Electronic and optical properties of 2D (atomically thin) InSe crystals

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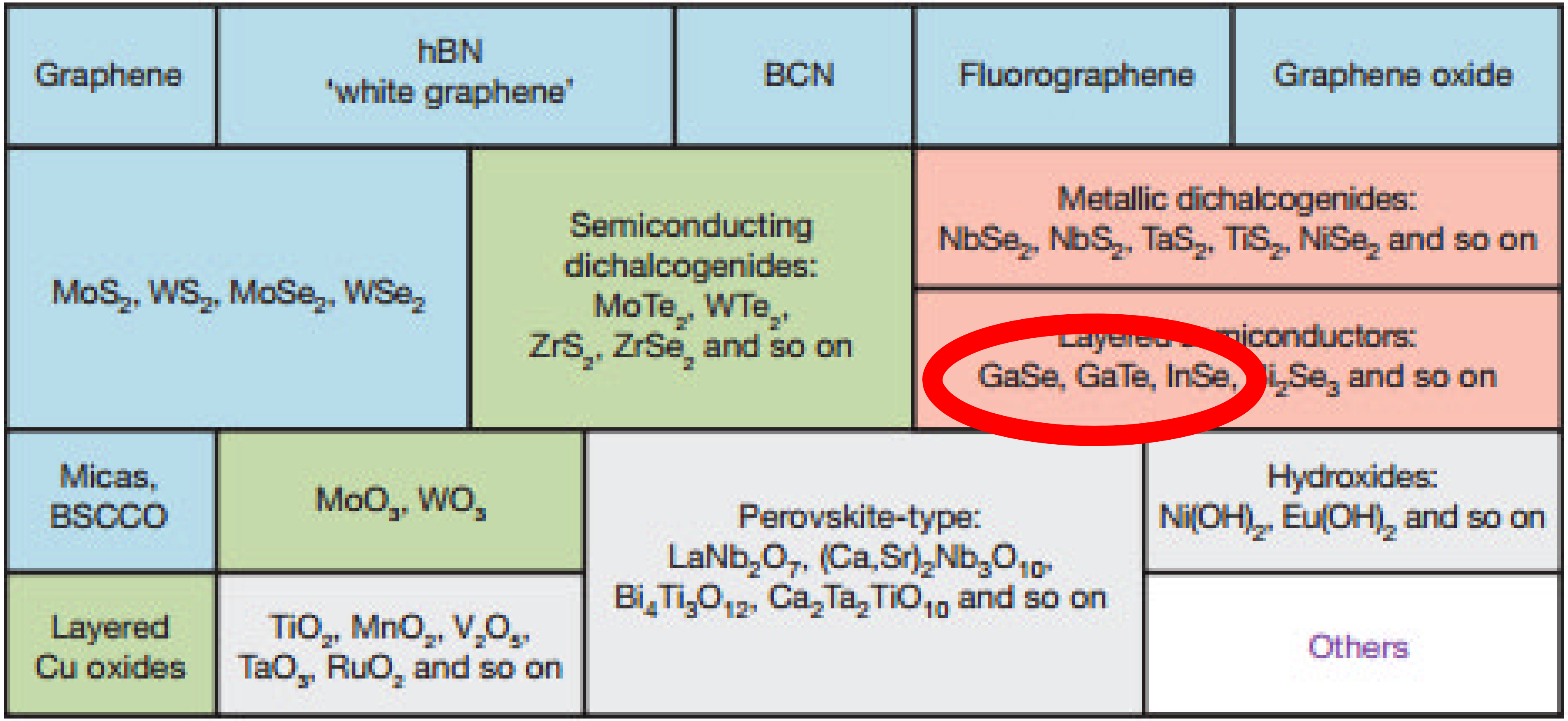
Zoo of 2D Materials

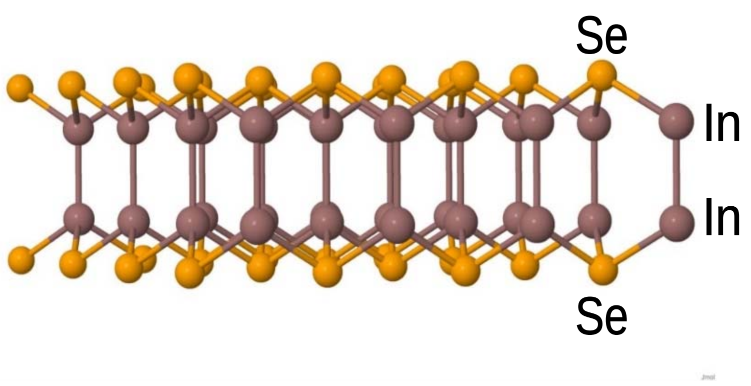


layered substances with covalent bonding within the layers and van der Waals coupling between the layers

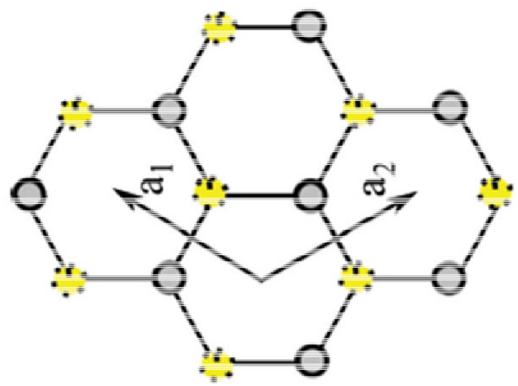
Electronic and optical properties of 2D (atomically thin) InSe crystals

- band and gaps for mono- and few-layer γ-InSe
- optical and transport properties (th+exp)

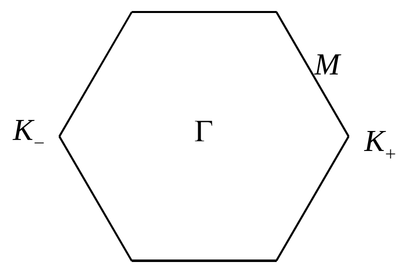
side view



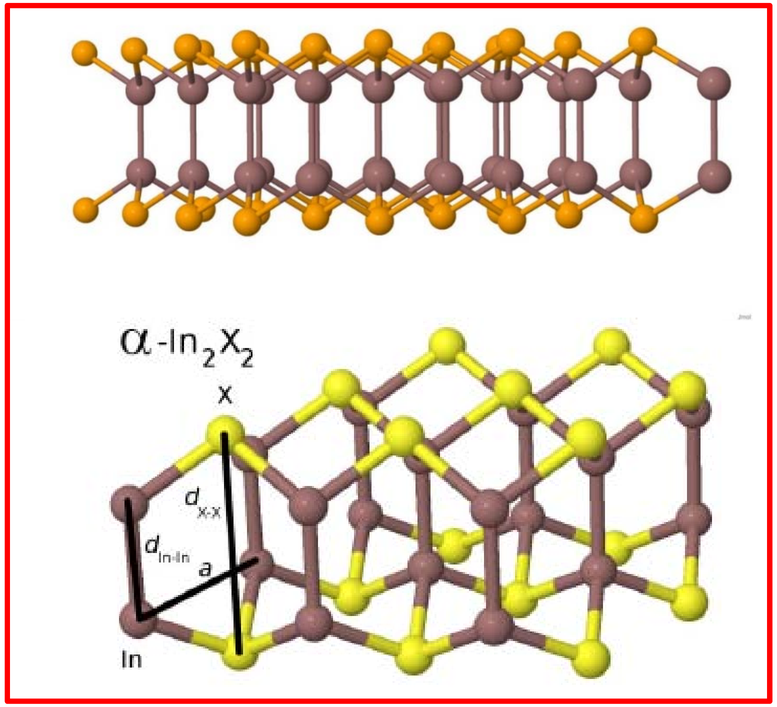
top view



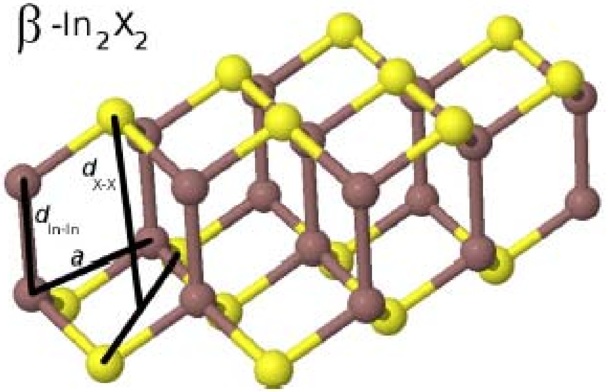
Brillouin zone



Monolayer (In_2Se_2)

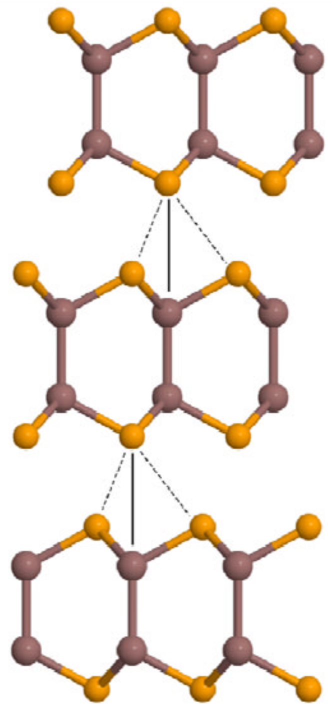


z→-z mirror symmetry



Zolyomi, Drummond, Fal'ko
PRB 89, 205416 (2014)

N-layer $In_{2N}Se_{2N}$



broken
z→-z mirror
symmetry

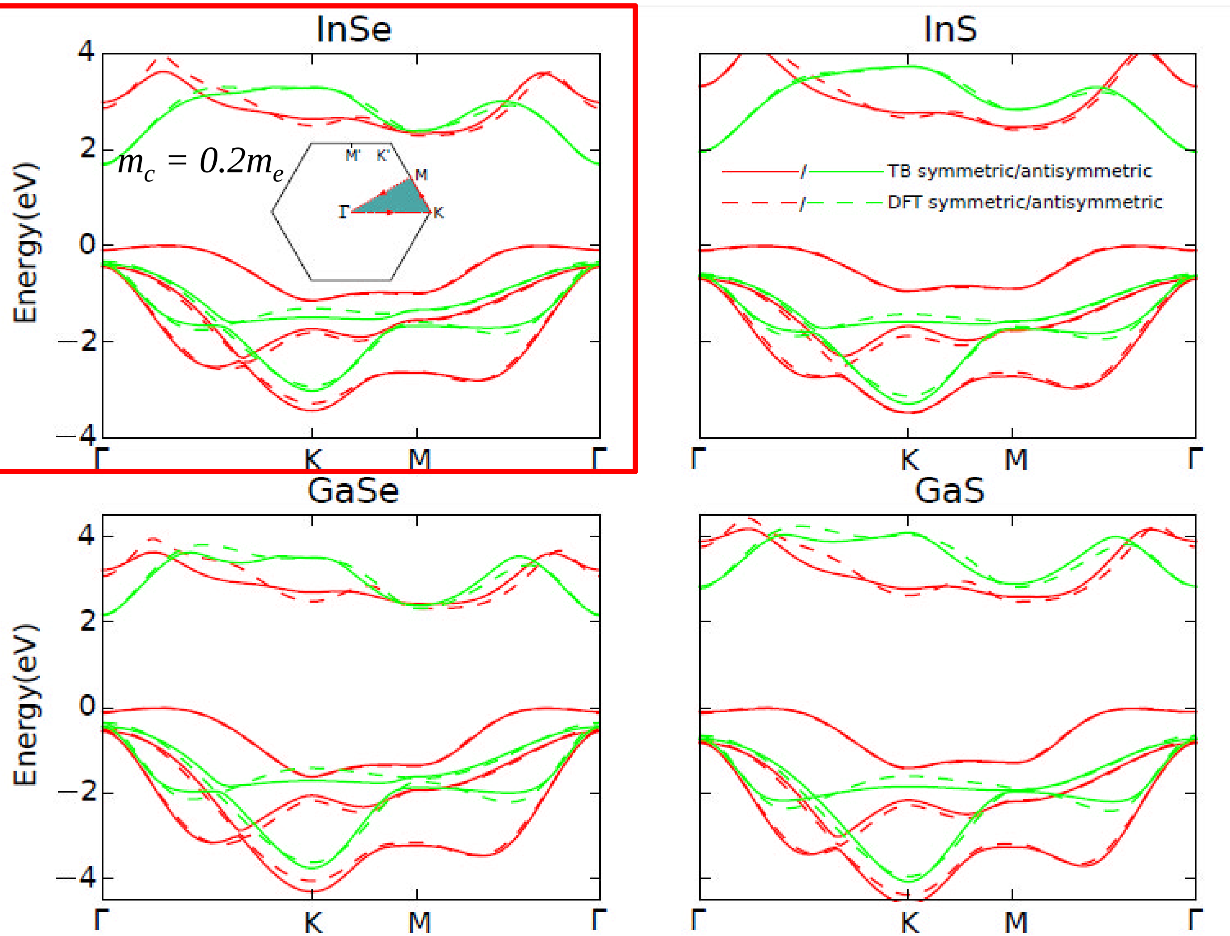
Monolayers In_2S_2, Ga_2X_2 have qualitatively very similar properties, but multilayer films have different lattice, bands/gaps and selection properties for optical transitions)

Zolyomi, Drummond, Fal'ko
PRB 87, 195403 (2013)

why InSe

DFT bands for monolayer M_2X_2

most stable in ambient conditions
bulk **γ**-InSe

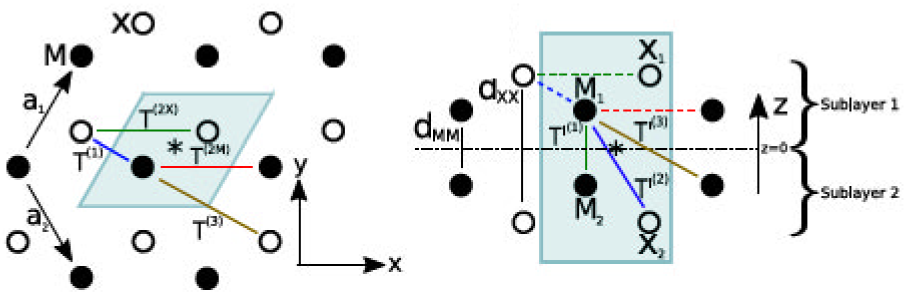


Relatively light electrons in conduction band:
good for high mobility

almost flat edge of the valence band:
opportunity to get strong correlations in hole-doped material

Zolyomi, Drummond, Fal'ko - PRB 87, 195403 (2013); PRB 89, 205416 (2014)

- Formulate tight binding model with all s and p orbitals and all nearest neighbour hoppings (MX, MM, XX) for monolayer (In_2Se_2) and bulk

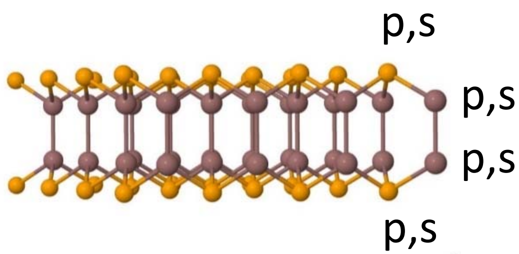


- Fit TB parameters to reproduce DFT bands in monolayer and bulk, after implementing a scissor correction to the band gap determined by comparing experimental and DFT gap values for bulk **γ**-InSe (1.45eV at low T and 1.25eV at room T)

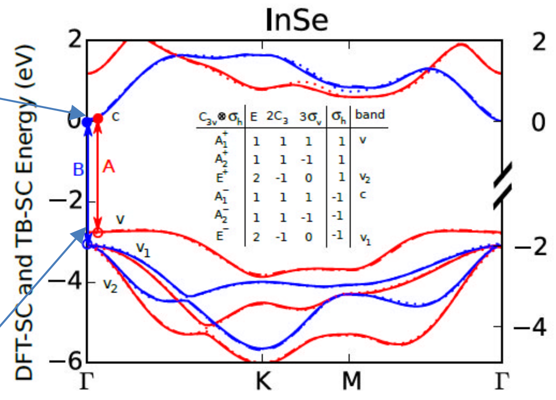
- Compute spectra of N-layer InSe ($In_{2N}Se_{2N}$) and matrix elements for z- and in-plane polarised optical transition, to compare with the experiment

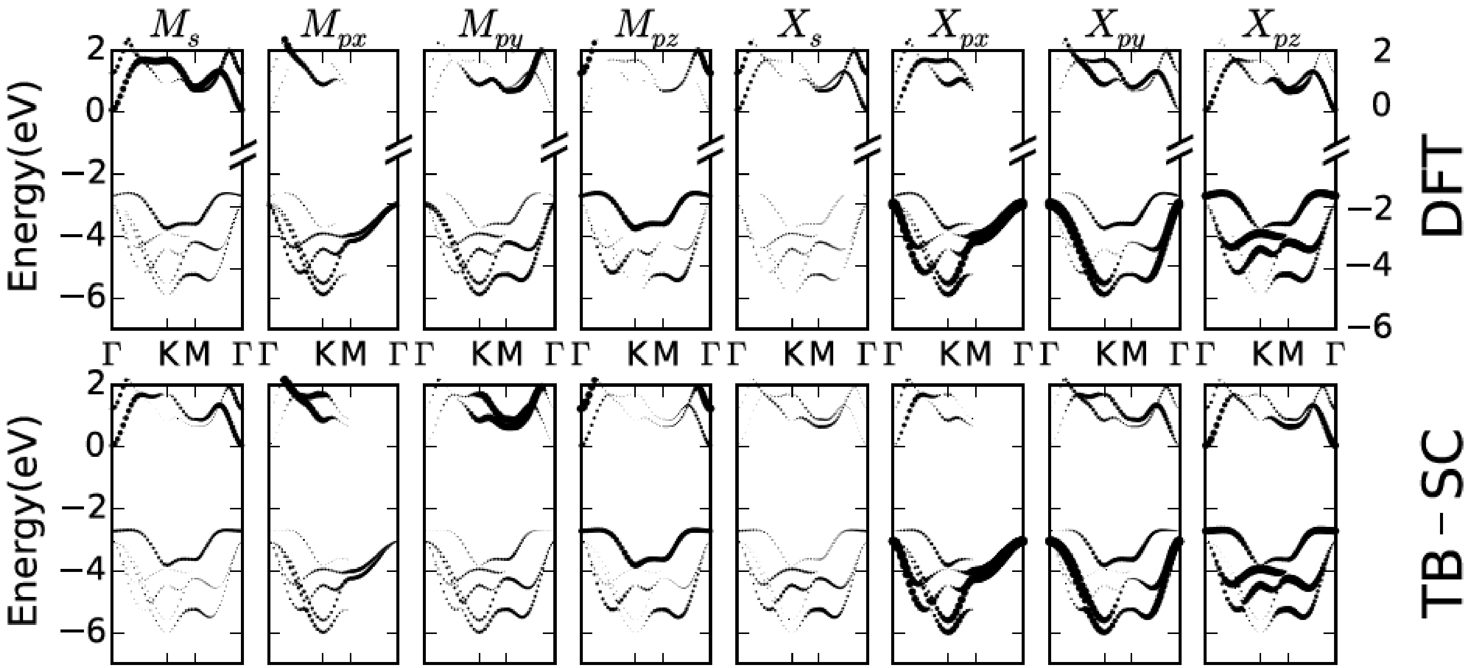
DFT-parametrised tight-binding model for $In_{2N}Se_{2N}$

$$0.115s^{\mathrm{In}} + 0.005p_z^{\mathrm{In}} + 0.059s^{\mathrm{Se}} + 0.088p_z^{\mathrm{Se}}$$



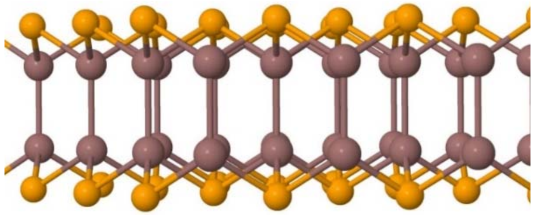
$$0.012s^{\mathrm{In}} + 0.043p_z^{\mathrm{In}} + 0.001s^{\mathrm{Se}} + 0.198p_z^{\mathrm{Se}}$$





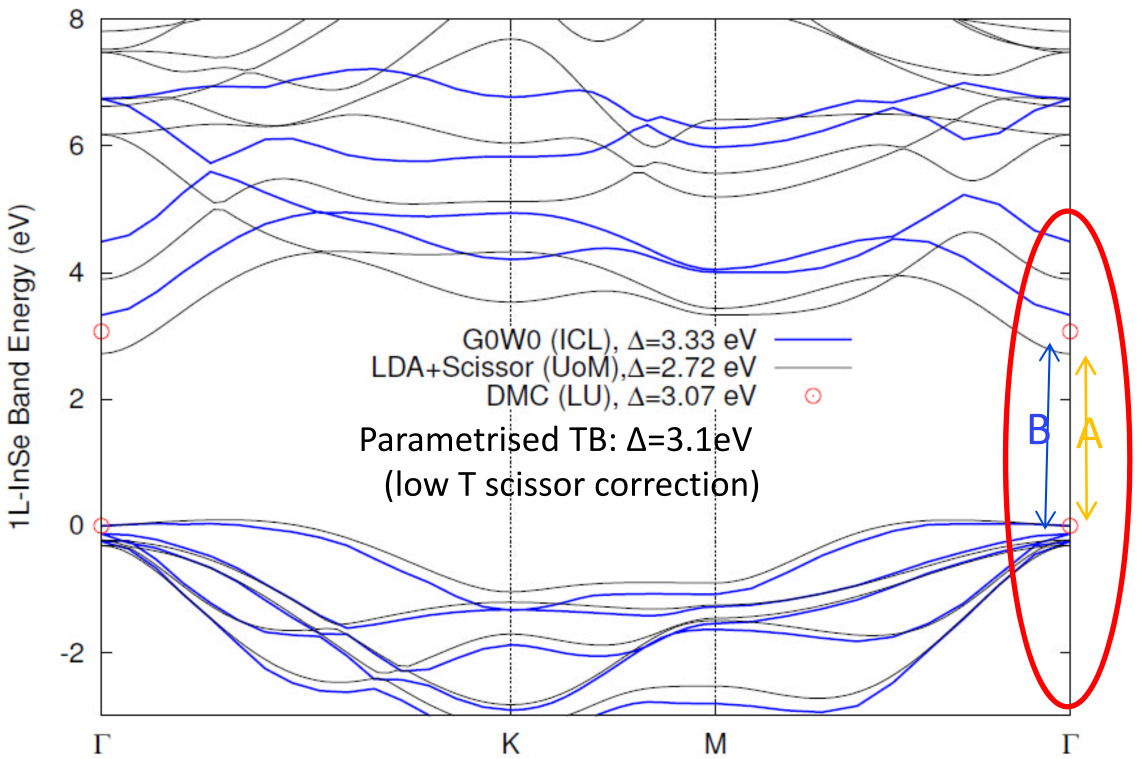
Magorrian, Zólyomi, Fal'ko - PRB 94, 245431 (2016)

DFT-parametrised tight-binding model for $In_{2N}Se_{2N}$



z→-z mirror symmetry

VB-CB transition 'A' across the gap is active in z-polarisation;
a higher-energy transition 'B' to a deeper double degenerate at the **Γ**-point valence band is active in the x-y polarisation



$0.115 s^{In} + 0.005 p_z^{In} + 0.059 s^{Se} + 0.088 p_z^{Se}$

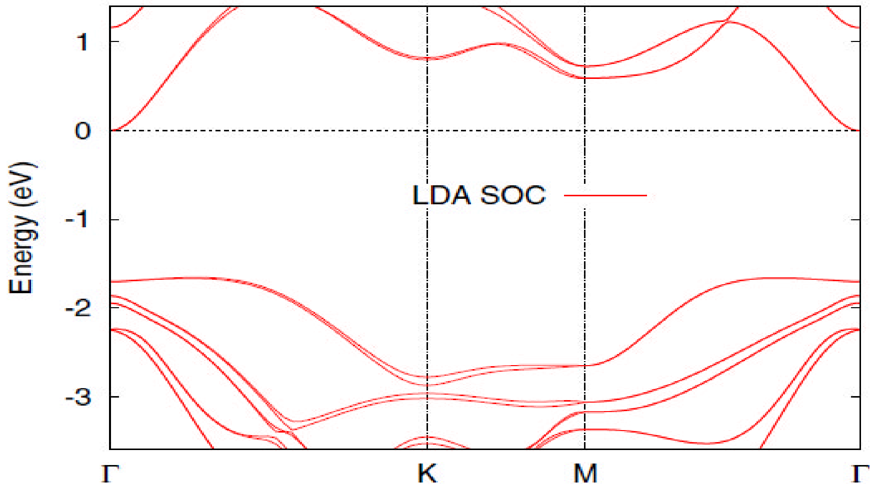
antisymmetric (odd)

symmetric (even)

$0.012 s^{In} + 0.043 p_z^{In} + 0.001 s^{Se} + 0.198 p_z^{Se}$

Role of spin-orbit coupling in monolayer InSe

$$\hat{H}_c = \hbar^2 k^2/2m_c + \gamma_c s_z k^3 \cos(3\phi)$$



$$\hat{H}_v(k,\phi) = E_0 + E_2 k^2 + E_4 k^4 + E_6 k^6 + E_8 k^8 + E_6' k^6 \cos(6\phi) + \gamma_v s_z k^3 \cos(3\phi)$$

odd/even z -> -z conduction/valence band states allow for

$$\frac{\hbar e}{c m_e} \beta_{sf} \vec{A} \times \vec{s}$$

spin-flip (due to atomic SO coupling) interband transition coupled with in-plane xy-polarised photons

4-band k•p theory for monolayer InSe

$$\hat{H}_c = \hbar^2 k^2/2m_c + \gamma_c s_z k^3 \cos(3\phi)$$

$$\hat{H}_v(k,\phi) = E_0 + E_2 k^2 + E_4 k^4 + E_6 k^6 + E_8 k^8 + E_6' k^6 \cos(6\phi) + \gamma_v s_z k^3 \cos(3\phi)$$

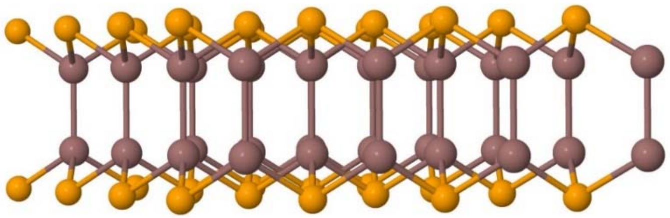
$$\hat{H} = \begin{pmatrix} H_c & E_z d_z + \frac{\hbar e}{c m_e}\beta_{sf}\vec{A}\times\vec{s} & \frac{\hbar e}{c m_e}\beta\vec{A} & 0 \\ E_z d_z + \frac{\hbar e}{c m_e}\beta_{sf}\vec{A}\times\vec{s} & H_v & 0 & \frac{\hbar e}{c m_e}\tilde{\beta}\vec{A} \\ \frac{\hbar e}{c m_e}\beta\vec{A} & 0 & H_{v_1} & 0 \\ 0 & \frac{\hbar e}{c m_e}\tilde{\beta}\vec{A} & 0 & H_{v_2} \end{pmatrix}$$

$$H_{v_{1(2)}} = E_0 + \frac{k^2}{2m} + \frac{k_x^2 - k_y^2}{2m'}\sigma_x + \frac{2k_x k_y}{2m'}\sigma_y$$

$C_{3v}\otimes\sigma_h$	E	$2C_3$	$3\sigma_v$	σ_h	band
A_1^+	1	1	1	1	v
A_2^+	1	1	-1	1	
E^+	2	-1	0	1	v_2
A_1^-	1	1	1	-1	c
A_2^-	1	1	-1	-1	
E^-	2	-1	0	-1	v_1

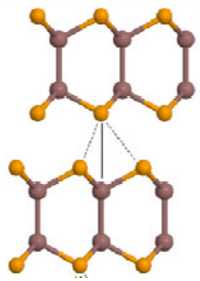
Electronic bands in $In_{2N}Se_{2N}$

Monolayer (In_2Se_2)

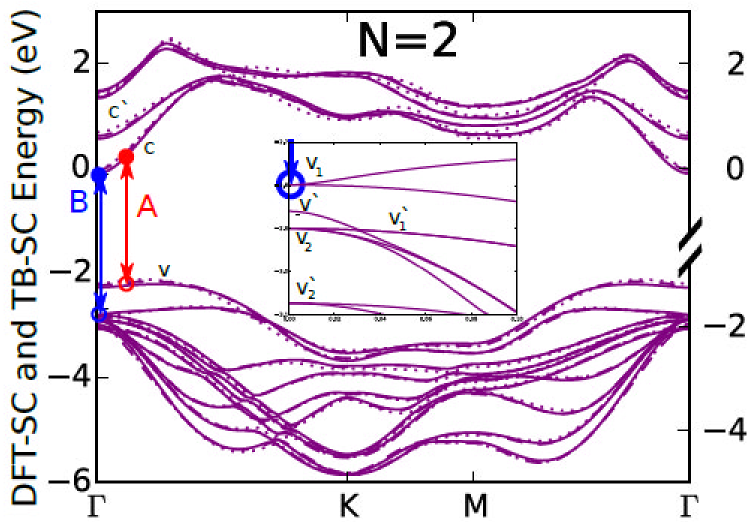


z→-z
mirror
symmetry

Bilayer (In_4Se_4)

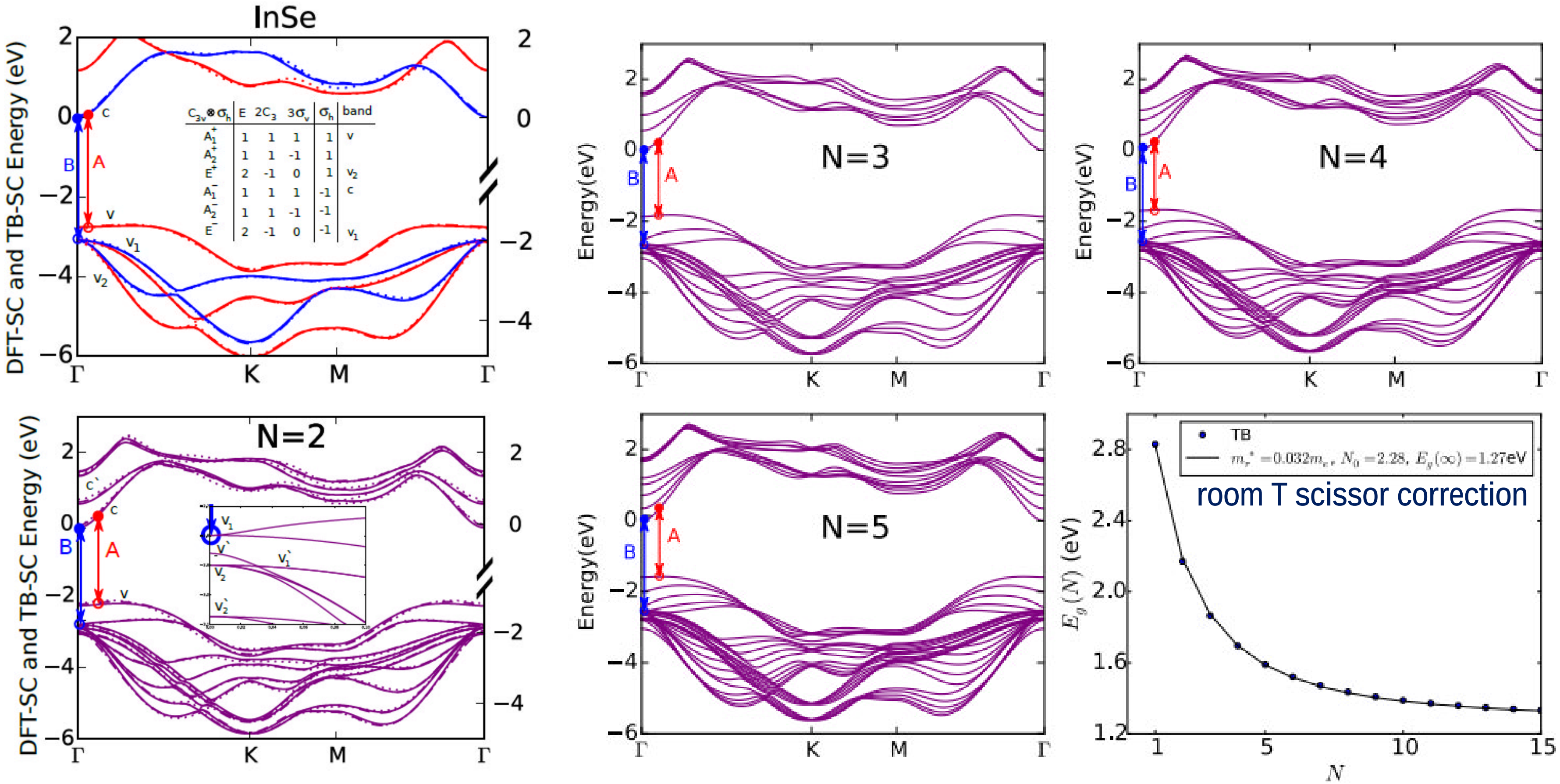


broken
mirror
symmetry



Magorrian, Zólyomi, Fal'ko - PRB 94, 245431 (2016)

Electronic bands in $In_{2N}Se_{2N}$

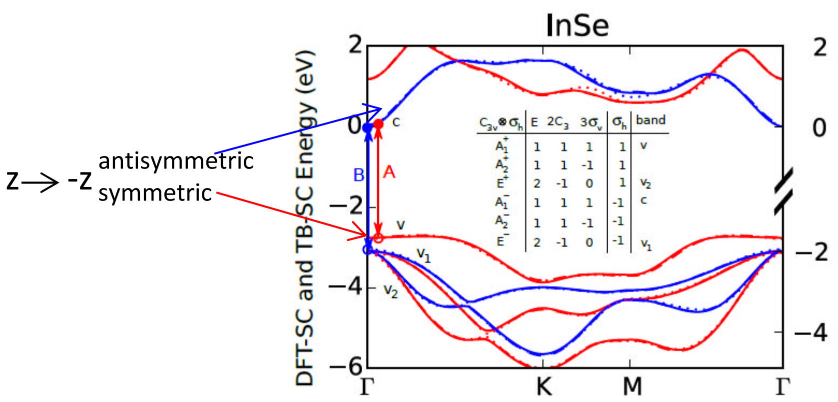


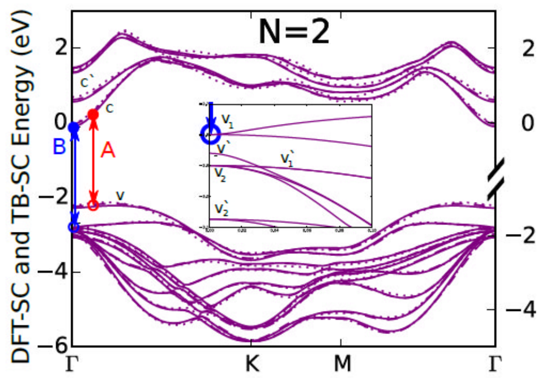
Relatively light electrons CB and wide interval of an almost flat VB edge also appear in few-layer InSe

Magorrian, Zólyomi, Fal'ko - PRB 94, 245431 (2016)

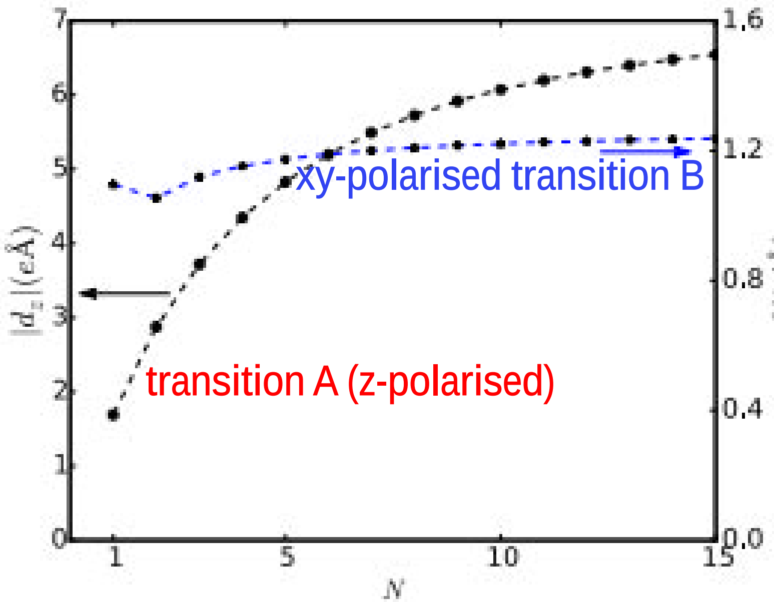
large variation of the band gap as a function of number of layers.

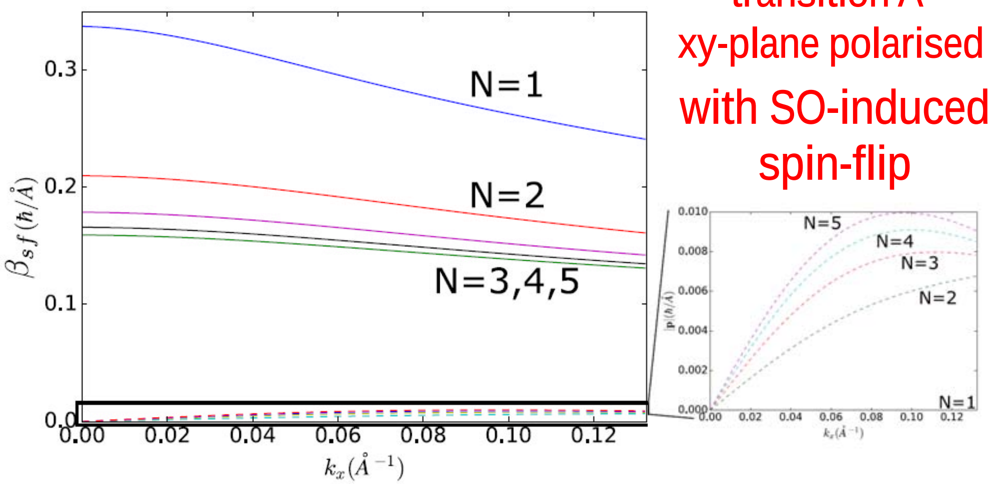
Optical transitions in $In_{2N}Se_{2N}$





broken
z→-z mirror
symmetry



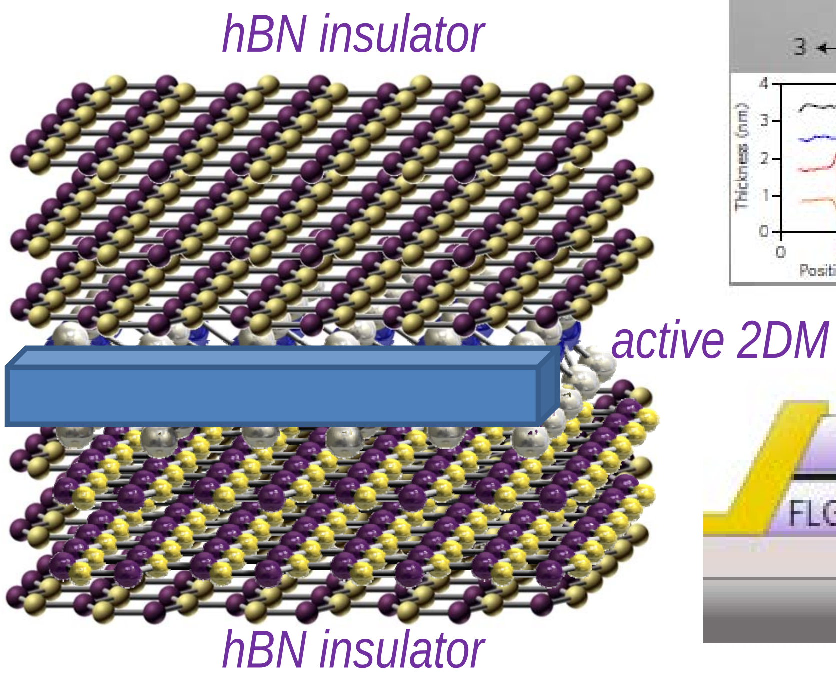


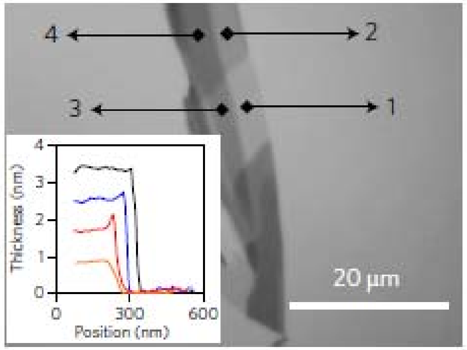
transition A
xy-plane polarised
with SO-induced
spin-flip

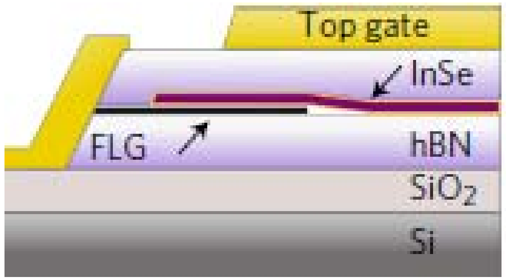
Magorrian, Zólyomi, Fal'ko - PRB 94, 245431 (2016)

hBN-encapsulated $In_{2N}Se_{2N}$

- hBN provides atomically flat substrate and clean encapsulation environment (low-resistance side contacts are possible)





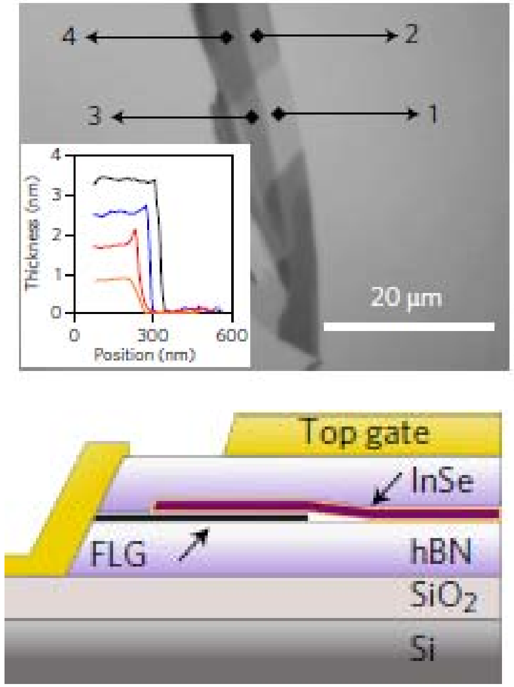


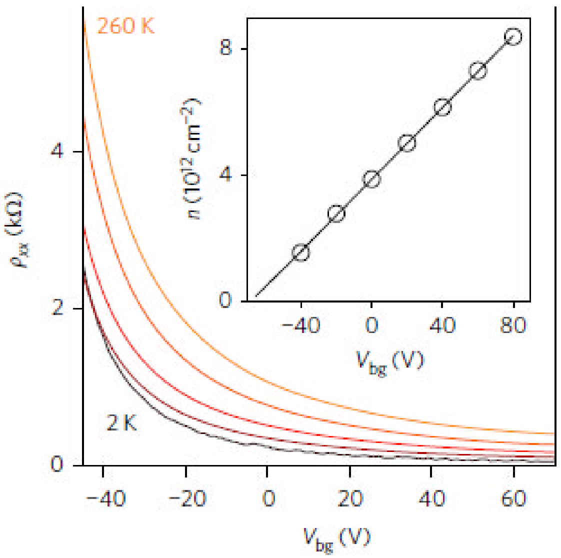
hBN is sp^2 – bonded insulator with a large band gap
 it is transparent and takes high voltage drop (useful for electrostatic gating)

- hBN and 2DM are glued together by weak van der Waals attraction, hence, they preserve their lattice structure and, hence, retain their basic physical properties.

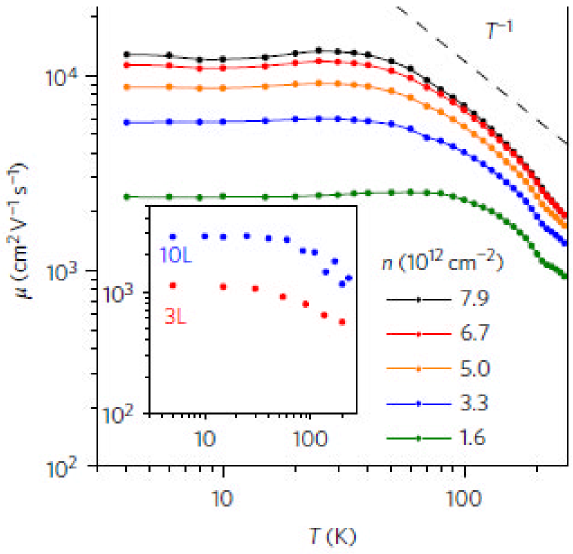
hBN-encapsulated $In_{2N}Se_{2N}$

- hBN provides atomically flat substrate and clean encapsulation environment (low-resistance side contacts are possible)



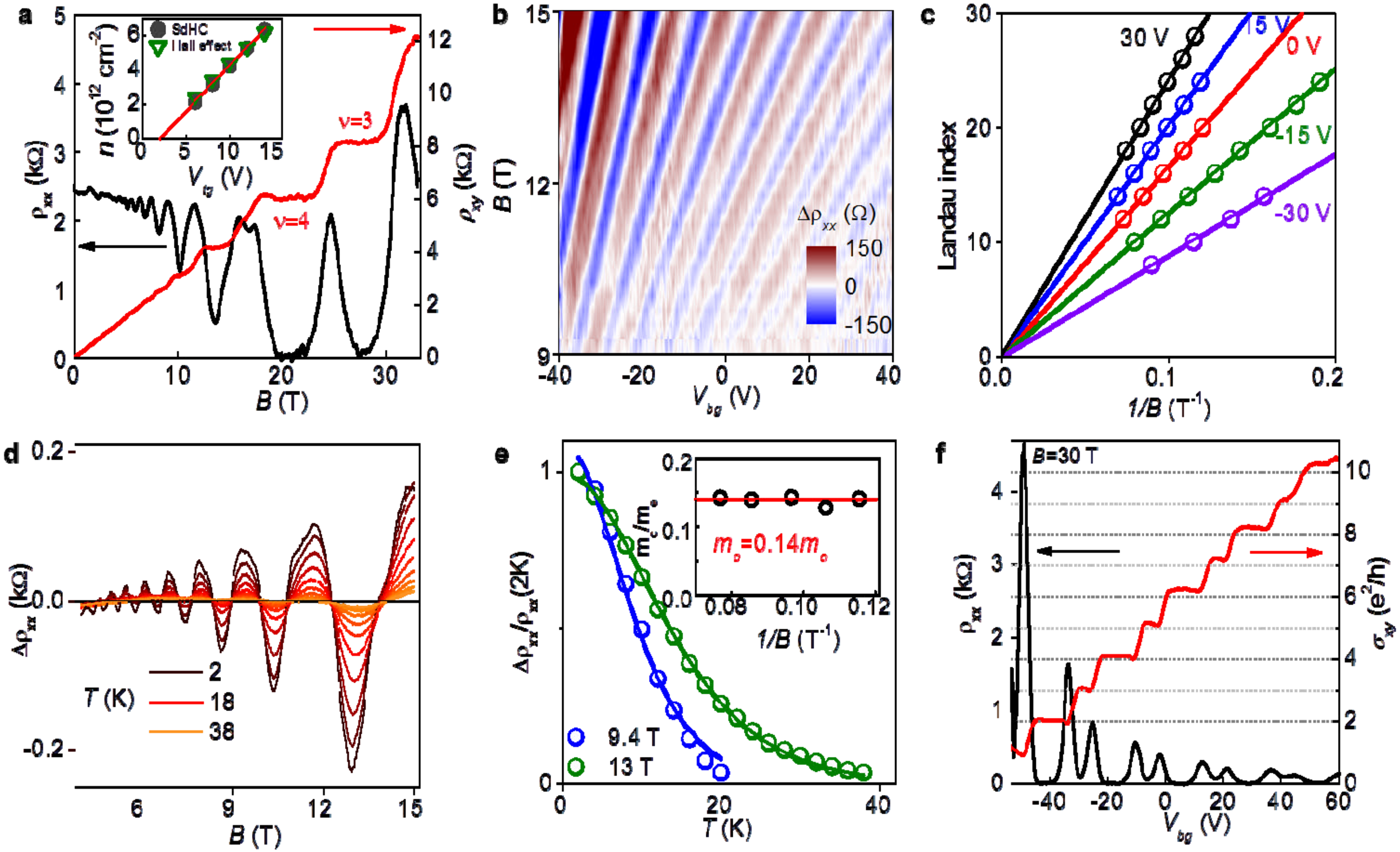


high mobility!



- hBN and 2DM are glued together by weak van der Waals attraction, hence, they preserve their lattice structures and, hence, retain their basic physical properties.

hBN-encapsulated 2D crystal: QHE in 6L-InSe

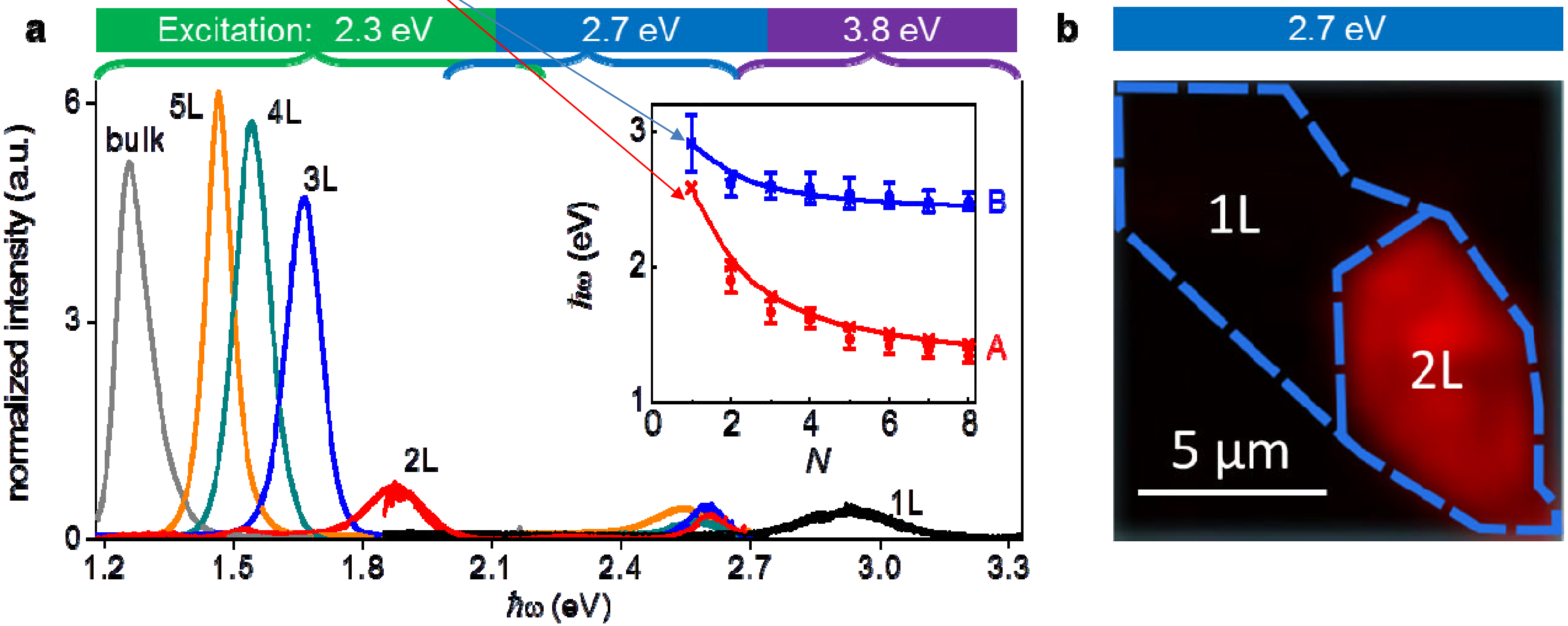


$m_c(N=3)=0.17m_e$
$m_c(N=6)=0.14m_e$

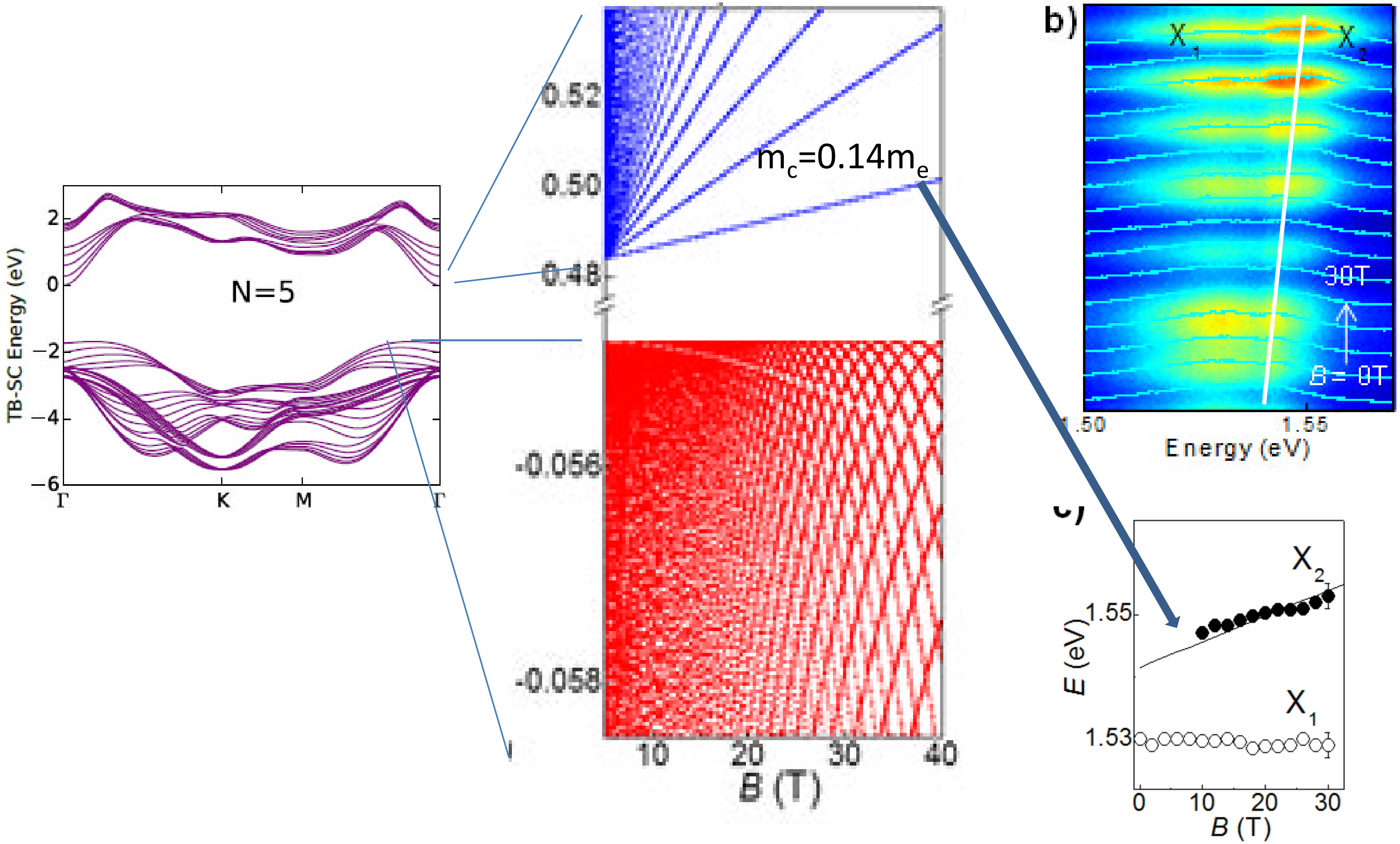
nature nanotechnology
LETTERS
PUBLISHED ONLINE: XX XX 2016 | DOI: 10.1038/NNANO.2016.242

Bandurin, Tyurnina, Yu, Mishchenko, Zólyomi, Morozov, Krishna Kumar, Gorbachev, Kudrynskyi
Pezzini, Kovalyuk, Zeitler, Novoselov, Patanè, Eaves, Grigorieva, Fal'ko, Geim, Cao

DFT/TB with scissor correction



Magnetoluminescence (non-encapsulated 5L)

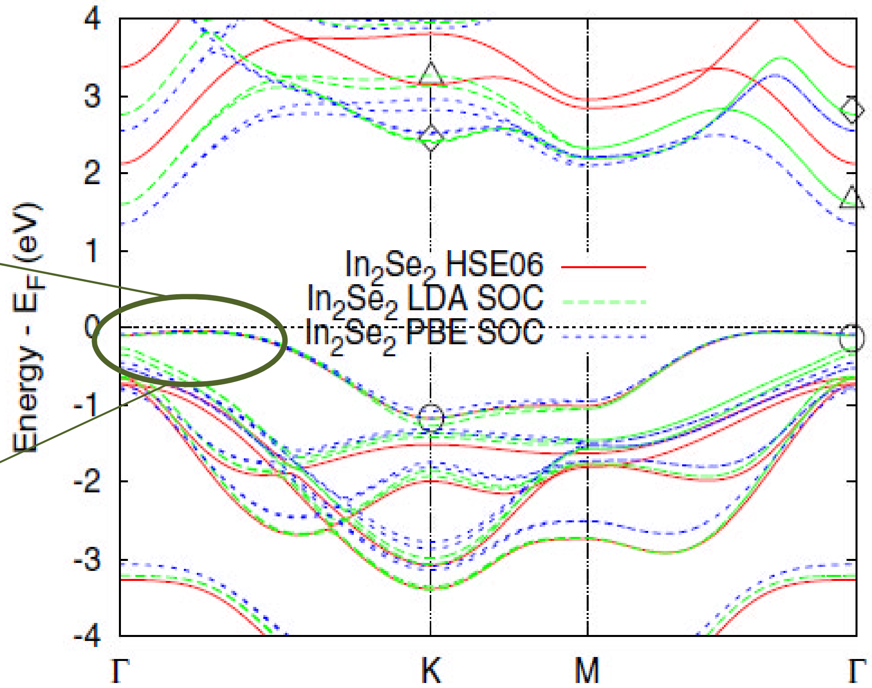


Mudd, Molas, Chen, Zolyomi, Nogajewski, Kudrynskyi, Kovalyuk, Yusa, Makarovsky, Eaves, Potemski, Fal'ko, Patane - Scientific Reports 6, 39619 (2016)

Two-dimensional InSe

- strong band gap variation with the number of layers
- light conduction band mass:
  potential for high mobility and quantum circuits
- potential for strongly correlated states
  in p-doped 2D InSe

P-doped: Wigner crystal?
Mott insulator? ferromagnetic state?
Peierls instability? superconductivity?



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