CrSBr: doping and magnetic control

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Outline

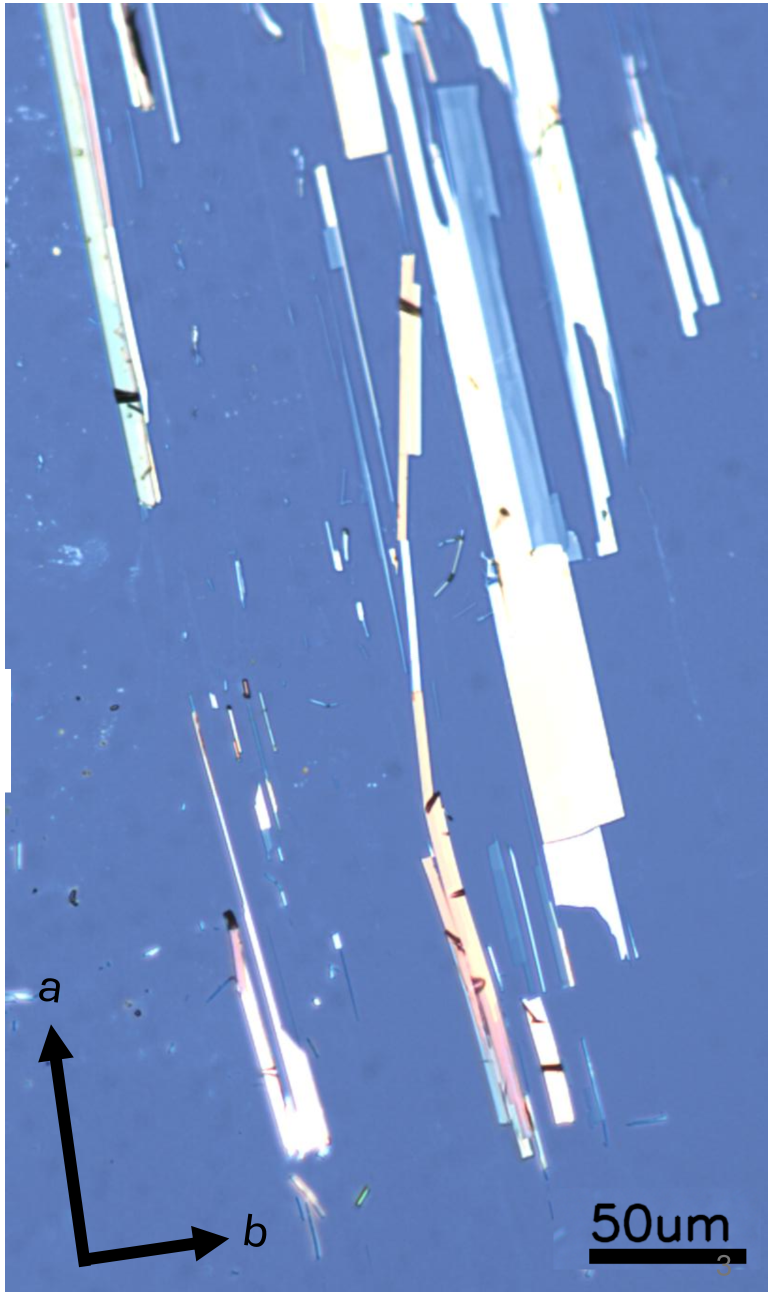
Motivation

Optical measurements

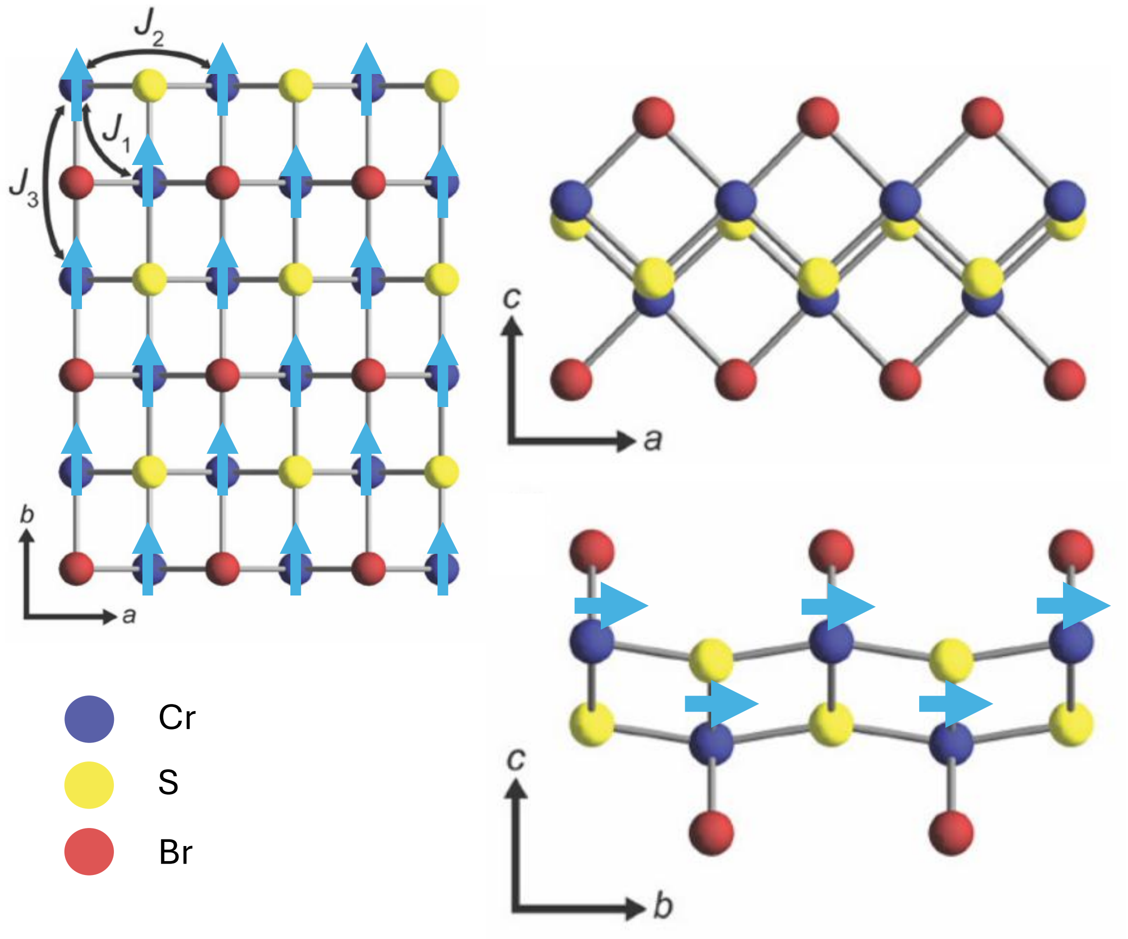
Conclusions and outlook

Properties of CrSBr

- 2D material
- Direct gap semiconductor
- A-type antiferromagnet
- Strong light-matter coupling
- Highly anisotropic



Properties of CrSBr: magnetism

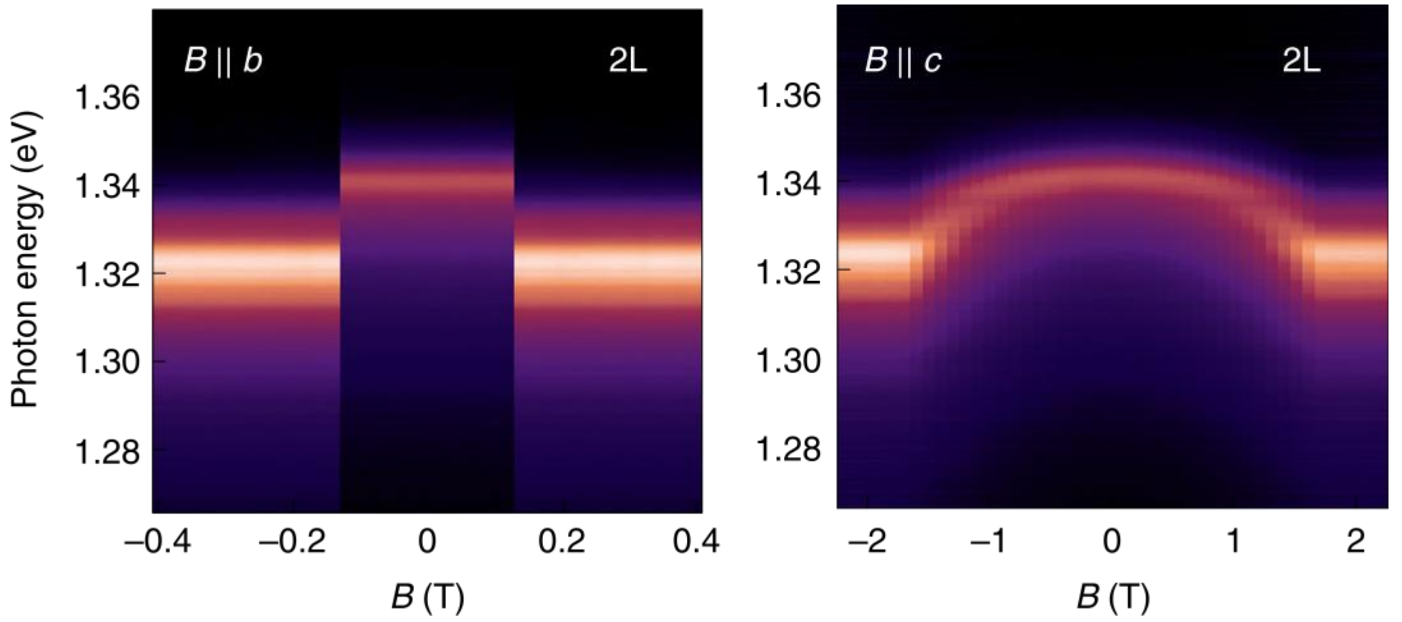


Michael E. Ziebel, Margalit L. Feuer, Jordan Cox, Xiaoyang Zhu, Cory R. Dean, and Xavier Roy
Nano Letters **2024** *24* (15), 4319-4329

- Intralayer ferromagnetism
- Interlayer antiferromagnetism
  $T_N \approx 140K$

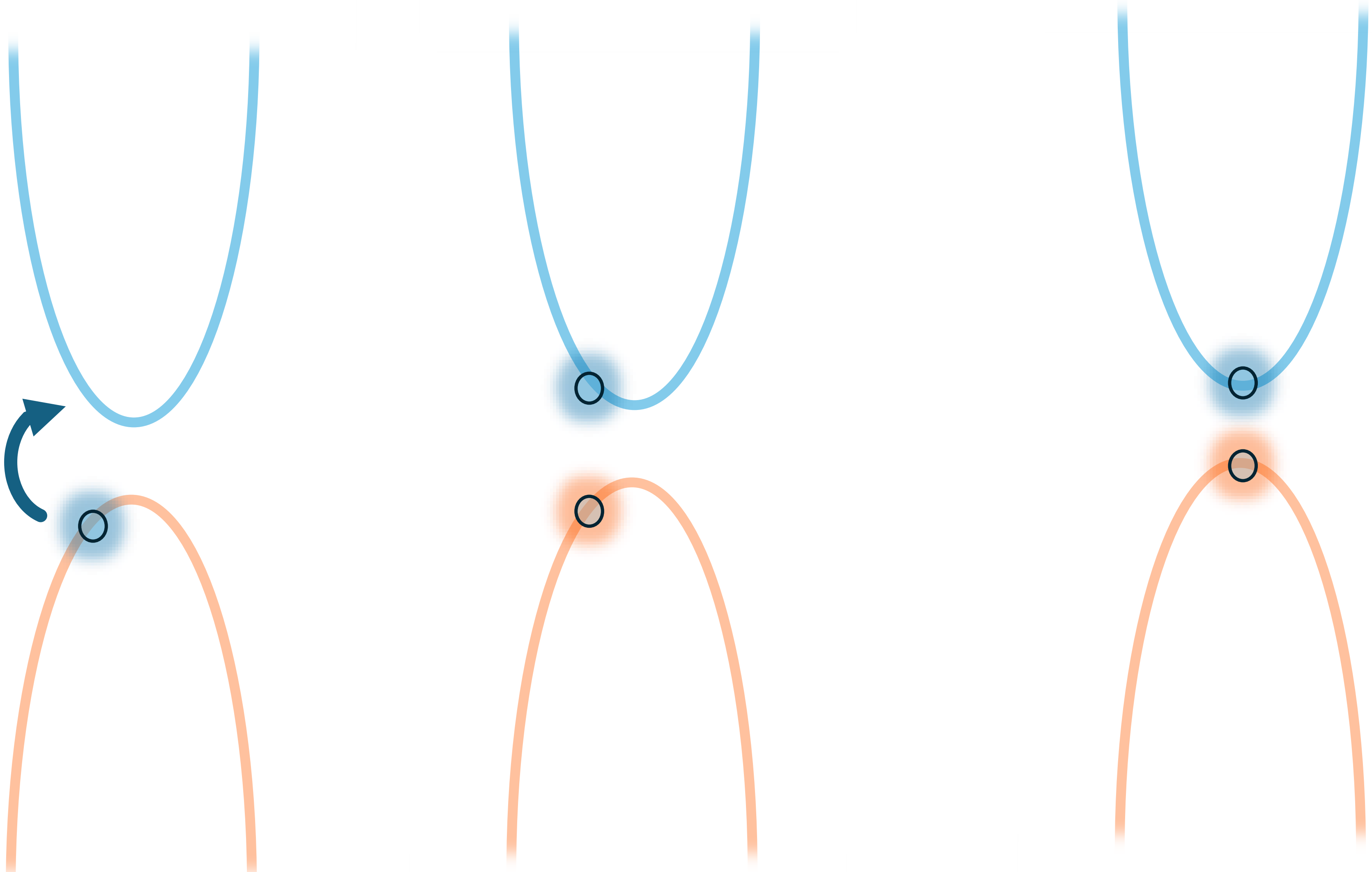
b	a	c
Easy	**Intermediate**	**Hard**

Exciton energy coupled to magnetic order

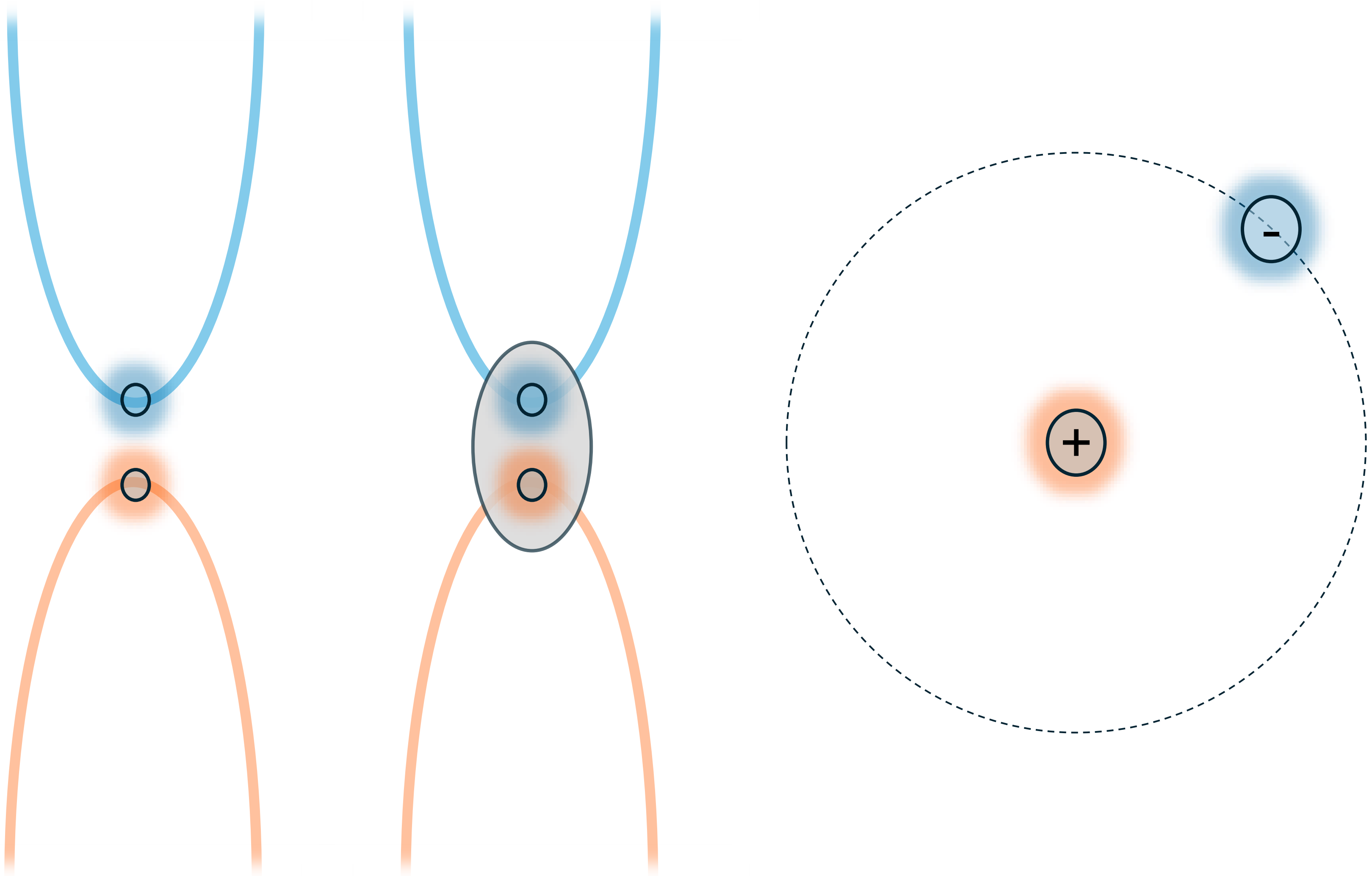


N. P. Wilson et al., Nat. Mater. 20, 1657 (2021)

Reminder: excitons



Reminder: excitons



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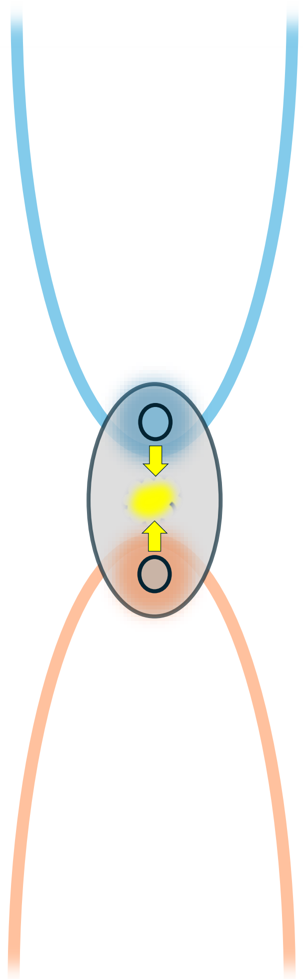
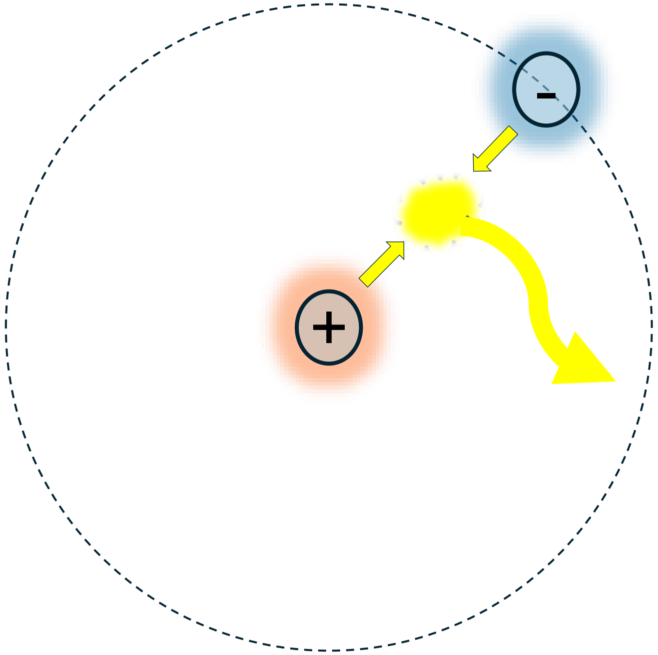
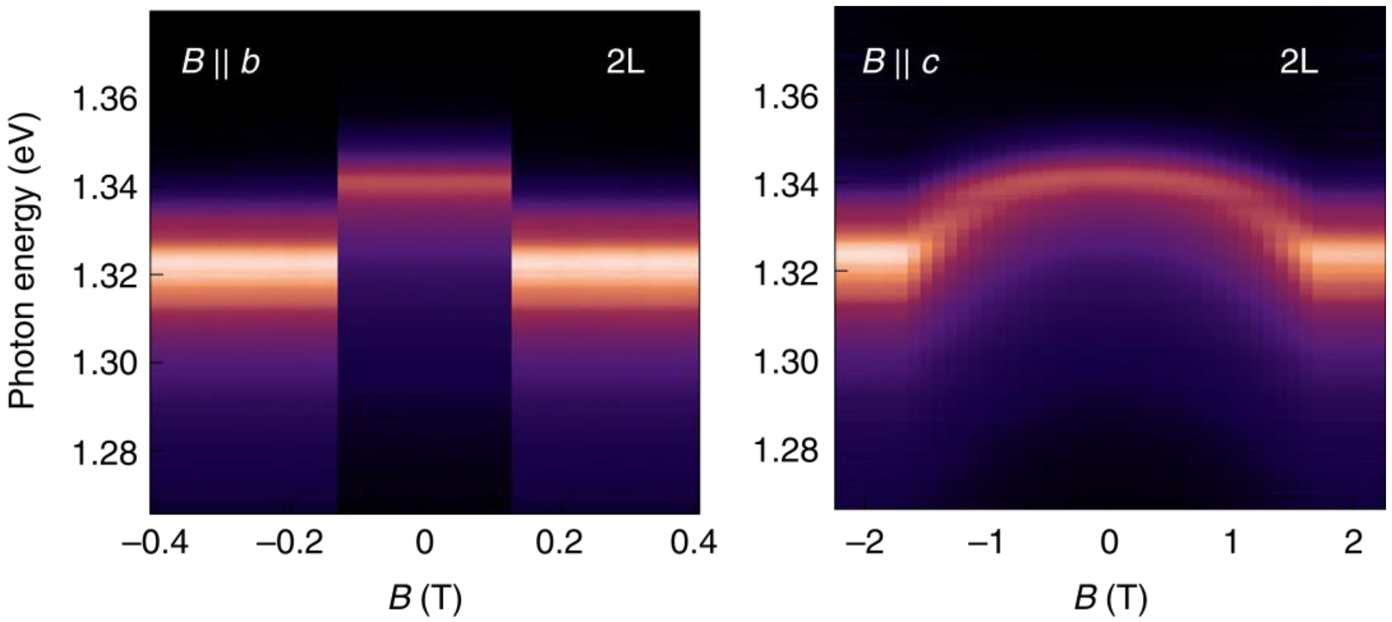


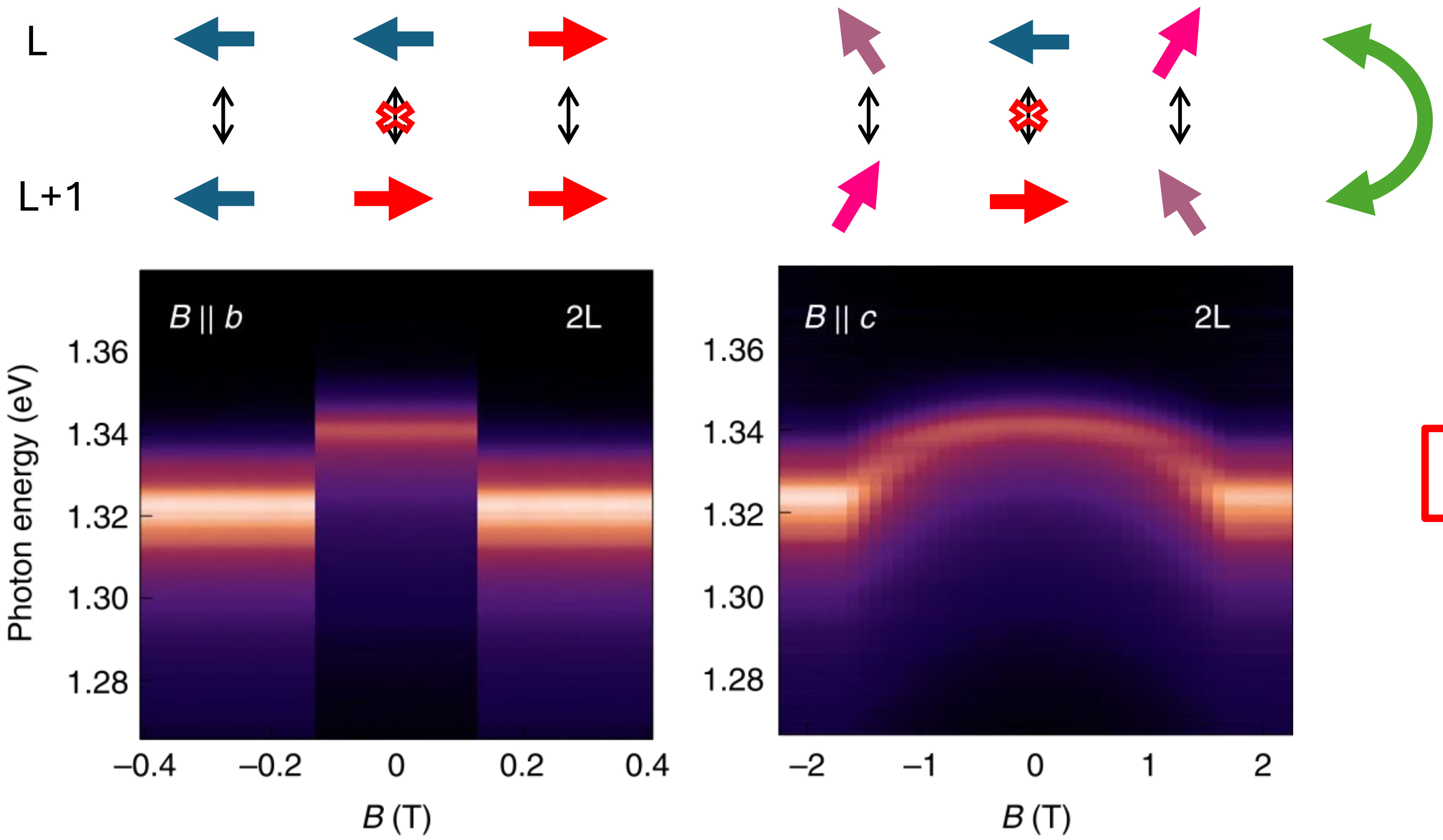


Exciton energy coupled to magnetic order



N. P. Wilson et al., Nat. Mater. 20, 1657 (2021)

Exciton energy coupled to magnetic order

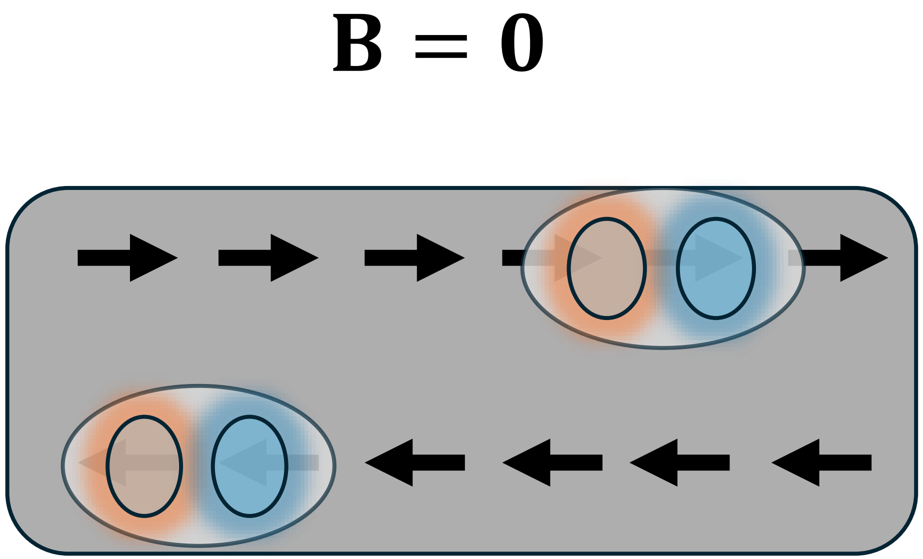


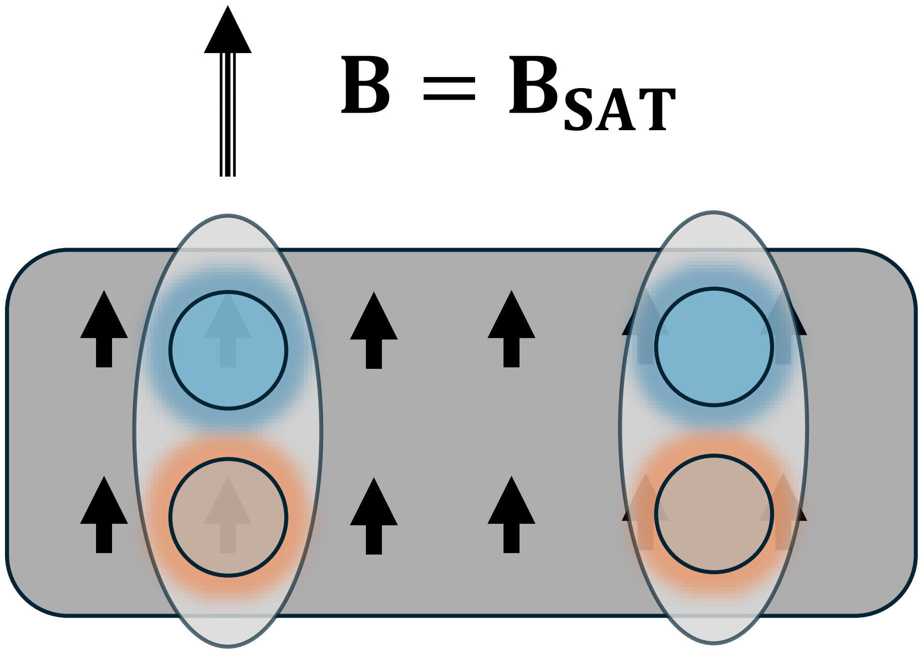
$$T_{L;L+1} \propto \langle S_L | S_{L+1} \rangle = \cos\left(\frac{\theta}{2}\right) \propto B$$

$$E_X = E_0 - \alpha |T_{L;L+1}|^2$$

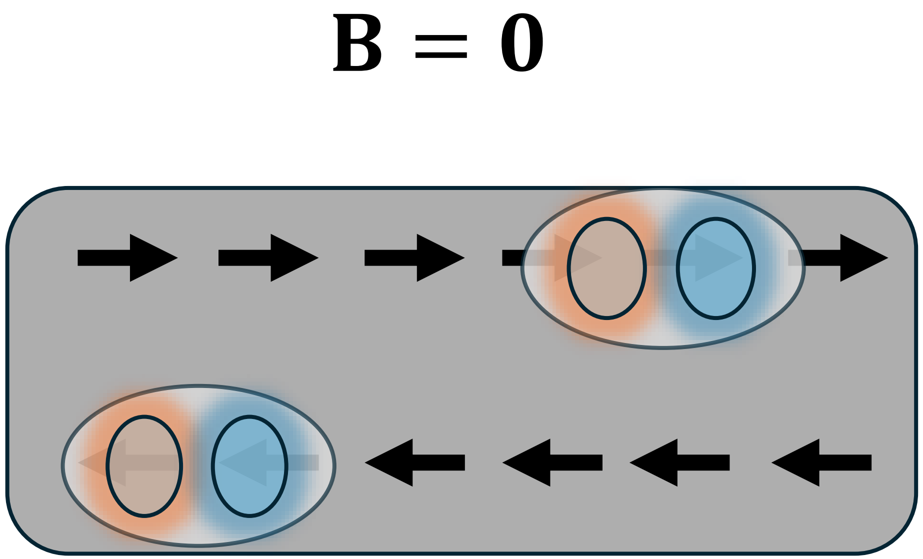
N. P. Wilson et al., Nat. Mater. 20, 1657 (2021)

How to verify this?

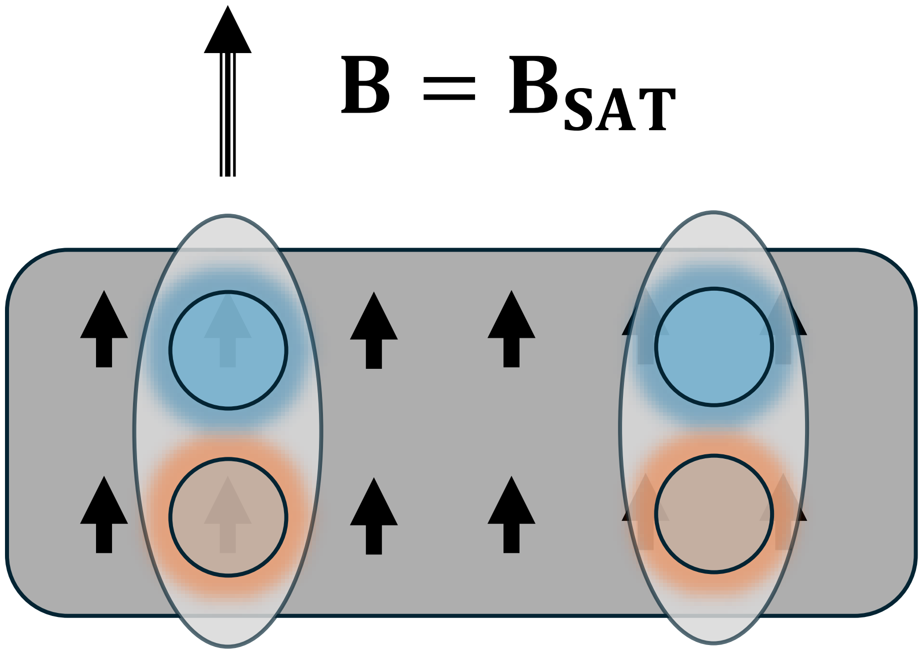




How to verify this?



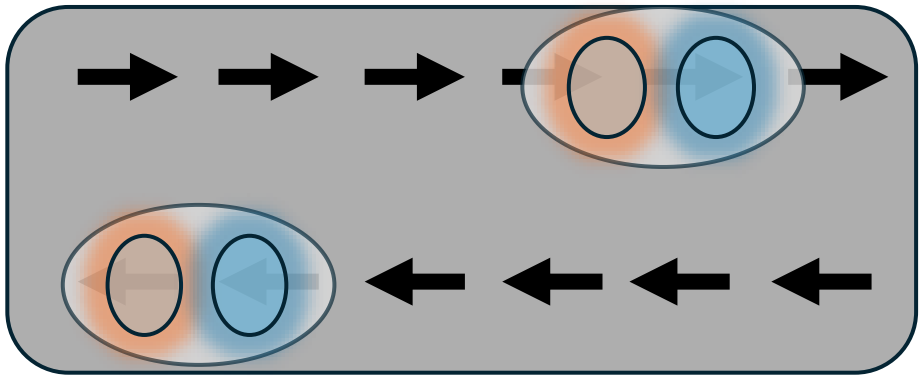
Small polarizability



Large polarizability

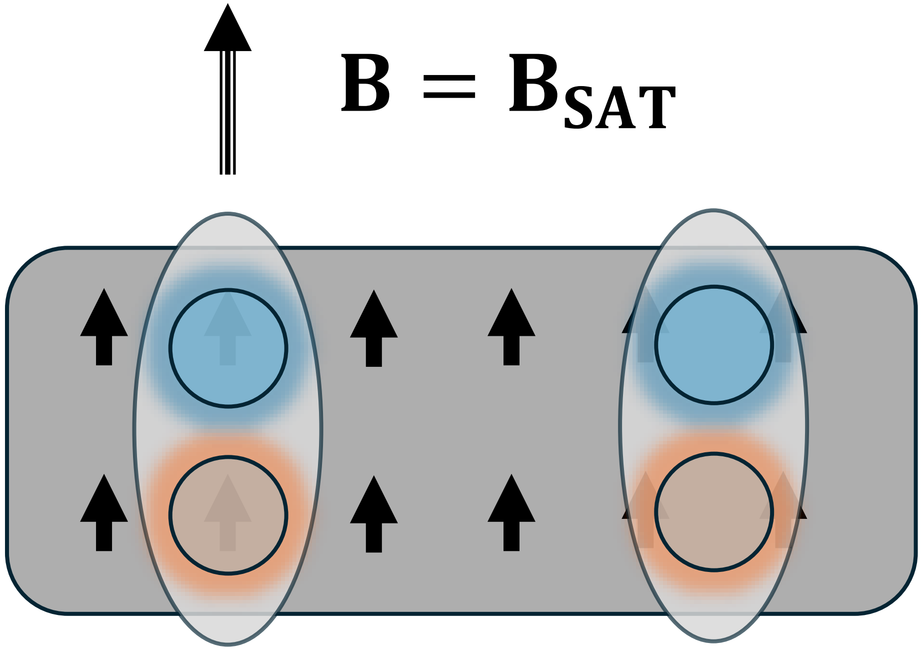
How to verify this?

$\mathbf{B = 0}$



Small polarizability

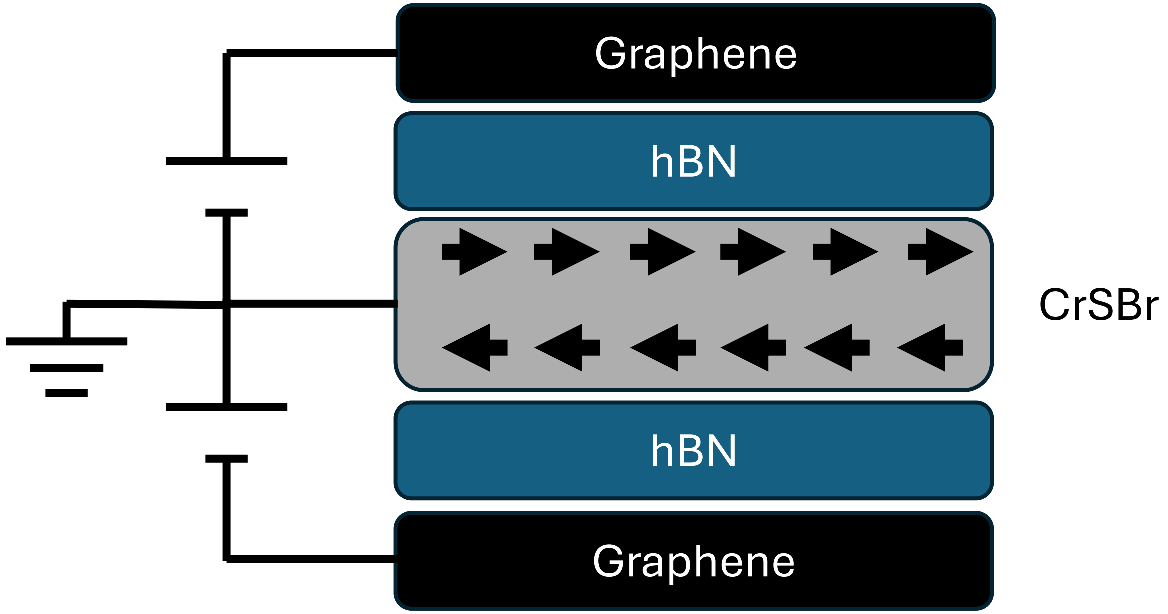
$\mathbf{B = B_{SAT}}$



Large polarizability

Apply E-field!

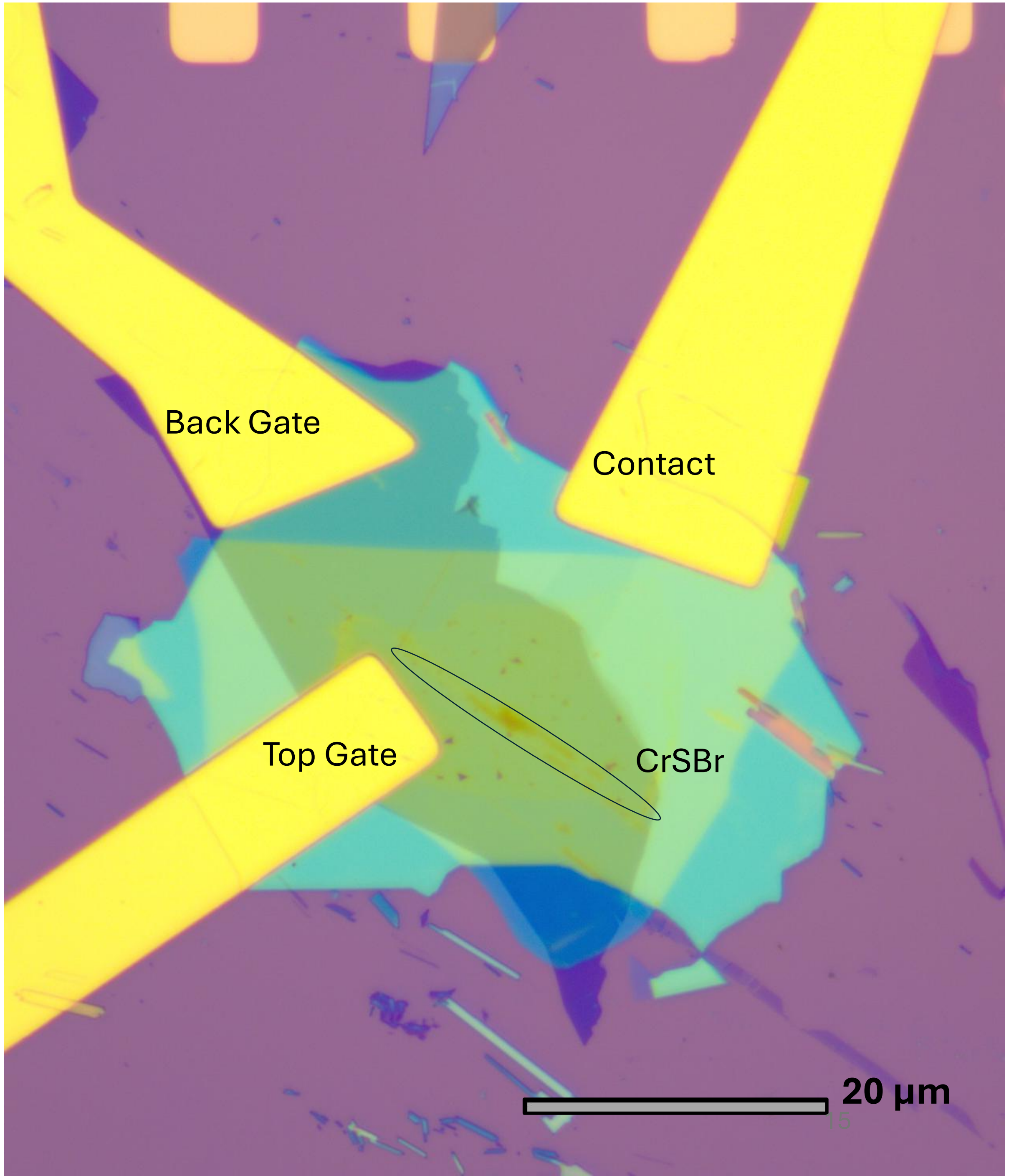
With such a device



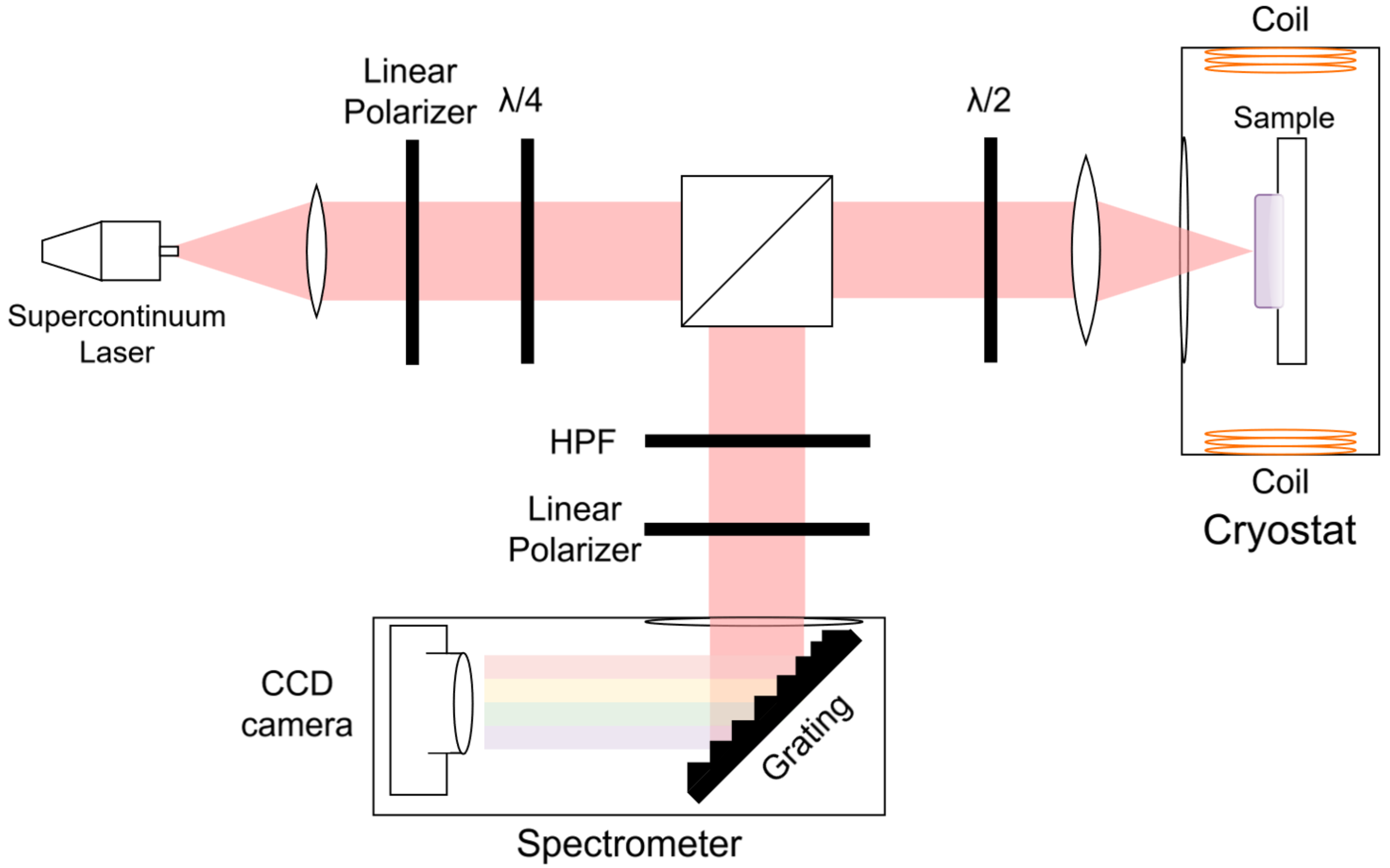
First CrSBr device

- Backgate not working

- Doping possible with contact and top gate

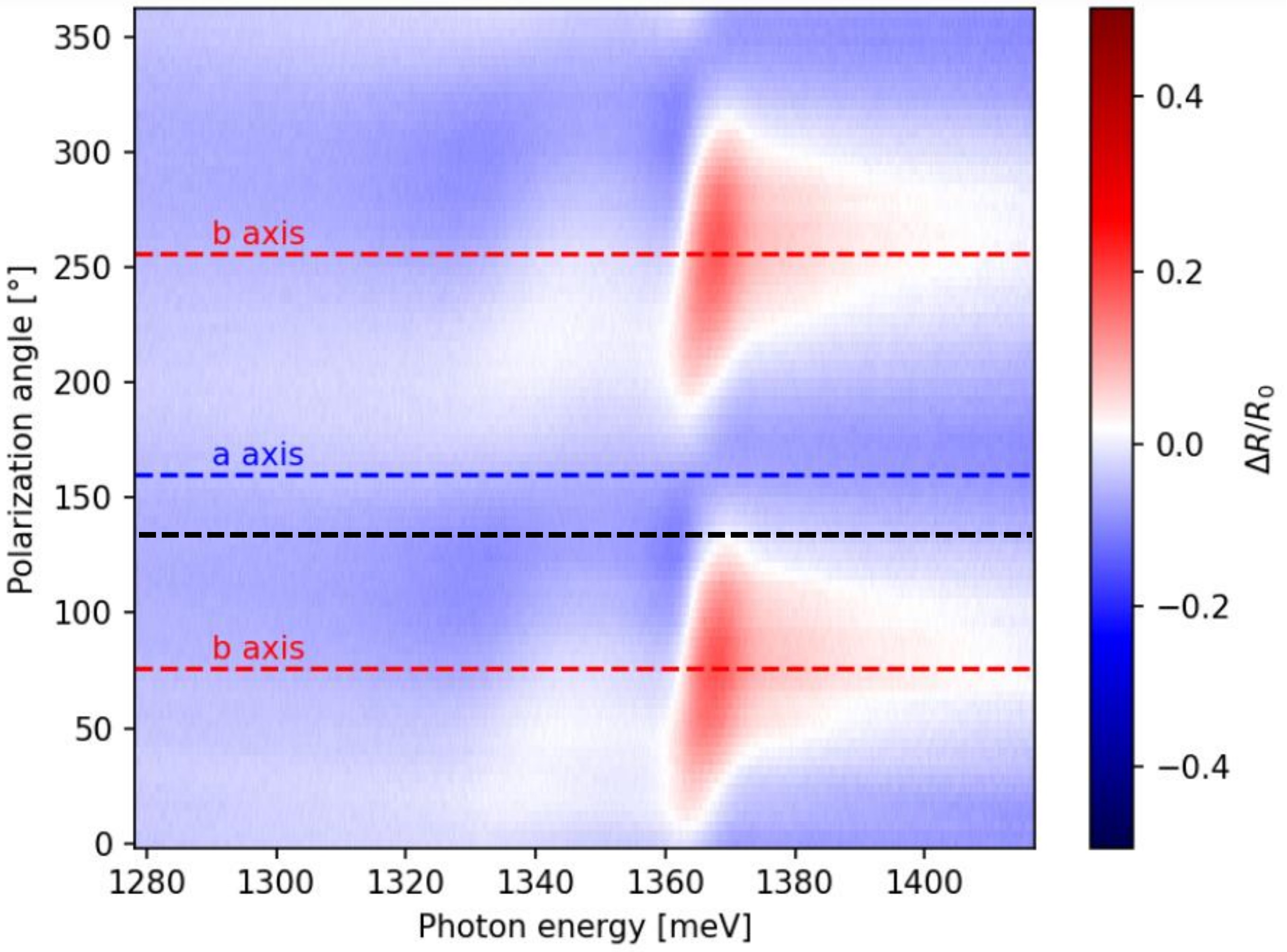


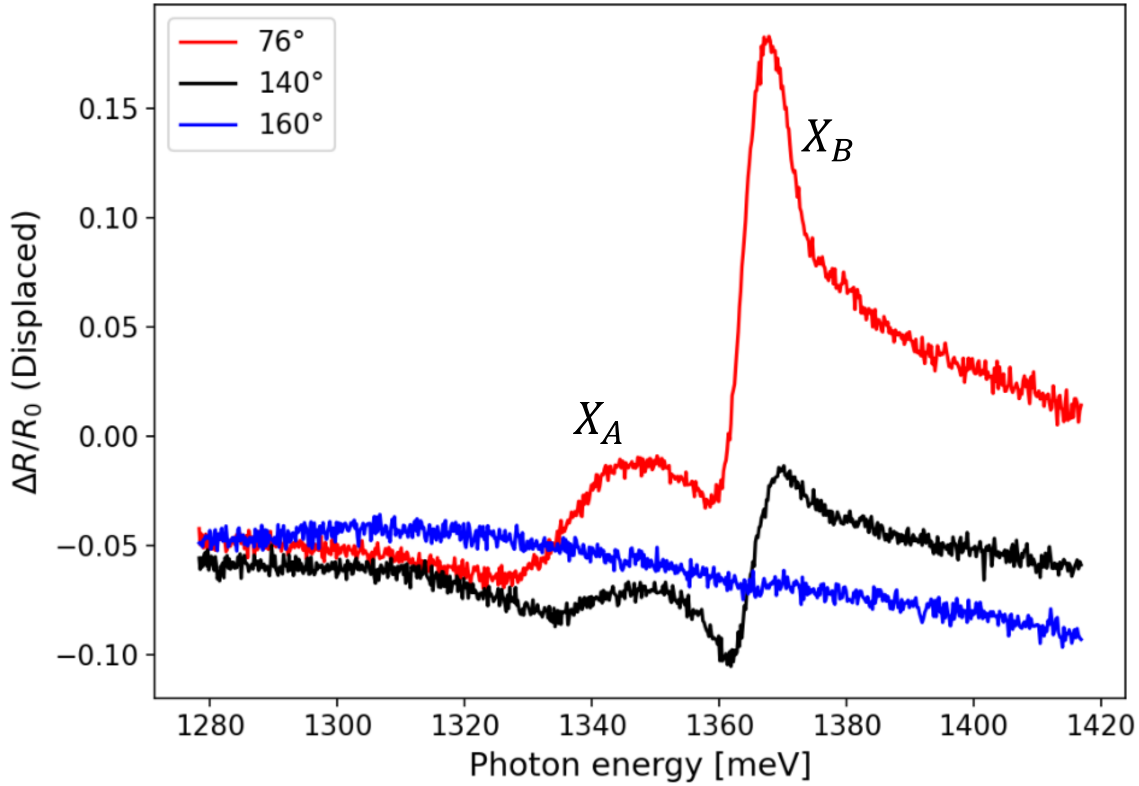
Polarization-Resolved Differential Reflectance Spectroscopy



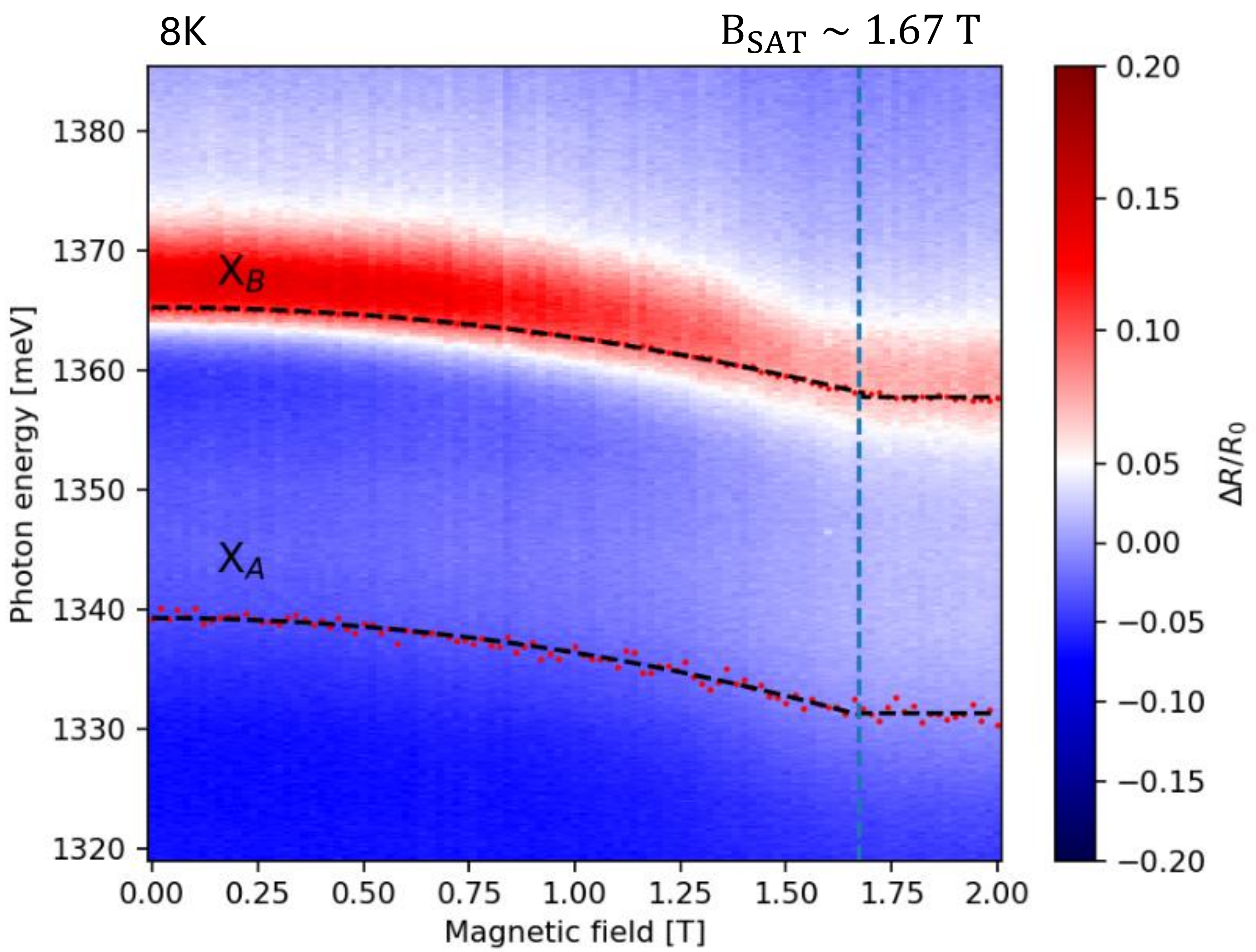
- Measure $\frac{\Delta R}{R_0} \propto Im(\chi(\omega))$
- Study exciton resonance

Complete linear polarization





Exciton energy vs magnetic field



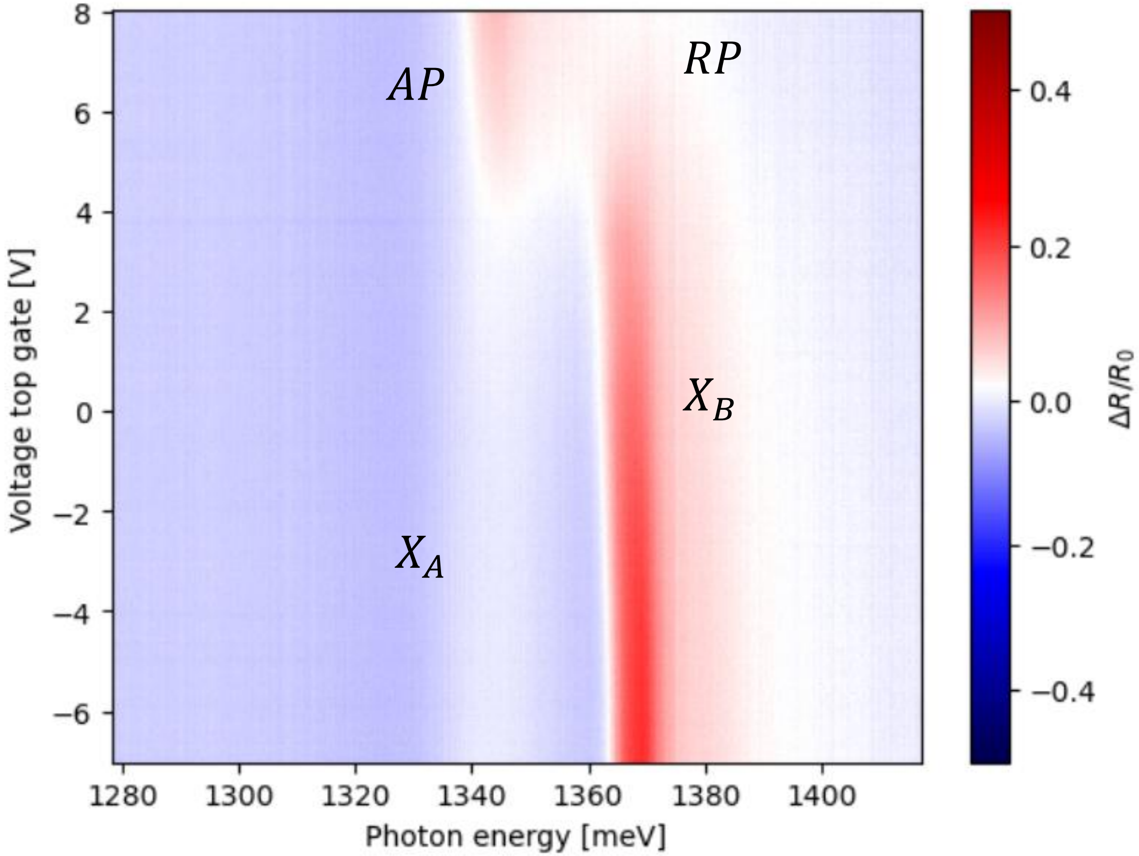
$$E_X = E_0 - \alpha B^2$$

$$\alpha(X_A) \sim 2.9\, meV \cdot T^{-2}$$

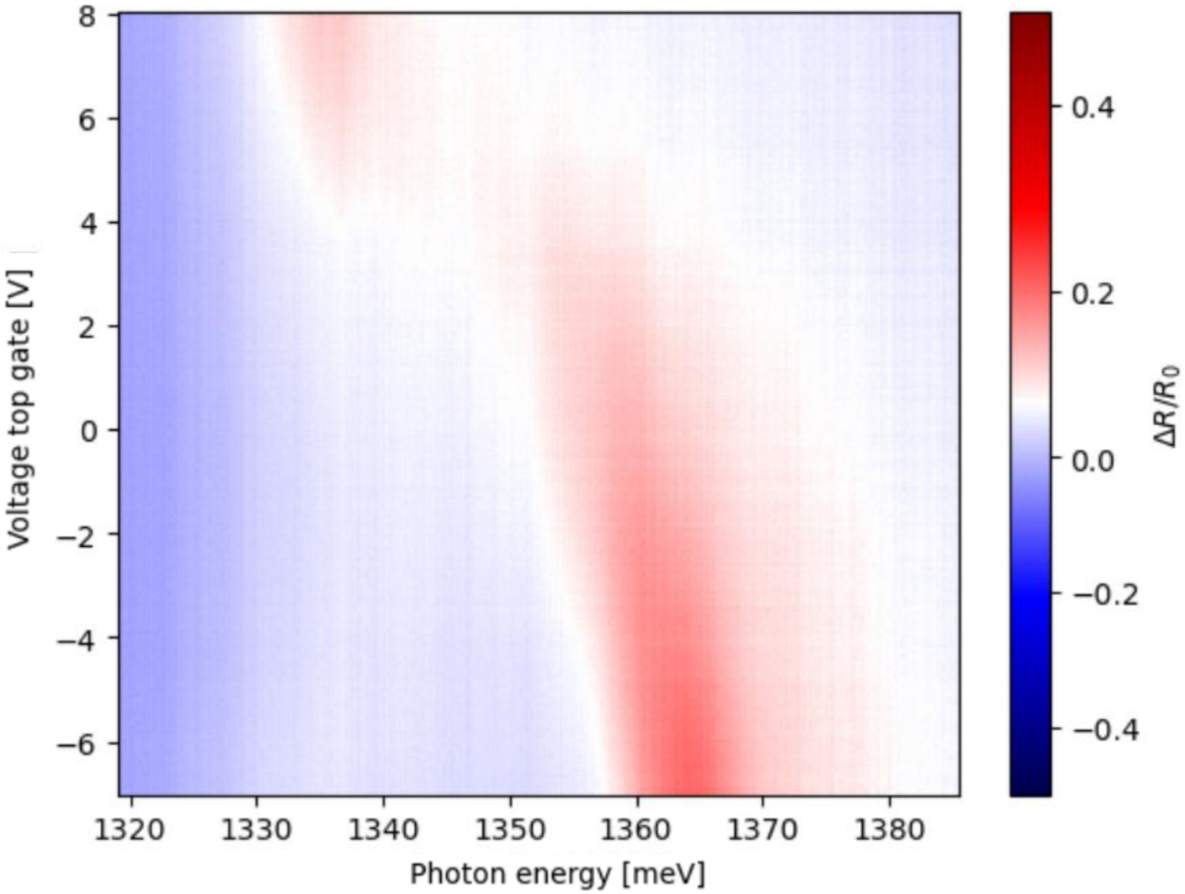
$$\alpha(X_B) \sim 2.5\, meV \cdot T^{-2}$$

Exciton energy vs top gate voltage

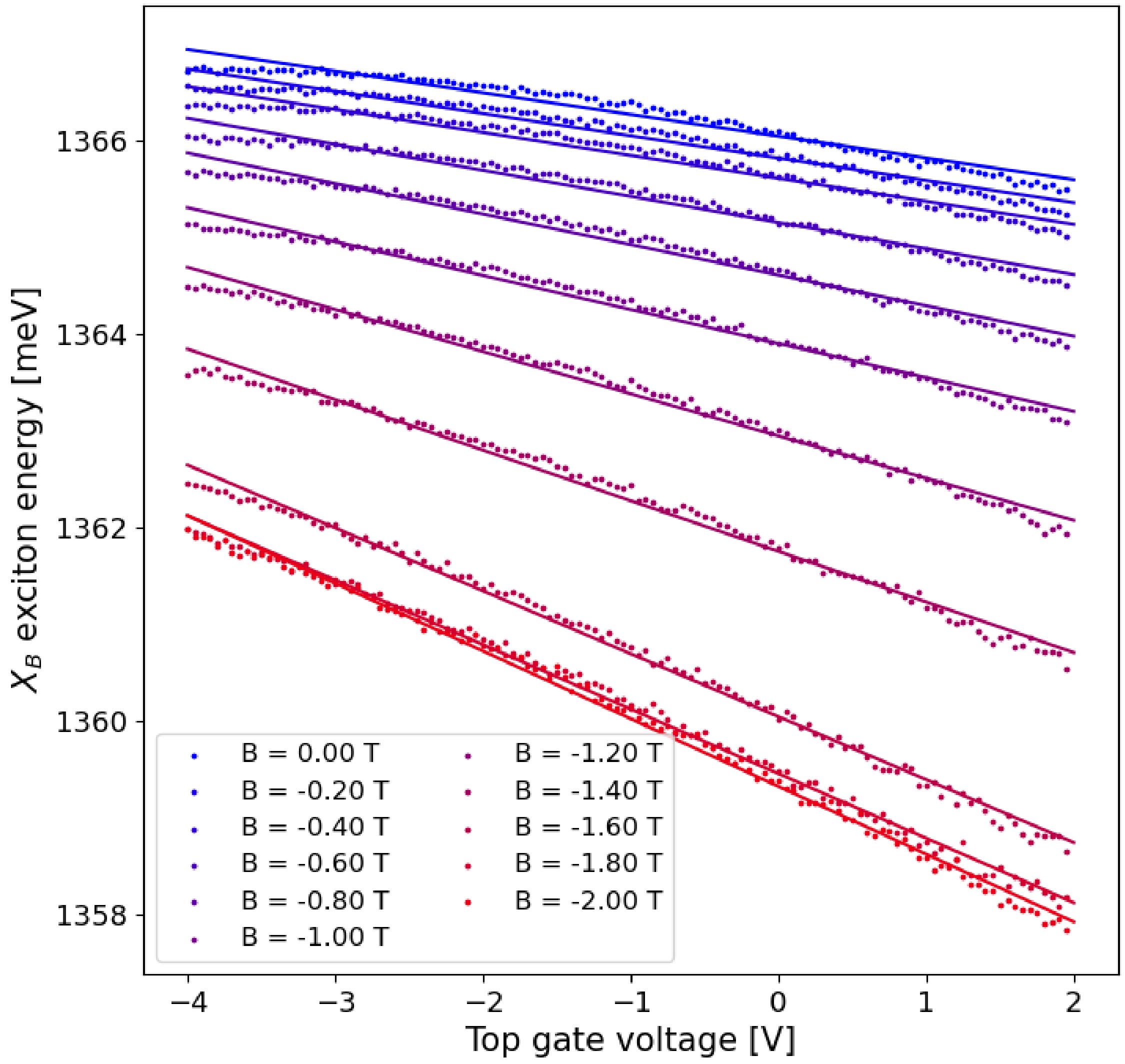
0T 8K



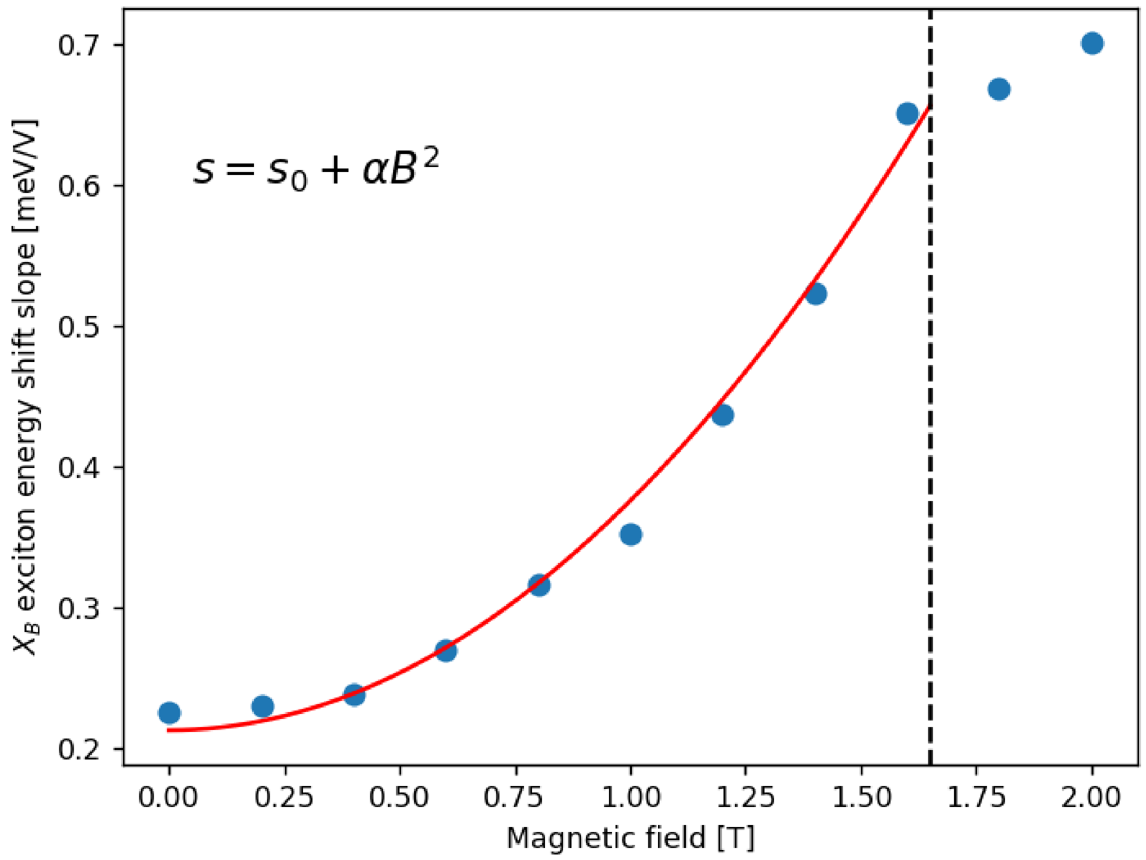
2T 8K



Slope for different magnetic fields

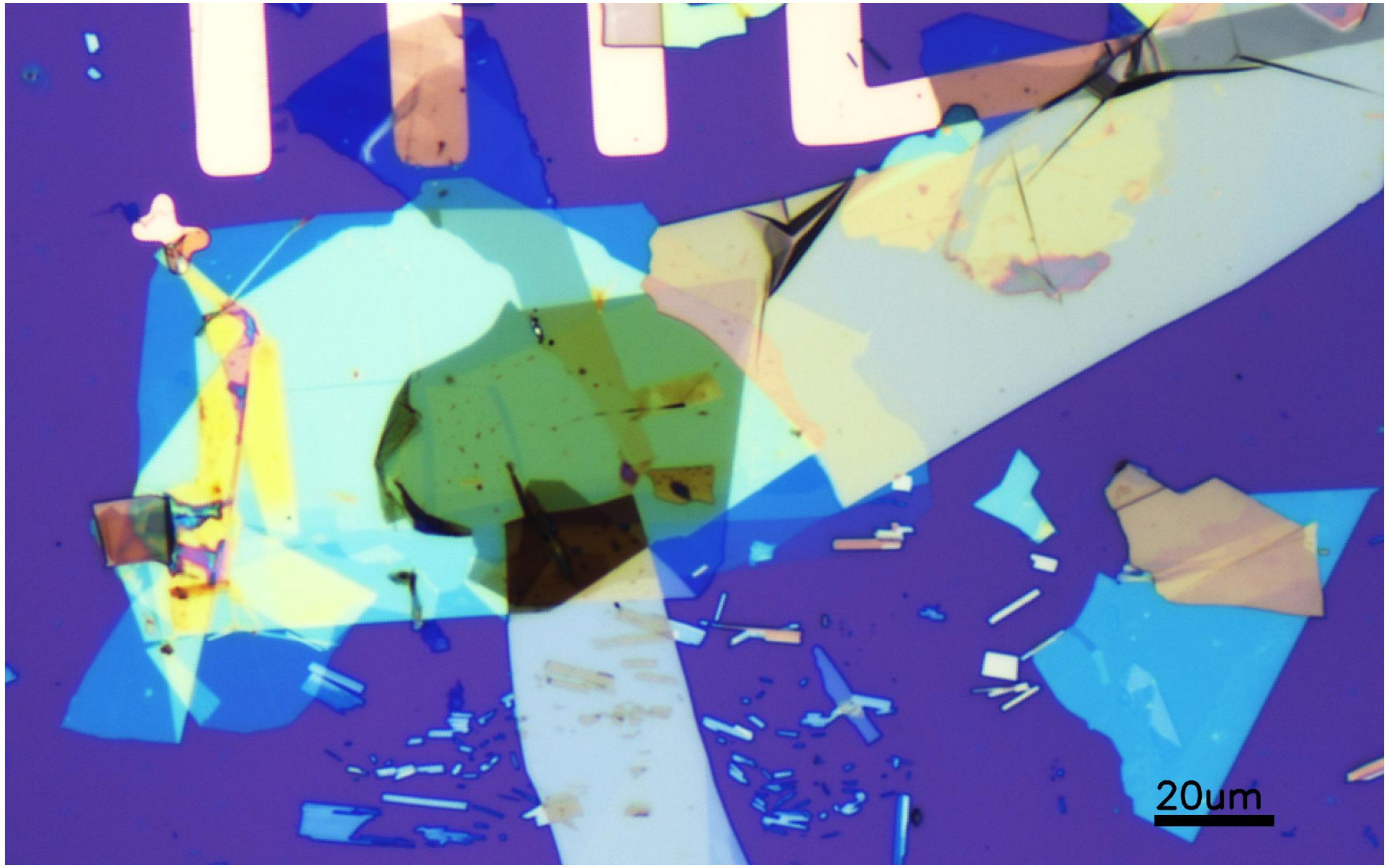


B_{SAT} ~ 1.65 T

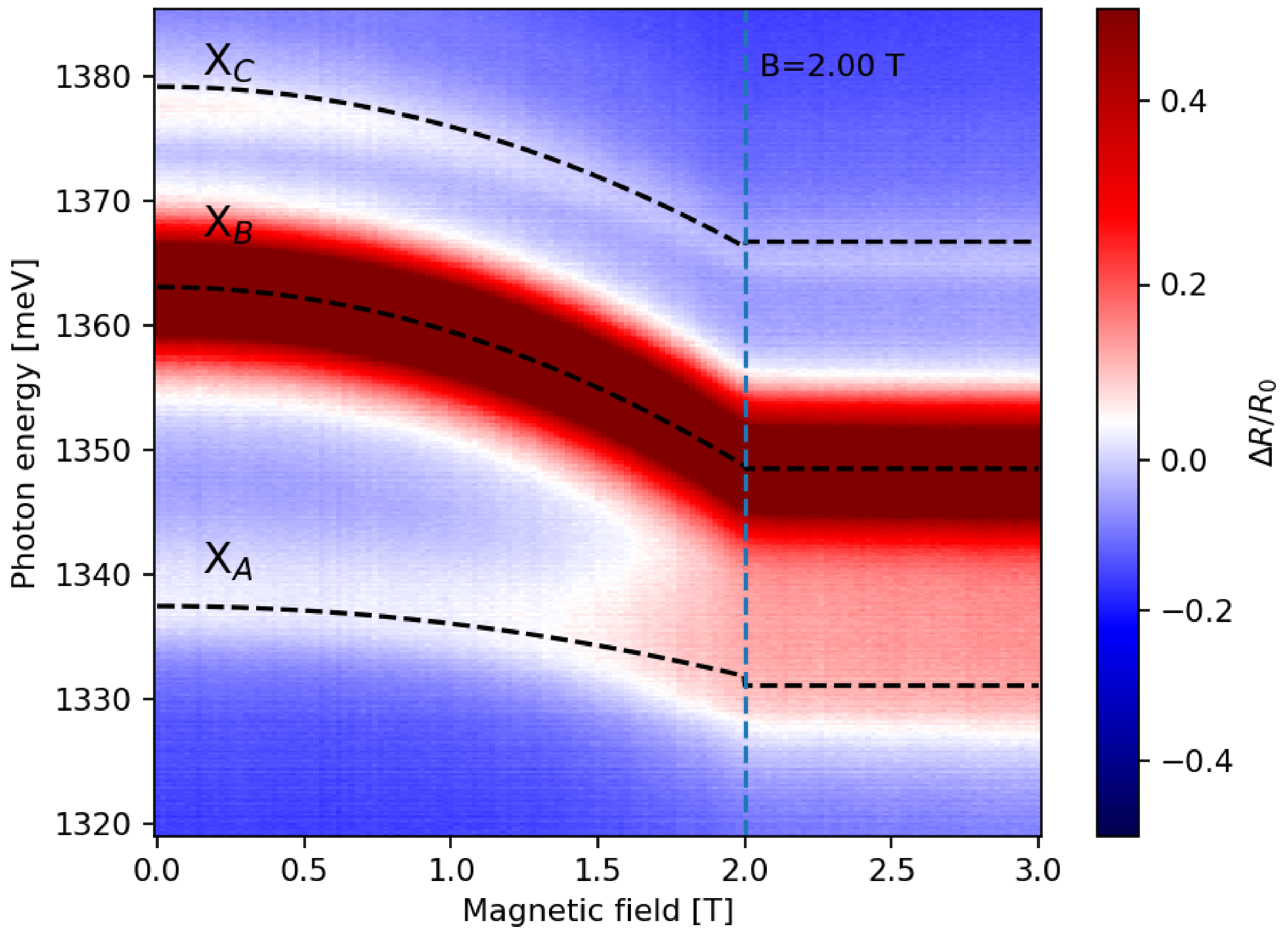


Second device

- 8 nm thick CrSBr
- Multiple graphite patches to avoid lithography



Exciton energy vs magnetic field



$$E_X = E_0 - \alpha B^2$$

First device

$$\alpha(X_A) \sim 2.9\, meV \cdot T^{-2}$$

$$\alpha(X_B) \sim 2.5\, meV \cdot T^{-2}$$

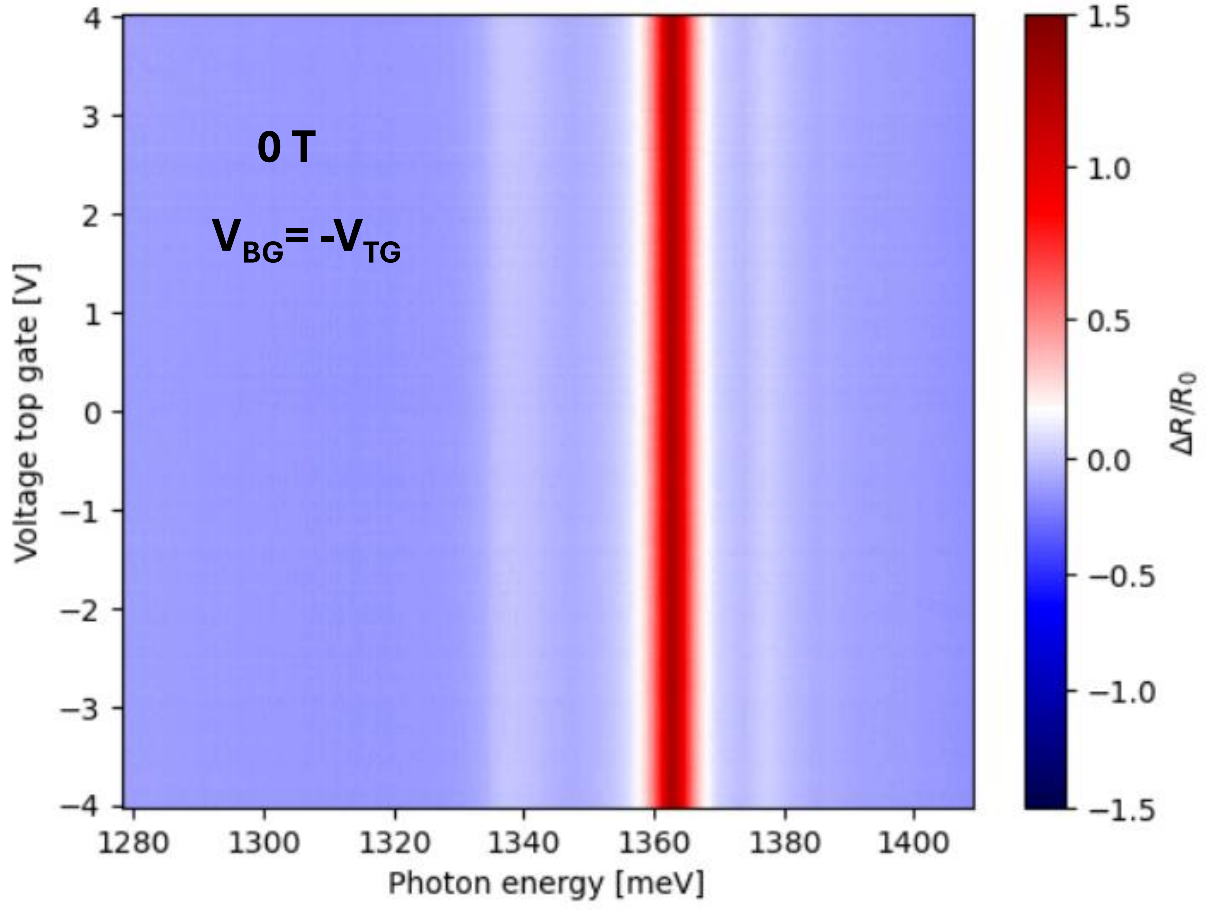
This device

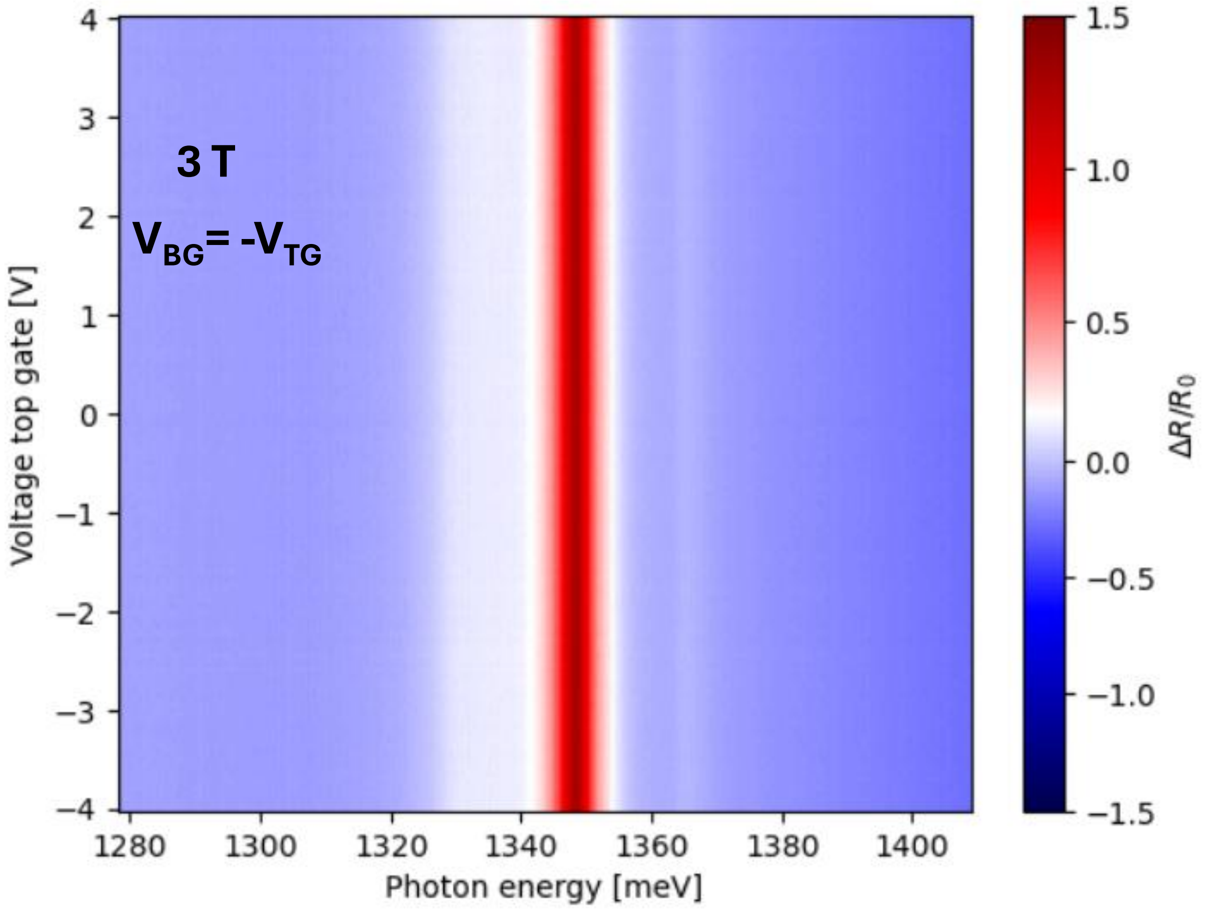
$$\alpha(X_A) \sim 1.4\, meV \cdot T^{-2}$$

$$\alpha(X_B) \sim 3.6\, meV \cdot T^{-2}$$

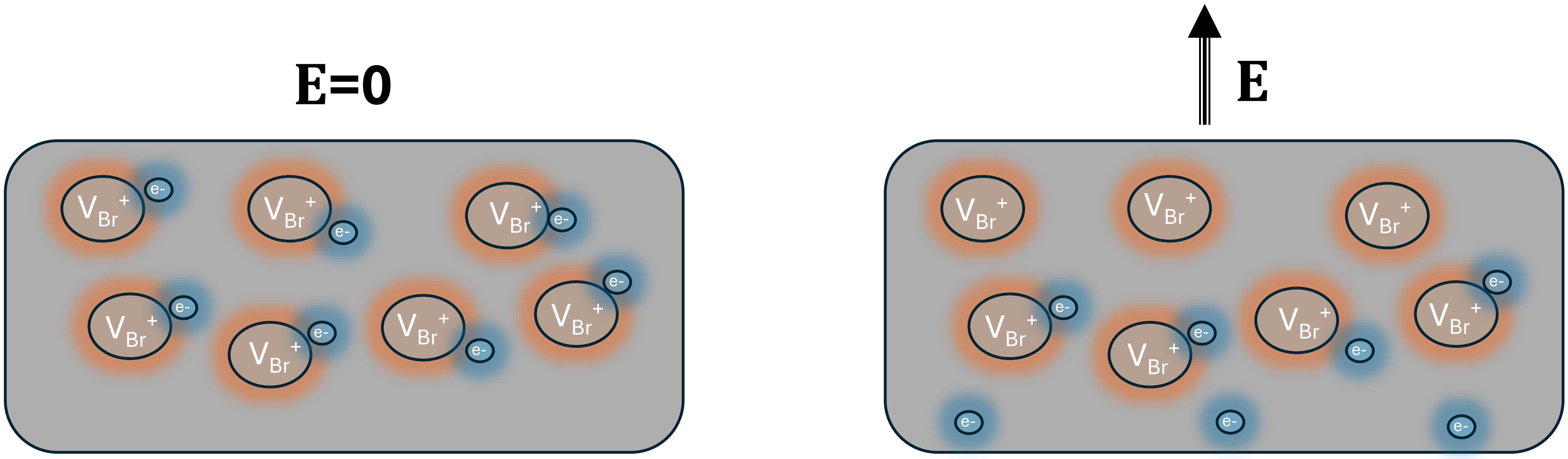
$$\alpha(X_C) \sim 3.2\, meV \cdot T^{-2}$$

Unfortunately…





New perspective: screening due to n-doping from vacancies



➢ **PL measurements could potentially verify this, but nothing found yet**

Conclusion

- Bilayer device with doping, and promising electric response
- No significant response to electrical field on thick devices
- Electrical control only seems to be possible in the thin flake limit
- Donor-like Bromine vacancies are a possible explanation

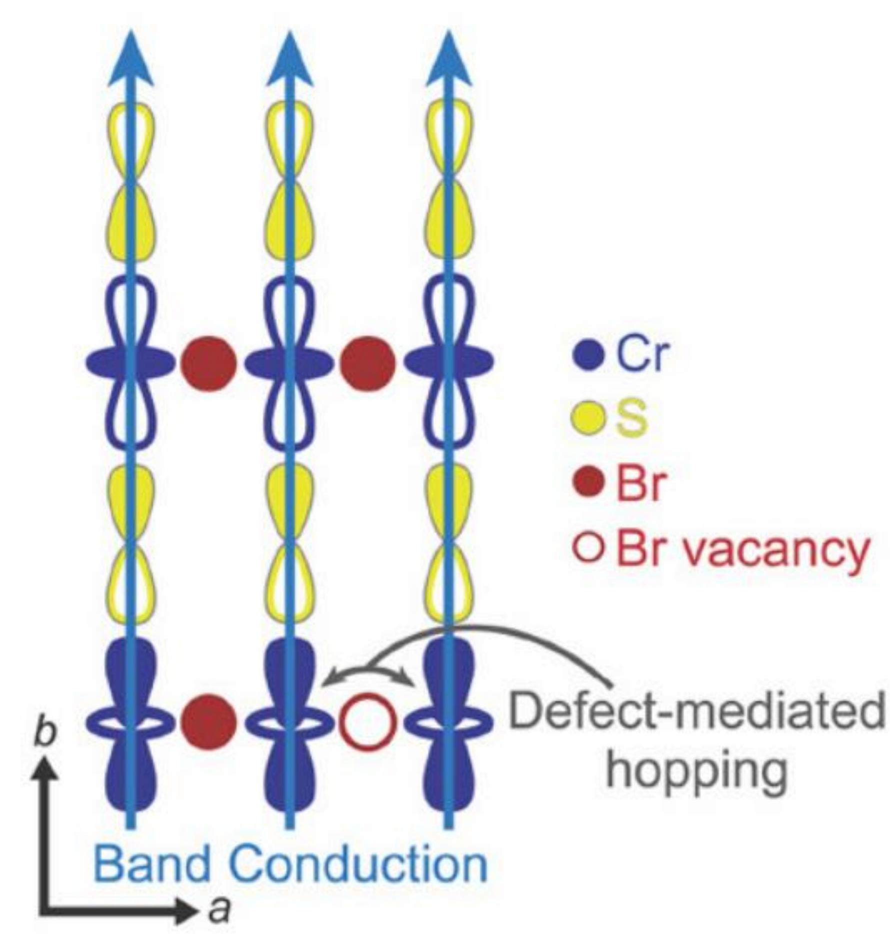
Outlook

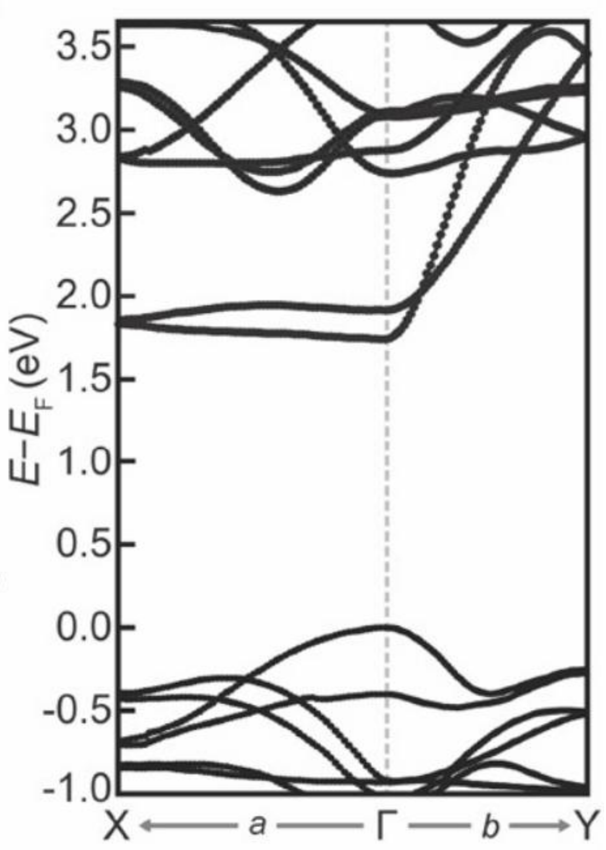
- Understanding the vacancies of this material
- New heterostructures: TMDs, twisted bilayers
- Exploring photonic integration of this material

Questions?

Acknoledgements

Properties of CrSBr: quasi-1D





$m_x^e/m_y^e \sim 50$

$m_x^h/m_y^h \sim 6$

Michael E. Ziebel, Margalit L. Feuer, Jordan Cox, Xiaoyang Zhu, Cory R. Dean, and Xavier Roy
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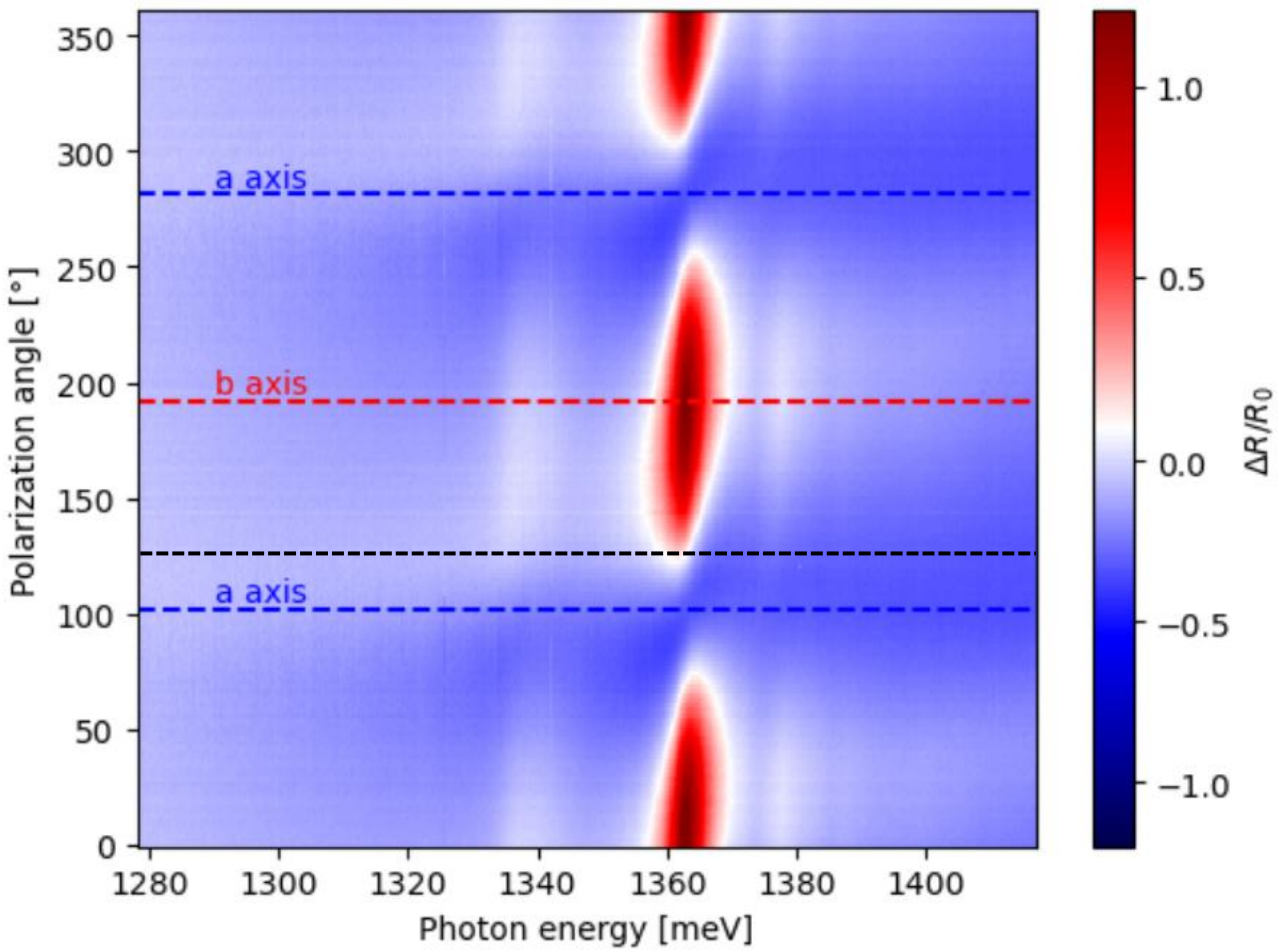
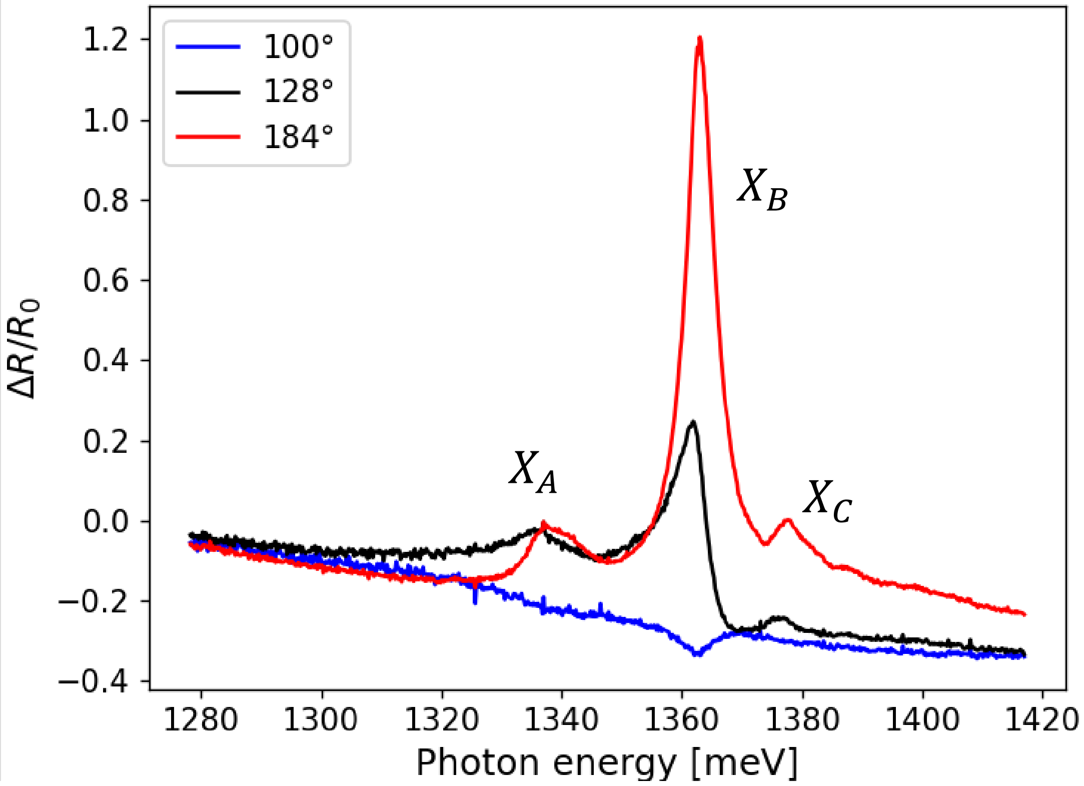
Complete linear polarization

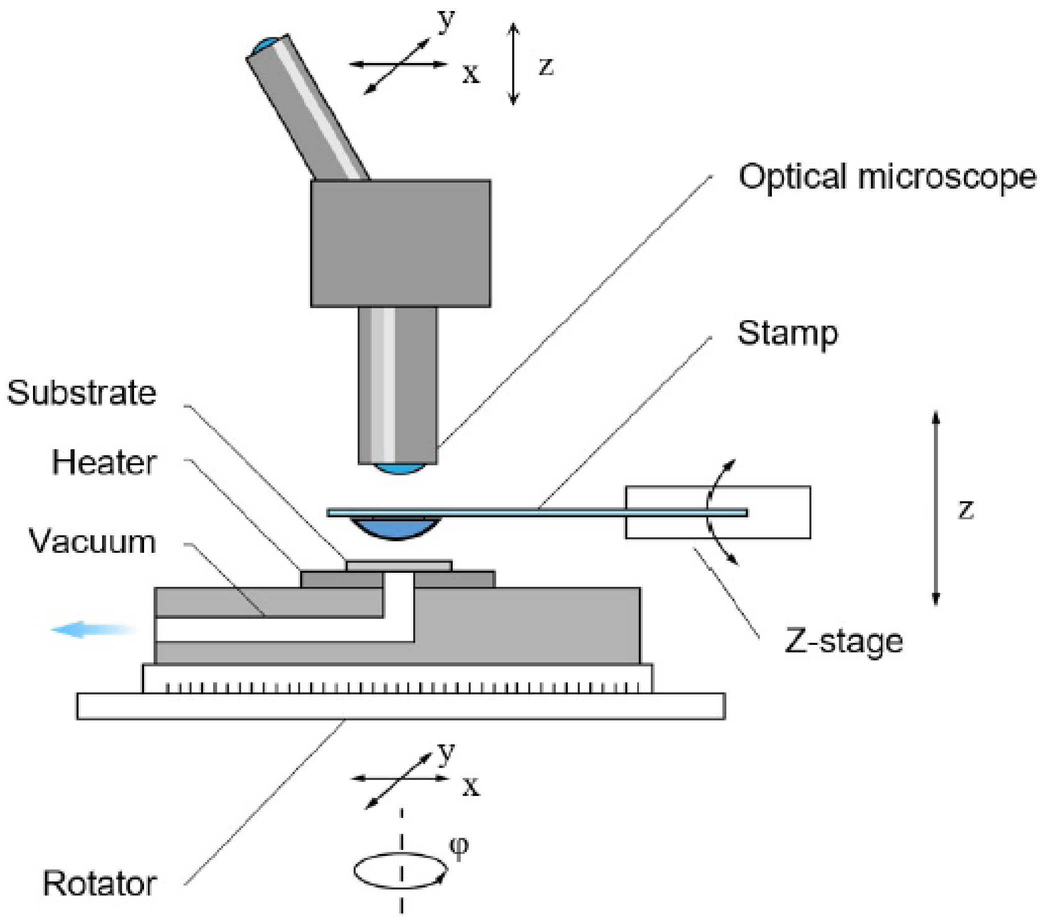
0T 8K



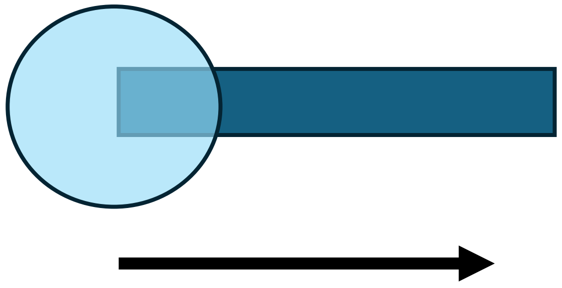


But first… fabrication!

Stacking setup



- PC on PDMS polymer stamp
- 70 → 140°C
- PC front along a-axis (long axis)



Temperature induced structural changes

Volatility of atomic species

- Air sensitivity of monolayers
- Up to $7x10^{13}$ cm^{-2} Bromine surface defects
- Increased surface roughness

Decreased top adhesion

Increased surface conformation

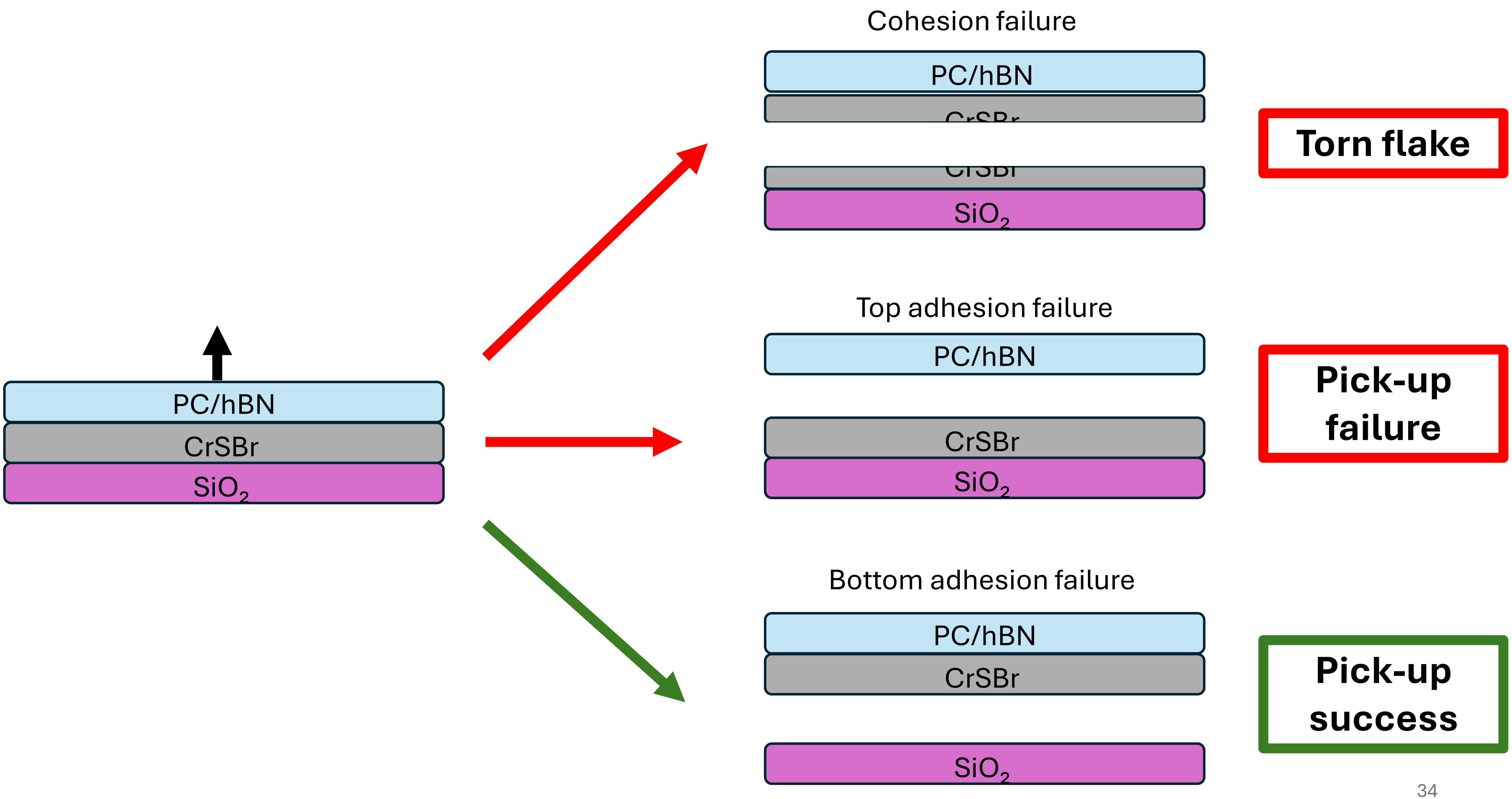
- Effect observed in graphene

Covalent bond formation

- SiO_2 surface hydroxyl groups tend to form covalent bonds with Bromine atoms

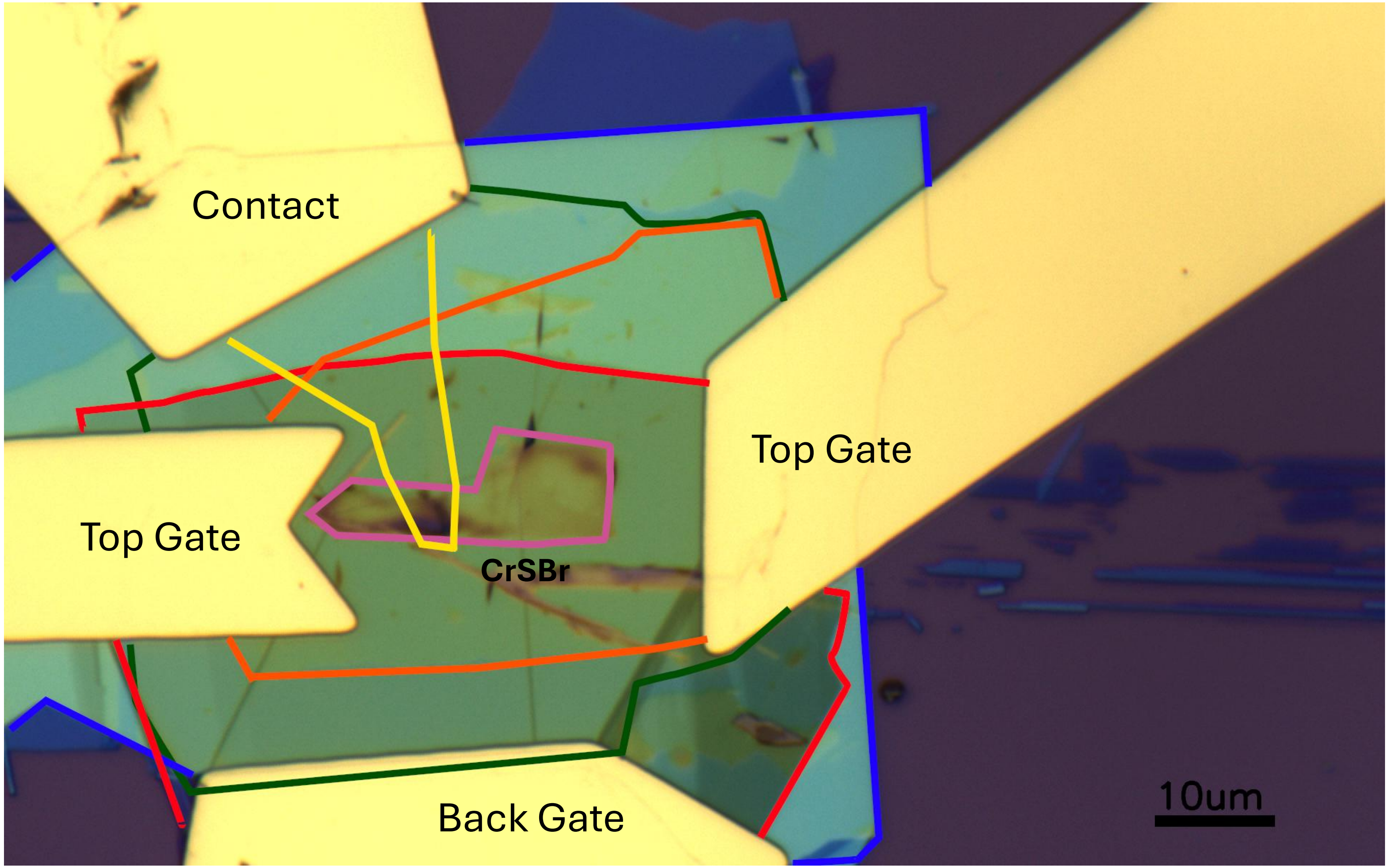
Increased bot adhesion

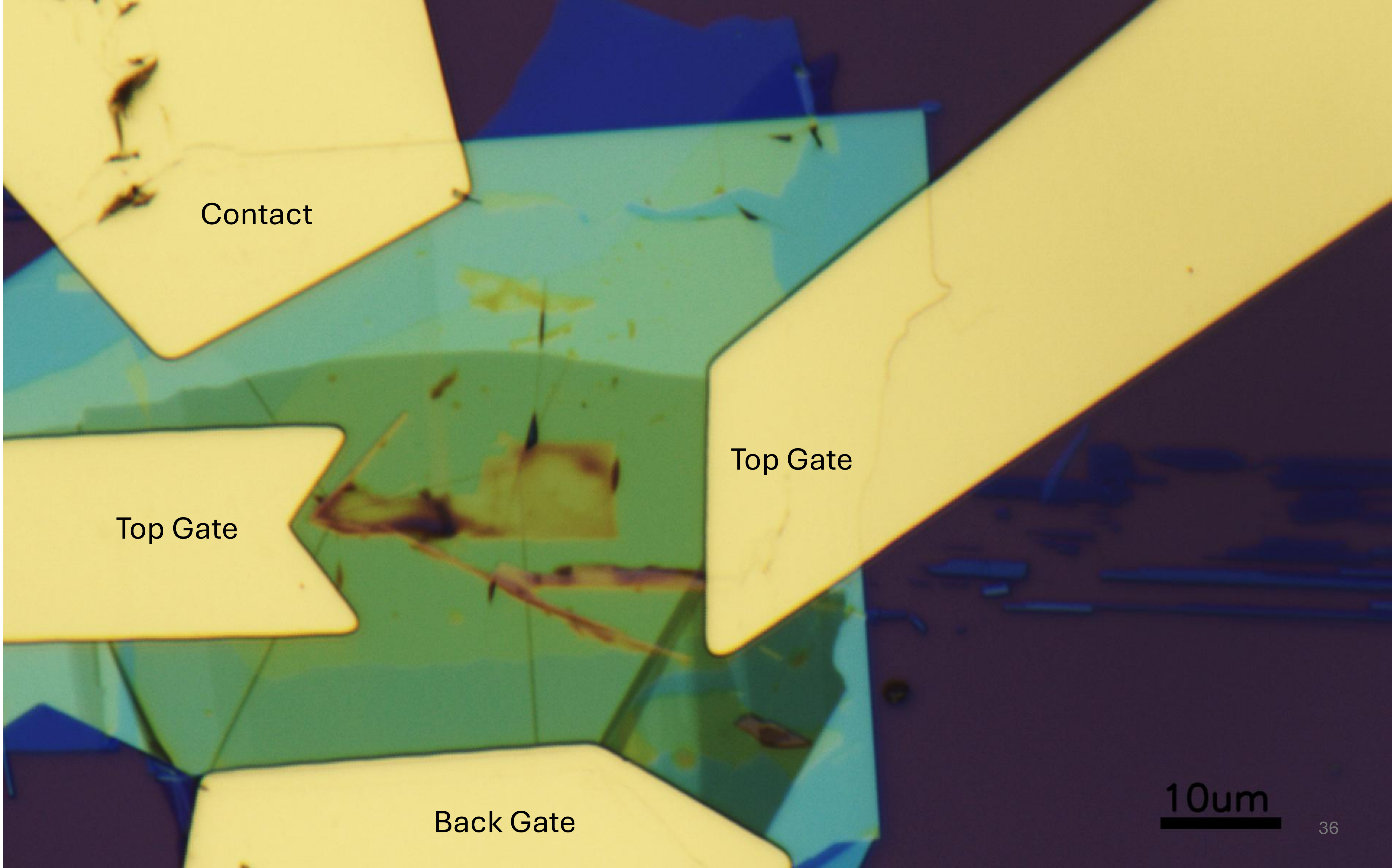
Sunmin Ryu, Li Liu, Stephane Berciaud, Young-Jun Yu, Haitao Liu, Philip Kim, George W. Flynn, and Louis E. Brus *Nano Letters* **2010** *10* (12), 4944-4951; Telford, E.J., Dismukes, A.H., Dudley, R.L. et al. Coupling between magnetic order and charge transport in a two-dimensional magnetic semiconductor. Nat. Mater. 21, 754–760 (2022); Adv Funct Materials, Volume: 33, Issue: 12, First published: 08 January 2023, DOI: (10.1002/adfm.202211366)


PC/hBN
CrSBr
SiO2
Cohesion failure
PC/hBN
SiO2
Torn flake
Top adhesion failure
PC/hBN
CrSBr
SiO2
Pick-up failure
Bottom adhesion failure
PC/hBN
CrSBr
SiO2
Pick-up success

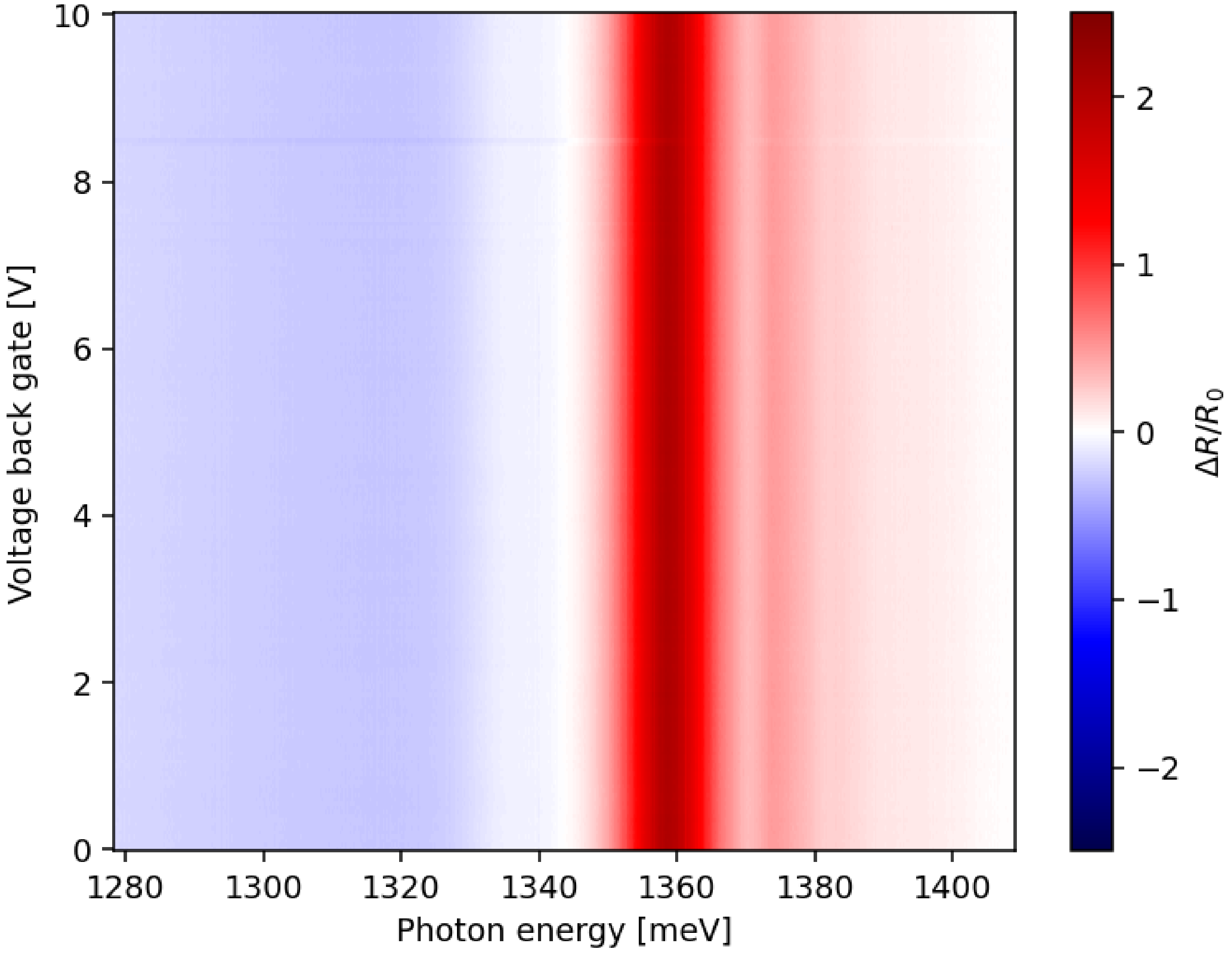
Third device

- 9 nm CrSBr



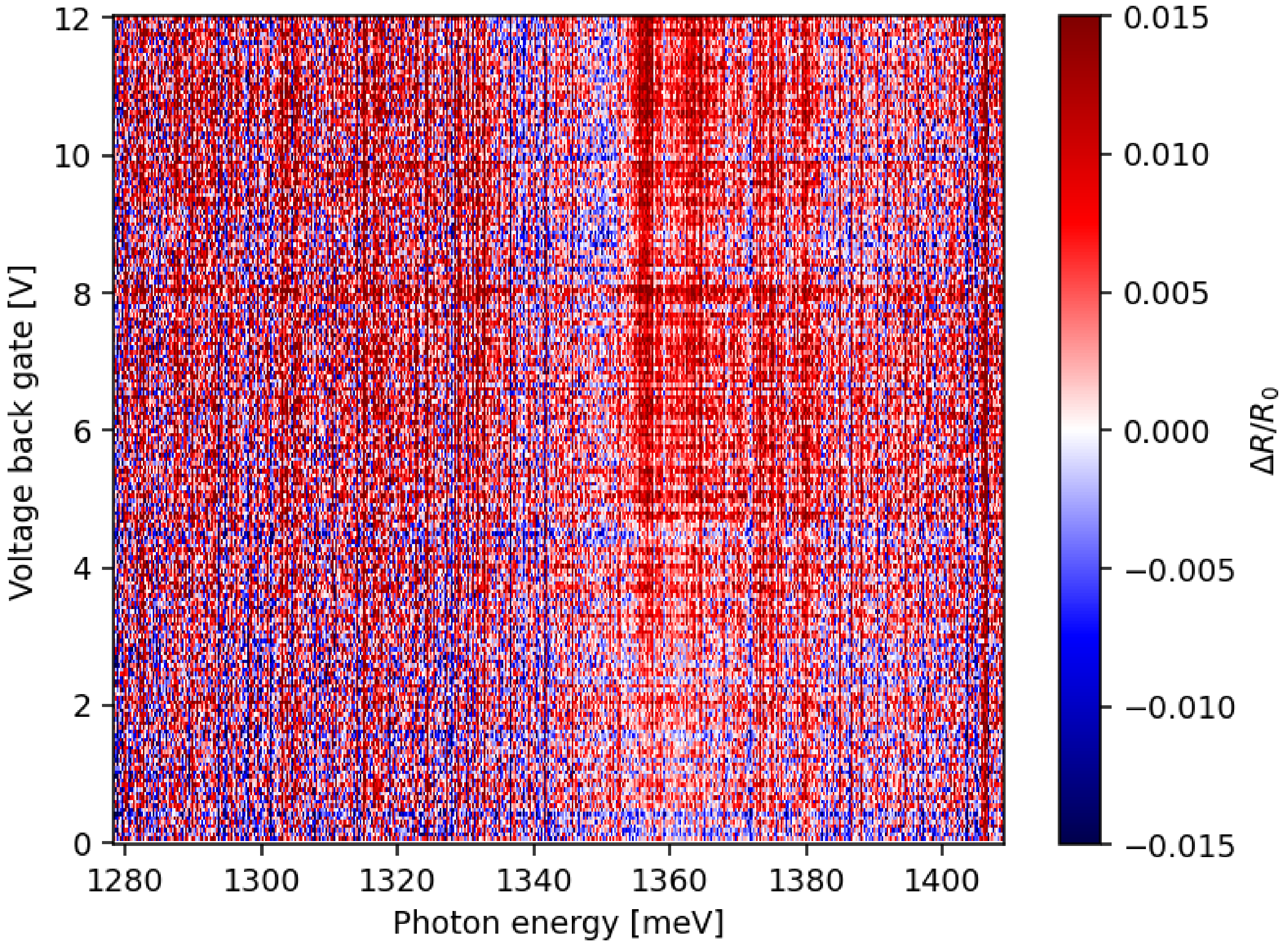

Contact
Top Gate
Top Gate
Back Gate
10um

Again…

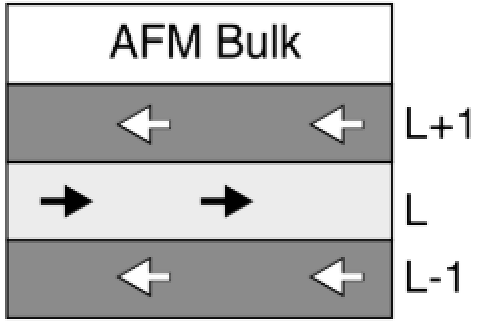


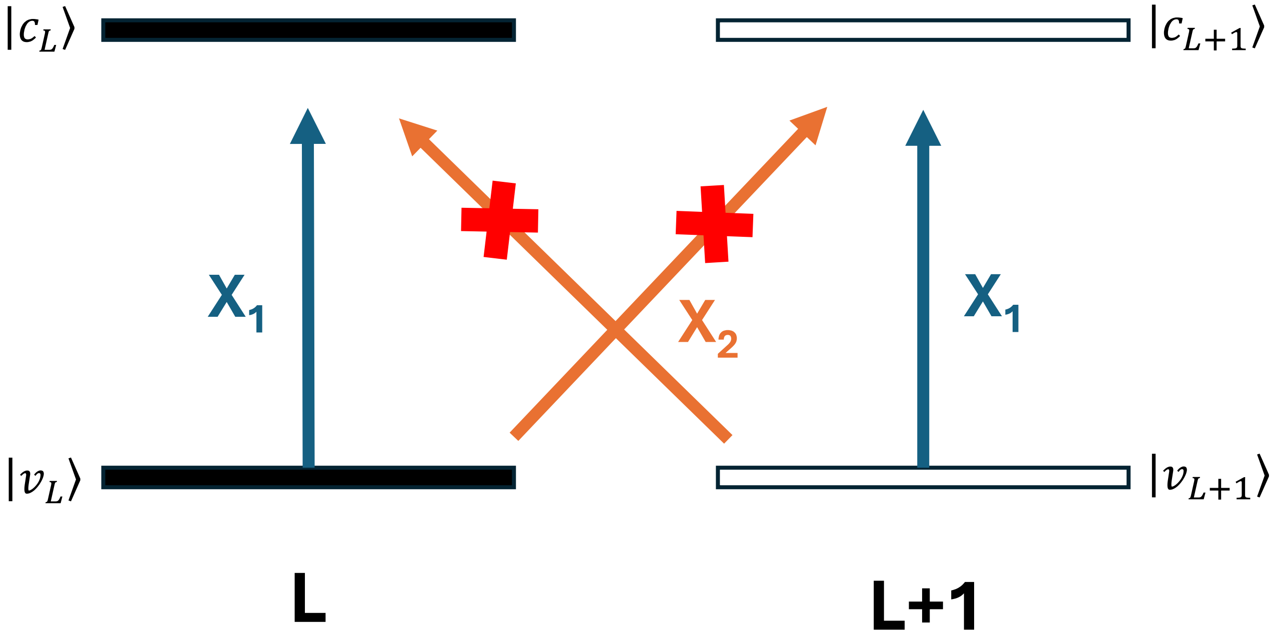
10
8
6
4
2
0
Voltage back gate [V]
1280
1300
1320
1340
1360
1380
1400
Photon energy [meV]
2
1
0
−1
−2
$\Delta R/R_0$

But…

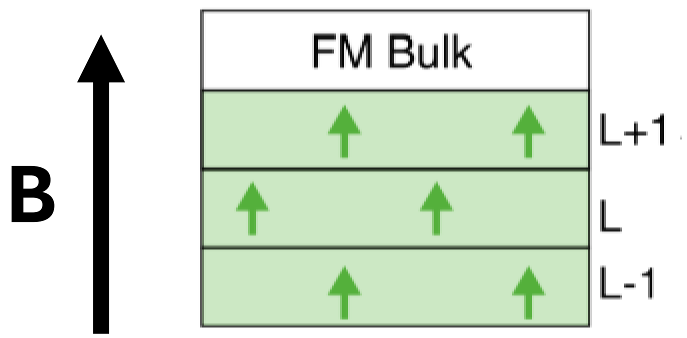


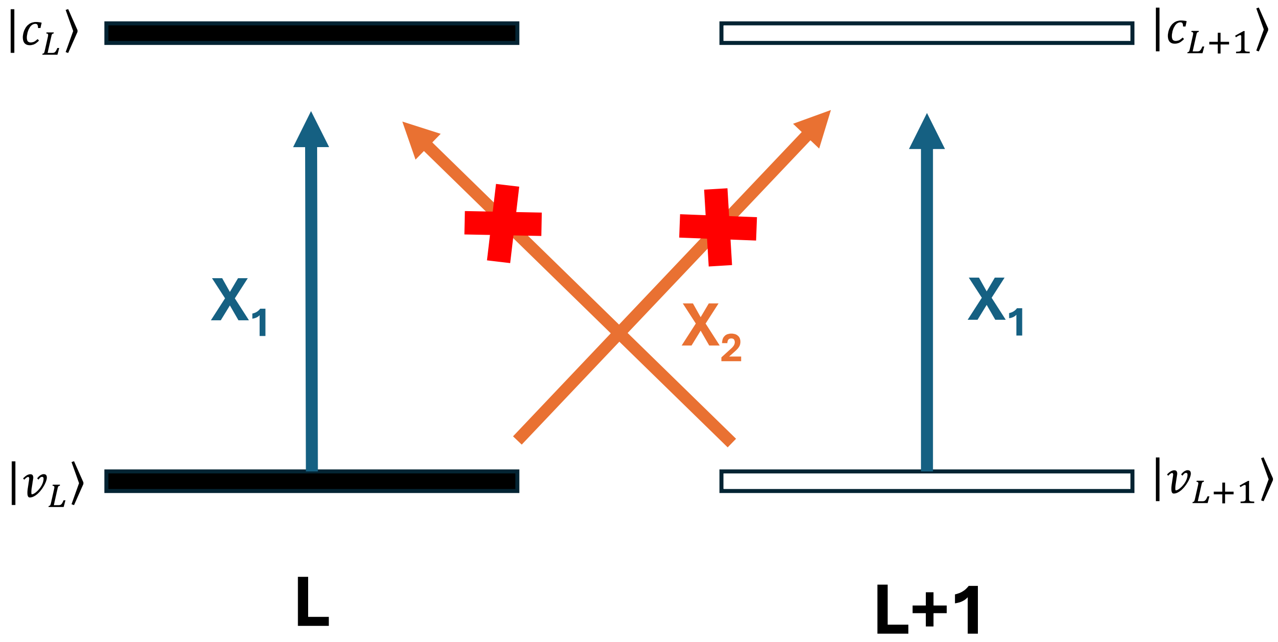
$$\frac{\frac{\Delta R}{R_0}(2L)}{\frac{\Delta R}{R_0}(9nm)} \sim 0.02$$





Marie-Christin Heißenbüttel, et al. (2024) https://doi.org/10.48550/arXiv.2403.20174





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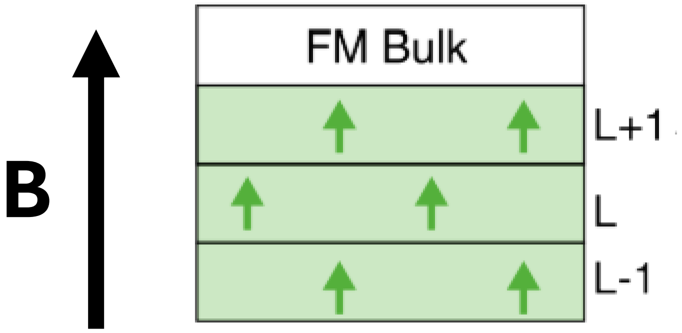
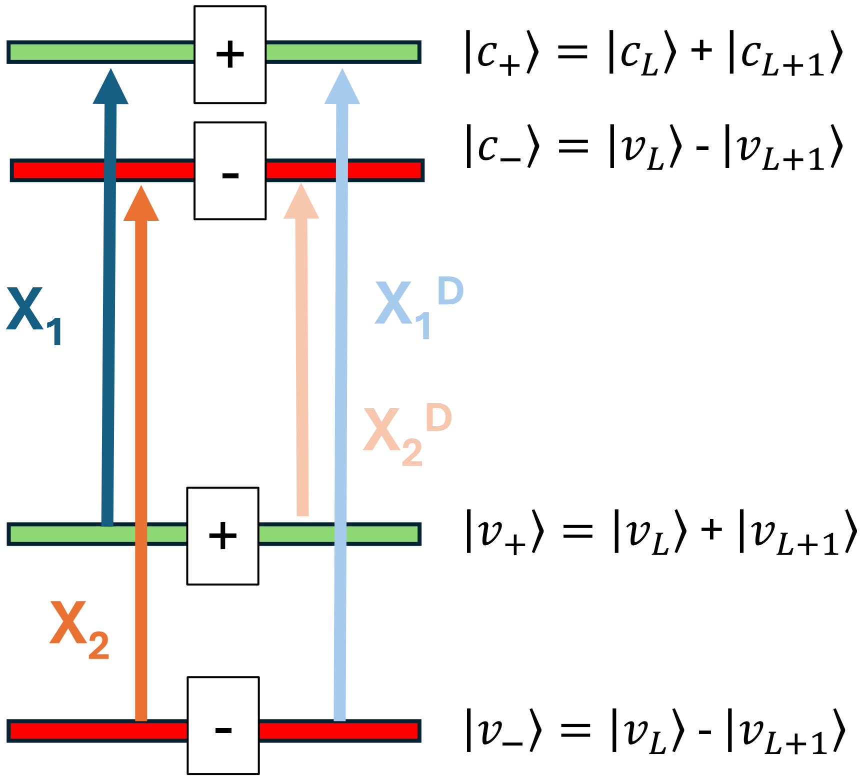
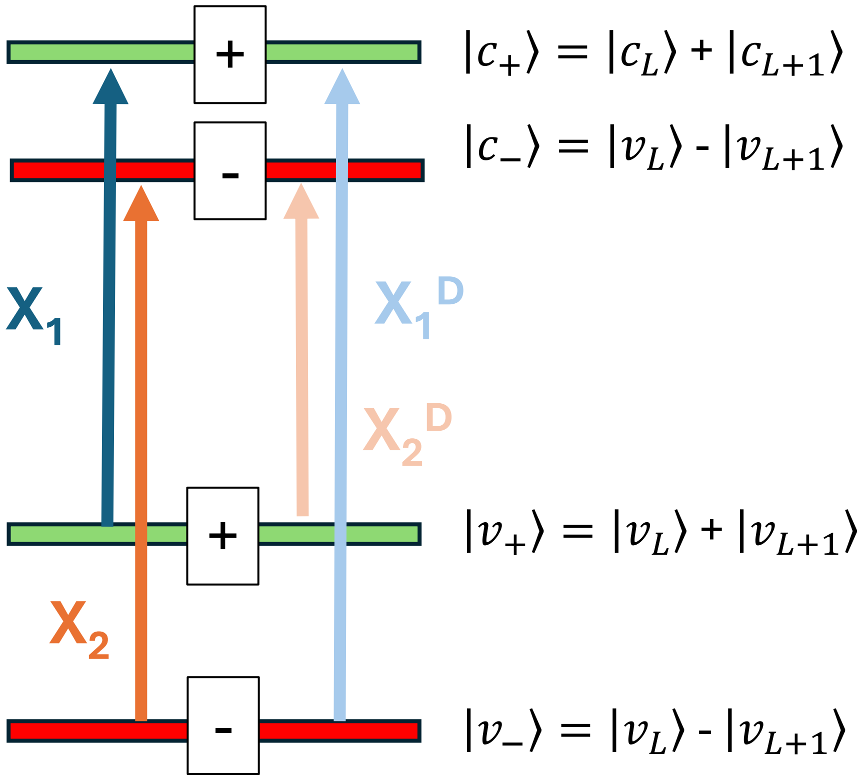




Marie-Christin Heißenbüttel, et al. (2024) https://doi.org/10.48550/arXiv.2403.20174

$$H^{\mathrm{el}}_{\boldsymbol{k}}(B) = \begin{pmatrix} E_{n\boldsymbol{k}} & t_{n\boldsymbol{k}}B \\ t_{n\boldsymbol{k}}B & E_{n\boldsymbol{k}} \end{pmatrix}$$



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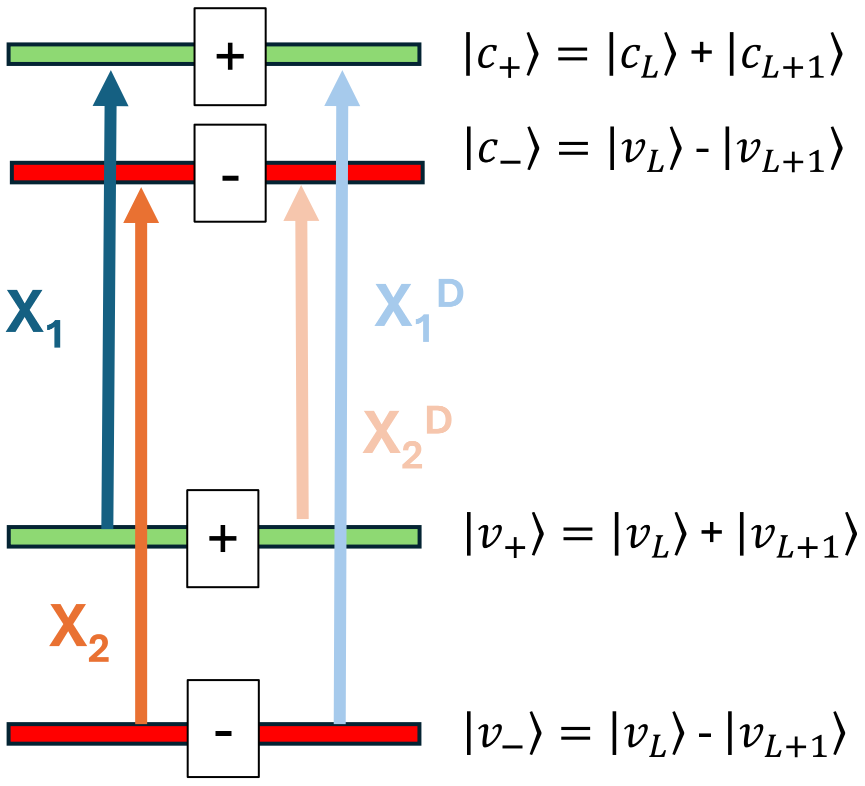
$$H^{\text{el}}_{\boldsymbol{k}}(B) = \begin{pmatrix} E_{n\boldsymbol{k}} & t_{n\boldsymbol{k}}B \\ t_{n\boldsymbol{k}}B & E_{n\boldsymbol{k}} \end{pmatrix}$$

Eigenvalues



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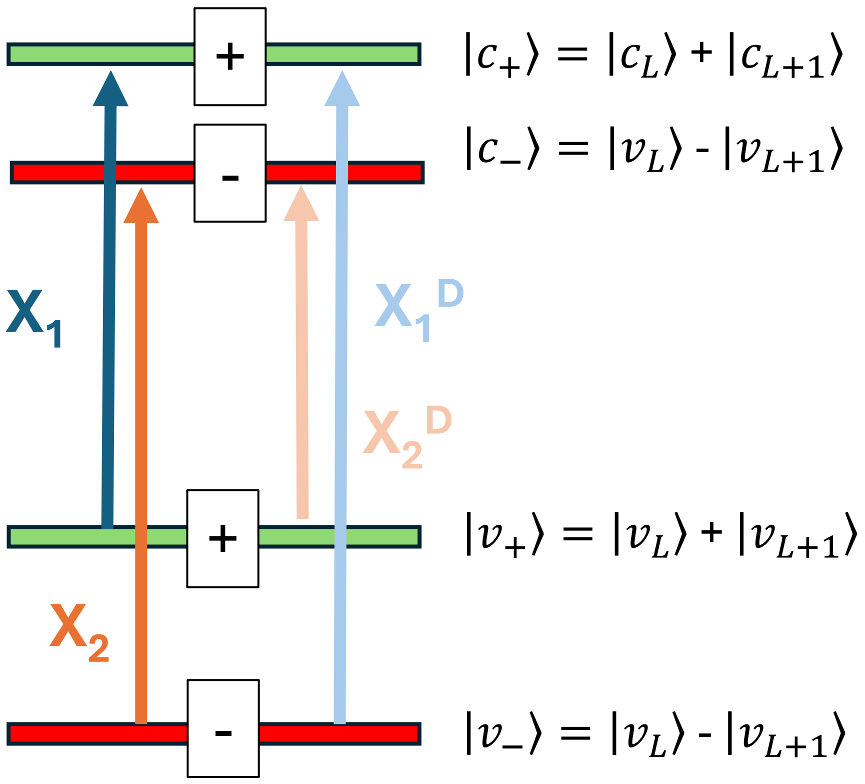
$$H^{\mathrm{el}}_{\boldsymbol{k}}(B) = \begin{pmatrix} E_{n\boldsymbol{k}} & t_{n\boldsymbol{k}}B \\ t_{n\boldsymbol{k}}B & E_{n\boldsymbol{k}} \end{pmatrix}$$

Eigenvalues

$$E_{n\boldsymbol{k},\pm} = E_{n\boldsymbol{k}} \pm t_{n\boldsymbol{k}}B$$



Marie-Christin Heißenbüttel, et al. (2024) https://doi.org/10.48550/arXiv.2403.20174

In the exciton basis...

$$H(B) = \begin{bmatrix} E_g + tB + V_{eh} & 0 \\ 0 & E_g - tB + V_{eh} \end{bmatrix}$$

Interaction

$$H(B) = \begin{bmatrix} E_g + tB + V_{eh} & V_I \\ V_I & E_g - tB + V_{eh} \end{bmatrix}$$

In the exciton basis...

$$H(B) = \begin{bmatrix} E_g + tB + V_{eh} & V_I \\ V_I & E_g - tB + V_{eh} \end{bmatrix}$$

Diagonalization

$$\lambda_{+,-} = E_g + V_{eh} \pm \sqrt{(tB)^2 + V_I^2}$$

Dominant B^2 contribution for strong enough interaction

hBN thicknesses

- CrSBr1:
  - Top hBN: 21.9 nm
  - Bottom hBN: 38.3 nm
- CrSBr5
  - Top hBN: 27.9 nm
  - Bottom hBN: 25.5 nm
- CrSBr8
  - Top hBN: 19 nm
  - Bottom hBN: 51.5 nm