Moiré Excitons in

Semiconducting Moiré Superlattices

Chenhao Jin, July 20 2022

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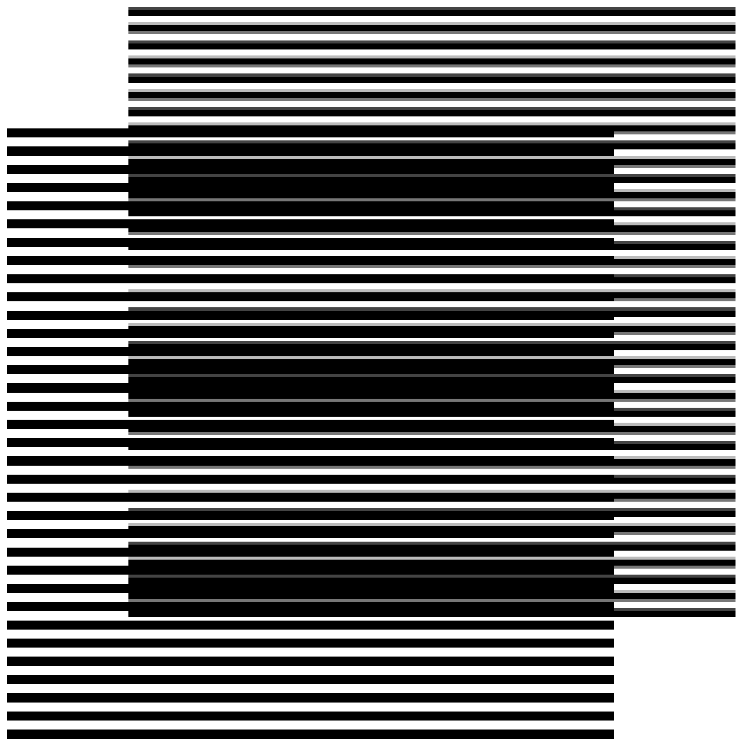
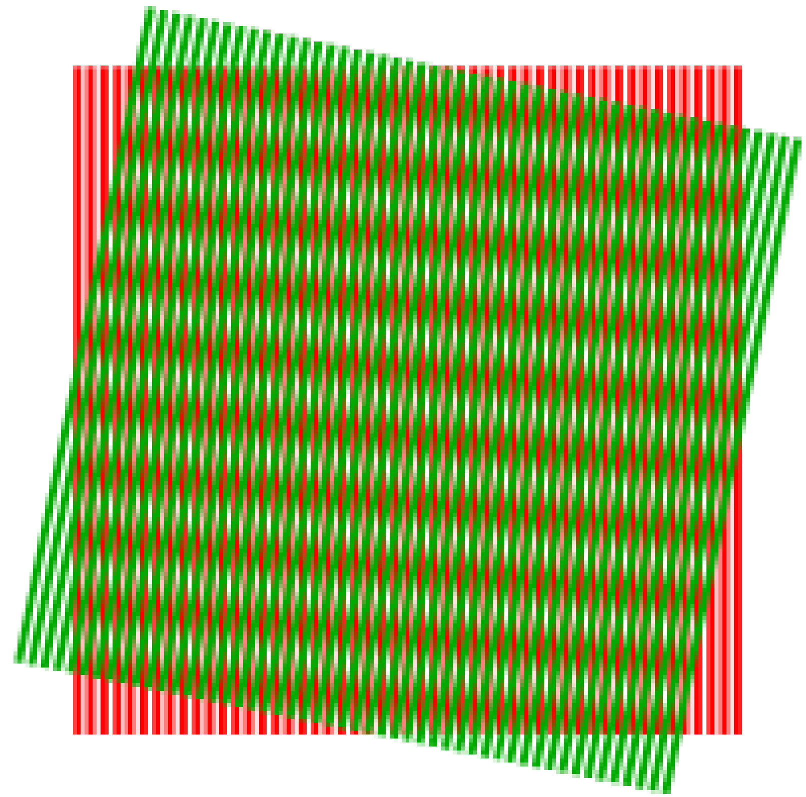
Outline

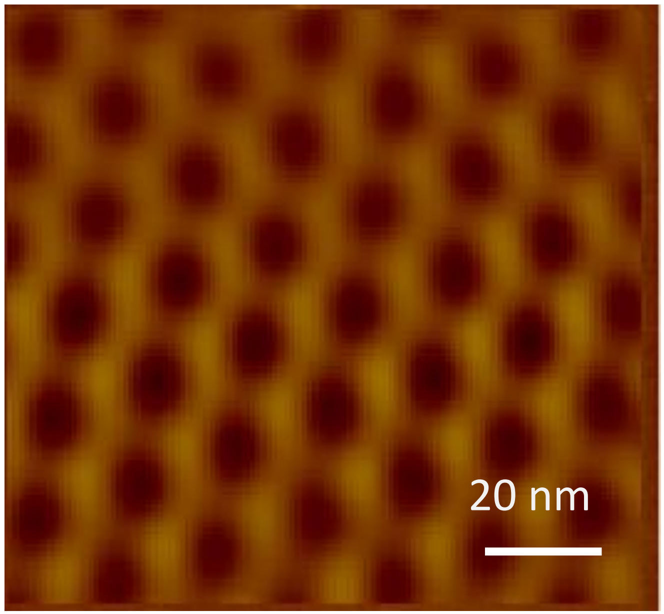
- **Introduction to moiré physics**
- **Moiré excitons: intralayer**
- **Moiré excitons: interlayer**

Moiré in Art:
interference produced by different patterns



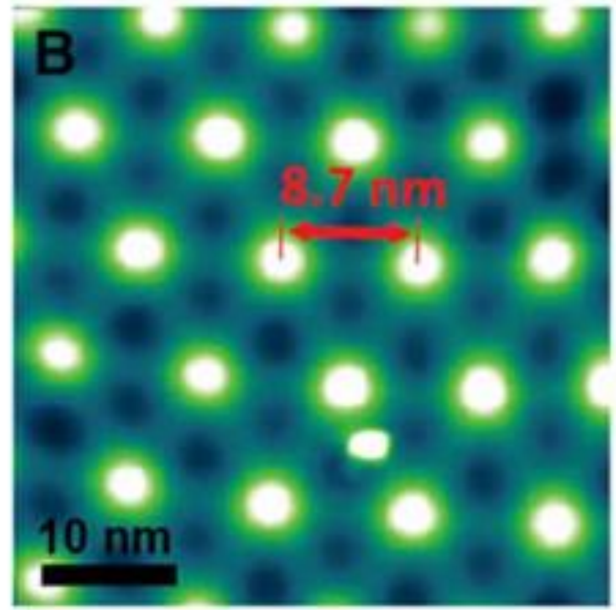


Moiré in Physics:
interference produced by different lattices



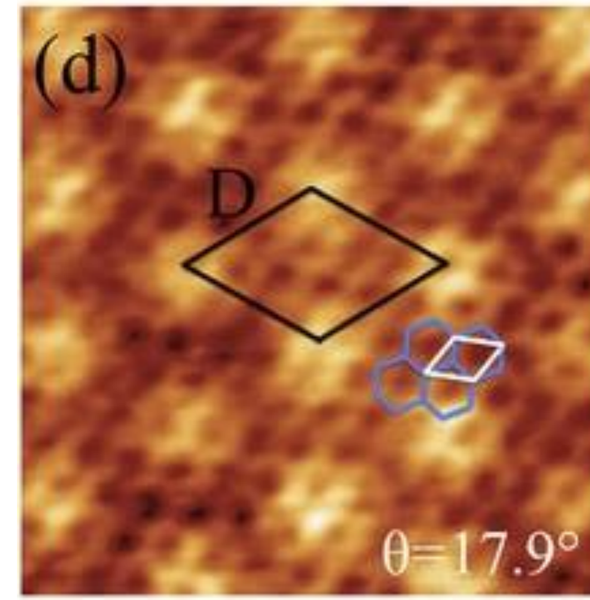
Nat. Phys. **10**, 743(2014)

Graphene/hBN



Sci. Adv. **3**, e1601459 (2017)

WSe2/MoS2

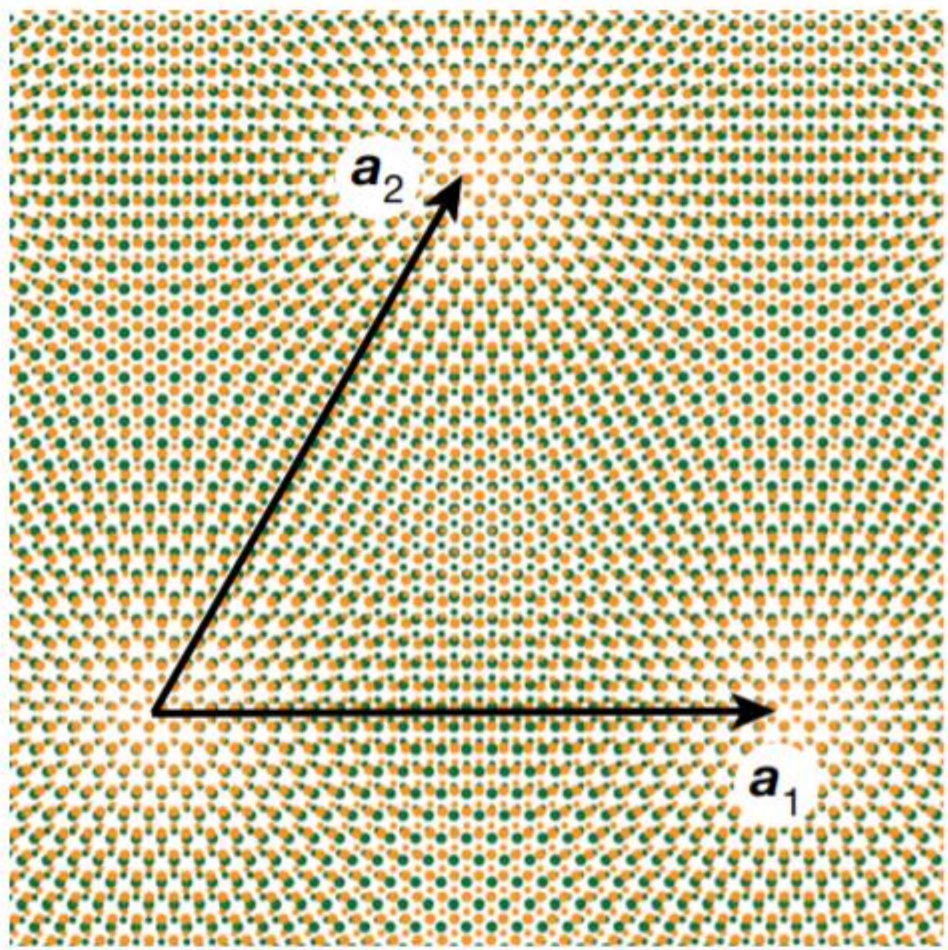


Sci. Rep. **6**, 27261 (2016)

Twisted bilayer graphene

2D van der Waals systems:
Unique platform for moiré physics

Rich physics from moiré pattern



$$H = H_0 + V_M$$

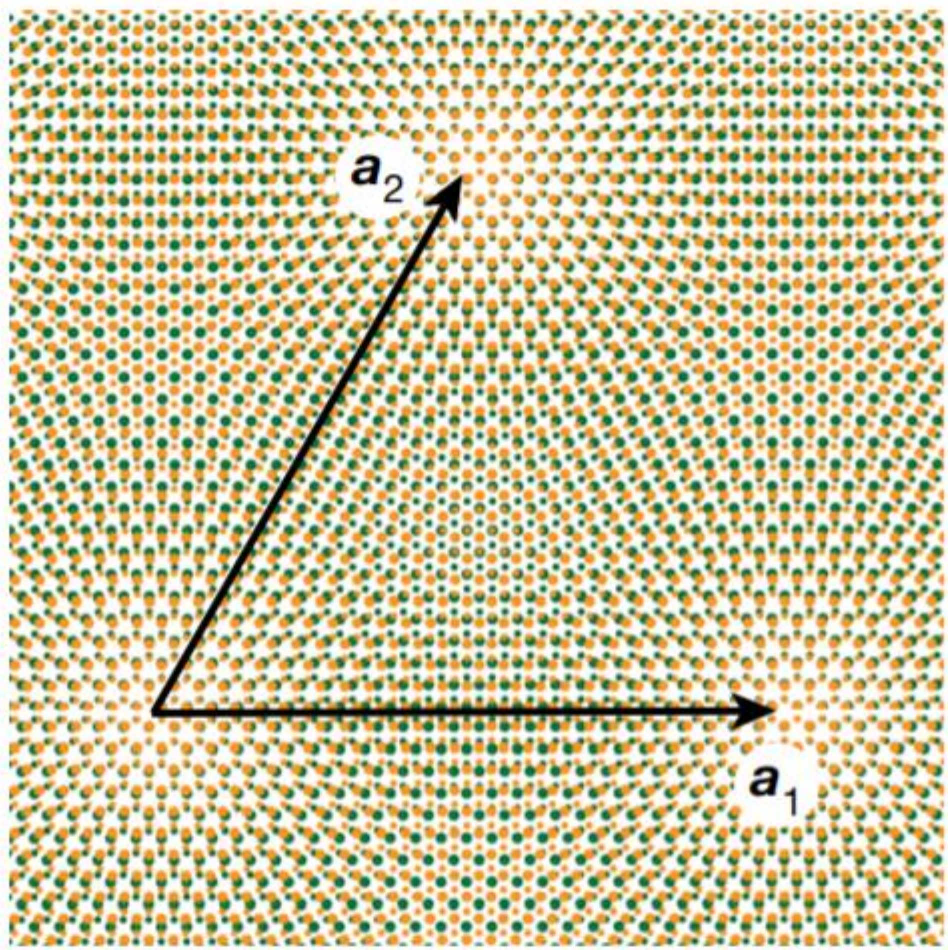
single layer

moiré potential

V_M has the periodicity of the moiré pattern

What will we get?

Rich physics from moiré pattern



$$H = H_0 + V_M$$

single layer   moiré potential

V_M has the periodicity of the moiré pattern

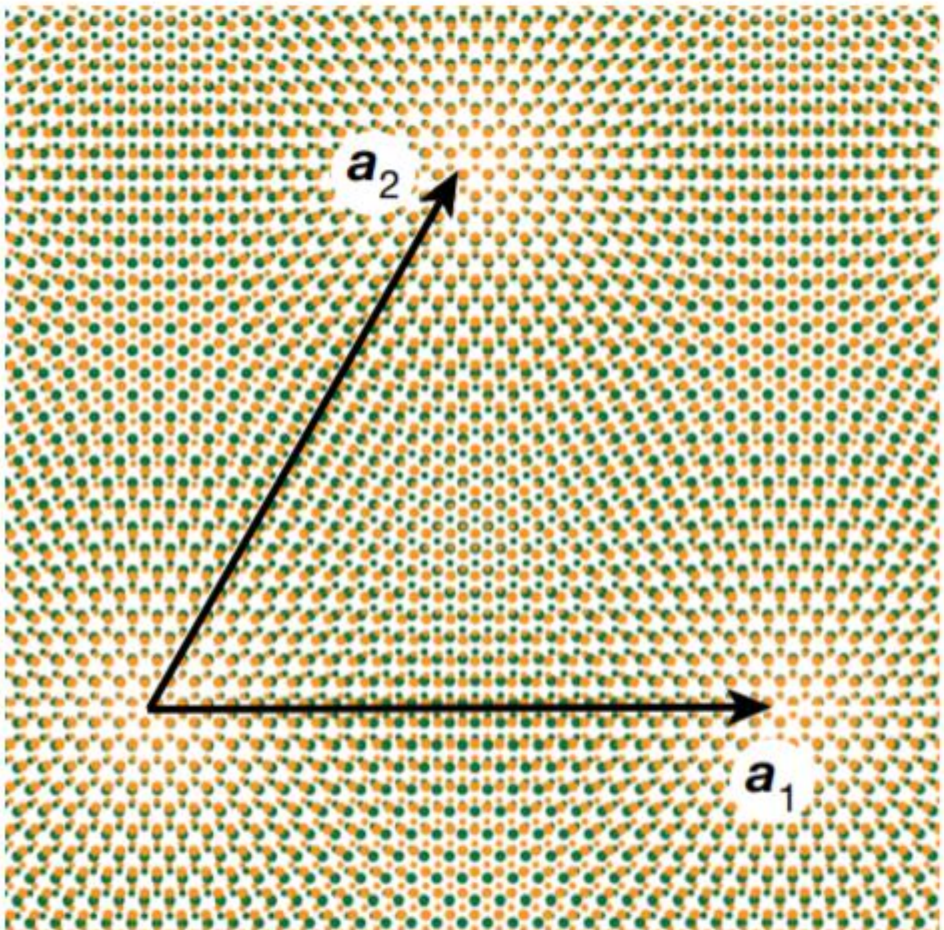
$$H = H_0 + V_L$$

kinetic energy   lattice potential

V_L has the periodicity of the crystal lattice

free electron -> Bloch bands

Artificial crystals from moiré superlattices



$$H = H_0 + V_M$$

single layer

moiré potential

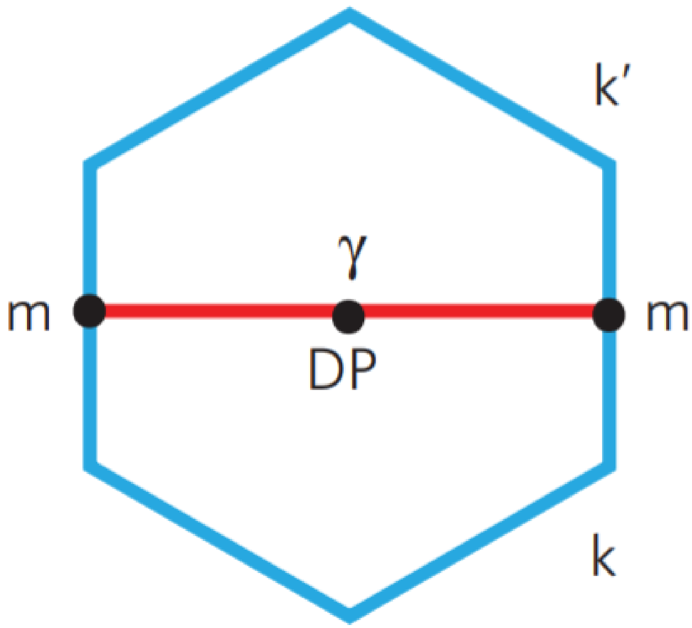
V_M has the periodicity of the moiré pattern

"free" electrons -> artificial solids

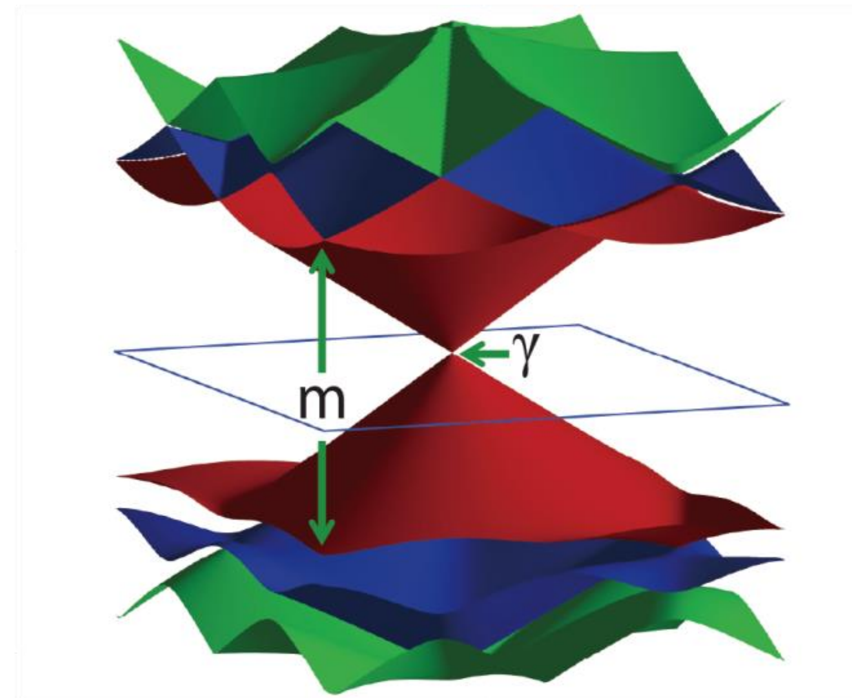
"superlattice" constant L_M defined by the moiré pattern

Artificial crystals from moiré superlattices

moiré/mini BZ



mini bands



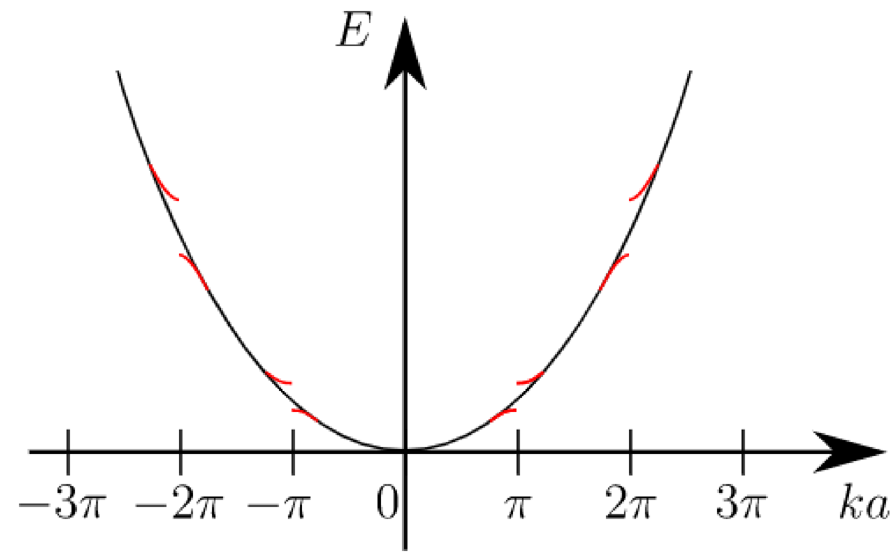
Rich physics should exist

What is missing?
A strong moiré potential

$$H = H_0 + O(V_L)$$

Weak potential

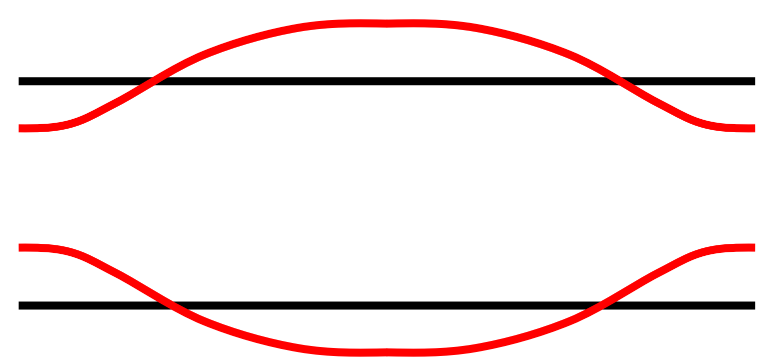
nearly-free electron picture



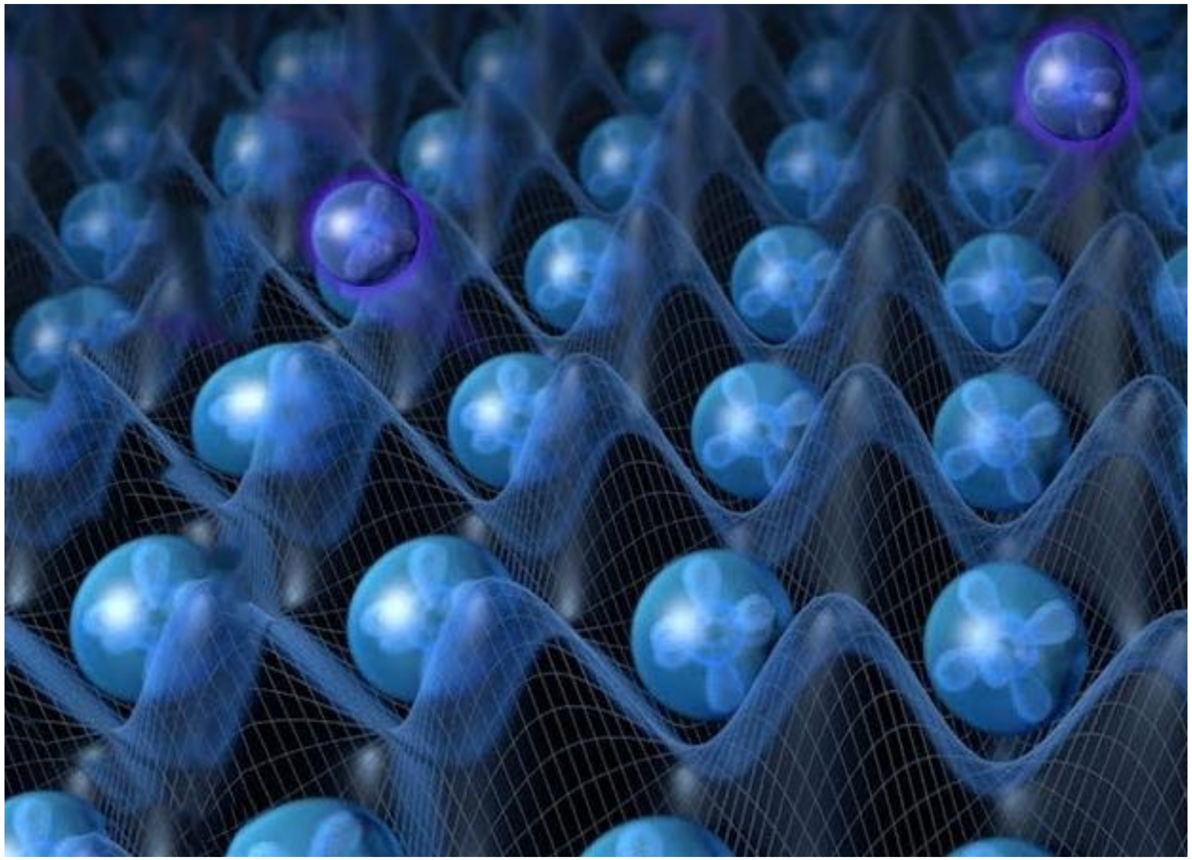
$$H = V_L + O(H_0)$$

Strong potential

tight-binding picture



With a strong moiré potential:
The "flat band" regime



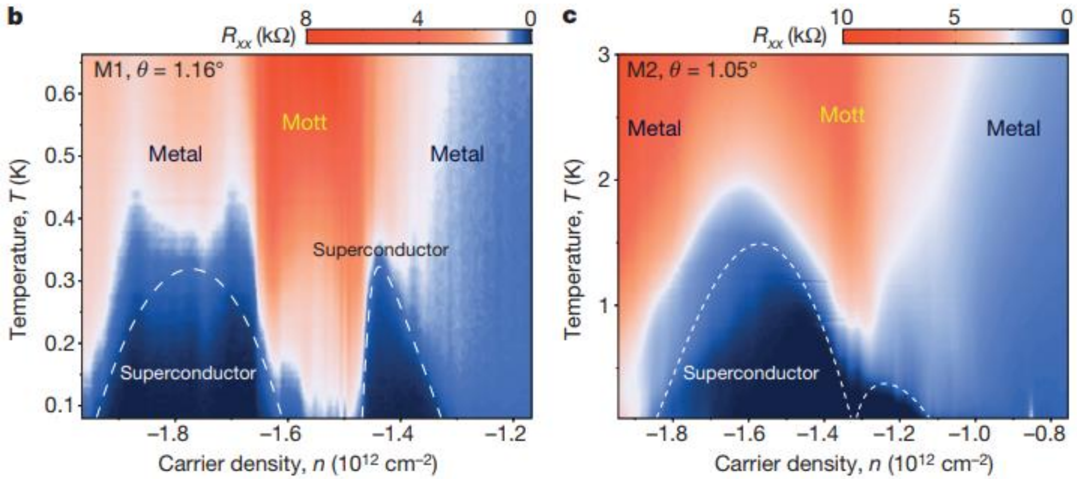
Artificial lattice from moiré trapping

trapping (moiré potential)	V_M determined by interlayer interaction
bandwidth (kinetic energy)	$t \sim \hbar^2/(mL_M^2)$ $\downarrow$ with V_M
on-site interaction (Coulomb energy)	$U \sim e^2/(\varepsilon L_M)$ $\uparrow$ with V_M

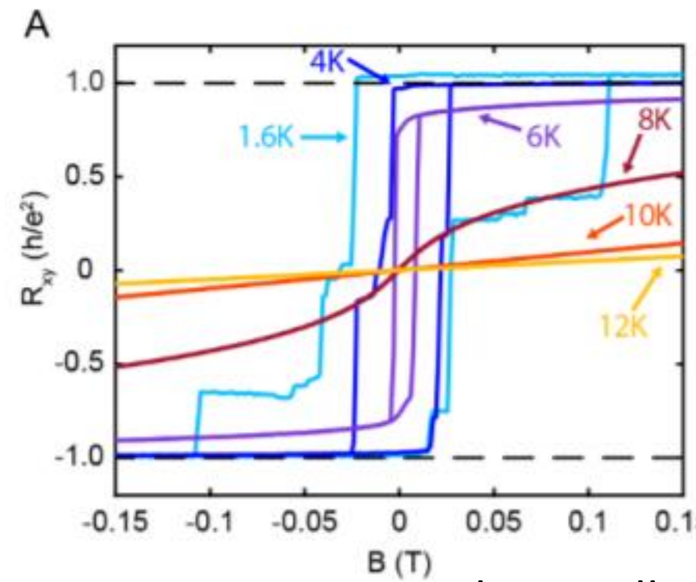
Tunability + Scalability, ~ meV energy scale

Image adapted from Sciencemag.org

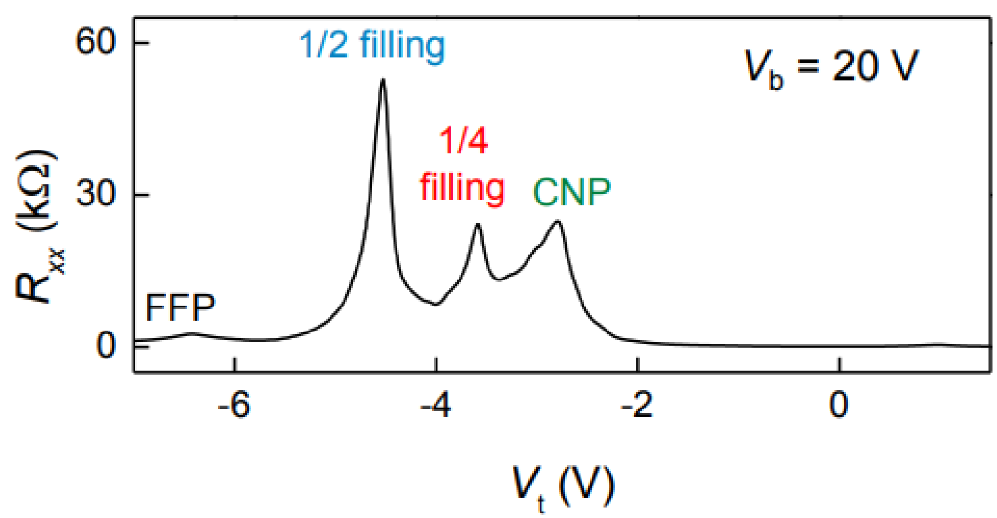
Rich physics from graphene-based moiré systems



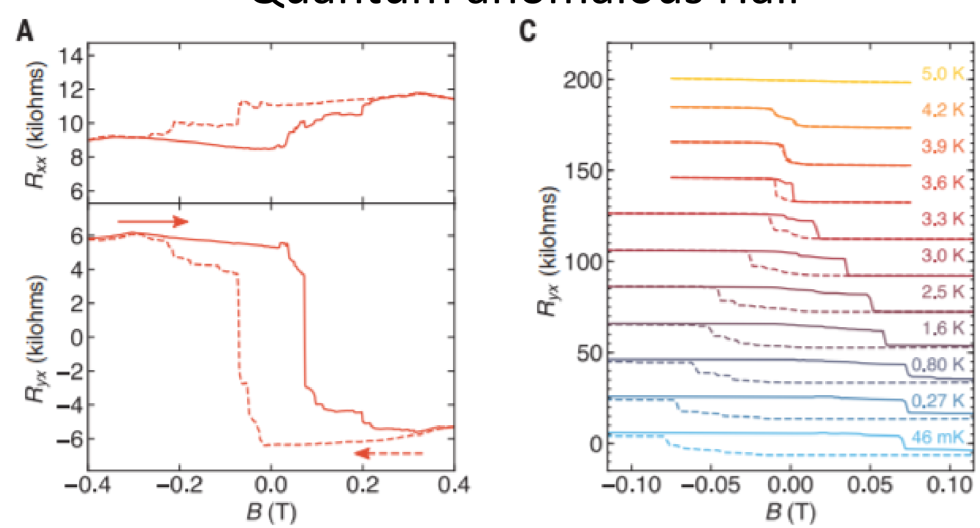
Superconductivity



Quantum anomalous Hall



Correlated insulator



Ferromagnetism

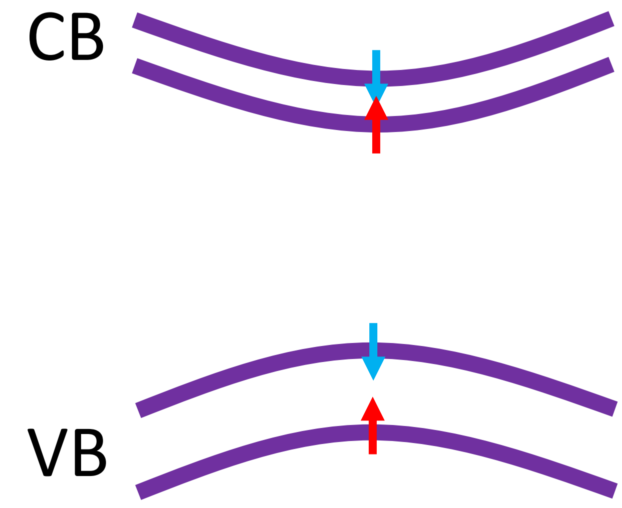
Pablo, Young, Wang, Gordon, Dean, …

Moiré superlattice in 2D semiconductors

- **Large SOC and electron mass**

  Single band Hubbard model

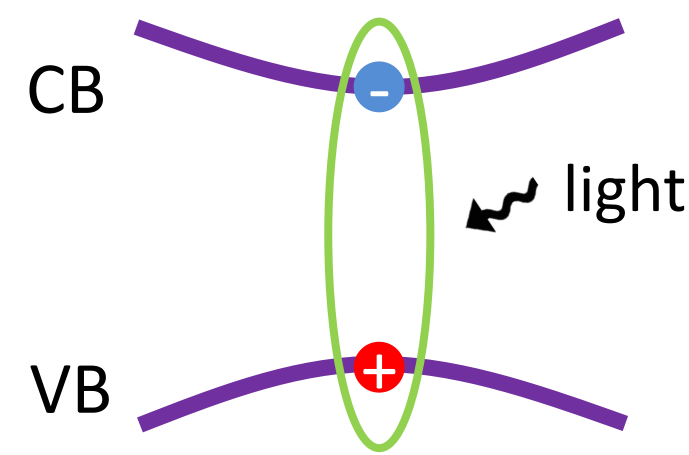
  Strong correlation effects



- **Strong light-matter interaction**

  New ways to create and probe

  excitations with light



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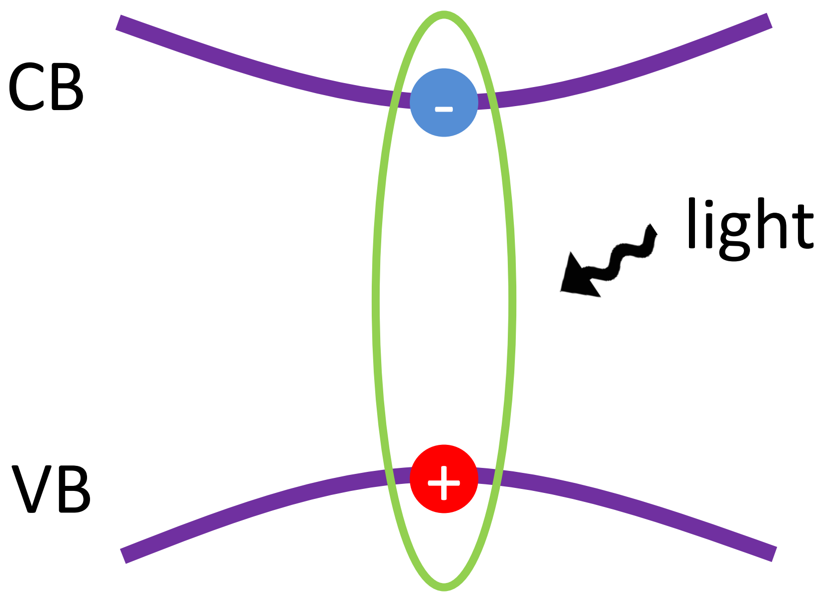
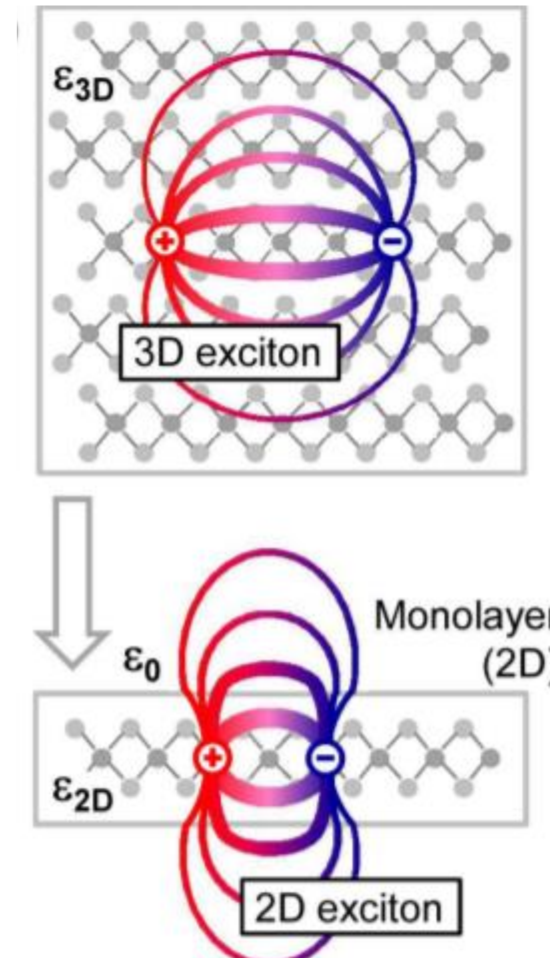
- Moiré excitons: interlayer

Excitons in 2D semiconductors

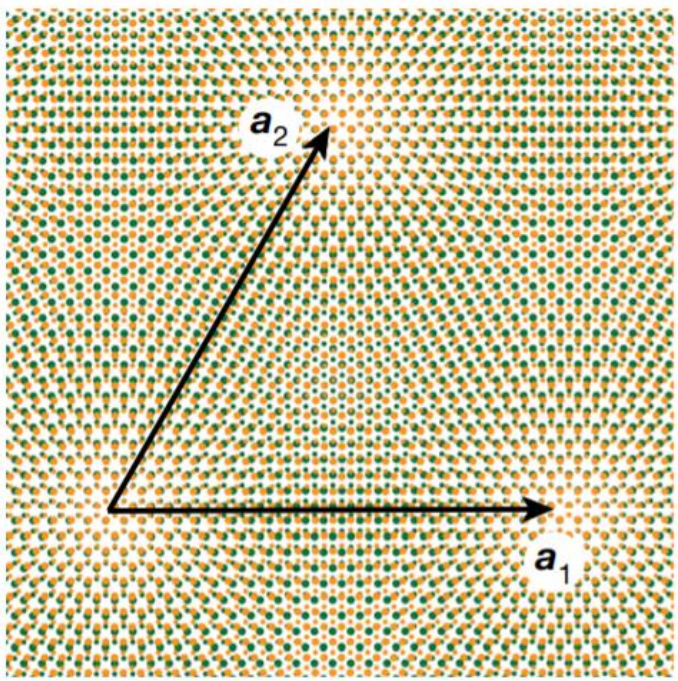




Heinz et al, PRL (2015)

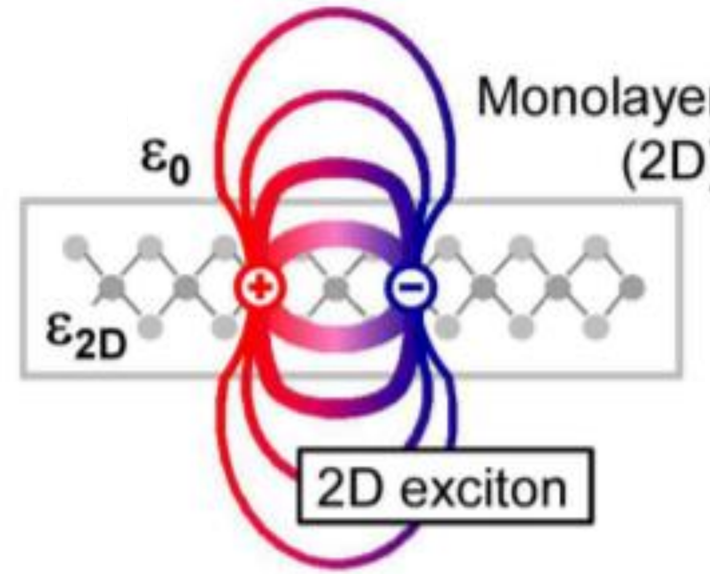
Unique opportunities in two-dimensional semiconductors

Tunable moiré superlattice



+

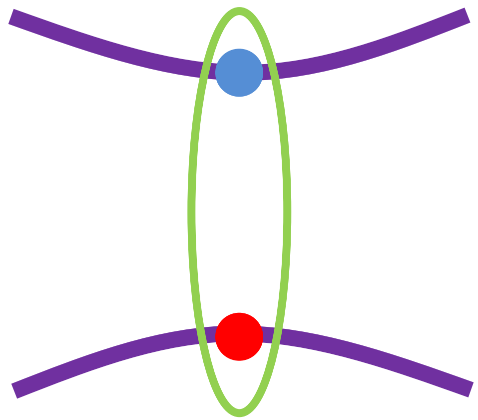
Ultra-stable excitons



Ideal for studying moiré excitons

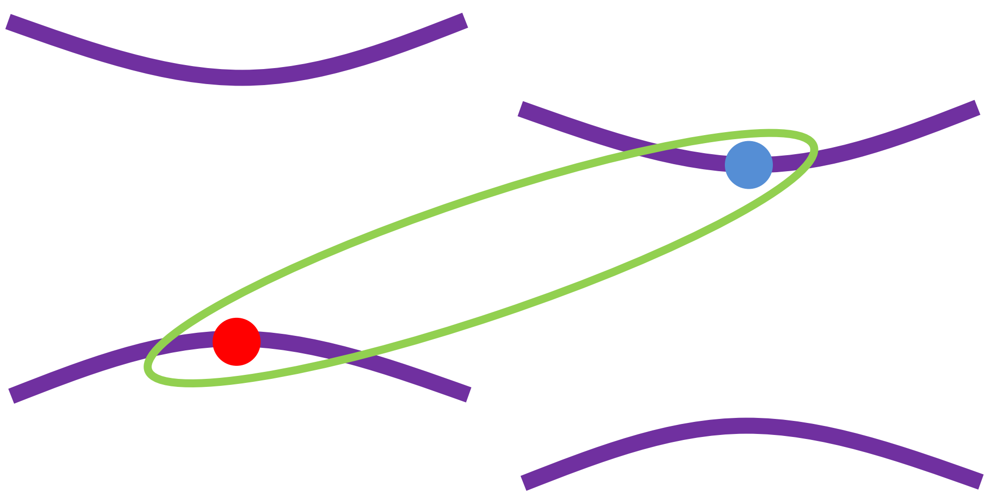
Excitons in two-dimensional systems

Intralayer exciton



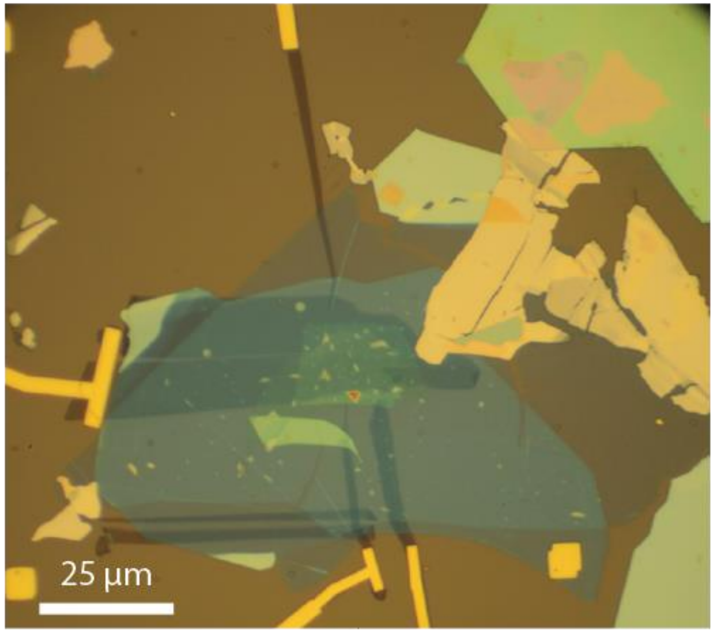
Hundreds of meV binding energy
Large oscillator strength
Strong light matter interaction

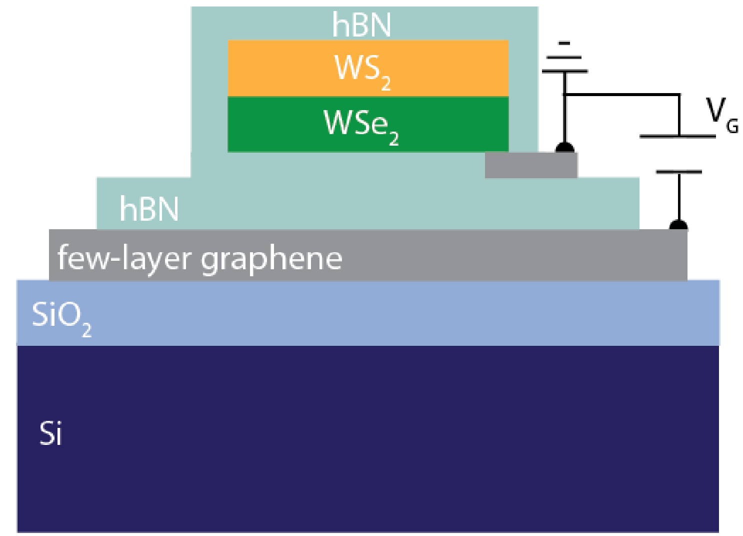
Interlayer exciton (type II)



Hundreds of meV binding energy
Long lifetime
Wide tunability
Unique selection rules

WSe_2/WS_2 heterostructure device



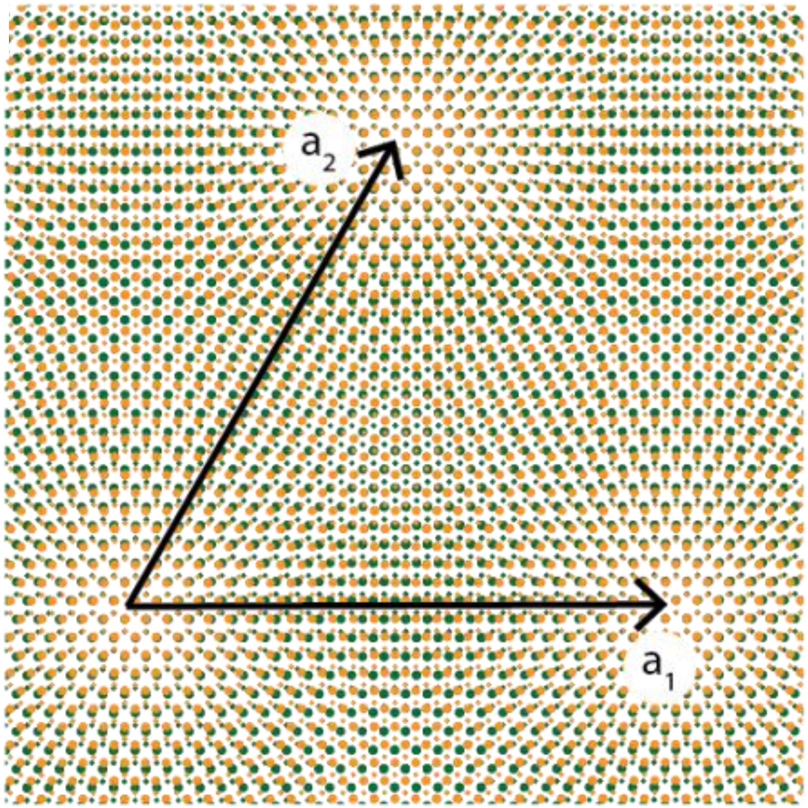


Optical microscope image.

Side view illustration. All contacts are grounded.

Jin, Regan, Wang et al., *Nature* (2019)

Moiré superlattice in aligned WSe_2/WS_2 heterostructure



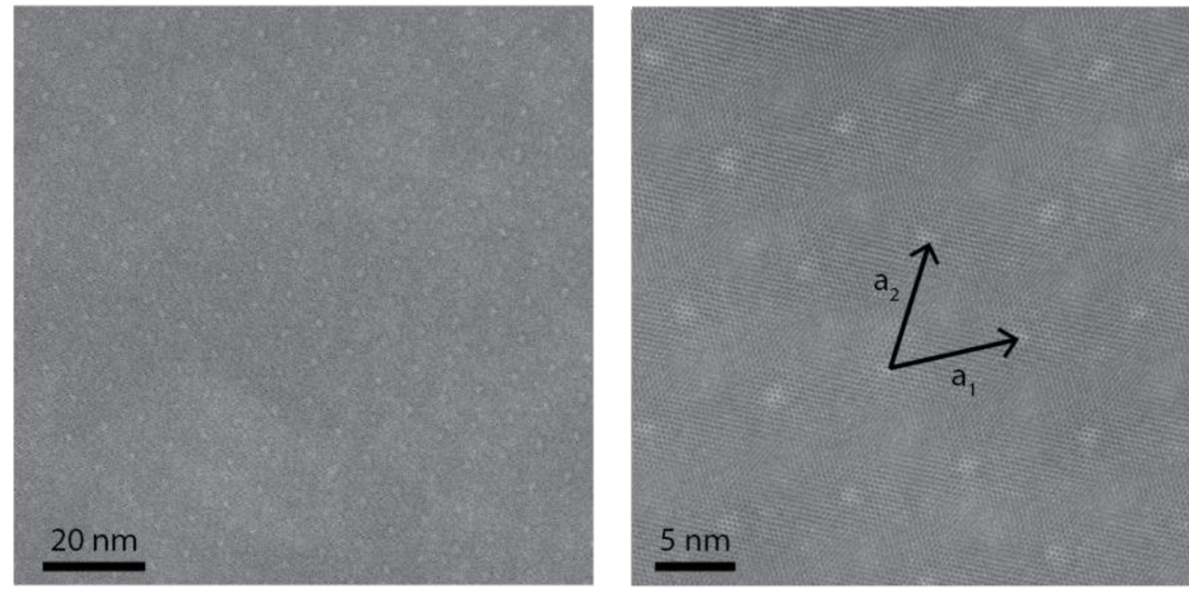
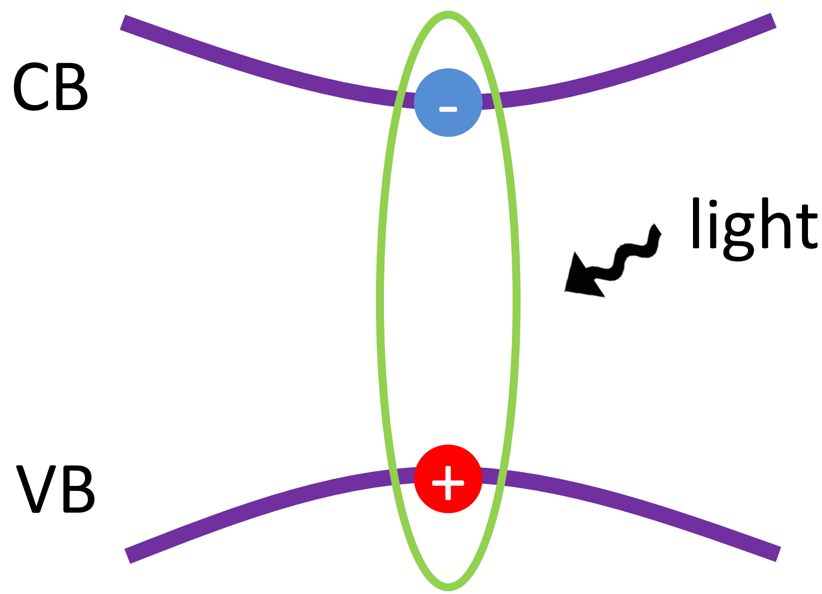


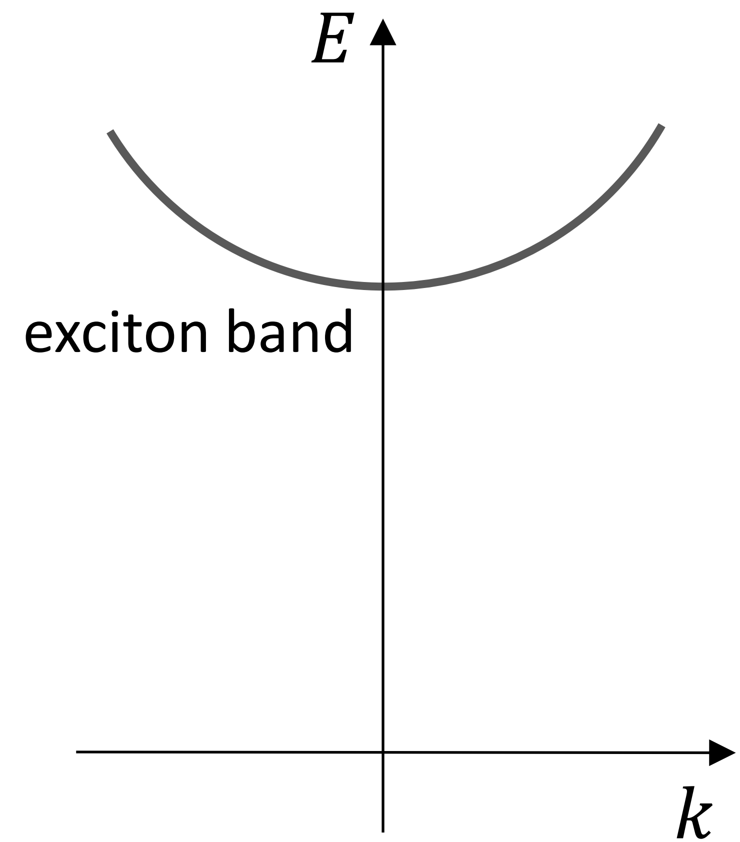
Illustration of the moiré superlattice with ~ 4% lattice mismatch.

Atomic-resolution STEM images.

Probing moiré excitons from optical spectroscopy

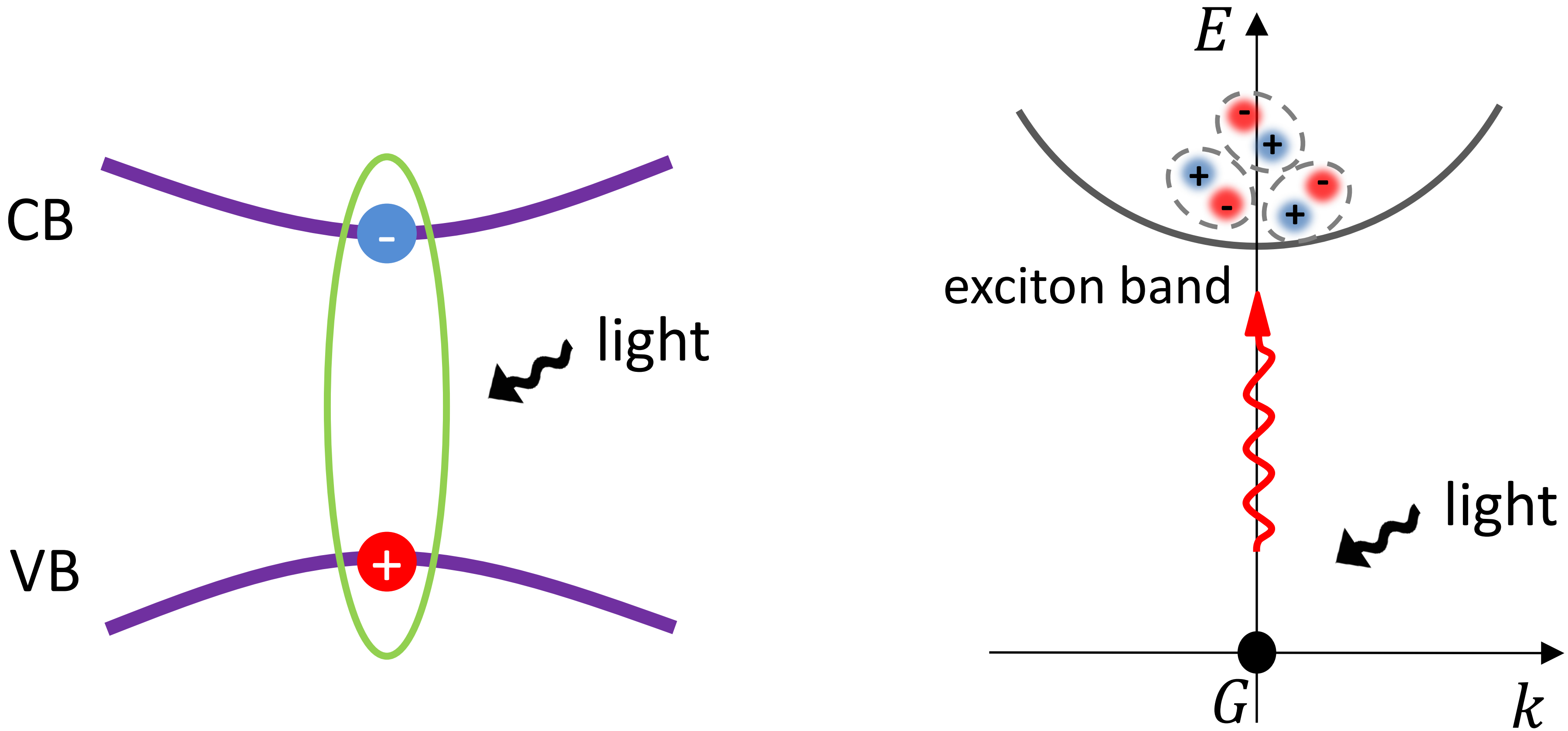


single particle picture



exciton picture

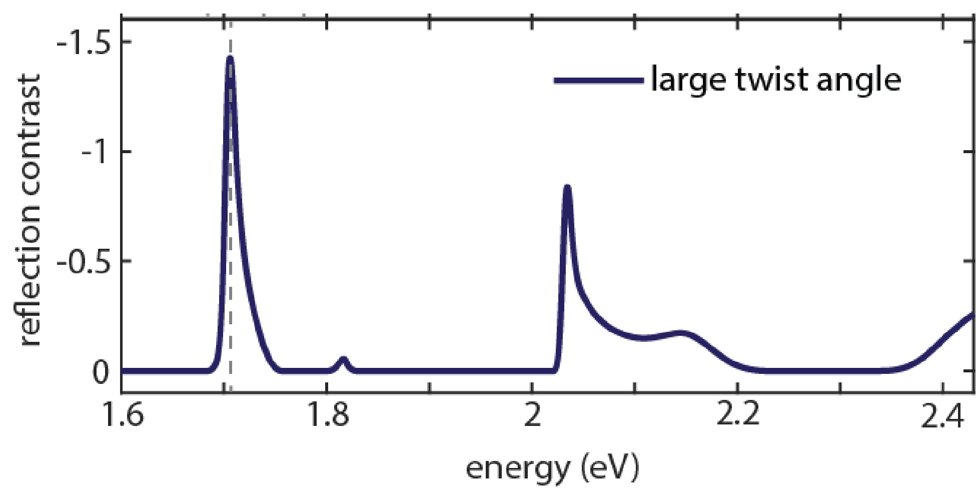
Probing moiré excitons from optical spectroscopy



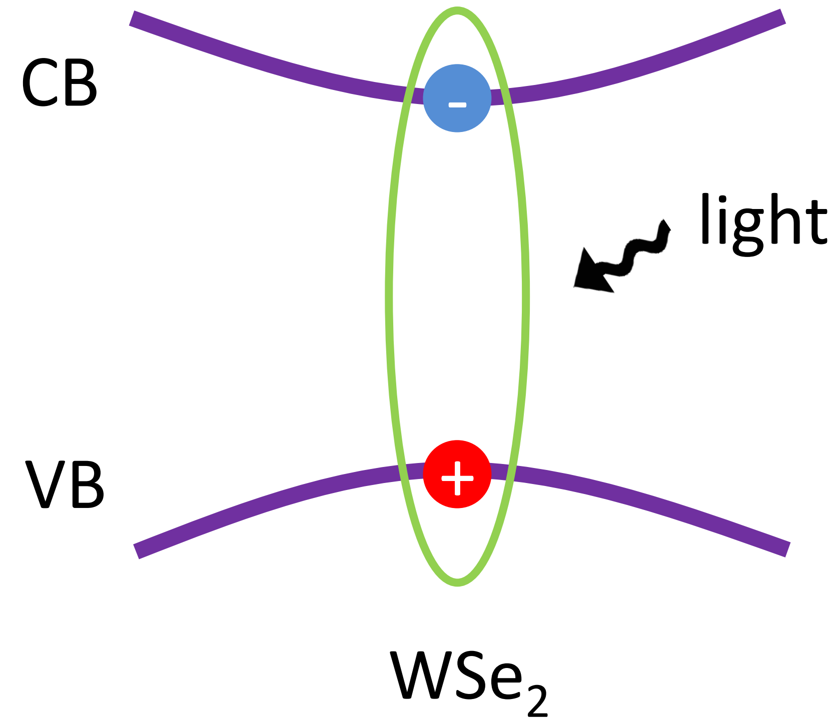
single particle picture

exciton picture

Probing moiré excitons from absorption

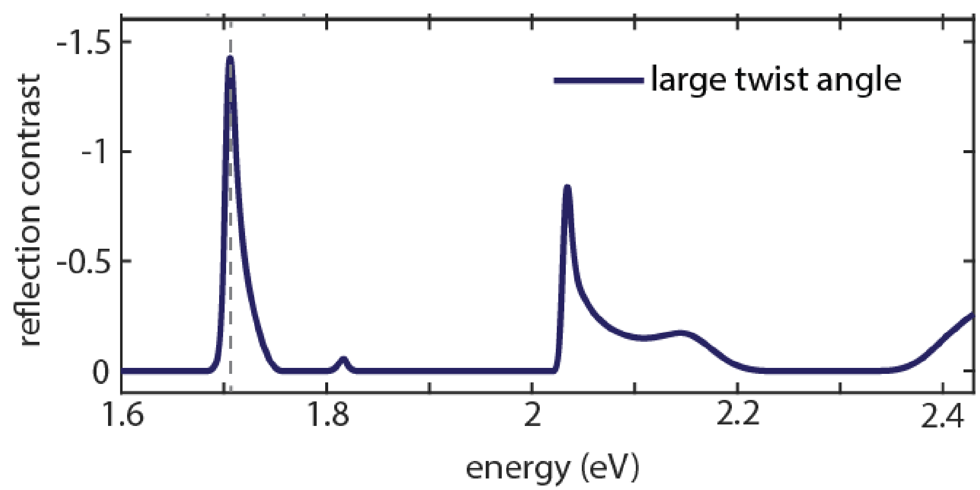


Absorption spectra

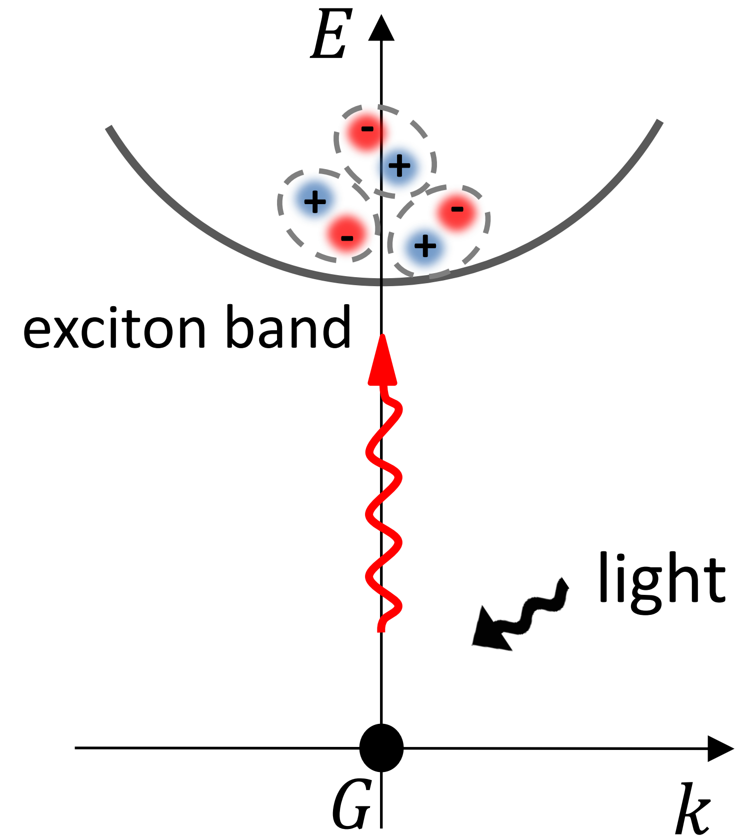


Transition at WSe_2 bandgap

Probing moiré excitons from absorption

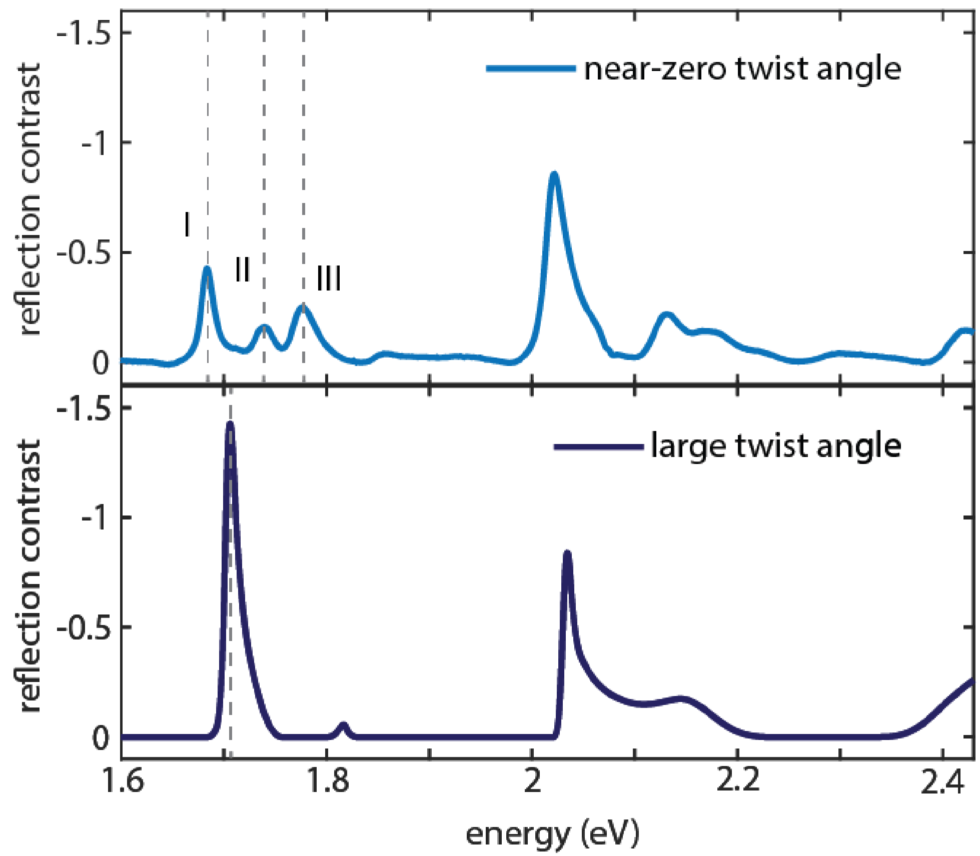


Absorption spectra



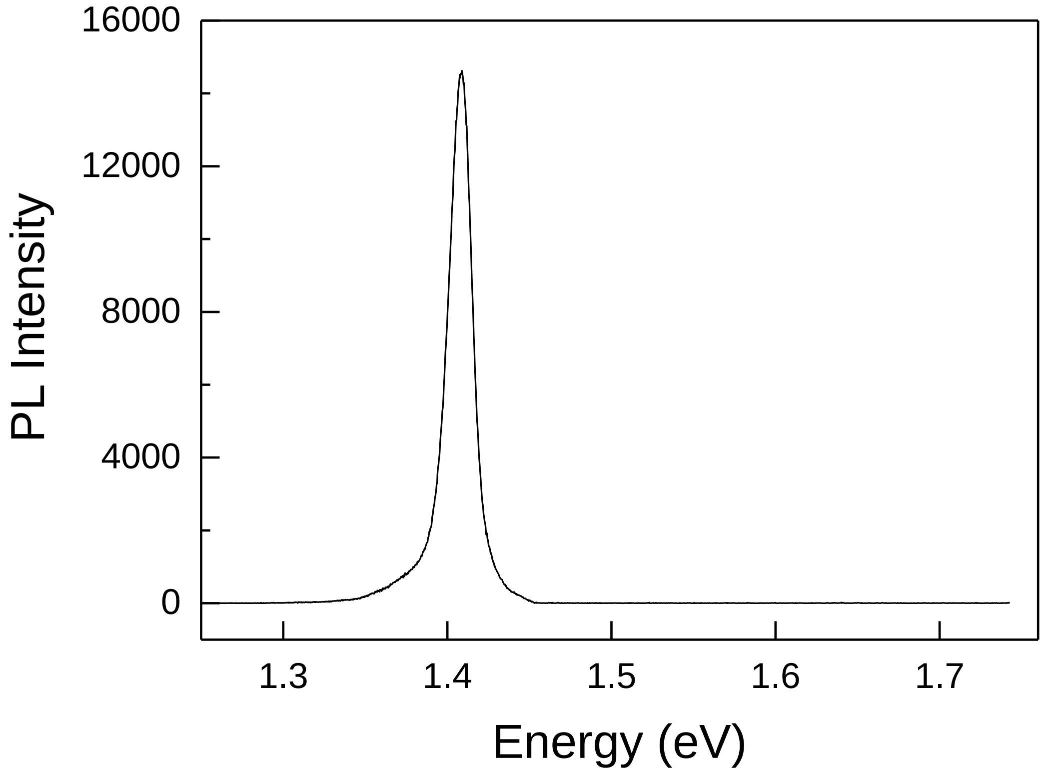
A exciton transition

Probing moiré excitons from absorption

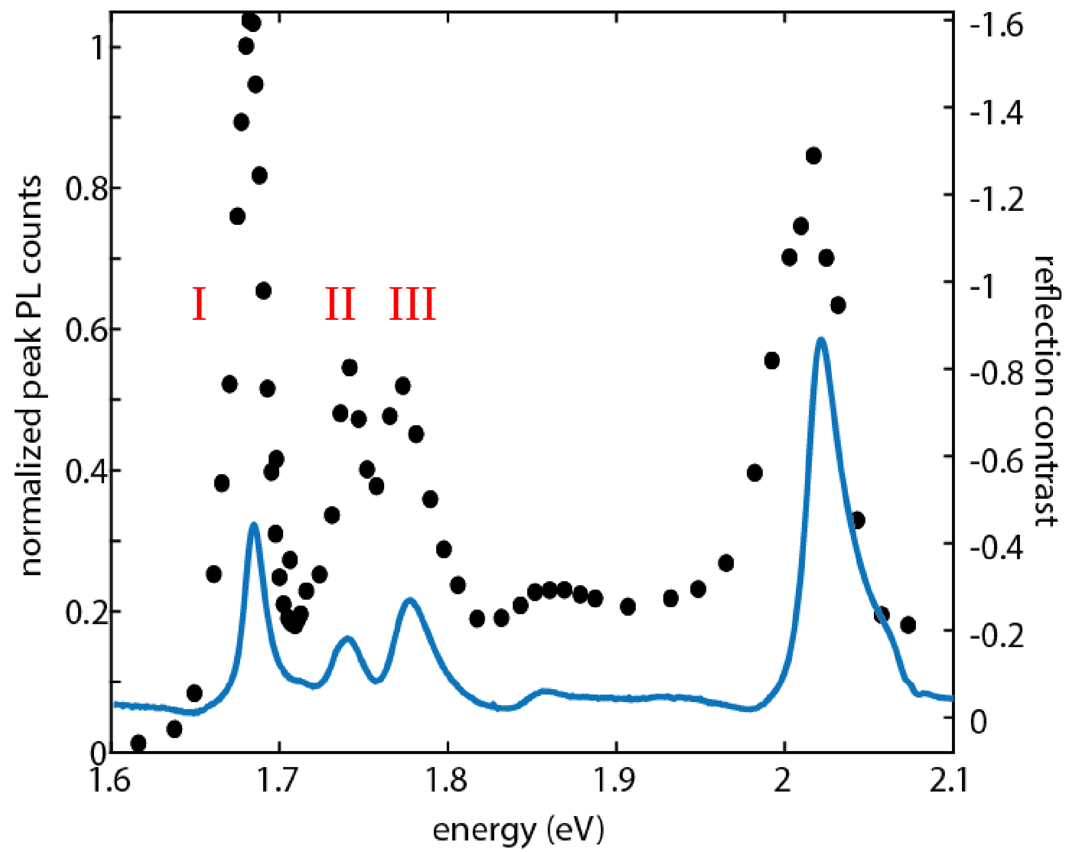


Absorption spectra

Photoluminescence excitation spectroscopy



PL of the HT shows a single peak



All the three absorption peaks strongly enhance the PL.

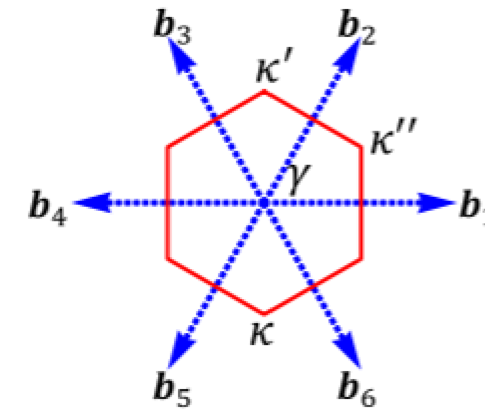
Origin of the moiré exciton peaks

"free" exciton

moiré potential

$$H = H_0 + \sum_{j=1}^{6} V_j \exp(i\boldsymbol{b_j} \cdot \boldsymbol{r}),$$

moiré momentum



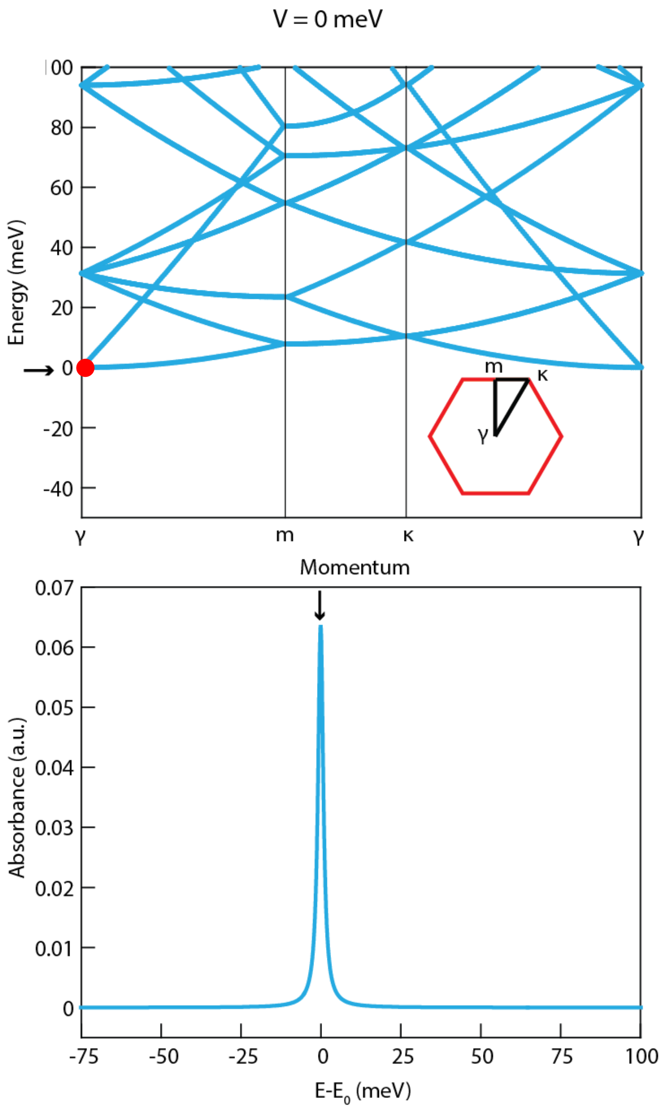
$$V_1 = V_3 = V_5 = V\exp(i\psi) \qquad V_2 = V_4 = V_6 = V\exp(-i\psi)$$

Modeling the moiré superlattice as an effective potential picture.

Due to symmetry, only two independent parameters.

Wu et al., PRL (2017)

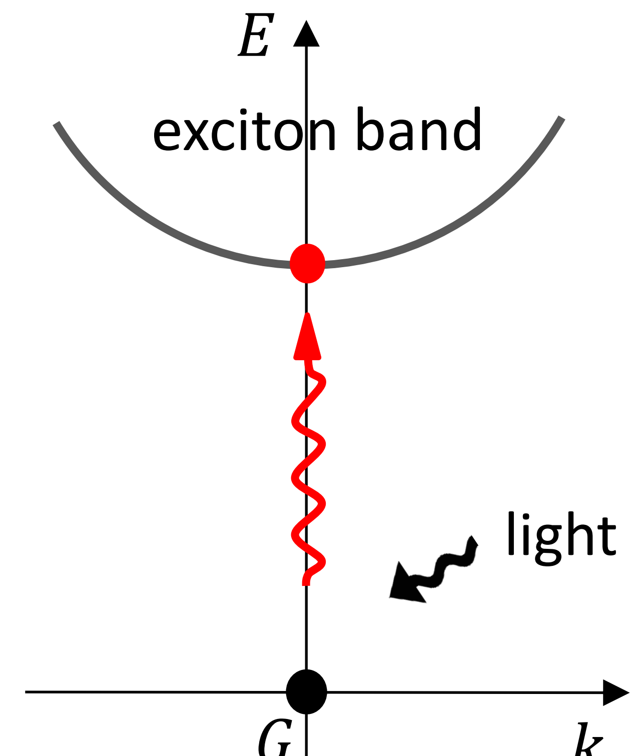
Origin of the moiré exciton peaks



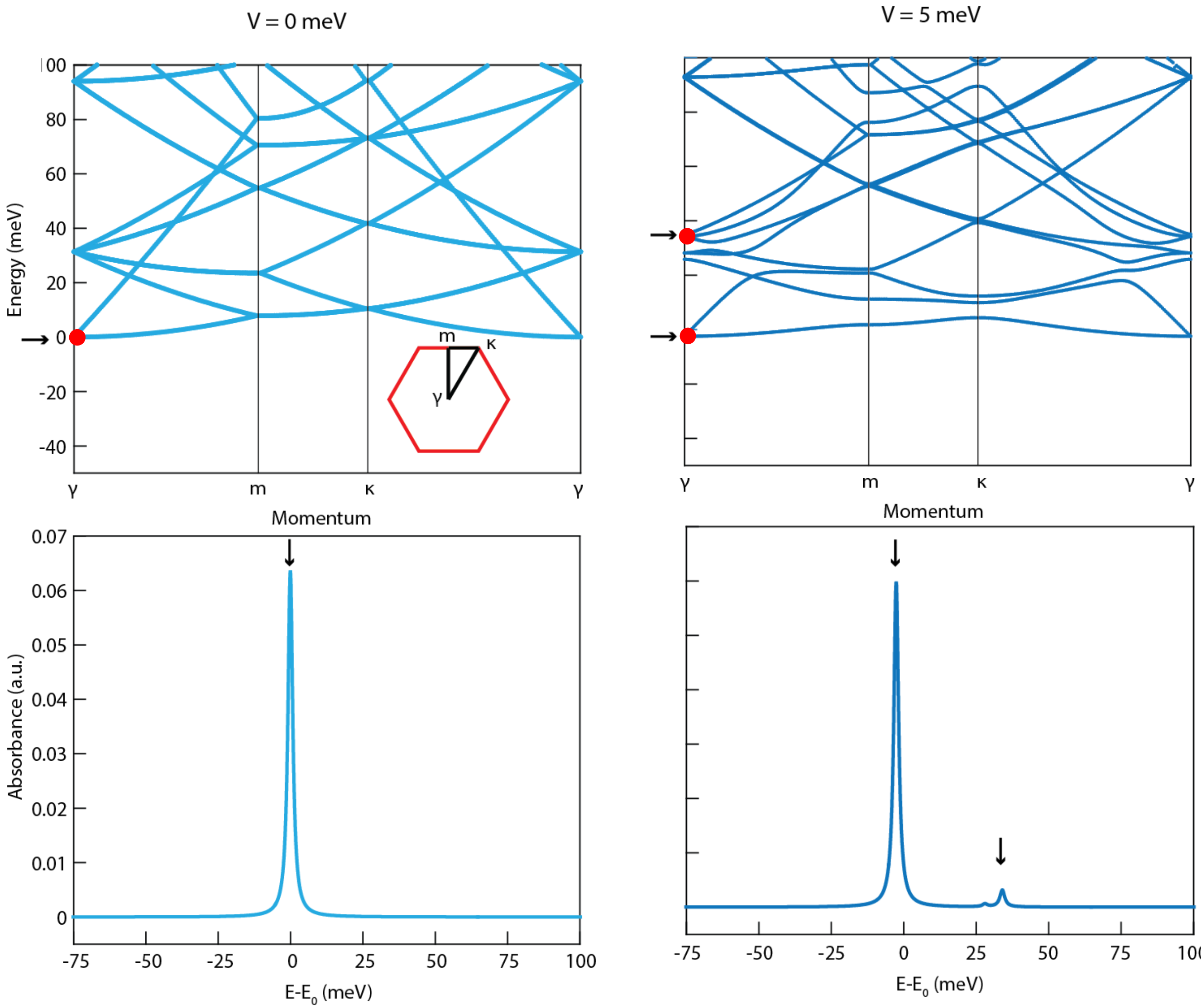
"No coupling"

Only lowest energy state is bright.

Single strong peak in absorption.



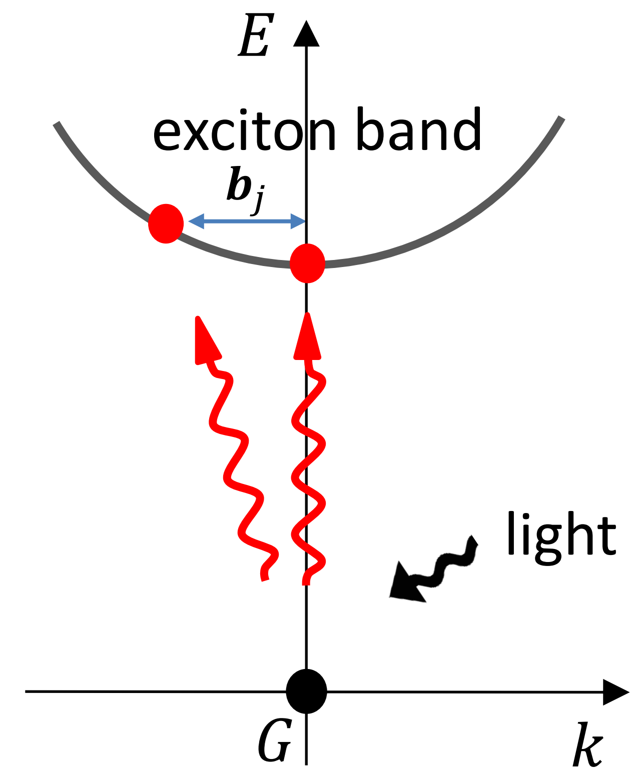
Origin of the moiré exciton peaks



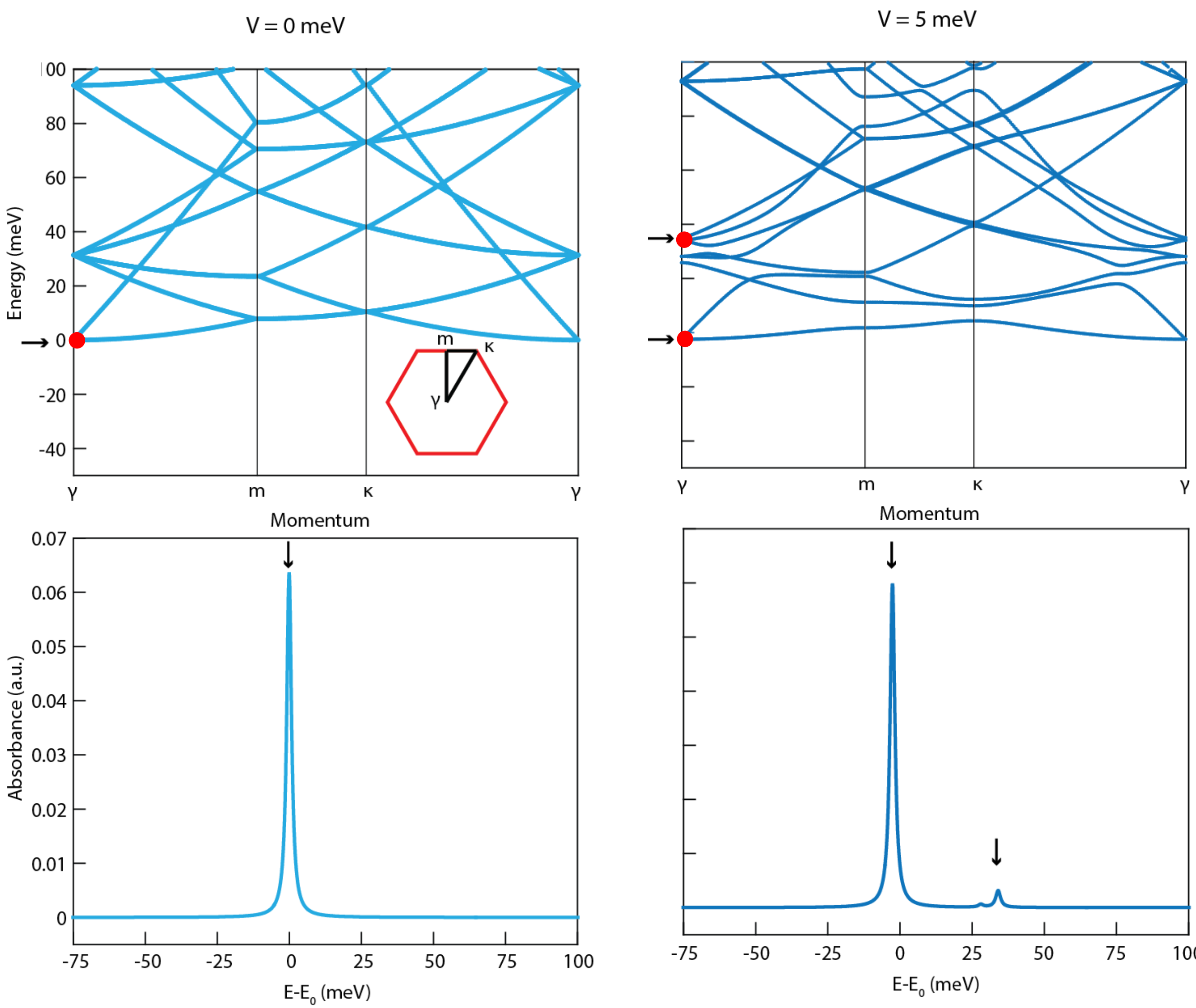
"Weak coupling"

Excitons states start mixing slightly.

$\boldsymbol{k} = \boldsymbol{b}_j$ states weakly brightened



Origin of the moiré exciton peaks



"Weak coupling"

Excitons states start mixing slightly.

$\boldsymbol{k} = \boldsymbol{b}_j$ states weakly brightened

Exciton dispersion remains largely the same.

Weak side peak at $E_m = \hbar^2 b_j^2/(8M)$ ~ 30 meV

Cannot explain our observation

Wu et al., PRL (2017)

Origin of the moiré exciton peaks

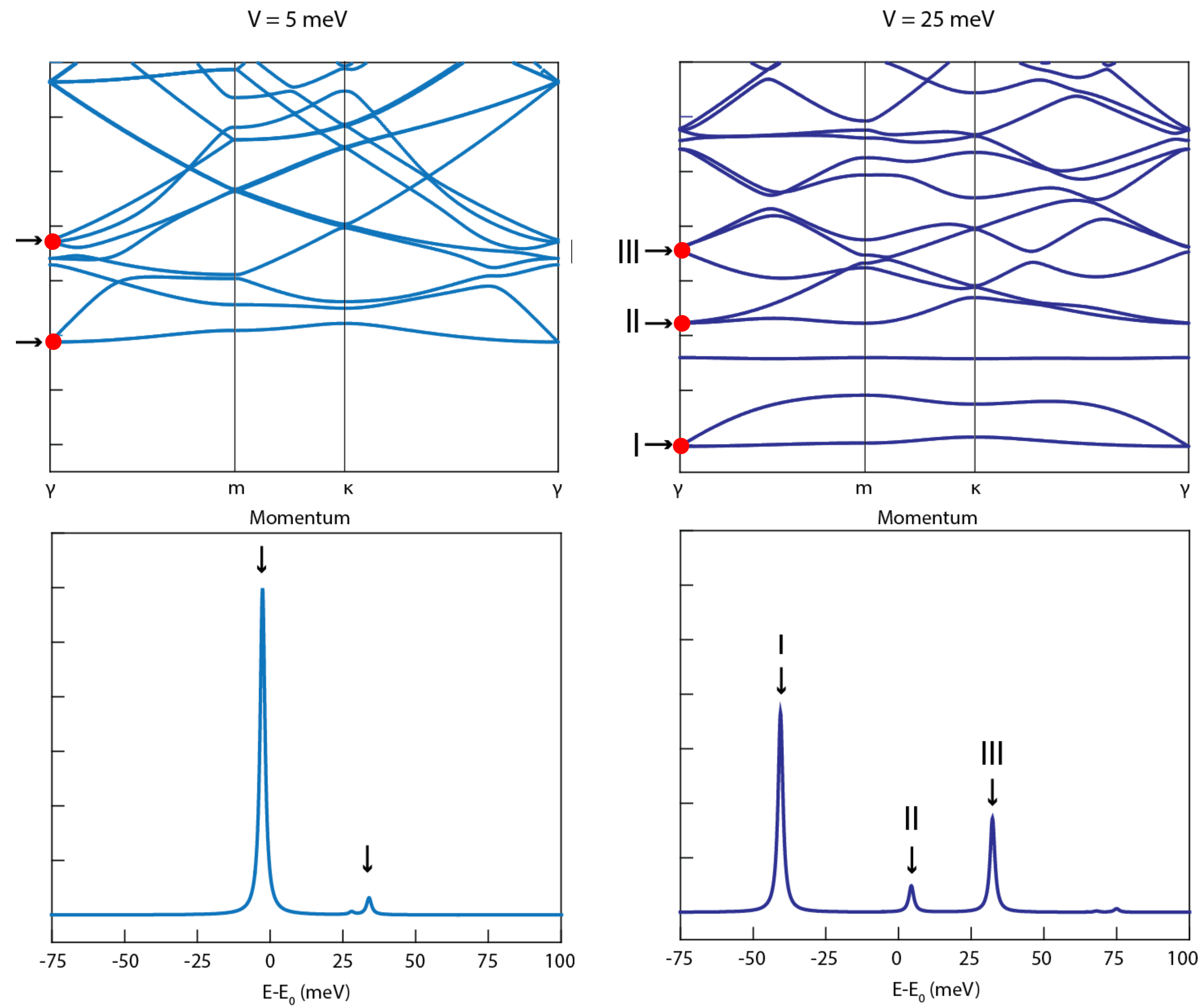
"Strong coupling"

Excitons states mix strongly

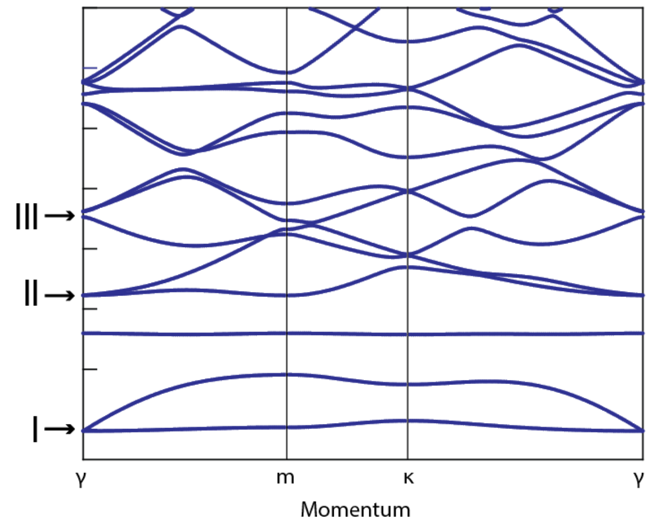
Different moiré exciton states can have comparable oscillator strength

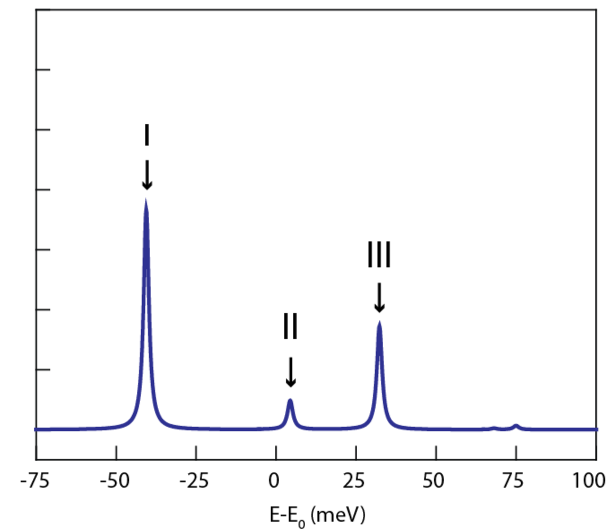
Exciton dispersion is strongly modified.

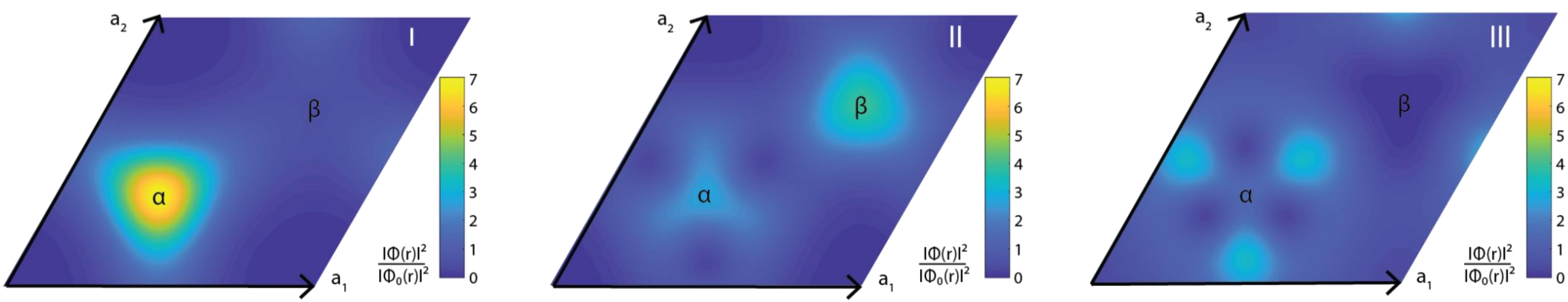
The energy of the states can vary significantly with potential parameters.



Moiré excitons in the real space





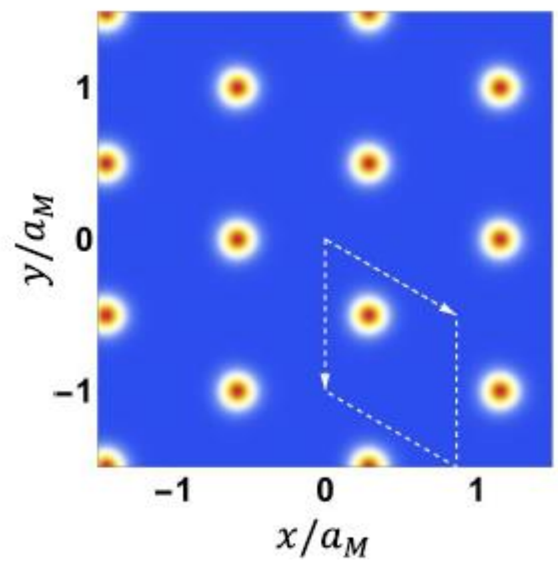


Emerging exciton lattice

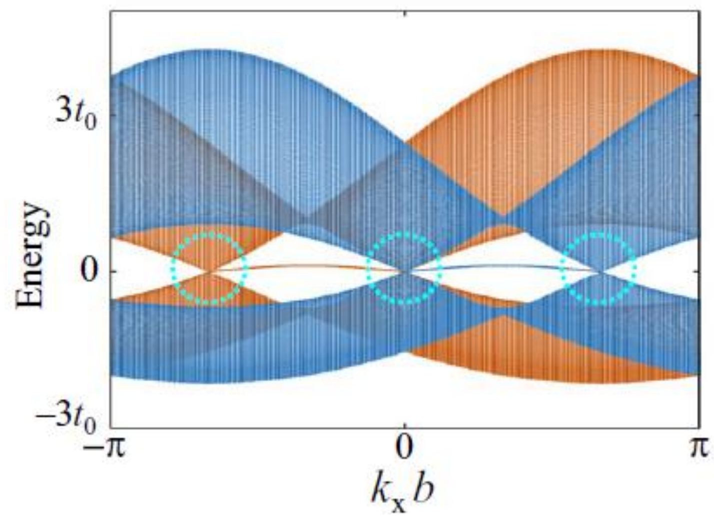
Jin, Regan, Wang, *Nature* (2019)

New opportunities from moiré excitons

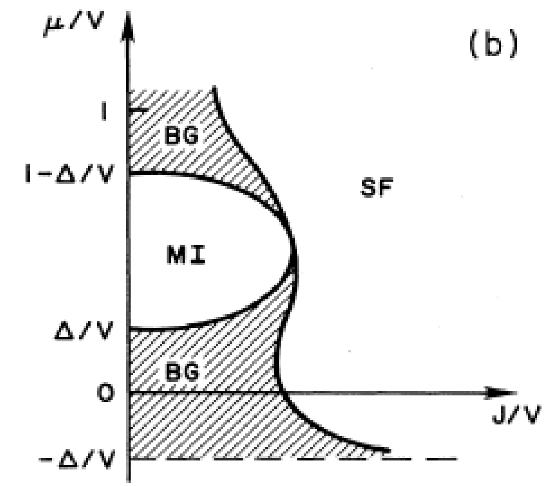
Exciton “solids”



Topological excitons

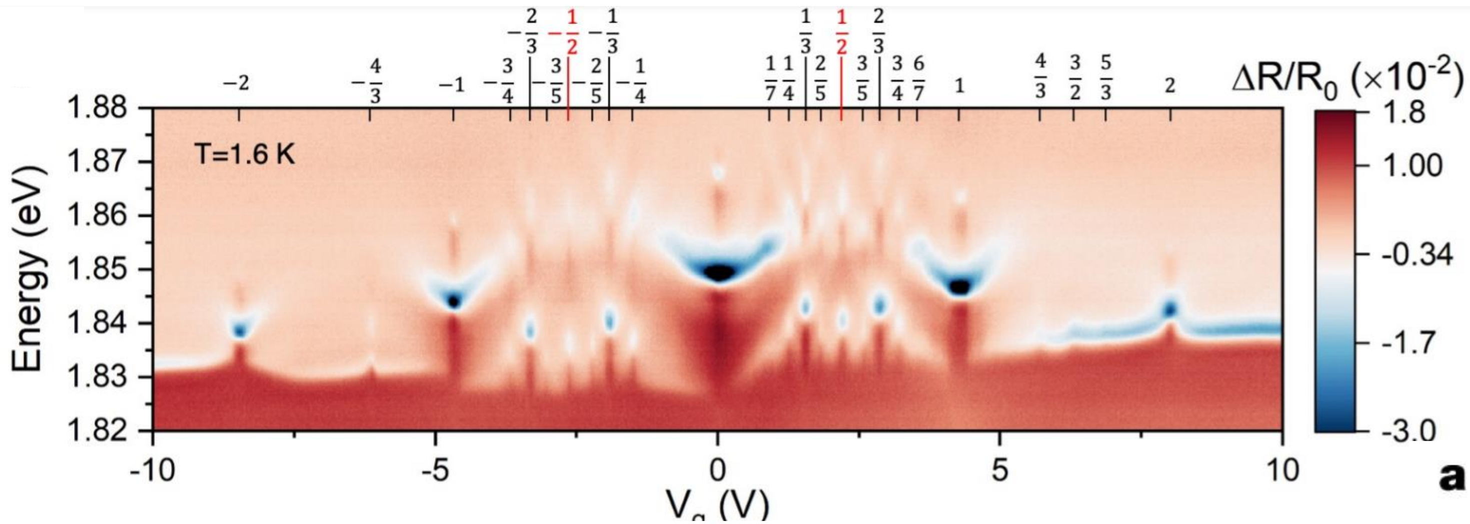


Bosonic physics

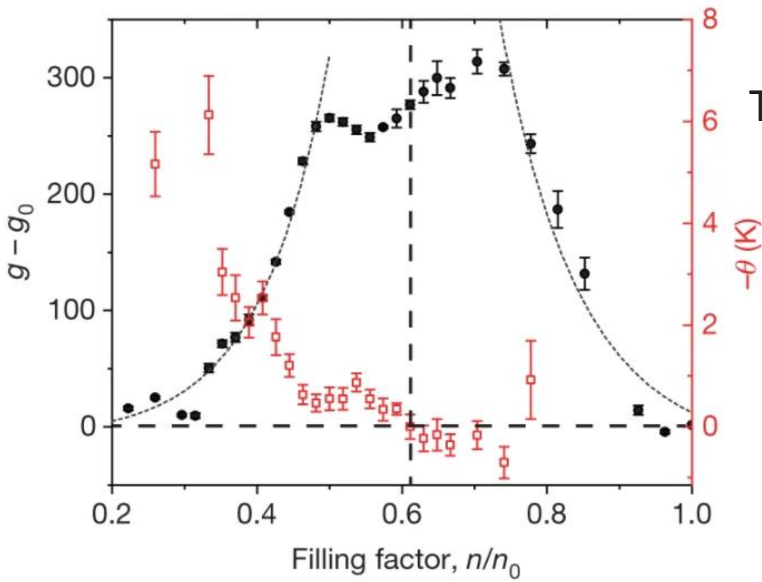


Fisher, et al., *PRB* (1989)
Wu et al., *PRL*, (2017)
Yu et al., *Sci. Adv.* (2017)

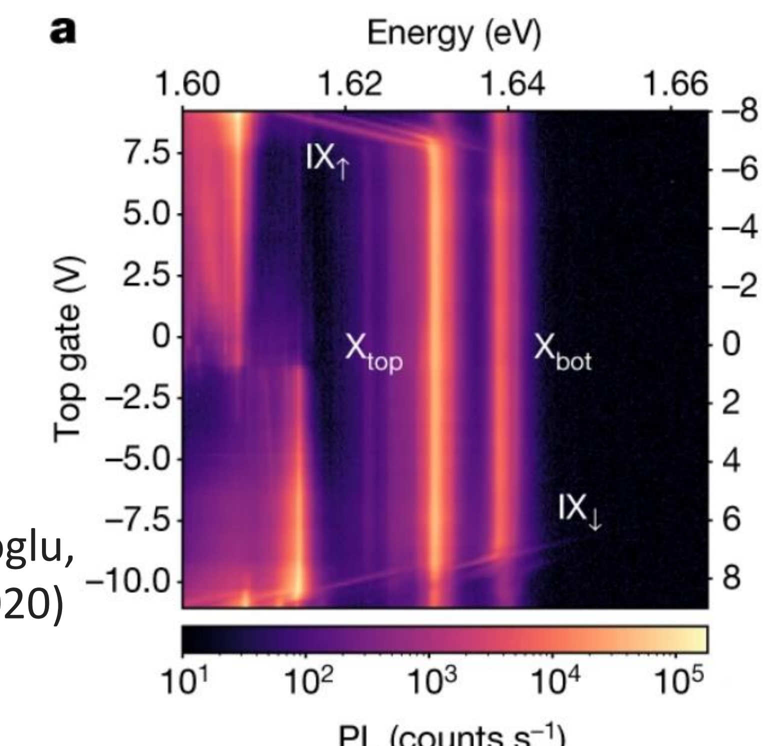
“Sensing” electrons with intralayer excitons



Xu, Mak, Shan, et al., *Nature* (2020)



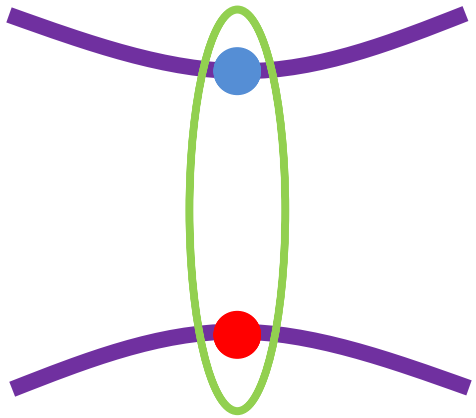
Tang, Shan, Mak, et al., *Nature* (2020)



Shimazaki, Imamoglu, et al., *Nature* (2020)

Excitons in two-dimensional materials

Intralayer exciton

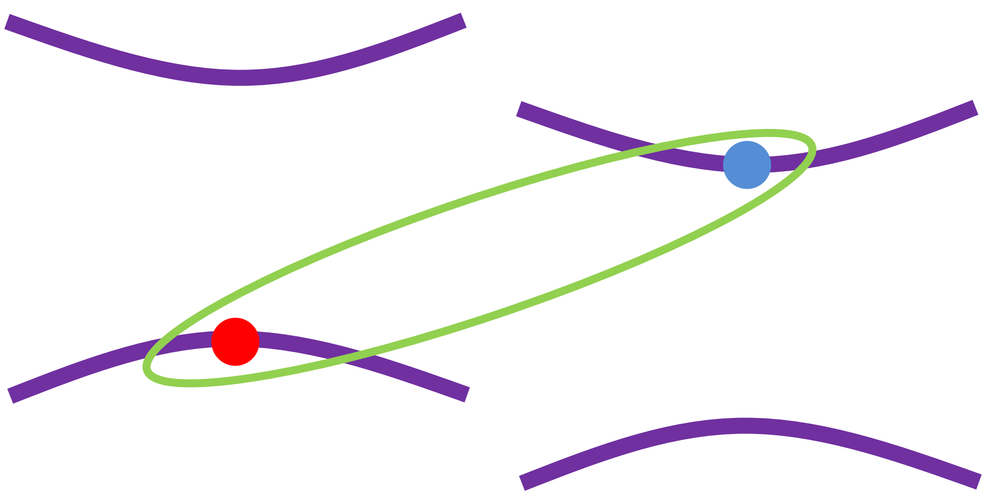


Hundreds of meV binding energy

Large oscillator strength

Strong light matter interaction

Interlayer exciton



Hundreds of meV binding energy

Long lifetime

Wide tunability

Unique selection rules

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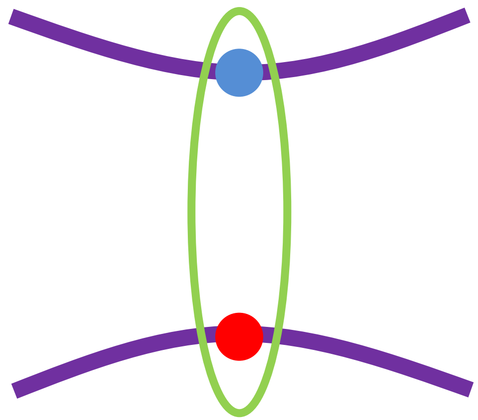
- **Moiré excitons: interlayer**

Excitons in two-dimensional materials

Intralayer exciton



Hundreds of meV binding energy

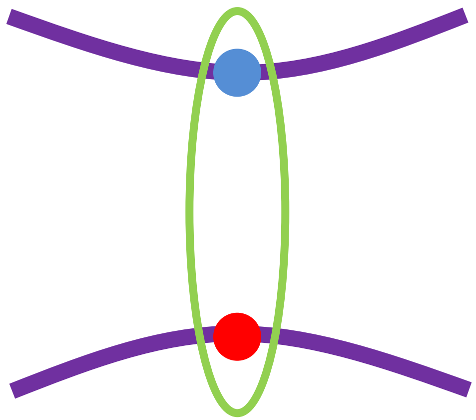
Large oscillator strength

Strong light matter interaction

Short lifetime (in type II HT)

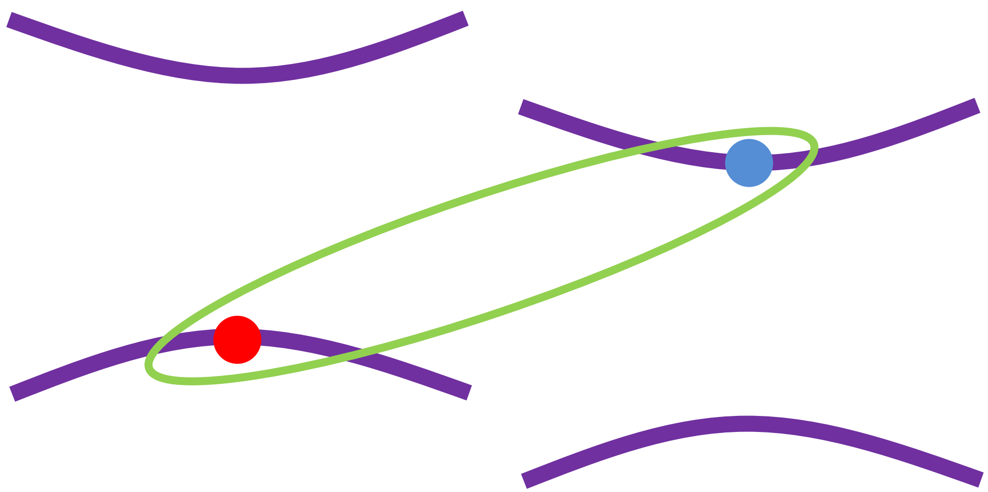
Excitons in two-dimensional materials

Intralayer exciton



Hundreds of meV binding energy
Large oscillator strength
Strong light matter interaction
Short lifetime (in type II HT)

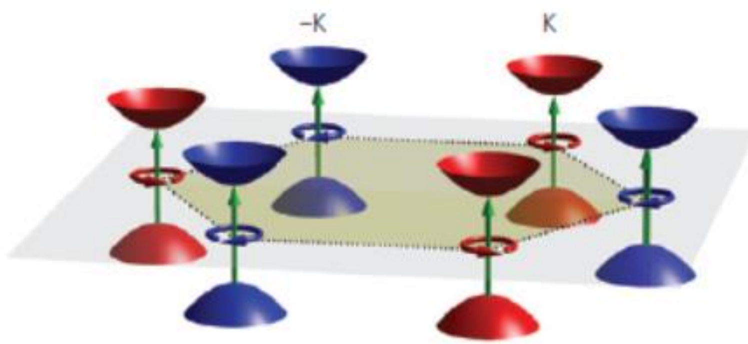
Interlayer exciton



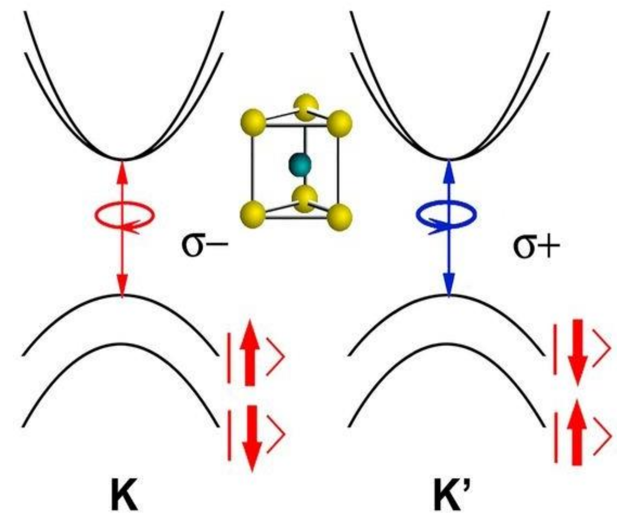
Hundreds of meV binding energy
Long lifetime
Wide tunability
Unique selection rules

An exciton lattice with "spin-orbit coupling"

Bright exciton does not have spin
But it has valley pseudospin in TMDC



Degenerate K/K' valley



Convenient addressing with polarized light.

Valley selection rule of interlayer exciton

Exciton wavefunction

Center of mass Relative Spin

$$\psi(\boldsymbol{r}_e, \boldsymbol{r}_h) = \Phi(\boldsymbol{R})\phi(\boldsymbol{r})\,S$$

Quasi angular momentum (QAM) conservation

$$C_3\psi(\boldsymbol{r}_e, \boldsymbol{r}_h) = e^{\frac{i2n\pi}{3}}\psi(\boldsymbol{r}_e, \boldsymbol{r}_h), n = ?$$

Spin singlet, 1s state: Determined by center-of-mass motion $\Phi(\boldsymbol{R})$, can be approximately understood from band to band transition picture.

Quasi-angular momentum of CBM electron

$$\psi_e^{\boldsymbol{k}}(\boldsymbol{r}_e) = \sum_{\boldsymbol{R_0}} \varphi(\boldsymbol{r}_e - \boldsymbol{R_0})\, e^{i\boldsymbol{k}\cdot\boldsymbol{R_0}}$$

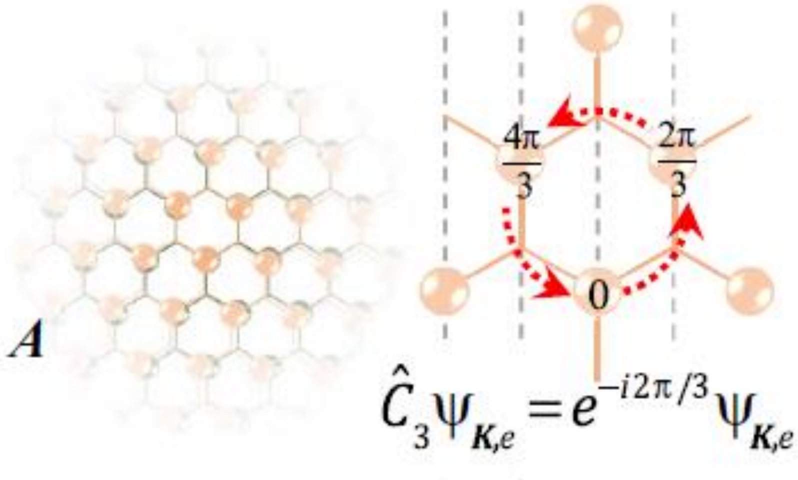
$$C_3\psi_e^{\boldsymbol{k}}(\boldsymbol{r}_e) = e^{\frac{i2n\pi}{3}}\psi_e^{\boldsymbol{k}}(\boldsymbol{r}_e), n = ?$$

Valley

$\varphi(\boldsymbol{r}_e - \boldsymbol{R_0})$:
Local (atomic) orbital,
angular momentum known

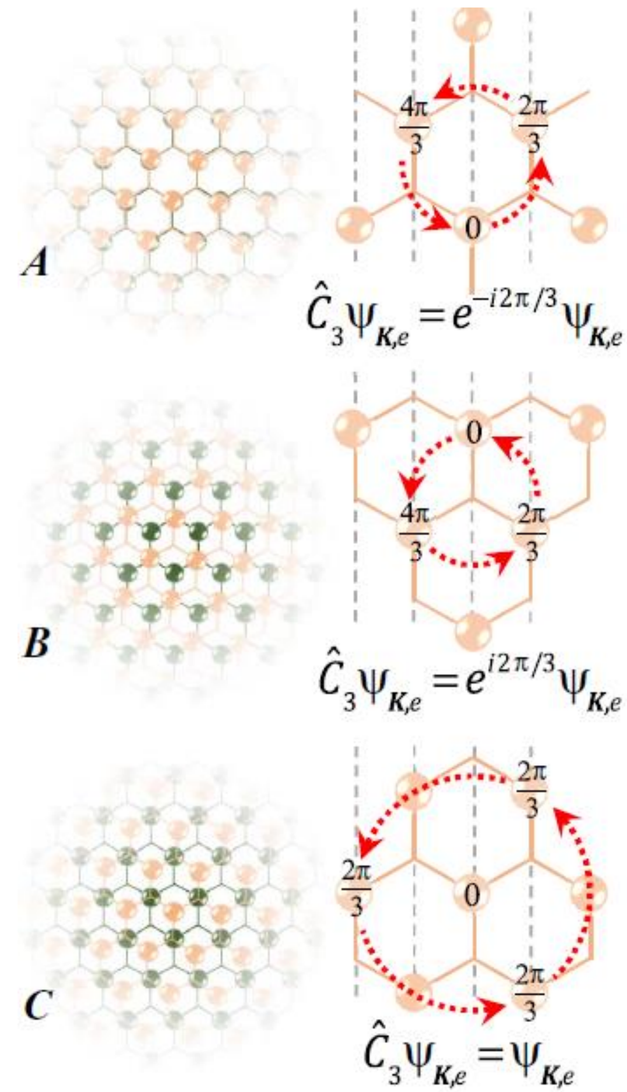
$e^{i\boldsymbol{k}\cdot\boldsymbol{R_0}}$:
Plane wave from Bloch theorem.
What is its contribution?

Valley selection rule of interlayer exciton



Additional angular momentum from the phase difference between unit cells

Intralayer exciton:
Electron and hole always cancel,
no overall contribution:
Only spin-valley contribution



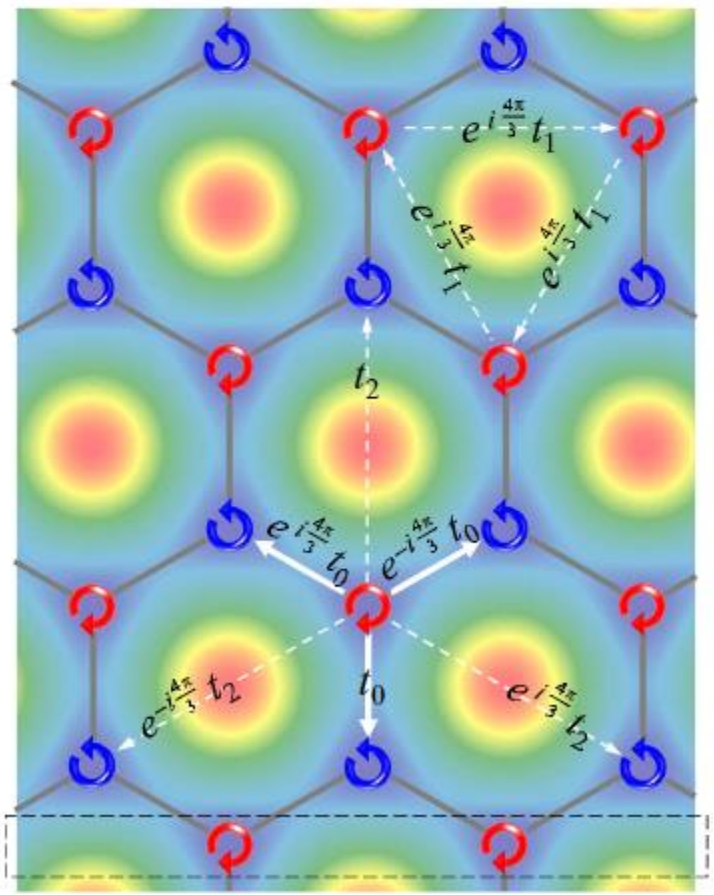
Interlayer exciton:

Depending on the local lattice configuration

Additional “moire” contribution

An exciton lattice with "spin-orbit coupling"

Interlayer excitons (of the same valley) at different moiré sites have different quasi-angular momentum



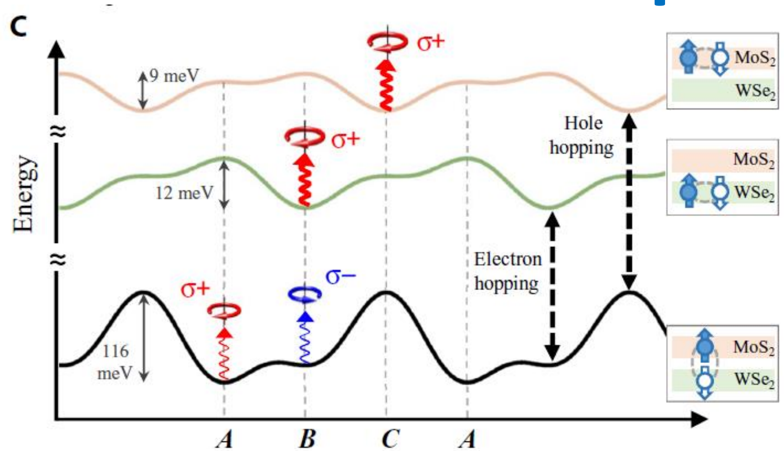
In this case…

NN coupling between ILE must be complex

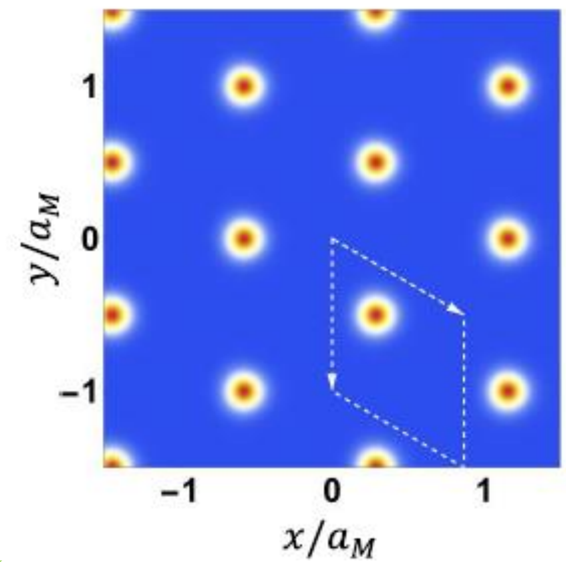
A large intrinsic "spin-orbit" coupling

Topological exciton states predicted to emerge

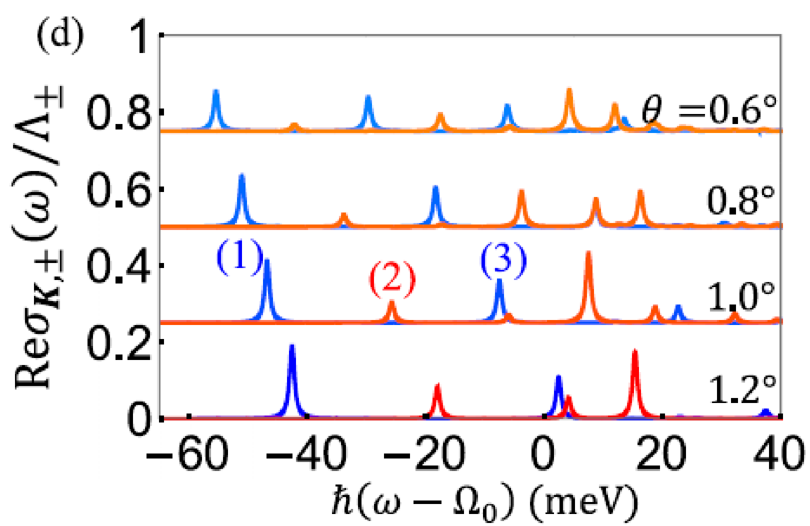
One last requirement: “localized” excitons



Different valley selection rule depending on local configuration



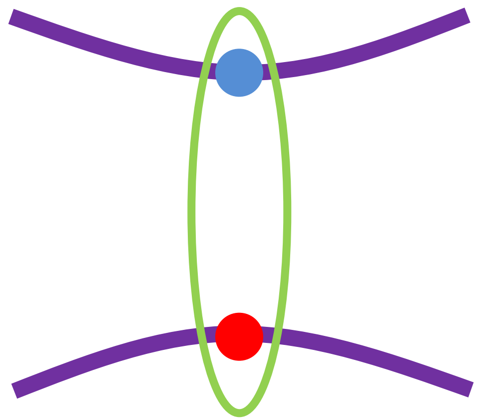
Interlayer exciton states are localized.



Multiple interlayer exciton states with opposite valley selection rule?

Excitons in two-dimensional materials

Intralayer exciton



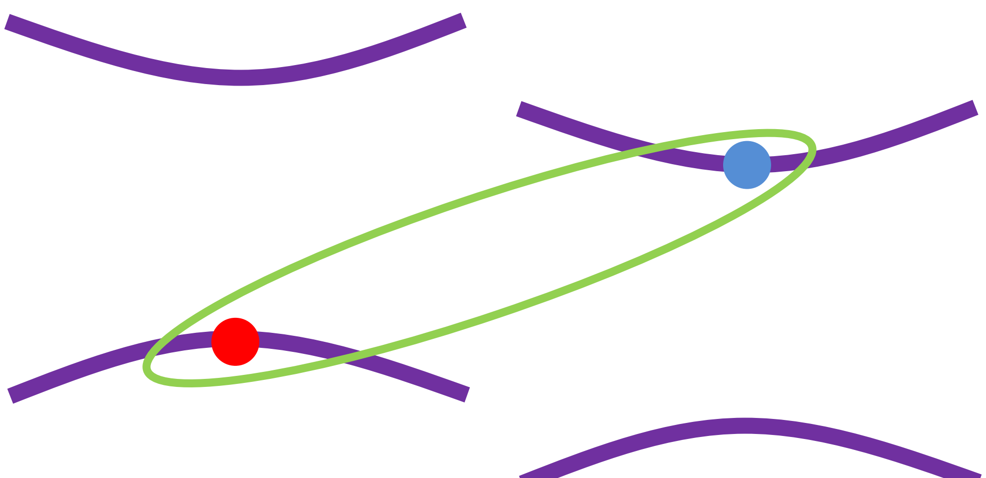
Hundreds of meV binding energy

Large oscillator strength

Strong light matter interaction

Short lifetime (in type II HT)

Interlayer exciton



Hundreds of meV binding energy

Long lifetime

Wide tunability

Unique selection rules

Small oscillator strength

Weak light matter interaction

How do we probe them?

PL (almost all literature):

- Difficult to probe high energy state
- Cannot determine the nature of the emitting state (momentum-indirect, spin-triplet…)
- Cannot extract the “moire” contribution

Absorption:

- Can probe high energy state
- Can differentiate dark/indirect exciton
- Difficult to do: small oscillator strength
- Cannot extract the “moire” contribution

How do we probe them?

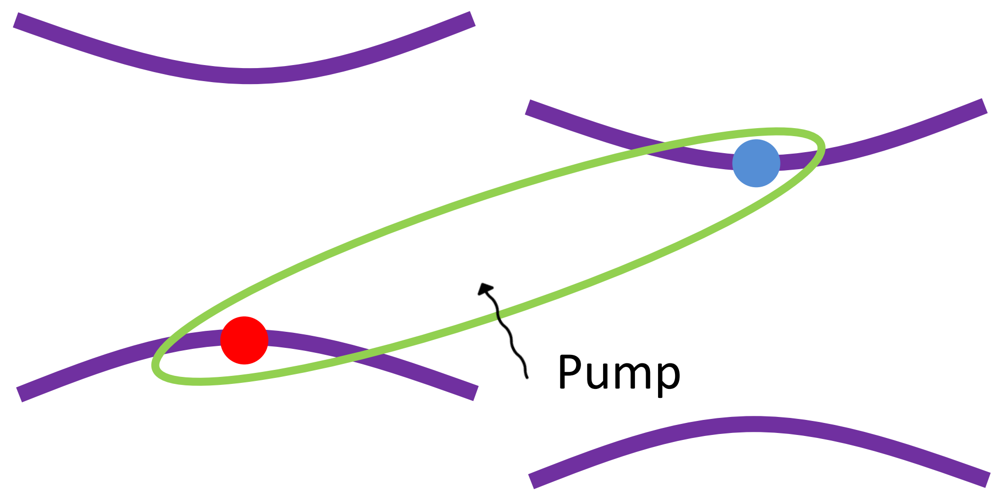
Problem:

Directly probe interlayer exciton always involve multiple contributions

Instead, one can determine the electron and hole **separately.**
e.g. Pumping interlayer exciton, probing intralayer exciton

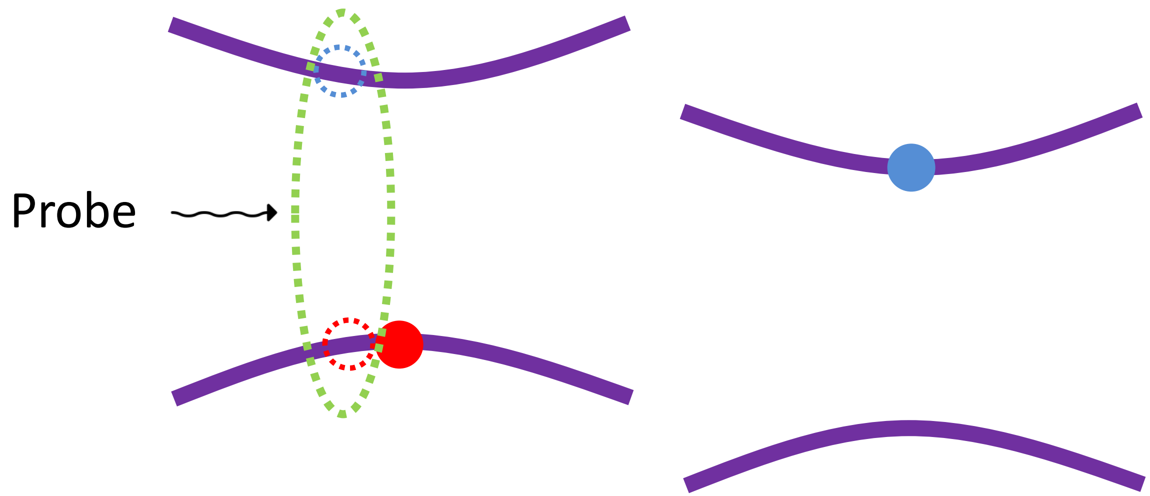
Probing interlayer exciton states

Resonant Pump probe: Interlayer exciton pump



Probing interlayer exciton states

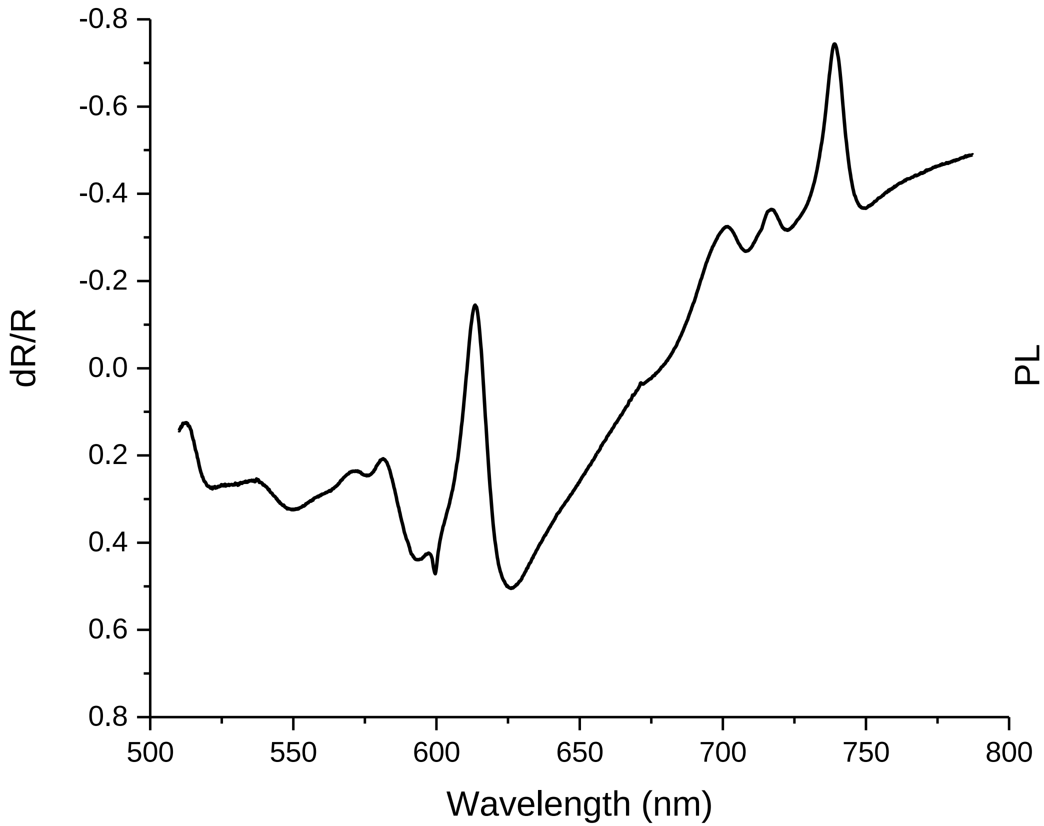
Resonant Pump probe: Intralayer exciton probe



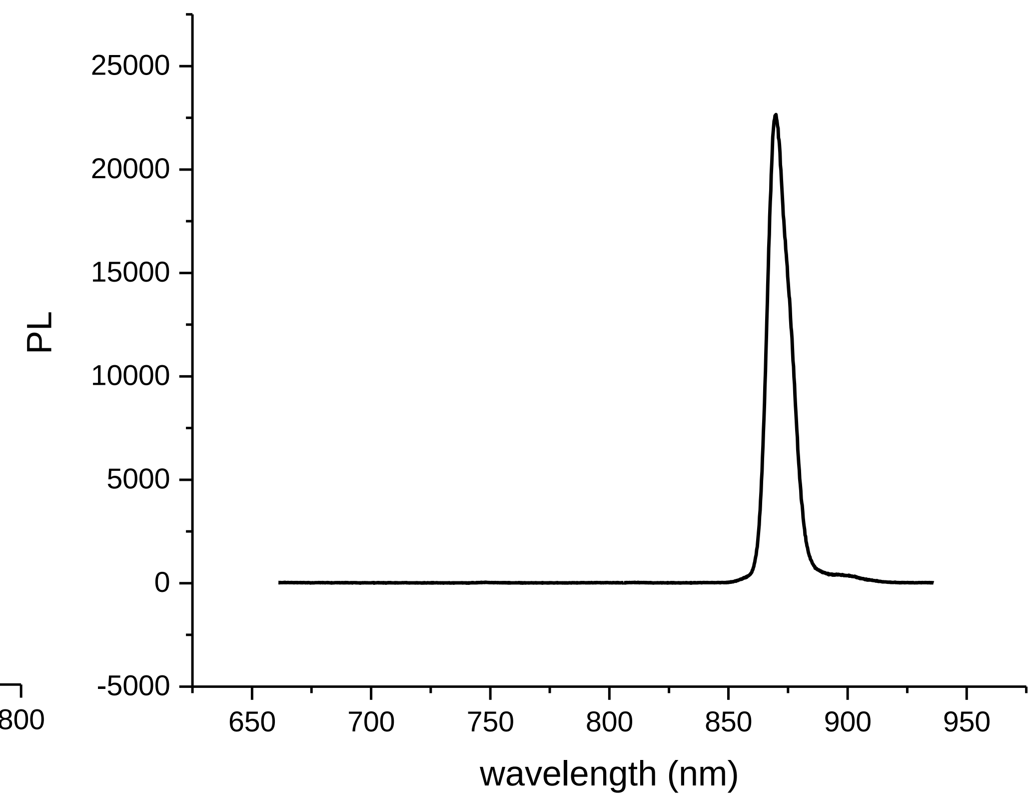
- Can probe high energy state
- Can differentiate dark/indirect exciton
- Can probe weak resonance: background free
- Can extract the "moire" contribution

Key: Intralayer exciton only contains spin-valley contribution

Basic characterization of a ~0 twist angle WSe2/WS2 device

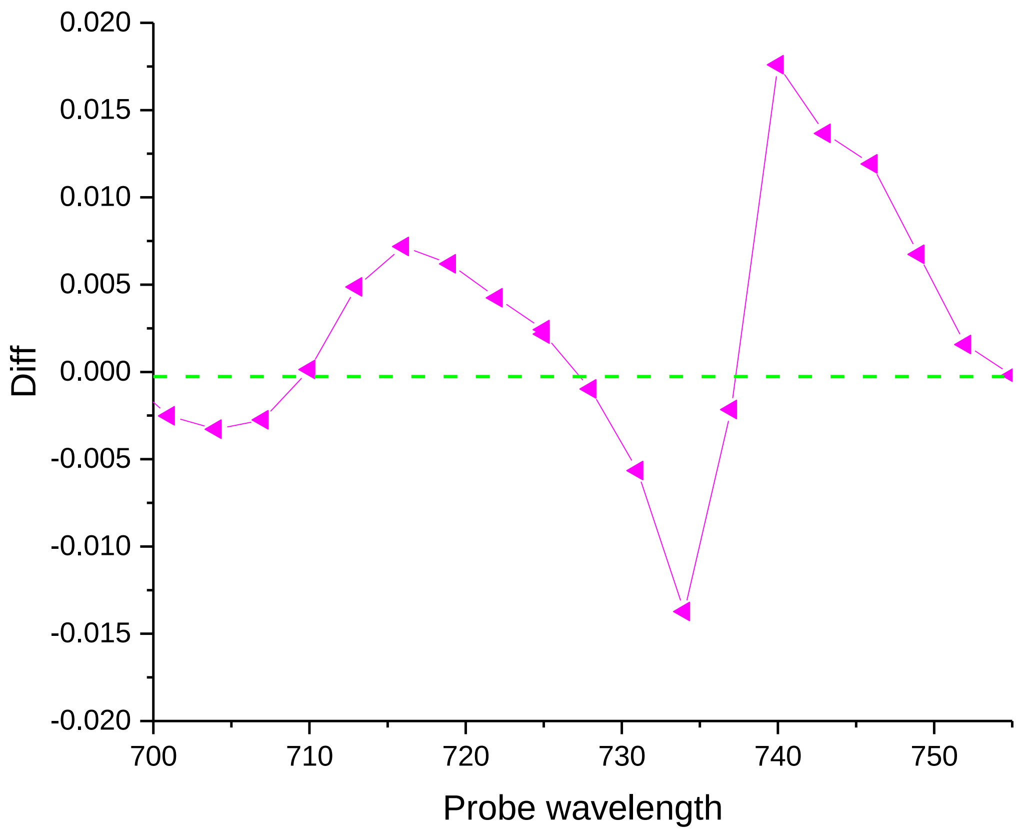


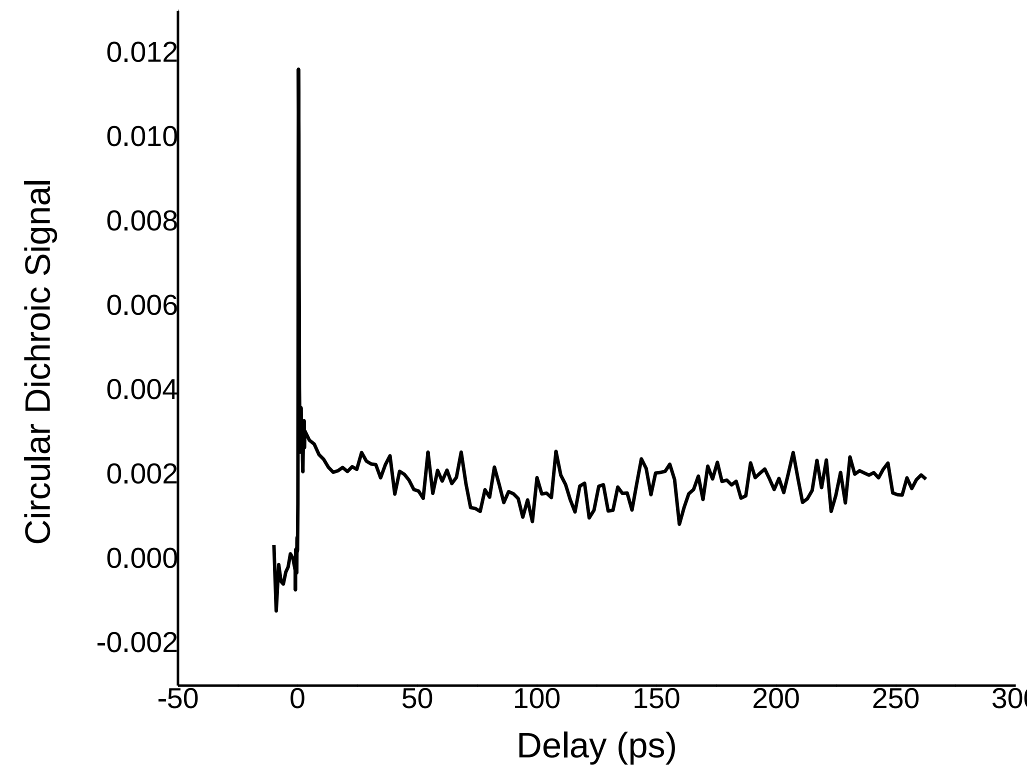
Reflection contrast: Multiple peaks from moire minibands.



PL with 532nm excitation: Single interlayer exciton emission peak at 870nm.

Calibration with intralayer pump – intralayer probe

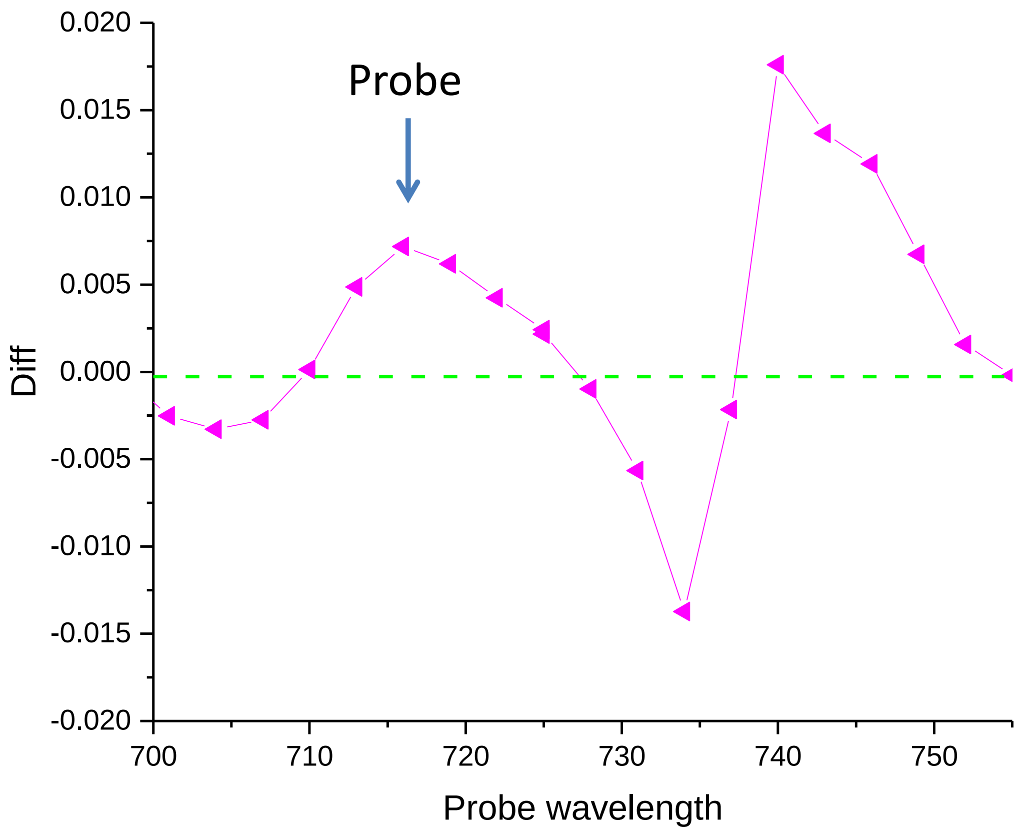


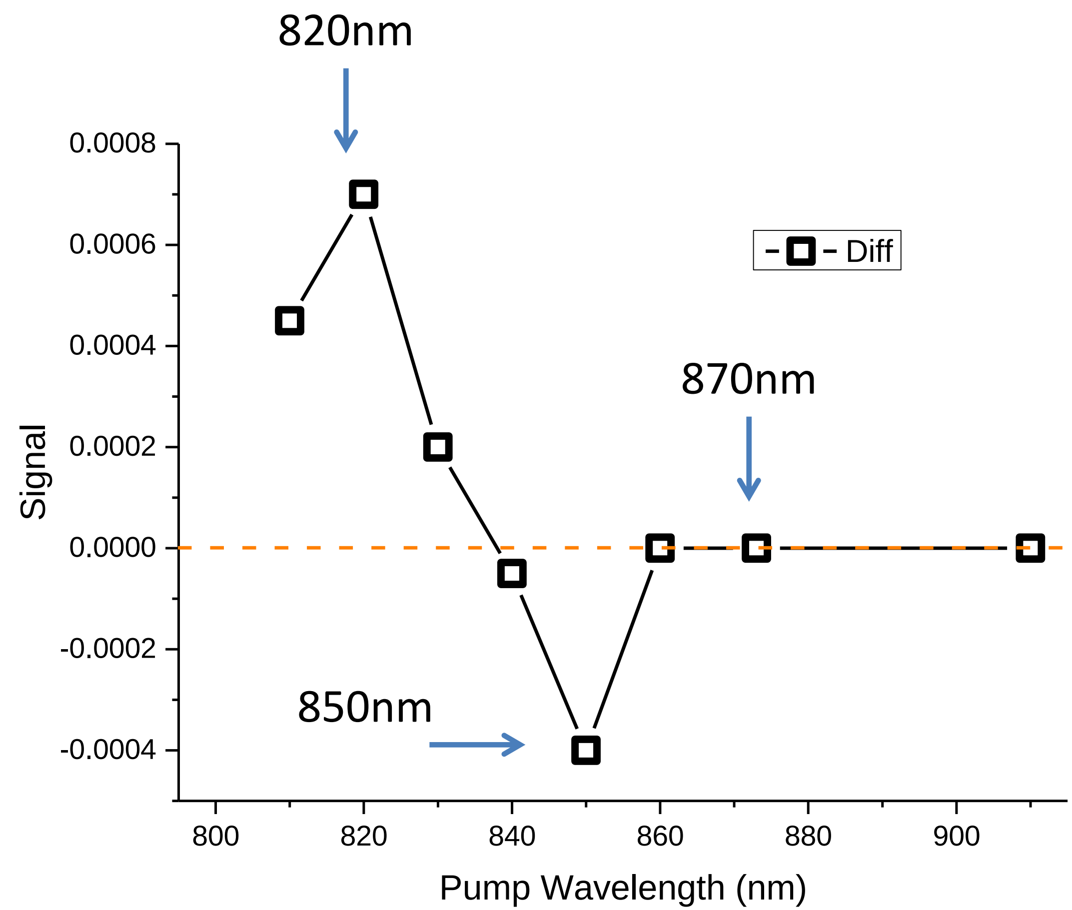


LCP Pump at 650nm creates **intralayer** excitons at K valley. K valley holes induce different response of K and K' valleys, as detected by the probe.

Representative dynamics, near constant over the whole range.

Intralayer Pump – intralayer probe

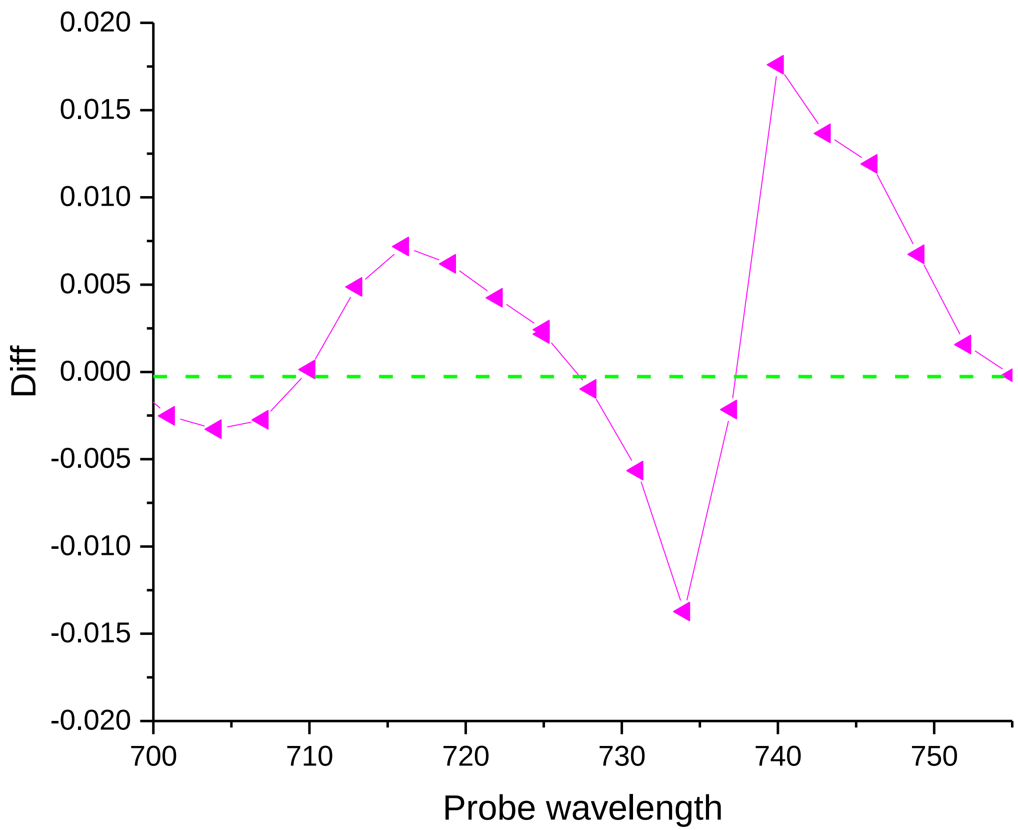




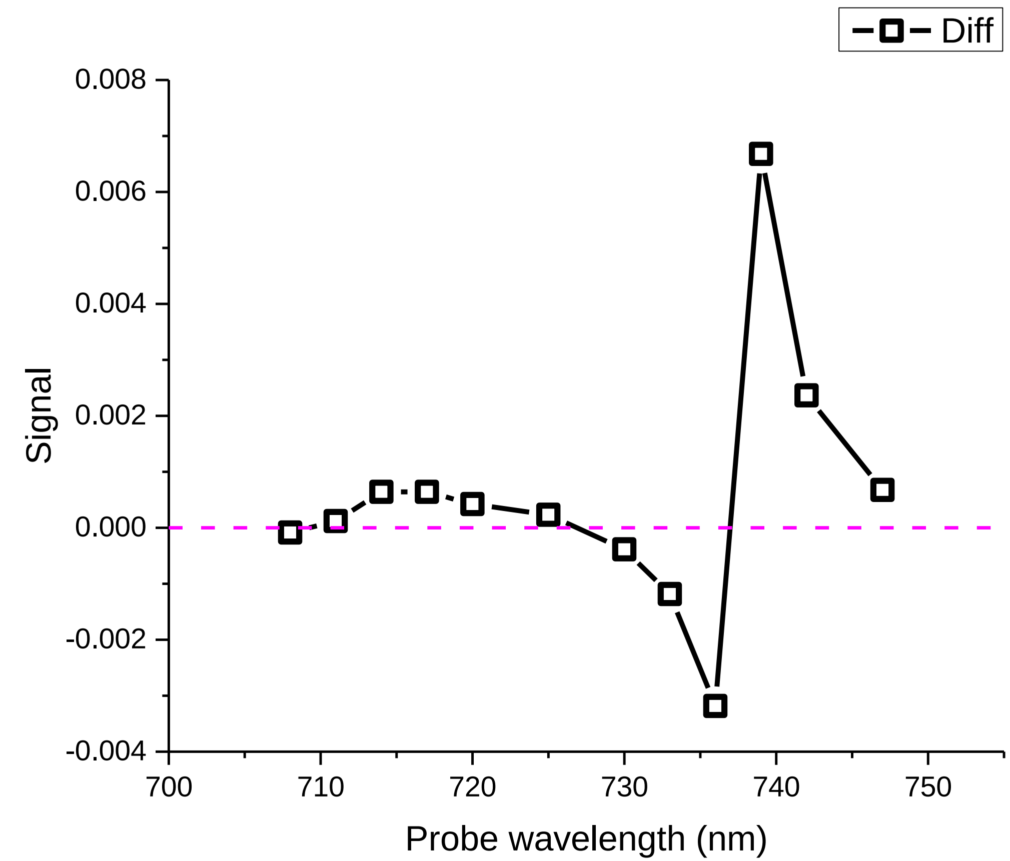
LCP Pump at 650nm creates **intralayer** excitons at K valley. K valley holes induce different response of K and K' valleys, as detected by the probe.

Fixed probe at 714nm
870nm : Weak absorption/valley-insensitive
820 and 850nm: Direct exciton states

Probing valley selection rule for 820nm state

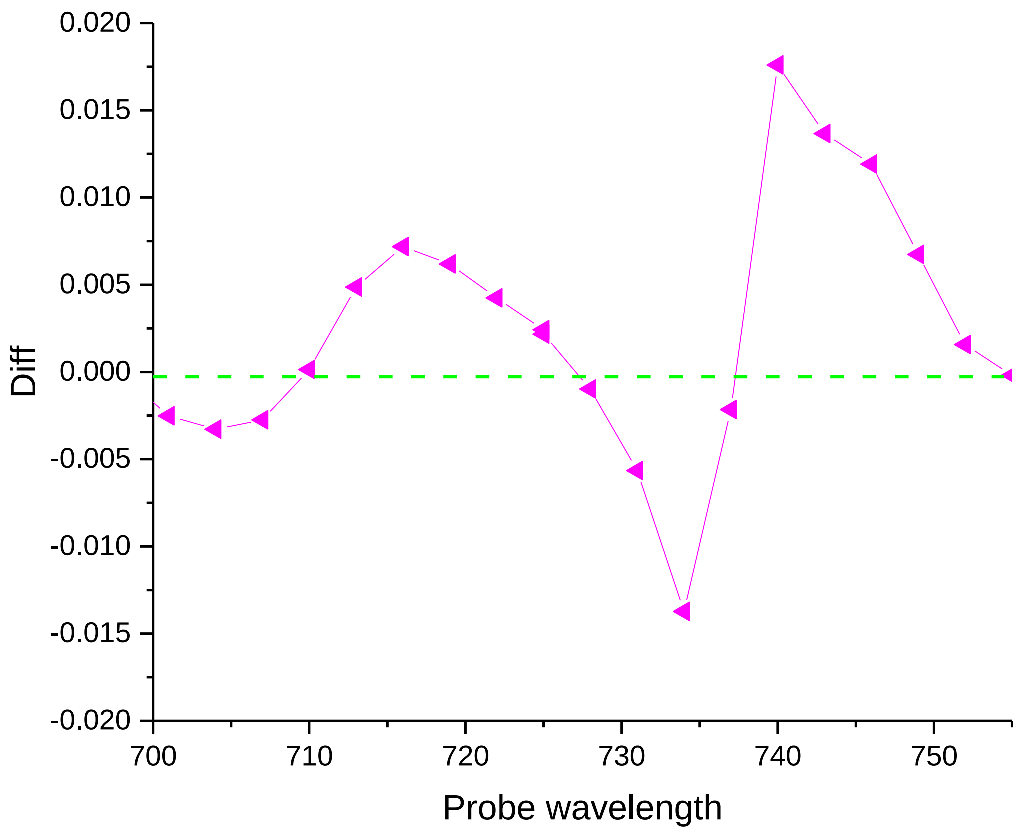


LCP pumping intralayer exciton:
Hole is in the K valley.

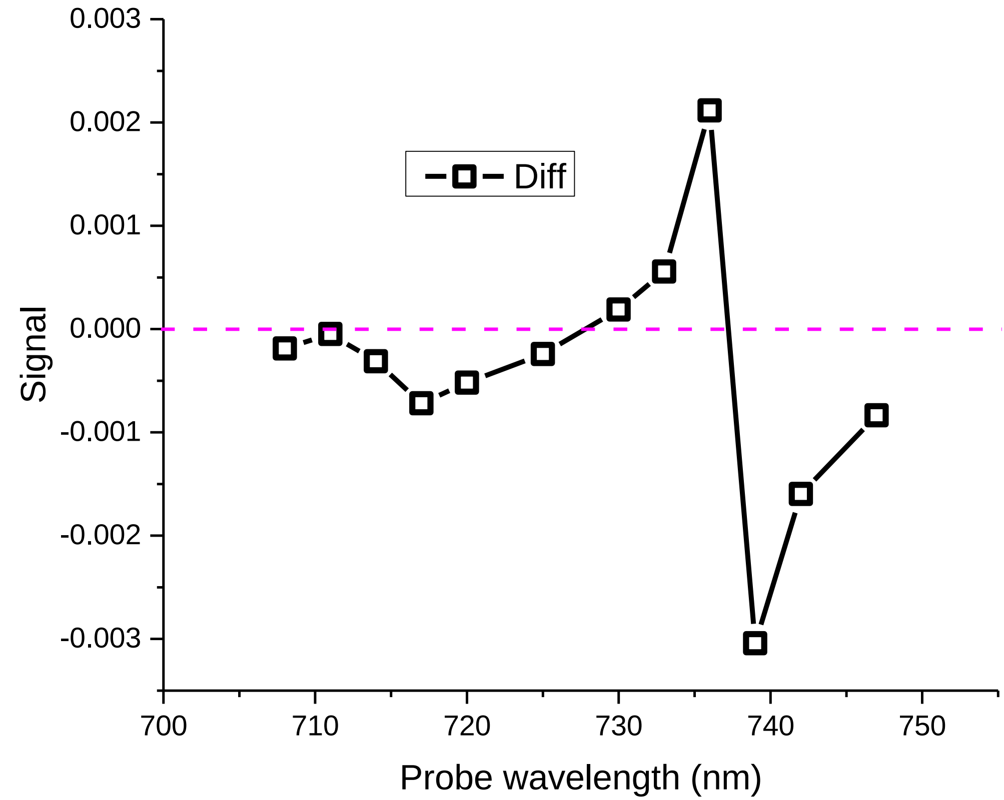


LCP pumping interlayer exciton at 820nm.
Hole is in the K valley.

Probing valley selection rule for 850nm state



LCP pumping intralayer exciton:
Hole is in the K valley.



LCP pumping interlayer exciton at 850nm.
Hole is in the K’ valley.

Extracting the moire contribution

Wavelength	Strong absorption	Band contribution	Moire contribution	Total
870nm				
850nm	Y			
820nm	Y			
Intralayer exciton	Y			
Comment For AA stacked case	Directly obtained from pump probe			

Extracting the moire contribution

Wavelength	Strong absorption	Band contribution	Moire contribution	Total
870nm				
850nm	Y			+1
820nm	Y			+1
Intralayer exciton	Y			+1
Comment For AA stacked case	Directly obtained from pump probe			Directly obtained from pump probe

Extracting the moire contribution

Wavelength	Strong absorption	Band contribution	Moire contribution	Total
870nm				
850nm	Y	K’ h↓; K’ e↓:-1		+1
820nm	Y	K h↑; K e↑: +1		+1
Intralayer exciton	Y	K h↑; K e↑: +1		+1
Comment For AA stacked case	Directly obtained from pump probe	Directly obtained from pump probe		Directly obtained from pump probe

Extracting the moire contribution

Wavelength	Strong absorption	Band contribution	Moire contribution	Total
870nm				
850nm	Y	K’ h↓; K’ e↓:-1	2	+1
820nm	Y	K h↑; K e↑: +1	0	+1
Intralayer exciton	Y	K h↑; K e↑: +1	0	+1
Comment For AA stacked case	Directly obtained from pump probe	Directly obtained from pump probe	Can be obtained by total minus band	Directly obtained from pump probe

Unambiguous determination of the “moiré” contribution.
Demonstration of exciton states with opposite valley selection rule.

Extracting the moire contribution

Wavelength	Bright?	Band contribution	Moire contribution	Total
870nm	N	K h↑; K e↓: 0	-2	-2 = +1
850nm	Y	K h↑; K e↑:+1	-2	-1
820nm	Y	K h↑; K e↑: +1	0	+1
Intralayer exciton	Y	K h↑; K e↑: +1	0	+1
Comment For AA stacked case	Directly obtained from pump probe	Directly obtained from pump probe	Can be obtained by total minus band	Directly obtained from pump probe

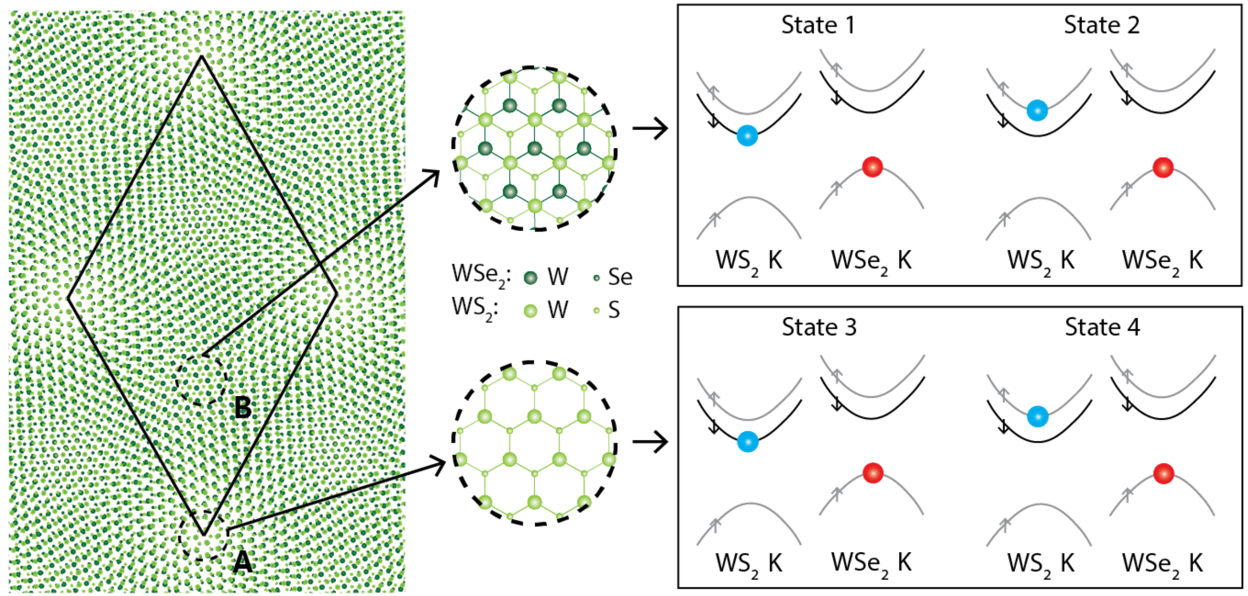
Dark interlayer exciton gains circular helicity from the moire contribution!

Multiple interlayer exciton states in the moire superlattice

State	Energy (eV)	Oscillator Strength	Total QAM	Hole	Electron	Spin QAM	Valley QAM	**Moiré QAM**	**Moiré Position**
1	1.43	Weak	+1 = -2	K ↑	K↓	-1	+1	**-2**	**B**
2	1.46	Strong	-1	K ↑	K ↑	0	+1	**-2**	**B**
3		Not observed	0	K ↑	K ↓	-1	+1	**0**	**A**
4	1.51	Strong	+1	K ↑	K ↑	0	+1	**0**	**A**
5	1.43	Weak	-1 = +2	K’ ↓	K’ ↑	1	-1	**+2**	**B**
6	1.46	Strong	+1	K’ ↓	K’ ↓	0	-1	**+2**	**B**
7		Not observed	0	K’ ↓	K’ ↑	1	-1	**0**	**A**
8	1.51	Strong	-1	K’ ↓	K’ ↓	0	-1	**0**	**A**

Jin, et al., *Nat. Phys.* (2019)

Multiple interlayer exciton states in the moire superlattice



Spin, Valley, Moiré degree of freedom:
$2 \times 2 \times 2 = 8$ interlayer exciton states!

4 states in the K valley (K' valley can be obtained from T symmtery).

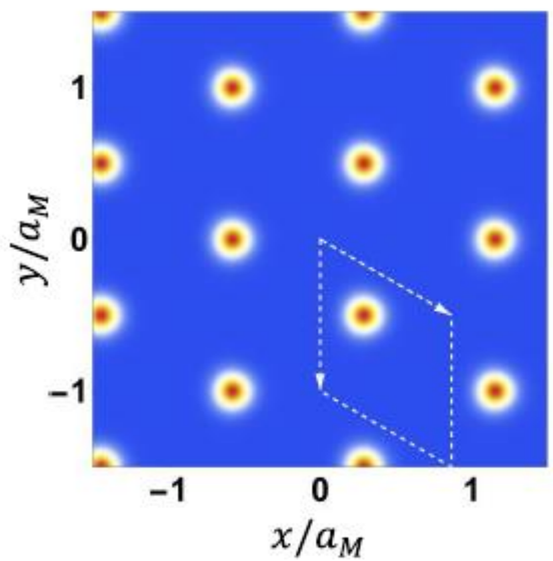
Summary

- Successfully measured interlayer exciton absorption to determine nature of all moire excitons.
- Unambiguously isolated the moire contribution to the QAM of the excitons, giving rise to distinctive selection rules at different sites
- This directly leads to an intrinsic "spin-orbit" coupling in the lattice
- Demonstrated the localization of interlayer exciton Wannier function.

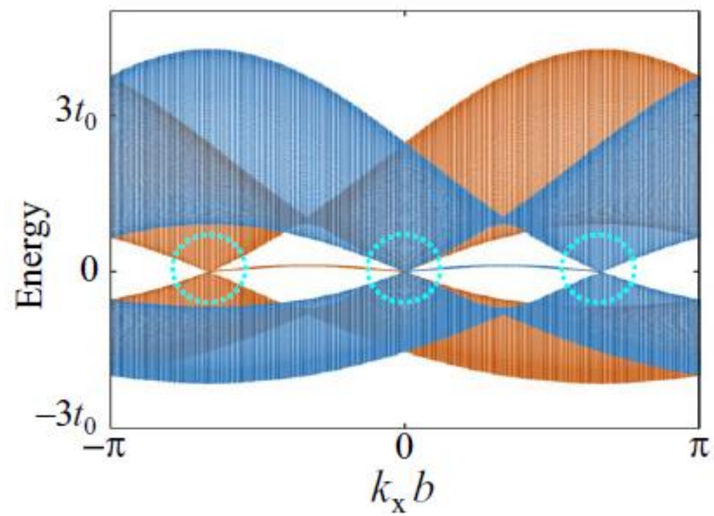
⇒ **Localized exciton lattice with "spin-orbit coupling".**

Localized exciton lattice with "spin-orbit coupling"

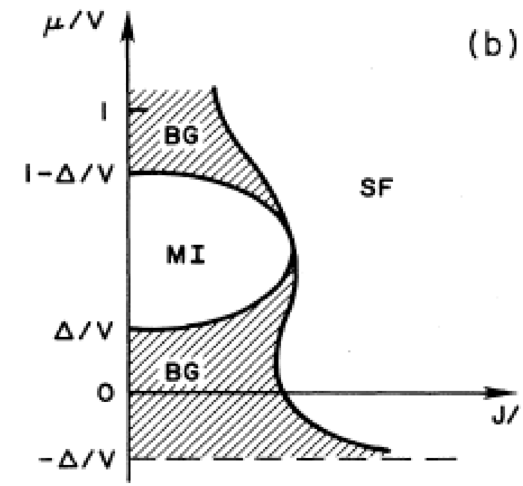
Exciton "solids"



Topological excitons



Bosonic physics



Fisher, et al., *PRB* (1989)
Wu et al., *PRL*, (2017)
Yu et al., *Sci. Adv.* (2017)

Thank you!