Effective tight binding Hamiltonian for monolayer MoS2

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Outline

A short Introduction to MoS2

Tight binding Hamiltonian

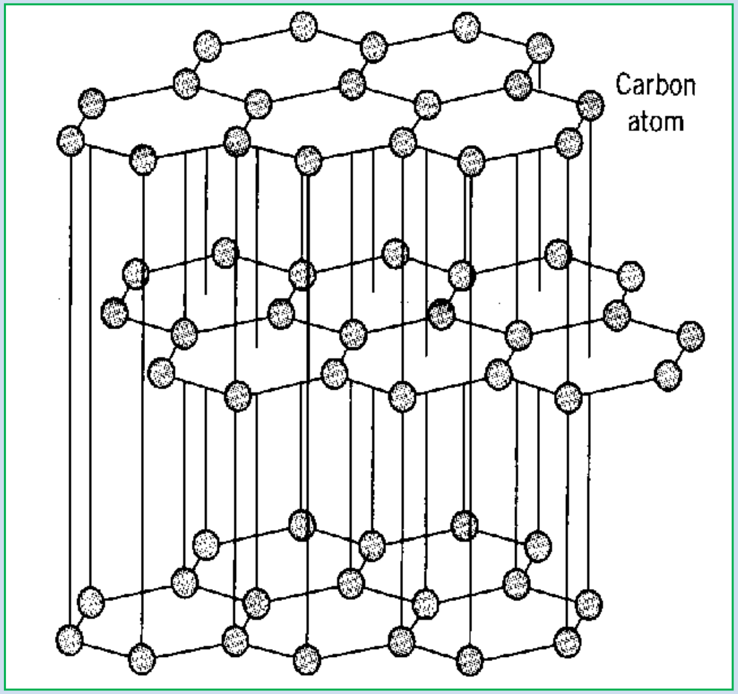
Two band Hamiltonian

Zeeman like for valley

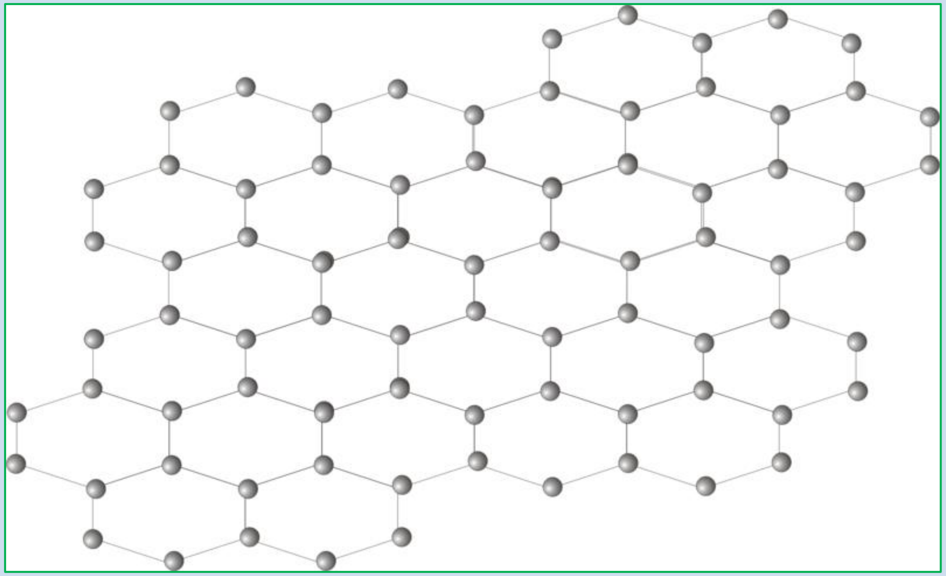
Conclusion

Layered Compounds: Graphite

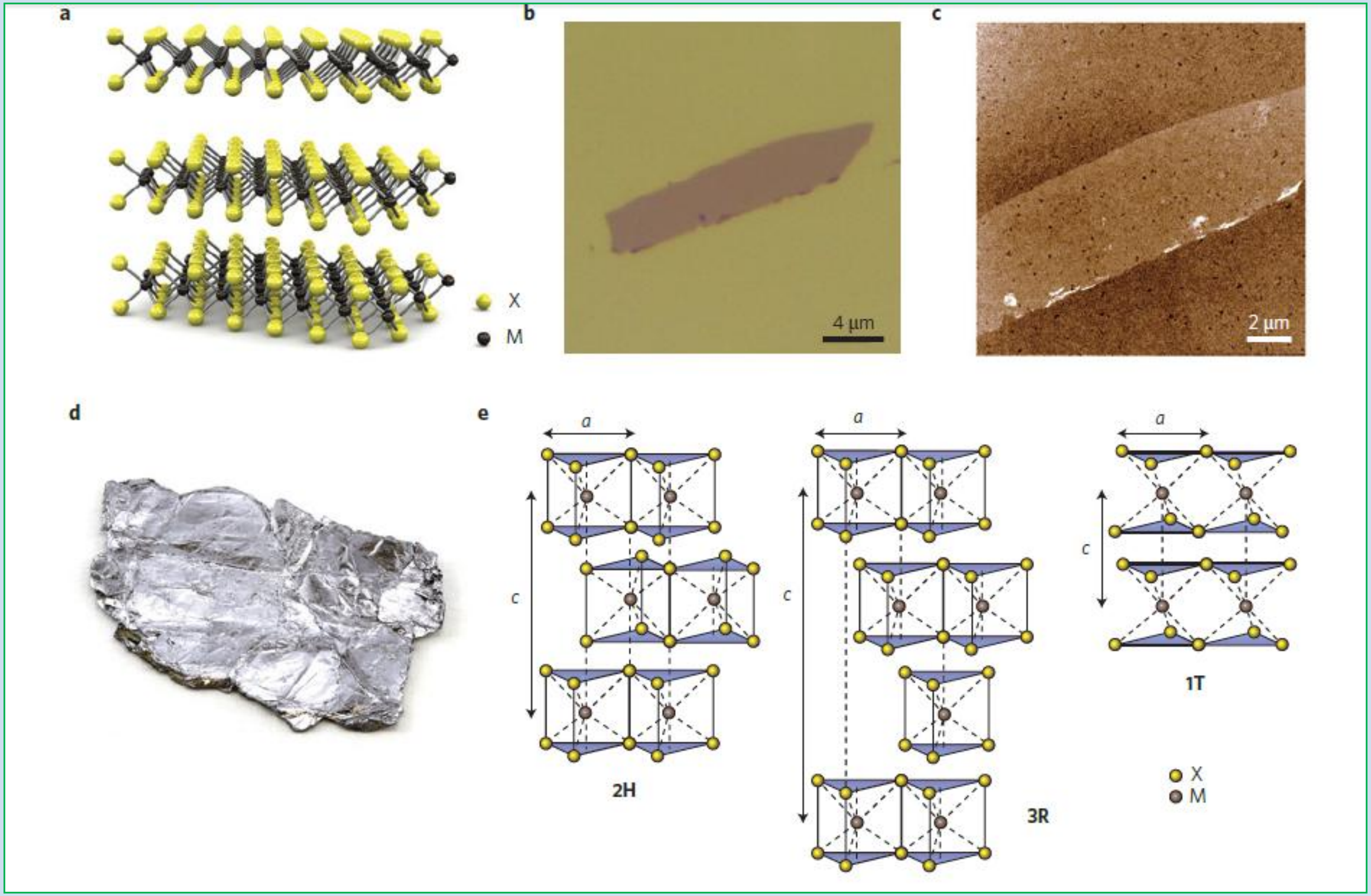
Graphite

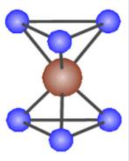


Graphene



Layered Compounds: Transition metal dichalcogenides (MX_2)





Wang et al, Nature Nanotech. 7, 699 (2012)

Layered Compounds: Transition metal dichalcogenides (MX_2)

MX_2
M = Transition metal
X = Chalcogen

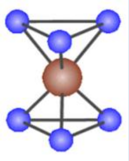
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg	3	4	5	6	7	8	9	10	11	12	Al	Si	P	**S**	Cl	Ar
K	Ca	Sc	**Ti**	**V**	Cr	Mn	Fe	**Co**	**Ni**	Cu	Zn	Ga	Ge	As	**Se**	Br	Kr
Rb	Sr	Y	**Zr**	**Nb**	**Mo**	**Tc**	Ru	**Rh**	**Pd**	Ag	Cd	In	Sn	Sb	**Te**	I	Xe
Cs	Ba	La - Lu	**Hf**	**Ta**	**W**	**Re**	Os	**Ir**	**Pt**	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac - Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fl	Uup	Lv	Uus	Uuo

HfS_2: *Insulator*

$TiSe_2, WTe_2$: *Semimetal*

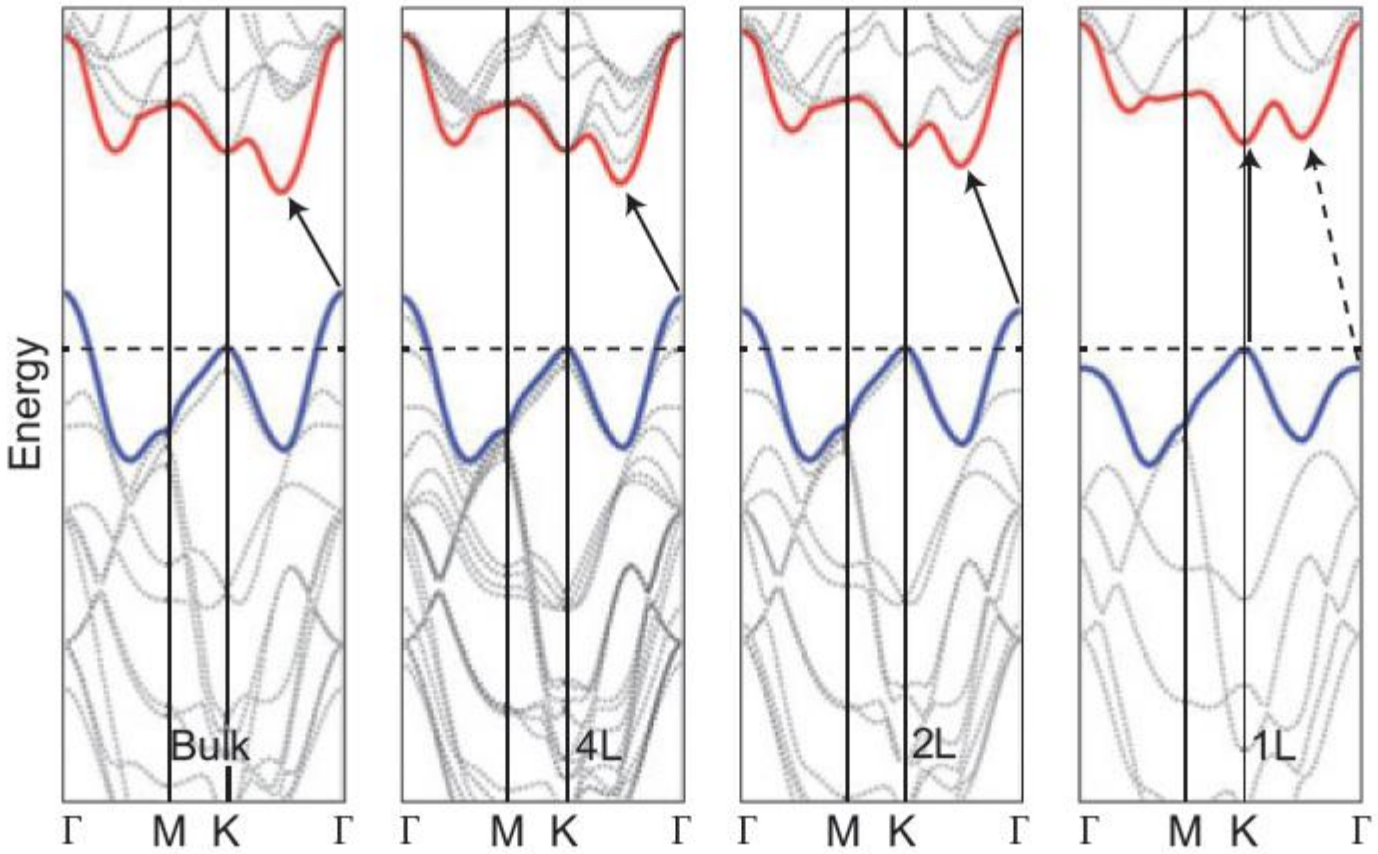
MoS_2, WS_2: *Semiconductor*

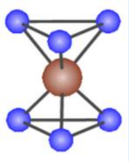
NbS_2, VSe_2: *Metal*



Chhowalla et al, Nature Chemistry 5, 263–275 (2013)

Band Structure: DFT





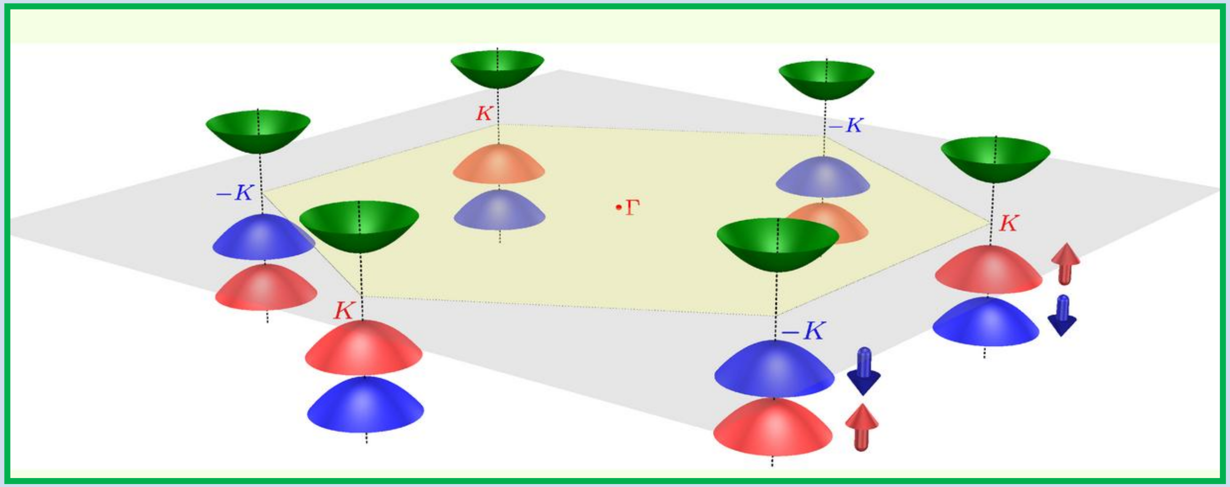
Nature Chemistry 5, 263–275 (2013)

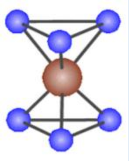
Xiao’s low-energy Hamiltonian

$\vec{K} \bullet \vec{P}$ model

$$|\phi_c\rangle = |d_{z^2}\rangle, \qquad |\phi_v^\tau\rangle = \frac{1}{\sqrt{2}}(|d_{x^2-y^2}\rangle + i\tau|d_{xy}\rangle),$$

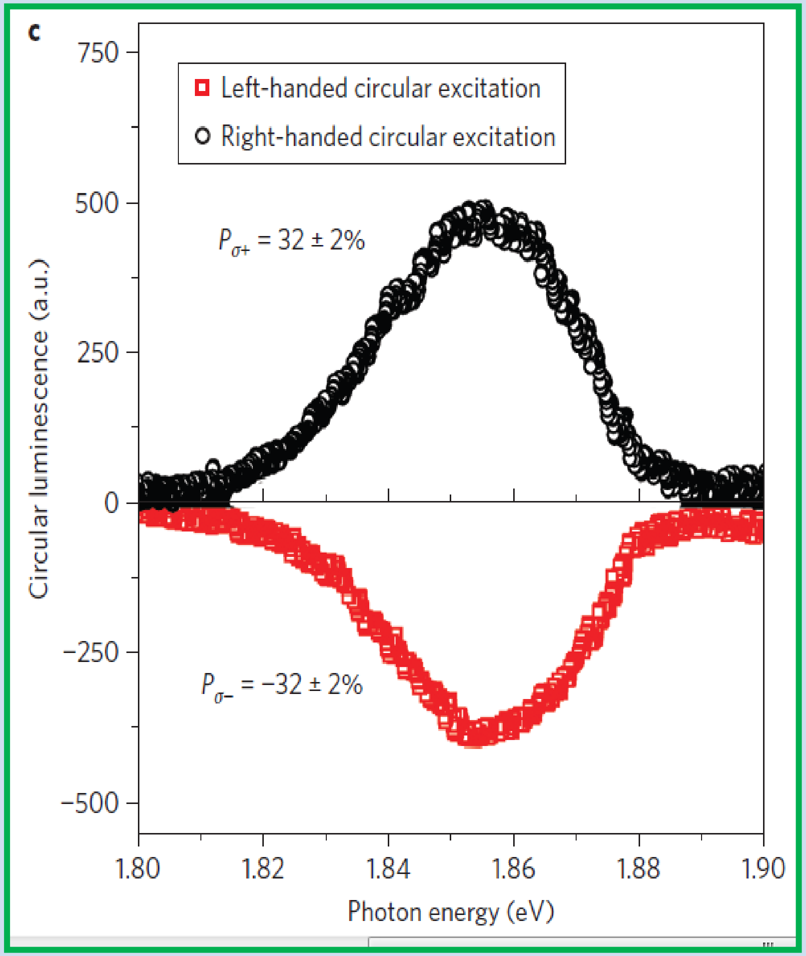
$$\hat{H} = at(\tau k_x \hat{\sigma}_x + k_y \hat{\sigma}_y) + \frac{\Delta}{2}\hat{\sigma}_z - \lambda\tau \frac{\hat{\sigma}_z - 1}{2}\hat{s}_z,$$





Di Xiao, *et al*, Phys. Rev. Lett. **108**,196802 (2012)

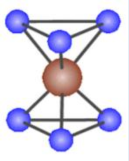
Circular dichroism



$$\mathcal{P}_\alpha(\boldsymbol{k}) \equiv m_0 \langle u_c(\boldsymbol{k})| \frac{1}{\hbar}\frac{\partial \hat{H}}{\partial k_\alpha} |u_v(\boldsymbol{k})\rangle$$

$$|\mathcal{P}_\pm(\boldsymbol{k})|^2 = \frac{m_0^2 a^2 t^2}{\hbar^2}\left(1 \pm \tau \frac{\Delta'}{\sqrt{\Delta'^2 + 4a^2t^2k^2}}\right)^2$$

$$P = \frac{|\mathcal{P}_+|^2 - |\mathcal{P}_-|^2}{|\mathcal{P}_+|^2 + |\mathcal{P}_-|^2}$$



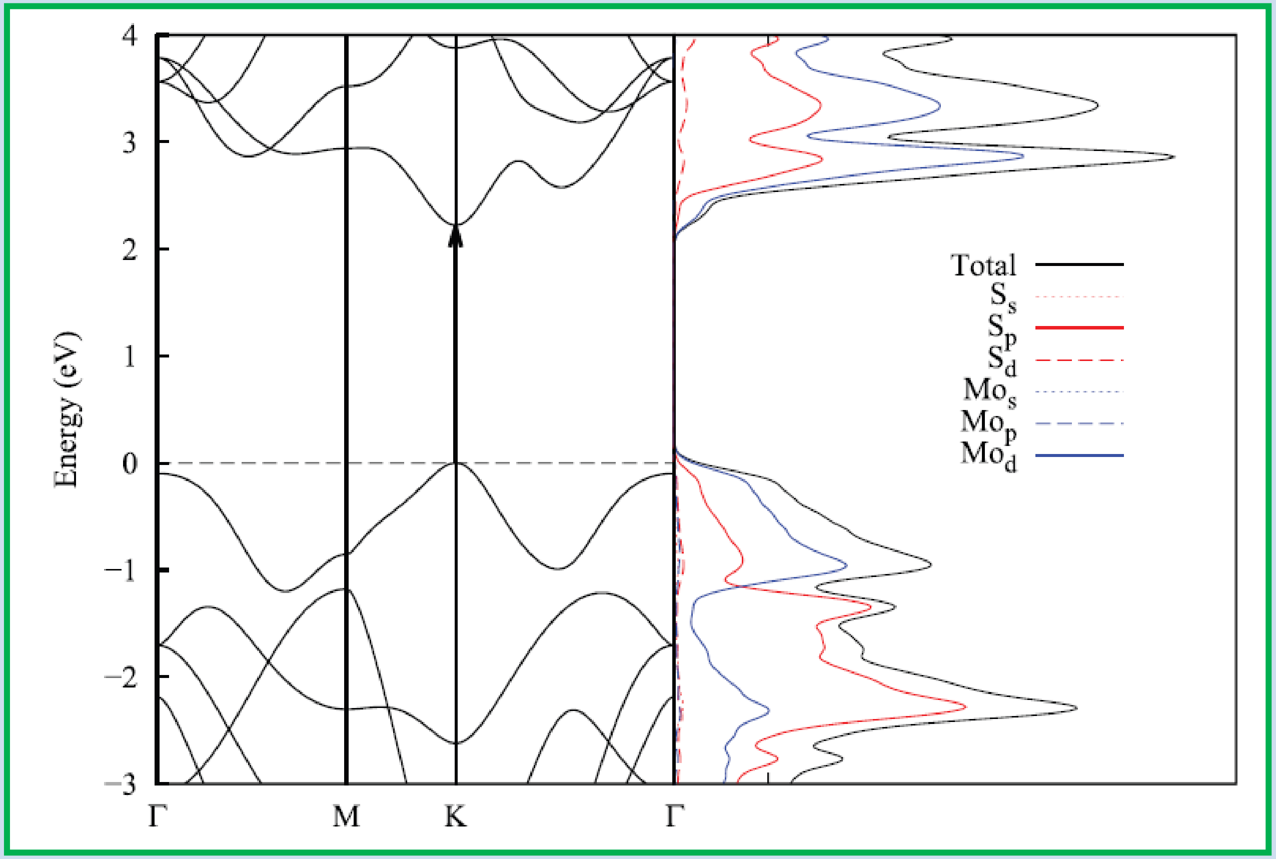
Zeng et al, Nature NanoTech. 7, 409 (2012)

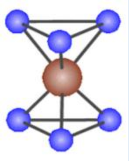
Di Xiao, *et al* , Phys. Rev. Lett. **108**,196802 (2012)

Relevant atomic orbitals

$Mo: [Kr]5s^1 4d^5$

$S: [Ne]\ 3s^2 3p^4$





Applied Physics Letters,99,261908(2011)

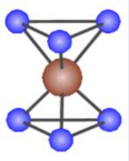
Tight binding Hamiltonian

Symmetry adopted basis orbitals

$$|1\rangle = |d_{z^2}\rangle \qquad |2\rangle = \frac{1}{\sqrt{2}}(|d_{x^2+y^2}\rangle + i|d_{xy}\rangle) \qquad |3\rangle = \frac{1}{\sqrt{2}}(|d_{x^2+y^2}\rangle - i|d_{xy}\rangle)$$

$$|d_{xz}\rangle \qquad |p_z\rangle \qquad |d_{yz}\rangle$$

$$|1'\rangle = \frac{1}{\sqrt{2}}(|p_x\rangle + i|p_y\rangle) \qquad |2'\rangle = \frac{1}{\sqrt{2}}(|p_x\rangle - i|p_y\rangle)$$



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~~$|d_{xz}\rangle$~~ ~~$|p_z\rangle$~~ ~~$|d_{yz}\rangle$~~

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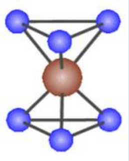


Tight binding Hamiltonian

$$\hat{H}_{TB} = \sum_{i\mu\nu}\{\epsilon^{a}_{\mu\nu}a^{\dagger}_{i\mu}a_{i\nu} + \epsilon^{b}_{\mu\nu}b^{\dagger}_{i\mu}b_{i\nu} + \epsilon^{b'}_{\mu\nu}b'^{\dagger}_{i\mu}b'_{i\nu}\} + \sum_{\langle ij\rangle,\mu\nu} t_{ij,\mu\nu}a^{\dagger}_{i\mu}(b_{i\nu}+b'_{i\nu}) + H.c.,$$

$$\hat{S} = \sum_{i\mu\nu}\sum_{\langle ij\rangle,\mu\nu} s_{ij,\mu\nu}a^{\dagger}_{i\mu}(b_{i\nu}+b'_{i\nu}) + H.c.,$$

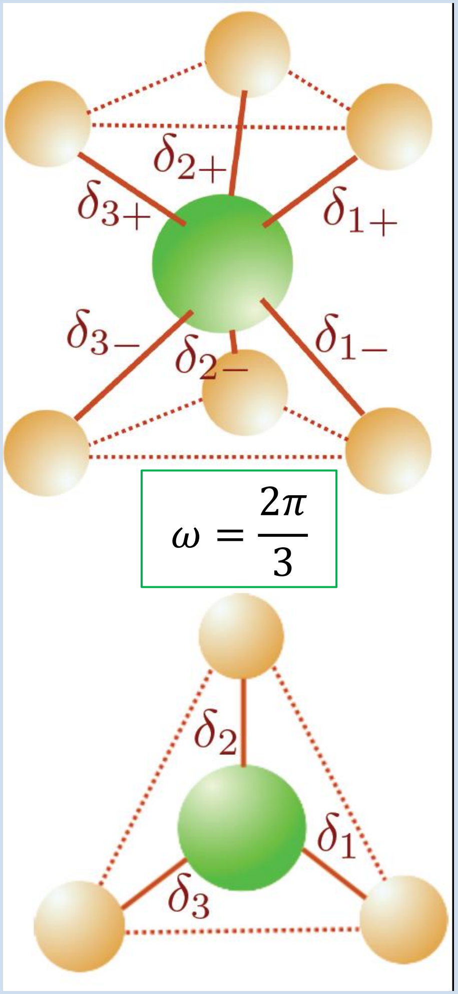
$a:$ Mo
$b, b':$ S

$$\begin{aligned}
\epsilon^{a}_{\mu\nu} &= \langle a,i,\mu|H|a,i,\nu\rangle \\
\epsilon^{b}_{\mu\nu} &= \langle b,i,\mu|H|b,i,\nu\rangle \\
\epsilon^{b'}_{\mu\nu} &= \langle b',i,\mu|H|b',i,\nu\rangle \\
t_{ij,\mu\nu} &= \langle a,i,\mu|H|b^{(')},j,\nu\rangle \\
s_{ij,\mu\nu} &= \langle a,i,\mu|b(b'),j,\nu\rangle
\end{aligned}$$

$$R_3 = e^{-i\frac{\omega L_z}{\hbar}}$$

$$\sigma_v : x \to -x, \quad y,z \to y,z$$

$$\sigma_h : z \to -z, \quad y,x \to y,x$$



$$\omega = \frac{2\pi}{3}$$

Rostami, Moghaddam, Asgari arXiv:1302.5901

Imposing symmetry constraints

Symmetries + SO coupling:

$$H = R_3^\dagger H R_3 \quad , \quad H = \sigma_v^{-1} H \sigma_v \quad , \quad H = \sigma_h^{-1} H \sigma_h$$

$$\langle 2|S.L|2\rangle \propto s \, , \langle 3|S.L|3\rangle \propto -s$$

Results:

$$\epsilon^a = \begin{pmatrix} A_1 & 0 & 0 \\ 0 & A_2 + \lambda s & 0 \\ 0 & 0 & A_2 - \lambda s \end{pmatrix} , \qquad \epsilon^b = \epsilon^{b'} = \begin{pmatrix} B & 0 \\ 0 & B \end{pmatrix} ,$$

$$t_{\delta_{1\pm}} = \begin{pmatrix} t_{11} & -e^{-i\omega} t_{11} \\ t_{21} & t_{22} \\ -t_{22} & -e^{i\omega} t_{21} \end{pmatrix} , \quad t_{\delta_{2\pm}} = \begin{pmatrix} e^{i\omega} t_{11} & -e^{i\omega} t_{11} \\ e^{-i\omega} t_{21} & t_{22} \\ -t_{22} & -e^{-i\omega} t_{21} \end{pmatrix} , \quad t_{\delta_{3\pm}} = \begin{pmatrix} e^{-i\omega} t_{11} & -t_{11} \\ e^{i\omega} t_{21} & t_{22} \\ -t_{22} & -t_{21} \end{pmatrix}$$

Rostami, Moghaddam, Asgari arXiv:1302.5901

k-space Hamiltonian

$$\mathcal{H}|\psi\rangle = E\mathcal{S}|\psi\rangle$$

$$\mathcal{H} = \begin{pmatrix} \hat{H}_a & \hat{H}_t & \hat{H}_t \\ \hat{H}_t^\dagger & \hat{H}_b & 0 \\ \hat{H}_t^\dagger & 0 & \hat{H}_{b'} \end{pmatrix}$$

$$\mathcal{S} = \begin{pmatrix} 1 & \hat{S}_t & \hat{S}_t \\ \hat{S}_t^\dagger & 1 & 0 \\ \hat{S}_t^\dagger & 0 & 1 \end{pmatrix}$$

$$\hat{H}_t = \begin{pmatrix} t_{11}f(\mathbf{k},\omega) & -e^{-i\omega}t_{11}f(\mathbf{k},-\omega) \\ t_{21}f(\mathbf{k},-\omega) & t_{22}f(\mathbf{k},0) \\ -t_{22}f(\mathbf{k},0) & -e^{i\omega}t_{21}f(\mathbf{k},\omega) \end{pmatrix}$$

$$\hat{H}_t \quad \xrightarrow{t_{\mu\nu} \to s_{\mu\nu}} \quad \hat{S}_t$$

$$\hat{H}_a = \epsilon^a + U^a \quad , \quad \hat{H}_{b^{(\prime)}} = \epsilon^{b^{(\prime)}} + U^{b^{(\prime)}}$$

Rostami, Moghaddam, Asgari arXiv:1302.5901

Empirical Parameters

$$\Delta = 1.9eV$$

$$\lambda = 80meV$$

Radisavljevic *et al*, nat. nanotech, **6**, 147 (2011)

Mak, *et al*, Phys. Rev. Lett. **105**, 136805 (2010)

$$m_e = 0.37\,m_0$$

$$m_h = -0.44\,m_0$$

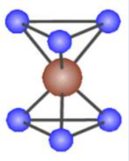
$$E_{VBM} = -5.73\,eV$$

Peeraers, Van de Walle, Phys. Rev. B **86**, 241401 (2012)

Yunguo Li, *et al*, arXiv: 1211.4052

$$s_{\mu\nu} = 0.1\,t_{\mu\nu}/eV$$

10% p-orbital mixing



Model's Parameters

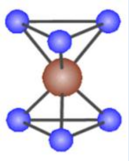
In the presence of σ_h symmetry, 7-band Hamiltonian will reduce to a 5-band one as:

$$\mathcal{H} = \begin{pmatrix} \hat{H}_a & 2\hat{H}_t \\ 2\hat{H}_t^\dagger & 2\hat{H}_b \end{pmatrix}, \qquad \mathcal{S} = \begin{pmatrix} 1 & 2\hat{S} \\ 2\hat{S}^\dagger & 2 \end{pmatrix}$$

By fitting the eigenvalue of above Hamiltonian with low energy Dispersion, one can find:

$$A_1 = -1.45 eV \qquad A_2 = -5.8 eV \qquad B = 5.53 eV$$

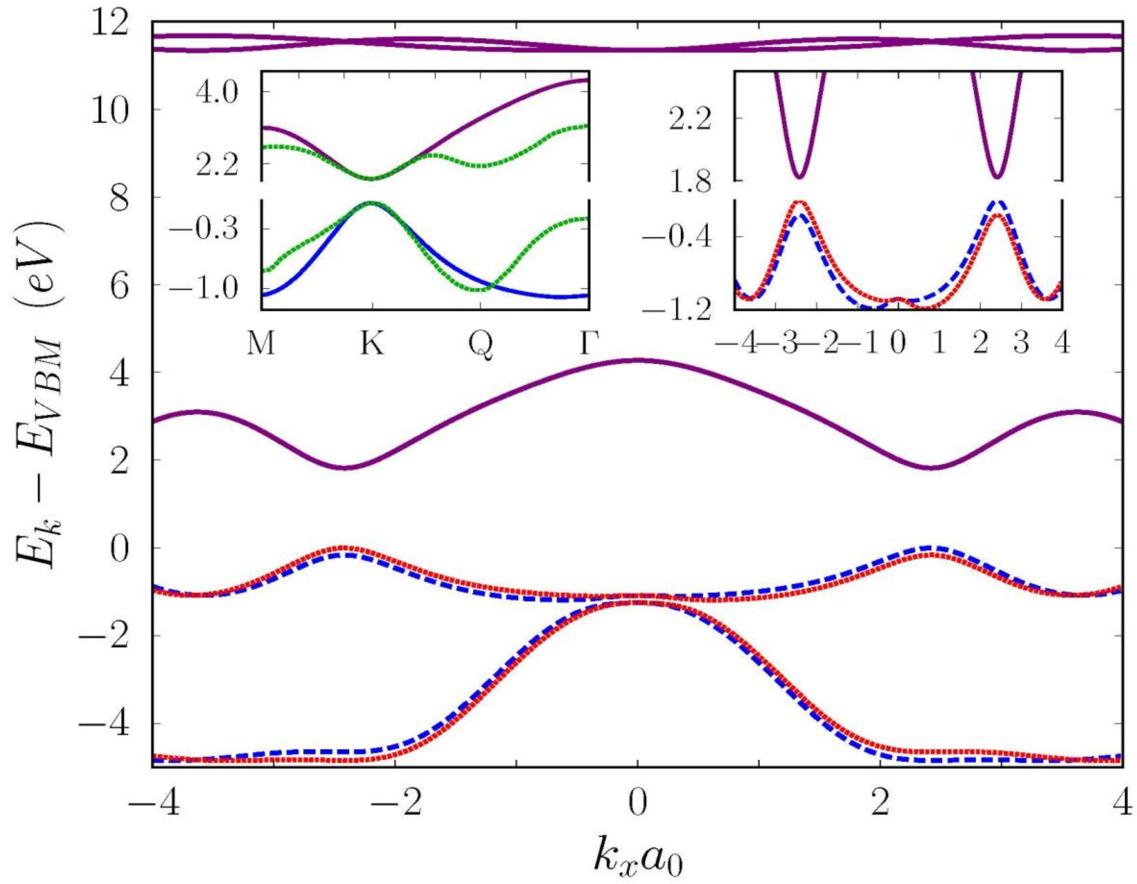
$$e^{i\pi/6} t_{11} = 0.82\, eV \qquad e^{-i\pi/6} t_{21} = -1 eV \qquad e^{-i\pi/2} t_{22} = 0.51\, eV$$



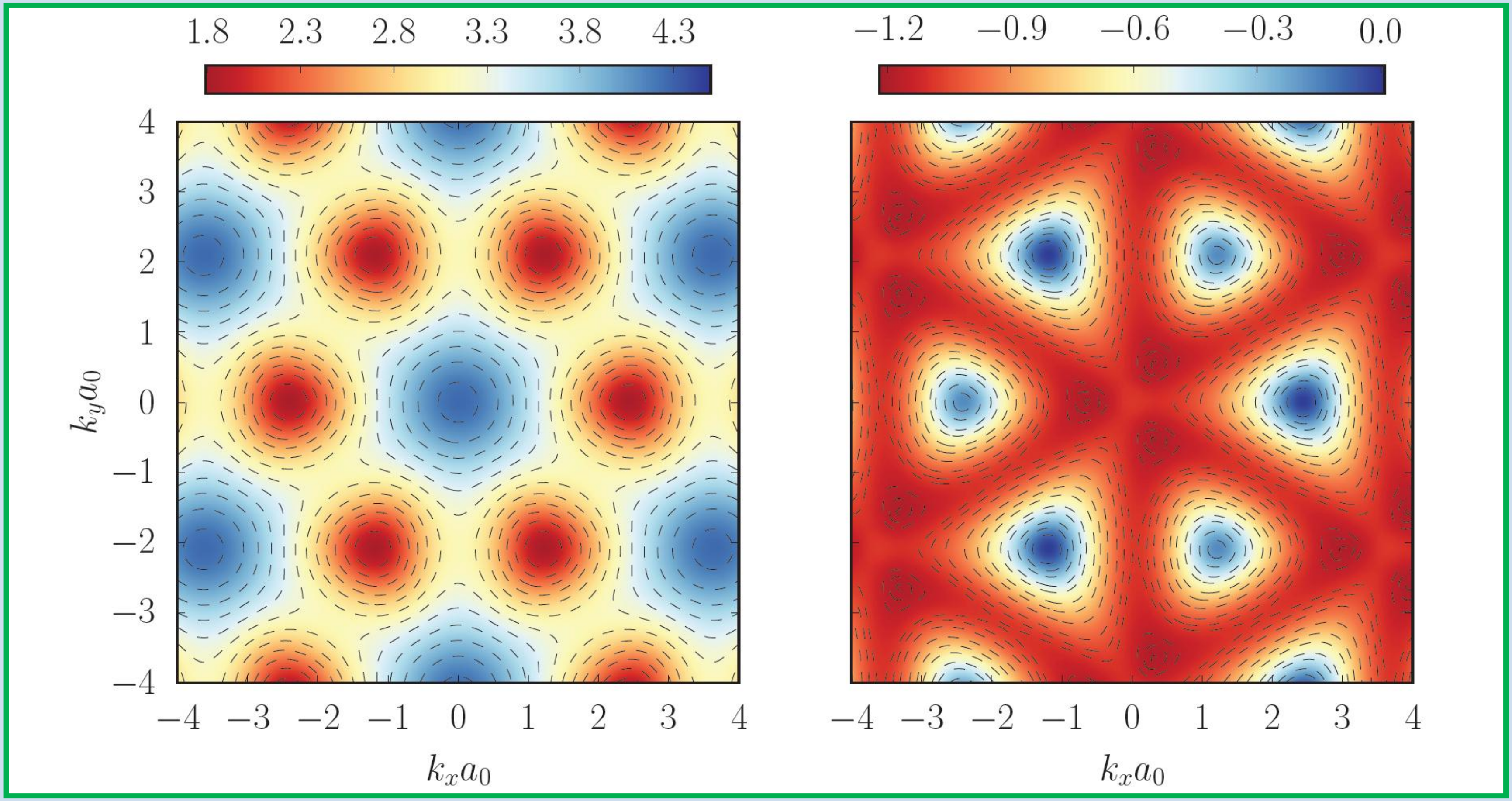
Energy dispersion relation

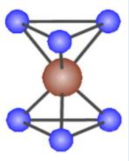
$$\mathcal{H} = \begin{pmatrix} \hat{H}_a & 2\hat{H}_t \\ 2\hat{H}_t^\dagger & 2\hat{H}_b \end{pmatrix}$$

$$\mathcal{S} = \begin{pmatrix} 1 & 2\hat{S} \\ 2\hat{S}^\dagger & 2 \end{pmatrix}$$



Trigonal Warping(beyond Low energy)





Two band Hamiltonian

Low-energy Hamiltonian

0-Expanding the Hamiltonian around the K-point(k=K+q)

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$$

1-Löwdin orthogonalization ($S^{-1/2}$)

$$\tilde{H} = \mathcal{S}^{-1/2}\mathcal{H}\mathcal{S}^{-1/2} \quad , \quad |\tilde{\psi}\rangle = \mathcal{S}^{1/2}|\psi\rangle$$

$$\mathcal{H}|\psi\rangle = E\mathcal{S}|\psi\rangle \quad \rightarrow \quad \tilde{H}|\tilde{\psi}\rangle = E|\tilde{\psi}\rangle$$

2-From orbital-space to band-space(U_0)

$$H' = U_0^\dagger \tilde{H} U_0 = H'_{\text{diag}} + V \quad \text{where} \quad U_0^\dagger \mathcal{S}_0^{-1/2}\mathcal{H}_0\mathcal{S}_0^{-1/2}U_0 \equiv \text{diagonal}$$

3-Löwdin partitioning($e^{-\mathcal{O}}$)

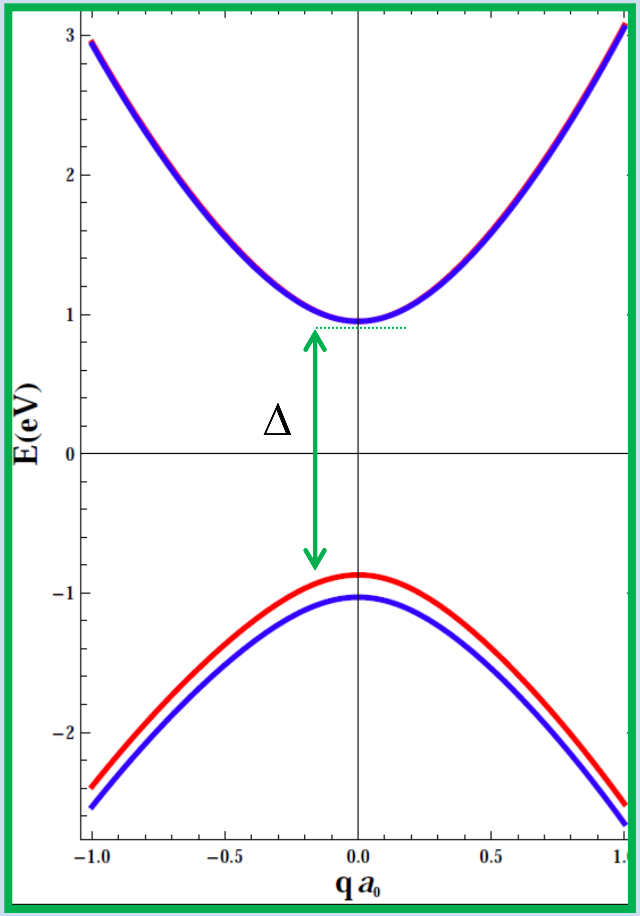
$$H = e^{-\mathcal{O}}H'e^{\mathcal{O}} = H'_{\text{diag}} + V + [H'_{\text{diag}}, \mathcal{O}] + [V, \mathcal{O}] + \frac{1}{2}[[H'_{\text{diag}}, \mathcal{O}], \mathcal{O}] + \cdots$$

$$V + [H'_{\text{diag}}, \mathcal{O}] = 0 \quad \text{restlts:} \quad \mathcal{O}$$

$$H = H'_{\text{diag}} + \frac{1}{2}[V, \mathcal{O}] + \cdots$$

Low-energy Hamiltonian

$$
\begin{aligned}
H_{\tau s} &= \frac{\Delta}{2}\sigma_z + \lambda\tau s\frac{1-\sigma_z}{2} + t_0 a_0 \mathbf{q}\cdot\sigma_\tau \\
&+ \frac{\hbar^2|\mathbf{q}|^2}{4m_0}(\alpha+\beta\sigma_z) \\
&+ t_1 a_0^2 \mathbf{q}\cdot\sigma_\tau^* \sigma_x \mathbf{q}\cdot\sigma_\tau^*,
\end{aligned}
$$

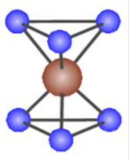


$$
\begin{aligned}
t_0 &= 1.68 eV \\
t_1 &= 0.1 eV \\
\alpha &= 0.43 \\
\beta &= 2.21
\end{aligned}
$$

$$\beta = \frac{m_0}{m_-} - \frac{4\, m_0 a_0^2 t_0^2}{(\Delta-\lambda)\hbar^2}$$

$$\alpha = \frac{m_0}{m_+}$$

$$m_\pm = \frac{m_e m_h}{m_h - m_e}$$



Zeeman like for Valley

Landau Level

$$H_{\tau s} = \frac{\Delta}{2}\sigma_z + \lambda\tau s\frac{1-\sigma_z}{2} + t_0 a_0 \vec{\pi}\cdot\vec{\sigma}_\tau + \frac{\hbar^2|\vec{\pi}|^2}{4m_0}(\alpha + \beta\sigma_z)$$

$$\vec{\pi} = \vec{p} + e\vec{A}$$

Valley degeneracy is broken

$$E^{\pm}_{n,\tau s} = \pm\sqrt{[\frac{\Delta - \lambda\tau s}{2} + \hbar\omega_c(\beta n - \frac{\alpha\tau}{2})]^2 + 2(\frac{t_0 a_0}{l_B})^2 n} + \frac{\lambda\tau s}{2} + \hbar\omega_c(\alpha n - \frac{\beta\tau}{2})$$

$$\omega_c = eB/2m_0 \quad , \quad l_B = \sqrt{\hbar/(eB)}$$

Valley polarization

$$E^{+}_{0,\tau s} = [\Delta - \hbar\omega_c \tau(\beta + \alpha)]/2 \quad , \quad E^{-}_{0,\tau s} = \lambda\tau s - [\Delta - \hbar\omega_c \tau(\beta - \alpha)]/2$$

Zeeman-like for valleys

$$\text{Landau level spacing:} \quad \delta E^{+} \approx 5.4\hbar\omega_c \quad , \quad \delta E^{-} \approx 4.6\hbar\omega_c$$

$$\text{Valley spliting in the conduction band:} \quad \delta E^{+}_{v} \approx g^{+}_{v}\hbar\omega_c$$

$$\text{Spin-Valley spliting in the valence band:} \quad \delta E^{-}_{s-v} \approx (g_s + g^{-}_{v})\hbar\omega_c$$

$$\text{Spin spliting(Zeeman effect):} \quad \delta E_s \approx g_s\hbar\omega_c$$

$$g_s = 2 \; , \quad g^{\pm}_{v} = \beta \pm \alpha$$

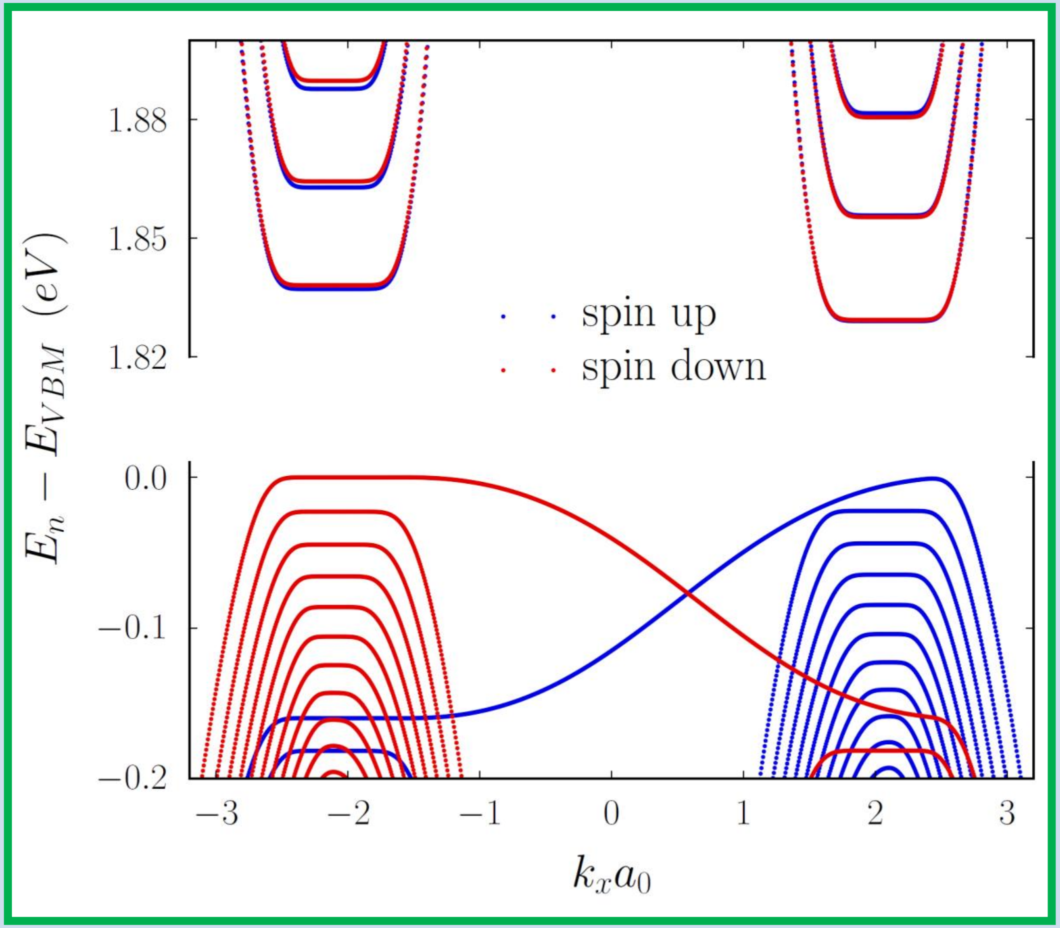
Valley polarization(Numerical)

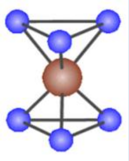
$$\hat{H}_{TB} = \sum_{i\mu\nu}\{\epsilon^{a}_{\mu\nu} a^{\dagger}_{i\mu} a_{i\nu} + \epsilon^{b}_{\mu\nu} b^{\dagger}_{i\mu} b_{i\nu} + \epsilon^{b'}_{\mu\nu} b'^{\dagger}_{i\mu} b'_{i\nu}\} + \sum_{\langle ij\rangle,\mu\nu} e^{i\phi_{ij}} t_{ij,\mu\nu} a^{\dagger}_{i\mu}(b_{i\nu} + b'_{i\nu}) + H.c.,$$

Note:
Zeeman effect is not added

Note:
Due to the absence of the symmetries at the edges and the contribution of other d-orbitals ,which are not Included in our model, the edgestates are not reliable.

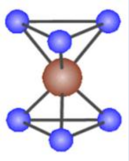
Note:
zigzag ribbon,
$W = 149a_0$,
$l_B = W/10$





Conclusion

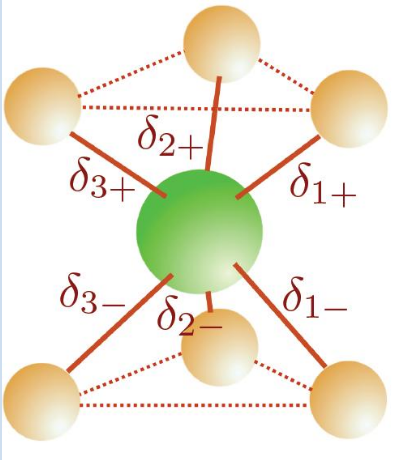
1. *Tight binding Hamiltonian* **based on low energy band structure**
2. *Two-band Hamiltonian* **in k- space**
3. **Quasi-particles are not Dirac Fermions**
4. **Valley degeneracy breaking in the presence of B (*Zeeman like for valley*)**
5. **The effect of *gate voltage* on the band structure**



Thank you!

Slater-Koster method

$$V_{ll'|m|} = < l', m, R' | H | l, m, R >$$



$$m = 0 \rightarrow \sigma$$

$$m = 1 \rightarrow \pi$$

$$m = 2 \rightarrow \delta$$

$$
\begin{aligned}
t_{11} &= -\frac{1}{\sqrt{2}}(n_x + i n_y)[\sqrt{3} n_z^2 V_{pd\pi} + \frac{1}{2}(1 - 3n_z^2) V_{pd\sigma}], \\
t_{21} &= \frac{1}{2}(n_x - i n_y)[\frac{\sqrt{3}}{2} V_{pd\sigma}(1 - n_z^2) + V_{pd\pi}(1 + n_z^2)], \\
t_{22} &= \frac{1}{2}(n_x - i n_y)^3(\frac{\sqrt{3}}{2} V_{pd\sigma} - V_{pd\pi}),
\end{aligned}
$$

$$\hat{n} = (n_x, n_y, n_z) = \frac{\vec{\delta}_1}{|\vec{\delta}_1|}$$

Rostami, Moghaddam, Asgari arXiv:1302.5901