

Flatland*

An introduction to 2D materials and quasiparticle dynamics in monolayer TMDCs

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September 12, 2026

*approximately

Where we are going

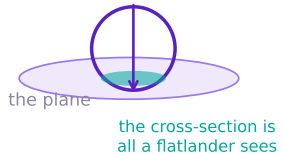
1. Flatland
2. What is a 2D material?
3. Confinement and quantum wells
4. Valleys
5. Quasiparticles
6. Dynamics: why femtoseconds
7. Experimental techniques

Flatland

A world with one dimension too few

- Abbott, 1884: a square who cannot perceive a third dimension.
- Our materials are Flatland. Light is the visitor from the dimension the electrons do not have.
- “(approximately)” — the layer has finite thickness, and its electric fields still live in 3D.
- That mismatch is where the physics comes from. It returns three times.

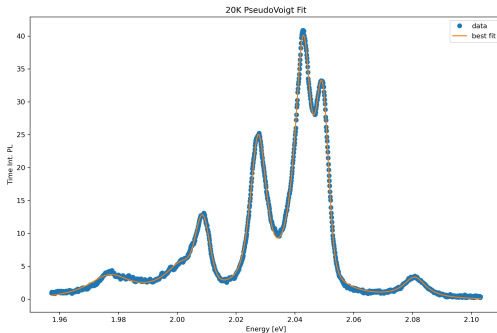
a sphere passes through Flatland



a monolayer, edge-on



The spectrum we are going to decode

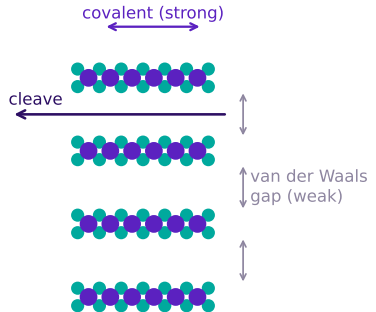


By the end: what (almost) every feature is, and how fast it lives.

What is a 2D material?

Layered van der Waals crystals

- Strong **covalent** bonds in plane.
- Weak **van der Waals** bonding between layers.
- Consequence: the crystal cleaves along the layers. (Sticky tape!)
- Down to a **single layer**, still crystalline, still stable in air (for the most part).



Transition metal dichalcogenides

- Formula MX_2 : M = Mo, W;
X = S, Se, Te.
- X–M–X sandwich, ~ 0.7 nm thick.

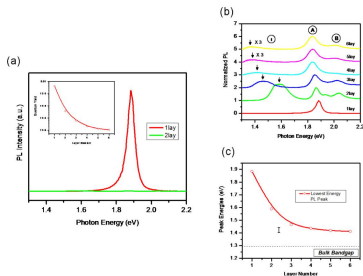
top view



side view: X–M–X sandwich



Indirect as bulk $\rightarrow$ direct as a monolayer



- PL quantum yield jumps by orders of magnitude from bilayer to monolayer [1, 2].
- The indirect gap shifts by more than 0.6 eV with thickness; the direct gap by only ~ 0.1 eV.

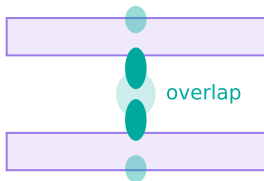
Why? Orbital character

- States at K : metal d_{z^2} , localised in the middle of the layer, screened by the chalcogens $\Rightarrow$ nearly *insensitive* to interlayer coupling.
- States at Γ and Λ : chalcogen p_z character, sticking out of the layer $\Rightarrow$ hybridise between layers, change with thickness.

K : metal d_{z^2} — stays mid-layer



Γ, Λ : chalcogen p_z — hybridises



Confinement and quantum wells

Particle in a box, briefly

- Confining a carrier costs energy, and the cost grows as $1/L^2$:

$$E_n = \frac{\hbar^2 \pi^2 n^2}{2m^* L^2}$$

- The criterion is: L small compared to the natural size of the state: $L \lesssim a_B$ [4].
- In a semiconductor a_B is a few nm, so nanometre-scale layers already do the job.

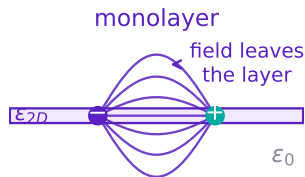
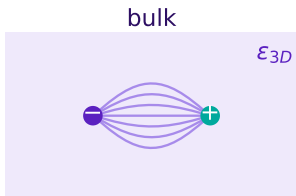
Grown wells vs. the monolayer

- Conventional QW: GaAs/AlGaAs, MBE-grown, $L \sim 10$ nm, lots of optimisation for an interface that is *nearly* atomically flat.
- Monolayer TMDC: $L \sim 0.7$ nm, relatively easy.
- 2D also gives a **step-like** density of states at the band edge as compared to $\sqrt{E}$ onset: sharp, strong optical transitions. **vacuum**.

Dielectric confinement - An important effect

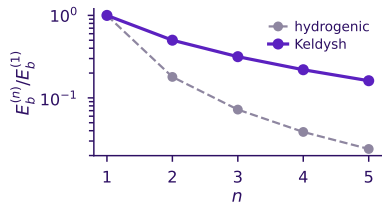
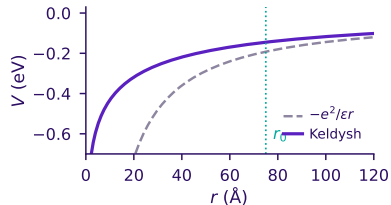
Dielectric confinement

The electron and the hole are trapped in the plane. Their attraction is not. The field lines **leave the layer**, pass through vacuum or substrate where nothing screens them, and come back. Confined, but not insulated [6].



Consequence: a non-hydrogenic Rydberg series

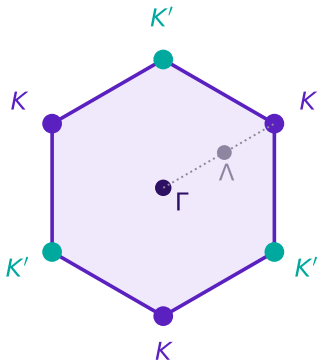
- Not $1/r$ but the Keldysh–Rytova potential: logarithmic at short range, $1/r$ beyond r_0 .
- So level spacings deviate from $1/n^2$. WS_2 on SiO_2 :
 $E_b = 0.32 \text{ eV}$, $E_g = 2.41 \text{ eV}$,
 $r_0 = 75 \text{ \AA}$ [6].
- $n = 3, 4, 5$ are hydrogenic; the deviation lives in $1s, 2s$.
- E_b and E_g shift together and cancel in E_{opt} : you cannot read E_b off the peak.



Valleys

Two corners of the Brillouin zone

- Hexagonal lattice $\Rightarrow$ hexagonal BZ, band extrema at the corners.
- Six corners, but only two are inequivalent: K and K' .
- They are related by time reversal: $K' = -K$.
- “Valley” = which corner a carrier lives in. A discrete label, like spin, but from the lattice [10].



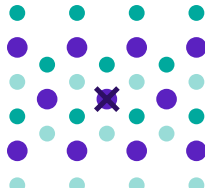
Broken inversion symmetry

- Bulk 2H and the bilayer are centrosymmetric.
- The monolayer is not: M and X sublattices are inequivalent.
- With inversion symmetry, any property at K must be identical at K' .

monolayer — no inversion centre

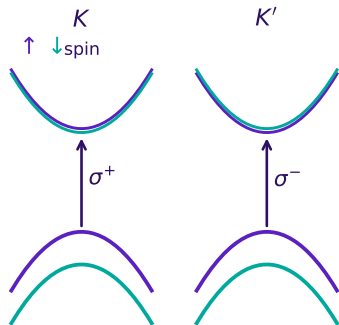


bilayer — inversion centre



Spin-orbit coupling and spin-valley locking

- Heavy metal atom $\Rightarrow$ strong SOC: the valence band splits by ~ 400 meV in WS_2 , ~ 150 meV in MoS_2 .
- The conduction band splits by tens of meV with opposite sign for Mo and W.
- Time reversal flips the splitting between K and K' , so spin and valley are locked: changing one means changing the other [3].



the splitting flips sign between valleys

Quasiparticles

What is a quasiparticle?

- A bare particle plus the disturbance it drags along, presented as a single new entity.
- Renormalised mass, renormalised energy, and, crucially, a finite lifetime.

The exciton

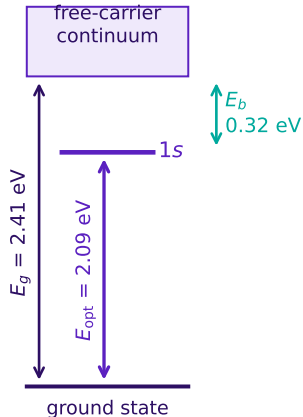
- An electron and a hole bound by their mutual Coulomb attraction.
- Wannier–Mott: radius $\gg$ lattice constant, delocalised, effective-mass description works. Frenkel: bound on a single site [4].

Optical gap vs. quasiparticle gap

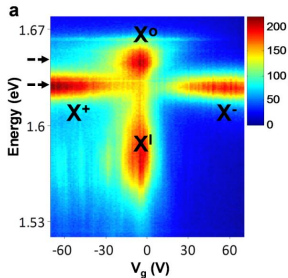
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$$E_{\text{opt}} = E_g - E_b$$

- E_g : cost of a *free* electron and a *free* hole.
- E_{opt} : where absorption and PL appear.



Trions, and gating

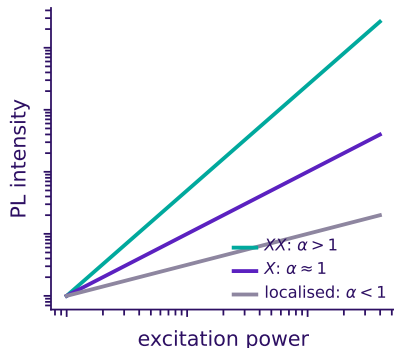


- A spare carrier can still bind: X^- or X^+ , red-shifted by 20–40 meV [5, 10].
- The sign of the doping picks which trion is formed.
- You can turn the quasiparticle on and off with a knob - the gate voltage.

- Spin-forbidden: electron and hole spins anti-aligned; emission needs a spin flip.
- Momentum-forbidden: electron and hole in different valleys (K - Λ , K - K'); emission needs a phonon.
- In W-based materials the lowest exciton state is *dark* [10]

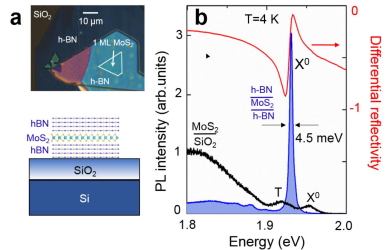
How to tell peaks apart

- Two excitons can bind into a biexciton; only at high density.
- Diagnostic: $I \propto P^\alpha$ — $\alpha > 1$ biexciton, $\alpha \approx 1$ exciton or trion, $\alpha < 1$ saturating localised state.
- Localised excitons: sharp sub-meV lines at low T ; some are single-photon emitters.



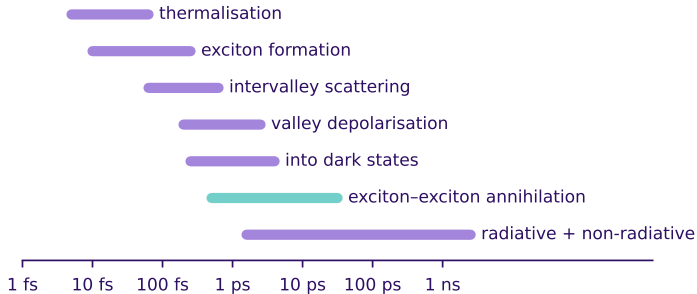
The hBN-encapsulation jumpscare

- Uniform dielectric environment $\Rightarrow$ no spatial scatter in E_b and E_g .
- Atomically flat, dangling-bond-free interface $\Rightarrow$ far less disorder and strain.
- Linewidths drop from ~ 50 meV to 2 meV at 4 K, which also bounds the lifetime at ≥ 0.33 ps [7].



Dynamics: why femtoseconds

Everything everywhere all at once, but at different timescales



- fs: carrier thermalisation, exciton formation from free carriers.
- sub-ps - ps: intervalley scattering, valley depolarisation, relaxation into dark states.
- ps - ns: radiative and non-radiative recombination, trion formation.
- Density-dependent: exciton-exciton annihilation [8].

The point of doing this in the time domain

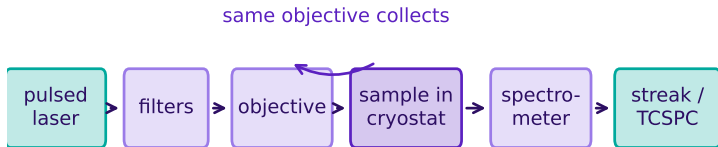
The population you detect is not the population you created

Between excitation and emission, carriers thermalise, form excitons, scatter between valleys, and fall into dark states. A time-integrated spectrum reports the average over all of it.

- Your time resolution has to beat the fastest process you care about.

Experimental techniques

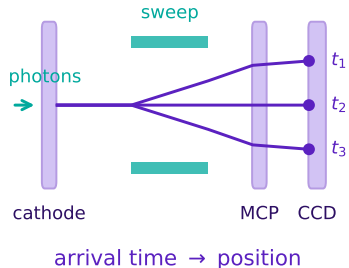
The generic setup



- Confocal geometry: the same objective excites and collects; spatial filtering rejects everything but the flake.
- Cryostat, because linewidth, dark-state occupation and lifetime are all strongly temperature-dependent, and you want to control your experiment (why would you not?)

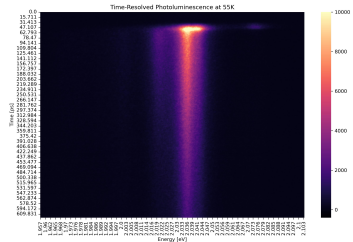
Streak camera: the principle

- Using an interesting proxy for photon generation time.
- Photon $\rightarrow$ photocathode $\rightarrow$ photoelectron: an optical timing problem becomes an electron-deflection problem.



Streak camera: what you get

- A single image is a 2D map of (λ, t) : slice horizontally for a time-resolved spectrum, vertically for a decay at one energy.



The instrument response function

- What you measure is the true decay **convolved** with the IRF:

$$I_{\text{meas}}(t) = \int \text{IRF}(t') I(t-t') dt'$$

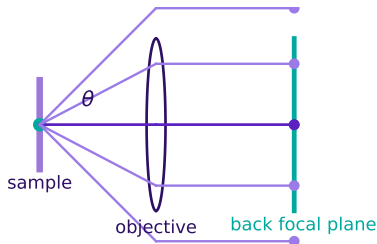
- Measure the IRF with scattered laser light, then **fit with reconvolution** rather than deconvolving numerically.
- You can extract lifetimes somewhat *below* the IRF width, not a good idea, though

Momentum-resolved (Fourier-plane) imaging

- The same trick as the streak camera, one axis over. There, time became position; here, **angle** becomes position.
- A high-NA objective maps emission angle onto position in the back focal plane:

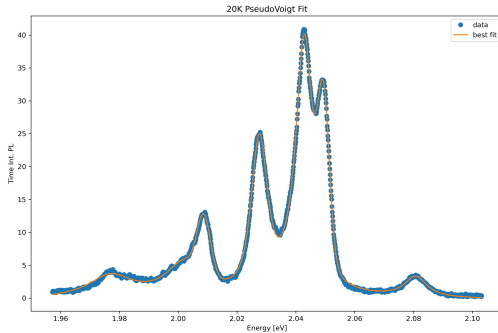
$$k_{\parallel} = \frac{2\pi}{\lambda} \sin \theta, \quad k_{\parallel}^{\max} = \frac{2\pi}{\lambda} \text{NA}$$

- Image the **BFP** rather than the sample plane. No new light source — just a different plane to look at.



angle $\rightarrow$ position, $r \propto \sin \theta$

The spectrum



- [1] K. F. Mak *et al.*, Atomically thin MoS₂: a new direct-gap semiconductor, *Phys. Rev. Lett.* **105**, 136805 (2010).
- [2] A. Splendiani *et al.*, Emerging photoluminescence in monolayer MoS₂, *Nano Lett.* **10**, 1271 (2010).
- [3] D. Xiao *et al.*, Coupled spin and valley physics in monolayers of MoS₂ and other group-VI dichalcogenides, *Phys. Rev. Lett.* **108**, 196802 (2012).
- [4] C. F. Klingshirn, *Semiconductor Optics*, 4th ed., Springer, Berlin (2012).
- [5] J. S. Ross *et al.*, Electrical control of neutral and charged excitons in a monolayer semiconductor, *Nat. Commun.* **4**, 1474 (2013).
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- [7] F. Cadiz *et al.*, Excitonic linewidth approaching the homogeneous limit in MoS₂-based van der Waals heterostructures, *Phys. Rev. X* **7**, 021026 (2017).
- [8] T. Mueller and E. Malic, Exciton physics and device application of two-dimensional transition metal dichalcogenide semiconductors, *npj 2D Mater. Appl.* **2**, 29 (2018).

- [9] G. Wang *et al.*, Colloquium: Excitons in atomically thin transition metal dichalcogenides, *Rev. Mod. Phys.* **90**, 021001 (2018).
- [10] A. Arora, Magneto-optics of layered two-dimensional semiconductors and heterostructures: progress and prospects, *J. Appl. Phys.* **129**, 120902 (2021).

Thank you

Questions?

Backup: Keldysh potential

$$V(r) = -\frac{\pi e^2}{2r_0} \left[H_0\left(\frac{r}{r_0}\right) - Y_0\left(\frac{r}{r_0}\right) \right]$$

- H_0 : Struve function. Y_0 : Bessel function of the second kind.
- $r \gg r_0$: recovers $1/r$. $r \ll r_0$: logarithmic.
- r_0 relates to the in-plane polarisability of the layer. For WS_2 on SiO_2 , the fit gives $r_0 = 75 \text{ \AA}$; *ab initio* for suspended WS_2 gives $38 \text{ \AA}$ and $E_b \approx 0.5 \text{ eV}$.