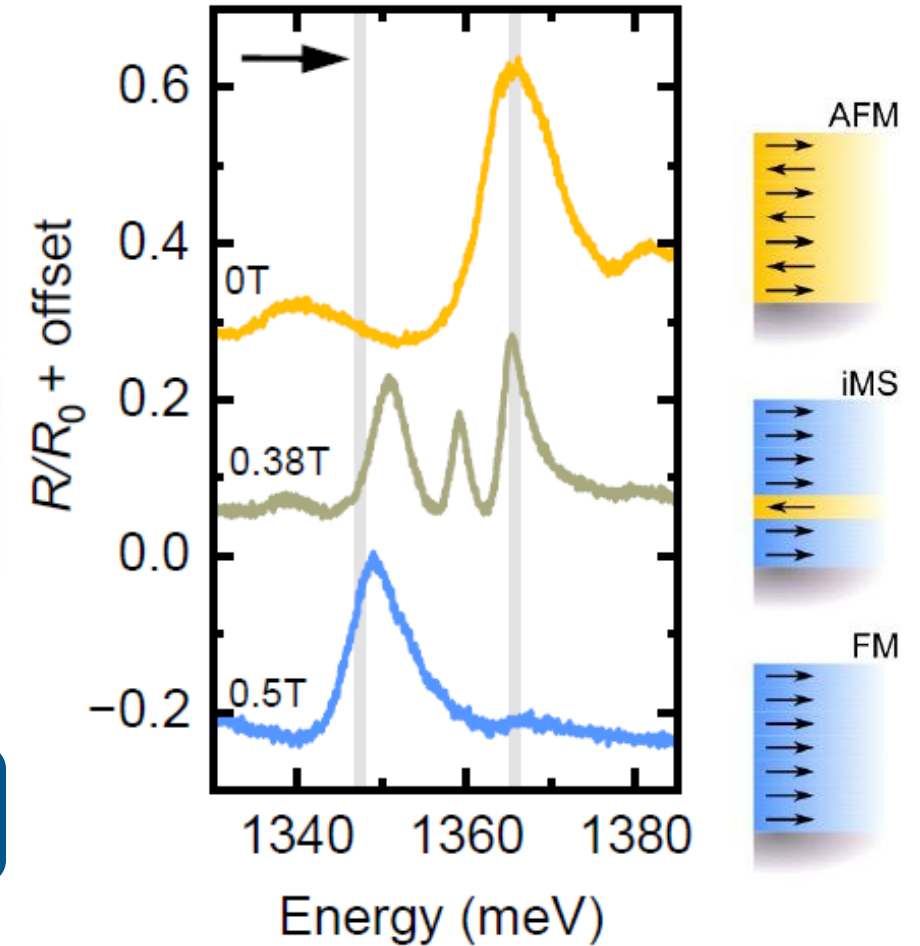
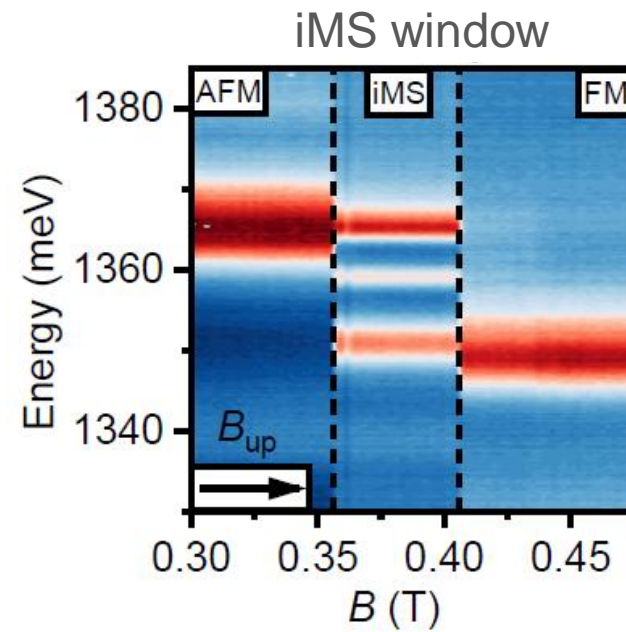
Non-binary switching (7L): multi-level intermediate states



- multi-level states
- reconfigurable, path-dependent (B_{\uparrow} vs B_{\downarrow})
- stable intermediate configurations

Intermediate magnetic states (iMS): out-of-plane domains

- layer-by-layer switching
- mixed AFM/FM sequence (variable domain thickness)

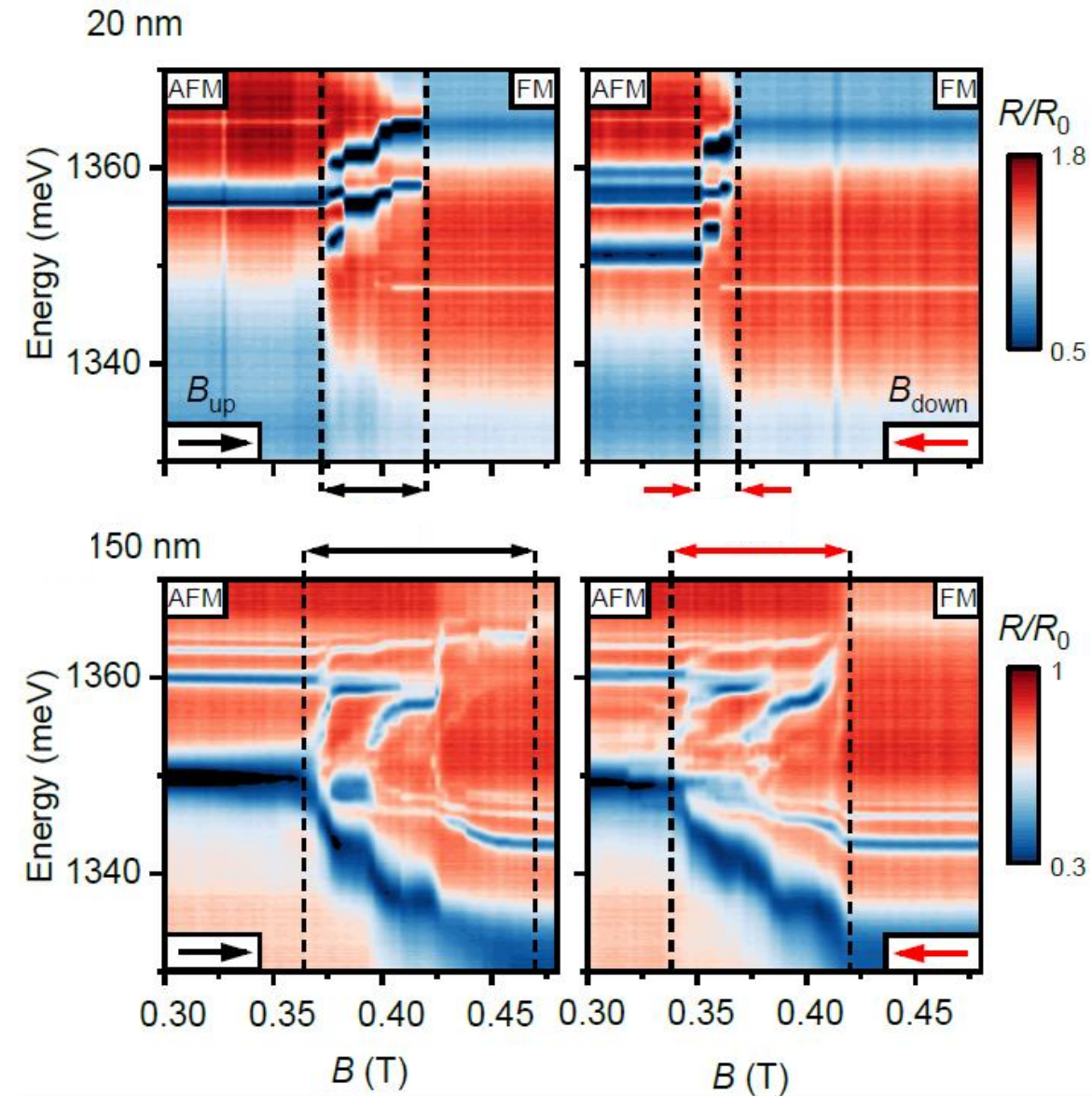


iMS = layered AFM/FM domain stack (out-of-plane)

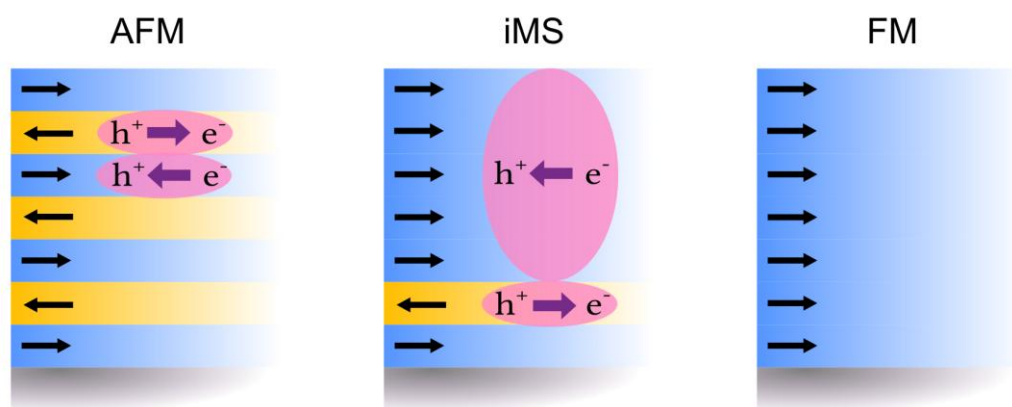
Non-binary switch: thicker flakes

- thicker flakes \rightarrow richer multi-level landscape.
- path-dependent access (B_{\uparrow} vs B_{\downarrow})

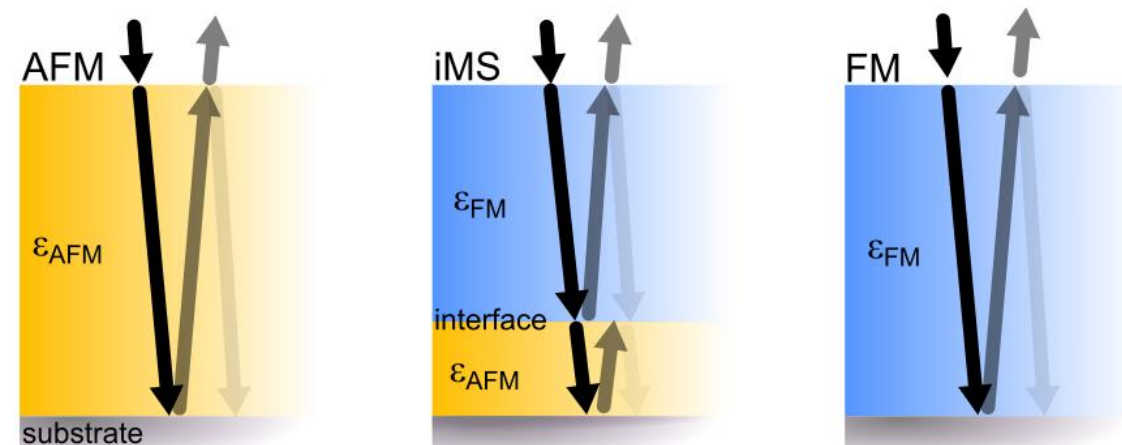
more accessible intermediate configurations



layer alignment \rightarrow interlayer hybridization
 \rightarrow excitonic $\epsilon(\omega)$

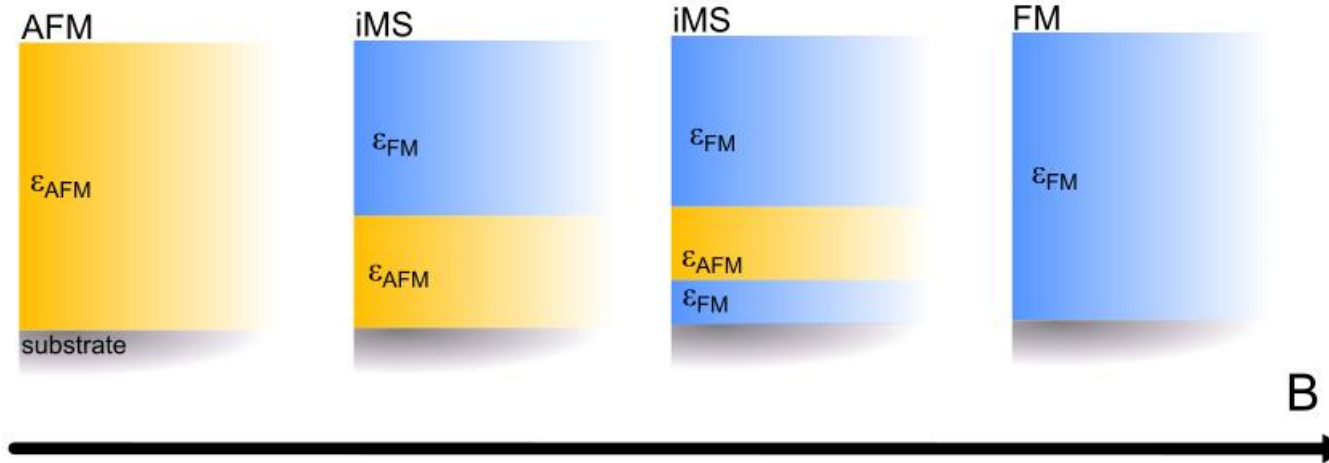


domain stack \rightarrow multilayer interference conditions

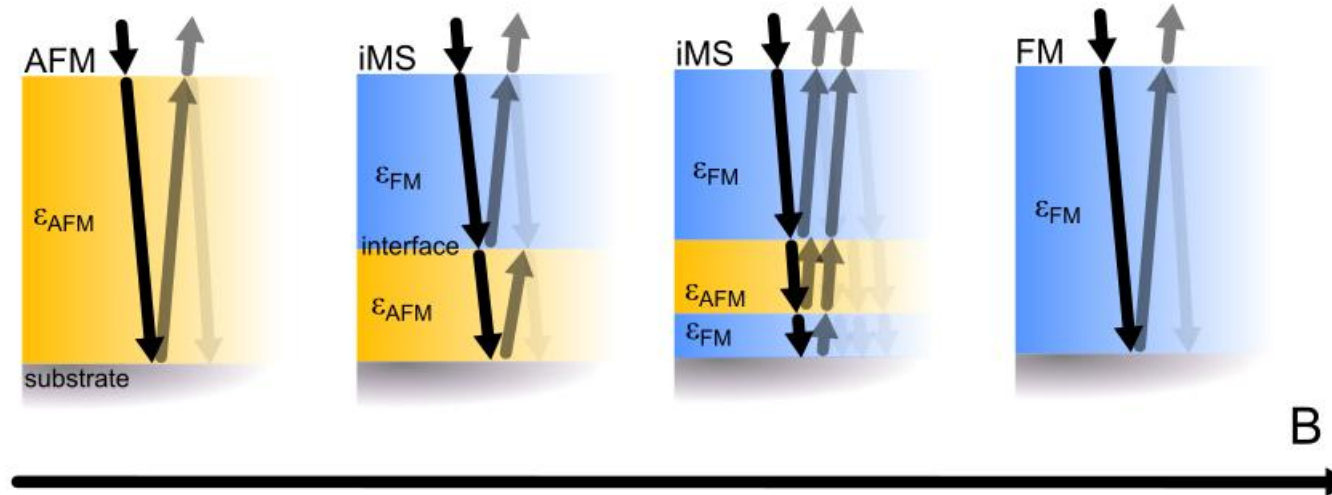


combined \rightarrow configuration-specific spectral signatures (readout)

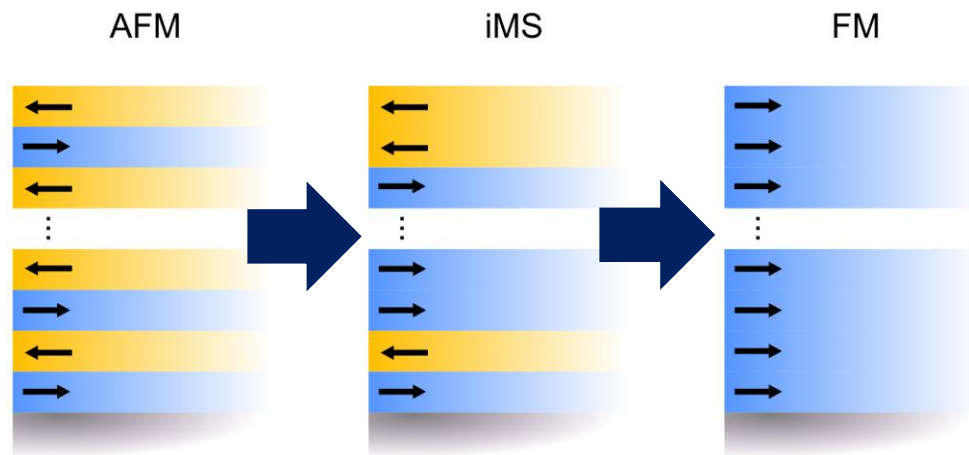
Why spectra change: configuration-dependent interference



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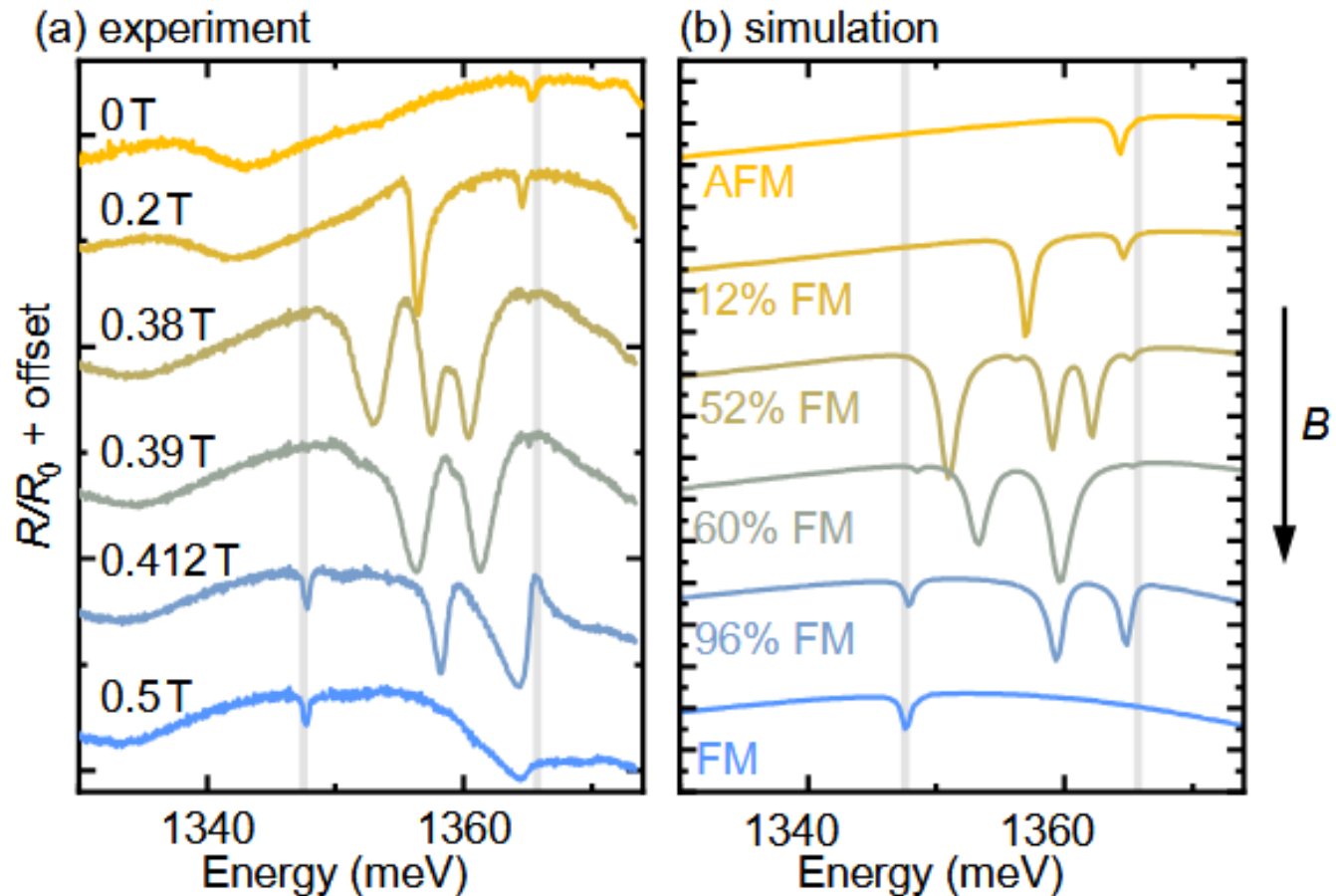


Minimal transfer-matrix model



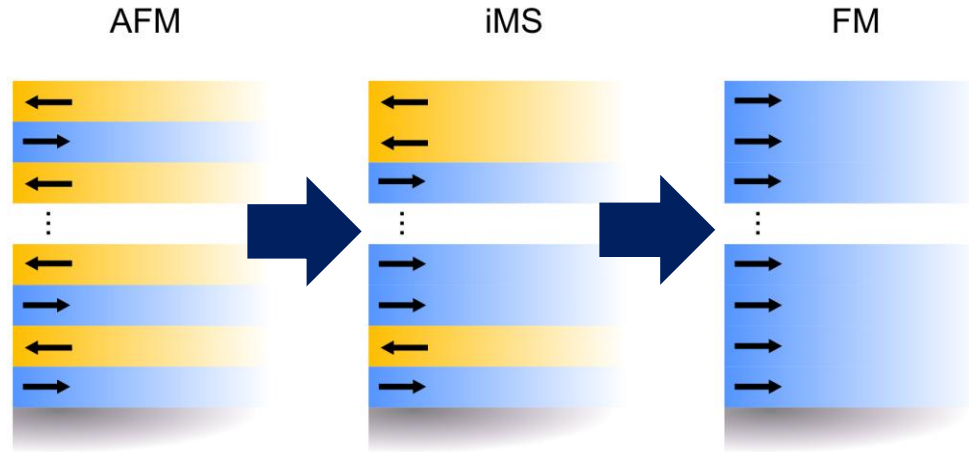
- single exciton resonance with different **AFM vs FM** energies \rightarrow different $\epsilon(\omega)$
- layer-by-layer switching \rightarrow mode multiplication

Spectral complexity = interference of one tunable exciton with layered stack



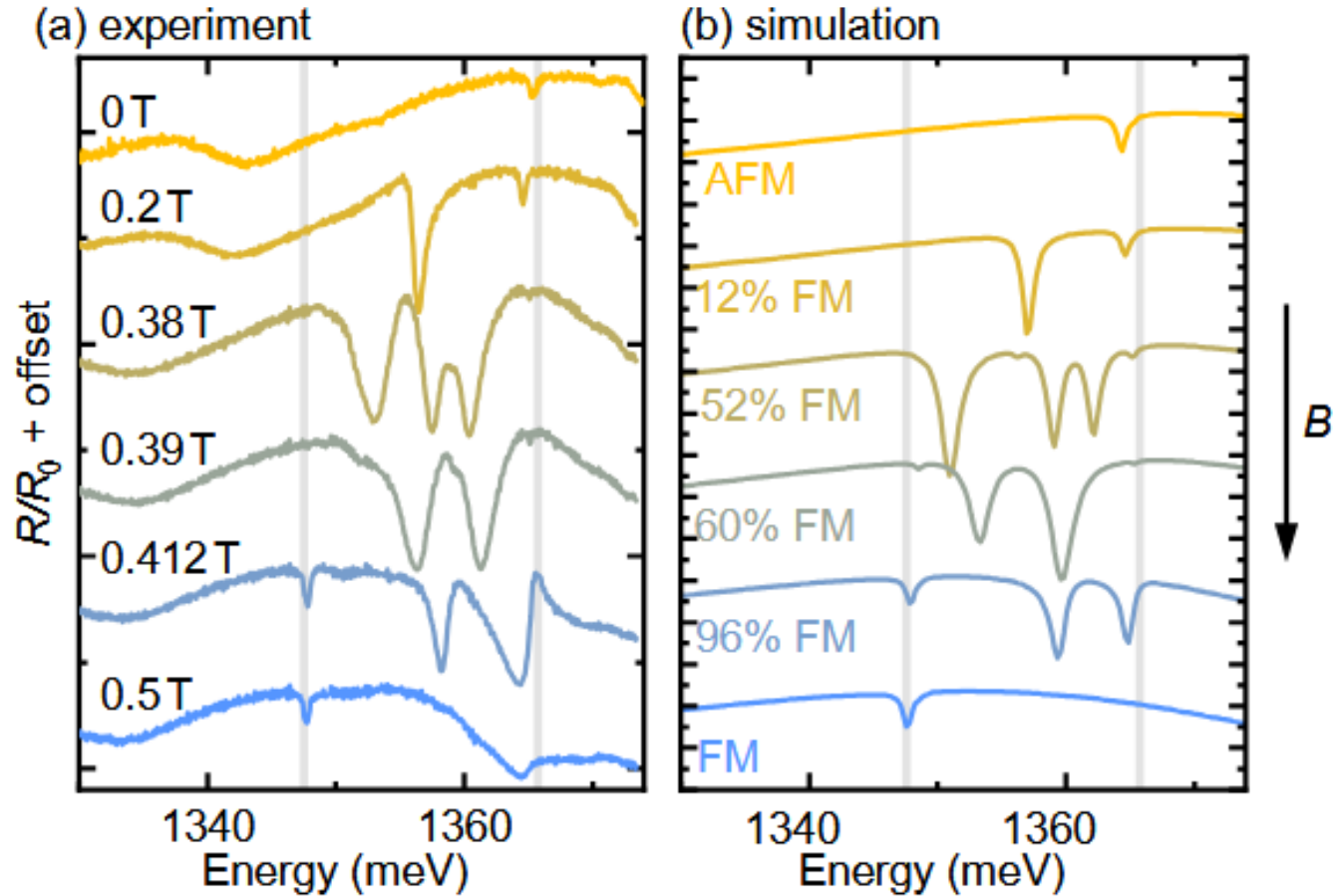
Minimal transfer-matrix model

- pureAFM
- ['FM','AFM'],[3,22]
- ['FM','AFM','FM','AFM','FM','AFM'], [3,3,8,2,2,7]
- ['FM','AFM','FM'], [12,10,3]
- ['FM','AFM','FM'],[22,1,2]
- pureFM



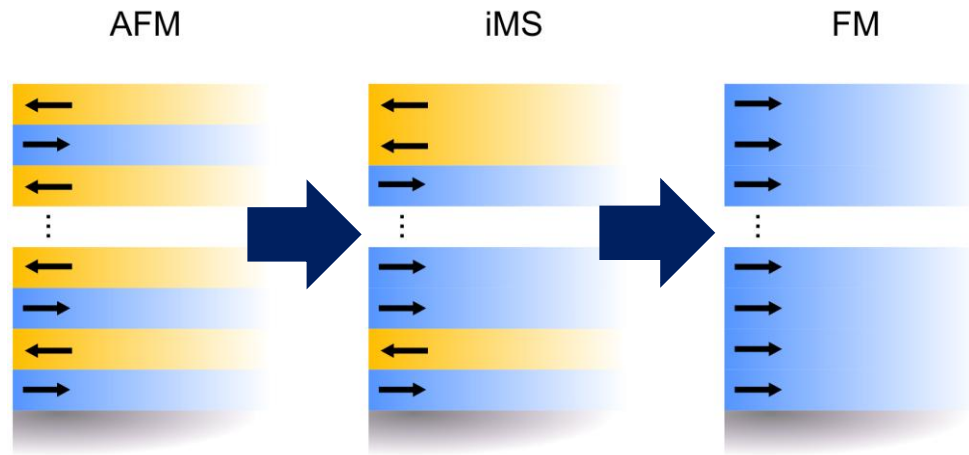
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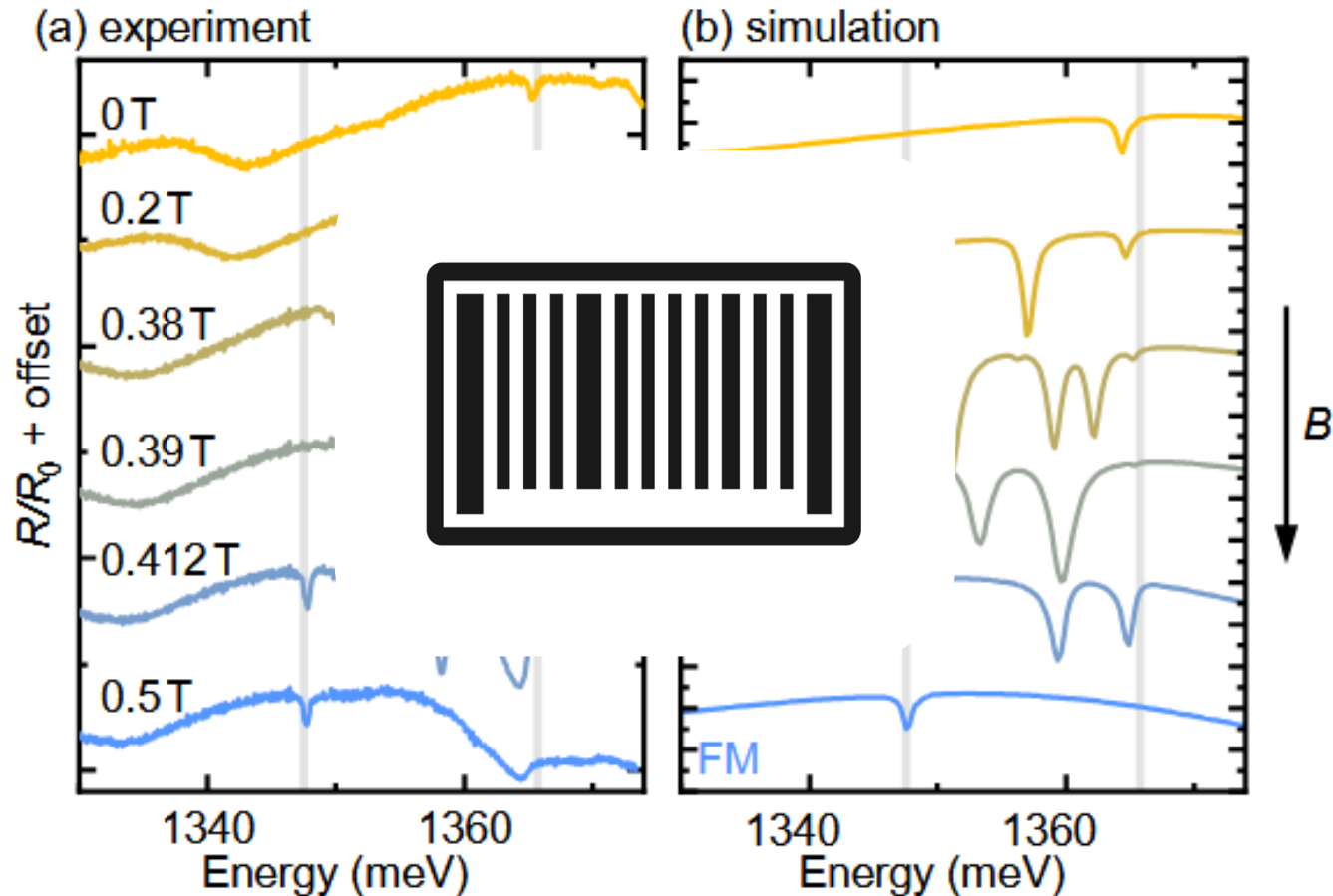
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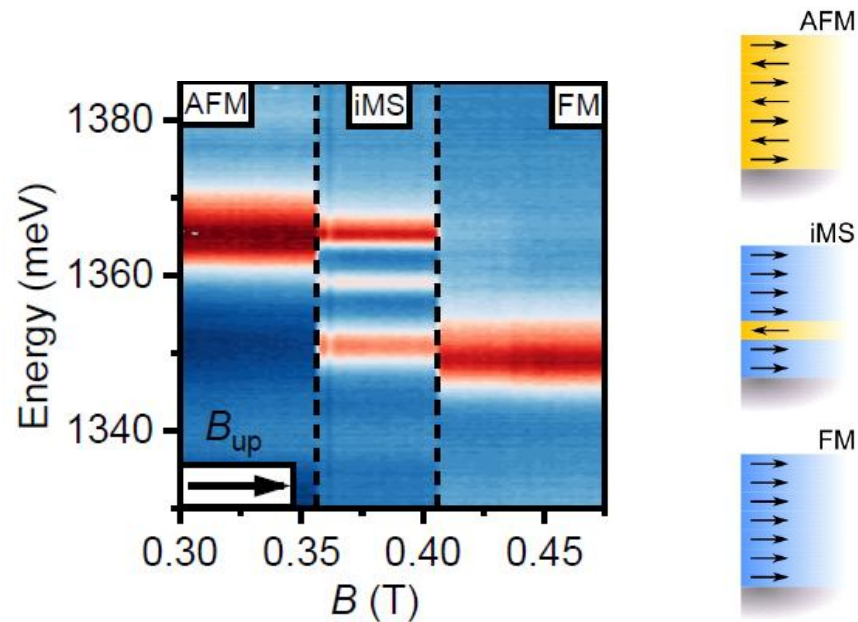


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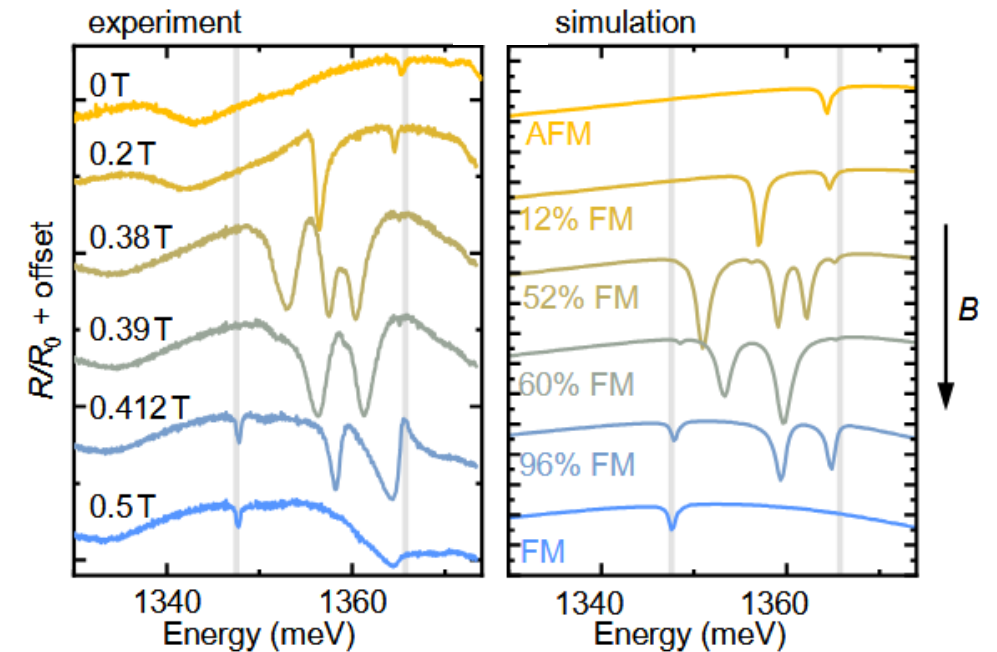
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Intermediate magnetic states: stable, readable, multi-level



- distinct out-of-plane domains
- stable intermediate regime
- history dependent (multi-level hysteresis / path dependence)

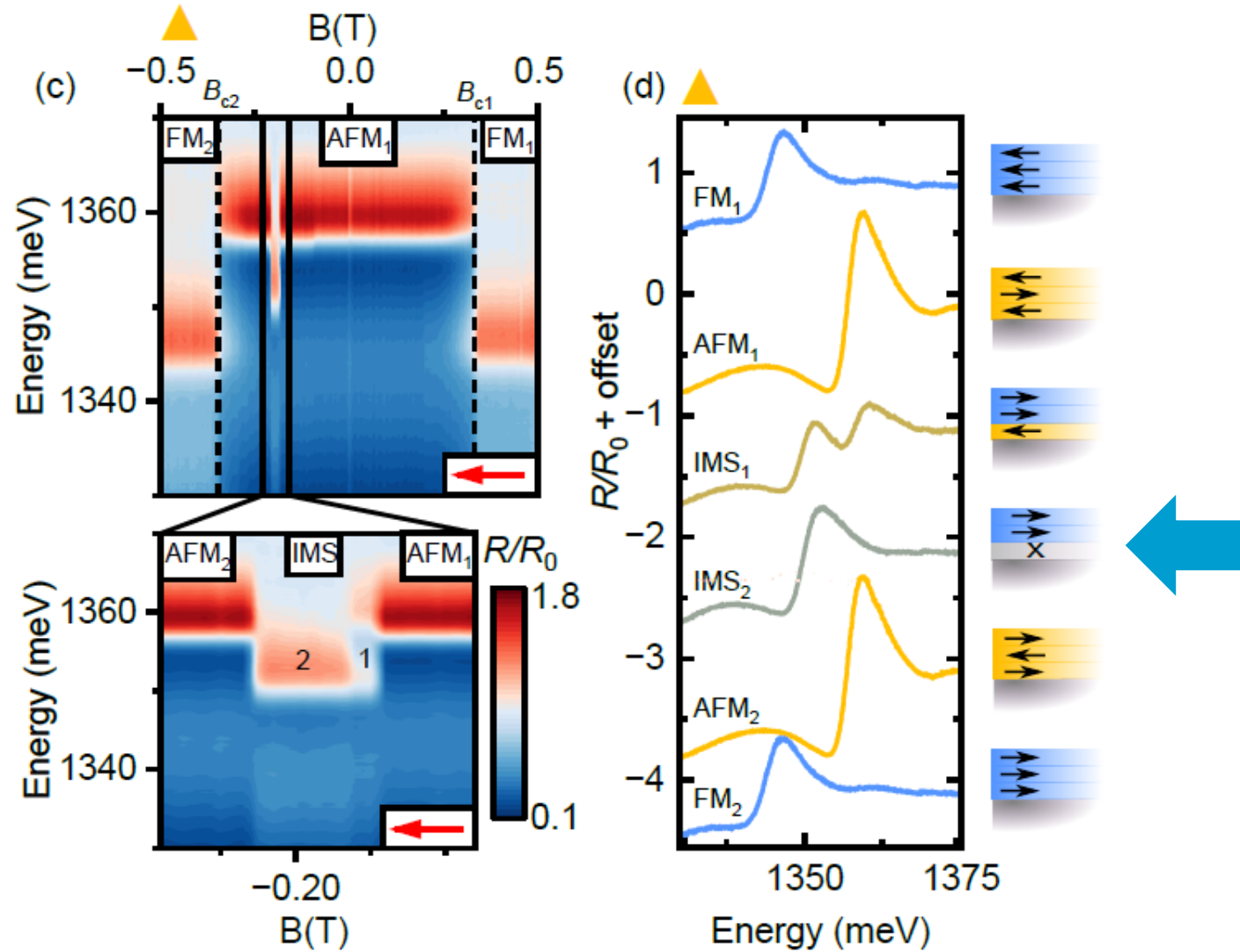


Optical barcode → non-contact **readout**

Next: what could this enable if we gain deterministic selection of configurations?

„Invisible” layer?

3L CrSBr





Outlook: towards intelligent matter

- structural: multi-level configurations
- sensing: optical barcode readout
- memory: multistability (partial)
- actuation: deterministic selection



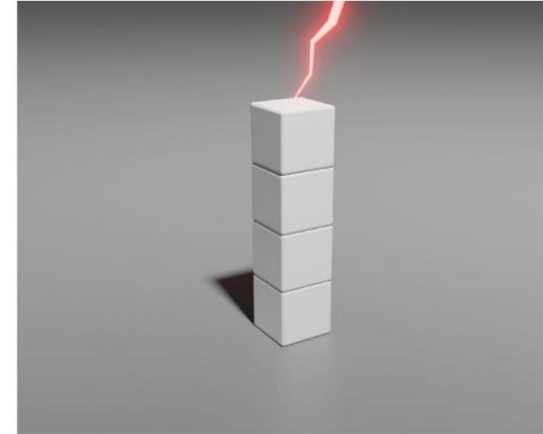
→ extra knobs: gating / strain / heterostructures

- network: coupling between elements

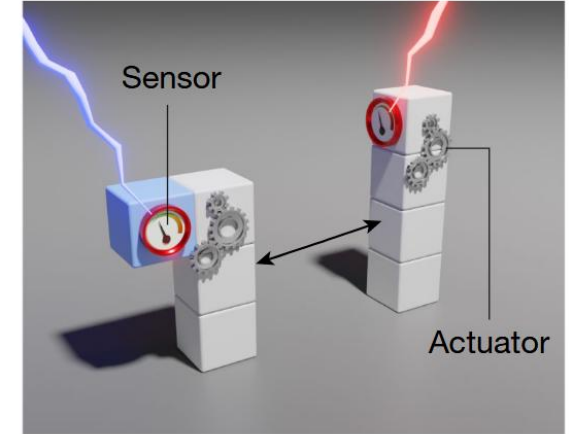


CrSBr: a step toward 'intelligent matter'
(requires deterministic control)

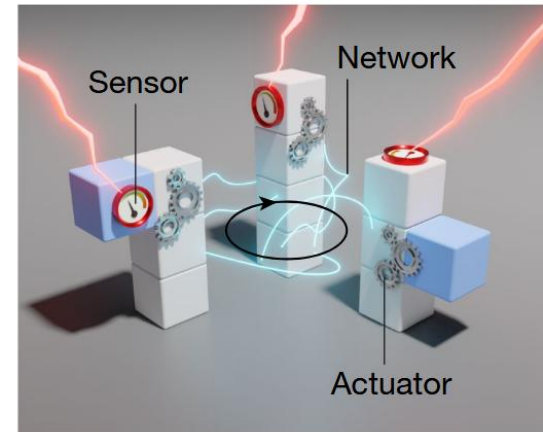
Structural



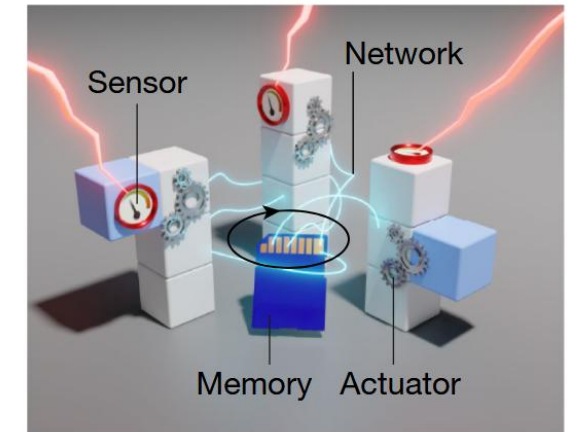
Responsive



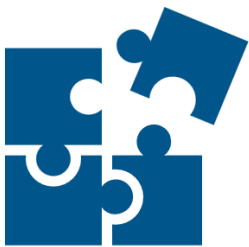
Adaptive



Intelligent

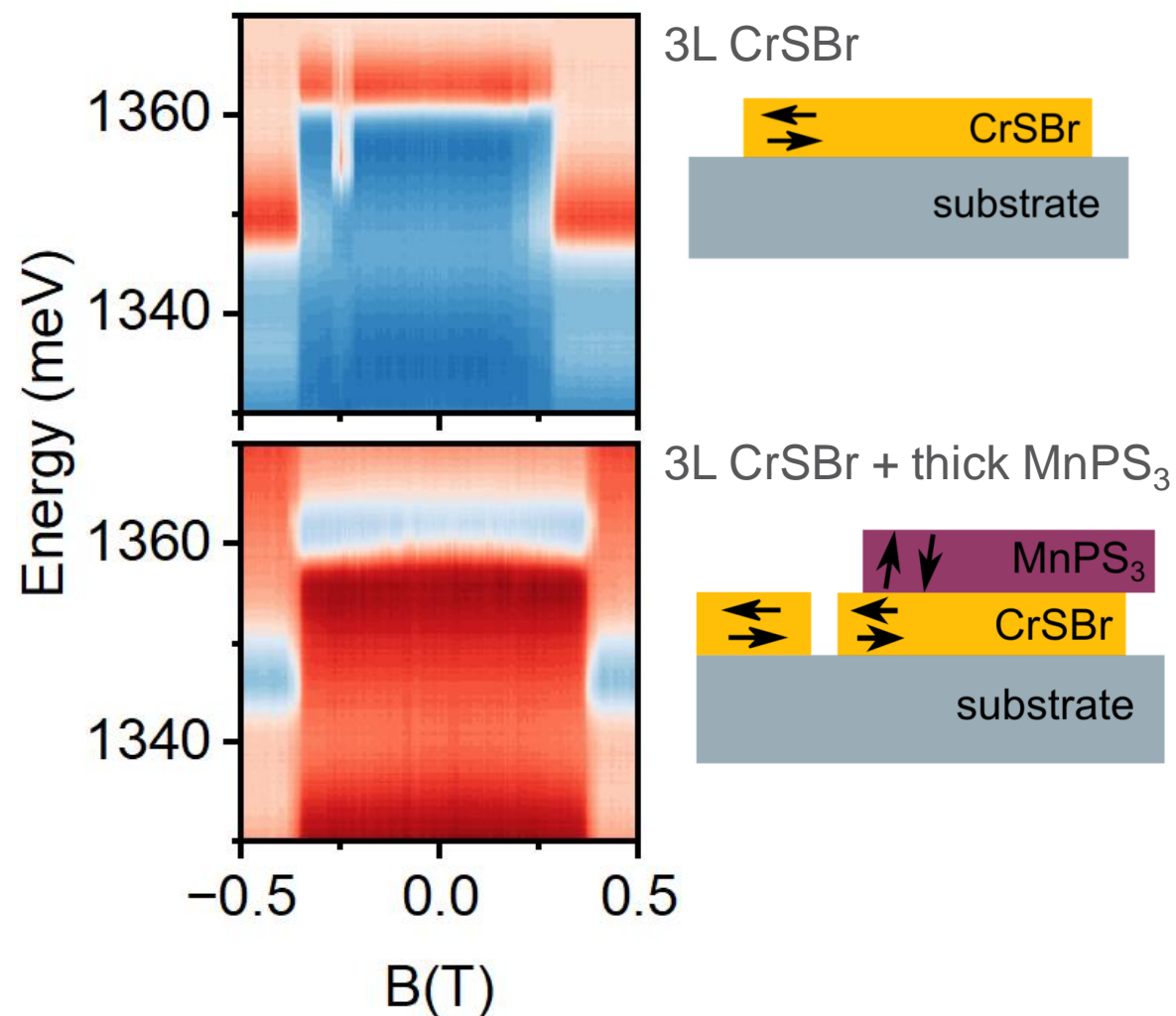


Kaspar *et al.*, "The rise of intelligent matter" (Nature 594, 345–355, 2021)



Outlook: tuning the intermediate regime via interfaces

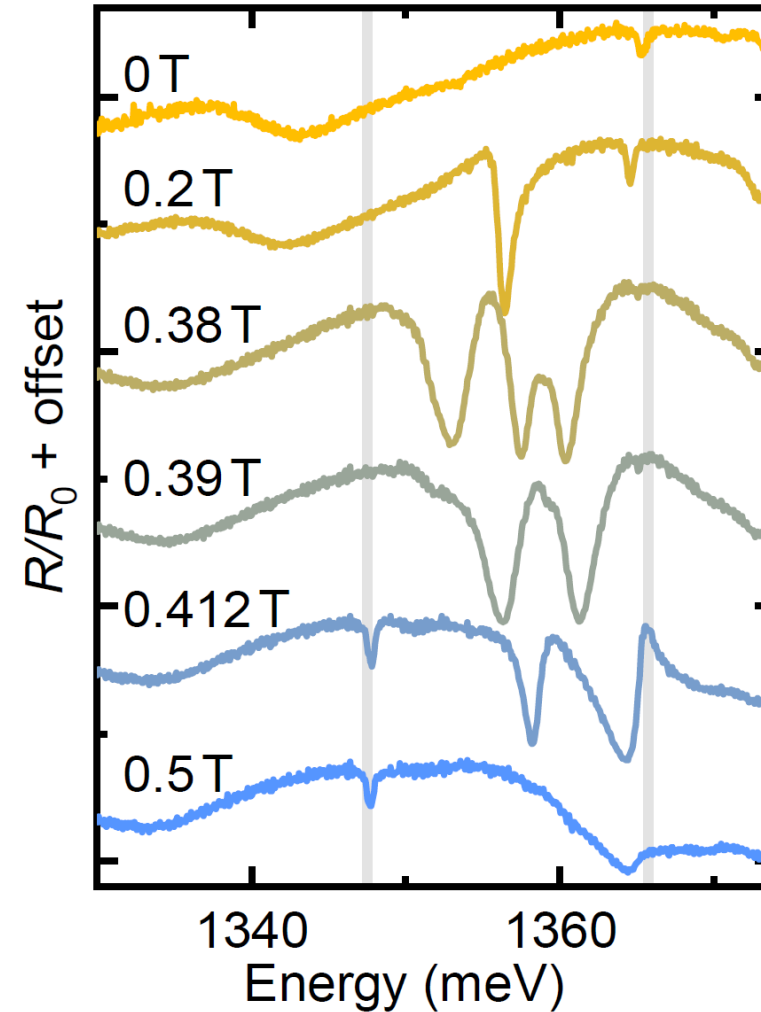
- interfacial coupling may pin and reshape intermediate configurations
- possible stabilization/suppression of iMS (reduced variability)





Take-home message

- **stable intermediate configurations** exist during easy-axis AFM→FM switching.
- **IMS** correspond to layered **out-of-plane domain** stacks formed by layer-by-layer switching.
- **out-of-plane magnetic domain sequence** creates a barcode which can be **readout** by **optics** in non-invasive way
- **interfacial coupling** is a promising handle to tune the intermediate regime (work in progress)

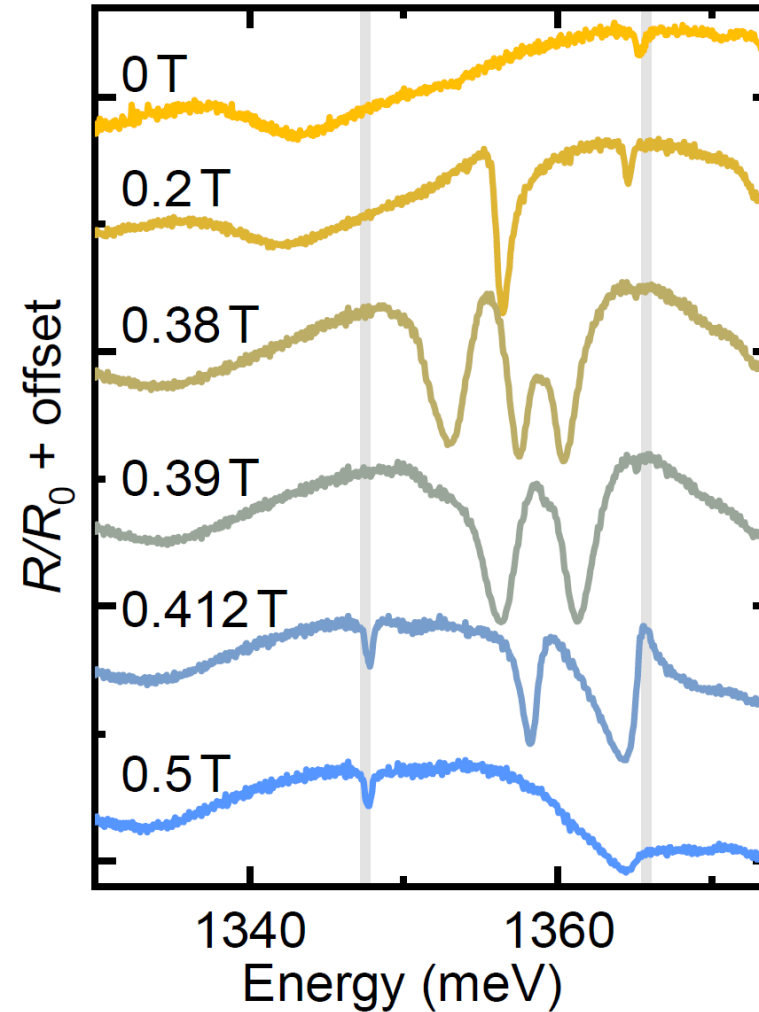




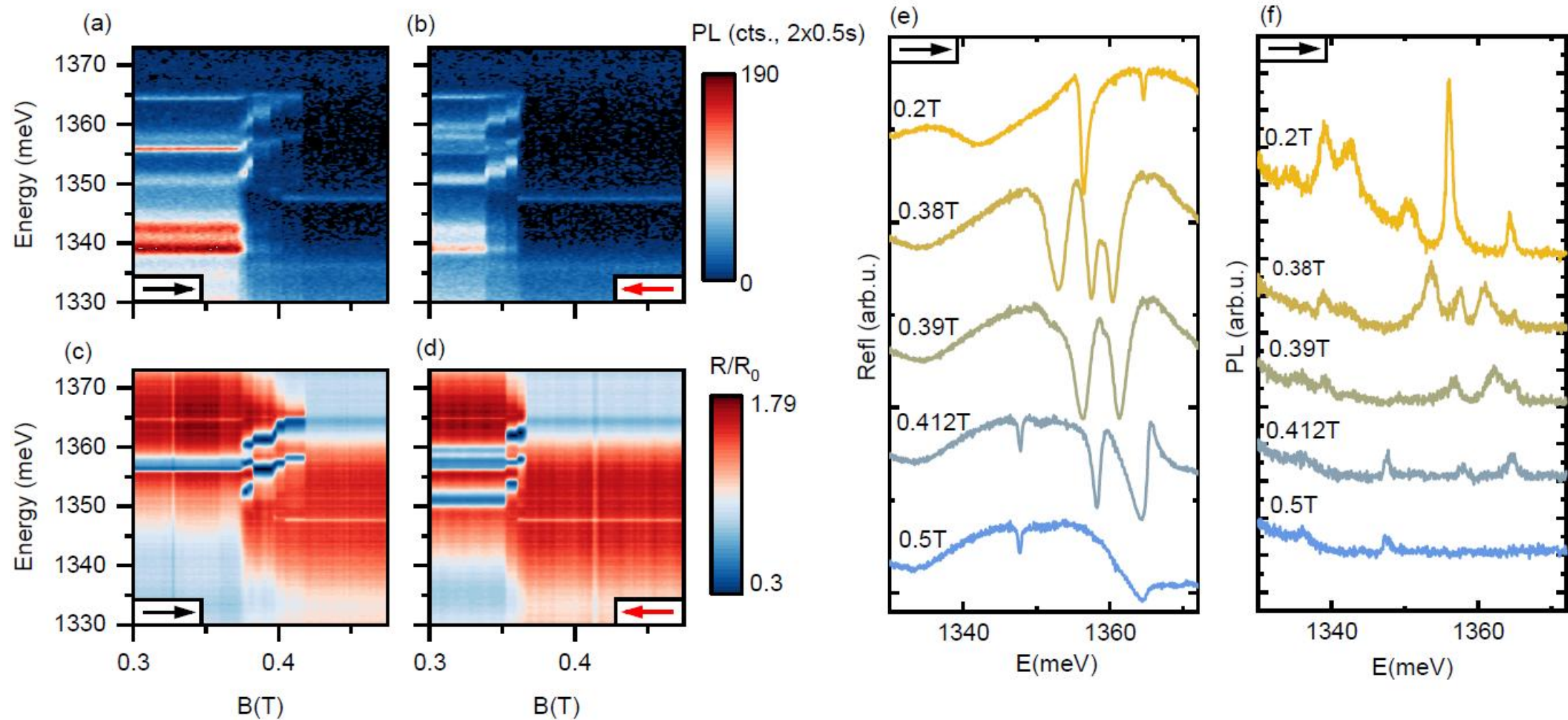
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Thank you

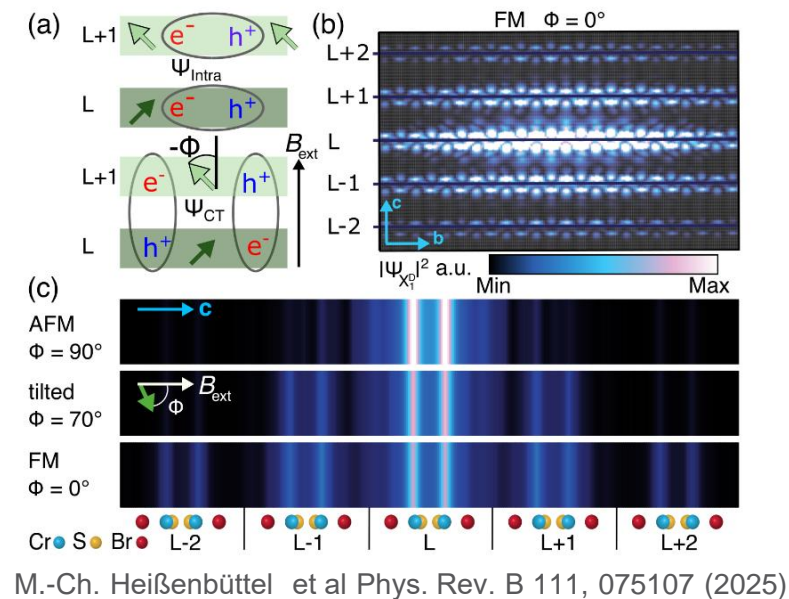


Non-binary switch – PL & Reflectance comparison

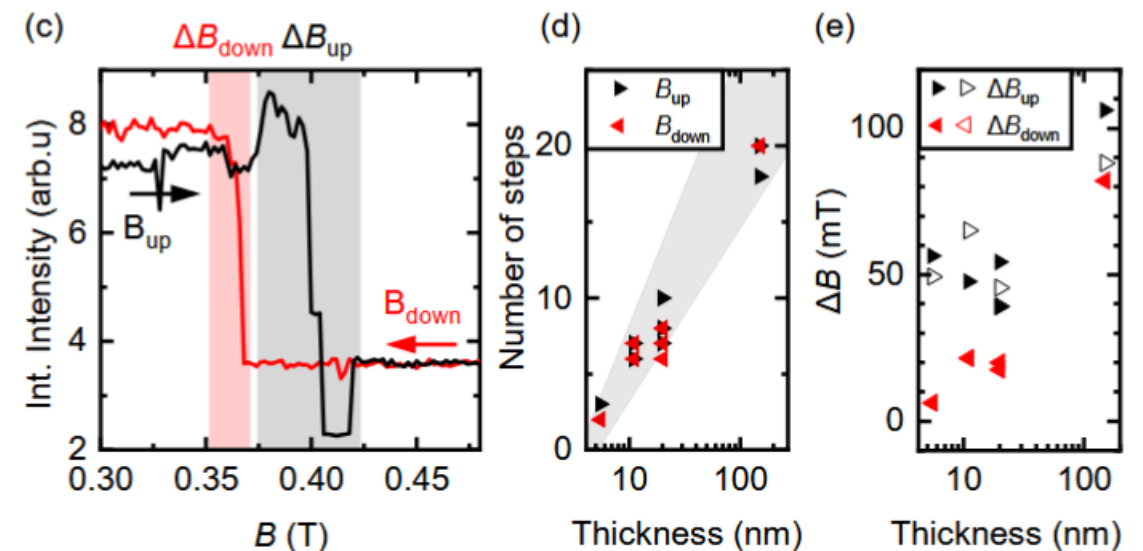


Microscopic origin

Change of the excitonic energy in AFM/FM states:
localization – delocalization of the exciton wavefunction



- Relocalization cost may explain asymmetry (FM→AFM vs AFM→FM). (?)



...or this is just magnetism (how spins are switching, spin pinning...)

TMM technicalities

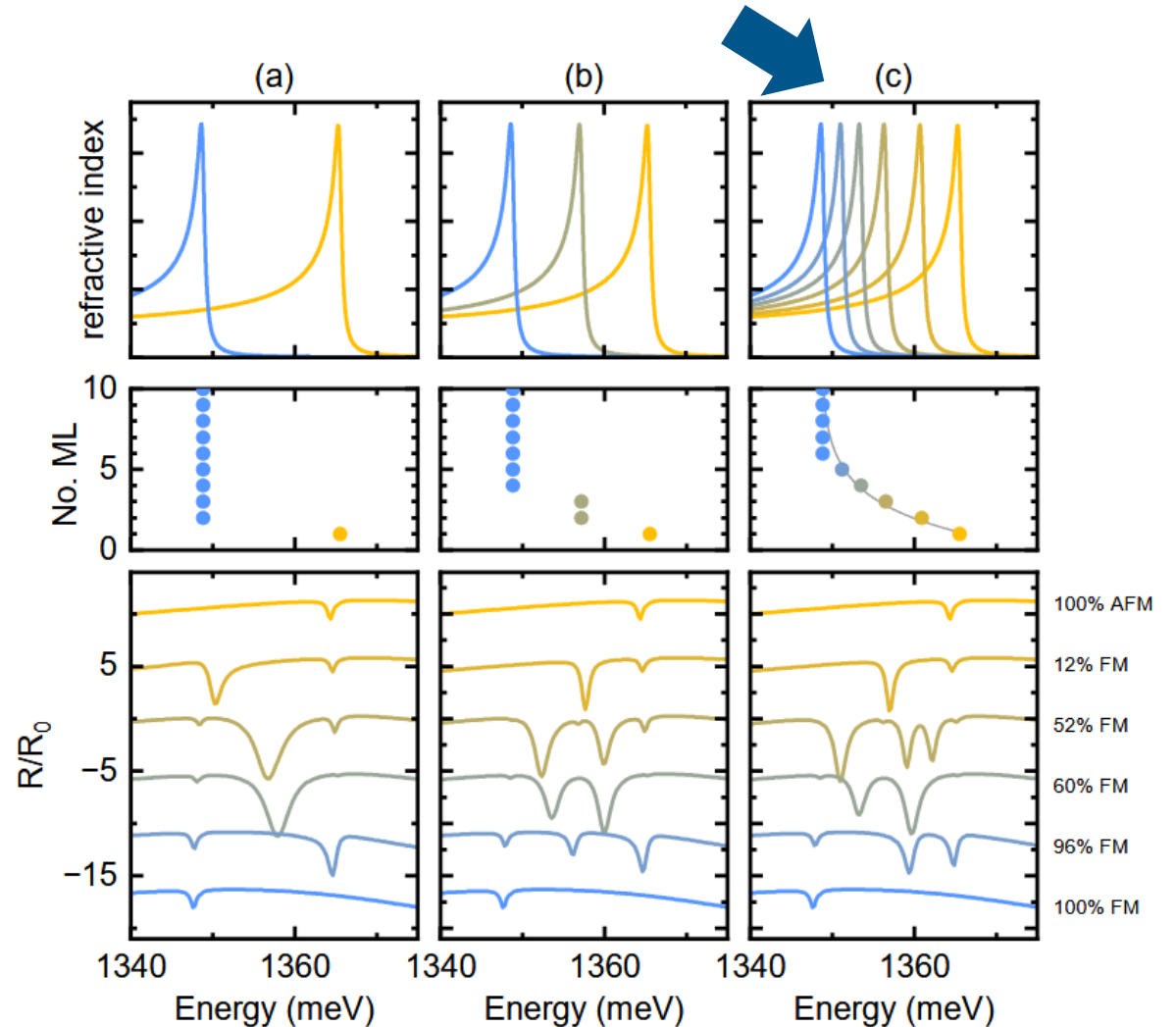
We assume:

- Monolayer have the same n and $k(E)$ as AFM bulk
- FM state (thick flake, in magnetic field) gives minimal energy for FM
- Thinner FM layers – something between

Issues:

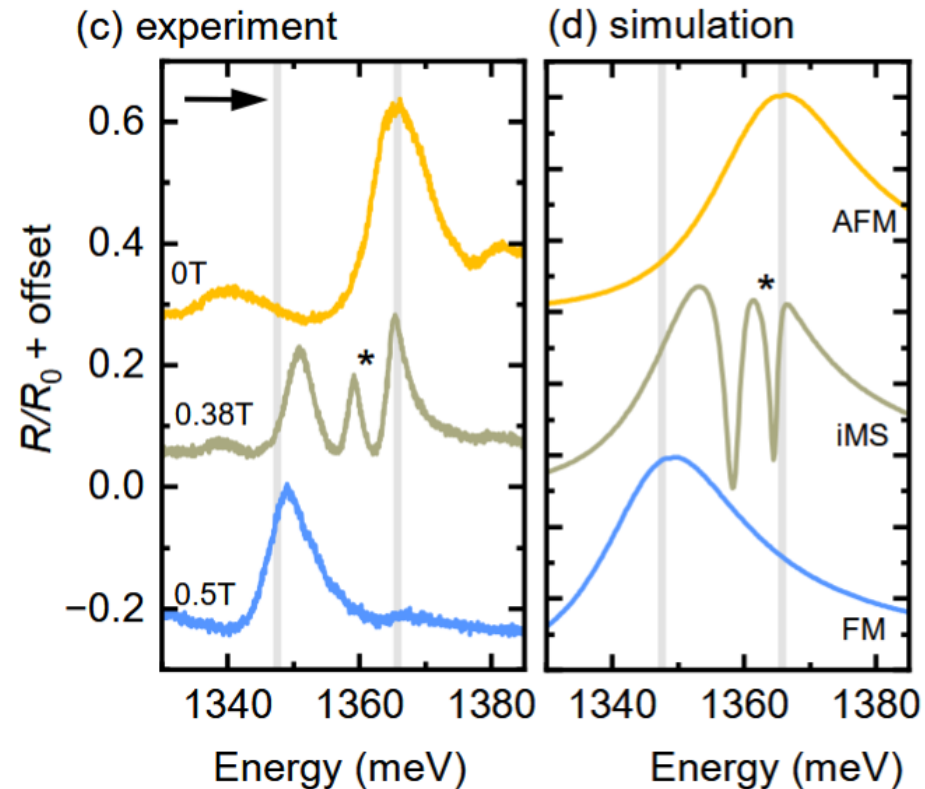
- Proper parameters for $n, k(E)$
- Phase
- Magnetism
- Is confinement enough?

energy change due to confinement in the thinner layer of CrSBr



Phase change

TMM as it is (this is in the paper)



TMM + phase (why it fits better?!)

