Polariton Anomalous Hall Effect in TMDs

Luis Martin-Moreno

In collaboration with
F. Guinea, L.Chirolli,
A. Gutierrez-Rubio (IMDEA, Madrid),
F. J. Garcia-Vidal (IFIMAC, Madrid)

Chiral modes in optics and electronics of 2D systems
Aussois, France, 27/11/2018

icma / Instituto de Ciencia de Materiales de Aragón /
CSIC Consejo Superior de Investigaciones Científicas
Universidad Zaragoza 1542

Outline

Excitons Polaritons have a Berry Curvature which can originate from:

(i) Exciton BC
(ii) Photon BC
(iii) Coupling between them

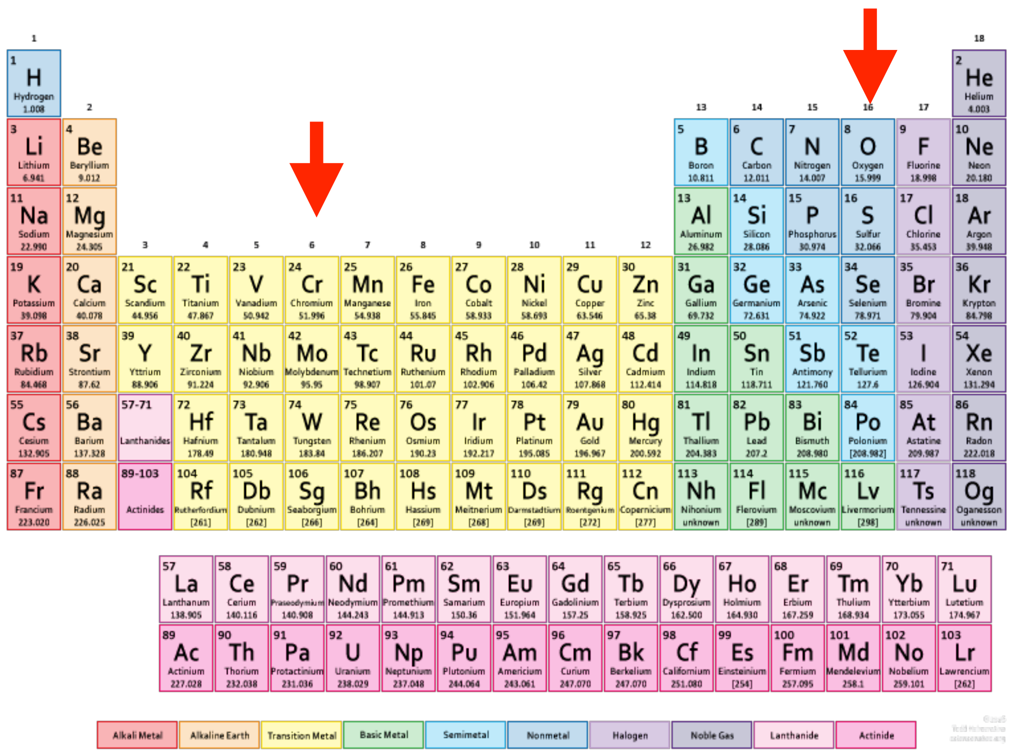
In this talk:

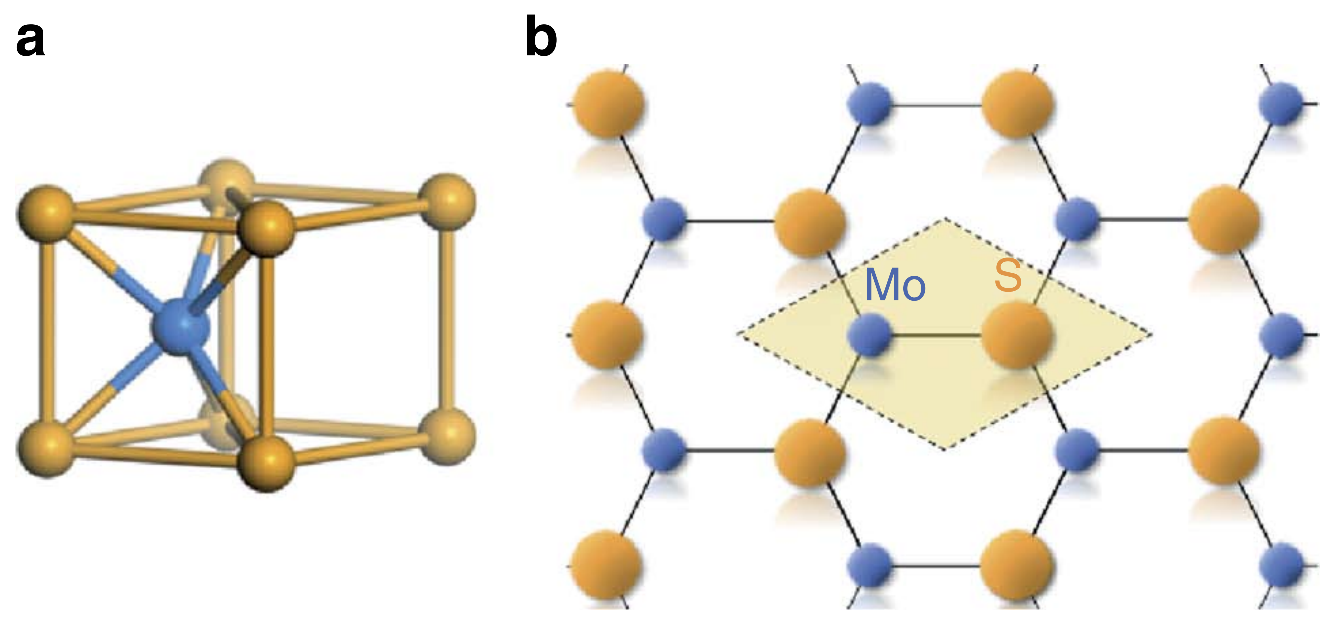
- Evaluation of the relative contributions
- Estimation of Lateral shifts upon Polariton motion

Model System:

Excitons in TMDs in a planar metallic cavity

Semiconductors: Transition Metal Dicalchogenides





<u>**Single Layer**</u>

- Direct bandgap 1.8 eV (in bulk, indirect gap of 1.3 eV)
- Strong light absorption and electroluminescence
- Strong exciton with large binding energy (stable at Room Temp.)

Excitons in 2D TMDs

While Excitons in 3D Materials (GaAs): ~ 100 nm, ~ 5 meV…

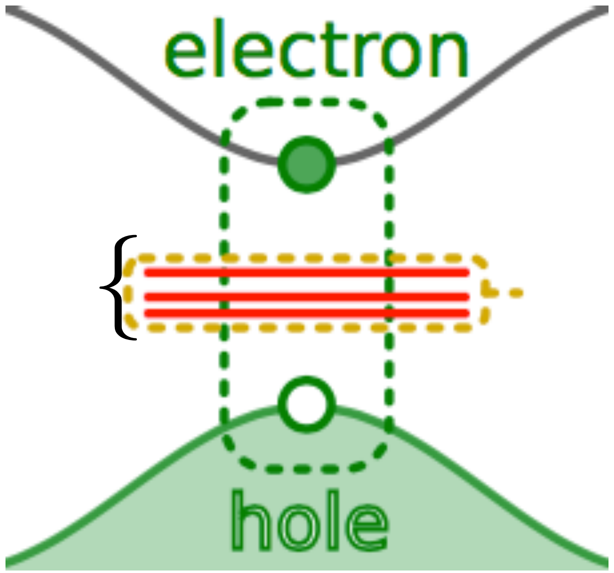
In TMDs: 2D character and weak dielectric screening:

enhanced Coulomb interactions

excitons dominate the **optical** and optoelectronic respond in TMDCs

Larger binding energy ~ 10 $k_B T_{room}$

Much smaller sizes ~ 1 nm



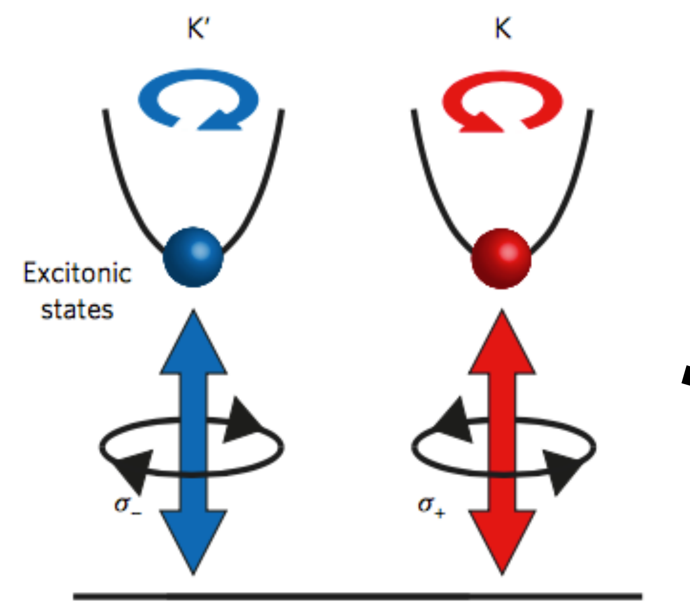
Effects of Diracness:
non-hydrogenic

Srivastava et al.,
PRL **115**, 166802 (2015)
J. Zhou et al.,
PRL **120**, 077401 (2018)

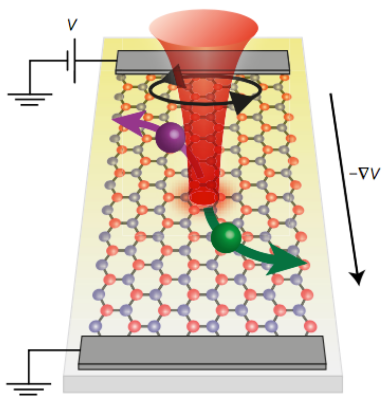
Splendiani et al., Nano Lett. **10**, 1271 (2010)
Berkelbach, PRB **88**, 045318 (2013)

Valley Hall and Exciton Anomalous Hall Effects in TMDCs

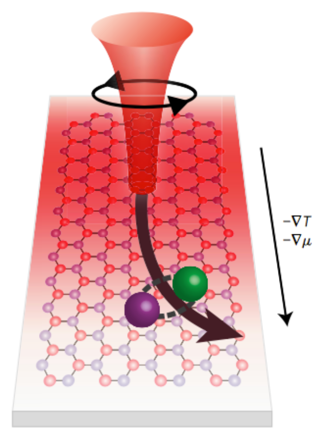
circular dichroism



Valley Hall effect



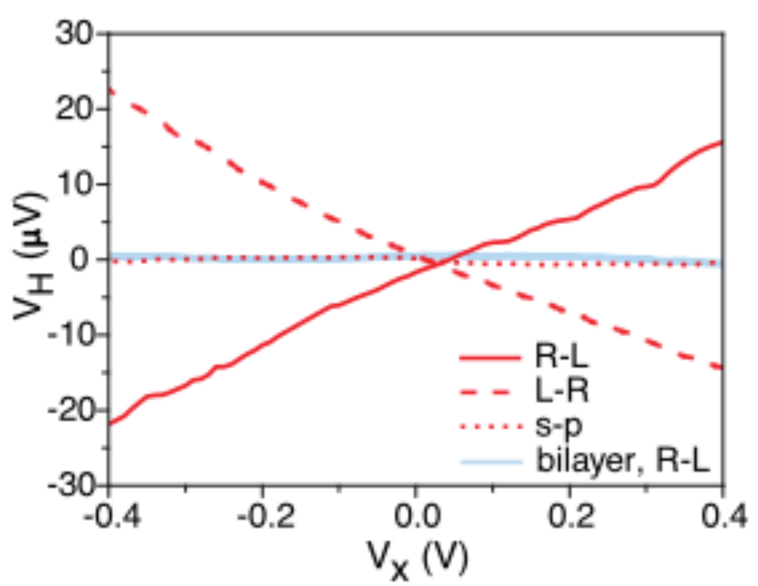
Exciton Hall effect

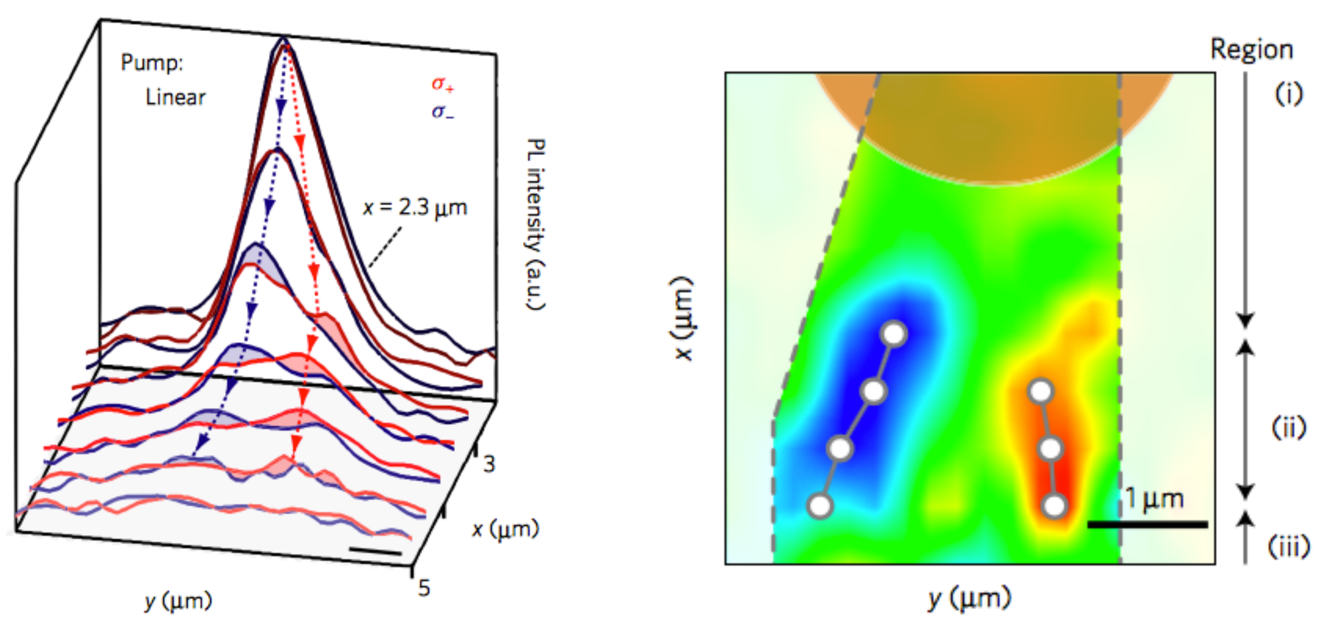


electrons and holes move in <u>opposite</u> directions

carrier's Berry curvature

electron-hole move in <u>same</u> directions

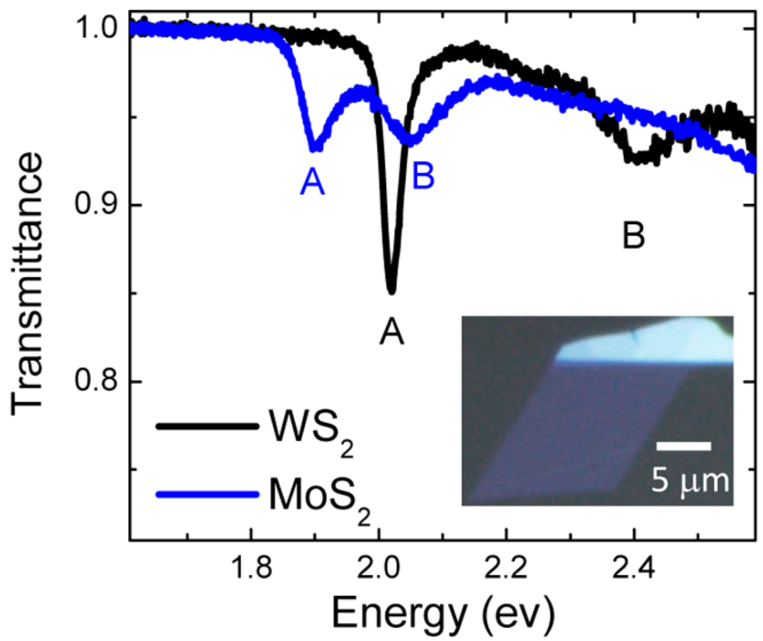




K. Mak et al. Science **344**, 1489 (2014)

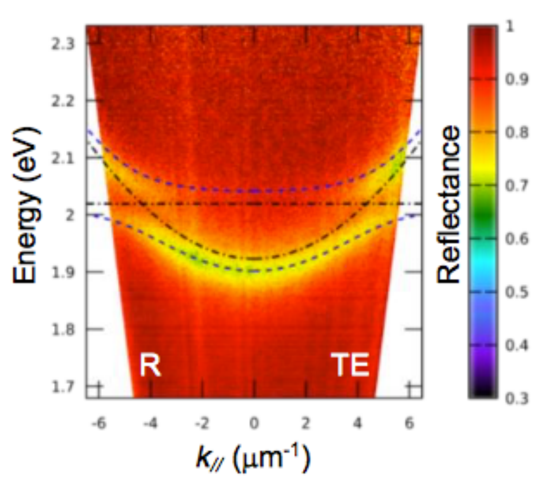
M. Onga et al. Nat. Materials **16**, 1193 (2017)

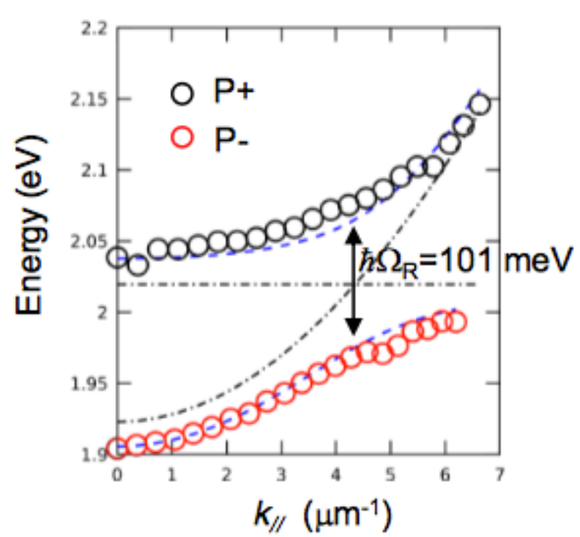
Monolayer in photonic cavity: Exciton-Polaritons

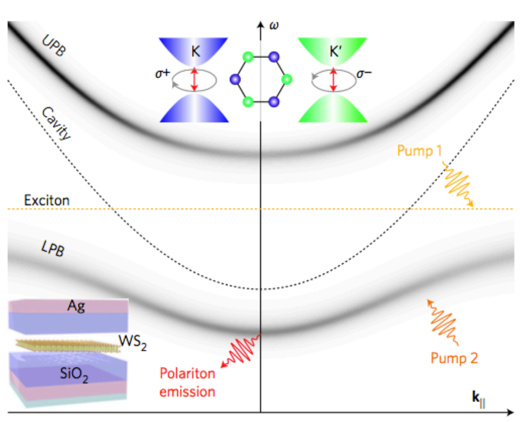


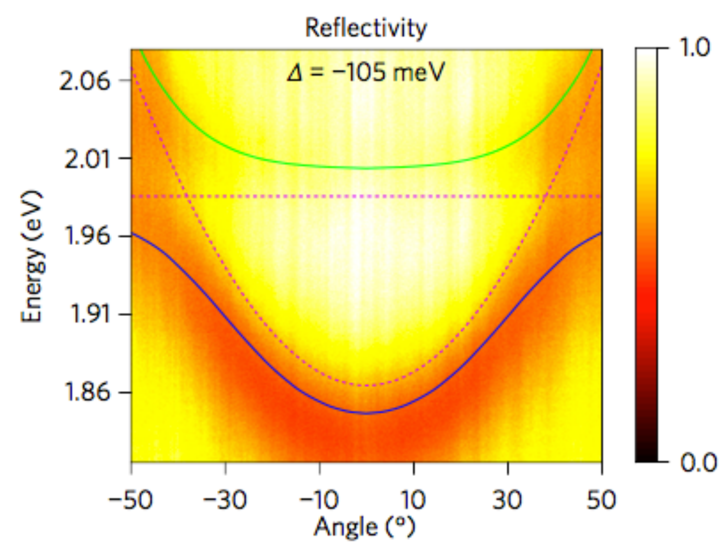
S. Wang et al. Nano Lett. **16 (7)**, 4368

Z. Sun et al. Nat. Photonics **11**, 491 (2017)





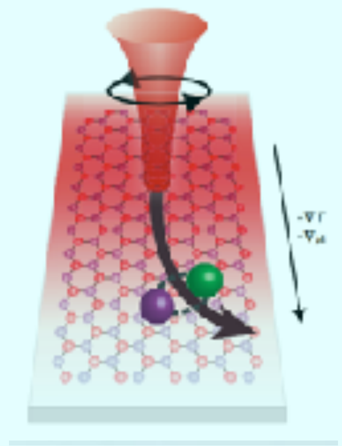




Berry Curvature in Exciton Polaritons.

Exciton: From the Berry curvature of electrons: $\Omega_c^{\tau}(\mathbf{q})$

$$\Omega_{\mathrm{ex}}^{\tau}(\mathbf{q}) = \frac{1}{4}\sum_{\mathbf{q}'} |\phi(q')|^2 \sum_{\beta=\pm 1} \Omega_c^{\tau}(\mathbf{q}' + \beta\mathbf{q}/2)$$



W. Yao and Q. Niu, PRL **101**, 106401 (2008)

Photon: Two polarizations.

$$\mathbf{A}_{\mathrm{ph}}^{\nu}(\mathbf{k}) = \nu(\cos\theta - 1)\mathbf{e}_{\phi}$$

$$\Omega_{\mathrm{ph}}^{\nu}(\mathbf{k}) = \nu k_z/k^3$$

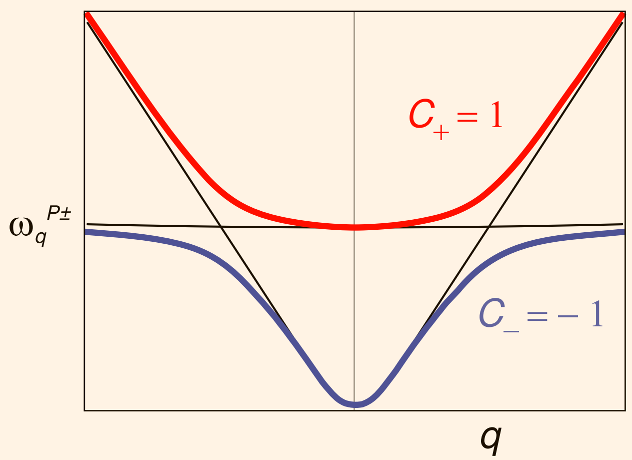
J. Segert, PRA **36**, 10 (1987)

M. Onoda, et al., PRL **36**, 083901 (2004)

Karzig et al PRX 5, 031001 (2015)

Polariton: the winding of exciton-photon coupling contributes

$$H = \sum_{\mathbf{q}} \omega_{\mathrm{ph},\mathbf{q}} a_{\mathbf{q}}^{\dagger} a_{\mathbf{q}} + \omega_{\mathrm{ex},\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} + (g_q e^{im\phi} b_{\mathbf{q}}^{\dagger} a_{\mathbf{q}} + \mathrm{H.c.})$$



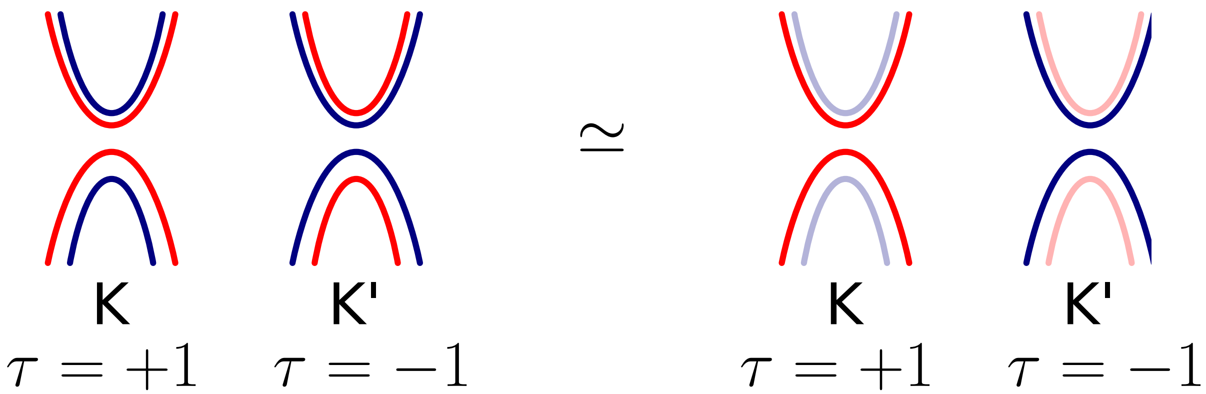
Which contribution dominates?

What is the Berry Curvature
of an Exciton polariton?

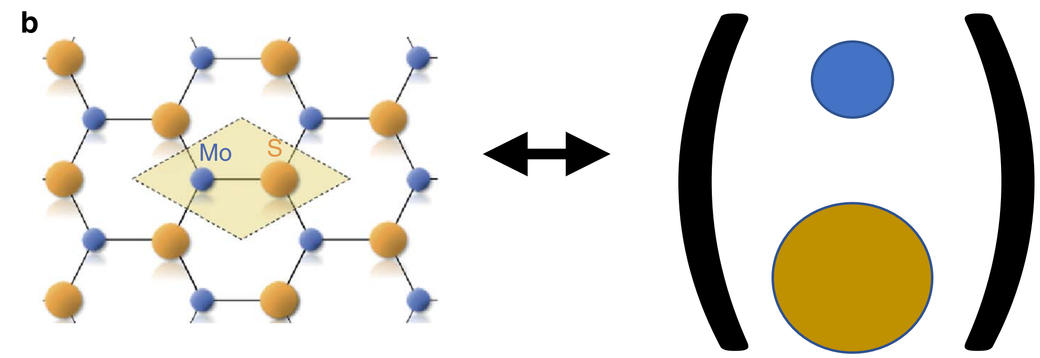
(i) We need the
exciton-photon hamiltonian

Electron Hamiltonian

Valley



Spinor



For each valley: $$H = h_0 + \vec{h} \cdot \vec{\sigma}$$

$$h_0 = \Delta/2,$$
$$\vec{h} = (\tau\, v\, k_x, v\, k_y, \Delta) = h(\sin\theta\cos\Psi, \sin\theta\sin\Psi, \cos\theta)$$

$$\tan\theta = \frac{vk}{\Delta}, \quad \tan\Psi = \frac{k_y}{\tau k_x}$$

$$\boxed{E = h_0 \pm h = h_0 \pm \sqrt{\Delta^2 + v^2 k^2}}$$

$$|\psi_{\mathbf{k}}^{c,\tau}\rangle = \begin{pmatrix} \cos\frac{\theta}{2} \\ \tau e^{i\tau\psi} \sin\frac{\theta}{2} \end{pmatrix}, \quad |\psi_{\mathbf{k}}^{v,\tau}\rangle = \begin{pmatrix} -\tau e^{-i\tau\psi} \sin\frac{\theta}{2} \\ \cos\frac{\theta}{2} \end{pmatrix}.$$

Wannier s-wave Excitons: Variational Wavefunction

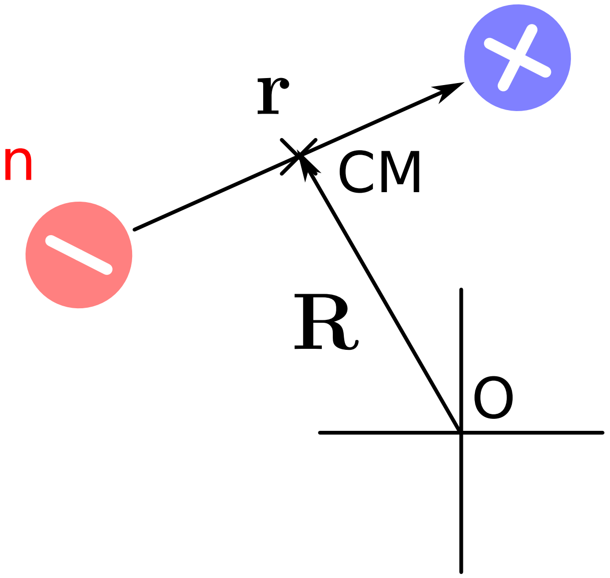
Center of mass: plane wave

$$|\psi_{\mathrm{ex},\tau}(\mathbf{Q})\rangle = \int d^2R \int d^2r \boxed{\frac{e^{i\mathbf{Q}\cdot\mathbf{R}}}{2\pi}} \boxed{\phi(\mathbf{r})} \psi^{\dagger}_{c,\tau}\left(\mathbf{R} - \frac{\mathbf{r}}{2}\right) \psi_{v,\tau}\left(\mathbf{R} + \frac{\mathbf{r}}{2}\right) |0\rangle$$

$$\phi(\mathbf{r}) = \sqrt{\frac{2}{a_{\mathrm{ex}}^2 \pi}} \exp[-r/a_{\mathrm{ex}}]$$

Relative motion: variational wavefunction

Prada, Elsa, et al. PRB 91, 245421 (2015)



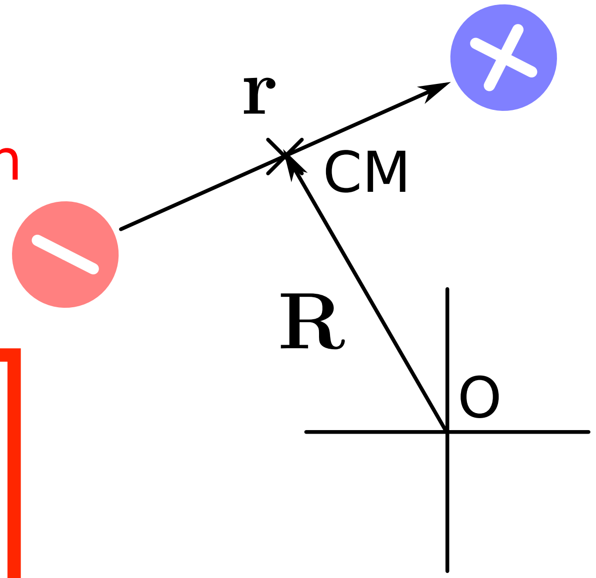
Wannier s-wave Excitons: Variational Wavefunction

Center of mass: plane wave

$$|\psi_{\mathrm{ex},\tau}(\mathbf{Q})\rangle = \int d^2R \int d^2r \boxed{\frac{e^{i\mathbf{Q}\cdot\mathbf{R}}}{2\pi}} \boxed{\phi(\mathbf{r})} \psi^\dagger_{c,\tau}\left(\mathbf{R}-\frac{\mathbf{r}}{2}\right)\psi_{v,\tau}\left(\mathbf{R}+\frac{\mathbf{r}}{2}\right)|0\rangle$$

Relative motion: variational wavefunction

$$\phi(\mathbf{r}) = \sqrt{\frac{2}{a_{\mathrm{ex}}^2\pi}}\exp[-r/a_{\mathrm{ex}}]$$



Prada, Elsa, et al. PRB 91, 245421 (2015)

$$|\psi_{\mathrm{ex},\tau}(\mathbf{Q})\rangle = \int d^2k\, \phi(\mathbf{k})\, c^\dagger_{c,\tau}\left(\mathbf{k}+\frac{\mathbf{Q}}{2}\right) c_{v,\tau}\left(\mathbf{k}-\frac{\mathbf{Q}}{2}\right)|0\rangle$$

$$\phi(\mathbf{k}) = \sqrt{\frac{2}{\pi}}a_{\mathrm{ex}}\frac{1}{[1+(a_{\mathrm{ex}}k)^2]^{3/2}}$$

Wannier s-wave Excitons: Variational Wavefunction

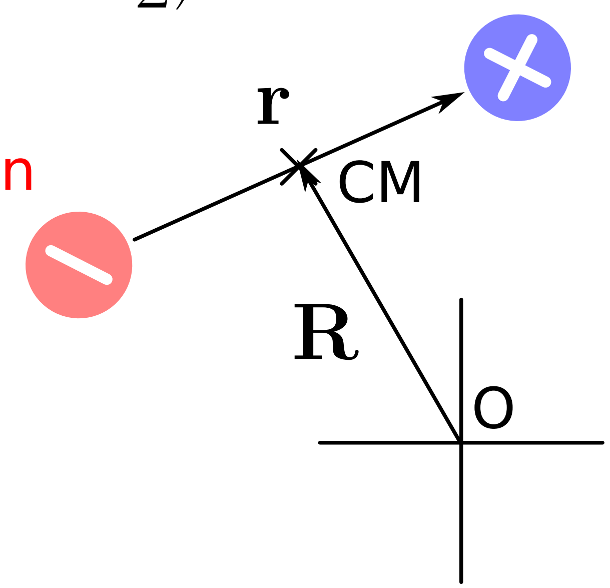
Center of mass: plane wave

$$|\psi_{\text{ex},\tau}(\mathbf{Q})\rangle = \int d^2R \int d^2r \, \frac{e^{i\mathbf{Q}\cdot\mathbf{R}}}{2\pi} \phi(\mathbf{r}) \psi^\dagger_{c,\tau}\left(\mathbf{R} - \frac{\mathbf{r}}{2}\right) \psi_{v,\tau}\left(\mathbf{R} + \frac{\mathbf{r}}{2}\right) |0\rangle$$

$$\phi(\mathbf{r}) = \sqrt{\frac{2}{a_{\text{ex}}^2 \pi}} \exp[-r/a_{\text{ex}}]$$

Relative motion: variational wavefunction

Prada, Elsa, et al. PRB 91, 245421 (2015)



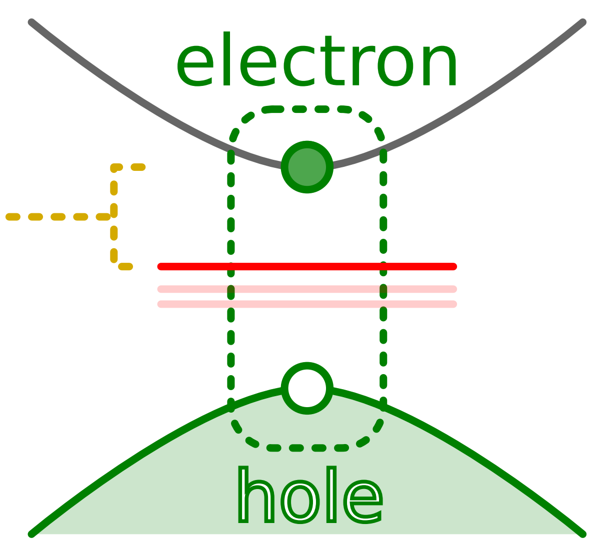
$$|\psi_{\text{ex},\tau}(\mathbf{Q})\rangle = \int d^2k \, \phi(\mathbf{k}) \, c^\dagger_{c,\tau}\left(\mathbf{k} + \frac{\mathbf{Q}}{2}\right) c_{v,\tau}\left(\mathbf{k} - \frac{\mathbf{Q}}{2}\right) |0\rangle$$

$$\phi(\mathbf{k}) = \sqrt{\frac{2}{\pi}} a_{\text{ex}} \frac{1}{[1 + (a_{\text{ex}}k)^2]^{3/2}}$$

$$\boxed{H_{\text{ex}} = \sum_{\tau} \int d^2Q \left[\frac{\hbar^2 Q^2}{2M_{\text{ex}}} + 2\Delta + E_b\right] b^\dagger_{\tau,\mathbf{Q}} b_{\tau,\mathbf{Q}}}$$

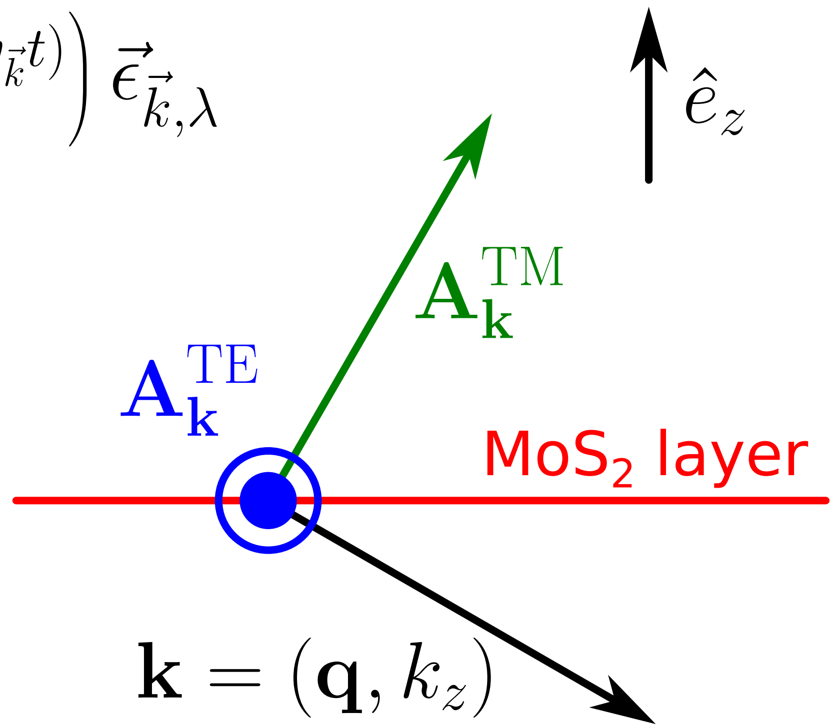
quadratic dispersion

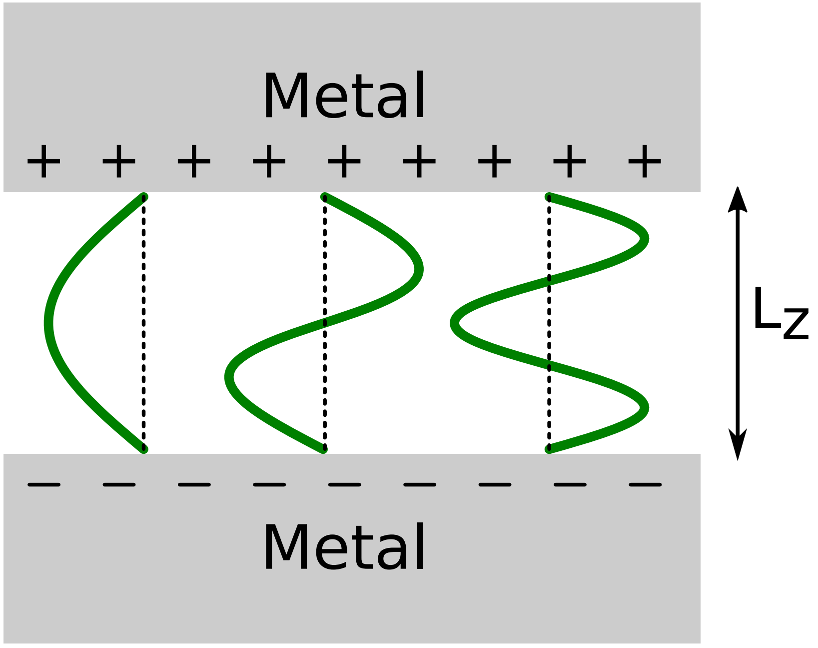
binding energy



Photons in a cavity

$$\vec{A}(\vec{r},t) = \frac{1}{\sqrt{\mathcal{V}}} \sum_{\vec{k}} \sum_{\lambda=1,2} \left(A_{\vec{k},\lambda} e^{i(\vec{k}\cdot\vec{r}-\omega_{\vec{k}} t)} + A^*_{\vec{k},\lambda} e^{-i(\vec{k}\cdot\vec{r}-\omega_{\vec{k}} t)} \right) \vec{\epsilon}_{\vec{k},\lambda}$$



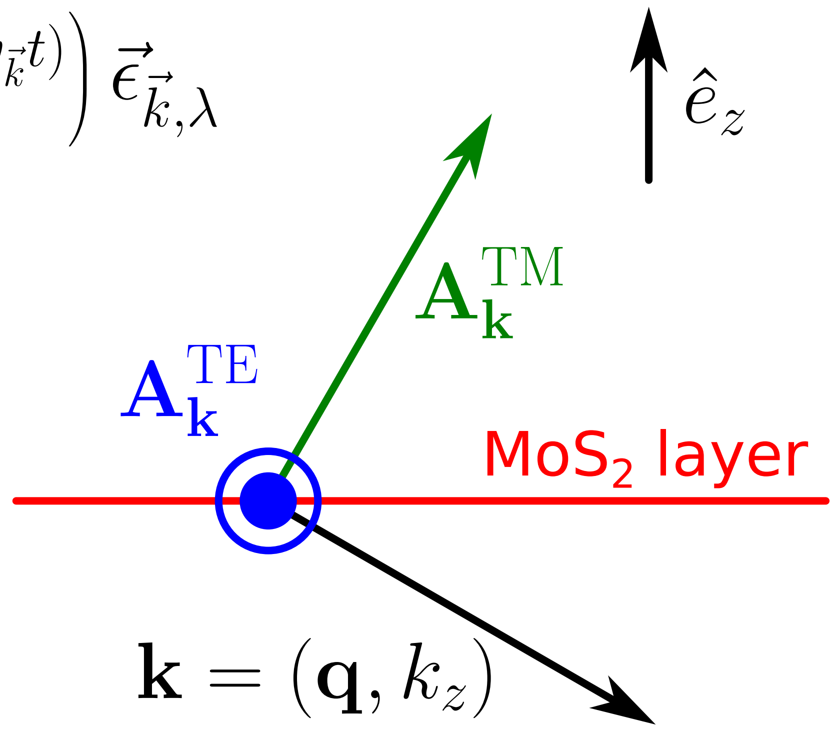


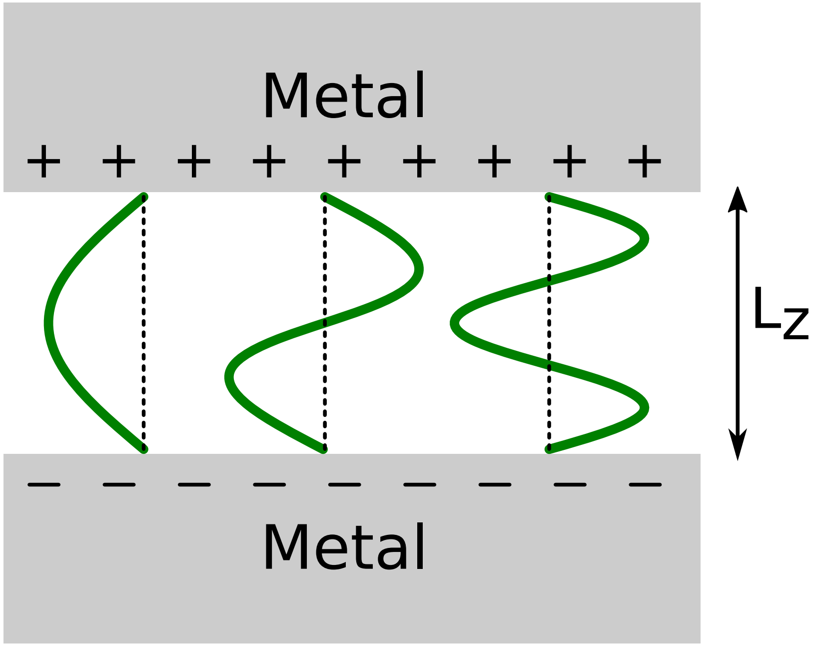
Photons in a cavity

$$\vec{A}(\vec{r},t) = \frac{1}{\sqrt{\mathcal{V}}} \sum_{\vec{k}} \sum_{\lambda=1,2} \left(A_{\vec{k},\lambda} e^{i(\vec{k}\cdot\vec{r}-\omega_{\vec{k}}t)} + A^{*}_{\vec{k},\lambda} e^{-i(\vec{k}\cdot\vec{r}-\omega_{\vec{k}}t)} \right) \vec{\epsilon}_{\vec{k},\lambda}$$

Perfect metal:

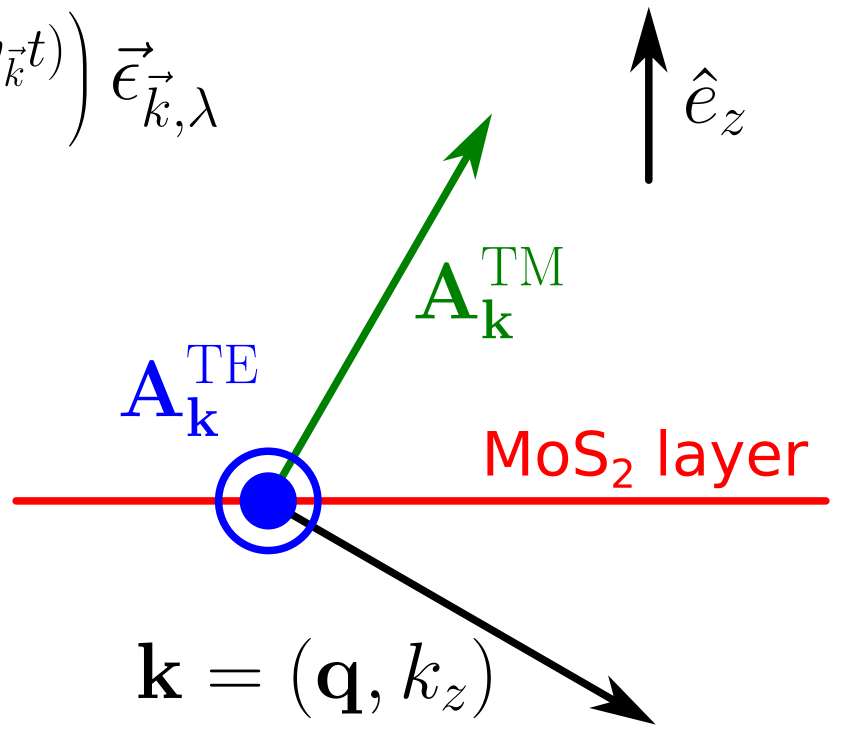
$$\vec{A}(\vec{r},t) = \sum_{\vec{q},k_z>0} F_{q,k_z} e^{i(\vec{q}\cdot\vec{r}-\omega_{\vec{q},k_z}t)} \Big\{ \vec{e}_{\vec{q}_\perp} \boxed{\sin(k_z z)} \hat{a}^{\mathrm{TE}}_{\vec{q},k_z}$$
$$+ \left[\hat{e}_{\vec{q}_\parallel} \boxed{\sin(k_z z)} \sqrt{1 - f^2_{q,k_z}} - i \hat{e}_z \cos(k_z z) f_{q,k_z} \right] \hat{a}^{\mathrm{TM}}_{\vec{q},k_z} \Big\}$$
$$+ \sum_{\vec{q}} \frac{F_{q,0}}{2i} \hat{e}_z \hat{a}^{\mathrm{TM}}_{\vec{q},0} e^{i(\vec{q}\cdot\vec{r}-\omega_{\vec{q},0}t)} + \mathrm{h.c.}$$





Photons in a cavity

$$\vec{A}(\vec{r},t) = \frac{1}{\sqrt{\mathcal{V}}} \sum_{\vec{k}} \sum_{\lambda=1,2} \left(A_{\vec{k},\lambda} e^{i(\vec{k}\cdot\vec{r}-\omega_{\vec{k}}t)} + A^*_{\vec{k},\lambda} e^{-i(\vec{k}\cdot\vec{r}-\omega_{\vec{k}}t)} \right) \vec{\epsilon}_{\vec{k},\lambda}$$



Perfect metal:

$$\vec{A}(\vec{r},t) = \sum_{\vec{q},k_z>0} F_{q,k_z} e^{i(\vec{q}\cdot\vec{r}-\omega_{\vec{q},k_z}t)} \Big\{ \vec{e}_{\vec{q}_\perp} \sin(k_z z) \hat{a}^{\mathrm{TE}}_{\vec{q},k_z}$$

$$+ \left[\hat{e}_{\vec{q}_\parallel} \sin(k_z z) \sqrt{1 - f^2_{q,k_z}} - i\hat{e}_z \cos(k_z z) f_{q,k_z} \right] \hat{a}^{\mathrm{TM}}_{\vec{q},k_z} \Big\}$$

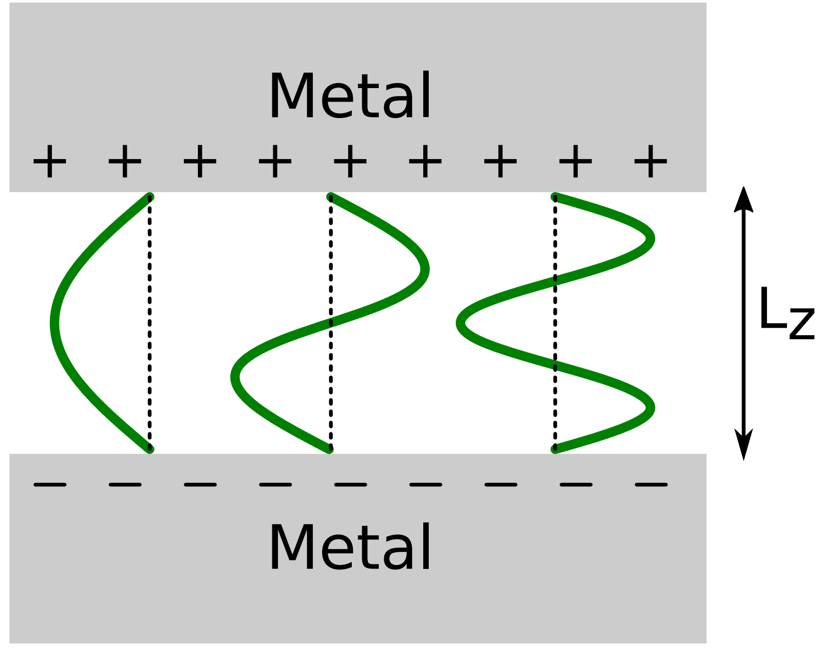
$$+ \sum_{\vec{q}} \frac{F_{q,0}}{2i} \hat{e}_z \hat{a}^{\mathrm{TM}}_{\vec{q},0} e^{i(\vec{q}\cdot\vec{r}-\omega_{\vec{q},0}t)} + \mathrm{h.c.}$$

$$\boxed{k_z = \frac{m\pi}{L_z}, \quad m \in \mathbb{Z}}$$

$$\boxed{f_Q = \frac{Q}{\omega/c}}$$

Cavity width encoded here

$$\omega_{\mathbf{Q},k_z} = \hbar c \sqrt{Q^2 + \left(\frac{m\pi}{L_z}\right)^2}$$



$$\boxed{H_{\mathrm{ph}} = \sum_{\mathbf{Q},k_z>0} \hbar\omega_{\mathbf{Q},k_z} \Big\{ \sum_{\nu} [\hat{a}^{\nu}_{\mathbf{Q},k_z}]^\dagger \hat{a}^{\nu}_{\mathbf{Q},k_z} + \mathrm{h.c.} \Big\}}$$

From electron-phonon to exciton-photon coupling

1. Electron-Photon coupling

(i) Expand Hamiltonian close to minima $\quad H(k) \approx H_0 + k_i H_I^i$

(ii) Minimal substitution $\vec{p} \rightarrow \vec{p} - e\vec{A} \quad H(k) \rightarrow H(k) - \frac{ev}{\hbar} A_i H_I^i$

$$\rightarrow \quad W_{EM} = -\frac{ev}{\hbar} A_i H_I^i \qquad \text{with } H_I^x = \tau\sigma_x, \quad H_I^y = \tau\sigma_y$$

2. Exciton-Photon coupling

(i) Project W_{EM} onto $|0_{\mathrm{ex}}\rangle \otimes |\gamma_{\mathbf{k},\nu}\rangle$ and $|\psi_{\mathrm{ex},\tau}(\mathbf{Q})\rangle \otimes |0_\gamma\rangle \qquad (\mathbf{k} = (\mathbf{Q}, k_z))$

(ii) Expand Electron Wavefunction to $O(Q^2)$ $\quad |\psi^{c(v),\tau}_{\mathbf{k}\pm\mathbf{Q}/2}\rangle \simeq |\psi^{c(v),\tau}_{\mathbf{k}}\rangle \pm \frac{1}{2} Q^i \partial_{k^i} |\psi^{c(v),\tau}_{\mathbf{k}}\rangle + \frac{1}{8} Q^i Q^j \partial^2_{k^i k^j} |\psi^{c(v),\tau}_{\mathbf{k}}\rangle$

$$H = H_{\mathrm{ex}} + H_{\mathrm{ph}} + H_{\mathrm{ph-ex}}$$

$$H_{\mathrm{ph-ex}} = \sum_{\nu,\tau} \sum_{k_z} \int d^2Q \, h^{\nu,\tau}_{\mathrm{ph-ex}}(\mathbf{Q}, k_z) \, a^\dagger_\nu(\mathbf{Q}, k_z) b_\tau(\mathbf{Q}) + \mathrm{h.c.}$$

$\nu = \{\mathrm{TE}, \mathrm{TM}\}$

$\tau = \pm 1$

Exciton-photon coupling

$$\boxed{\begin{aligned} h^{TE,\tau}_{ph-ex}(\vec{Q},k_z) &= \gamma\ \exp^{-\imath\tau\phi_{\vec{Q}}} \\ h^{TM,\tau}_{ph-ex}(\vec{Q},k_z) &= \imath\,\frac{k_z}{k}\,\gamma\ \exp^{-\imath\tau\phi_{\vec{Q}}} \end{aligned}} + O\left(\frac{Q^2}{(\Delta/v)^2}\right)$$

For MoS_2

$$\boxed{\frac{\mathcal{O}(Q^2)}{\mathcal{O}(Q^0)} \sim 10^{-8},\ \propto \left[\frac{v}{\Delta}\right]^2}$$

$$\gamma = \frac{e\kappa\Delta}{\sqrt{\pi\hbar\omega_k\epsilon_0 L_z}} F_0(\kappa) \qquad F_0(\kappa) = \frac{1}{\kappa}\left[\frac{1}{\kappa} + \frac{1}{\kappa+1}\right] \qquad \kappa = \frac{a_{ex}\Delta}{v} \approx 3.7 \text{ for } \mathrm{MoS}_2$$

$$F_0 \approx 0.13$$

Changing polarization basis onto the circular polarization basis set (with respect to $\vec{k}$, not to $\vec{e}_z$!!):

$$a_+ = \frac{1}{\sqrt{2}}\left[a_{TM} - \imath\, a_{TE}\right] e^{-\imath\phi_{\vec{k}}}$$

$$a_- = \frac{1}{\sqrt{2}}\left[a_{TM} + \imath\, a_{TE}\right] e^{\imath\phi_{\vec{k}}}$$

Polariton Hamiltonian in circular polarization basis.

$$
H = \begin{array}{cccc}
\tau = 1 & \nu = 1 & \tau = -1 & \nu = -1 \\
\end{array}
$$

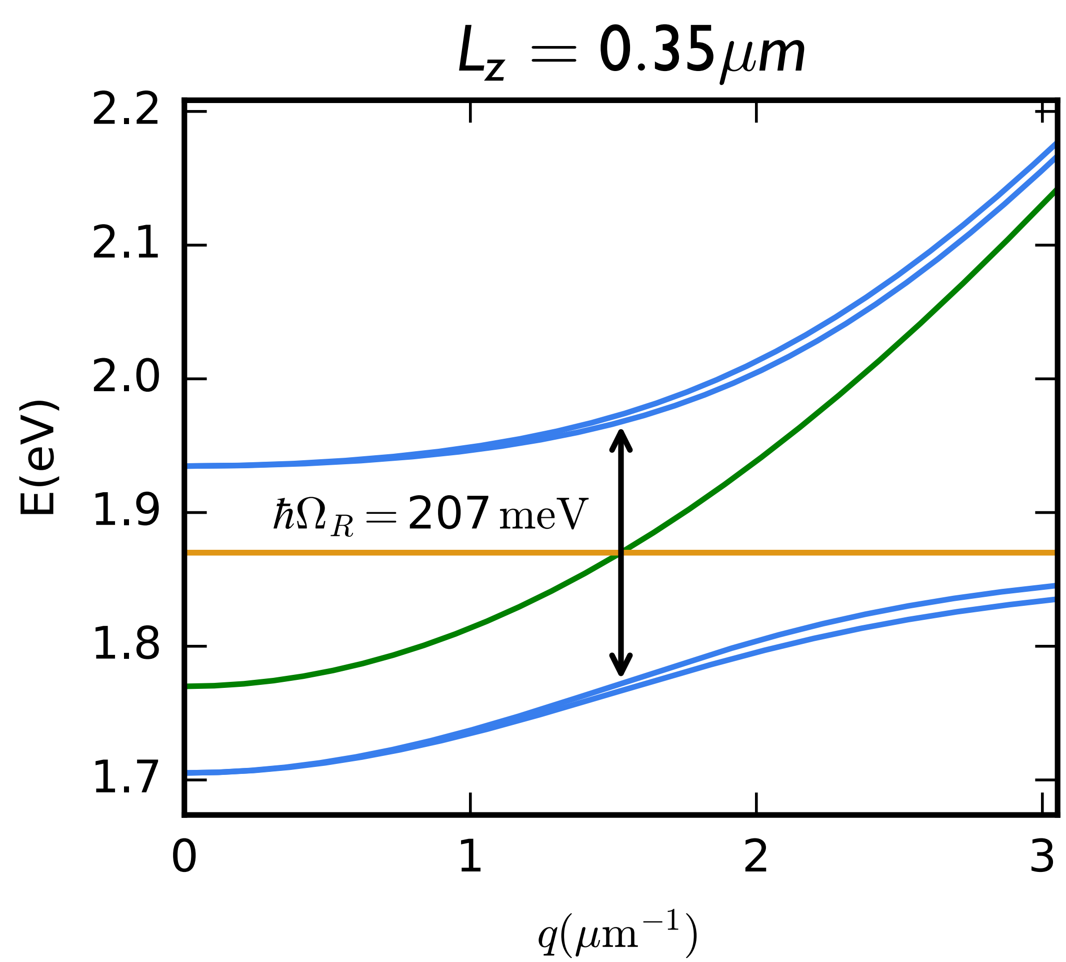
$$
H = \begin{bmatrix}
\omega_{\rm ex} & i\gamma_0 & 0 & i\gamma_0 \frac{q^2}{4k_z^2} e^{2i\phi} \\
-i\gamma_0 & \omega_{\rm ph} & i\gamma_0 \frac{q^2}{4k_z^2} e^{2i\phi} & 0 \\
0 & -i\gamma_0 \frac{q^2}{4k_z^2} e^{-2i\phi} & \omega_{\rm ex} & -i\gamma_0 \\
-i\gamma_0 \frac{q^2}{4k_z^2} e^{-2i\phi} & 0 & i\gamma_0 & \omega_{\rm ph}
\end{bmatrix}
$$

small q expansion

winding phase

At strong coupling

$q/k_z \approx 0.3$



fine splitting $5\ \mathrm{meV}$

compatible with photon linewidth

S. Dufferwiel et al

Nat. Comm. **6**, 8579 (2015)

Berry Curvature of composite particles

Composite-particle state: $|n\rangle = \sum_i \psi_n^i \, |e_i\rangle$

$|e_i\rangle$ elementary constituent

ψ_n^i encode the coupling

Berry connection of composite particle state: $\vec{A}_n = \imath \, \langle n | \nabla_{\vec{q}} | n \rangle$

Berry Curvature: $\vec{\Omega}_n = \vec{\nabla}_{\vec{q}} \times \vec{A}_n$

Defining the "intrinsic" Berry connection $\vec{A}_{jk}^{int} = \imath \, \langle e_j | \nabla_{\vec{q}} | e_k \rangle$

$$\vec{A}_n = \underbrace{\imath \sum_j \psi_n^{j*} \, \nabla_{\vec{q}} \, \psi_n^j}_{\text{extrinsic}} + \underbrace{\sum_{j,k} \psi_n^{j*} \, \vec{A}_{jk}^{int} \, \psi_n^k}_{\text{intrinsic}}$$

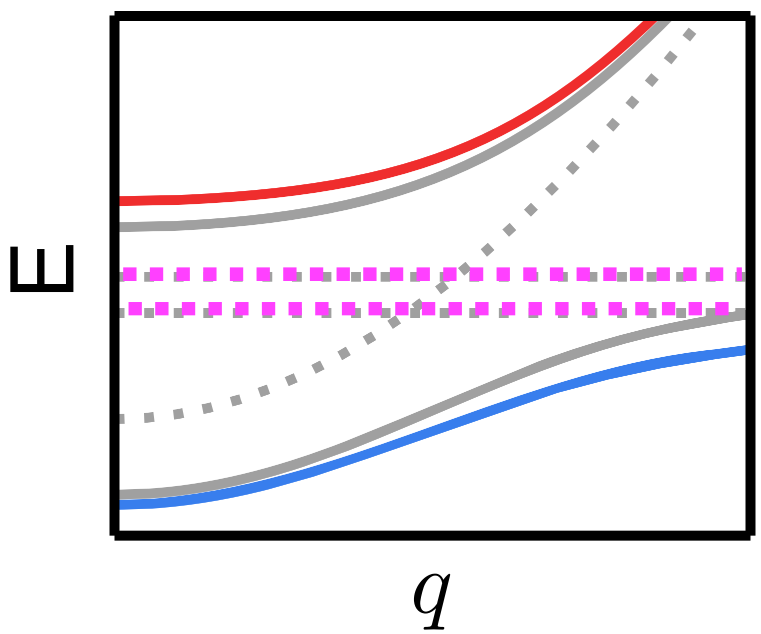
A. Gutierrez et al., Phys. Rev. Lett. **121**, 137402 (2018)

Effective Description of Polaritons.

Due to the "wrong" valley-polarization coupling: $\Omega_n = 0$ opposite valley/polarization cancel

need to break time-reversal symmetry → apply magnetic field

Spectrum



Schrieffer-Wolf transformation

$$H_{\text{eff}} = \epsilon_0(q) + \begin{pmatrix} \Delta & \alpha q^2 e^{2i\phi} \\ \alpha q^2 e^{-2i\phi} & -\Delta \end{pmatrix}$$

($\Delta \propto V_z$ exciton Zeeman and α depends on γ)

Two Effective 2x2 Hamiltonians (for LPs and for UPs)

analogous to gapped bilayer graphene ⇒ analytical results

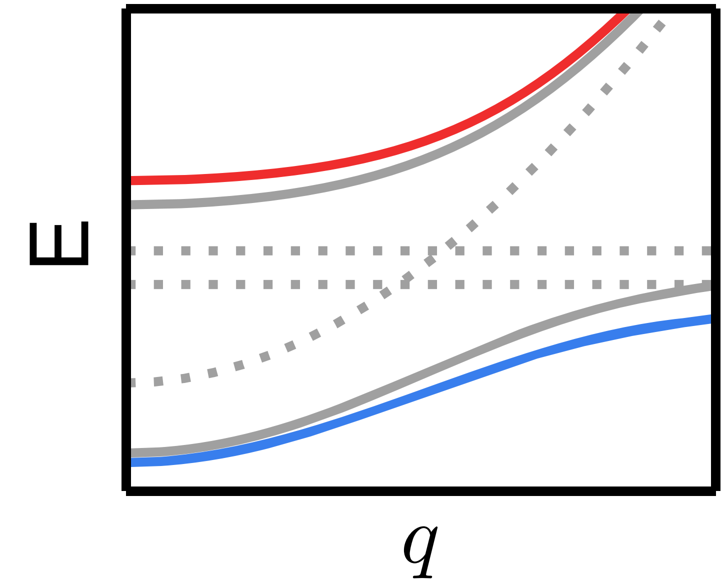
see also O. Bleu et al., PRL 121, 020401 (2018)
and the poster in this conference!

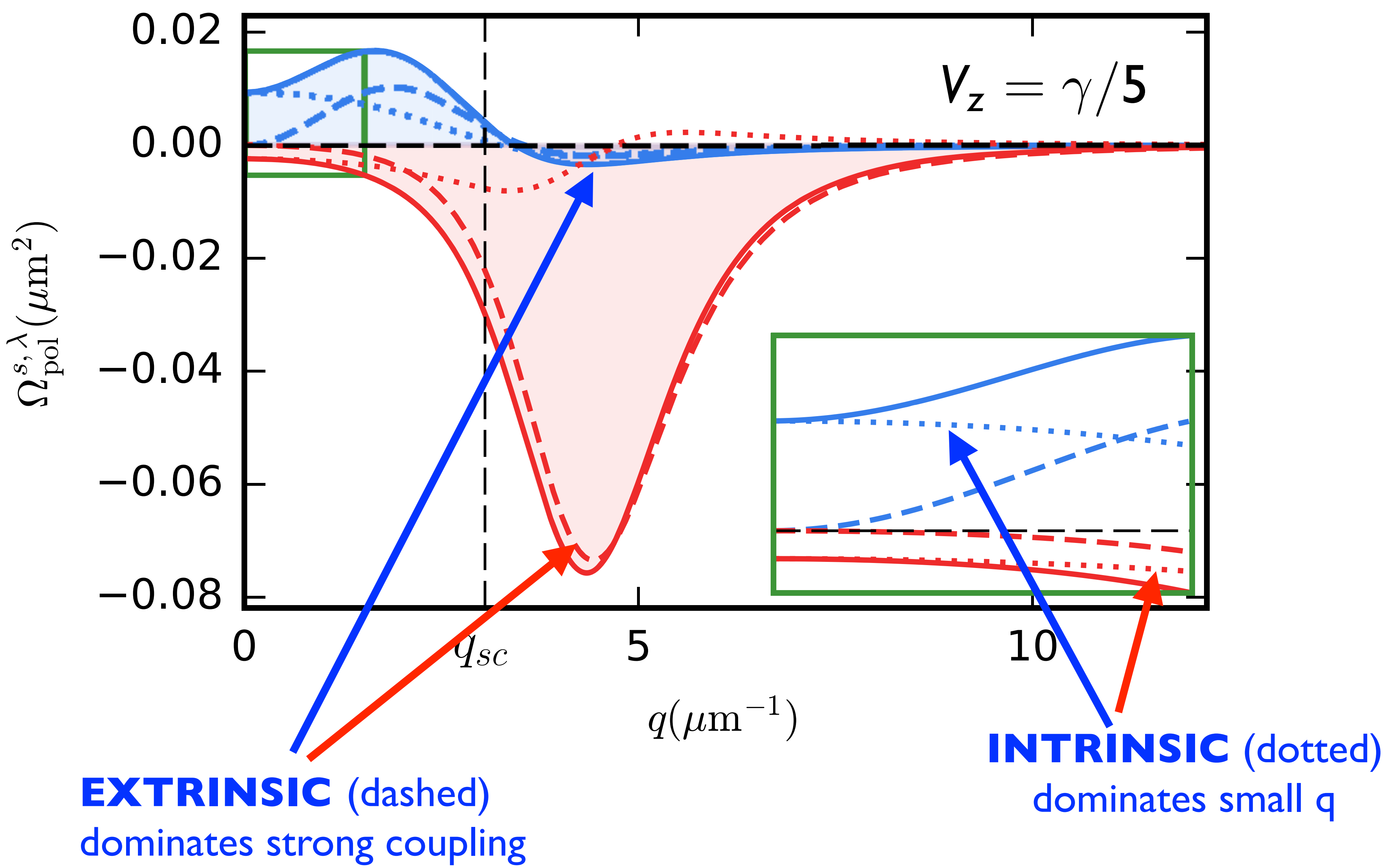
A. Gutierrez et al., Phys. Rev. Lett. **121**, 137402 (2018)

Composite Berry Curvature

- The Exciton Berry curvature is negligible

- "Coupling" Berry curvature $\Omega^{s,\lambda}_{ext} = \dfrac{2\lambda\Delta_s}{\alpha}\dfrac{q^2}{\left[q^2+(\Delta_s/\alpha)^2\right]^{3/2}}$

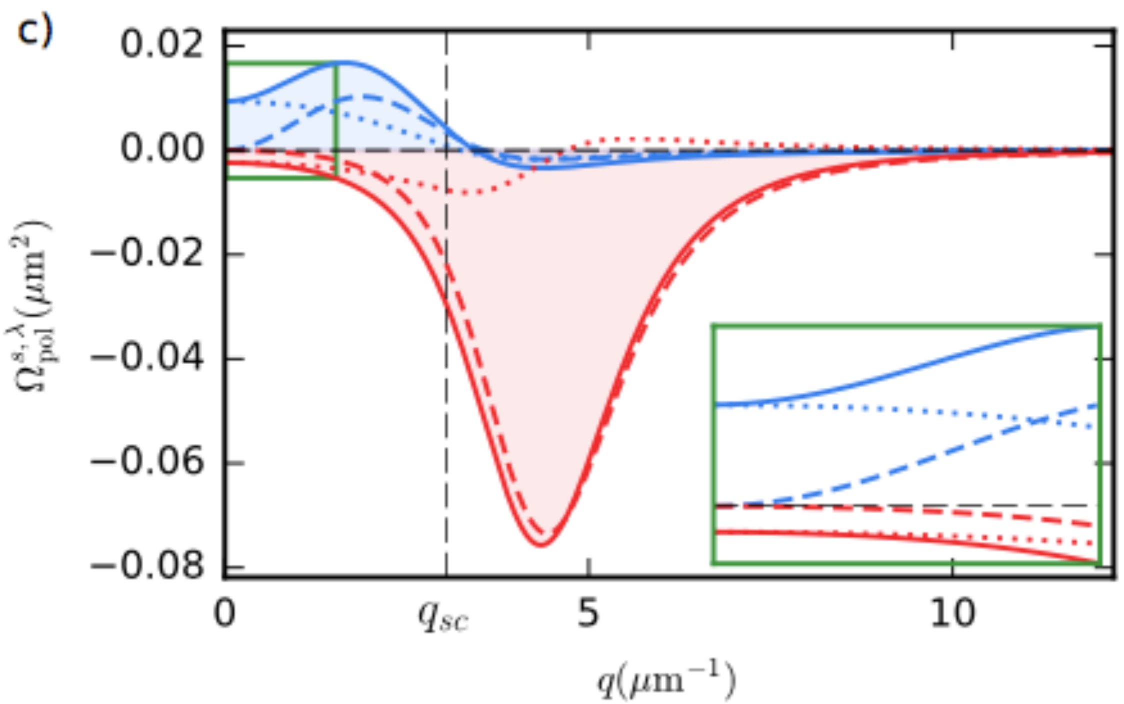


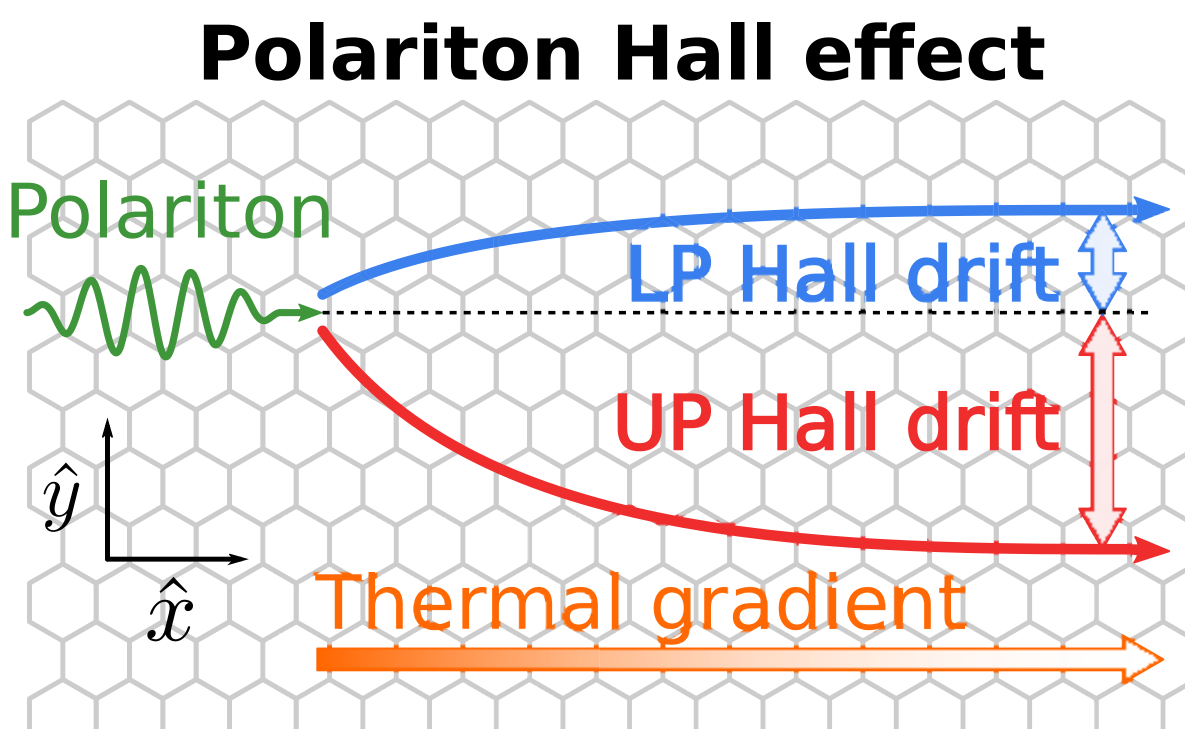


Exciton-Polariton Anomalous Hall Effect

quasi-classical equation of motion for wavepacket

$$\dot{\mathbf{r}}_c = \frac{\partial E_{\mathrm{pol}}}{\partial \mathbf{q}_c} - \dot{\mathbf{q}} \times \mathbf{\Omega}_{\mathrm{pol}}(q_c) \qquad \dot{\mathbf{q}}_c = -\frac{\partial E_{\mathrm{pol}}}{\partial \mathbf{r}_c}$$





$$\Delta_y \approx \begin{array}{l} 0.2\,\mu m\,(UP) \\ 0.03\,\mu m\,(LP) \end{array}$$

Summary

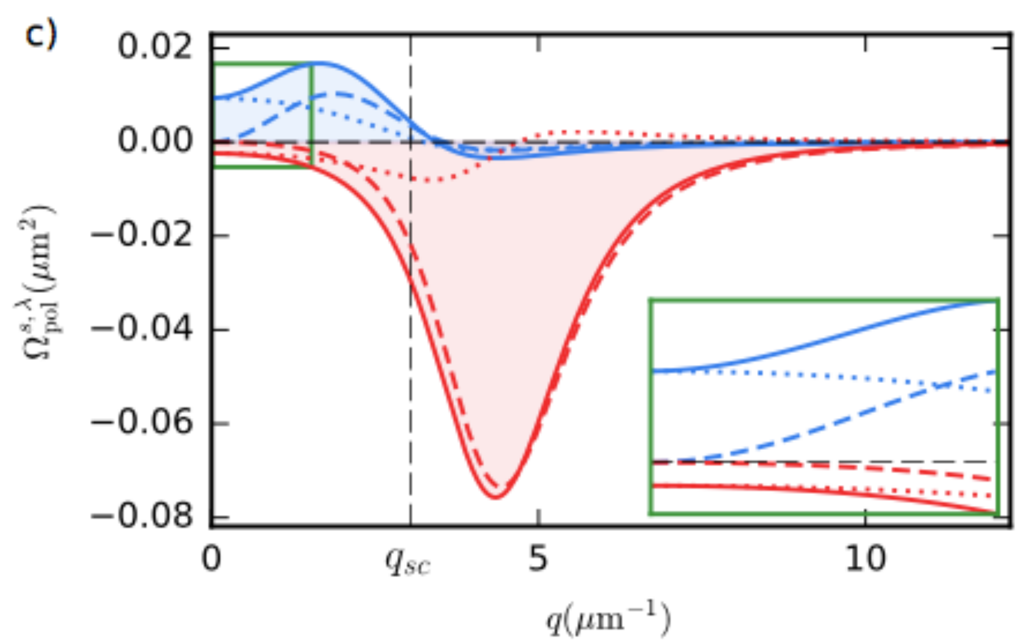
- Cavity-modified selection rules at finite momentum.

$$H = \begin{array}{c} \begin{array}{cccc} \tau=1 & \nu=1 & \tau=-1 & \nu=-1 \end{array} \\ \begin{bmatrix} \omega_{\text{ex}} & i\gamma_0 & 0 & i\gamma_0\frac{q^2}{4k_z^2}e^{2i\phi} \\ -i\gamma_0 & \omega_{\text{ph}} & i\gamma_0\frac{q^2}{4k_z^2}e^{2i\phi} & 0 \\ 0 & -i\gamma_0\frac{q^2}{4k_z^2}e^{-2i\phi} & \omega_{\text{ex}} & -i\gamma_0 \\ -i\gamma_0\frac{q^2}{4k_z^2}e^{-2i\phi} & 0 & i\gamma_0 & \omega_{\text{ph}} \end{bmatrix} \end{array}$$

- Polariton fine splitting.

- Berry curvature of composite particles.

- Exciton Polaritons have a Berry curvature arising from the photon and the coupling ones.



A. Gutierrez et al., Phys. Rev. Lett. **121**, 137402 (2018)