# Quantum Geometry and Related Phenomena in 2D Materials

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# Introduce using First-principles Theory

## I. New Class of Spectroscopies based on Quantum Geometry

### Demonstration of a Vibrational Spectroscopy In 2D Materials

**R Bhuvaneswari**, M M Deshmukh and U V Waghmare, Physical Review B 110, 014305 (2024)

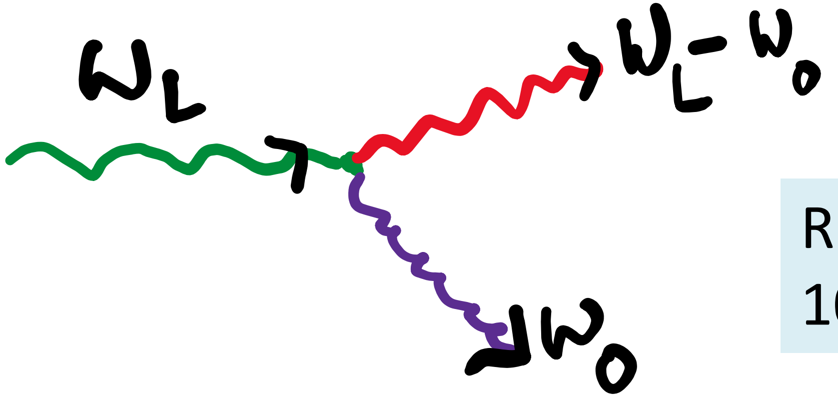
## II. Anomalous Hall Transistor

### *Graphene:*$CrTe_2$ heterostructure

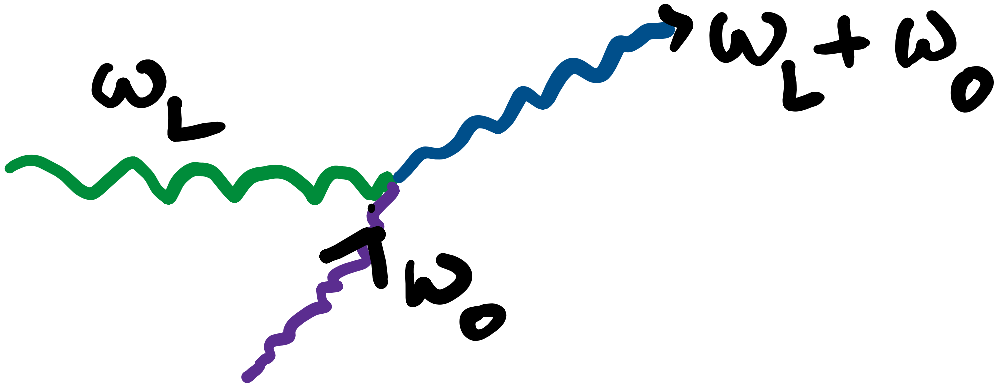
**S Menon** and U V Waghmare, Nanoscale (under revision)

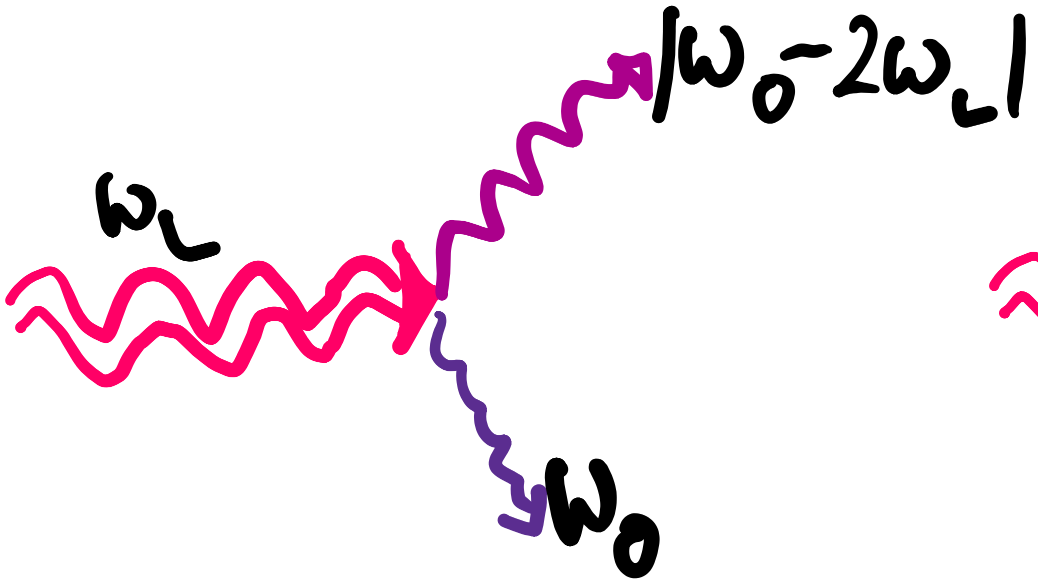
# Sensing Vibrations Using Quantum Geometry of Electrons

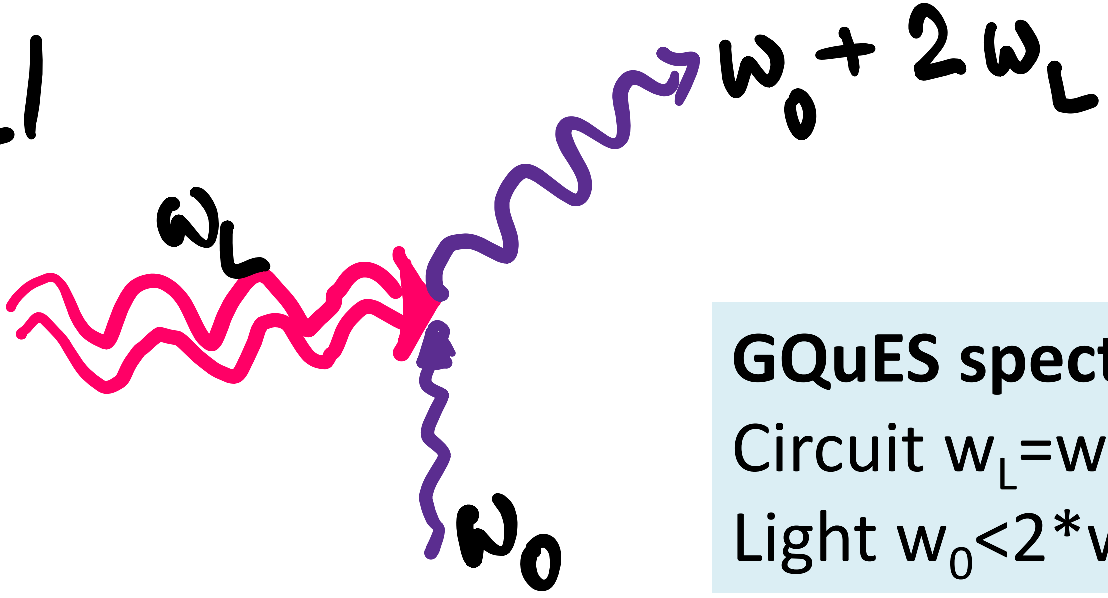
R Bhuvaneswari, M M Deshmukh and U V Waghmare, Physical Review B 110, 014305 (2024)




Raman
100 years!



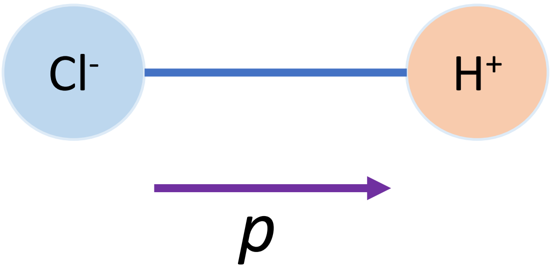



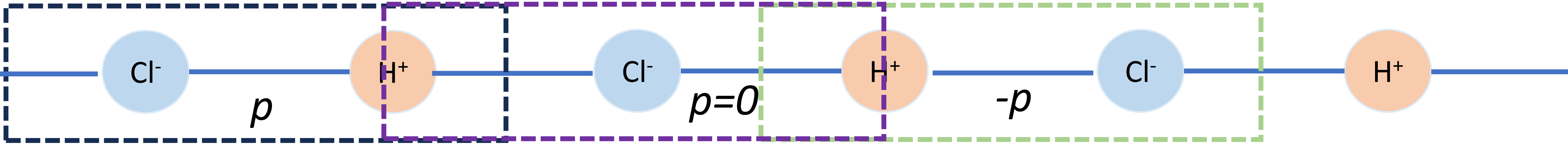

**GQuES spectra**
Circuit $w_L = w_{AC}$
Light $w_0 < 2 * w_L$

# Puzzle of Electric Polarization

Electric dipole

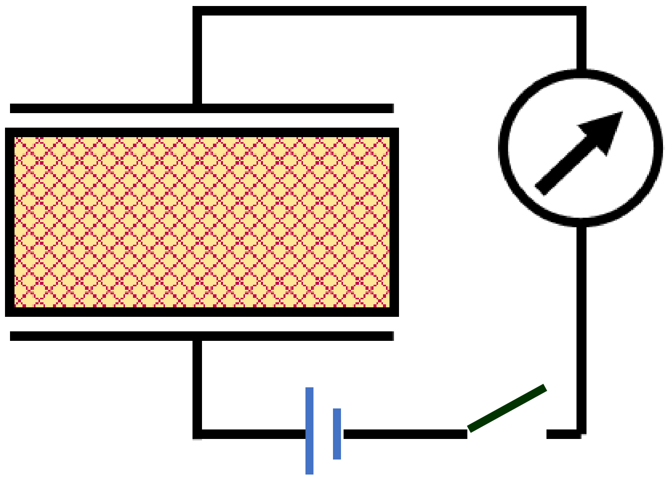



Polarization of a crystal: dipole moment/unit cell




- Dependence of P on the *choice of Unit Cell*
- Even more tricky when one realizes that the quantum electronic density is a *continuous* periodic distribution, not a set of discrete charges!

# Measurement of Polarization



Difference in polarization $\Delta P$
accessible to measurement of macroscopic current

Bulk properties: *derivatives* of P with respect to field ($\lambda$)
e.g. dielectric constant, piezoelectric, pyroelectric constants

Change in polarization, $\Delta P = \int_0^L \frac{\partial P}{\partial \lambda} d\lambda = P(\lambda = L) - P(\lambda = 0)$

Redistribution of electrons within a crystal in response to time-varying field λ involves flow of *adiabatic* **current**:
density of current $j$ is the time rate change of polarization $\frac{\partial P}{\partial t}$

Change in polarization, $\Delta P = \int_0^T \frac{\partial P}{\partial t} dt = \int_0^T j \, . dt$

*Path Dependent* $\oint j dt = nP_{Quantum}$

R Resta, Ferroelectric 136, 51 (1992)

Rabe & Waghmare, Ferroelectric 136, 147 (1992)

# Quantum Theory of Polarization: $\Delta P$

Change in polarization $\Delta P$ of an insulator with adiabatic change in the Hamiltonian ($\lambda$) obtained as integrated current analytically [1]

$$\Delta P = P(\lambda = L) - P(\lambda = 0), \text{ where } P(\lambda) \propto i \int_{-\pi/a}^{\pi/a} d\boldsymbol{k} \left\langle u_{\boldsymbol{k}}^{\lambda} \middle| \frac{\partial}{\partial k} \middle| u_{\boldsymbol{k}}^{\lambda} \right\rangle$$

$$\gamma = i \int_{-\pi/a}^{\pi/a} d\boldsymbol{k} \left\langle u_{\boldsymbol{k}}^{\lambda} \middle| \frac{\partial}{\partial k} \middle| u_{\boldsymbol{k}}^{\lambda} \right\rangle \sim \frac{\langle x \rangle}{a}(2\pi)$$

is the quantum geometric (Berry) phase

Connection with the **Pancharatnam phase** in optics[2]

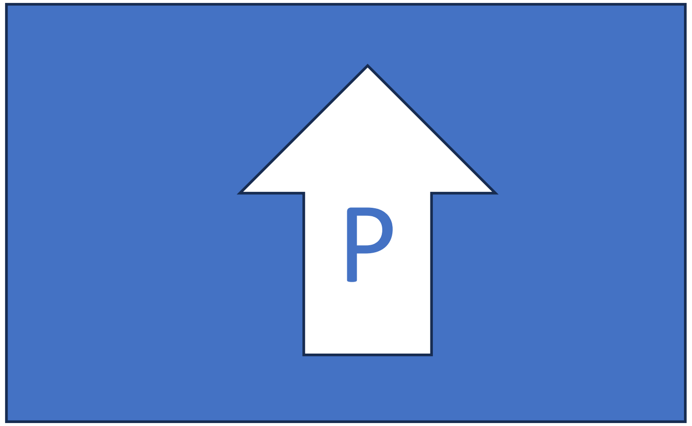
The integrand is Berry connection $A(\boldsymbol{k})$[3]

Most practical calculations use discretized formulation of the integral


Ref. 1. R. D. King-Smith and D. Vanderbilt, Theory of polarization of crystalline solids, *Phys. Rev. B.* **47**, 3 (1993).

Ref. 2. S. Pancharatnam, Generalized theory of interference and its applications. Part I. Coherent pencils, *Proc. Ind. Acad. Science* **A44**, 247 (1956).

Ref. 3. M. V. Berry, Quantal Phase factors accompanying adiabatic changes, *Proc. Roy. Soc. (London)* **392**, 45 (1984).

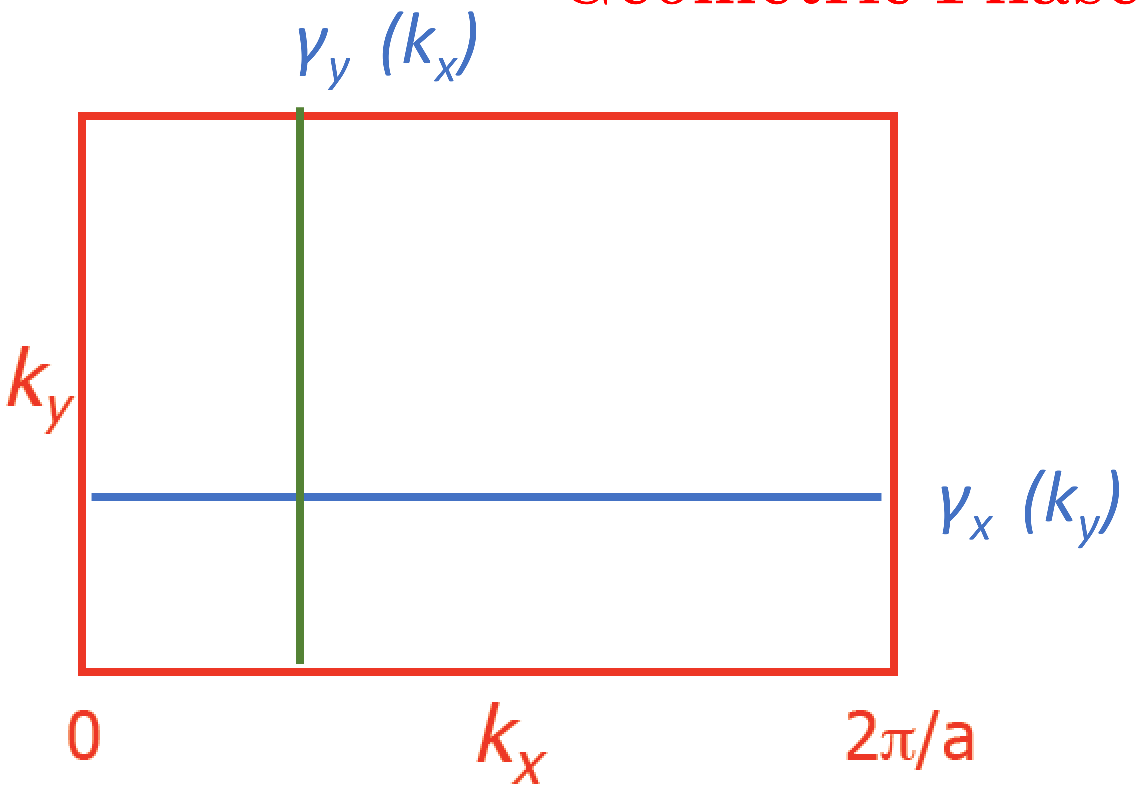



Measurement of P
Flow of **Electric Current**

Quantum Theory of P:
**Geometric Phase**

$$\gamma_\alpha = i\int_{-\pi/a}^{\pi/a} d\boldsymbol{k} \left\langle u_{\boldsymbol{k}} \middle| \frac{\partial}{\partial k_\alpha} \middle| u_{\boldsymbol{k}} \right\rangle \sim \frac{\langle r_\alpha \rangle}{a}(2\,\pi)$$

# Geometric Phases in 2D




Berry potential:

$$\mathbf{A}(\mathbf{k}) = -\mathrm{Im}\,\langle u_{\mathbf{k}}|\nabla_{\mathbf{k}}|u_{\mathbf{k}}\rangle$$

Berry phase:

$$\phi = \oint \mathbf{A}(\mathbf{k}) \cdot d\mathbf{k}$$

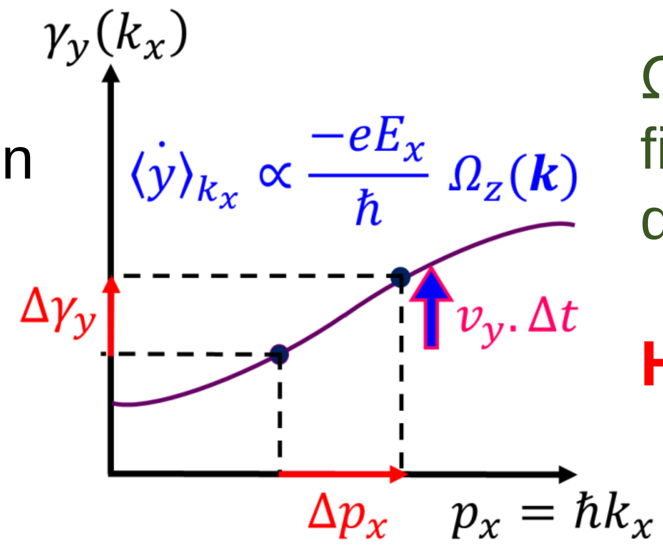
Berry curvature:

$$\Omega(\mathbf{k}) = \nabla \times \mathbf{A}$$

*$\gamma_\alpha$ : <x> and <y>*

Applied Electric Field, $E_x$, causes change in momentum $\hbar\Delta k_x$ results in shift in <y> if $\Omega \neq 0$, change in Berry phase $\gamma_y(k_x)$

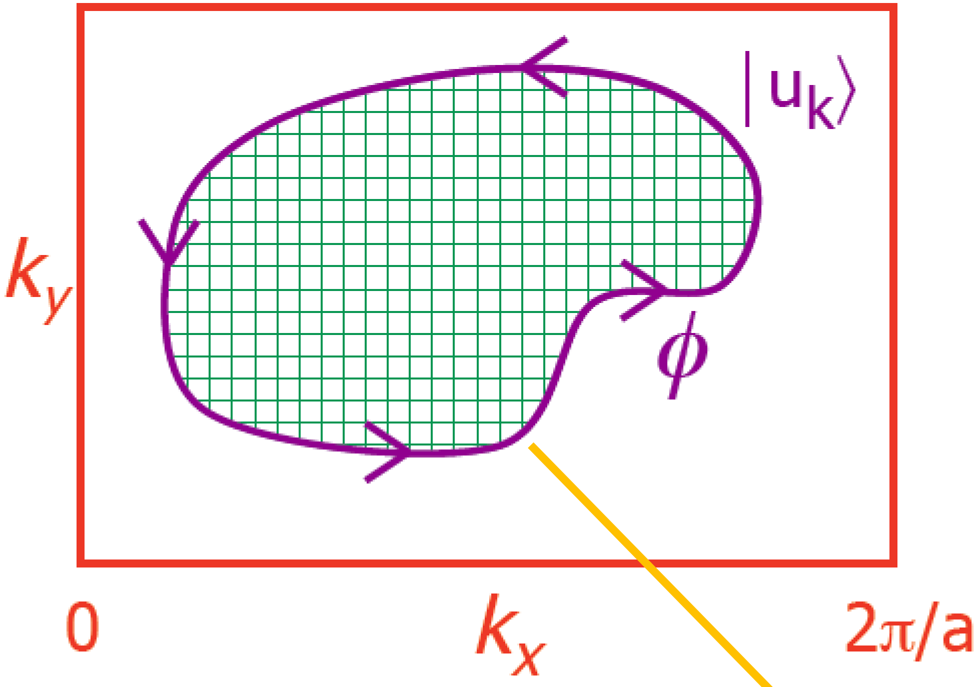
*anomalous velocity $v_y$*




$\Omega \neq 0$ acts like a magnetic field emerging from quantum geometry

**Hall Effect** with $B_{ext}$=0

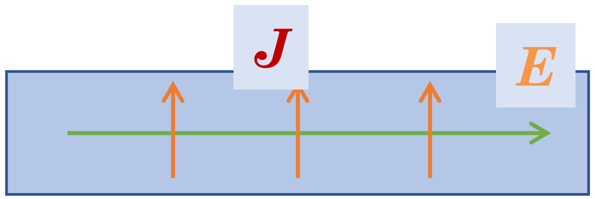
# Geometric Phases and Anomalous Hall Conductivity:
## Hall effect with out magnetic field!




$$\vec{v} = -\frac{e}{c}\vec{E} \times \vec{\Omega}(\vec{k})$$

$E_x$ causes $j_y$

Fermi Surface of a metal



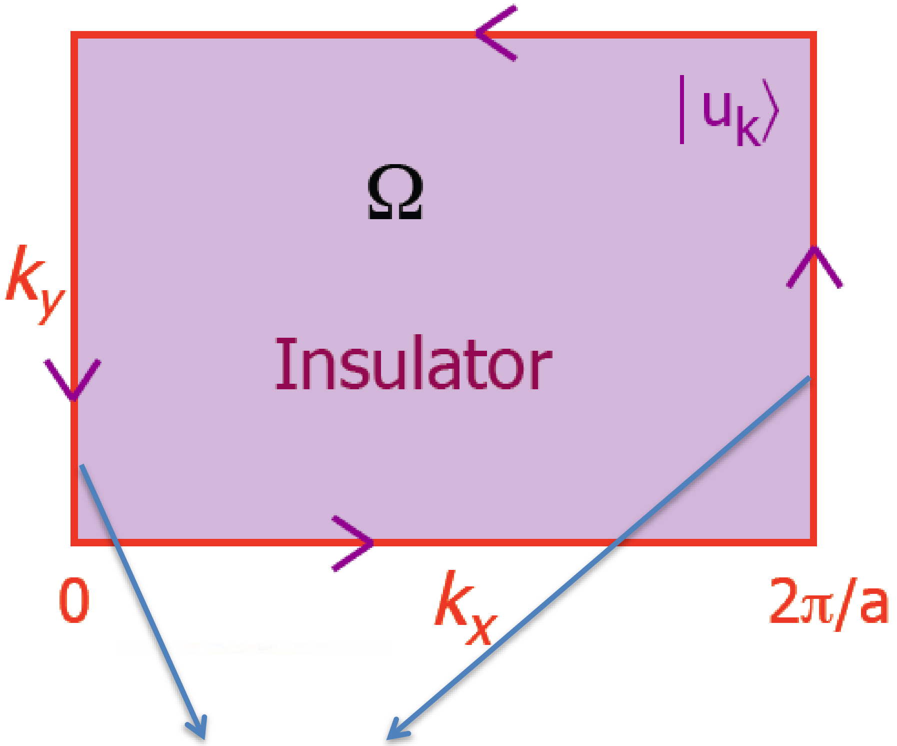

Stokes Theorem: $$\phi = \int d^2k\, \Omega_z(k)$$

Anomalous Hall Conductivity $$\sigma_{xy} = \frac{-e^2}{2\pi\hbar}\phi$$

Karplus & Luttinger; Sundaram & Niu

# Geometry and Topology of Electrons in a Crystal

Ref. D. J. Thouless, M. Kohmoto, M. P. Nightingale, and M. den Nijs, Quantized Hall conductance in a two-dimensional periodic potential, *Phys. Rev. Lett.* **49**, 405 (1982).




$$\phi = \int d^2k\, \Omega_z(k) = 2\pi C$$

C: Chern Number, integer

$$\sigma_{xy} = -\frac{e^2}{h} C$$

Similarity to *Integer Quantum Hall Effect*

Equivalence between the two edges is broken if C≠0

$$\mathrm{k} = \frac{\pi}{a}, and\ k = -\frac{\pi}{a}$$

Time reversal symmetry has to be broken in Chern TI's.

Quantum Anomalous Hall (Chern) Insulator

# Consequences of Symmetry

Time Reversal Symmetry
$t \rightarrow -t$

$$\Omega(-k) = -\Omega(k)$$

Inversion Symmetry
$(xyz) \rightarrow -(xyz)$

$$\Omega(-k) = \Omega(k)$$

In centrosymmetric, non-magnetic crystals $\Omega(k) = 0!$
Most metals, Si, …

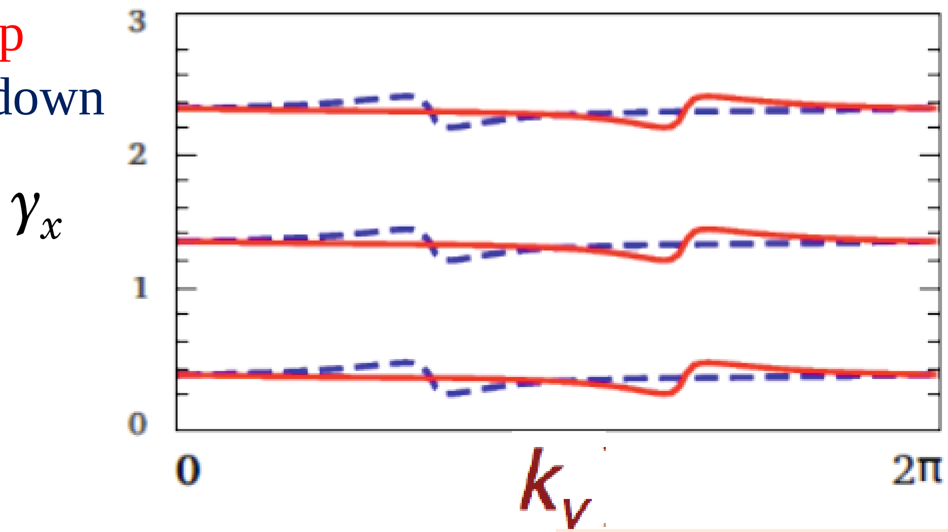
# $Z_2$ Topological Insulator

Chern Number $C = 1$ for spin up electrons,
$= -1$ for spin down electrons

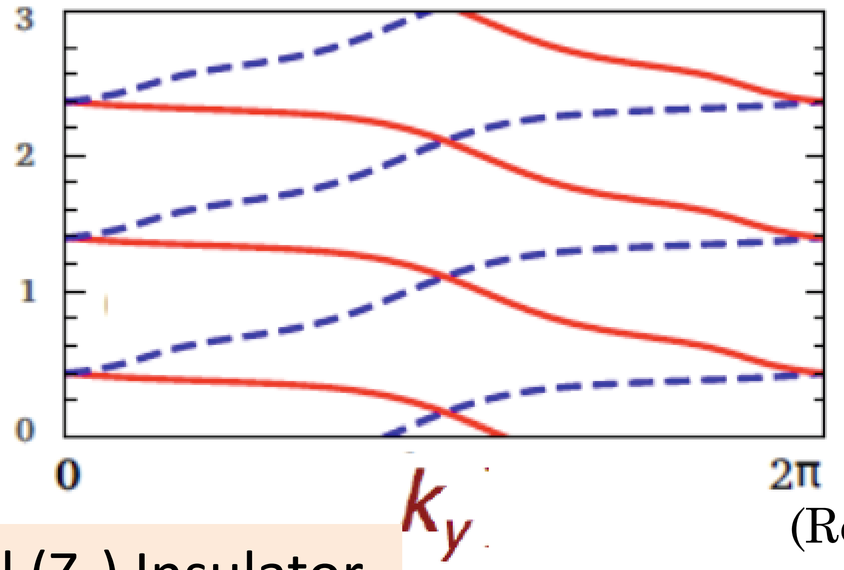
It obeys time reversal symmetry
total $C=0$
$Z_2$ invariant ($C_u$-$C_d$) is odd
needs spin-orbit coupling

Red: Spin up
Blue: Spin down

Normal Insulator

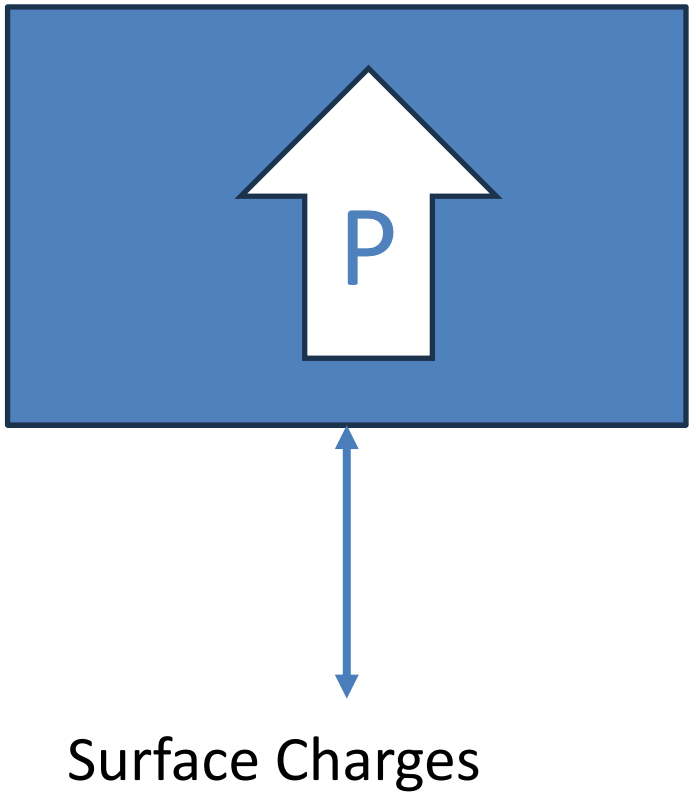



$Z_2$Topological Insulator



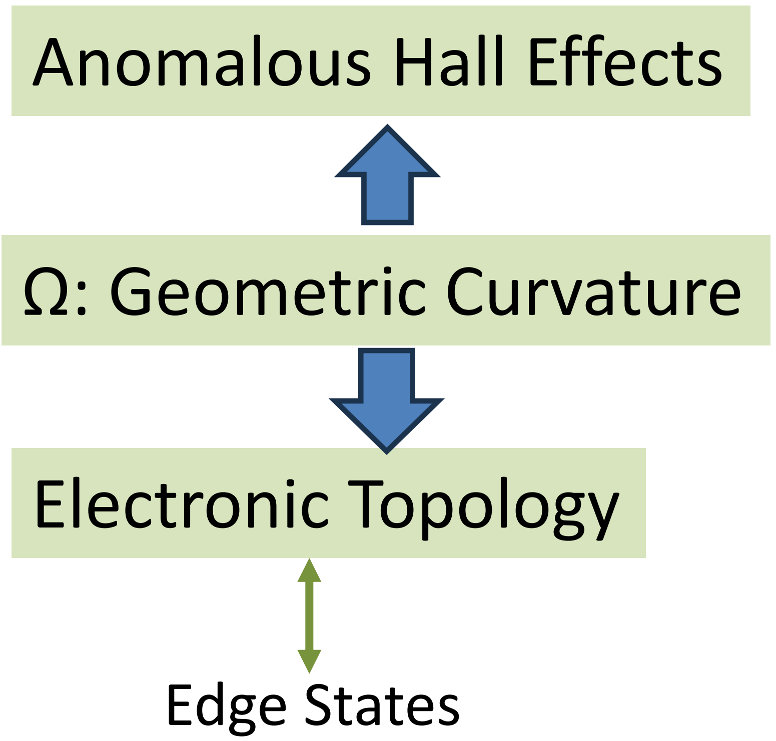

(Ref. A Soluyanov)

Quantum Spin Hall ($Z_2$) Insulator




Measurement of P
Flow of **Electric Current**

Quantum Theory of P:
**Geometric Phase**



# Quantum Geometry & Topology of Electrons: *Emerging Fields*

| Geometric band theory | Electromagnetism (gauge fields) |
|---|---|
| Geometric Pancharatnam-Berry phase, $\gamma(C) = \oint_C d\boldsymbol{k}.A(\boldsymbol{k}) = \int_S d^2\boldsymbol{k}\,\Omega(\boldsymbol{k})$ | Aharonov-Bohm phase, $-\frac{e}{\hbar}\oint d\boldsymbol{r}\,A(\boldsymbol{r}) = -\frac{e}{\hbar}\int d\boldsymbol{S}\,B(\boldsymbol{r})$ |
| Berry connection, $A(\boldsymbol{k}) = \langle u_{\boldsymbol{k}} \vert i\partial/\partial\boldsymbol{k} \vert u_{\boldsymbol{k}}\rangle$ | Vector potential, $A(\boldsymbol{r})$ |
| Berry curvature, $\Omega(\boldsymbol{k}) = \nabla_{\boldsymbol{k}} \times A(\boldsymbol{k})$ | Magnetic field strength, $B(\boldsymbol{r}) = \nabla_{\boldsymbol{r}} \times A(\boldsymbol{r})$ |
| Chern number, $C_n = \oiint_{BZ} d^2\boldsymbol{k}\,\Omega(\boldsymbol{k})\,/(2\pi)$ = integer | Dirac's monopole charge, $\oint_S d^2\boldsymbol{r}\,B(\boldsymbol{r}) = integer\,\frac{e}{h}$ |

# Consequences of Symmetry

Time Reversal Symmetry
$t \rightarrow -t$

$$\Omega(-k) = -\Omega(k)$$

Inversion Symmetry
$(xyz) \rightarrow -(xyz)$

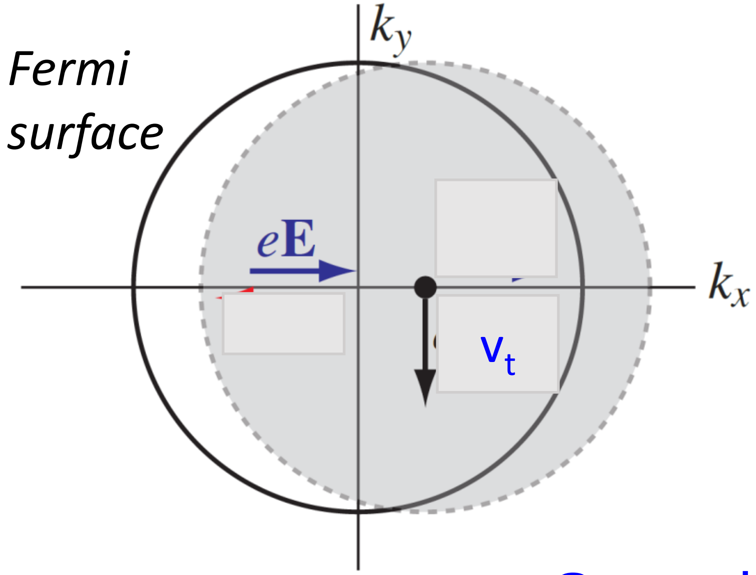
$$\Omega(-k) = \Omega(k)$$

In centrosymmetric, non-magnetic crystals $\Omega(k) = 0!$
Most metals, Si, …

# Nonlinear Hall effect

Anomalous <*linear*> Hall Effect $\sigma_{xy}$: **Broken time-reversal symmetry**

$$\boldsymbol{\Omega}(-\boldsymbol{k}) = -\boldsymbol{\Omega}(\boldsymbol{k})$$




Applied E-field lowers the inversion symmetry

Shift in the Fermi surface: asymmetry in occupied states

First moment of $\boldsymbol{\Omega}(\boldsymbol{k})$ can be nonzero.

Theoretical Prediction

Second order non-linear Hall current: $j_y \propto E_x^2 . D$

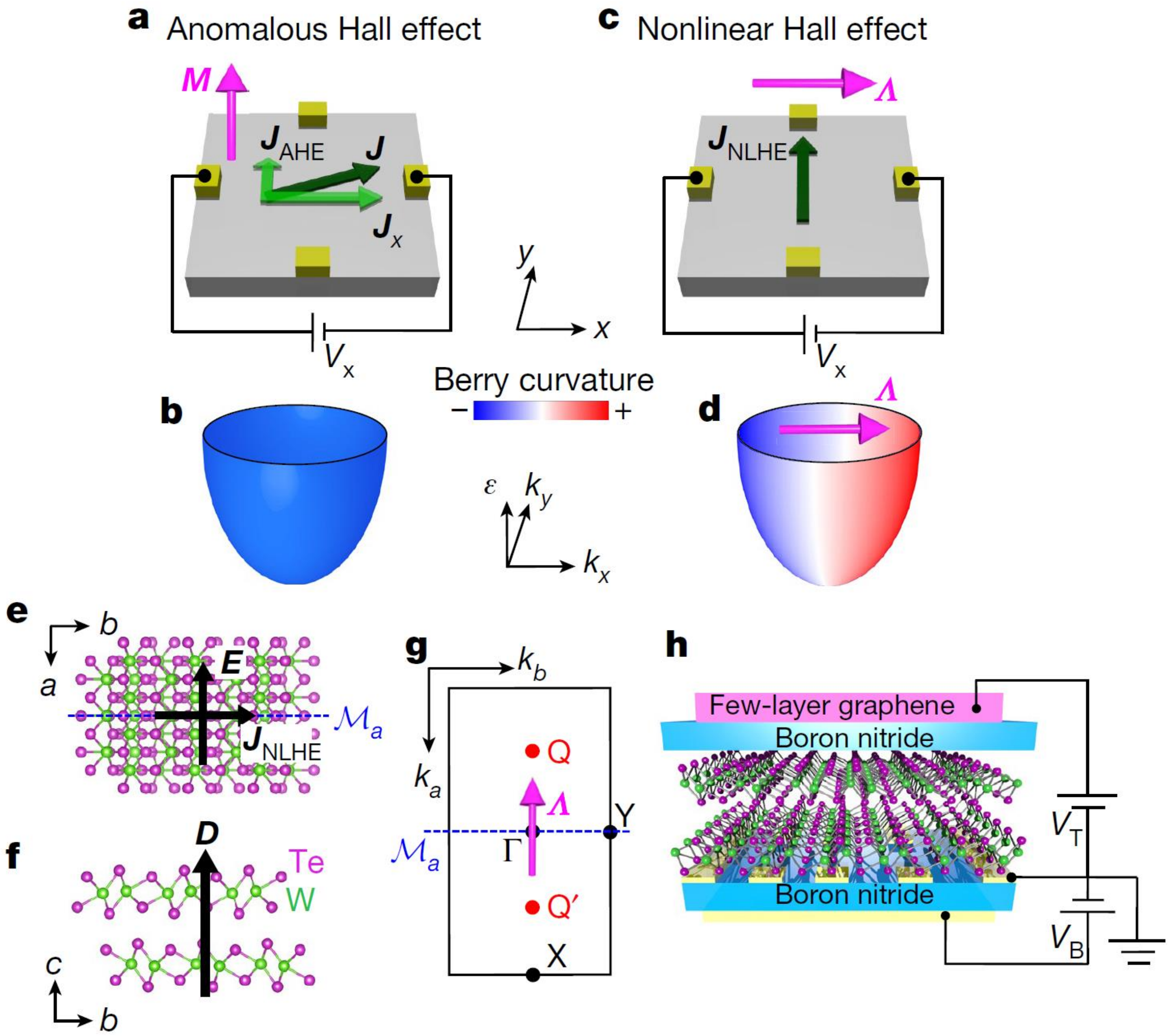
where $D$ is the first moment of Berry curvature (Berry curvature dipole)

$$D = \iint f(\varepsilon_{\boldsymbol{k}}) \left[ \frac{\partial}{\partial k_x} \Omega_z(\boldsymbol{k}) \right] = -\frac{1}{\hbar} \iint v_x(\boldsymbol{k}) \left[ \frac{\partial}{\partial \varepsilon_k} f(\varepsilon_{\boldsymbol{k}}) \right] \Omega_z(\boldsymbol{k})$$

Generates w=0 rectification or 2w (SHG) Hall signal for $E_x$(w)

Ref. I. Sodemann and L. Fu, Quantum Nonlinear Hall Effect induced by Berry Curvature Dipole in Time-Reversal Invariant Materials, *Phys. Rev. Lett.* **115**, 216806 (2015).

# Nonlinear Hall effect




Demonstrated experimentally
in noncentrosymmetric
$T_d$-$WTe_2$ type-II Weyl semimetal
$\Lambda = D \neq 0$

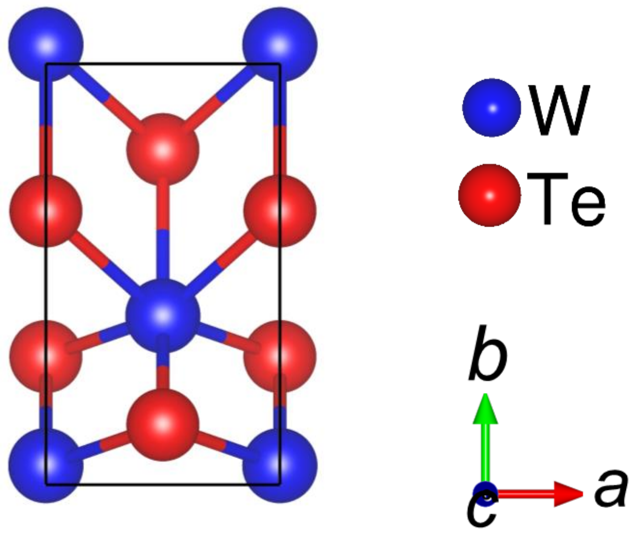
Q Ma et al, Nature 565, 337 (2019)

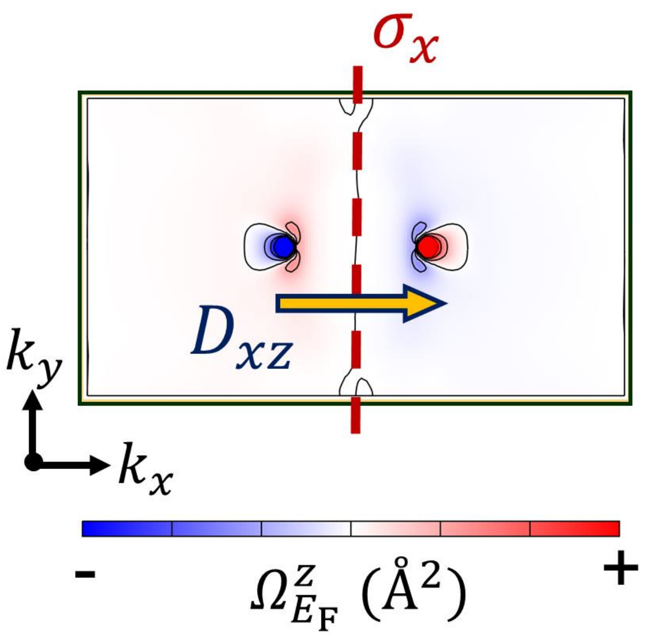
# Nonlinear Hall effect

Time reversal symmetry ✓

Band-gap should be *small*: $\Omega \neq 0$

Crystal structural symmetry: **Low**



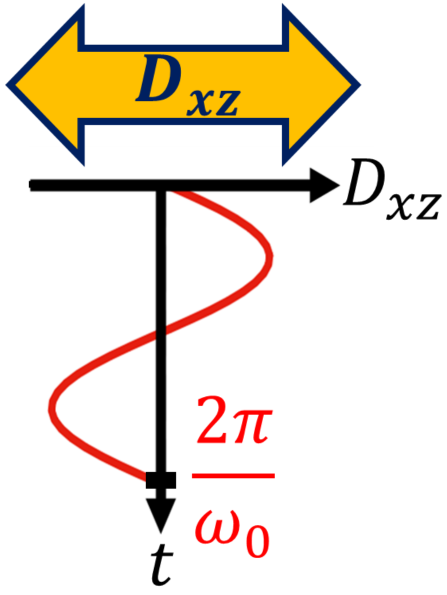

Bhuvaneswari, Deshmukh, Waghmare (2024)

$WTe_2$ Monolayer:

low-symmetry structure (only $\sigma_x$),

**Our work: Crystal Structural Symmetry can be *dynamically* lowered!**

# Dynamical Lowering of Crystal Symmetry




Dynamical Excitations:

Vibration of a lattice lowers its symmetry (function of t): induce oscillations in the quantum geometry of electrons

If $\partial D/\partial u \neq 0$

where $u$ is the amplitude of vibrational mode at $w$=$w_0$

Frequency-dependent non-linear Hall current proposed in the work:

$$j_y(\omega) \propto E_x^2 \left\{ 2\boldsymbol{D}[\delta(\omega - 2\omega_{AC}) + \delta(\omega)] + u_0 \left.\frac{\partial \boldsymbol{D}}{\partial \boldsymbol{u}}\right|_{u=0} \begin{bmatrix} \delta(\omega - (\omega_0 + 2\omega_{AC})) + \\ \delta(\omega - |\omega_0 - 2\omega_{AC}|) + \\ 2\delta(\omega - \omega_0) \end{bmatrix} \right\}$$

Even a centrosymmetric, non-magnetic material that leads to vanishing $\Omega$:

has nontrivial $j_y(\omega)$ through $\frac{\partial D}{\partial u}$

**Present Work:**

*First-principles* Theory and Simulations

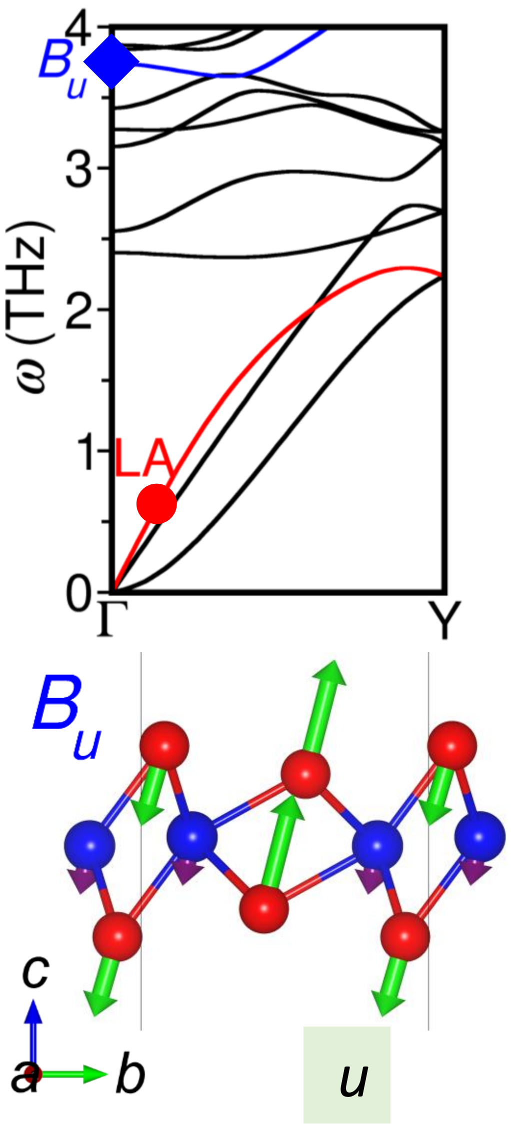
Use Density Functional Theory for Quantum motion of e

Obtain (a) electronic structure

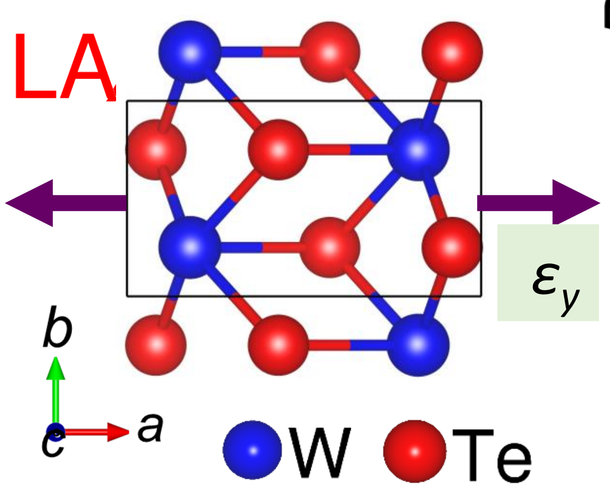
(b) interatomic interactions (hence phonons)

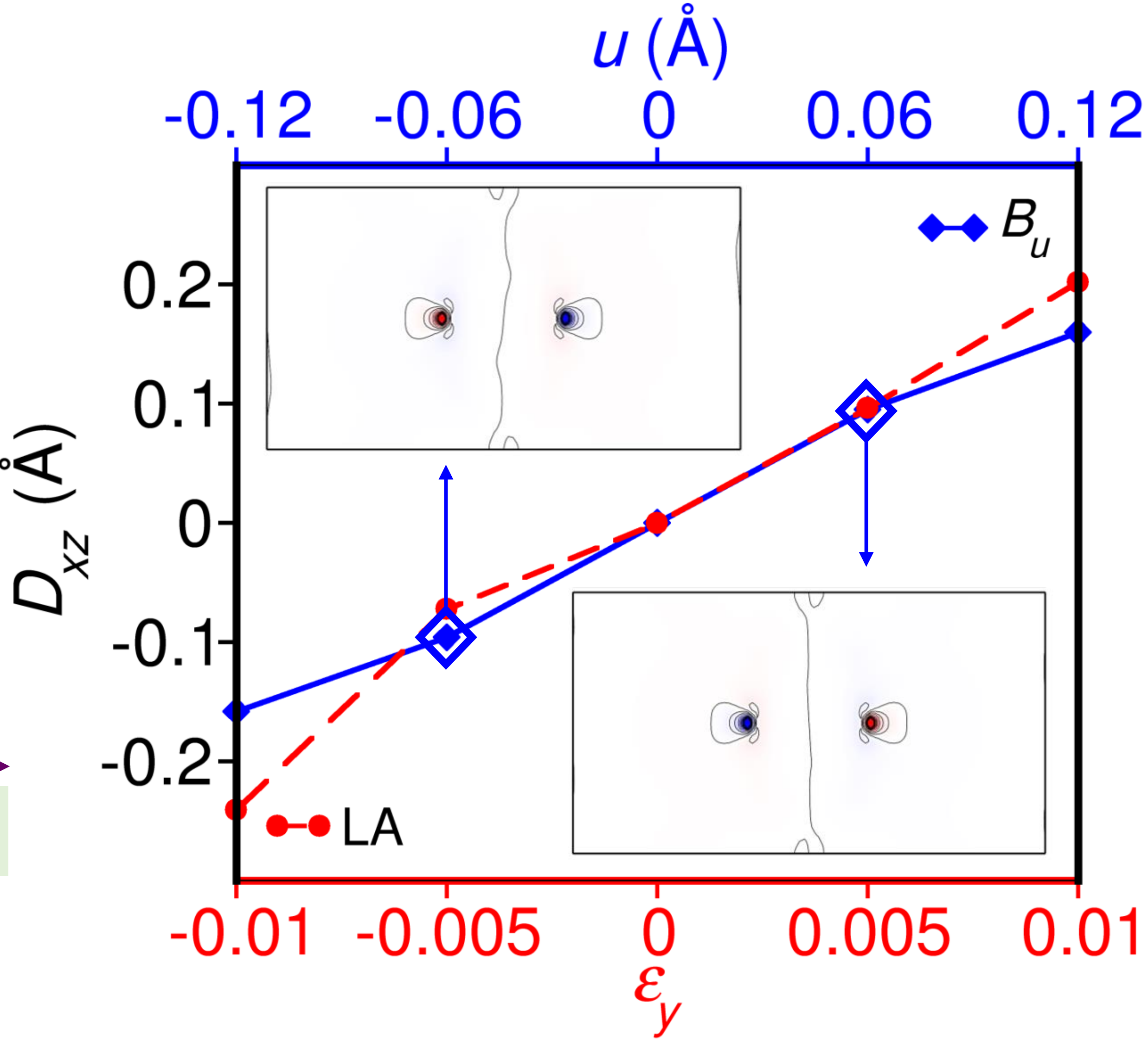
from scratch (no experimental input).

# GQuES-active Modes of Centrosymmetric $T'$-$WTe_2$ Monolayer



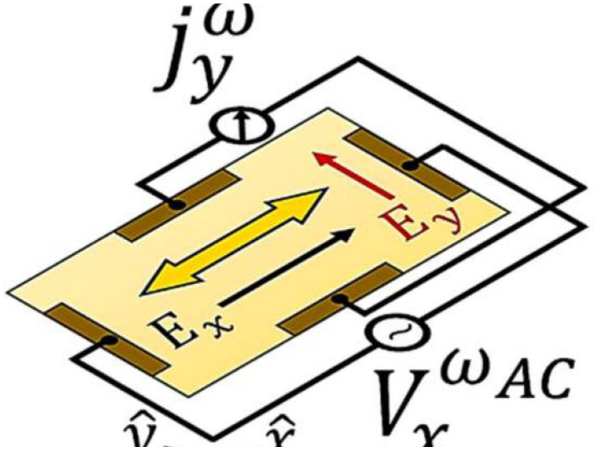

GQuES-active $B_u$ and LA mode *dynamically* lower the inversion symmetry of $T'$-$WTe_2$ → $D \neq 0$ with distortions

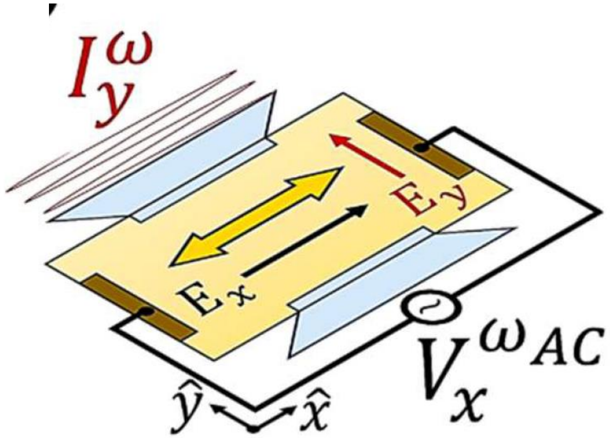


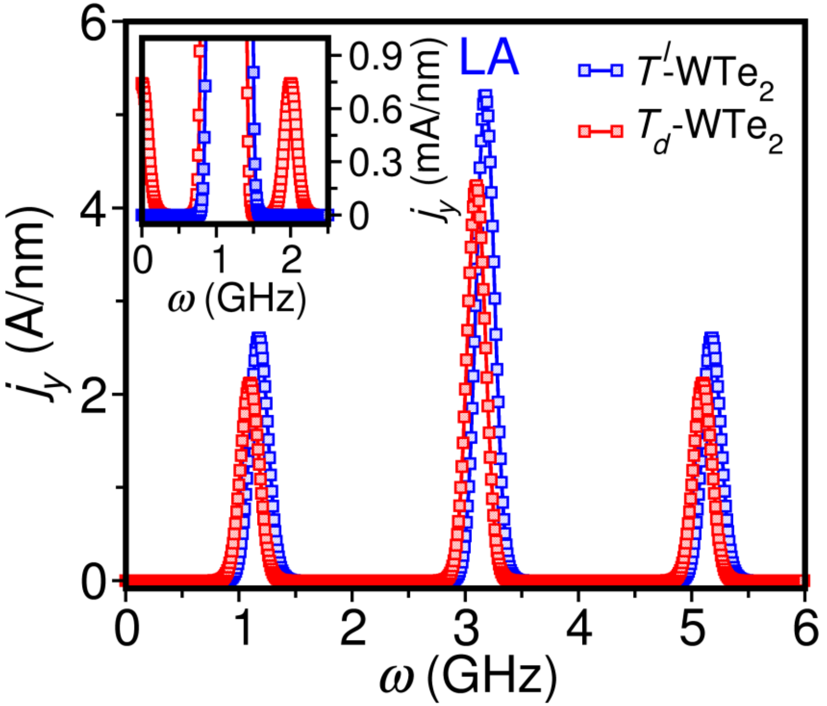
# GQuES-spectra of $WTe_2$ Monolayers

$\omega_{AC}$=1 GHz, GQuES-spectra of centrosymmetric $T'$-$WTe_2$ and non-centrosymmetric $T_d$-$WTe_2$ monolayer

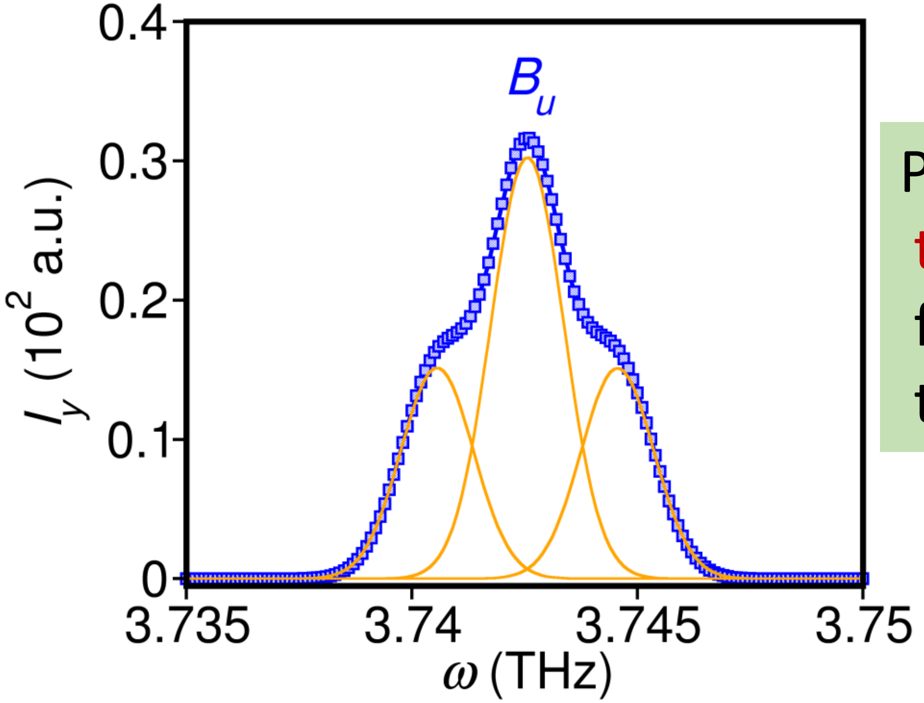



Similarity with IR and Raman spectroscopies




Strain mode (4 μ sample)




Optic Mode

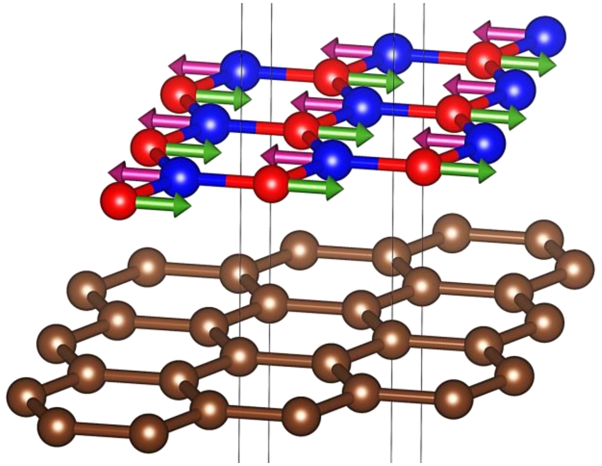
Possibility to translate siganls from GHz to THz

Our idea of dynamical lowering of symmetry works:
Introduces a new type of spectroscopy

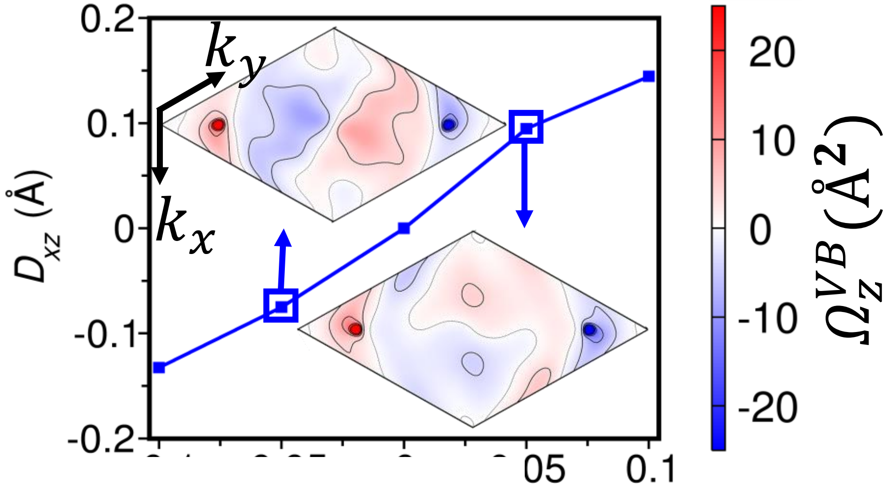
However, the ideas appear still ***restrictive*** to narrow band gap crystals… and those which have the potential of nontrivial quantum geometry!

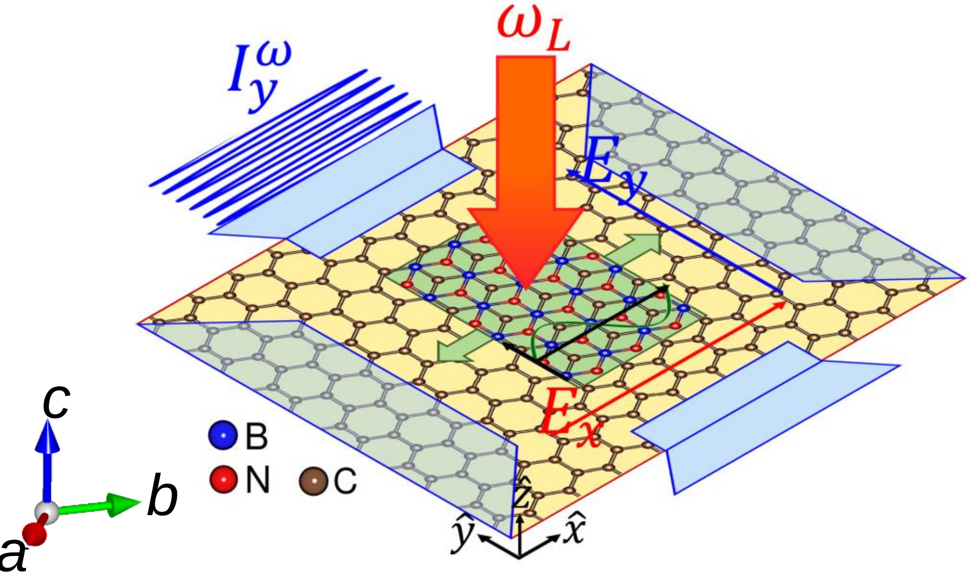
We overcome this by interfacing an ***inert*** material
(e.g. large band-gap h-BN)
with one with a narrow gap or nontrivial quantum geometry!
(e.g. graphene)

# Substrate-induced GQuES activity in aligned gr-hBN

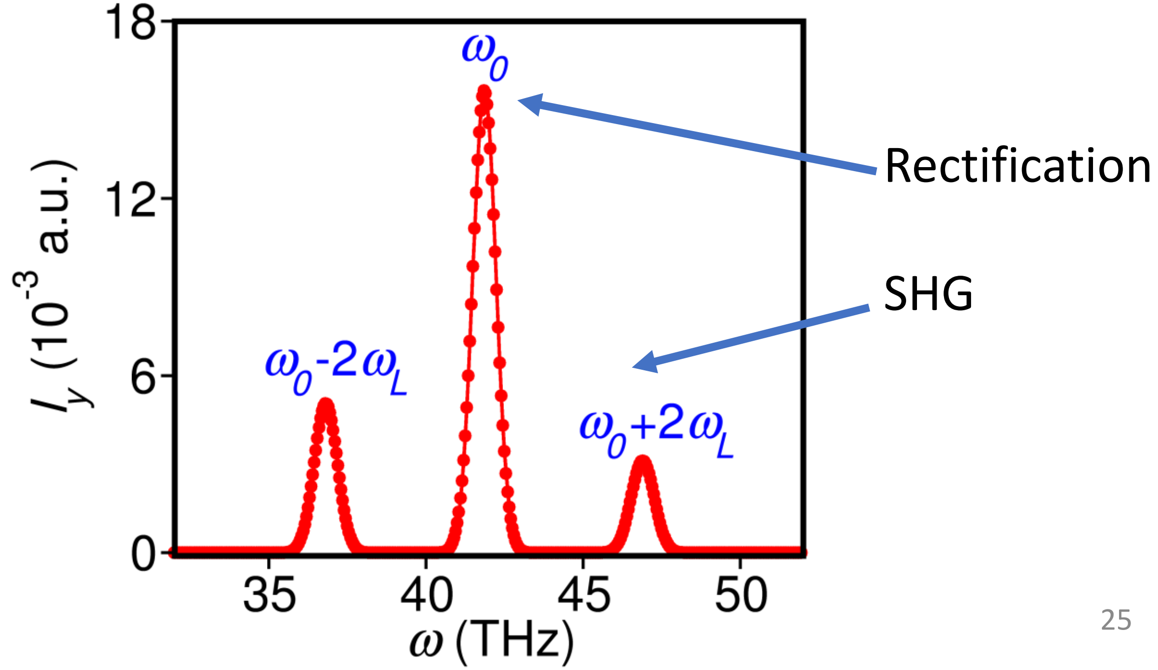


*E* mode of aligned hBN dynamically lowers the $C_{3z}$ symmetry and induces $D \neq 0$

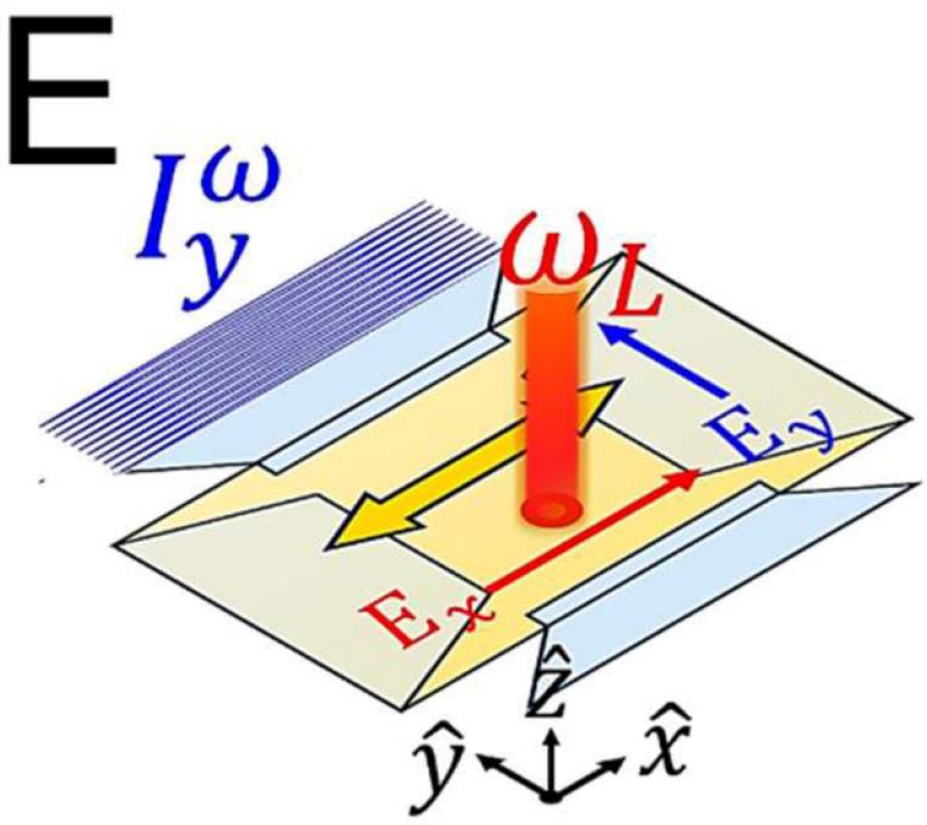



LASER frequency $\omega_L$=2.52 THz
($\lambda_L$=118.9 μm)



# GQuES spectrum of $WTe_2$

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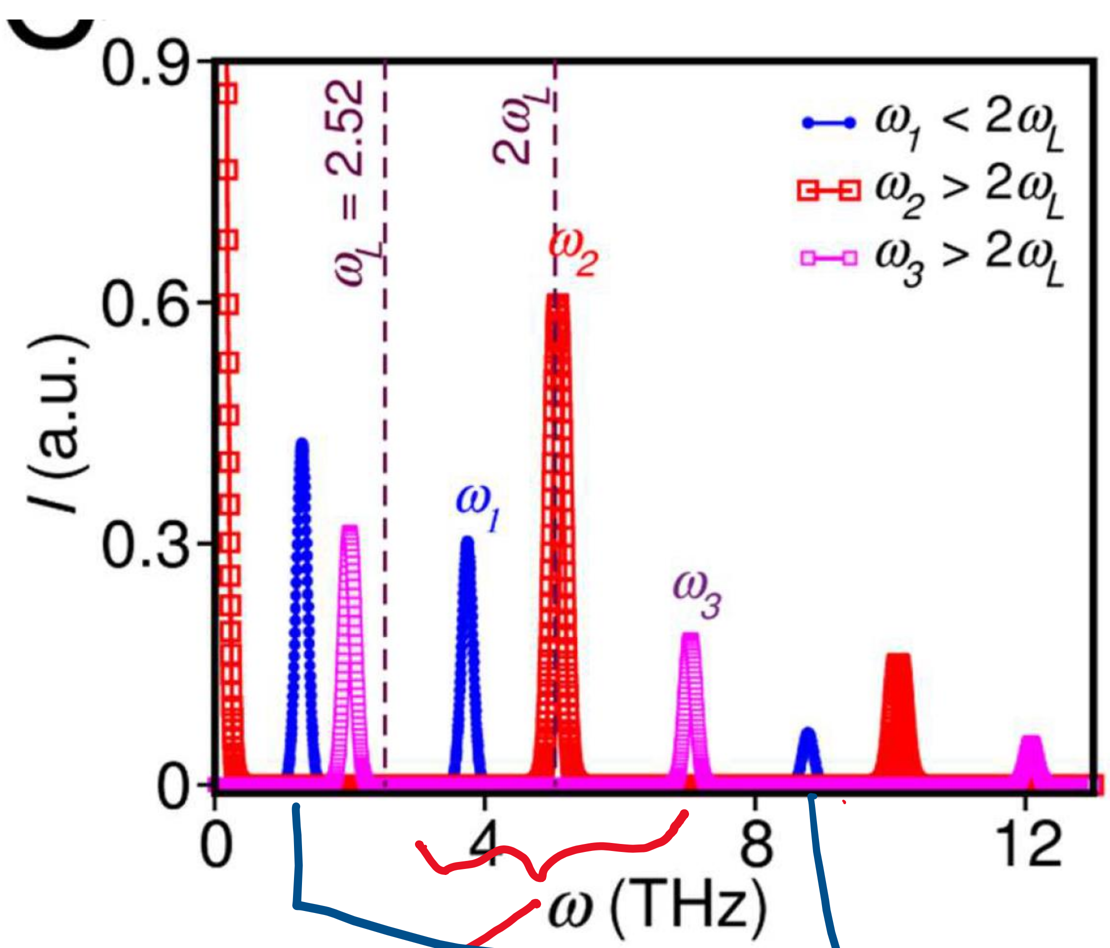
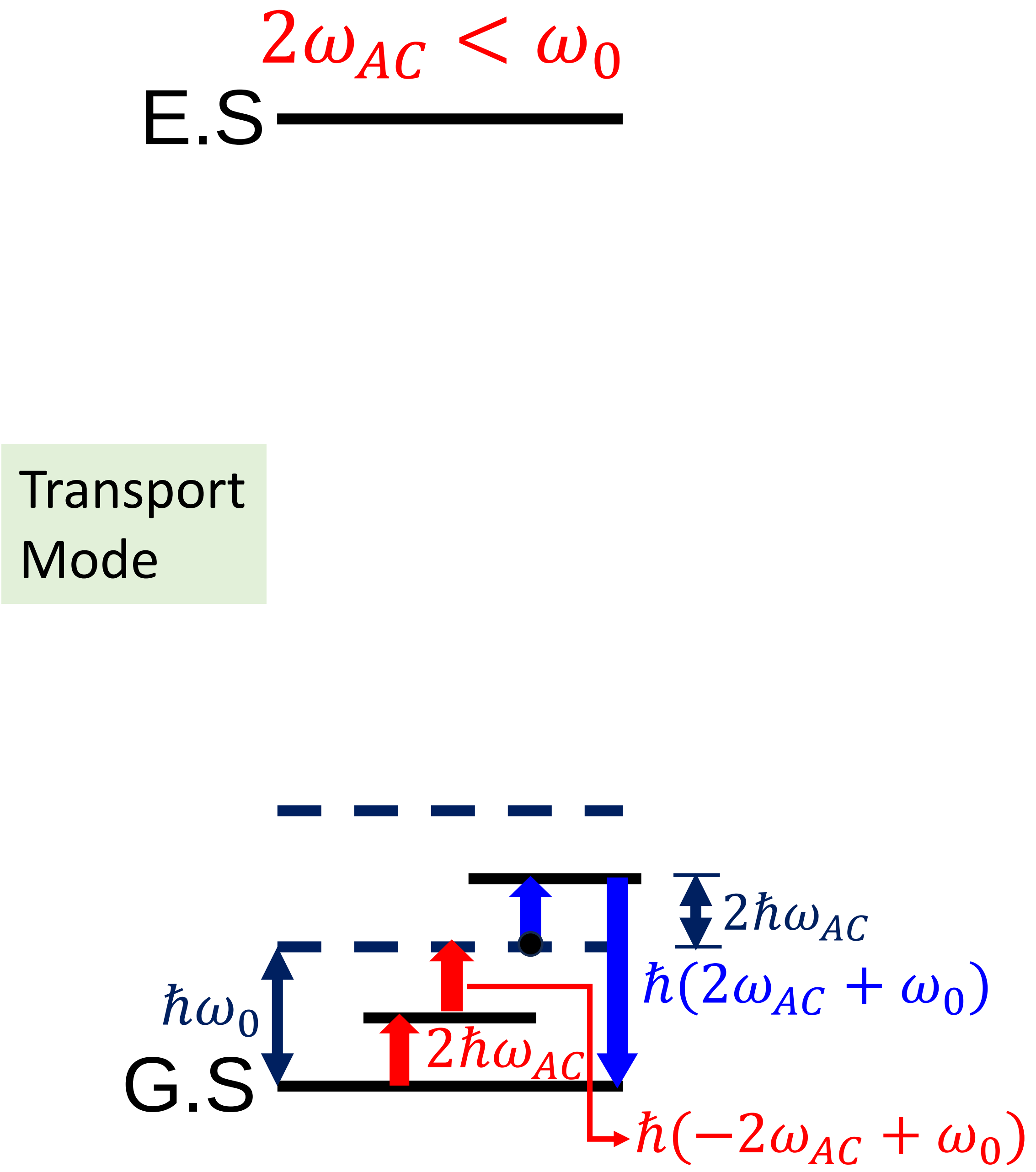
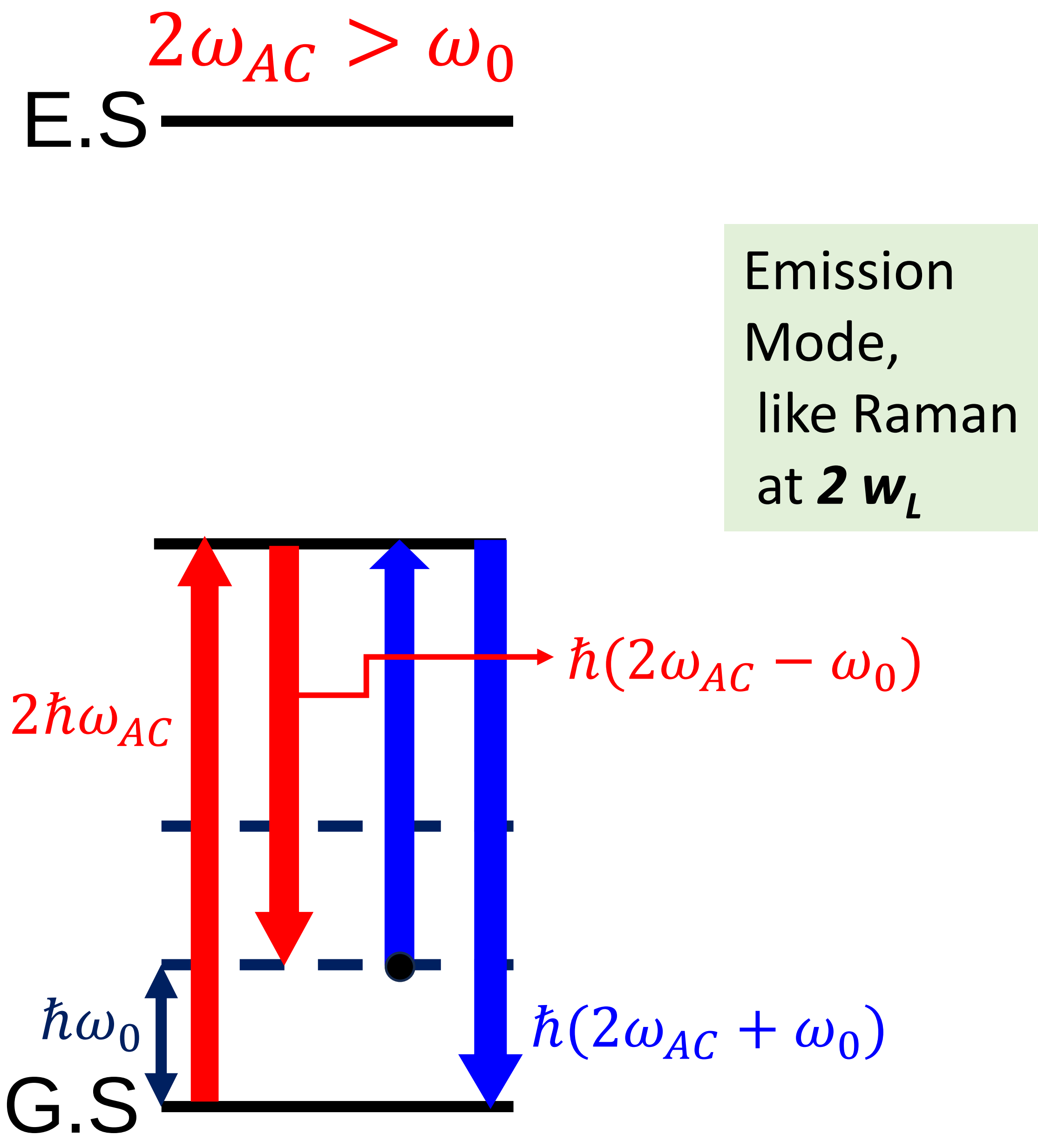


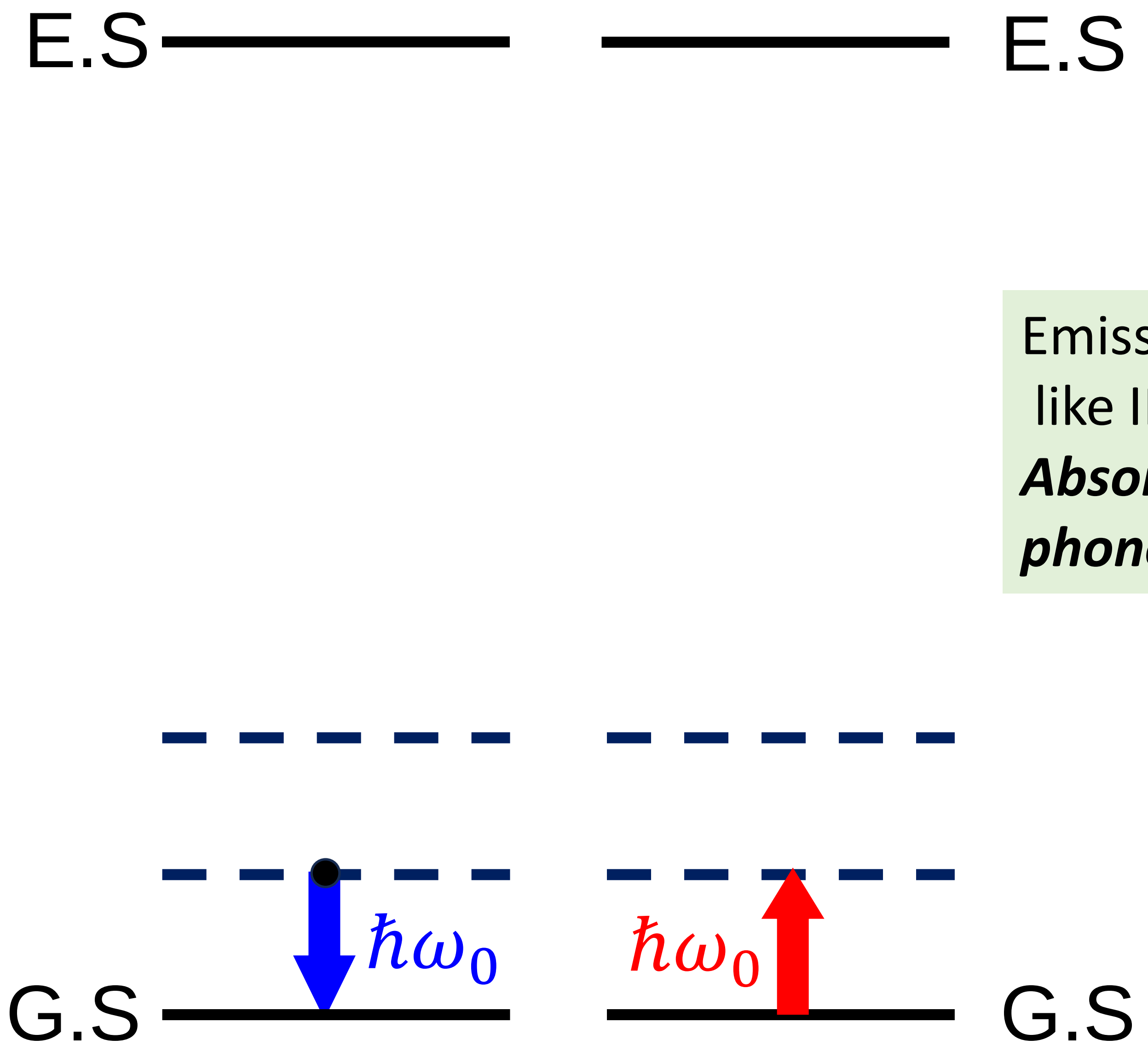

Similarity to IR and
Raman ($w_1$ Stokes and Antistokes centered at 2 $w_L$)

# Quantum Picture of GQuES



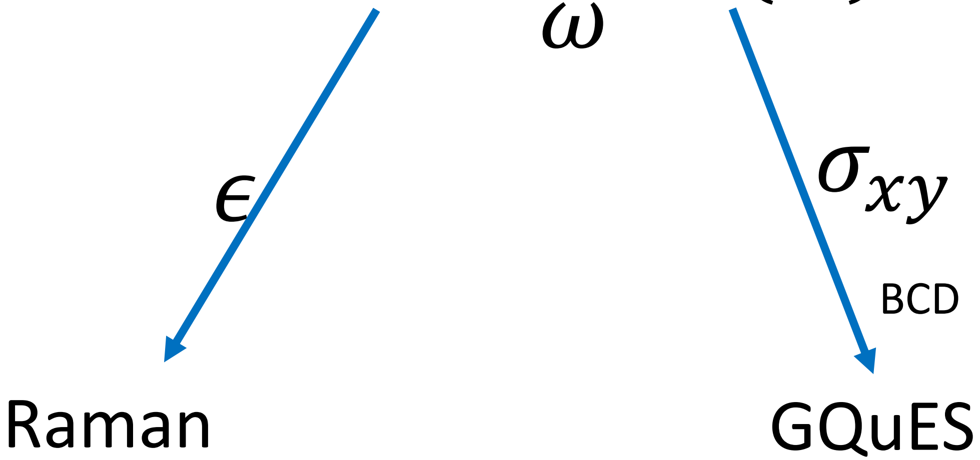



# Quantum Picture of GQuES (Rectification)




Emission Mode,
like IR:
***Absorption and emission of phonon at $w_0$***

$$\epsilon + \frac{4\pi}{\omega} i\sigma(\omega)$$



# Summary (Part I)

- Introduced **GQuES vibrational spectroscopy**: transport and emission modes
  Combines capabilities of IR, Raman and Brillouin

- Can be generalized to *other dynamical excitations* (eg magnon, plasmon)

- Quantum Geometry ideas applied to wider set of systems, including 3D crystals

R Bhuvaneswari, M M Deshmukh and U V Waghmare arXiv: 2403.05872 (2024)

Physical Review B 110, 014305 (2024)

# Introduce using First-principles Theory

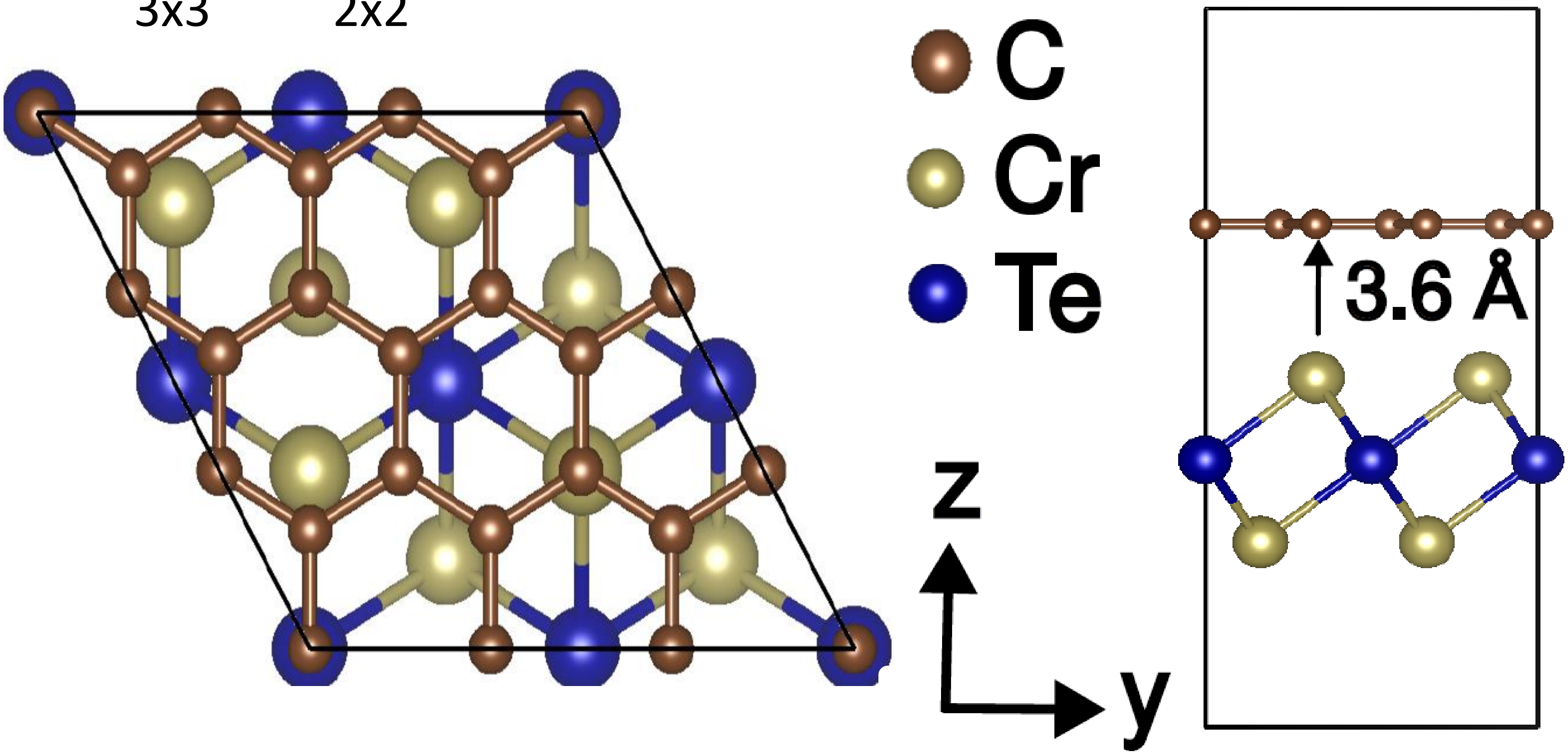
## II. Anomalous Hall Transistor

*Graphene:CrTe*$_2$ heterostructure

S Menon and U V Waghmare, Nanoscale (under review);
Application for Indian Patent

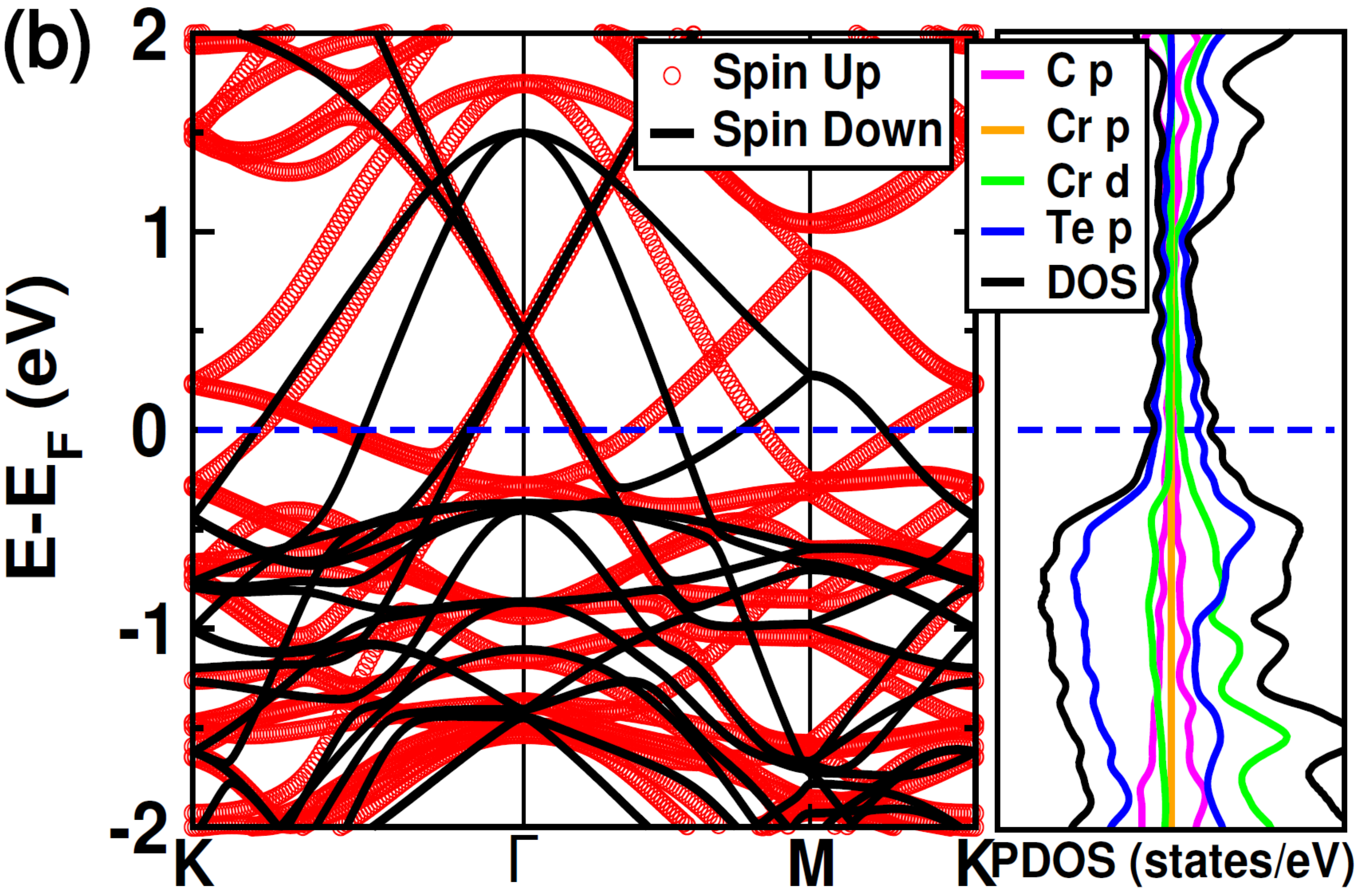
# *Graphene:$CrTe_2$* heterostructure

3x3 2x2



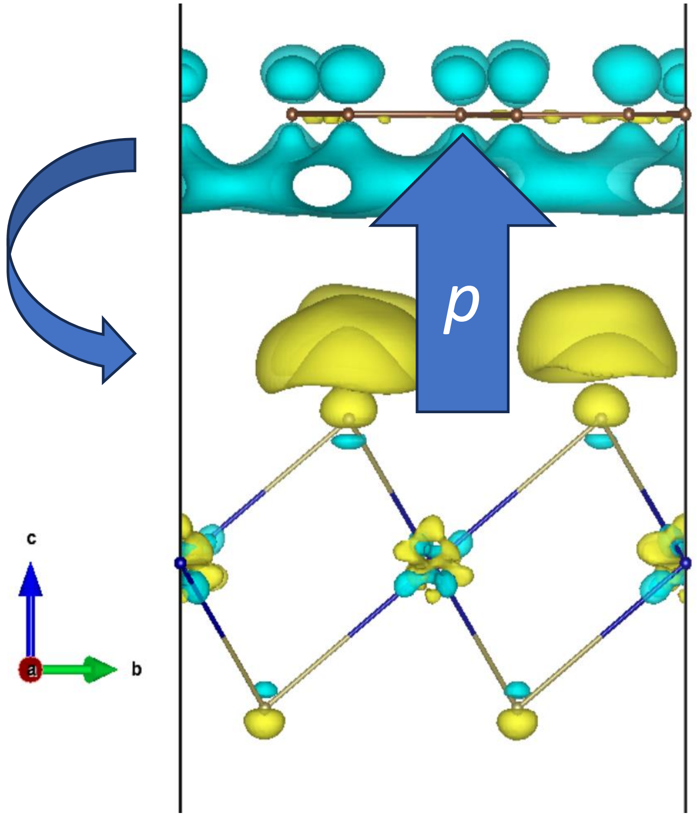

Model Structure: lattice matched within 0.5 %

# Electronic Structure




Almost rigid shift in energy

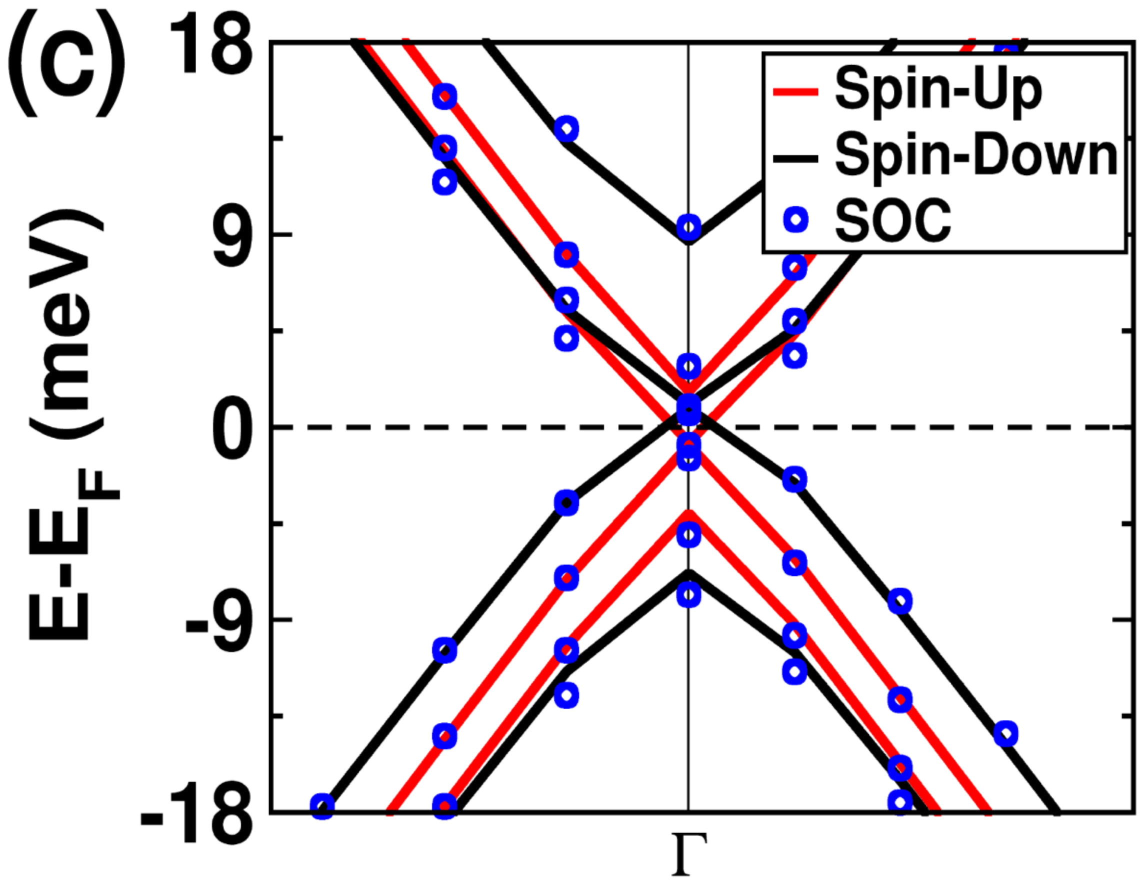
e transfer from G to $CrTe_2$




$P = 1.69\ \mu C/cm^2$

- $P$ couples with electric field $E_z$
- $E_z$ control on charge carrier concentration

# Electronic Structure near the Dirac Points of Graphene




Crystal Field:

Split bands at K and K'

Mix bands at K with bands at K'

*6 meV*

Exchange interaction:

Split spin-degeneracy

*3 meV*

8 bands (folded to Γ point)

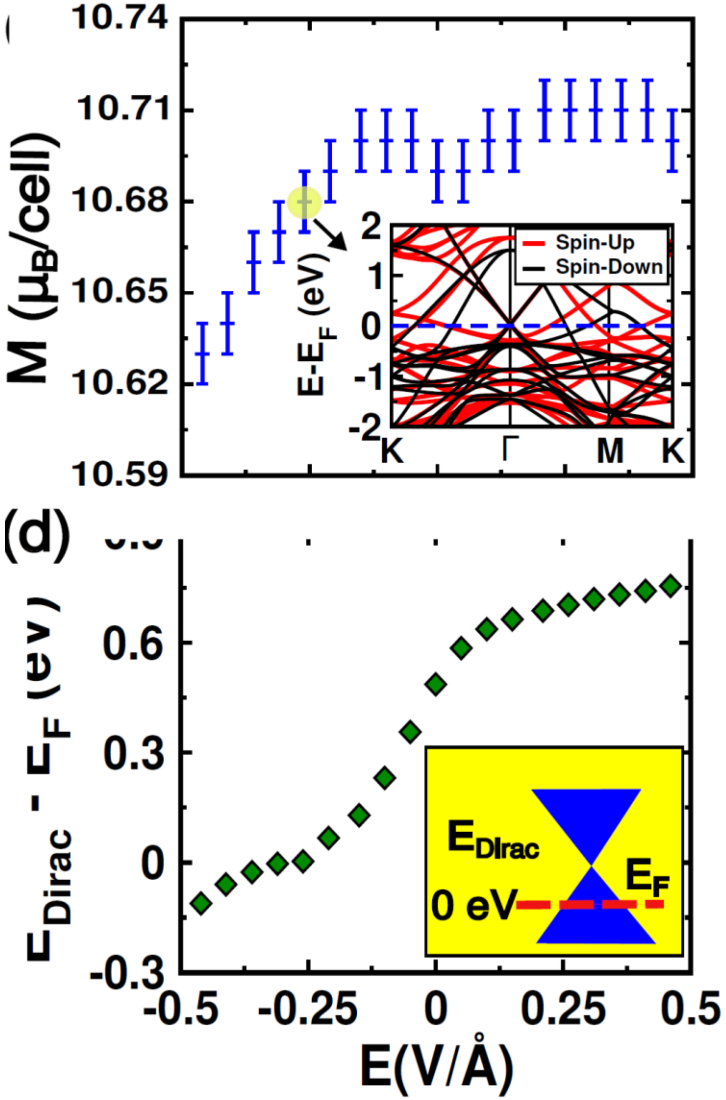
Splitting due to SOC is weaker ≤ 1 meV

4-band model (per spin)

$$H_o = \hbar v_f \begin{pmatrix} (\vec{k}.\vec{\sigma})^* & 0 \\ 0 & (\vec{k}.\vec{\sigma}) \end{pmatrix} \qquad H_c = \frac{1}{|\vec{k}|} \begin{pmatrix} (\vec{k}.\vec{\sigma})^* & \delta \\ \delta^\dagger & (\vec{k}.\vec{\sigma}) \end{pmatrix}, \delta = \begin{pmatrix} 0 & k_- \\ -k_+ & 0 \end{pmatrix}$$

# Broken Time Reversal and Inversion Symmetries:

## Linear Magnetoelectric Effect




$$\alpha_{ij} = \frac{\partial P_i}{\partial H_j} = \mu_0 \frac{\partial M_i}{\partial E_j}$$

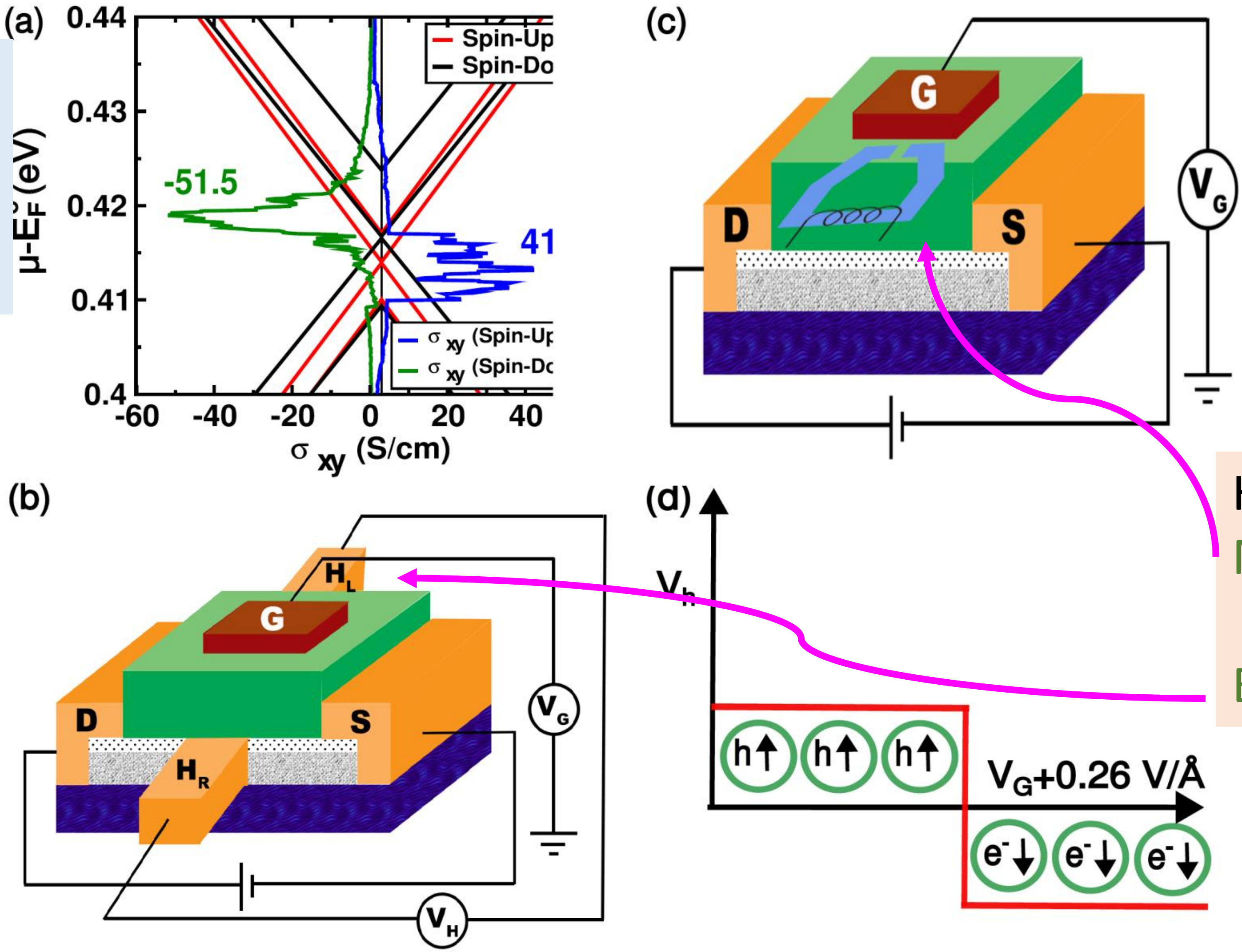
$\alpha$ is $\sim$ 0.80 ps/m,

Compare $Cr_2O_3$ (4.13 ps/m at 298 K)

Asymmetry due to asymmetry in the shift of Dirac point in response to electric field $E_z$

# Graphene:$CrTe_2$ Heterostructure: *Anomalous Hall Transistor*

*Spin-polarized*
Hall conductivity:
Switchable by
 $E_z$




Hall signal readable
Magnetically
 and
Electrically

Menon
 and
Waghmare,
Under Review.

Magnetically and Electrically Readable Memory Device

# Summary (Part II)

## Graphene:$CrTe_2$ Heterostructure

*Gate Field E couples with P: control carrier concentration*

*Emergence of New Functional Properties:*

1. *Linear* Magnetoelectric Effect
2. E-field Switchable Spin-polarized Hall Conductivity <*Graphene*>

Anomalous Hall Transistor

An electrically and magnetically readable memory device

Menon and Waghmare, Under Review (2024).

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