Hybrid moiré excitons and trions

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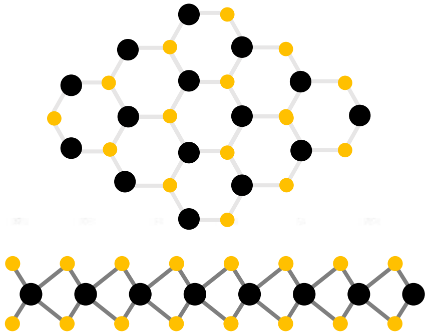
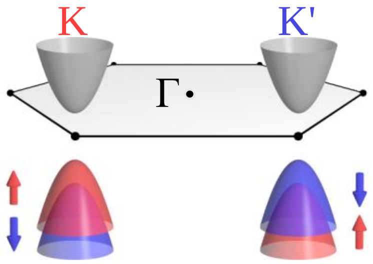
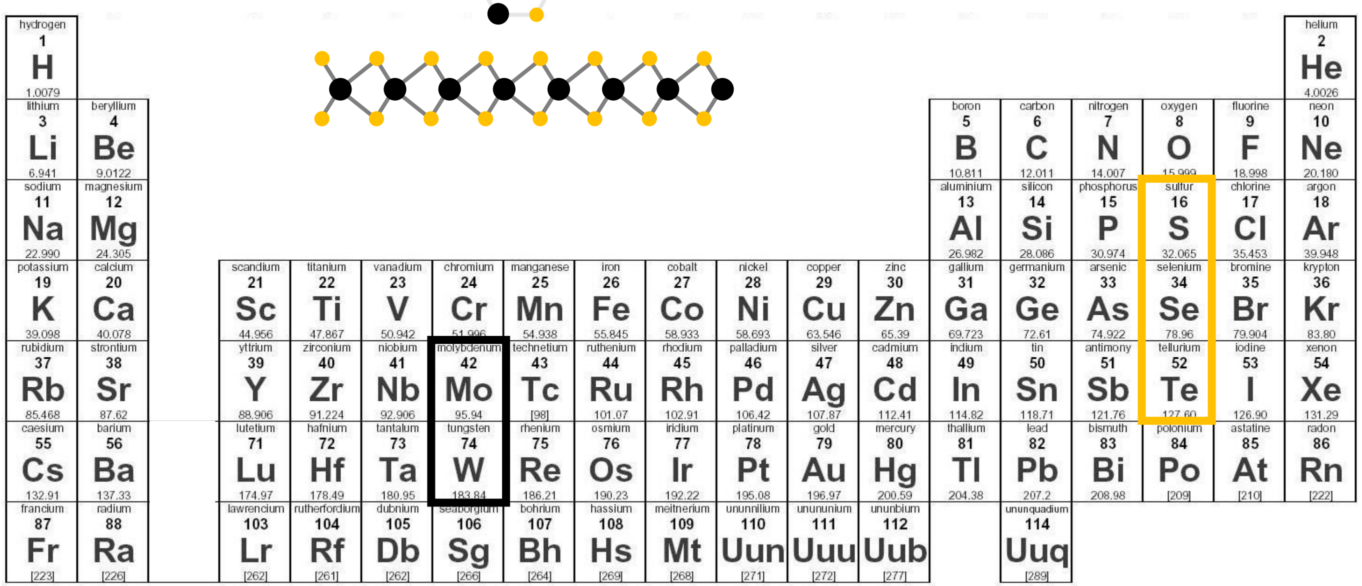
Monolayer 2D semiconductors

Transition Metal Dichalcogenides:

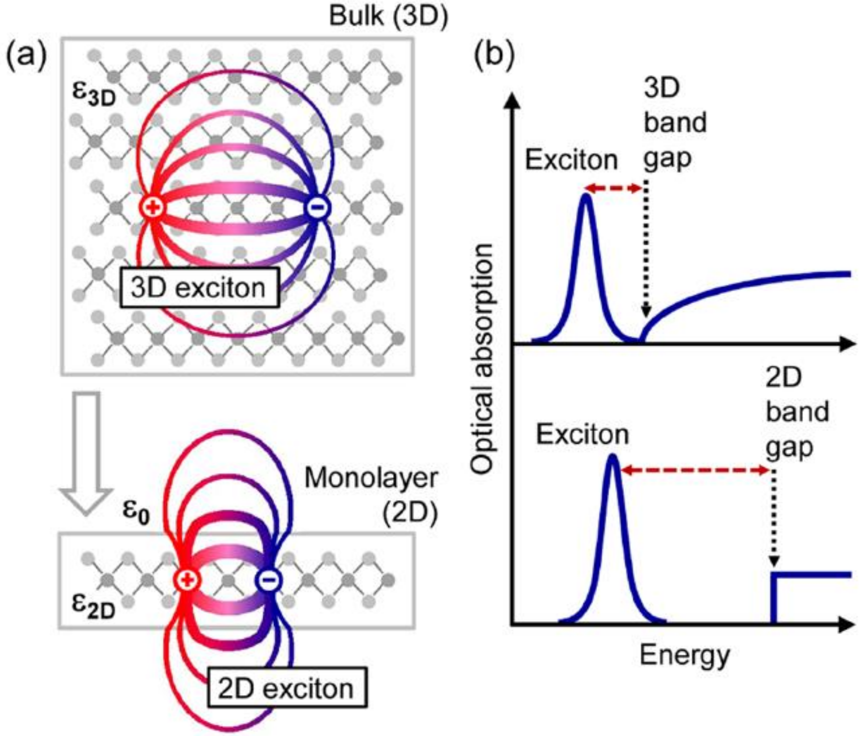
MoS_2
$MoSe_2$
$MoTe_2$
WS_2
WSe_2





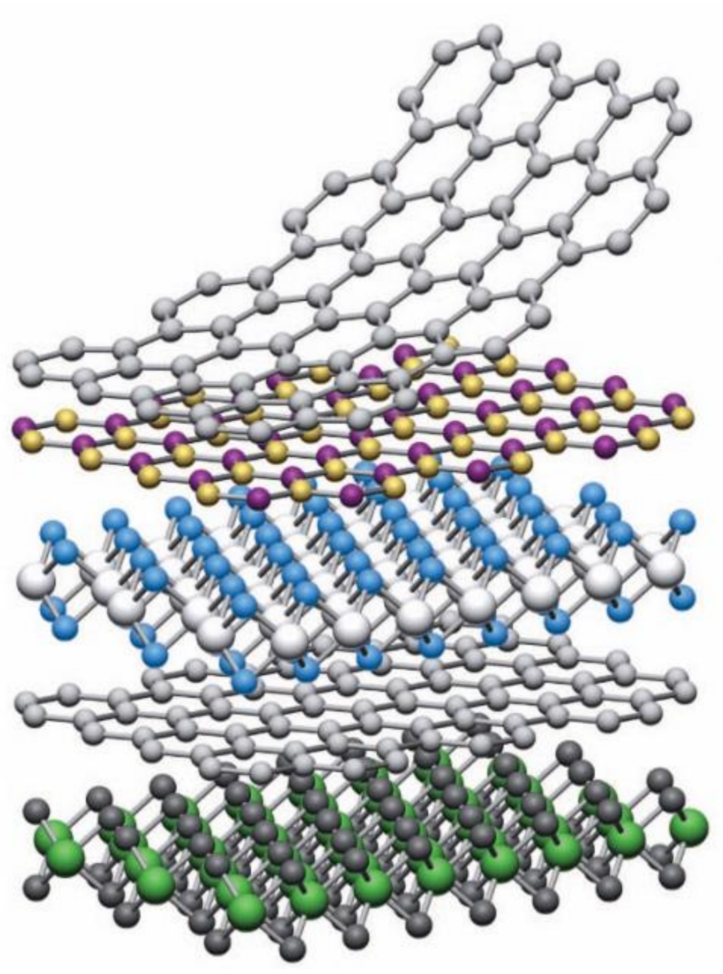


- Direct semiconductors at K valleys
- Huge exciton binding energies

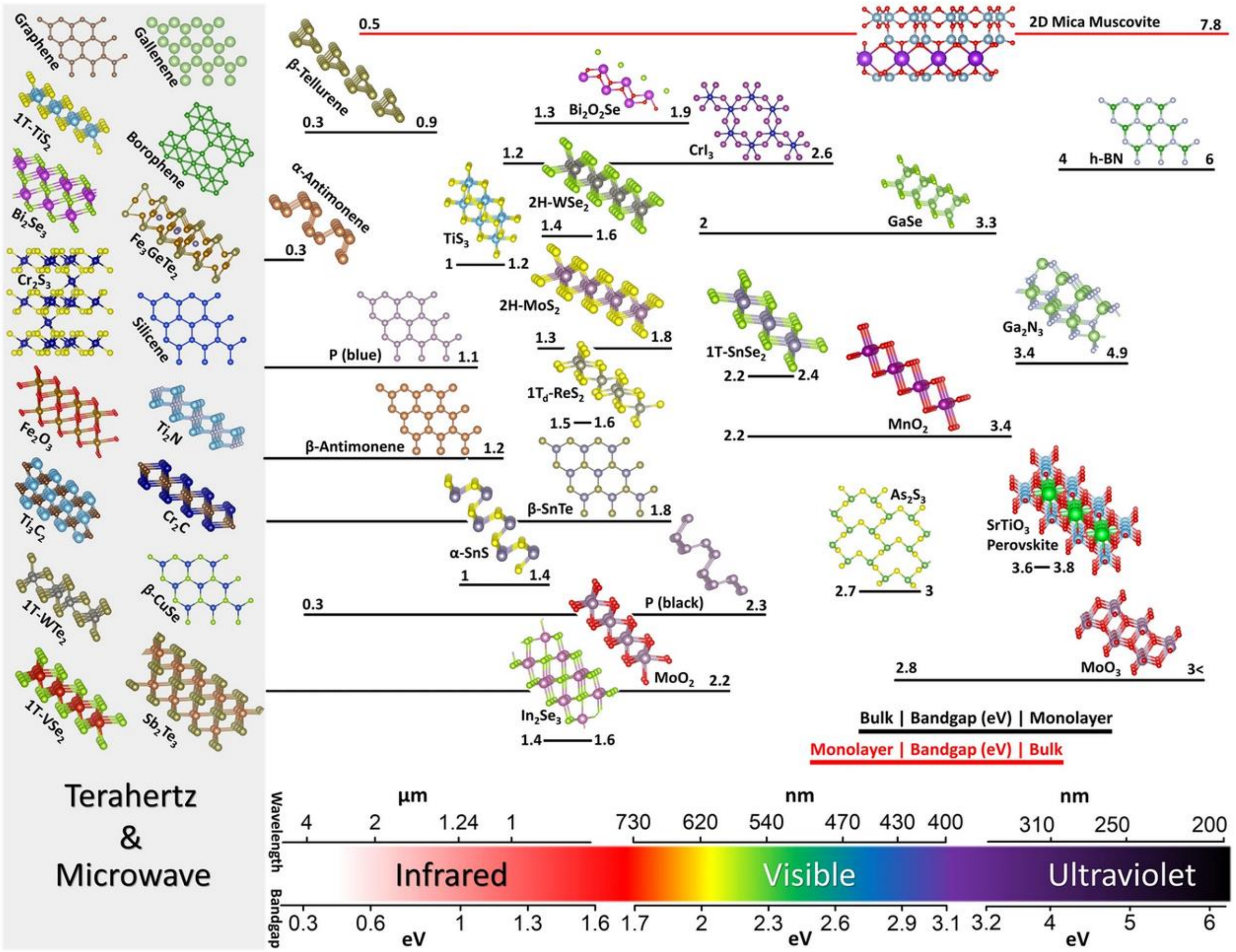


Chernikov et al., PRL 113, 076802 (2014)

van der Waals Lego for material engineering

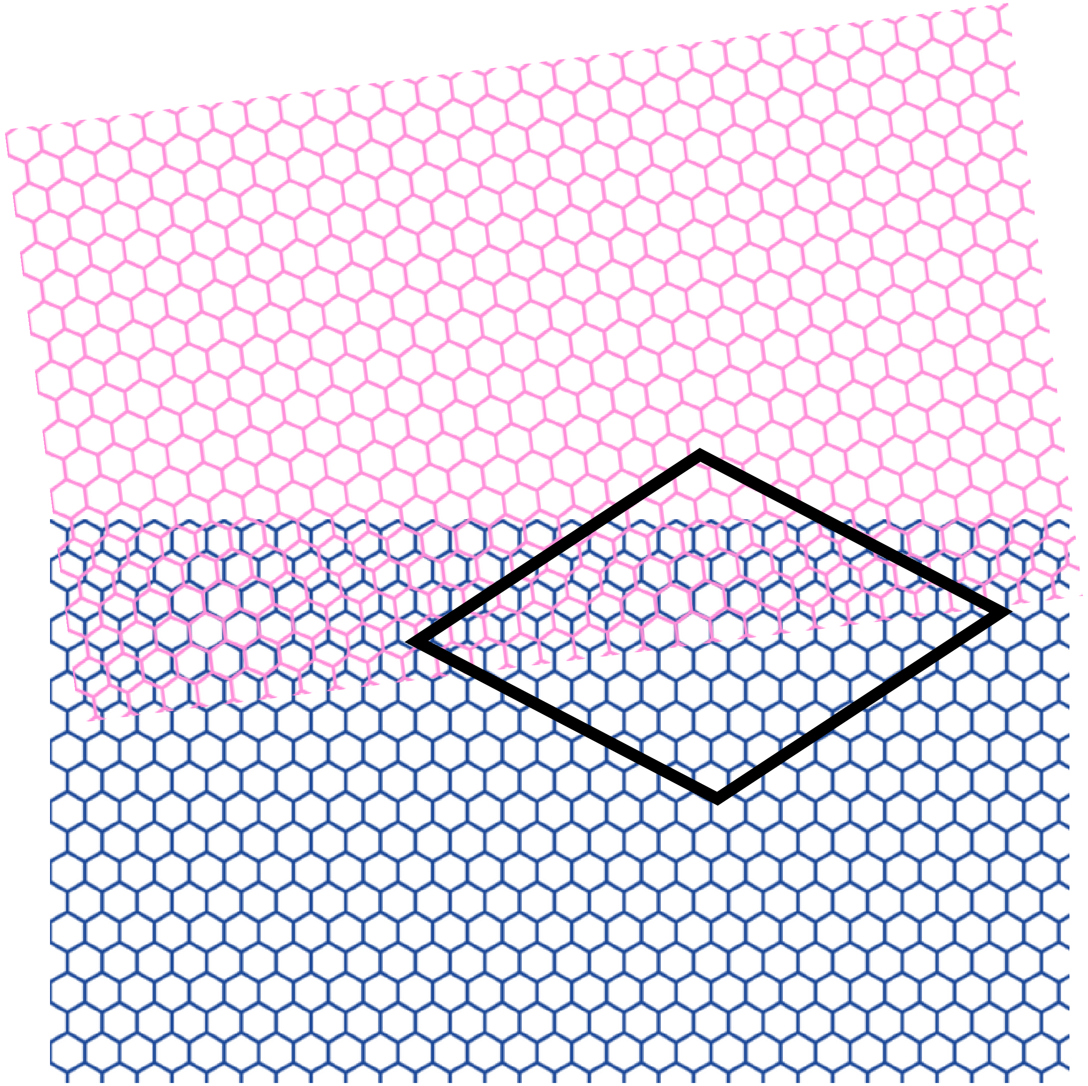


Geim et al., Nature 499, 419 (2013)

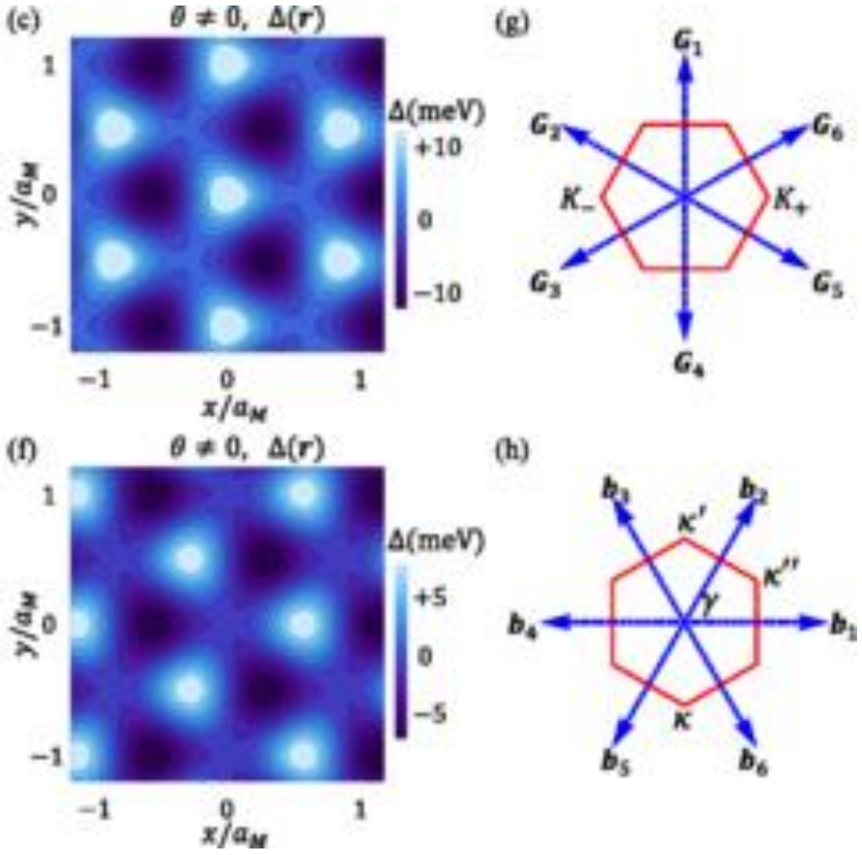


Chaves et al., npj 2D Mater. Appl. 4, 29 (2020)

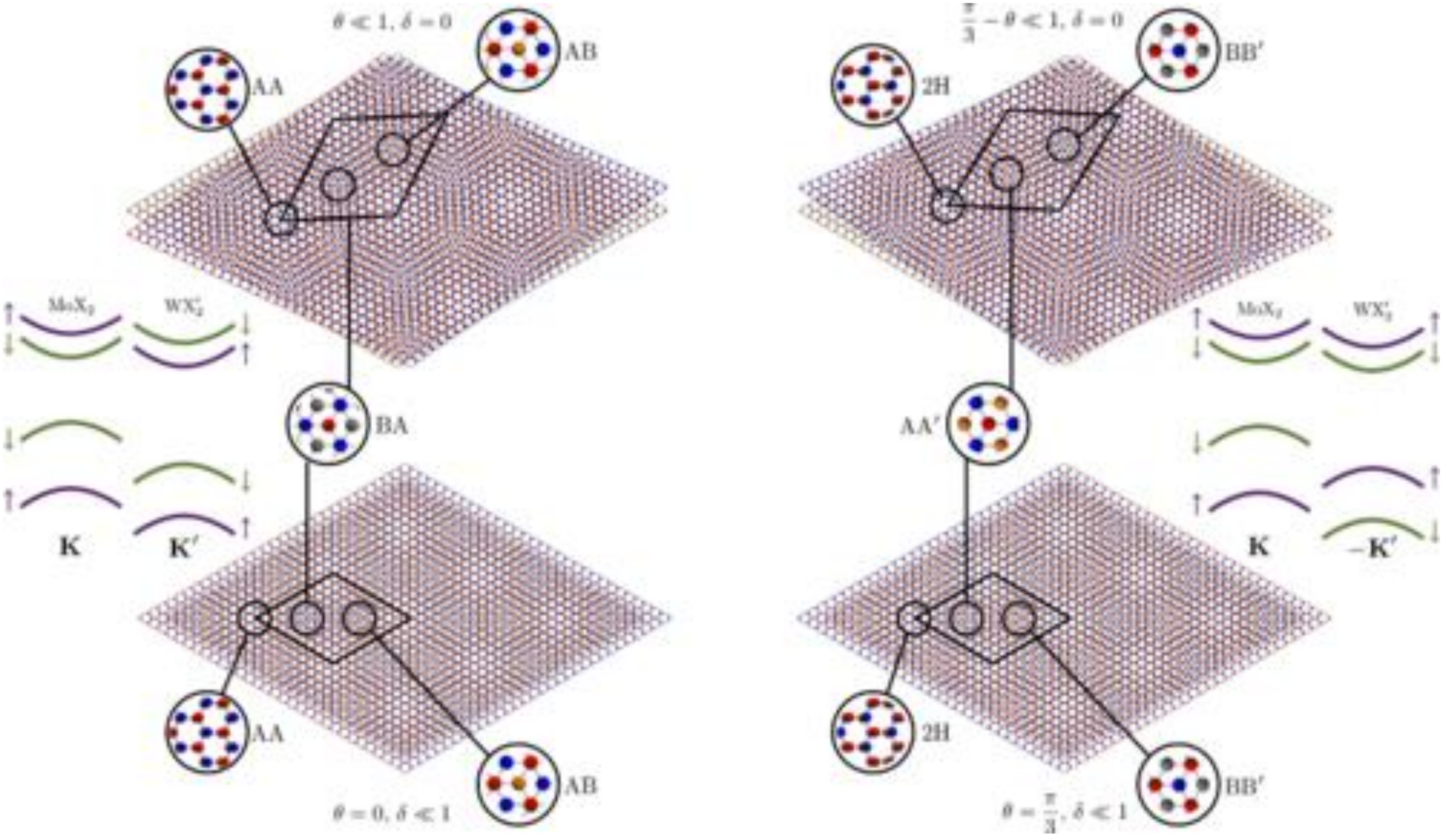
Moiré superlattice formation in bilayers



Moiré theory



F. Wu, T. Lovorn, and A. H. MacDonald, PRL 118, 147401 (2017)



D. A. Ruiz-Tijerina and V. I. Fal’ko, PRB 99, 125424 (2019)

Heterobilayer $MoSe_2/WS_2$

- Lattice mismatch ~ 4%
- $MoSe_2$ and WS_2 have (almost) resonant CBs, allowing for intra- & interlayer exciton hybridization
- Holes are in $MoSe_2$ layer

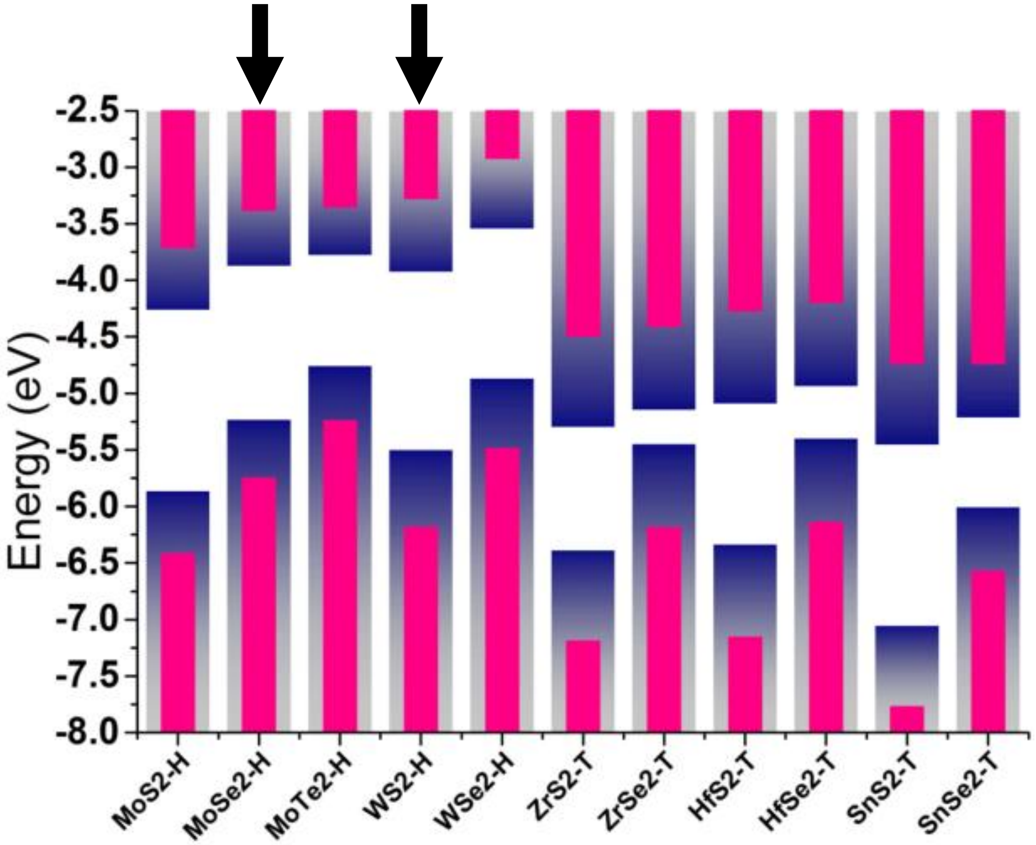
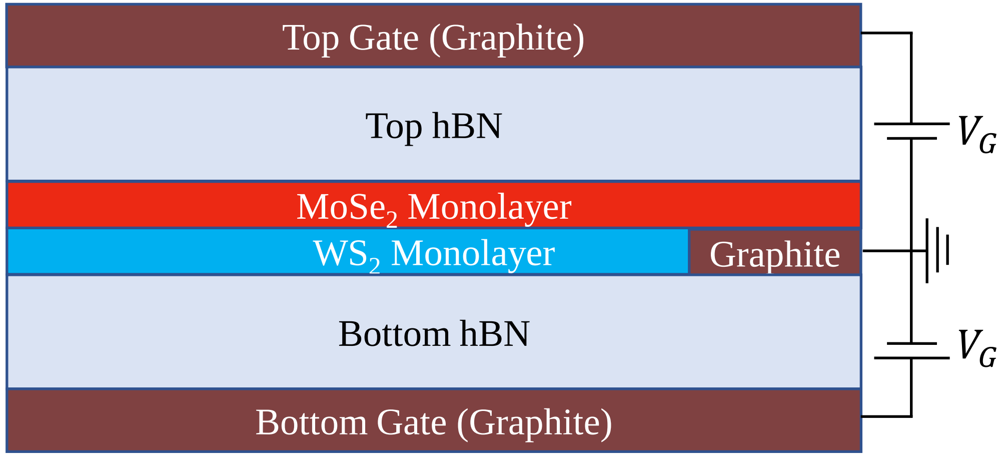


Figure 2. Position of band edges for stable semiconducting TMDs with respect to vacuum. The band edge of DFT-PBE data and G_0W_0 data are indicated by filled navy blue gradient column and pink solid column, respectively. The vacuum level is set to 0 eV.

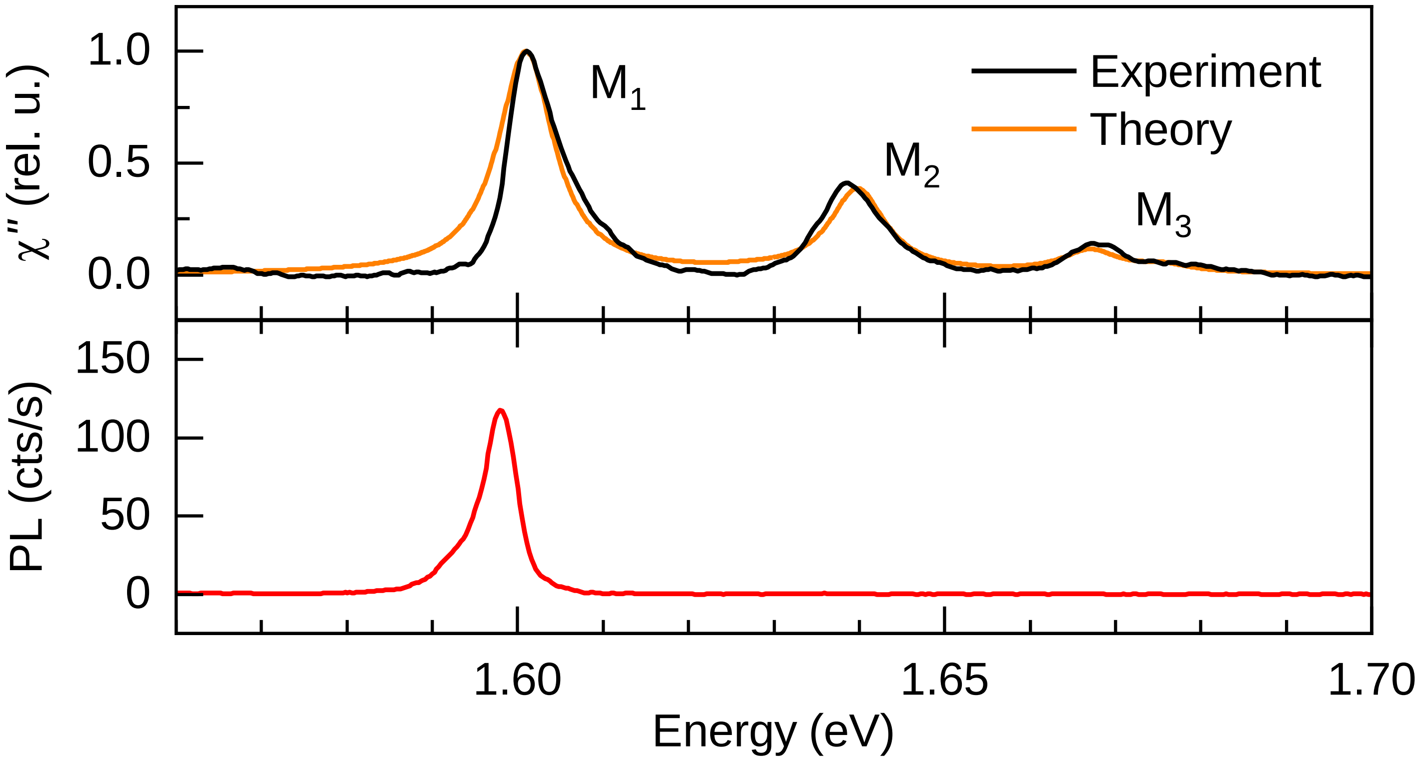
Zhang et al., 2D Mater. 4, 015026 (2016)

Our experiment

Rotationally aligned $MoSe_2/WS_2$ in anti-parallel stacking (H-type) in a dual-gate field-effect device

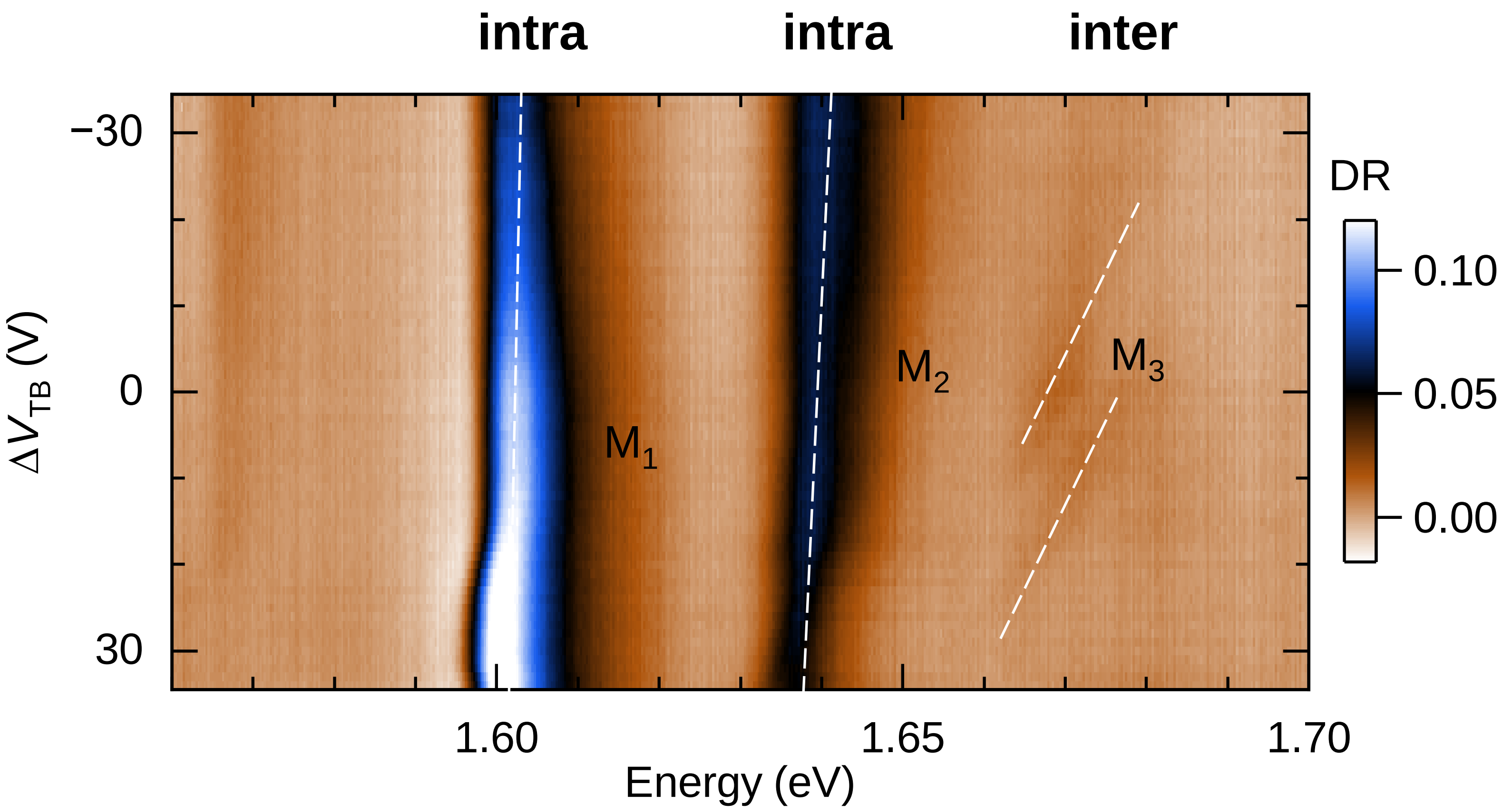


White-light DR and PL Data



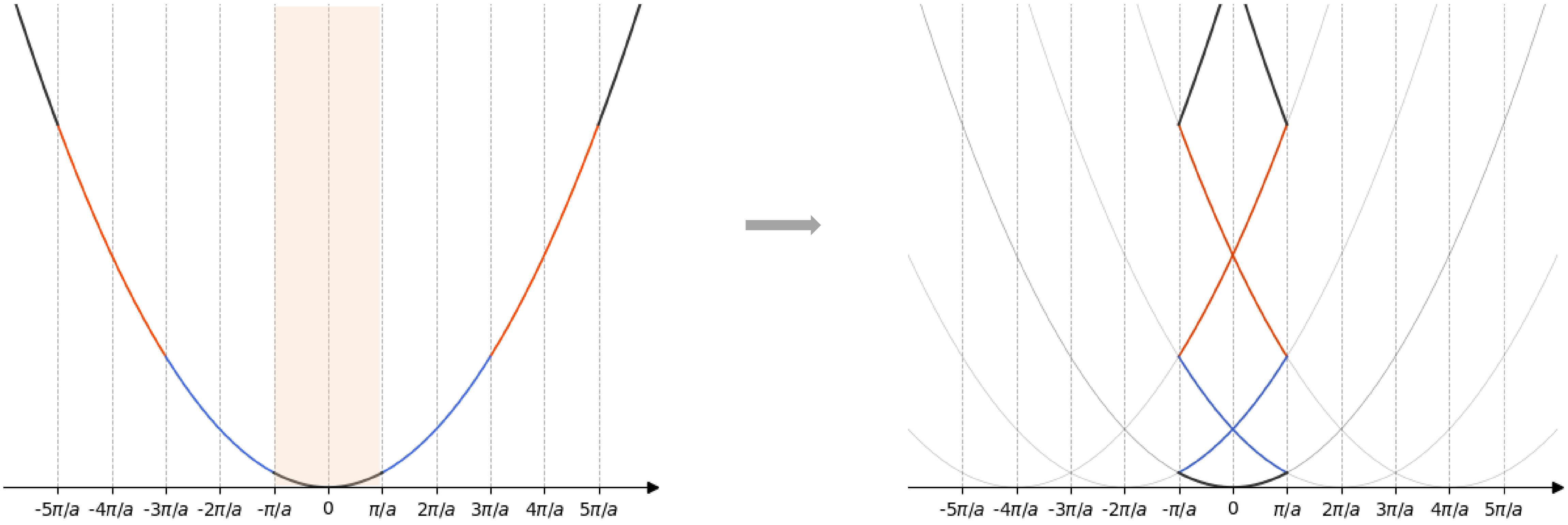
Polovnikov, … , Baimuratov, PRL 32, 076902 (2024)

E-field dependence of white light DR

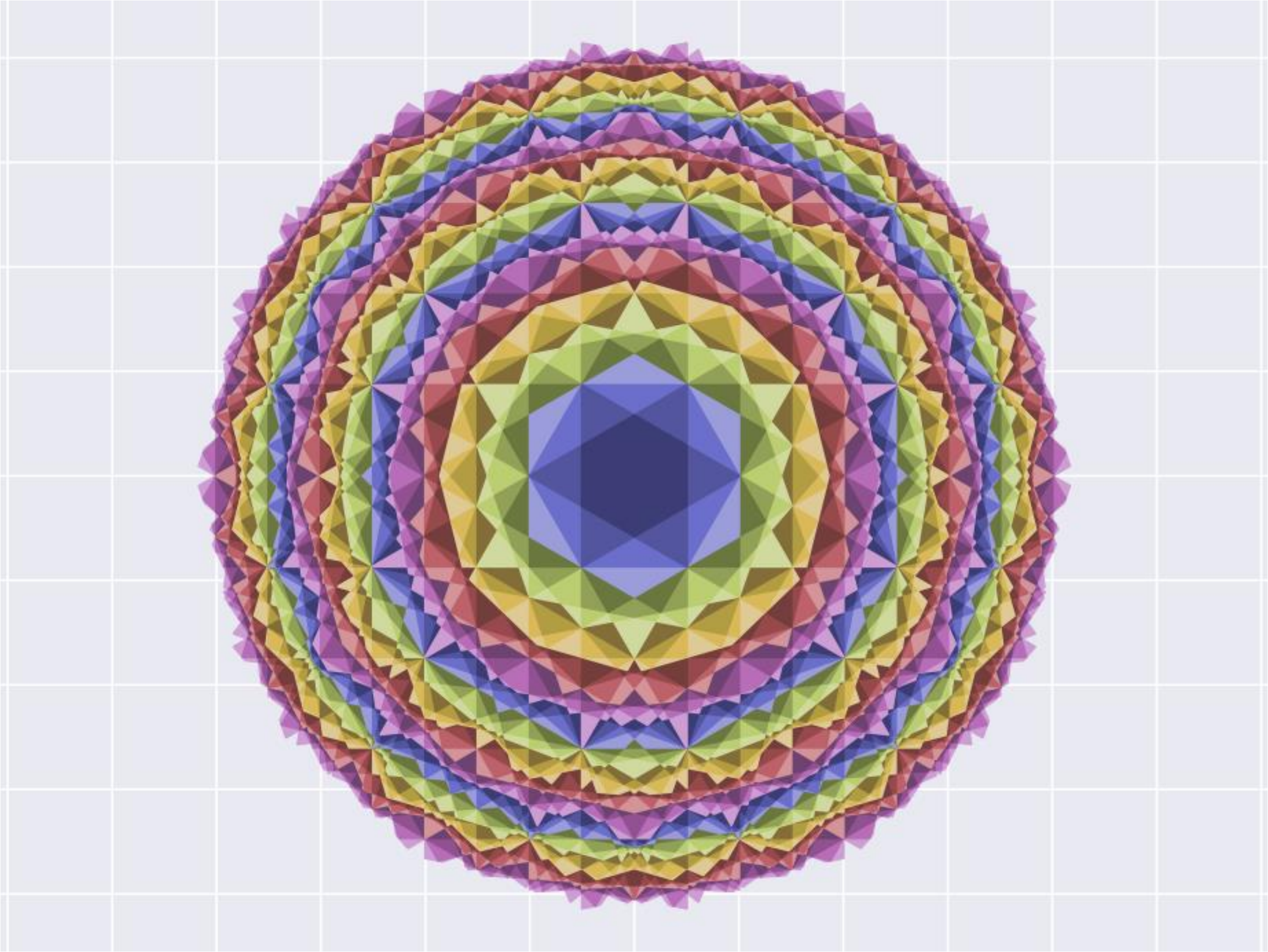


$$\Delta V_{TB} = V_B - V_T$$

Band folding in periodic lattices

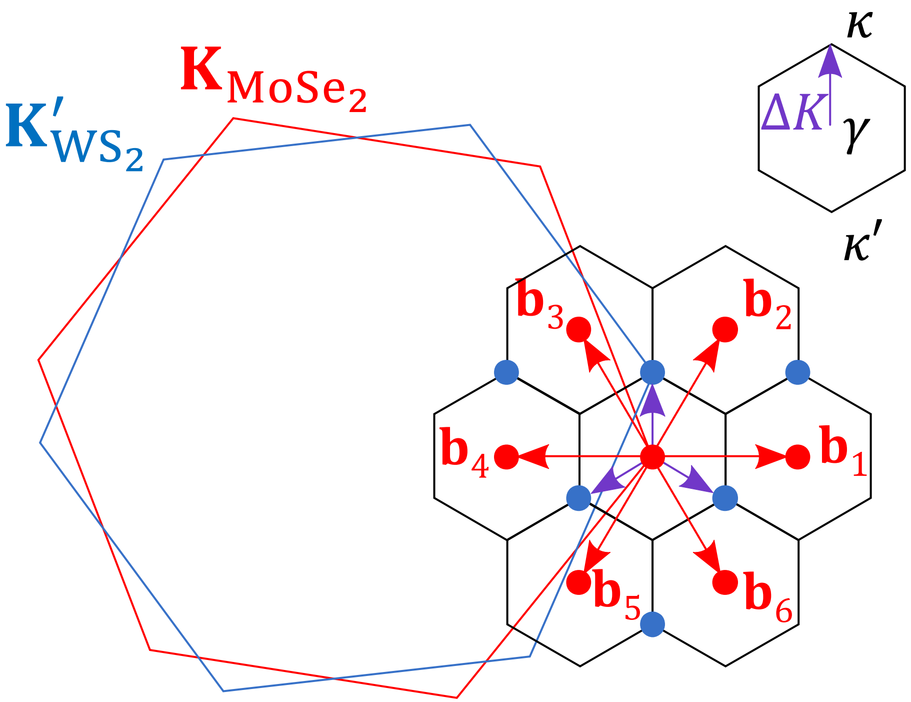


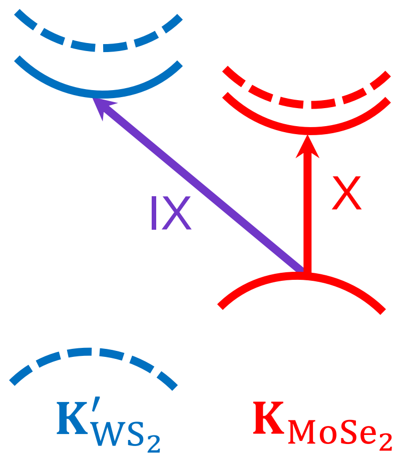
60 brillouin zones of a hexagonal lattice



https://github.com/hamdav/brillouinzones

Effective model for intra- and interlayer moiré excitons





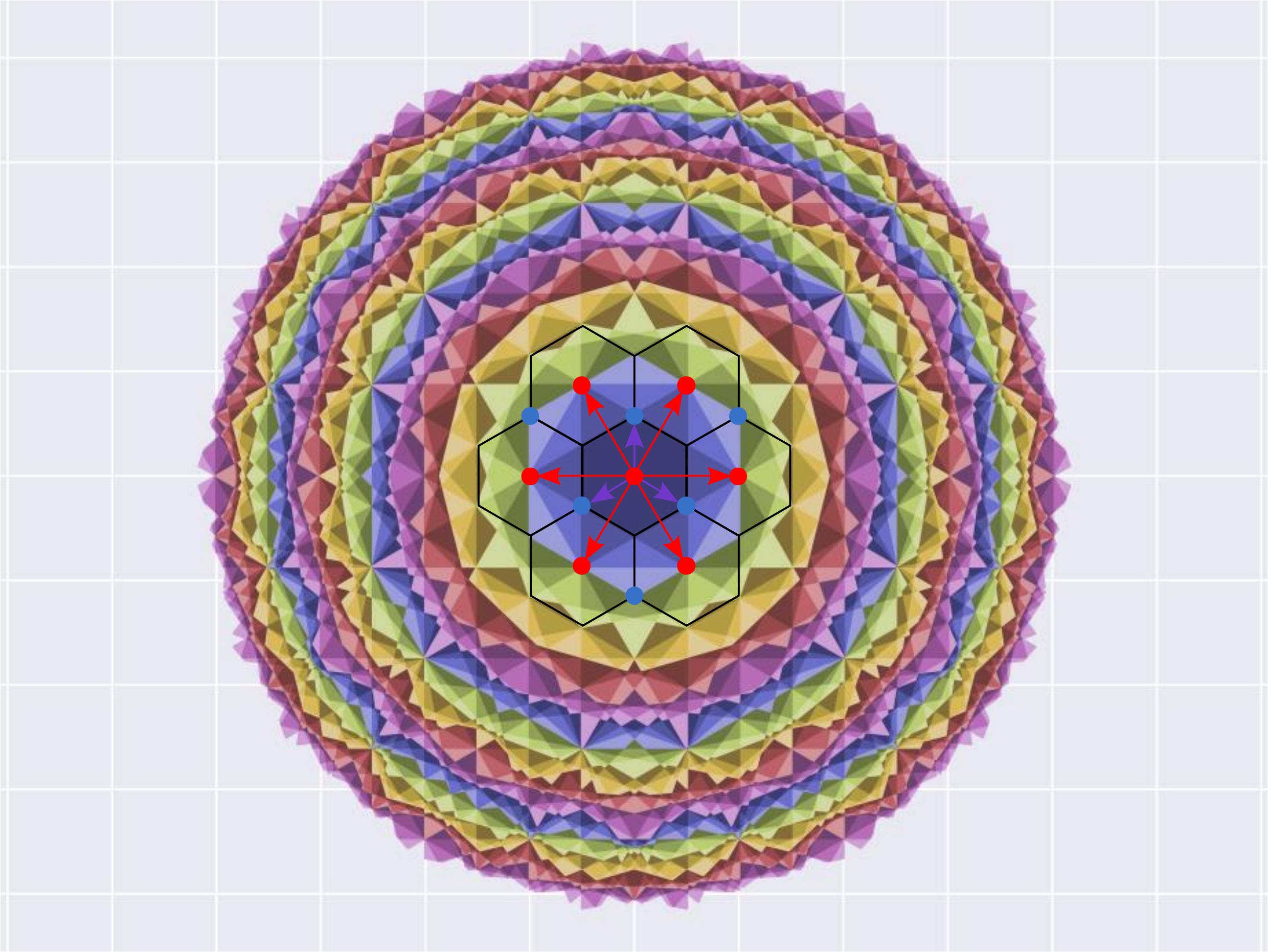
Intralayer potential

$$V(\mathbf{r}) = \sum^{6} V_j \exp\left(i\mathbf{b}_j\mathbf{r}\right)$$

Interlayer Coupling

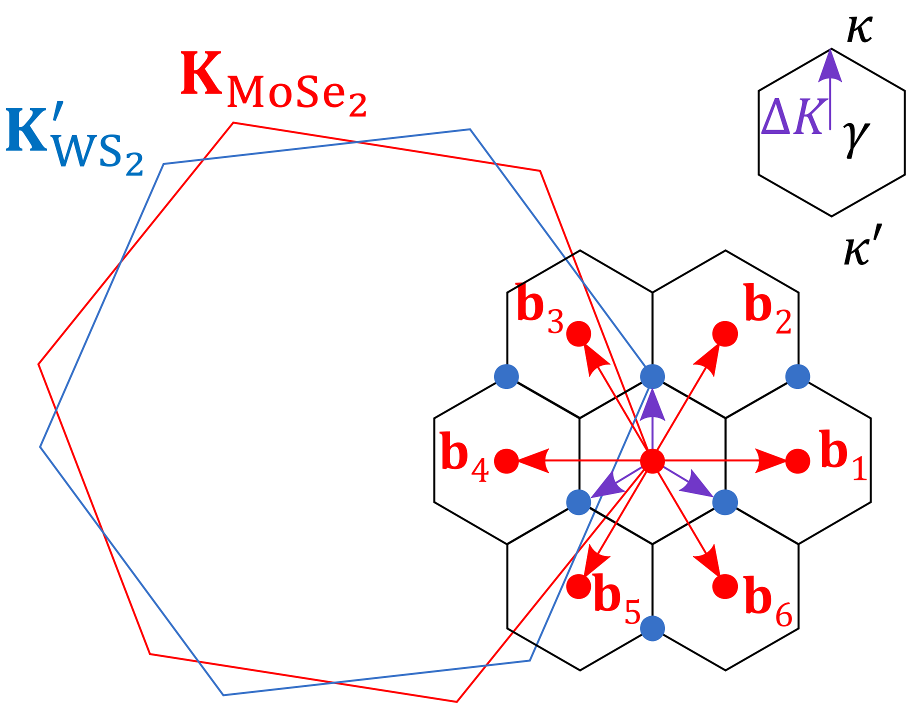
$$\langle \mathrm{IX}, \mathbf{k}' + \mathbf{g}' | T | \mathrm{X}, \mathbf{k} + \mathbf{g} \rangle = \sum_{\eta=0}^{2} t\delta_{\mathbf{k}+\mathbf{g}-\mathbf{k}'-\mathbf{g}', C_3^{\eta}\Delta\mathbf{K}}$$

Four mini Brillouin zones



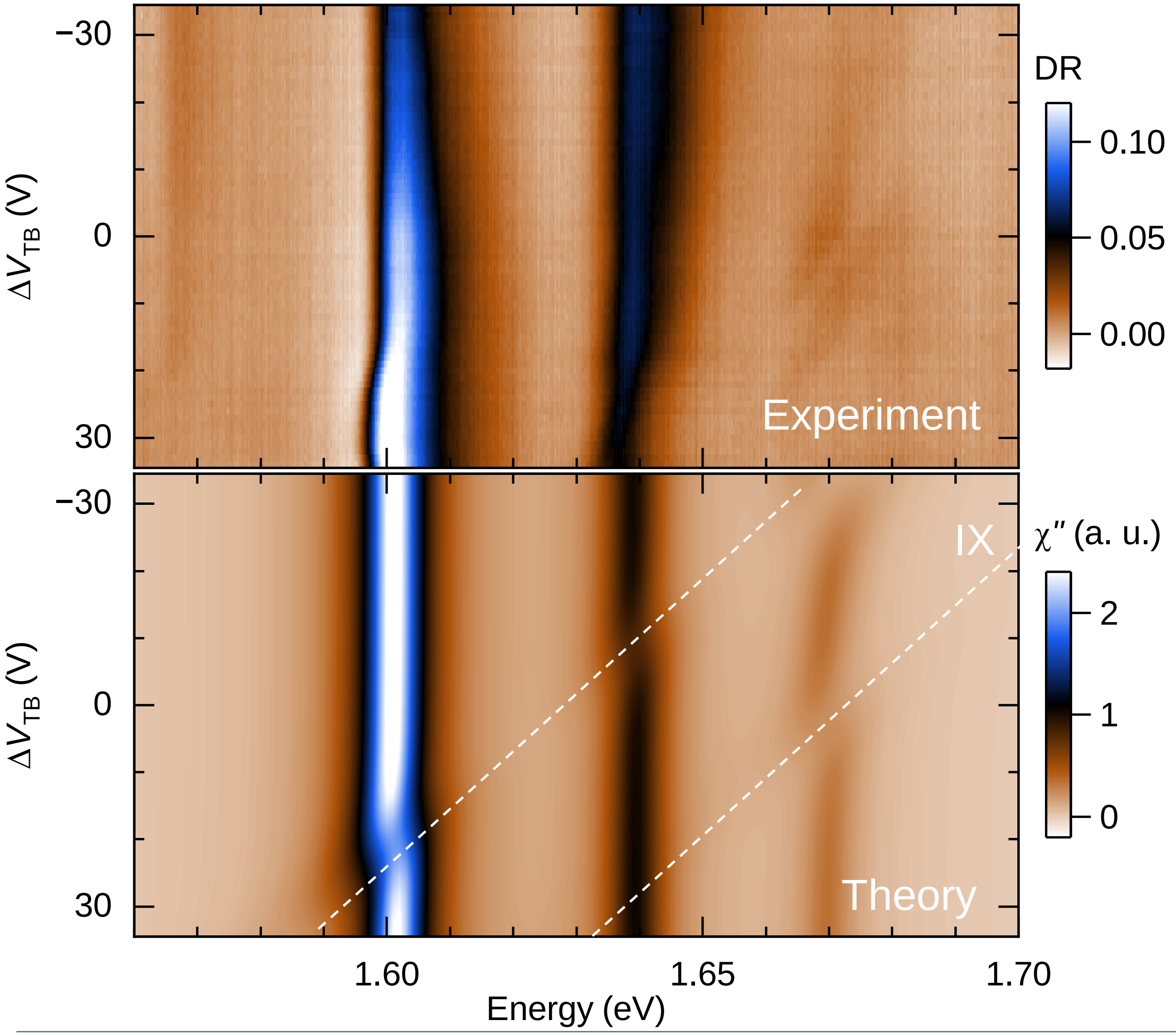
https://github.com/hamdav/brillouinzones

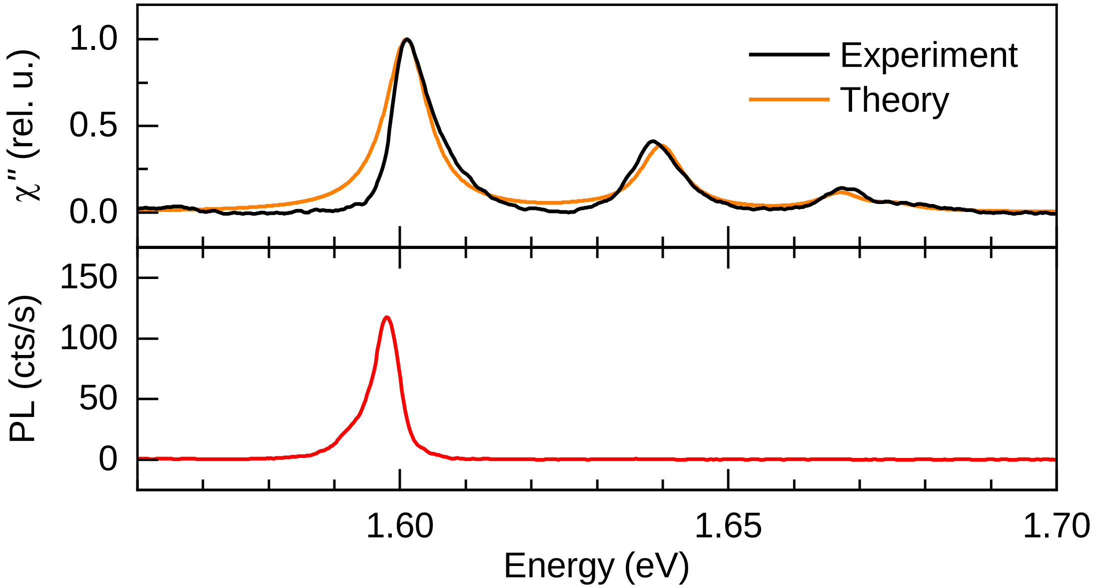
13-miniband Hamiltonian for moiré excitons



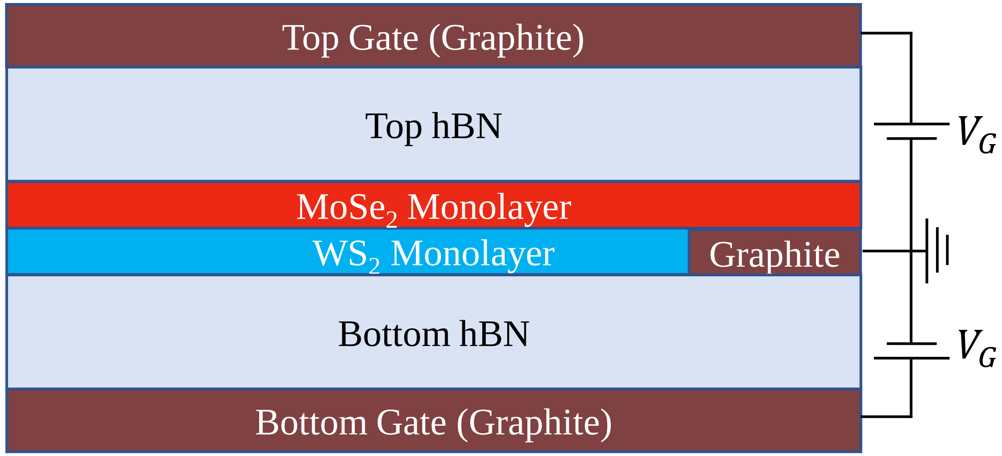
$$H(\mathbf{k}) = \begin{pmatrix}
E_0 & V & V^* & V & V^* & V & V^* & t & t & t & 0 & 0 & 0 \\
V^* & E_1 & V & 0 & 0 & 0 & V & 0 & 0 & t & 0 & t & 0 \\
V & V^* & E_2 & V^* & 0 & 0 & 0 & t & 0 & 0 & 0 & t & 0 \\
V^* & 0 & V & E_3 & V & 0 & 0 & t & 0 & 0 & 0 & 0 & t \\
V & 0 & 0 & V^* & E_4 & V^* & 0 & 0 & t & 0 & 0 & 0 & t \\
V^* & 0 & 0 & 0 & V & E_5 & V & 0 & t & 0 & t & 0 & 0 \\
V & V^* & 0 & 0 & 0 & V^* & E_6 & 0 & 0 & t & t & 0 & 0 \\
t^* & 0 & t^* & t^* & 0 & 0 & 0 & \mathcal{E}_0 & 0 & 0 & 0 & 0 & 0 \\
t^* & 0 & 0 & 0 & t^* & t^* & 0 & 0 & \mathcal{E}_1 & 0 & 0 & 0 & 0 \\
t^* & t^* & 0 & 0 & 0 & 0 & t^* & 0 & 0 & \mathcal{E}_2 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & t^* & t^* & 0 & 0 & 0 & \mathcal{E}_3 & 0 & 0 \\
0 & t^* & t^* & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \mathcal{E}_4 & 0 \\
0 & 0 & 0 & t^* & t^* & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \mathcal{E}_5
\end{pmatrix}$$

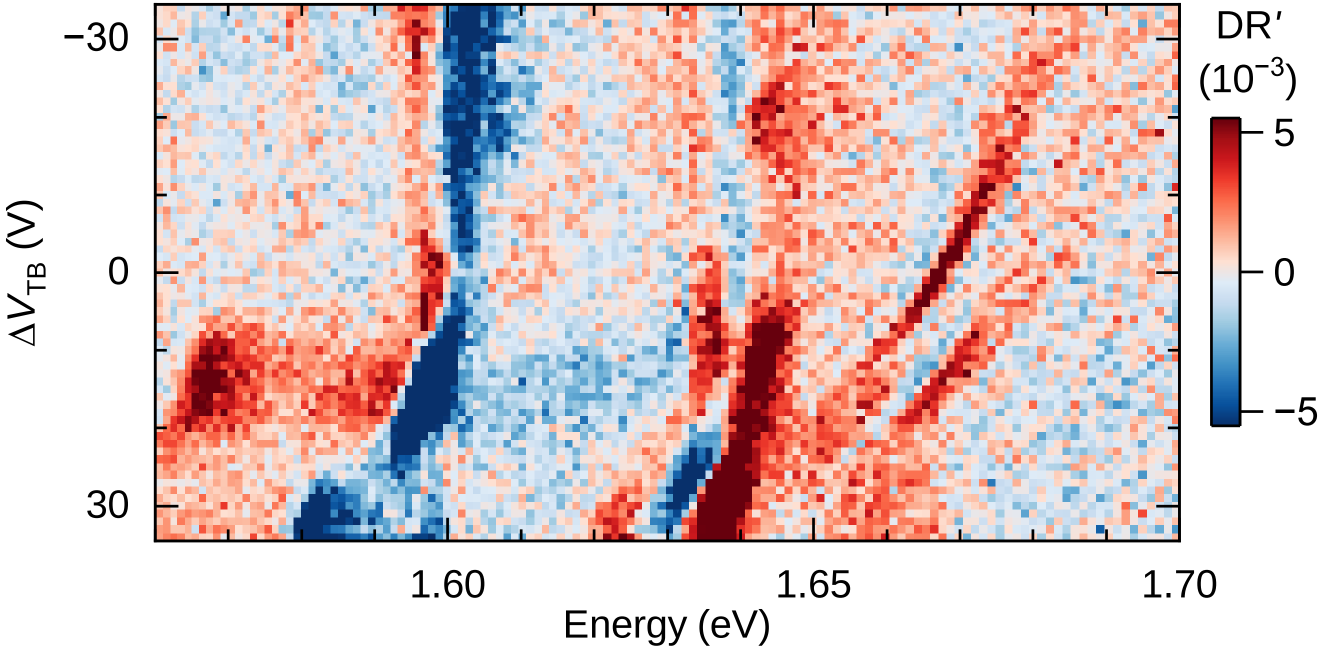
Fitting to E-field dependent data



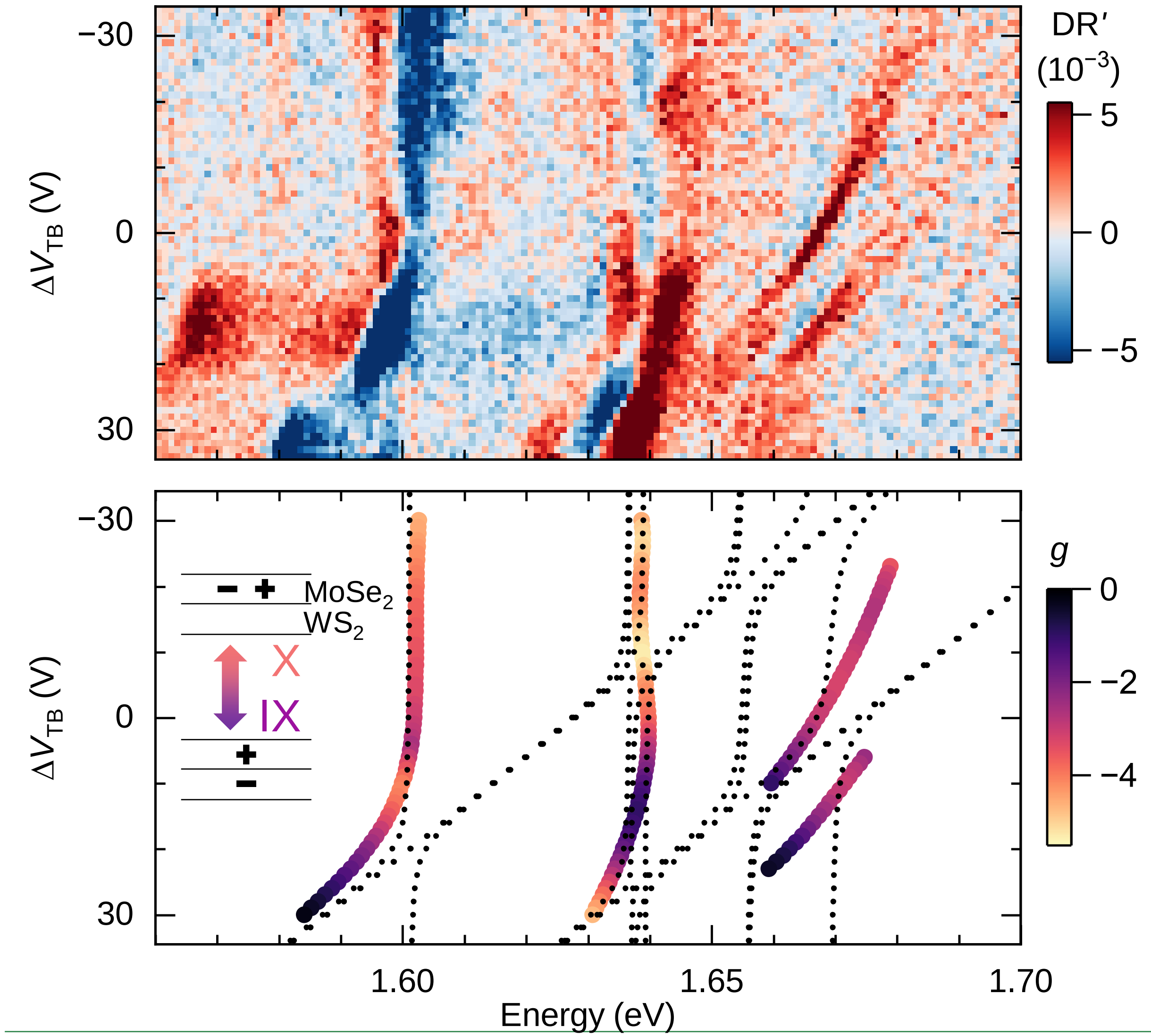


Making the interlayer excitons visible





Making the interlayer excitons visible

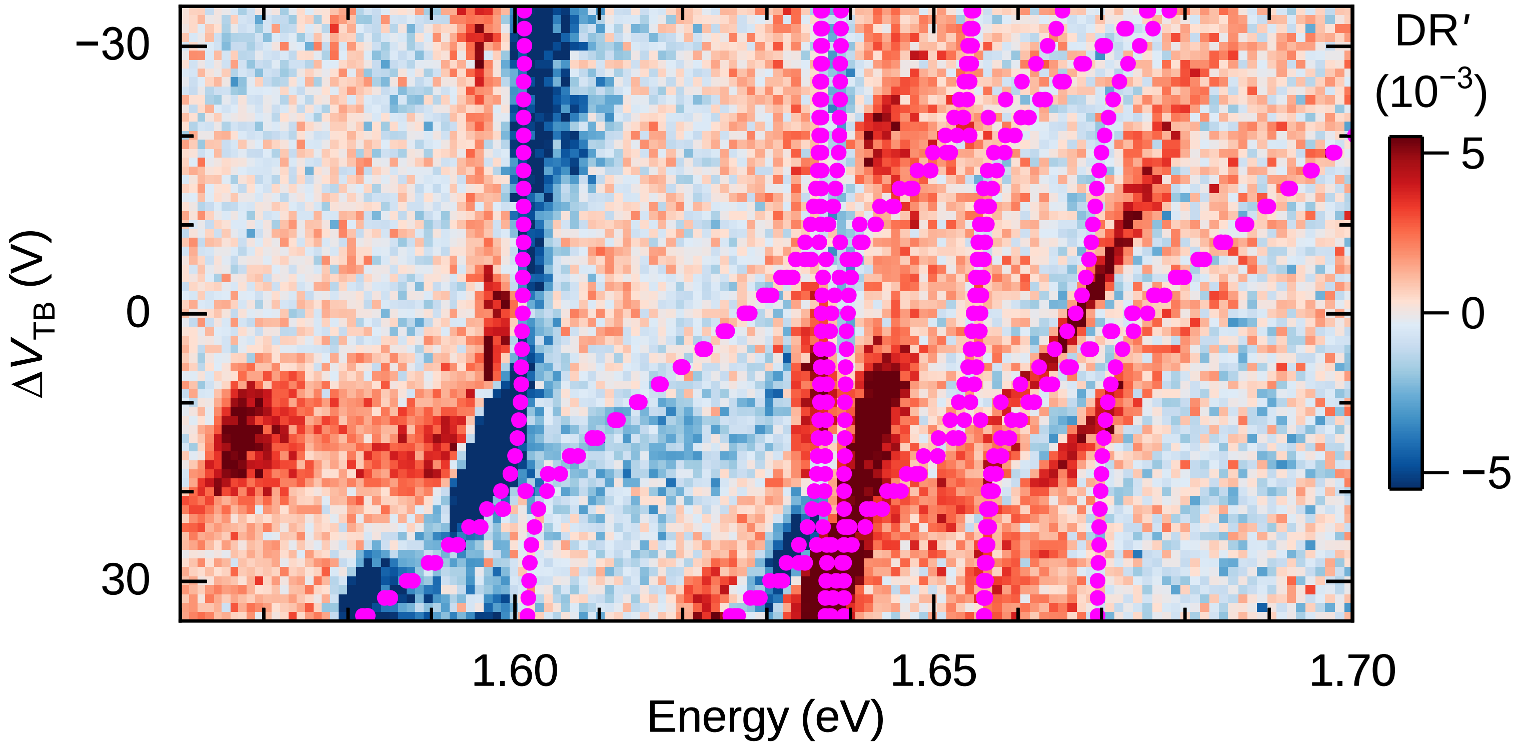


Dotted Lines: Eigenvalues of the Hamiltonian irrespective of the oscillator strength

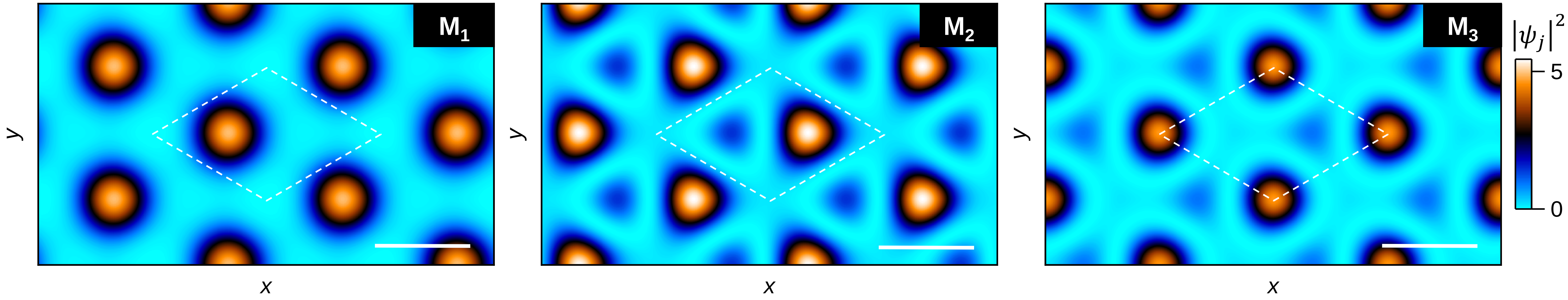
Colors of the data points: deviations of the exciton Landé g-factor from the intralayer value g = -4

Band alignment

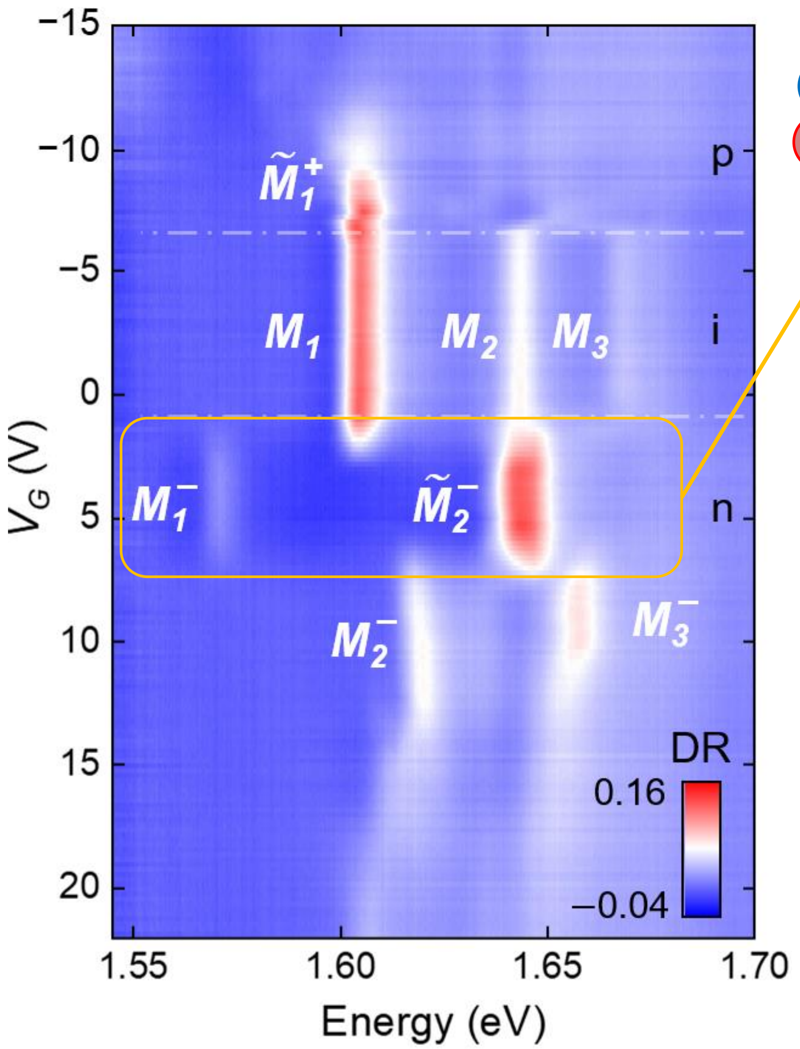
- At electic field ~0.05 V/nm the intra- and interlayer states exciton are resonant

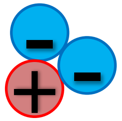


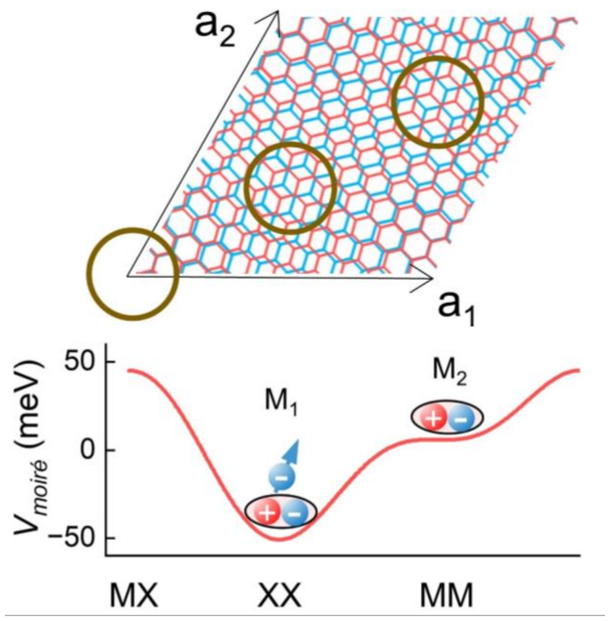
Spatial distribution of bright moiré excitons



Charge doping effects on moiré excitons







Summary

- A simple effective model combining phenomenological moiré potentials with resonant interlayer hopping describes intra- and interlayer moiré excitons simultaneously;
- $MoSe_2/WS_2$ is of type I alignment, and the first IX state lies ~30meV above the intralayer ground state exciton;
- A field of ~0.05 V/nm is sufficient to bring the intra- and interlayer excitons into resonance, and beyond that type II alignment can be reached;
- Moiré excitons localize in real space and can act as reporters on band alignment or charge order.

Editors' Suggestion

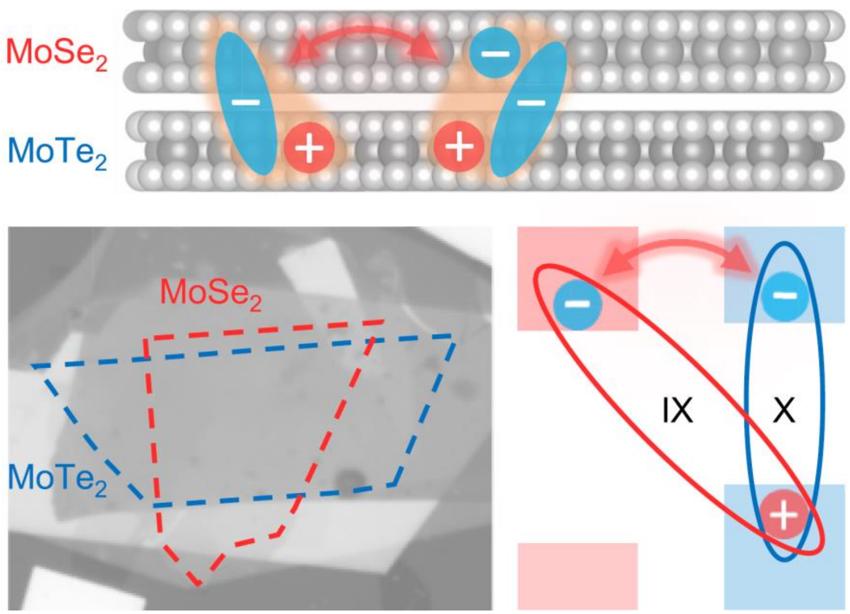
Field-Induced Hybridization of Moiré Excitons in $MoSe_2/WS_2$ Heterobilayers

Borislav Polovnikov, Johannes Scherzer, Subhradeep Misra, Xin Huang, Christian Mohl, Zhijie Li, Jonas Göser, Jonathan Förste, Ismail Bilgin, Kenji Watanabe, Takashi Taniguchi, Alexander Högele, and Anvar S. Baimuratov

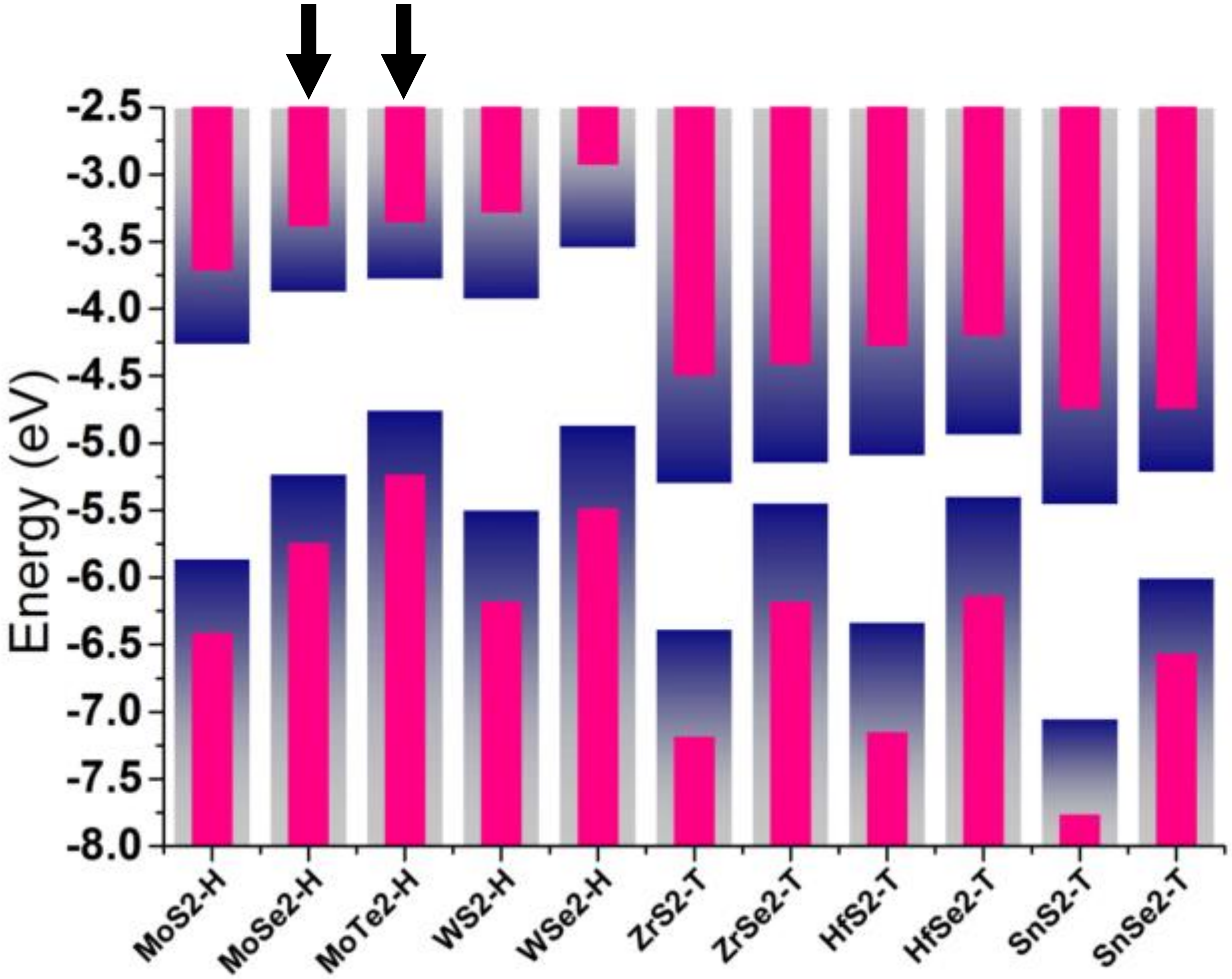
Phys. Rev. Lett. **132**, 076902 – Published 16 February 2024

Heterobilayer $MoTe_2$ / $MoSe_2$

- Lattice mismatch ~ 7%
- $MoTe_2$ and $MoSe_2$ have (almost) resonant CBs, allowing for intra- & interlayer exciton hybridization
- Holes are in $MoTe_2$ layer

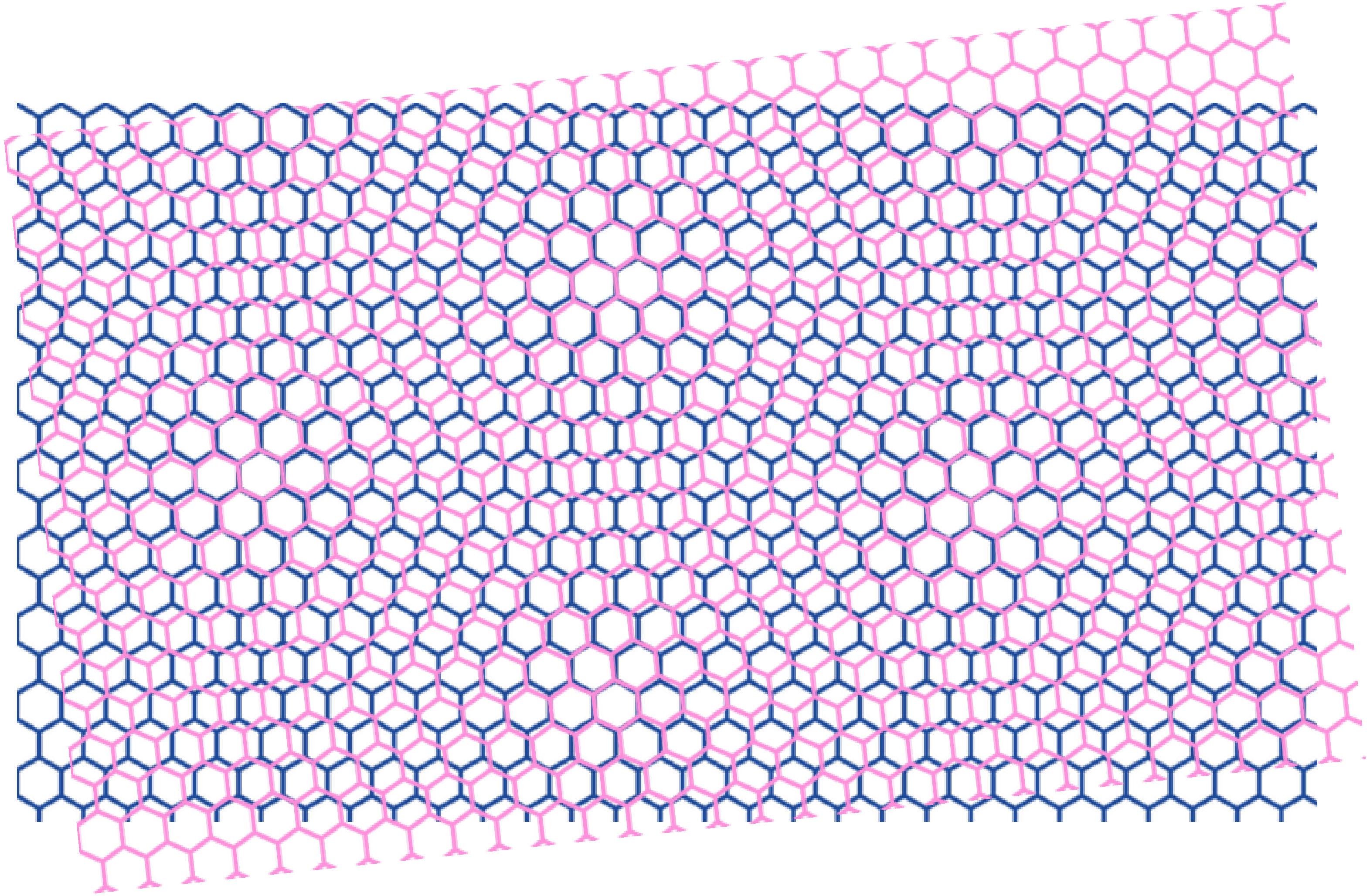


Zhao, Huang, …, Högele, Baimuratov, Nano Lett. 24, 4917 (2024)

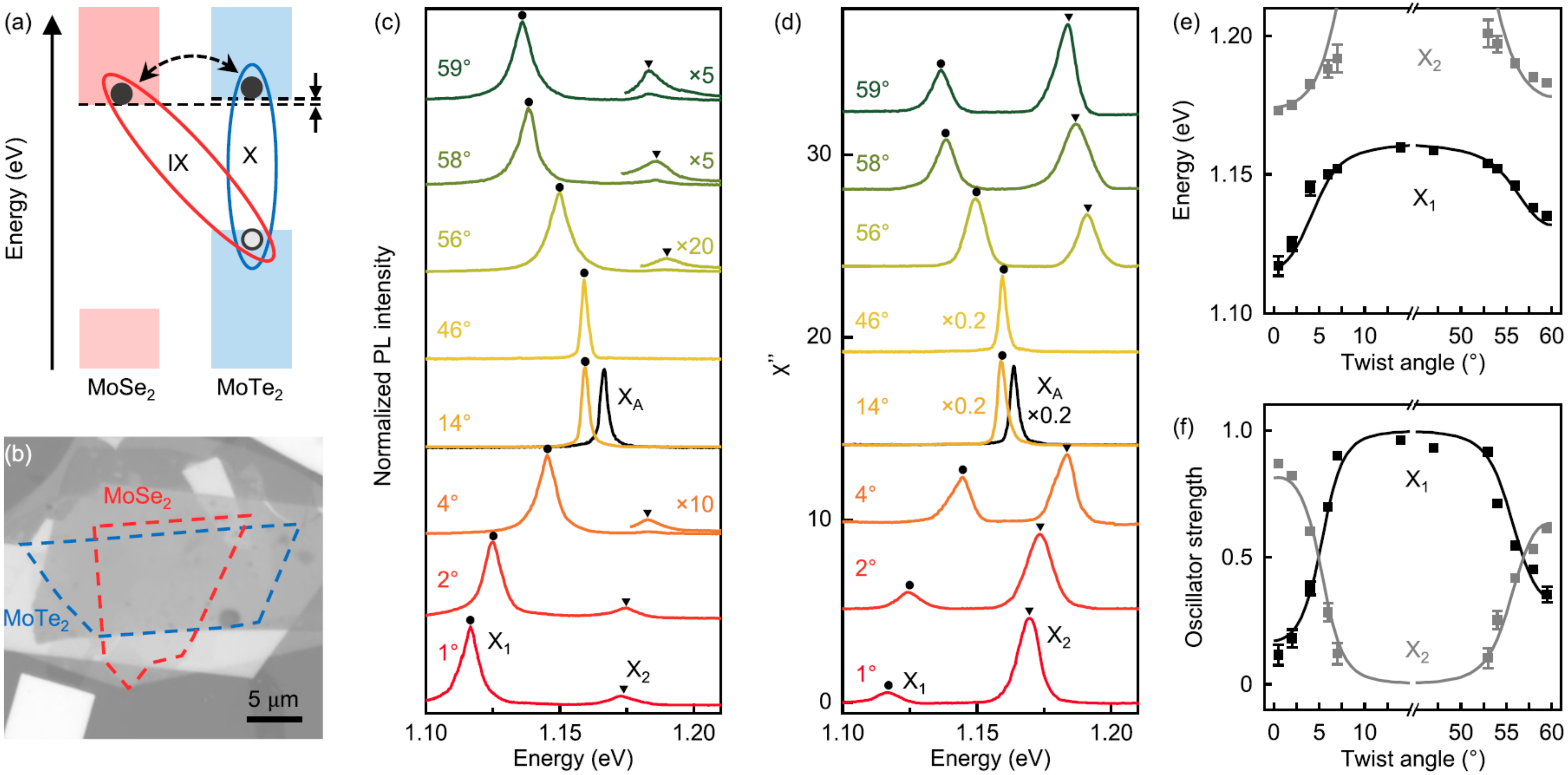


Zhang et al., 2D Mater. 4, 015026 (2016)

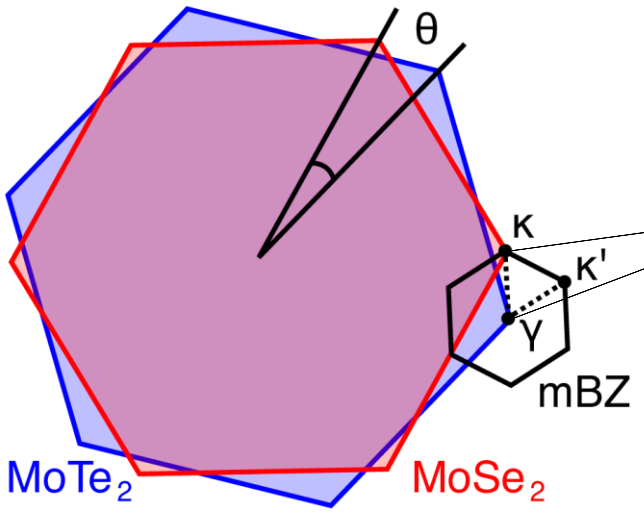
Heterobilayer $MoTe_2$ / $MoSe_2$



Heterobilayer $MoTe_2$ / $MoSe_2$



Theory for hybridization



$$\mathbf{b}_{1,2} = \frac{\sqrt{3}}{2}\begin{bmatrix}\sqrt{3} & \pm 1 \\ \mp 1 & \sqrt{3}\end{bmatrix}\Delta\mathbf{K}$$

Momentum mismatch

Intralayer excitons in $MoTe_2$

$$H = \begin{pmatrix} H_{\mathrm{X}} & T \\ T^* & H_{\mathrm{IX}} \end{pmatrix}$$

Hopping

Interlayer excitons

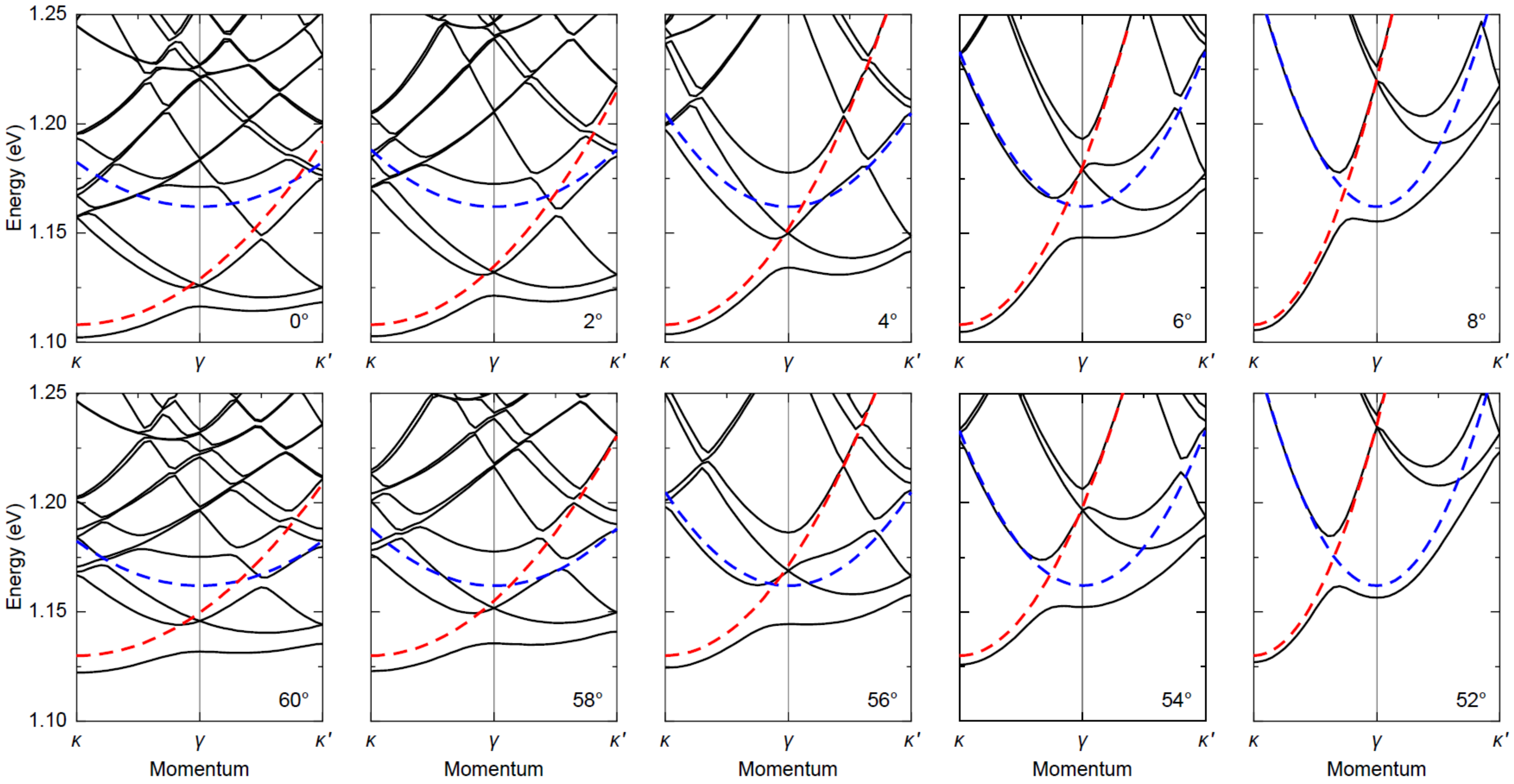
$$\langle \mathrm{X}, \mathbf{k}+\mathbf{g}'|H_{\mathrm{X}}|\mathrm{X}, \mathbf{k}+\mathbf{g}\rangle = \delta_{\mathbf{g},\mathbf{g}'}\left(E_{\mathrm{X}} + \frac{\hbar^2|\mathbf{k}+\mathbf{g}|^2}{2M_{\mathrm{X}}}\right) + \sum_{j=1}^{6} V_j \delta_{\mathbf{g}-\mathbf{g}',\mathbf{G}_j}$$

$$\langle \mathrm{IX}, \mathbf{k}'+\mathbf{g}'|H_{\mathrm{IX}}|\mathrm{IX}, \mathbf{k}'+\mathbf{g}\rangle = \delta_{\mathbf{g},\mathbf{g}'}\left(E_{\mathrm{IX}} + \frac{\hbar^2|\mathbf{k}+\mathbf{g}'|^2}{2M_{\mathrm{IX}}}\right)$$

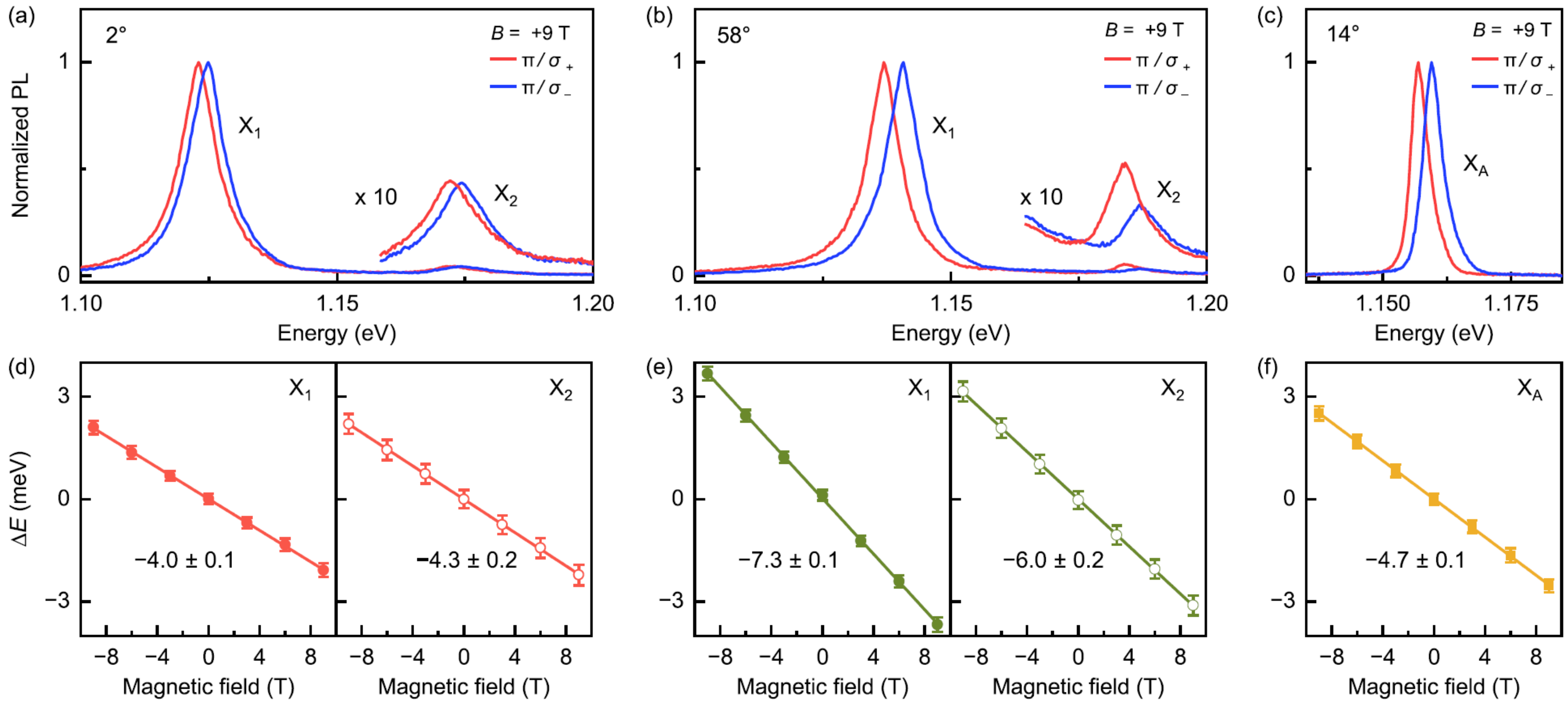
$$\langle \mathrm{IX}, \mathbf{k}'+\mathbf{g}'|T|\mathrm{X}, \mathbf{k}+\mathbf{g}\rangle = t(\delta_{\mathbf{k}+\mathbf{g},\mathbf{k}'+\mathbf{g}'+\Delta\mathbf{K}} + \delta_{\mathbf{k}+\mathbf{g},\mathbf{k}'+\mathbf{g}'+C_3^1\Delta\mathbf{K}} + \delta_{\mathbf{k}+\mathbf{g},\mathbf{k}'+\mathbf{g}'+C_3^2\Delta\mathbf{K}})$$

Polovnikov, … , Baimuratov, PRL 32, 076902 (2024)

Twist angle tuning



g-factors of hybrid excitons



g-factors of hybrid excitons

	m_c/m_0	m_{c+1}/m_0	m_v/m_0	Δ_{SO}	L_c	L_{c+1}	L_v
$MoTe_2$	0.58	0.67	−0.68	69	1.586	1.204	3.872
$MoSe_2$	0.55	0.63	−0.64	23	1.798	1.526	3.977

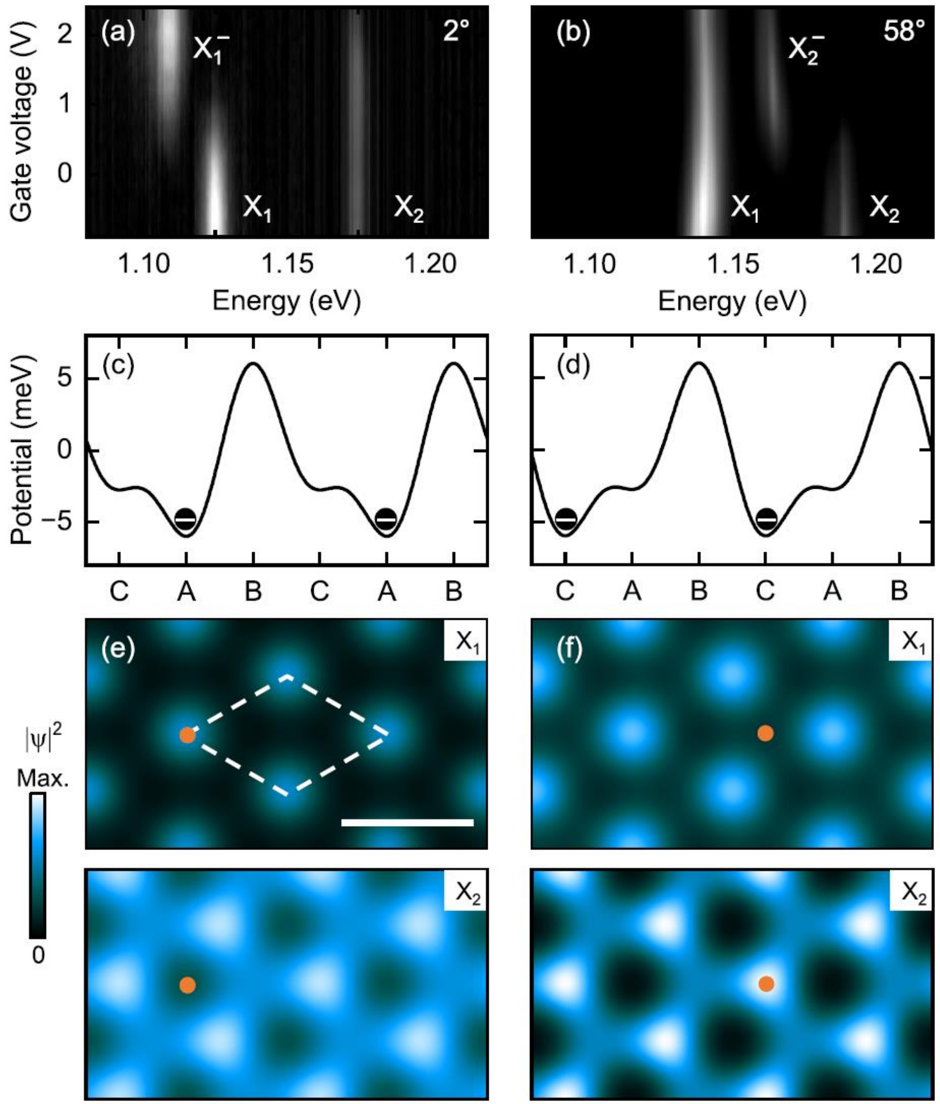
$$g_{\mathrm{A}} = 2(L_c - L_v)$$

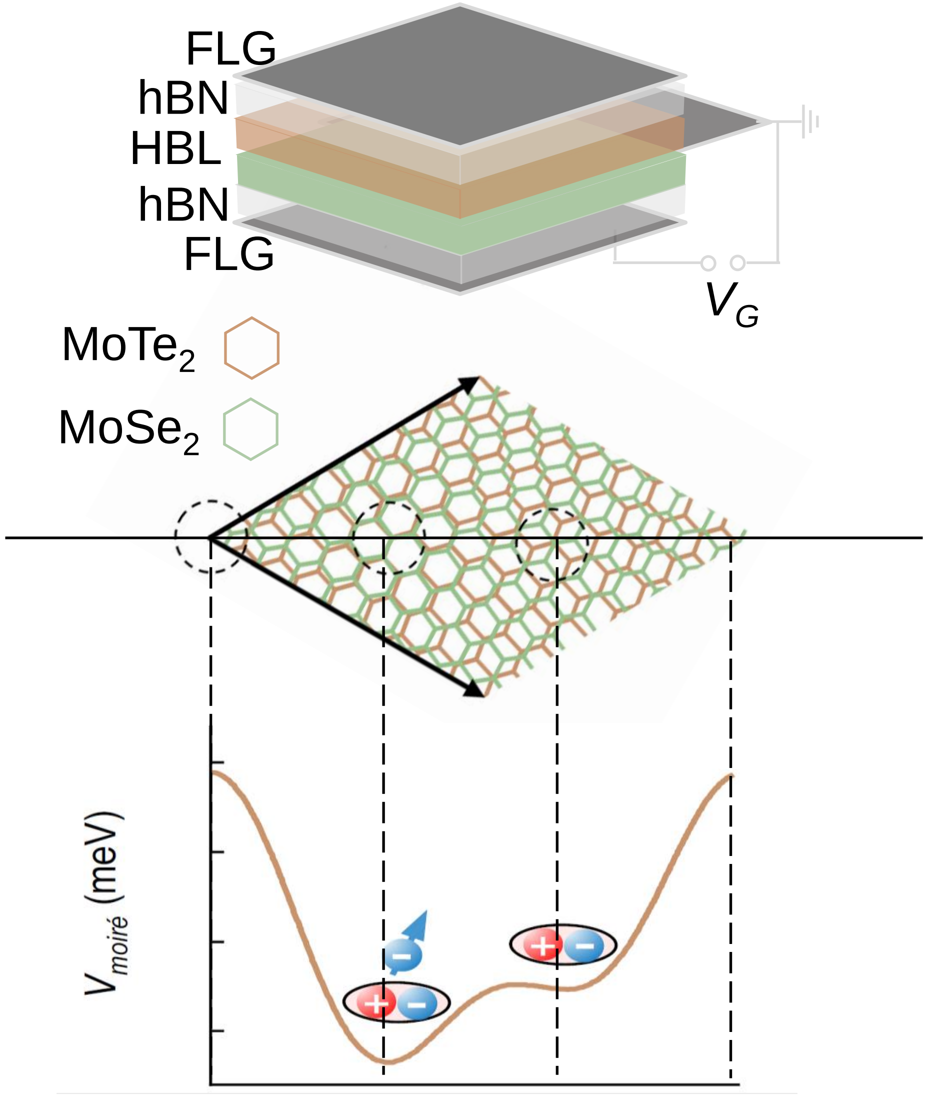
$$g_i^{(\mathrm{R})} = 2\left(f_i^{(\mathrm{X})} L_c + f_i^{(\mathrm{IX})} L_{c'} - L_v\right)$$

$$g_i^{(\mathrm{H})} = 2\left(f_i^{(\mathrm{X})} L_c - f_i^{(\mathrm{IX})} L_{c'+1} - L_v\right)$$

Twist angle	Exciton X_1		Exciton X_2	
	Exp.	Theory	Exp.	Theory
14°(R-type) and ML $MoTe_2$	4.7	4.6	-	-
2° (R-type)	4.0	4.2	4.3	4.4
58° (H-type)	7.3	8.0	6.0	6.8

Doping and trion formation





Summary

- We study properties of $MoTe_2/MoSe_2$ heterobilayer and compare our data with the developed model;
- Heterobilayer $MoTe_2/MoSe_2$ demonstrates strong effect of hybridization without additional application of electric field;
- Our work provides fundamental understanding of hybrid moiré excitons and trions in $MoTe_2$/ $MoSe_2$ heterobilayers and establishes the material system as a prime candidate for optical studies of correlated phenomena in moiré lattices.

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Hybrid Moiré Excitons and Trions in Twisted $MoTe_2$–$MoSe_2$ Heterobilayers

Shen Zhao,△ Xin Huang,*,△ Roland Gillen, Zhijie Li, Song Liu, Kenji Watanabe, Takashi Taniguchi, Janina Maultzsch, James Hone, Alexander Högele,* and Anvar S. Baimuratov*

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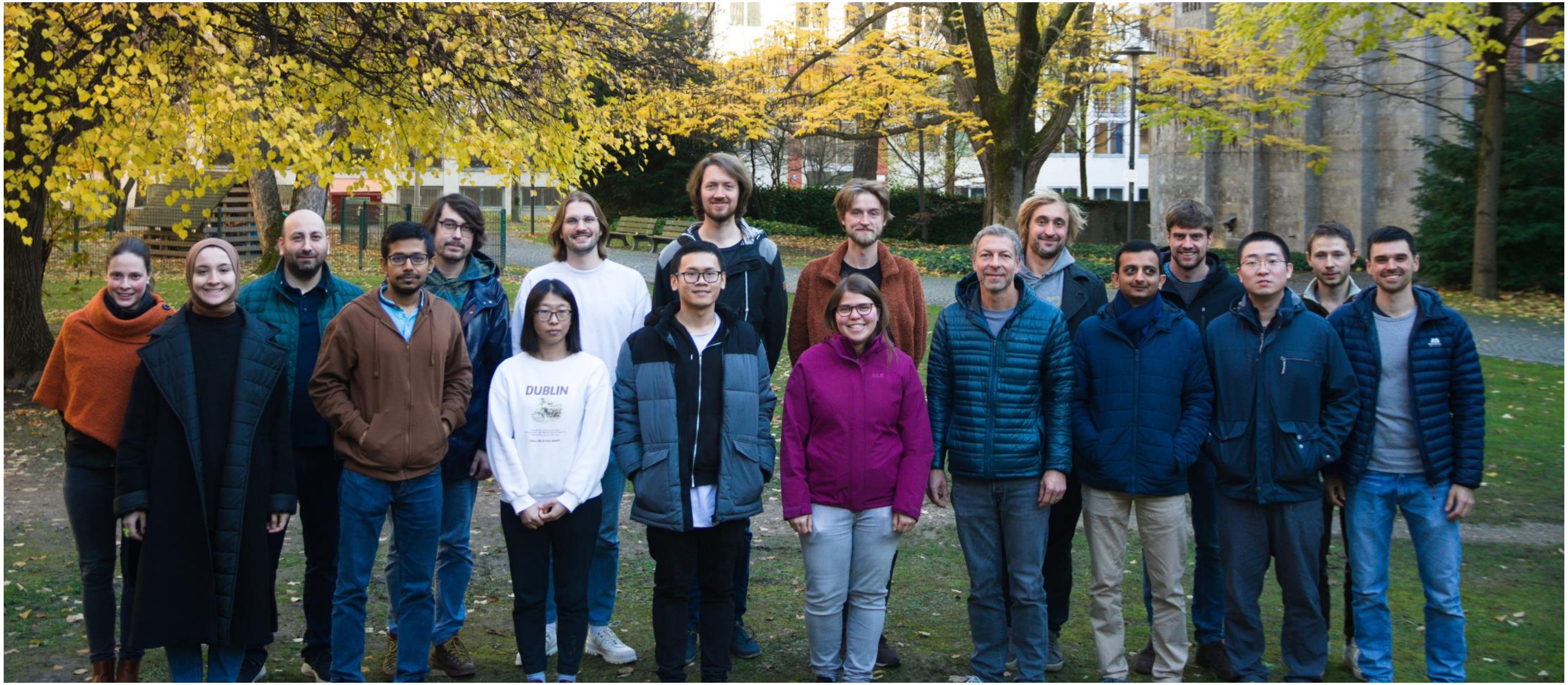
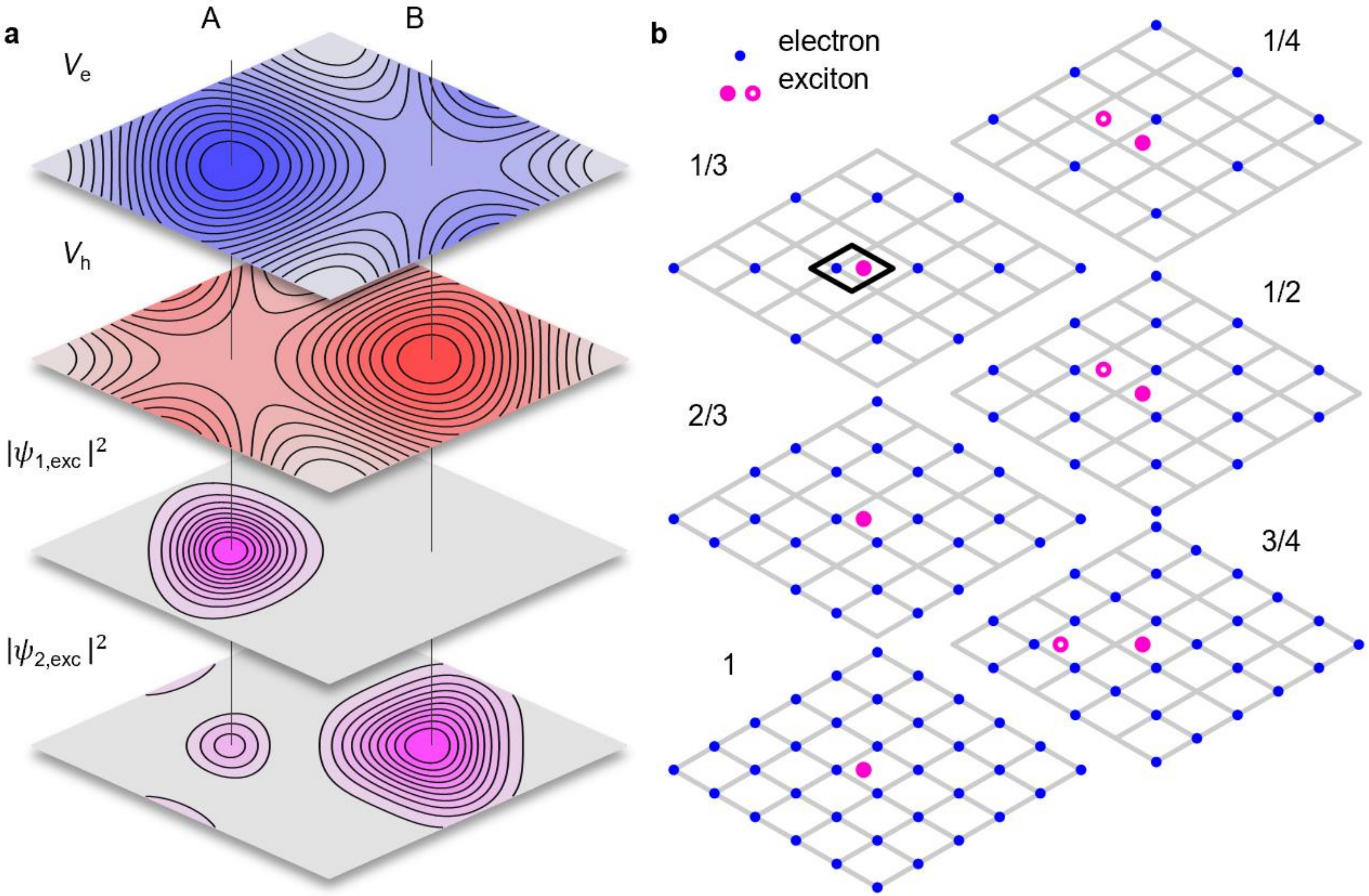


Remote excitons as sensors of correlations



Magnetization of the electron lattice