Antiferromagnetic order and magnetoelectricity of charge carriers in quantum wells

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in collaboration with:

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Outline

- Introduction & Motivation
  - basics of the magnetoelectric effect
  - quasi-2D/quantum well structures
- Magnetoelectricity in quasi-2D itinerant-electron para-, ferro-, and antiferromagnets
- Antiferromagnetic order of charge carriers
  - new quantity $\langle \boldsymbol{\tau} \rangle$: AFM analog of spin polarisation $\langle \boldsymbol{\sigma} \rangle$
- multi-band $\mathbf{k}\cdot\mathbf{p}$ model for diamond AFM
- Conclusions

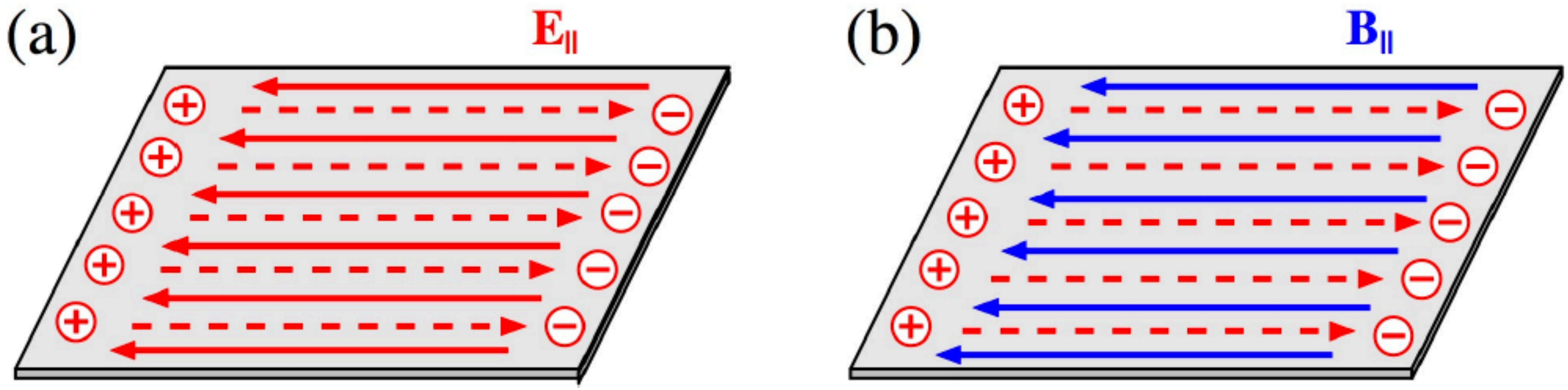
Introduction:
Magnetoelectric effect

Magnetoelectric effect

- magnetoelectric media: unusual electromagnetic response   O'Dell, *The Electrodynamics of Magneto-electric Media* (1970)

$$\mathcal{P} = \epsilon_0\, \underline{\chi}_{\mathcal{E}} \cdot \boldsymbol{\mathcal{E}} + \underline{\alpha} \cdot \boldsymbol{\mathcal{B}}$$

$$\mathcal{M} = \mu_0\, \underline{\alpha}^{\mathrm{T}} \cdot \boldsymbol{\mathcal{E}} + \mu_0^{-1}\, \underline{\chi}_{\mathcal{B}} \cdot \boldsymbol{\mathcal{B}}$$



Magnetoelectric effect

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$$\mathcal{P} = \epsilon_0 \underline{\chi}_{\mathcal{E}} \cdot \mathcal{E} + \underline{\alpha} \cdot \mathcal{B}$$

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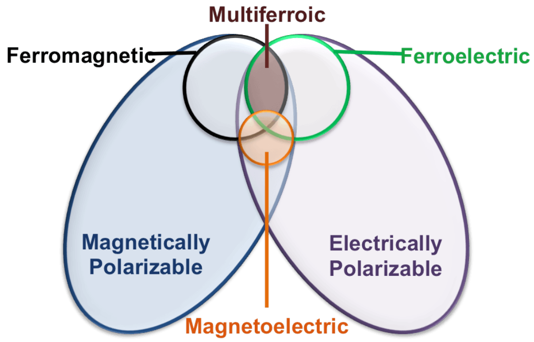


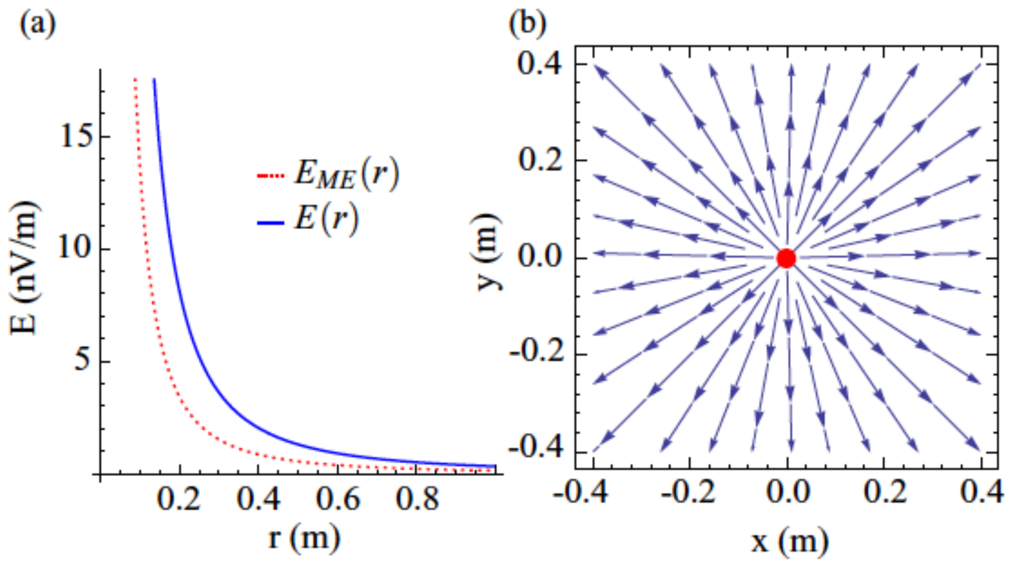
- magnetoelectric tensor: $\alpha_{ij} = a\,\delta_{ij} + \tilde{\tau}_k\,\varepsilon_{ijk} + q_{ij}$
  - can be separated into the sum of a uniform (axion) part, a toroidal part, and a quadrupolar part
  - requires broken inversion and time-reversal symmetry
  - typically in complex materials: multiferroics Fiebig et al, Nat Rev Mat (2016), topological insulators Qi & Zhang, PRB (2008)

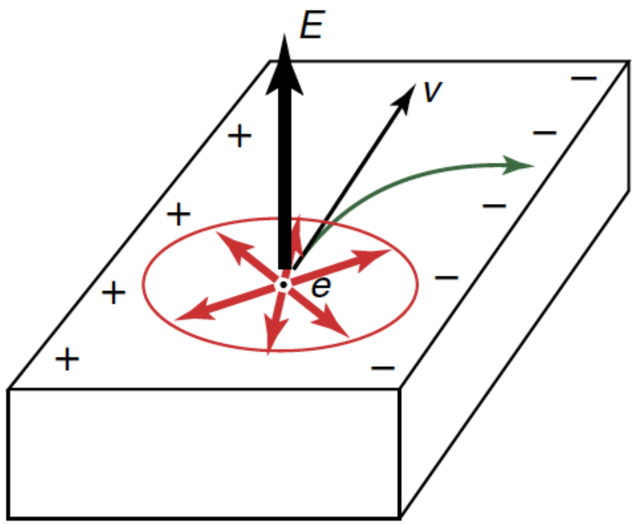
Unusual electromagnetism in magnetoelectrics

- electromagnetism turned on its head/perfectly dual!

  Fechner et al., PRB (2014); Khomskii, Nat Comms (2014)
  - electric charge generates magnetic-monopole field
  - thus magnetic fields accelerate an electric charge
  - electric Hall effect, magneto-photovoltaic effect, ...

Fechner et al., PRB **89**, 184415 (2014)





Khomskii, Nat Comms **5**, 4793 (2014)

Questions

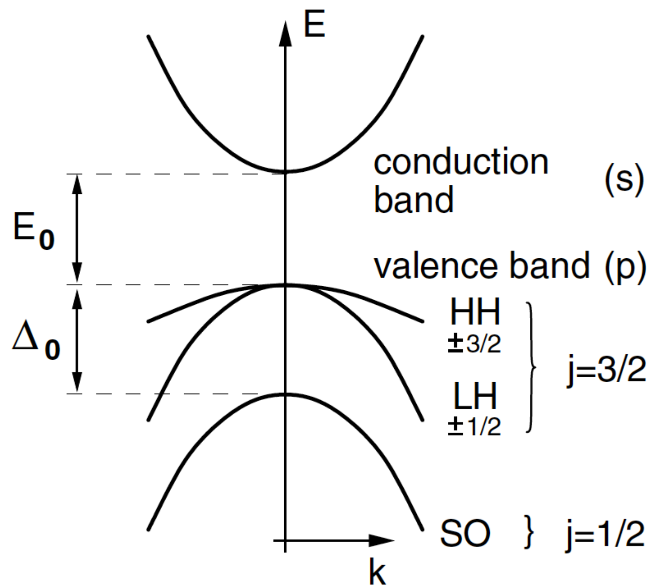
- Magnetoelectricity in more ordinary materials?
  - especially in (low-D) conductors ⇒ spintronics!
  - equilibrium magnetoelectric responses ⇒ alternative to dissipative Edelstein effect!
- How can magnetoelectricity be manipulated?
  - multiferroics: only tuneable by temperature!
  - topological insulators: quantized!
- Can we obtain a unified microscopic description for both of the dual magnetoelectric responses?
  - most works focused either on magnetically induced polarisation or electrically induced magnetisation!

Introduction:
Quantum wells realised in semiconductor heterostructures

Quantum wells: Quasi-2D electron systems

- band-gap engineering in semiconductor hetero-structures realises tunable quantum-well potential
  - perpendicular electric field shifts quasi-2D bound state
  - magnetoelectricity: in-plane magnetic field does same!

Image credit: Roland Winkler



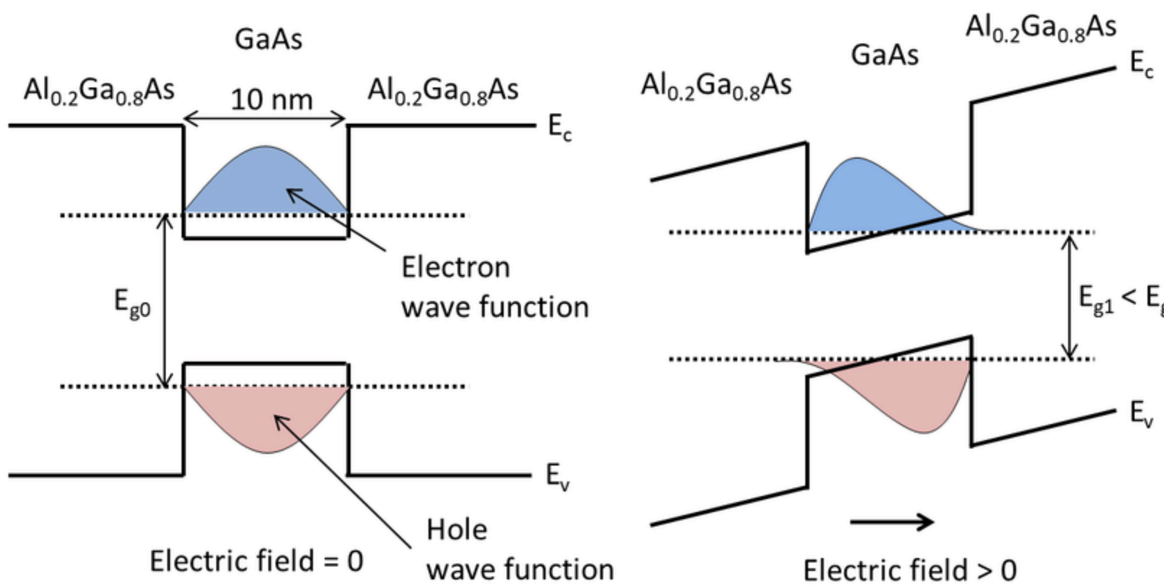
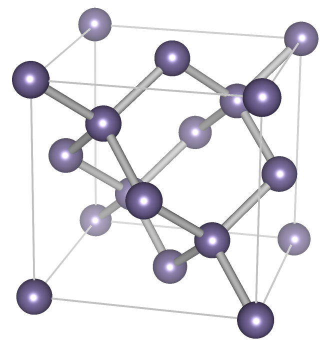
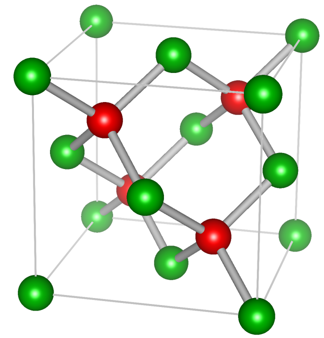


Image credit: Vignesh Devaki Murugesan (U Ulm)

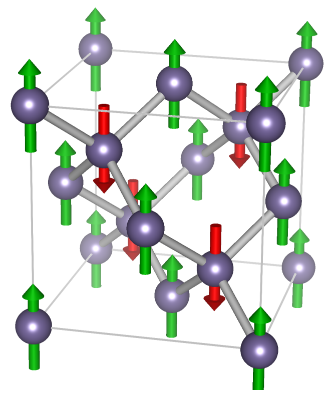
Bulk materials: Variants of diamond structure



- diamond structure: Si, Ge, …
  - inversion & time-reversal symmetric
  - not magnetoelectric



- zincblende structure: GaAs, InSb, …
  - broken inversion/intact time-reversal symmetry
  - becomes magnetoelectric when magnetised



- diamond antiferromagnet: $CoRh_2O_4$
  - broken inversion & time-reversal symmetry
  - combined inversion*time-reversal symmetric
  - has all ingredients for magnetoelectricity

Electric/magnetic responses in 2D systems

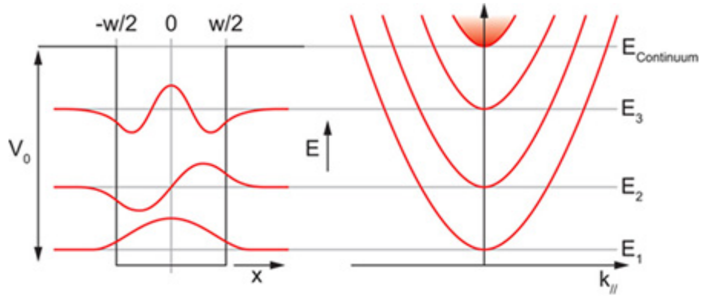


Image credit: Paul Townsend

- solve Schrödinger equation with
  - symmetric confining potential $V(z)$
  - electric potential $e\,\mathcal{E}_z\, z$
  - vector potential $\boldsymbol{\mathcal{A}} = z\,\boldsymbol{\mathcal{B}}_{\|} \times \hat{\mathbf{z}}$
- obtain eigenstates $\Psi_{n\mathbf{k}_\|}(\mathbf{r}) = \dfrac{\mathrm{e}^{i\mathbf{k}_\|\cdot\mathbf{r}}}{2\pi}\,\Phi_{n\mathbf{k}_\|}(z)$
  - subband index n
  - in-plane wave vector $\mathbf{k}_\|$
- perp. polarisation: $\mathcal{P}_z = -\dfrac{e}{w}\sum_n \int \dfrac{d^2k_\|}{(2\pi)^2}\, f(E_{n\mathbf{k}_\|})\,\langle z\rangle_{n\mathbf{k}_\|}$
- in-plane magnetisation:

$$\boldsymbol{\mathcal{M}}_\| = -\frac{e}{w}\sum_n \int \frac{d^2k_\|}{(2\pi)^2}\, f(E_{n\mathbf{k}_\|})\ \hat{\mathbf{z}} \times \langle\{z\,,\mathbf{v}_\|\}\rangle_{n\mathbf{k}_\|}$$

Analytical model for the conduction band

FM quantum well

$$\mathsf{H} = \mathsf{H}_k + V(z) + \mathsf{H}_\mathrm{D} + \mathsf{H}_\mathcal{Z} + e\mathcal{E}_z z$$

$$\mathsf{H}_k = \frac{\hbar^2 k^2}{2m}$$

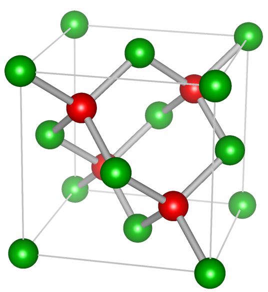
$$\mathsf{H}_\mathrm{D} = d\left(\{k_x, k_y^2 - k_z^2\}\,\sigma_x + \mathrm{c.p.}\right)$$

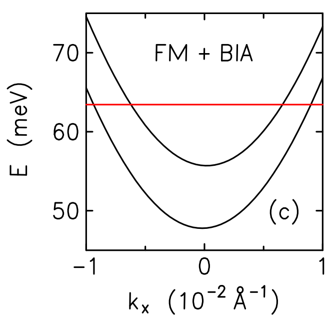
$$\mathsf{H}_\mathcal{Z} = \boldsymbol{\mathcal{Z}} \cdot \boldsymbol{\sigma}$$

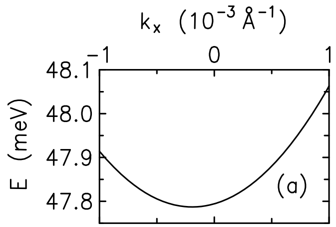
$\boldsymbol{\mathcal{Z}}$… Zeeman/exchange field

$$\mathsf{H} = \frac{\hbar^2 k_z^2}{2m} + V(z) + \frac{\hbar^2}{2m}\left(\mathbf{k}_\parallel - \mathbf{k}_0\right)^2 - \frac{\hbar^2 k_0^2}{2m} + \mathcal{Z}\,\sigma_z + e\mathcal{E}_z z$$

$$\mathbf{k}_0 = \frac{m}{\hbar^2}\, d\, k_z^2 \left[\begin{pmatrix} \cos\varphi_\mathcal{Z} \\ -\sin\varphi_\mathcal{Z} \end{pmatrix}\sigma_z - \begin{pmatrix} \sin\varphi_\mathcal{Z} \\ \cos\varphi_\mathcal{Z} \end{pmatrix}\sigma_x\right]$$







AFM quantum well

$$\mathsf{H} = \mathsf{H}_k + V(z) + \mathsf{H}_\mathcal{N} + e\mathcal{E}_z z$$

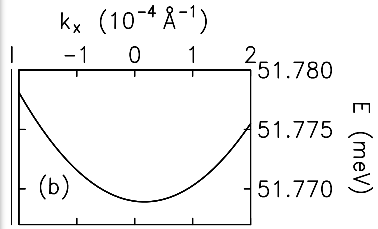
$$\mathsf{H}_k = \frac{\hbar^2 k^2}{2m}$$

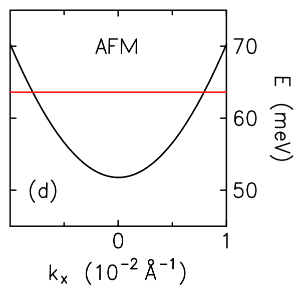
$$\mathsf{H}_\mathcal{N} = \tilde{d}\left(\{k_x, k_y^2 - k_z^2\}\,\mathcal{N}_x + \mathrm{c.p.}\right)$$

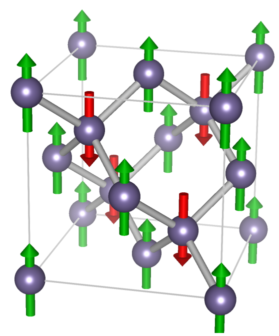
$\boldsymbol{\mathcal{N}} = \boldsymbol{\mathcal{Y}}/\mathcal{Y}$; $\boldsymbol{\mathcal{Y}}$… staggered exchange field

$$\mathsf{H} = \frac{\hbar^2 k_z^2}{2m} + V(z) + \frac{\hbar^2}{2m}\left(\mathbf{k}_\parallel - \mathbf{k}_0\right)^2 - \frac{\hbar^2 k_0^2}{2m} + e\mathcal{E}_z z$$

$$\mathbf{k}_0 = \frac{m}{\hbar^2}\, \tilde{d}\, k_z^2 \begin{pmatrix} \cos\varphi_\mathcal{N} \\ -\sin\varphi_\mathcal{N} \end{pmatrix}$$





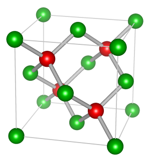


Magnetoelectric response I:

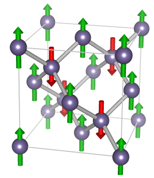
Electric-field-induced in-plane magnetisation

Analytical results for conduction band

- perturbative treatment of $\mathcal{E}_z$ yields results for the in-plane magnetisation:

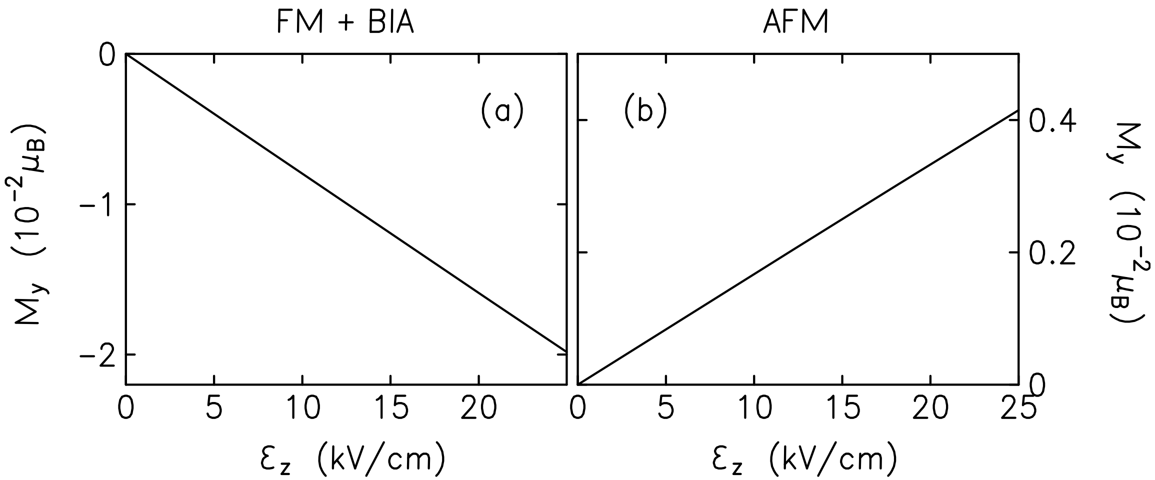


$$\boldsymbol{\mathcal{M}}_{\parallel} = \mathcal{M}_0 \; e\mathcal{E}_z w \; \lambda_d \; \xi(\mathcal{Z}) \begin{pmatrix} \sin\varphi_{\mathcal{Z}} \\ \cos\varphi_{\mathcal{Z}} \end{pmatrix} \quad \Bigg| \quad \boldsymbol{\mathcal{M}}_{\parallel} = -\mathcal{M}_0 \; e\mathcal{E}_z w \; \lambda_{\tilde{d}} \begin{pmatrix} \sin\varphi_{\mathcal{N}} \\ \cos\varphi_{\mathcal{N}} \end{pmatrix}$$



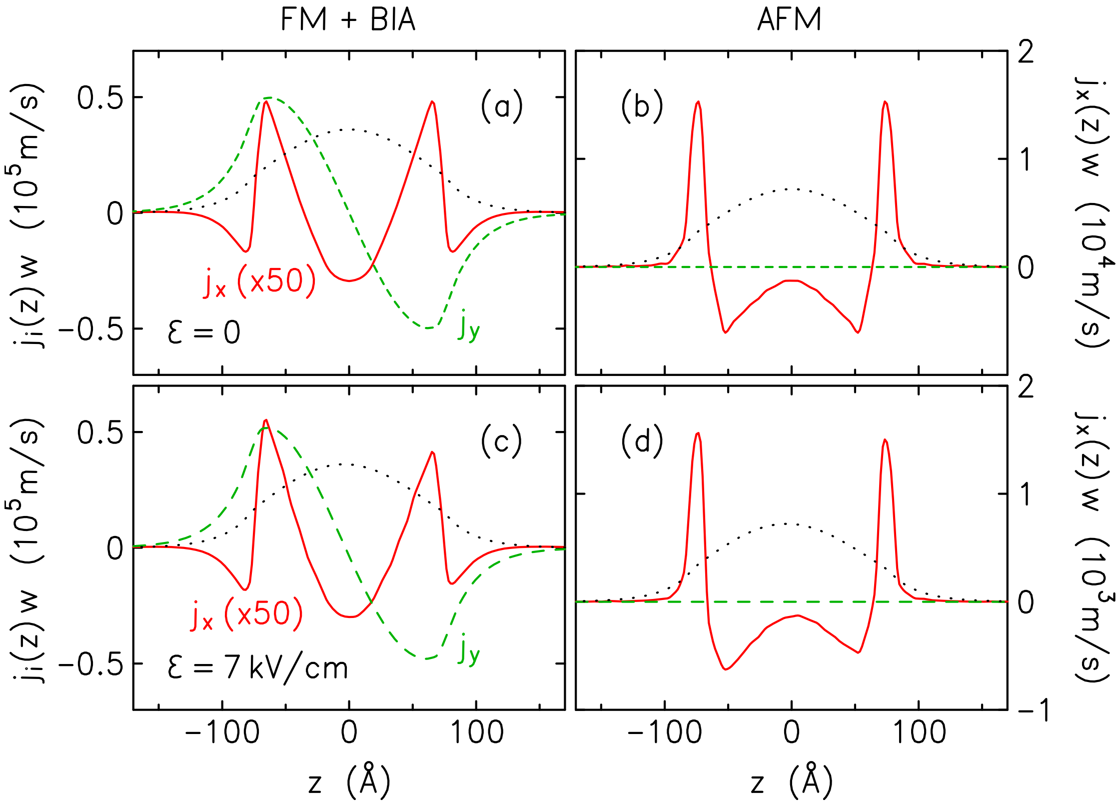
  - direction determined by (staggered) exchange field
  - details of quantum-well structure enter $\lambda_d, \lambda_{\tilde{d}}$
  - overall scale: $\mathcal{M}_0 = -\mu_{\mathrm{B}} N_{\mathrm{s}}/w$

- small magnitude (per particle)



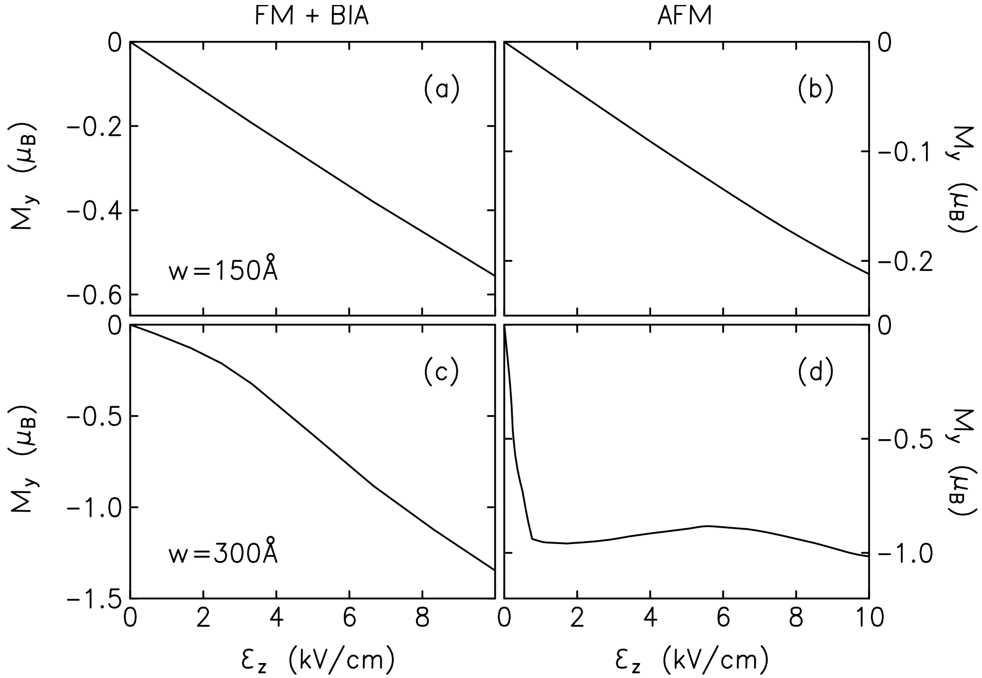
Physical picture

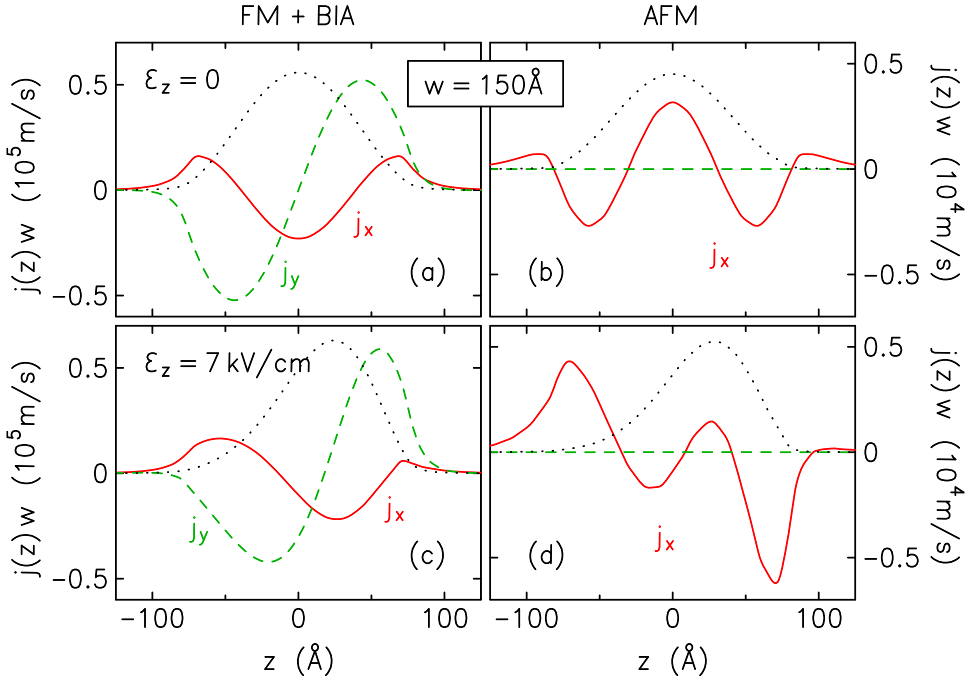
- electric field modifies the equilibrium-current distribution in quasi-2D ferro-/antiferromagnets
  - zero $\mathcal{E}_z$: quadrupolar equilibrium currents
  - finite $\mathcal{E}_z$ distorts the quadrupolar equilib. currents, generates a dipolar component
  - $\mathcal{E}_z$-induced in-plane magnetisation!



Results for valence-band carriers (holes)

- same basic physics, but much larger magnitude!
  - induced magnetisation/particle reaches $\sim 1\mu_B$
- peculiarities of valence-band structure relevant



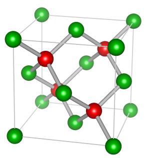


Magnetoelectric response II:

Magnetic-field-induced out-of-plane polarisation

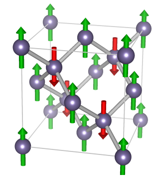
Analytical results for conduction band

- perturbative treatment of $\mathcal{B}_{\|}$ yields results for the out-of-plane electric polarisation:



$$\mathcal{P}_z = \mathcal{P}_0 \; \mu_\mathrm{B} \mathcal{B}_{\|} \; \lambda_d \; \xi(\mathcal{Z}) \; \sin(\varphi_\mathcal{Z} + \varphi_{\|})$$

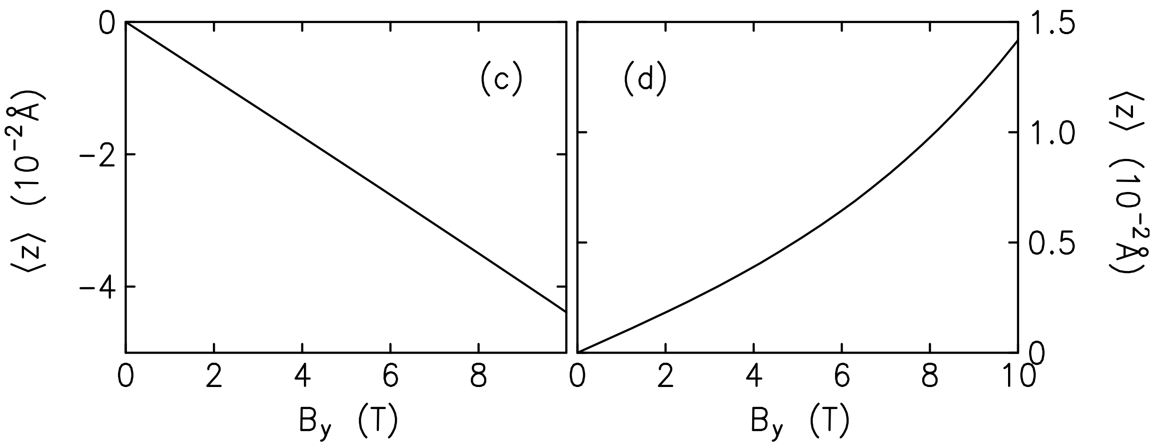
$$\mathcal{P}_z = \mathcal{P}_0 \; \mu_\mathrm{B} \mathcal{B}_{\|} \; \lambda_{\tilde{d}} \; \sin(\varphi_\mathcal{N} + \varphi_{\|})$$



  – modulated by direction of (staggered) exchange field
  – details of quantum-well structure enter $\lambda_d, \lambda_{\tilde{d}}$
  – overall scale is

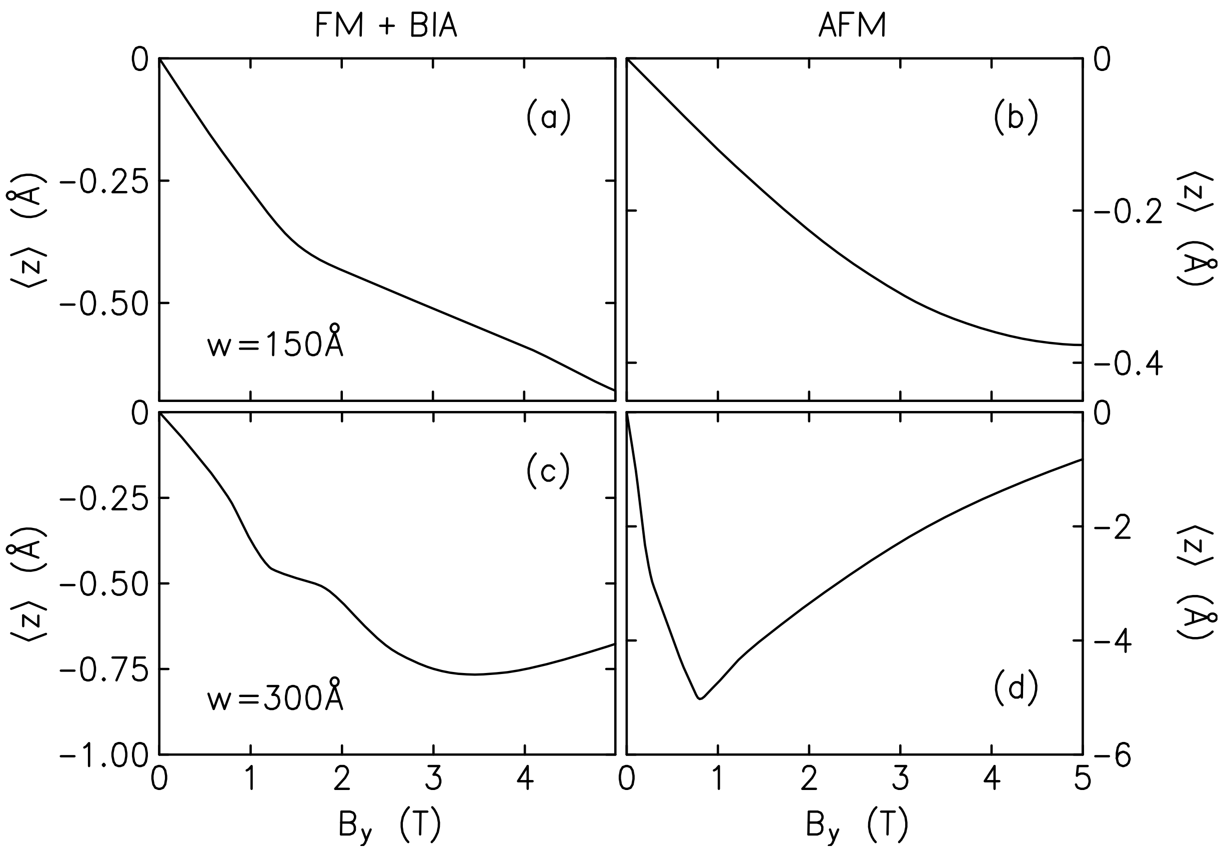
$$\mathcal{P}_0 = -e N_\mathrm{s}$$

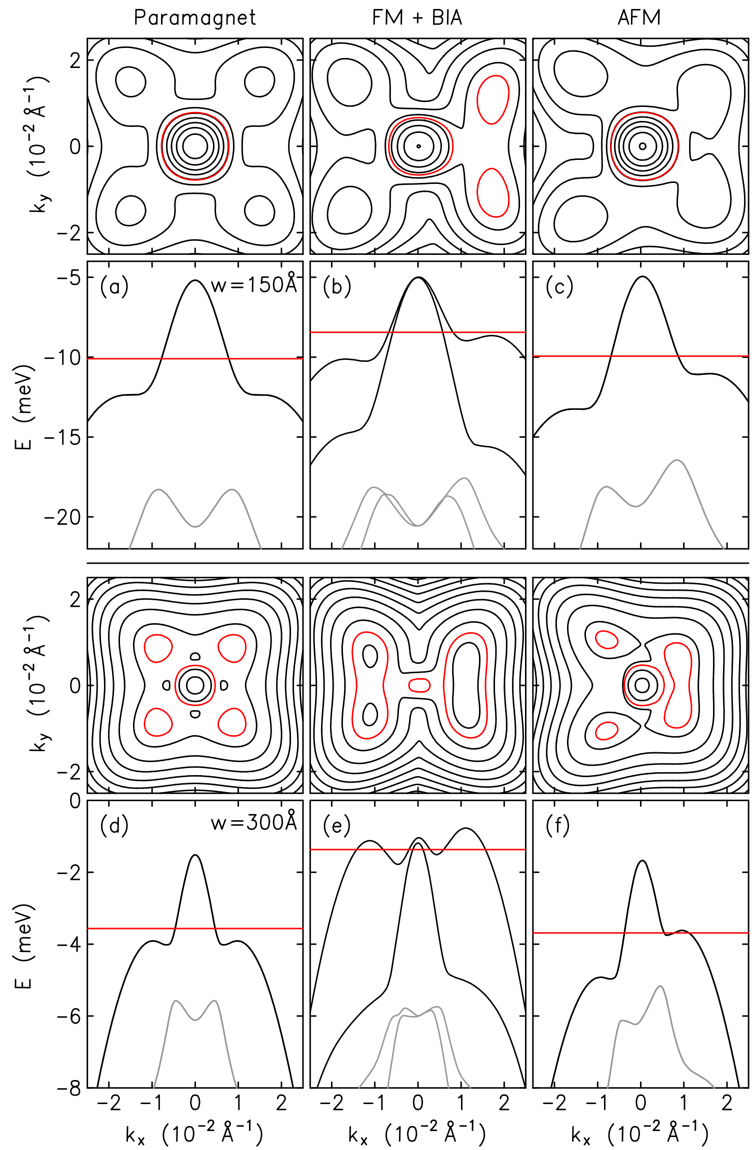
- typically a very small magnitude



Results for holes

- larger magnitude; complexity of subband structure matters





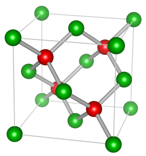
Antiferromagnetic order of charge carriers in ferro- and antiferromagnetic 2D systems

Quantifying AFM order of charge carriers

- ferromagnetism manifests in the band structure as an exchange field $\boldsymbol{\mathcal{X}}$
  - relevant term in Hamiltonian: $\mathsf{H}_{\mathcal{X}} = \boldsymbol{\mathcal{X}} \cdot \boldsymbol{\sigma}$
  - generates spin polarisation $\langle \boldsymbol{\sigma} \rangle$ of the charge carriers
- antiferromagnetism is associated with staggered exchange field $\boldsymbol{\mathcal{Y}}$
  - term in Hamiltonian: $\mathsf{H}_{\mathcal{Y}} = \dfrac{\tilde{d}}{\mathcal{Y}} \left(\{k_x, k_y^2 - k_z^2\} \, \mathcal{Y}_x + \text{c.p.} \right)$
  - rewrite as $\mathsf{H}_{\mathcal{Y}} = \boldsymbol{\mathcal{Y}} \cdot \boldsymbol{\tau}$
  - quantity $\langle \boldsymbol{\tau} \rangle$ represents antiferromagnetic order of the charge carriers generally, not only in antiferromagnets!

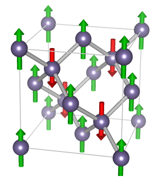
Antiferromagnetic order in quantum wells

- for conduction-band electrons: $\boldsymbol{\tau} = q_\tau\, k_z^2 \begin{pmatrix} k_x \\ -k_y \end{pmatrix}$
  – results for quantum wells:

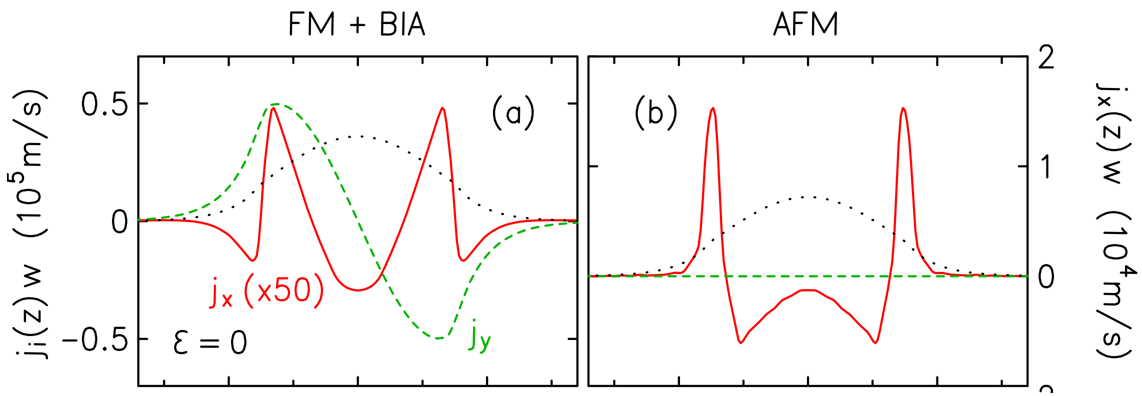


$$\langle \boldsymbol{\tau} \rangle = 2\pi\, q_\tau\, d\, N_\mathrm{s}\, \xi(\mathcal{Z}) \begin{pmatrix} \cos\varphi_\mathcal{Z} \\ \sin\varphi_\mathcal{Z} \end{pmatrix} \sum_{\nu' \neq 0} \frac{|\langle \nu' | k_z^2 | 0 \rangle|^2}{E_{\nu'}^{(0)} - E_0^{(0)}}$$

$$\langle \boldsymbol{\tau} \rangle = 2\pi\, q_\tau\, \tilde{d}\, N_\mathrm{s} \begin{pmatrix} \cos\varphi_\mathcal{N} \\ \sin\varphi_\mathcal{N} \end{pmatrix} \sum_{\nu' \neq 0} \frac{|\langle \nu' | k_z^2 | 0 \rangle|^2}{E_{\nu'}^{(0)} - E_0^{(0)}}$$



  – vector $\langle \boldsymbol{\tau} \rangle$ is parallel to (staggered) exchange field
- collinear antiferromagnetic order of charge carriers measured by $\langle \boldsymbol{\tau} \rangle$
- reflects quadrupolar equilibrium currents



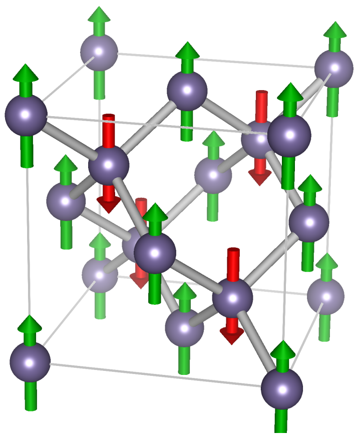
Envelope-function theory for charge carriers in diamond antiferromagnets

Band structure of diamond antiferromagnet

- tight-binding model for diamond, resp. zincblende structures + staggered exchange field $\boldsymbol{\mathcal{Y}}$
- obtain 14×14 extended Kane model description for the band structure
- additional terms involving $\boldsymbol{\mathcal{Y}}$

$$\begin{aligned}
\mathcal{H}^{\mathcal{Y}}_{8c\,8v} &= (2i/3)\,\mathcal{Y}(\mathcal{N}_x J_x + \mathrm{cp}) \\
\mathcal{H}^{\mathcal{Y}}_{8c\,7v} &= -2i\,\mathcal{Y}(\mathcal{N}_x U_x + \mathrm{cp}) \\
\mathcal{H}^{\mathcal{Y}}_{7c\,7v} &= (-i/3)\,\mathcal{Y}(\mathcal{N}_x \sigma_x + \mathrm{cp}) \\
\mathcal{H}^{\mathcal{Y}}_{6c\,6c} &= d(\{k_x, k_y^2 - k_z^2\}\mathcal{N}_x + \mathrm{cp}) \\
\mathcal{H}^{\mathcal{Y}}_{8v\,8v} &= \mathcal{D}^1_{88}(\{k_x, k_y^2 - k_z^2\}\mathcal{N}_x + \mathrm{cp}) \\
&\quad + \mathcal{D}^2_{88}[(\mathcal{N}_y k_y - \mathcal{N}_z k_z)J_x^2 + \mathrm{cp}] \\
&\quad + \mathcal{D}^3_{88}[(\mathcal{N}_x k_y - \mathcal{N}_y k_x)\{J_x, J_y\} + \mathrm{cp}] \\
&\quad + \mathcal{D}^4_{88}[(\mathcal{N}_y \mathcal{E}_z - \mathcal{N}_z \mathcal{E}_y)\{J_x, J_y^2 - J_z^2\} + \mathrm{cp}] \\
&\quad + \mathcal{D}^5_{88}[(\mathcal{N}_y \mathcal{E}_z + \mathcal{N}_z \mathcal{E}_y)J_x + \mathrm{cp}] \\
&\quad + \mathcal{D}^6_{88}[(\mathcal{N}_y \mathcal{E}_z + \mathcal{N}_z \mathcal{E}_y)J_x^3 + \mathrm{cp}] \\
&\quad + \mathcal{D}^7_{88}(\mathcal{N}_x \mathcal{E}_x + \mathrm{cp})(J_x J_y J_z + J_z J_y J_x) \\
\mathcal{H}^{\mathcal{Y}}_{7v\,7v} &= \mathcal{D}^1_{77}(\{k_x, k_y^2 - k_z^2\}\mathcal{N}_x + \mathrm{cp})
\end{aligned}$$

$$\boldsymbol{\mathcal{N}} = \boldsymbol{\mathcal{Y}}/\mathcal{Y}$$



Conclusions

- studied equilibrium magnetoelectric effects for charge carriers in quasi-2D systems
  - interplay of ferro-/antiferromagnetic exchange fields, quantum confinement, inversion-asymmetric bulk
  - sizable, tuneable, microscopically describable
- derived envelope-function theory for diamond-antiferromagnet bands (generalised Kane model)
  - widely applicable to study AFMs' electronic properties
- derived quantity $\langle \boldsymbol{\tau} \rangle$ that represents AFM order of charge carriers: analog of $\langle \boldsymbol{\sigma} \rangle$ that's finite in FMs!

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