Old 2degs with new tricks: Antiferromagnetic order and magnetoelectricity of 2D charge carriers

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Outline

- Introduction
  - basics of the magnetoelectric effect
  - quasi-2D charge carriers in semiconductor quantum wells
- Multi-band **k·p** (envelope-function) theory
  - zincblende-structure FM
  - diamond-structure AFM
- Magnetoelectricity exhibited by quasi-2D charge carriers
  - is tunable and sizable (~1μ_B/carrier)
  - due to AFM order of charge carriers
- AFM order of quasi-2D charge carriers
  - signaled by new quantity $\langle\boldsymbol{\tau}\rangle$: AFM analog to spin polarisation in FM
- Conclusions

Introduction

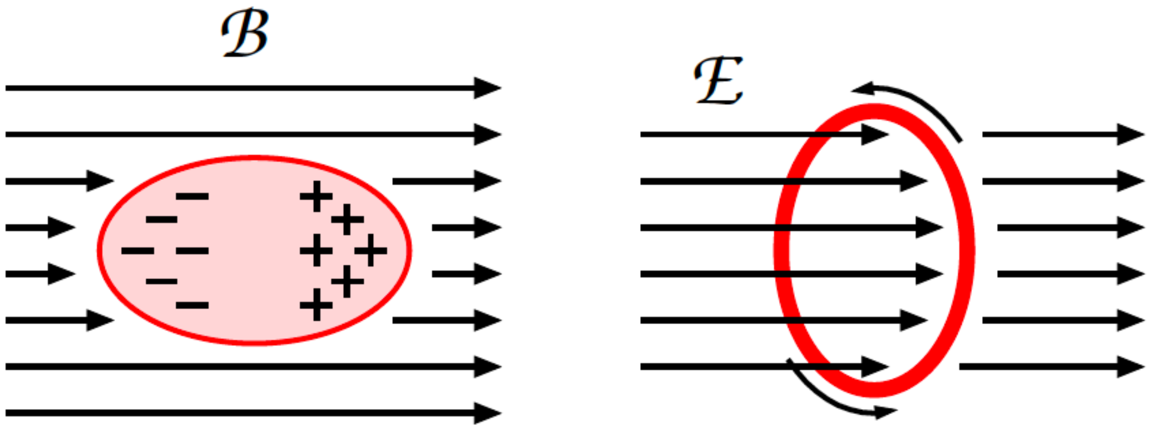
Magnetoelectric effect

- magnetoelectric media exhibit unusual electromagnetic response

O'Dell, *The Electrodynamics of Magneto-electric Media* (1970); Fiebig, J. Phys. D (2005)

$$\mathcal{P}_i = \chi^{\mathcal{E}}_{ij}\,\mathcal{E}_j + \alpha_{ij}\,\mathcal{B}_j$$

$$\mathcal{M}_i = \alpha_{ji}\,\mathcal{E}_j + \chi^{\mathcal{B}}_{ij}\,\mathcal{B}_j$$



- magnetoelectric tensor α_{ij} governs both $\boldsymbol{\mathcal{B}} \to \boldsymbol{\mathcal{P}}$ and $\boldsymbol{\mathcal{E}} \to \boldsymbol{\mathcal{M}}$ effects
- requires broken space-inversion and time-reversal symmetries
- is an equilibrium phenomenon
  - free energy: $F(\boldsymbol{\mathcal{E}}, \boldsymbol{\mathcal{B}}) = F(\mathbf{0}, \mathbf{0}) - \frac{1}{2}\chi^{\mathcal{E}}_{ij}\,\mathcal{E}_j\,\mathcal{E}_j - \frac{1}{2}\chi^{\mathcal{B}}_{ij}\,\mathcal{B}_i\,\mathcal{B}_j - \alpha_{ij}\,\mathcal{E}_i\,\mathcal{B}_j - \ldots$

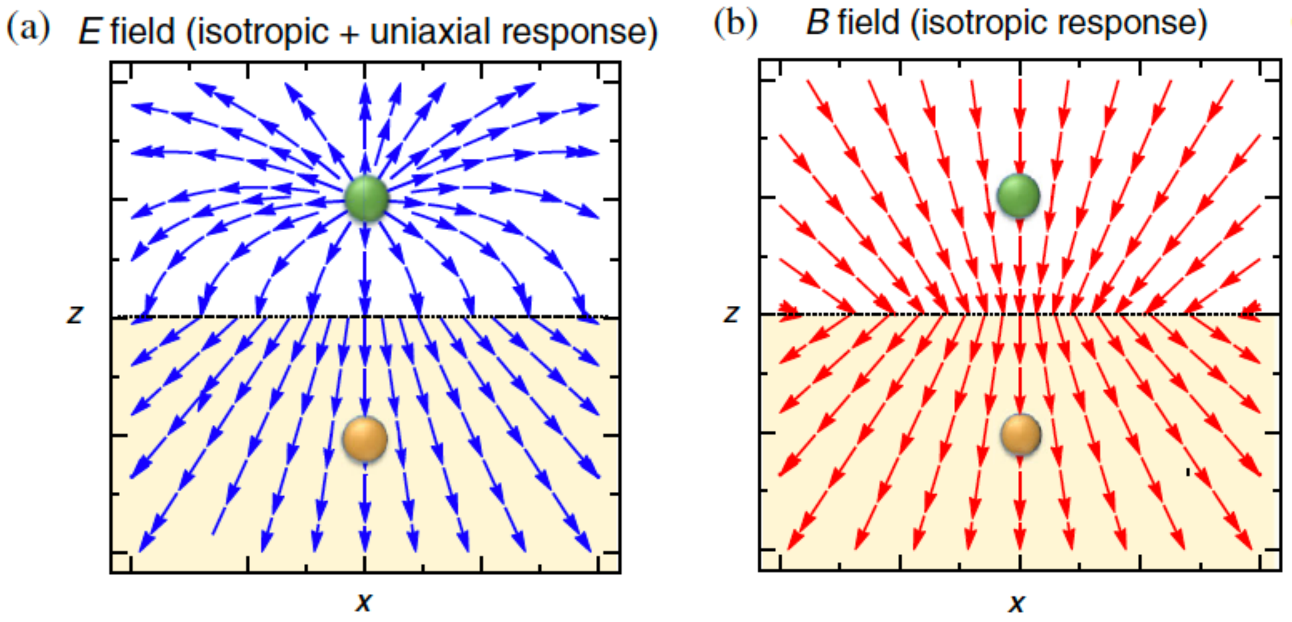
Landau and Lifshitz, *Electrodynamics of Continuous Media* (1957)

Unusual electromagnetism in magnetoelectrics

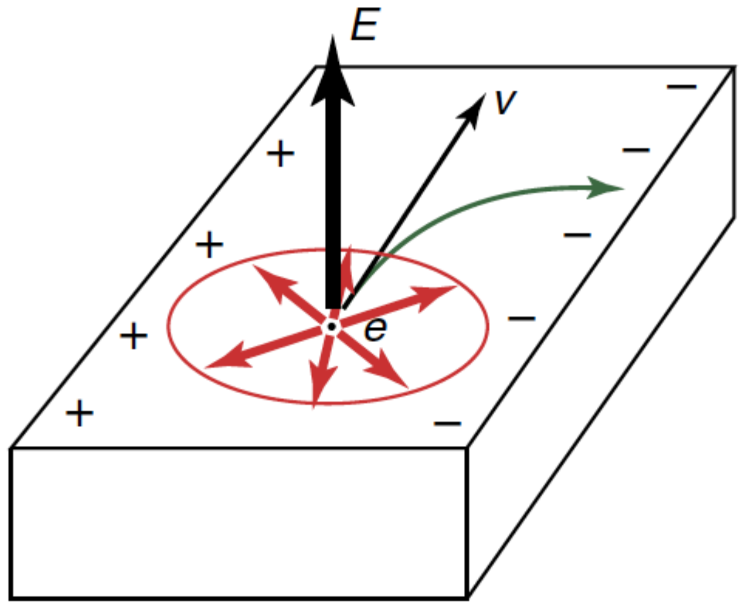
- electromagnetism turned on its head/perfectly dual!

Fechner et al., Phys. Rev. B (2014); Khomskii, Nat. Commun. (2014)

  - electric charge generates magnetic-monopole field
  - thus magnetic fields accelerate an electric charge
  - electric Hall effect, magneto-photovoltaic effect, ...



Meier et al., Phys. Rev. X **9**, 011011 (2019)



Khomskii, Nat. Commun. **5**, 4793 (2014)

Survey of materials and magnitudes

single-phase materials

Cr$_2$O$_3$

$$|\alpha_{ij}| \lesssim 3\times10^{-4}\sqrt{\epsilon_0/\mu_0}$$

$$|\mathcal{M}| \lesssim 0.001\ \mu_B/\text{Cr atom}$$

Hehl et al., Phys. Rev. A (2008)

TbPO$_4$

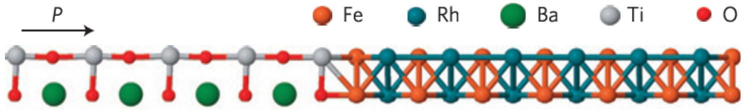
$$|\alpha_{ij}| \lesssim 9\times10^{-2}\sqrt{\epsilon_0/\mu_0}$$

$$|\mathcal{M}| \lesssim 2\ \mu_B/\text{Tb atom}$$

Rado et al., Phys. Rev. B (1984)

multiferroics

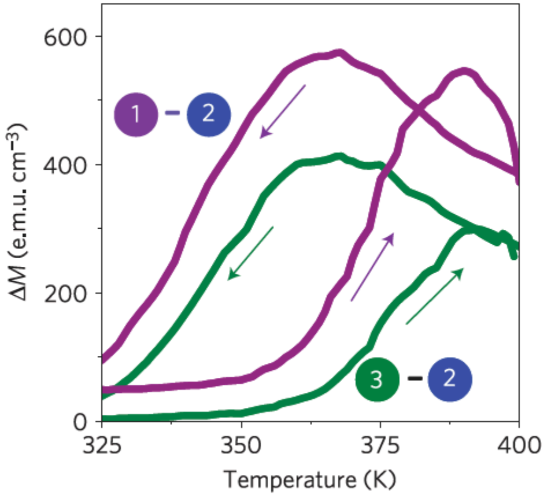
BaTiO$_3$/FeRh



$$|\alpha_{ij}| \lesssim 4{,}800\sqrt{\epsilon_0/\mu_0}$$

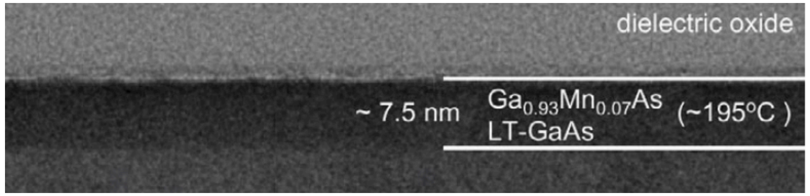
$$|\mathcal{M}| \lesssim 2\ \mu_B/\text{Fe atom}$$

Cherifi et al., Nat. Mater. (2014)



magnetic semiconductors

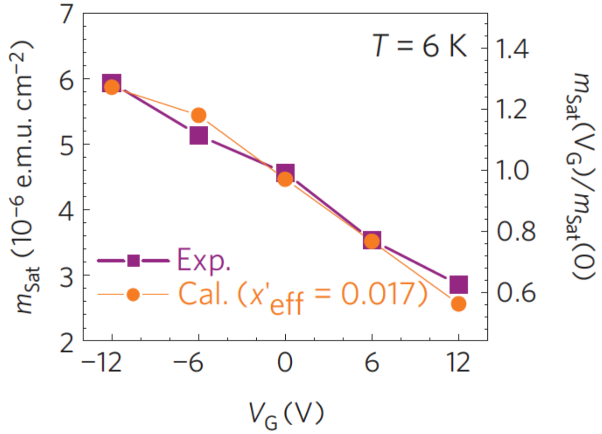
GaMnAs



$$|\alpha_{ij}| \lesssim 0.004\sqrt{\epsilon_0/\mu_0}$$

$$|\mathcal{M}| \lesssim 2\ \mu_B/\text{Mn atom}$$

Sawicky et al., Nat. Phys. (2009)



Magnetoelectric effect vs. Edelstein effect

magnetoelectric effect ☑

$$\mathcal{M}_i = \alpha_{ji}\,\mathcal{E}_j$$

- equilibrium phenomenon

Dzyaloshinski, JETP (1960)

- duality

  electric field $\boldsymbol{\mathcal{E}}$ → magnetisation $\boldsymbol{\mathcal{M}}$

  magnetic field $\boldsymbol{\mathcal{B}}$ → polarisation $\boldsymbol{\mathcal{P}}$

Landau and Lifshitz, *Electrodynamics of Continuous Media* (1957)

- requires broken time reversal

  → only in FM or AFM materials

Schmid, Int. J. Magn. (1973)
Watanabe and Yanase, Phys. Rev. B (2018)

Edelstein effect ☒

$$\mathcal{M}_i = \eta_{ik}\,\mathcal{J}_k = \eta_{ik}\,\sigma_{kj}\,\mathcal{E}_j$$

- nonequilibrium phenomenon

Ivchenko and Pikus, JETP Lett. (1978); Levitov et al., JETP (1985); Edelstein, Solid State Commun. (1990)

- Onsager reciprocity

  current $\boldsymbol{\mathcal{J}}$ → magnetisation $\boldsymbol{\mathcal{M}}$

  magnetisation $\boldsymbol{\mathcal{M}}$ → current $\boldsymbol{\mathcal{J}}$

Shen et al. Phys. Rev. Lett. (2014)

- broken time reversal not needed

  → occurs in nonmagnetic materials

  → spin-orbit torque in magnets

Manchon et al., Rev. Mod. Phys. (2019)

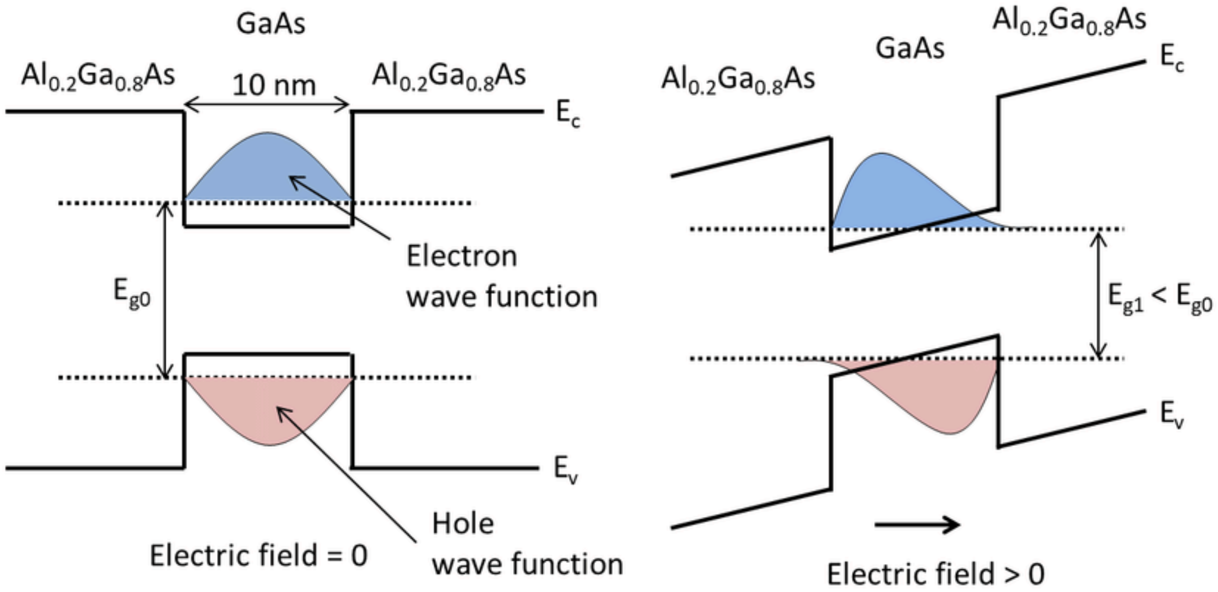
Quantum wells: Quasi-2D electron and hole systems

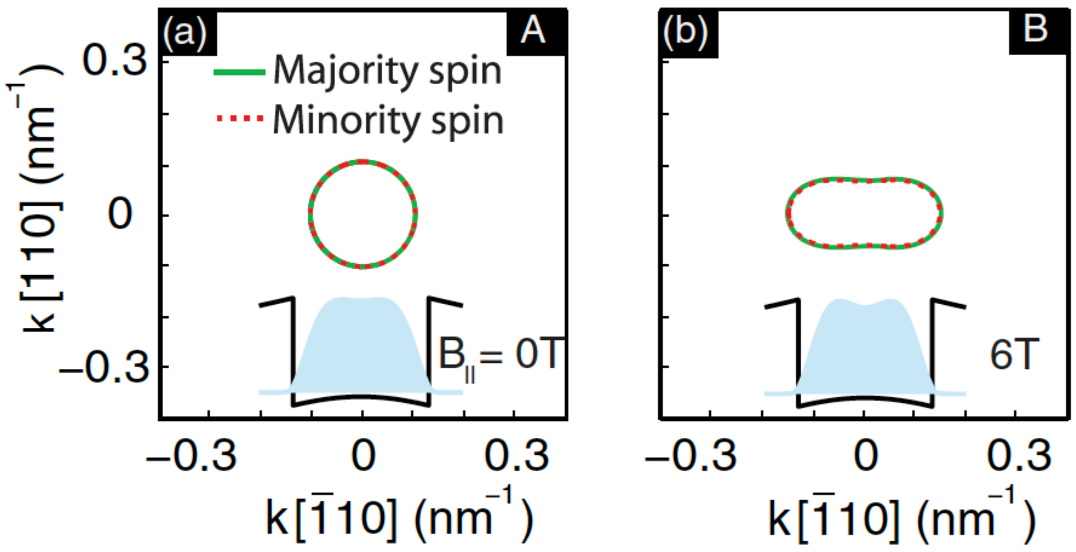
- band-gap engineering in semiconductor heterostructures

  Davies, *The Physics of Low-Dimensional Semiconductors* (1998)

- 2D bound state affected by electric and magnetic fields
  - perpendicular electric field $\mathcal{E}_z$ shifts density profile   Bastard et al., Phys. Rev. B (1983)
  - in-plane magnetic field $\boldsymbol{\mathcal{B}}_{\|}$ splits density profile Smrčka and Jungwirth, J. Phys. CM (1995)
  - this work: coupling of $\mathcal{E}_z$ or $\boldsymbol{\mathcal{B}}_{\|}$ to bound state → magnetoelectricity

Image credit: Vignesh Devaki Murugesan (U Ulm)





Mued et al., Phys. Rev. Lett. (2015)

Electric/magnetic responses of quasi-2D electron systems

- Schrödinger equation with
  - symmetric confining potential $V(z)$
  - electric potential $e\,\mathcal{E}_z\,z$
  - vector potential $\boldsymbol{\mathcal{A}} = z\,\boldsymbol{\mathcal{B}}_\parallel \times \hat{\mathbf{z}}$

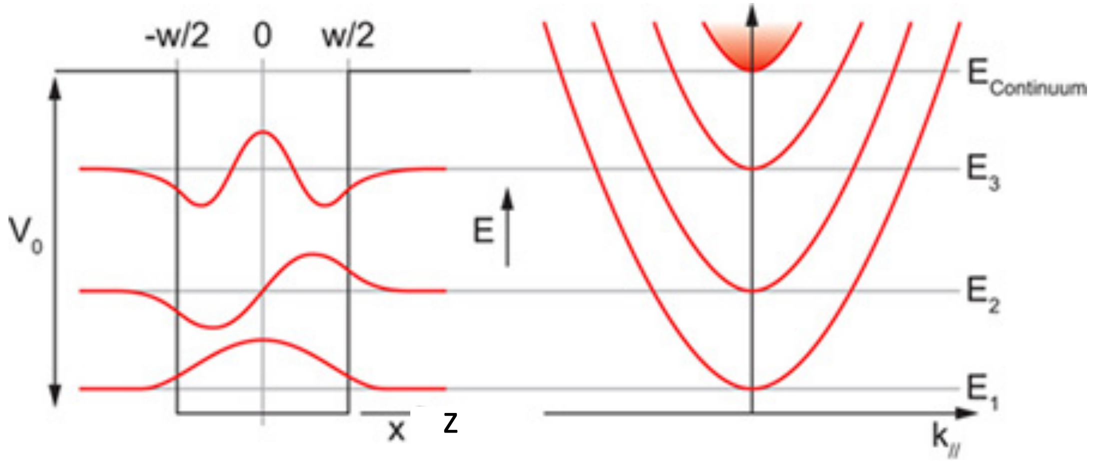
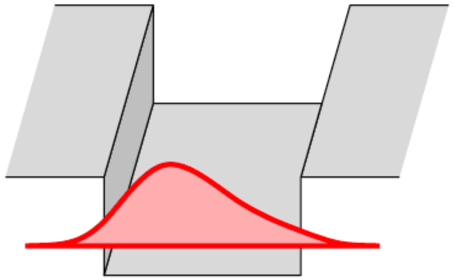
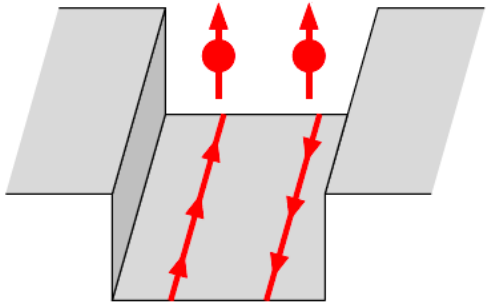


Image credit: Paul Townsend

- eigenstates $\Psi_{n\mathbf{k}_\parallel}(\mathbf{r}) = \dfrac{\mathrm{e}^{i\mathbf{k}_\parallel\cdot\mathbf{r}}}{2\pi}\,\Phi_{n\mathbf{k}_\parallel}(z)$
- perpendicular electric polarisation:

$$\mathcal{P}_z = -\frac{1}{w}\sum_n \int \frac{d^2k_\parallel}{(2\pi)^2}\, f(E_{n\mathbf{k}_\parallel})\; e\,\langle z\rangle_{n\mathbf{k}_\parallel}$$





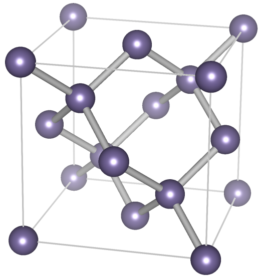
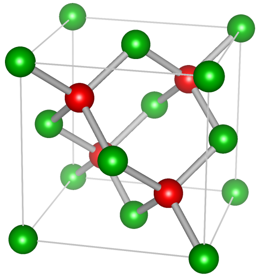
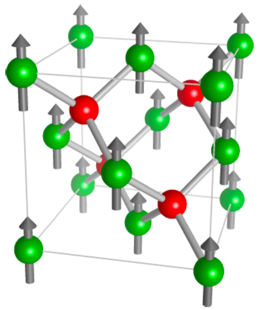
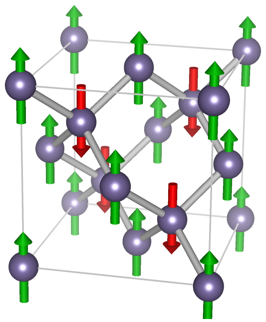
- in-plane magnetisation:

$$\boldsymbol{\mathcal{M}}_\parallel = -\frac{1}{w}\sum_n \int \frac{d^2k_\parallel}{(2\pi)^2}\, f(E_{n\mathbf{k}_\parallel})\;\left[e\,\hat{\mathbf{z}} \times \langle\{z\,,\mathbf{v}_\parallel\}\rangle_{n\mathbf{k}_\parallel} + \frac{g}{2}\,\mu_\mathrm{B}\,\langle\sigma\rangle_{n\mathbf{k}_\parallel}\right]$$

Envelope-function theory for charge carriers in semiconductors

(especially antiferromagnets)

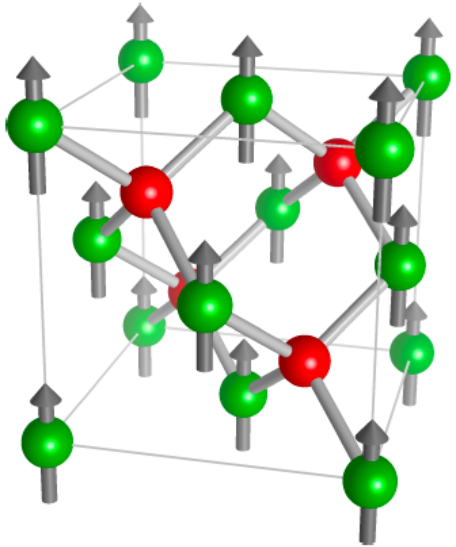
Bulk materials: Variants of the diamond structure

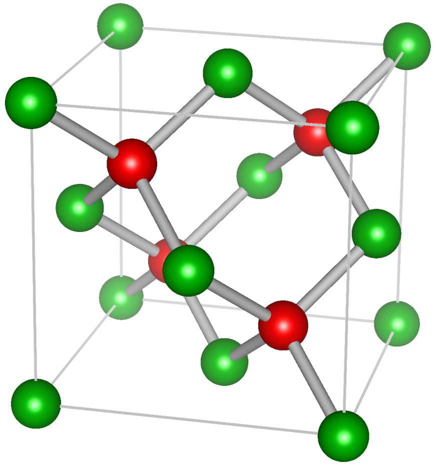
		space inversion	*time reversal*	*magneto-electric?*
• diamond structure Si, Ge, …		okay	okay	no
• zincblende structure GaAs, InSb, …		broken	okay	no
• zincblende FM (Ga, Mn)As, (In, Mn)Sb, …		broken	broken	yes
• diamond AFM $CoRh_2O_4$, …		broken	broken	yes

Band structure of a zincblende FM

- zincblende semiconductors: well-established extended-Kane-model

  Winkler, *Spin-Orbit-Coupling Effects in 2D Electron and Hole Systems* (2003)
- FM exchange field included via a Zeeman-type spin splitting   Jungwirth et al., Rev. Mod. Phys. (2006); Dietl and Ohno, *ibid.* (2014)
- conduction band: tractable analytically (two-band model)
- valence band: amenable only to numerics





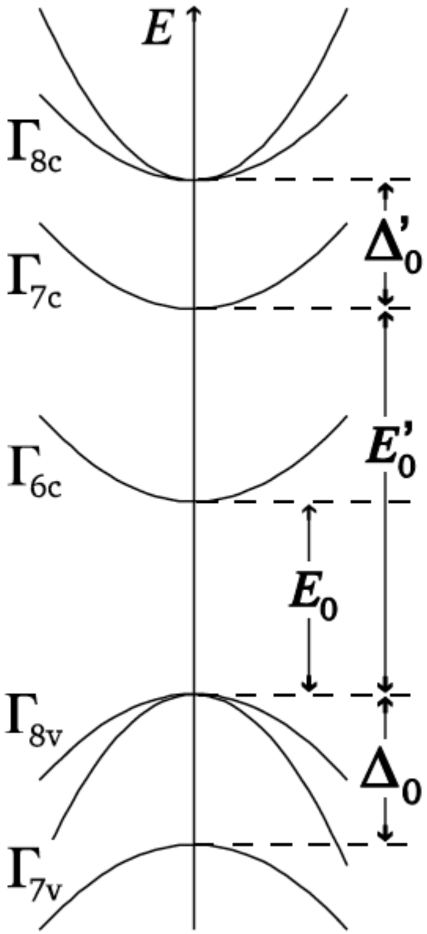
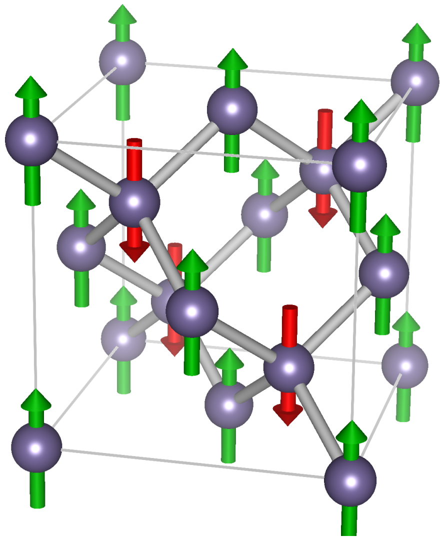


Image credit: Roland Winkler

Band structure of a diamond AFM

- apply well-known tight-binding model for diamond and zincblende structures with added ingredient: staggered exchange field $\boldsymbol{\mathcal{Y}}$
  - obtain 14 × 14 extended Kane model for the AFM system
- Kane Hamiltonian contains new terms depending on $\boldsymbol{\mathcal{Y}}$
  - one of our major new results!



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

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

Analytical two-band model for the conduction band

FM quantum well

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

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

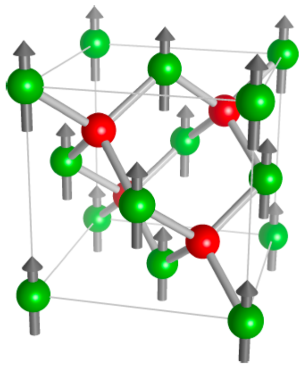
$$\mathsf{H}_\mathrm{D} = d\left(\{k_x, k_y^2 - k_z^2\}\,\sigma_x + \text{c.p.}\right)$$

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

$\boldsymbol{\mathcal{Z}}$ … 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

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

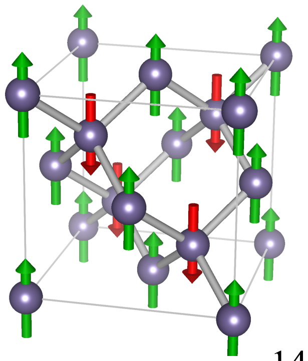
$$\mathsf{H}_k = \frac{\hbar^2 k^2}{2m}$$

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

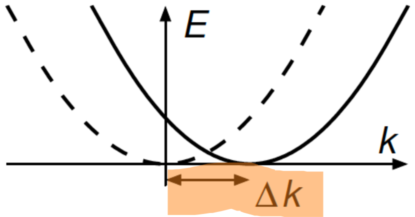
$\boldsymbol{\mathcal{Y}}$ … staggered exchange field

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

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



$$E(-\mathbf{k}) \neq E(\mathbf{k})$$

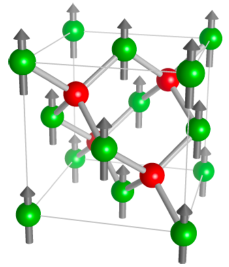


Magnetoelectric response:

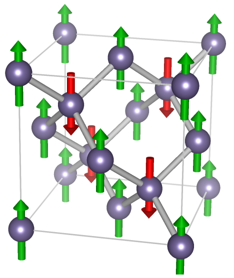
In-plane magnetization induced by a perpendicular electric field

Analytical results for the conduction band

- perturbative treatment of $\mathcal{E}_z \rightarrow$ in-plane magnetisation

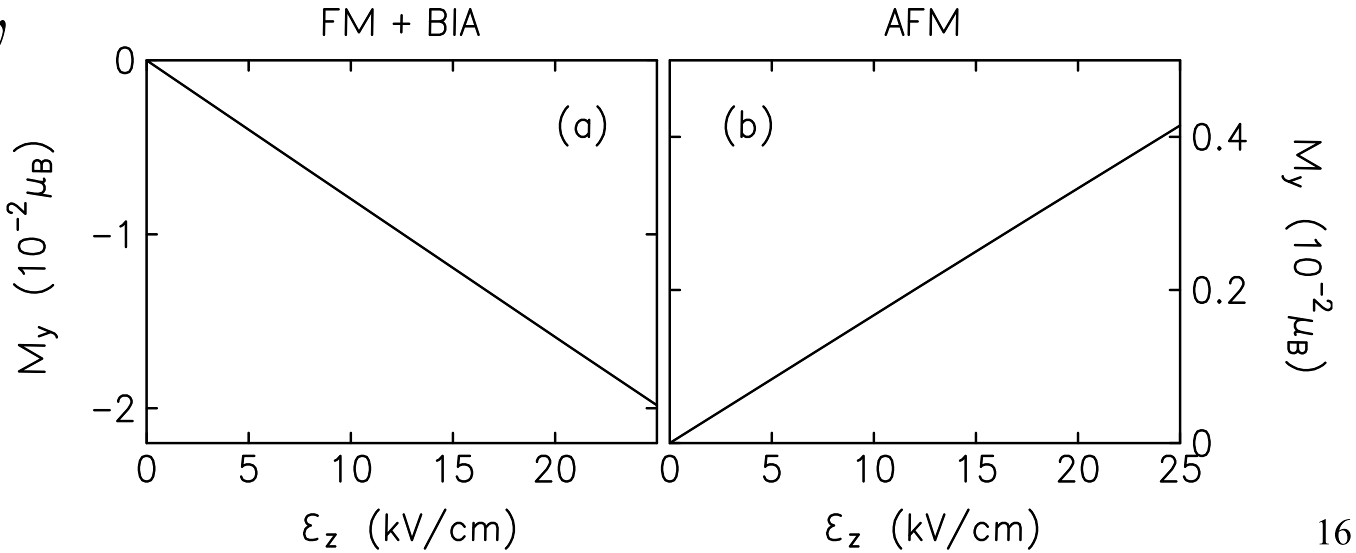


$$\boldsymbol{\mathcal{M}}_{\parallel} = \mathcal{M}_0 \, e\mathcal{E}_z w \, \lambda_d \, \xi(\mathcal{Z}) \begin{pmatrix} \sin\varphi_{\mathcal{Z}} \\ \cos\varphi_{\mathcal{Z}} \end{pmatrix} \quad \Big| \quad \boldsymbol{\mathcal{M}}_{\parallel} = -\mathcal{M}_0 \, e\mathcal{E}_z w \, \lambda_d \begin{pmatrix} \sin\varphi_{\mathcal{Y}} \\ \cos\varphi_{\mathcal{Y}} \end{pmatrix}$$



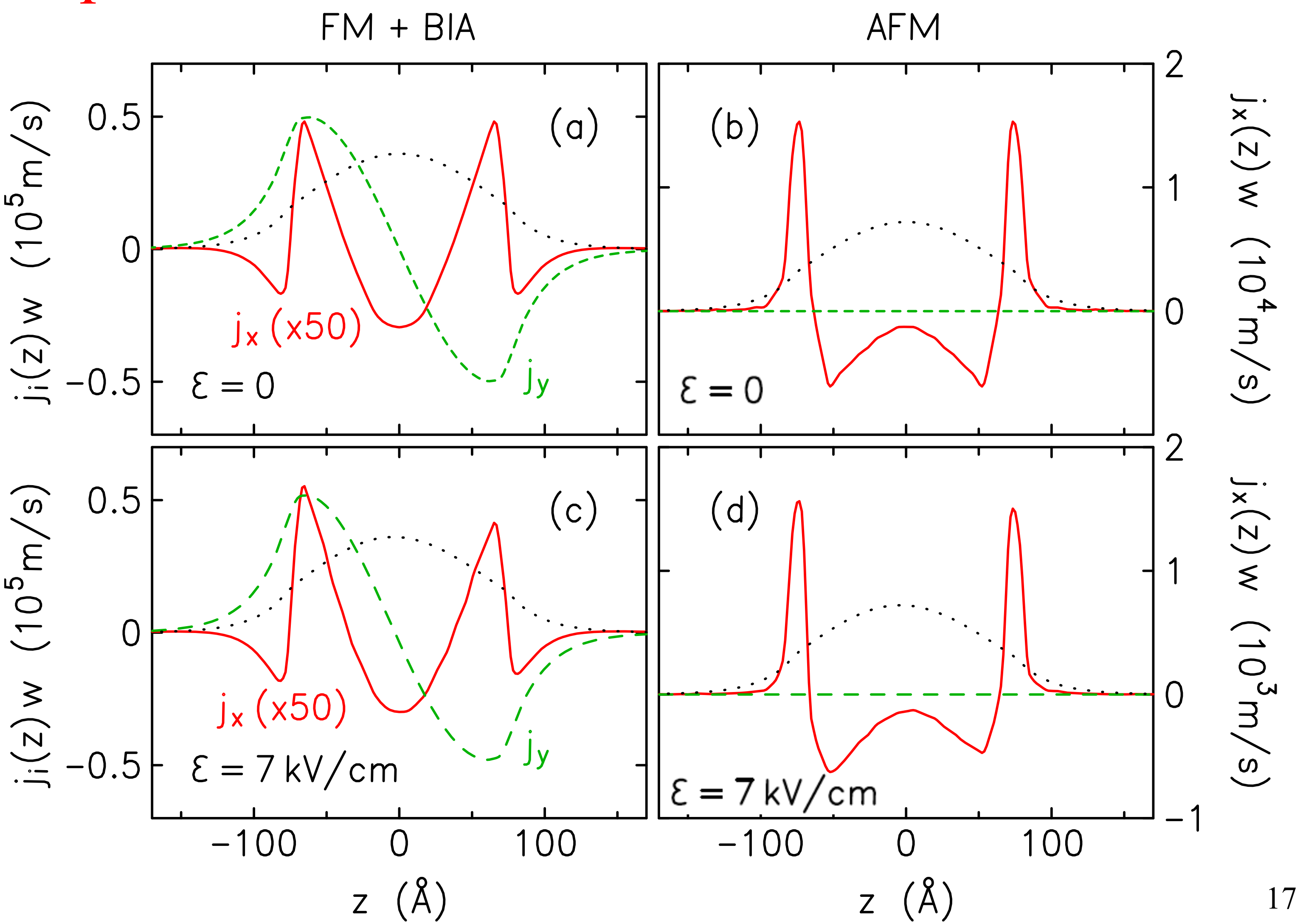
  - direction determined by exchange field $\boldsymbol{\mathcal{Z}}$ / staggered exchange field $\boldsymbol{\mathcal{Y}}$
  - details of quantum-well structure enter via λ_d
  - overall scale: $\mathcal{M}_0 = -\mu_{\mathrm{B}} N_{\mathrm{s}}/w$

- small magnetic moment per particle ($\sim 10^{-2}\,\mu_{\mathrm{B}}$)



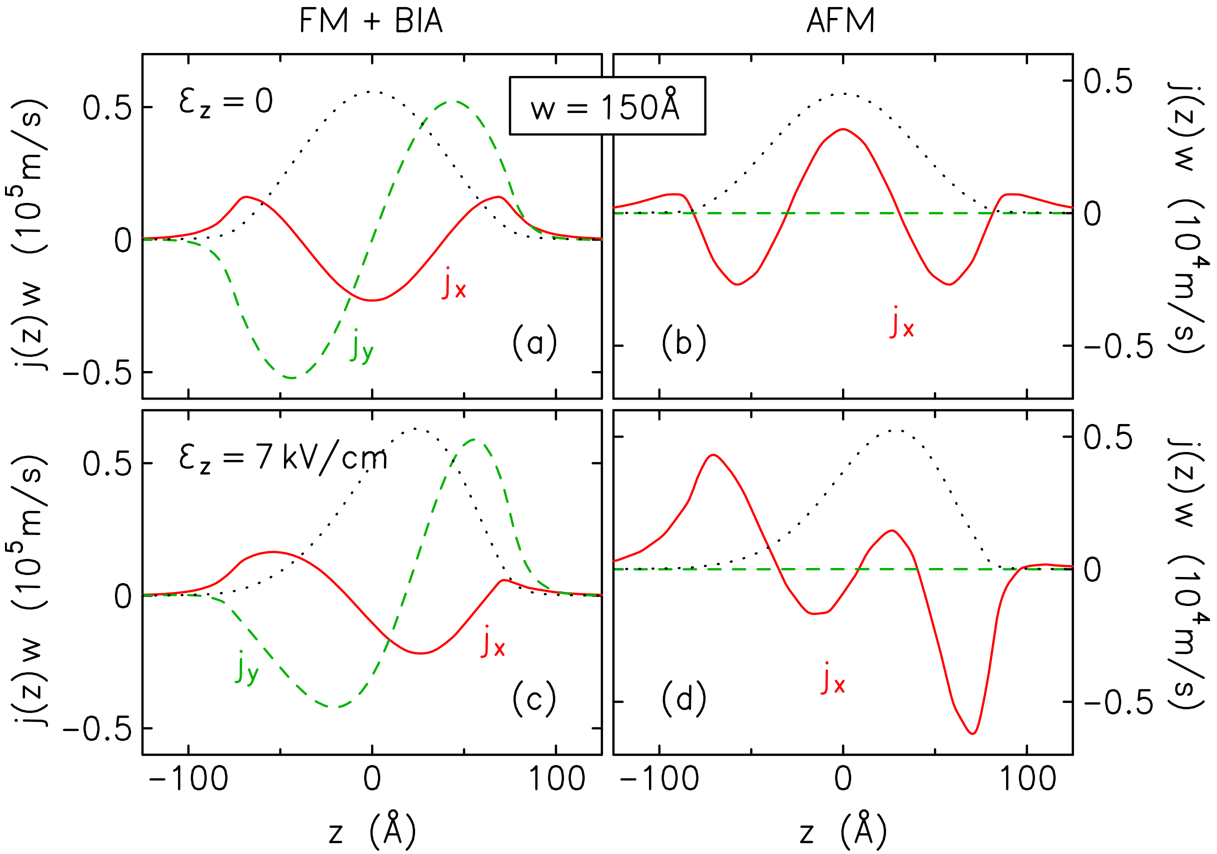
Physical picture

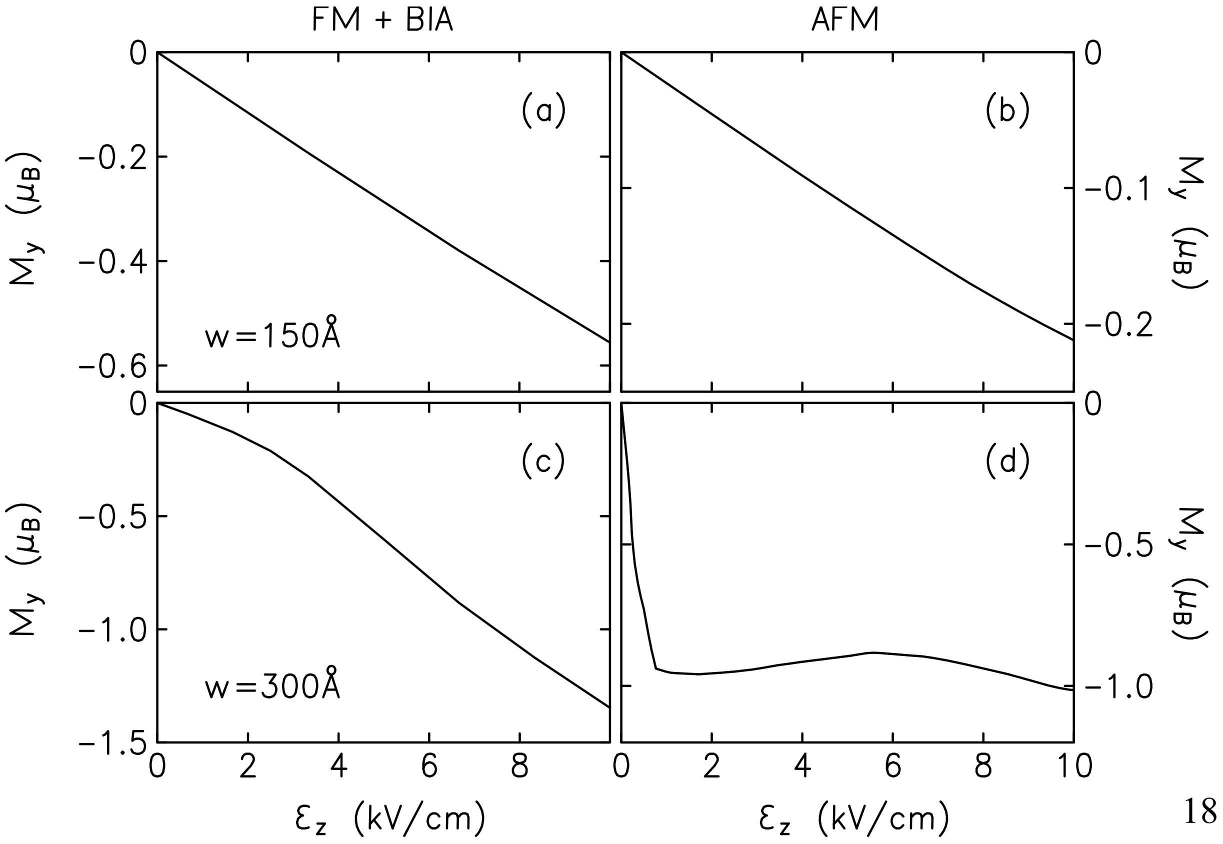
- electric field modifies the equilibrium-current distribution
- zero $\mathcal{E}_z$: quadrupolar equilibrium currents
  - AFM order also in FM!
- finite $\mathcal{E}_z$ distorts these quadrupolar currents
  - generates finite dipolar equilibrium current
  - finite magnetisation!



Results for valence-band carriers (holes)

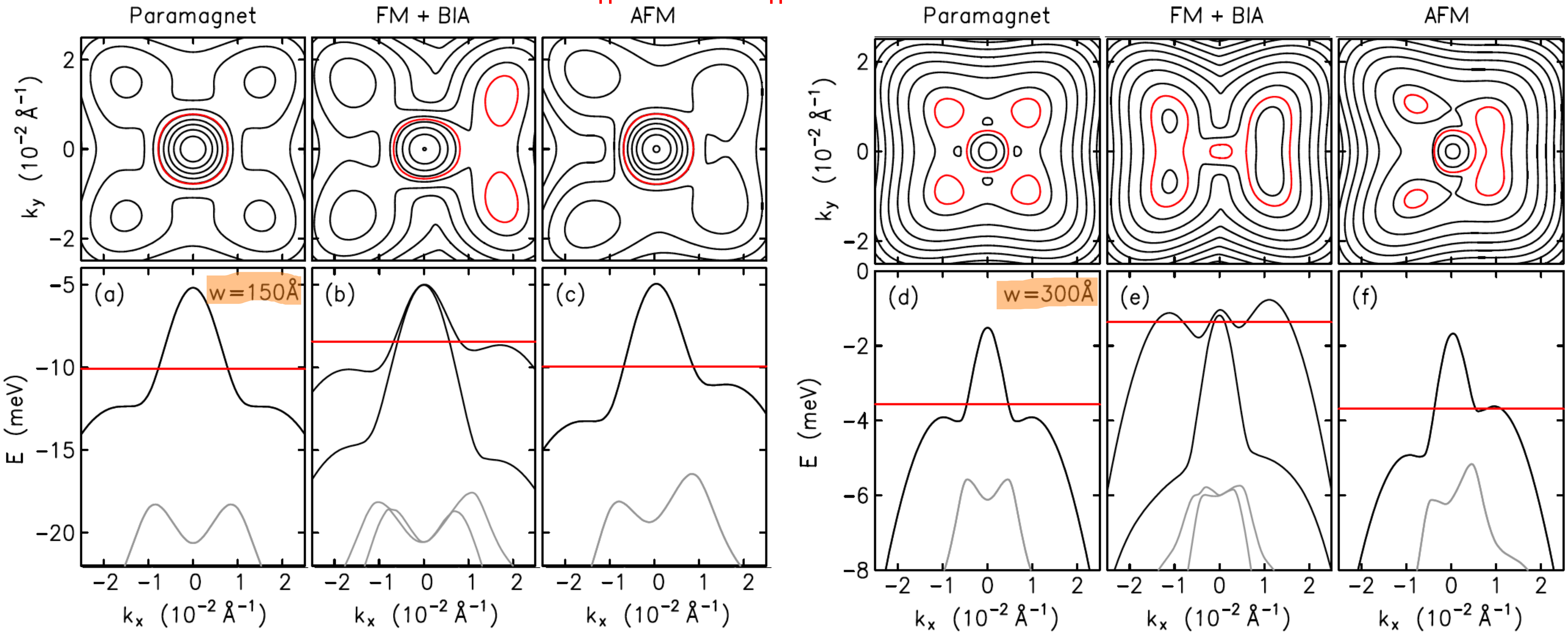
- same basic physics but much larger magnetic moment/particle ($\sim 1\,\mu_B$)
- peculiarities of valence-band structure relevant
  - strong nonlinear effects!





Why the strong magnetoelectricity in 2D holes?

- strong asymmetry $E_{n,-k_{||}} \neq E_{n,k_{||}}$ in subbands of FM/AFM material



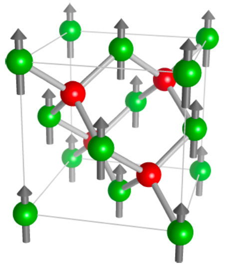
AFM order of charge carriers in FM and AFM 2D systems

Quantifying AFM order of charge carriers

- FM order manifests for charge carriers as an exchange field $\boldsymbol{\mathcal{X}}$
  - relevant term in Hamiltonian: $\mathsf{H}_{\mathcal{X}} = \boldsymbol{\mathcal{X}} \cdot \boldsymbol{\sigma}$
  - thus spin operator satisfies $\boldsymbol{\sigma} = \dfrac{\partial \mathsf{H}}{\partial \boldsymbol{\mathcal{X}}}$
  - finite spin polarisation $\langle \boldsymbol{\sigma} \rangle$ of charge carriers signals FM order
- AFM is associated with staggered exchange field $\boldsymbol{\mathcal{Y}}$
  - term in Hamiltonian: $\mathsf{H}_{\mathcal{Y}} = d\left(\left\{k_x, k_y^2 - k_z^2\right\} \mathcal{Y}_x + \text{c.p.}\right) \equiv \boldsymbol{\mathcal{Y}} \cdot \boldsymbol{\tau}$
  - thus find Néel operator $\boldsymbol{\tau} = \dfrac{\partial \mathsf{H}}{\partial \boldsymbol{\mathcal{Y}}}$
  - finite toroidal moment $\langle \boldsymbol{\tau} \rangle$ of charge carriers signals AFM order
- operators $\boldsymbol{\sigma}$ and $\boldsymbol{\tau}$ are fixed for any given band (structure)
  - measure FM/AFM order <u>even in absence of corresponding exchange fields</u>!

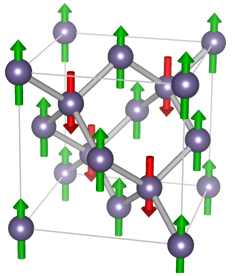
AFM order in quantum wells

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



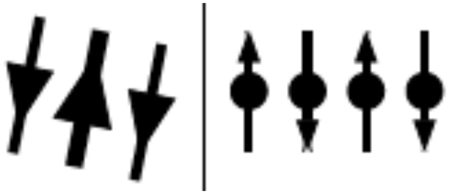
$$\langle \boldsymbol{\tau} \rangle = 2\pi \, q_\tau \, d \, N_\mathrm{s} \, \xi(\mathcal{Z}) \begin{pmatrix} \cos\varphi_\mathcal{Z} \\ \sin\varphi_\mathcal{Z} \end{pmatrix} \sum_{\nu' \neq 0} \frac{|\langle \nu' | k_z^2 | 0 \rangle|^2}{E_{\nu'}^{(0)} - E_0^{(0)}}$$

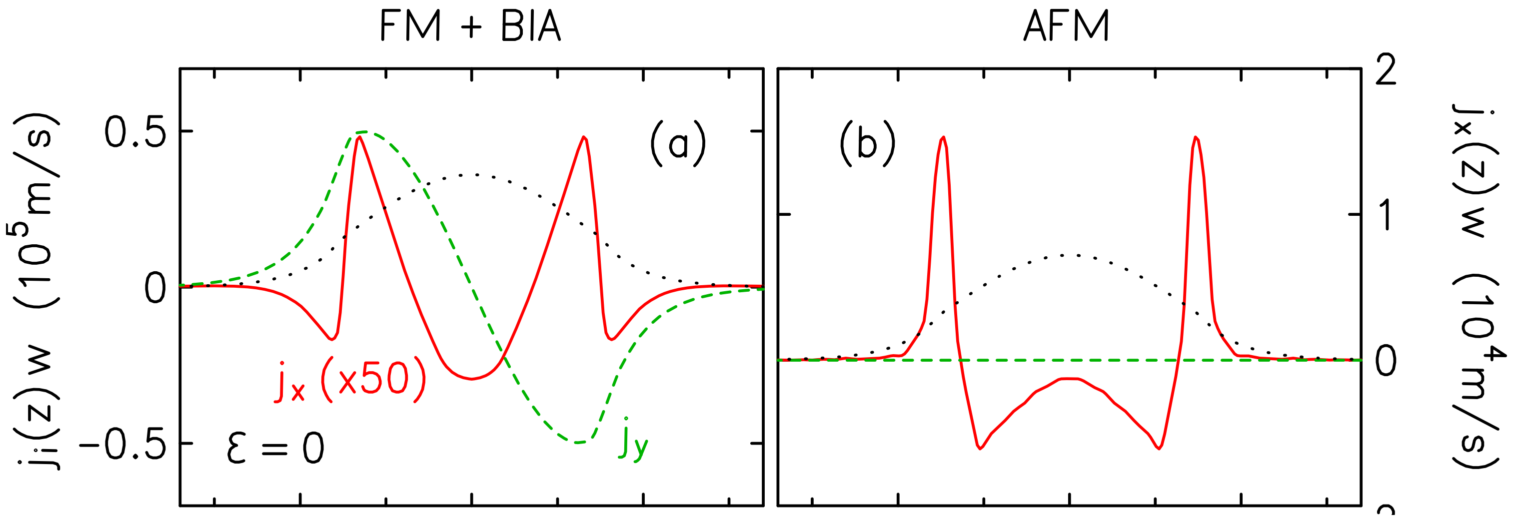
$$\langle \boldsymbol{\tau} \rangle = 2\pi \, q_\tau \, d \, N_\mathrm{s} \begin{pmatrix} \cos\varphi_\mathcal{Y} \\ \sin\varphi_\mathcal{Y} \end{pmatrix} \sum_{\nu' \neq 0} \frac{|\langle \nu' | k_z^2 | 0 \rangle|^2}{E_{\nu'}^{(0)} - E_0^{(0)}}$$



  - AFM order revealed by $\langle \boldsymbol{\tau} \rangle \neq 0$
  - vector $\langle \boldsymbol{\tau} \rangle$ parallel to exchange field $\boldsymbol{\mathcal{Z}}$ / staggered exchange field $\boldsymbol{\mathcal{Y}}$

- reflects quadrupolar equilibrium currents
  - like staggered spins!





Conclusions

- discussed equilibrium magnetoelectric effects in quasi-2D systems
  - interplay of FM/AFM exchange fields, confinement & no inversion symmetry
  - sizable, tuneable, microscopically describable & experimentally accessible
- derived envelope-function theory for band structure of diamond AFM
  - generalised Kane model to include effect of staggered magnetization
  - widely applicable to study electronic properties of (diamond) AFM
- identified toroidal moment $\langle \boldsymbol{\tau} \rangle$ of charge carriers
  - represents AFM order
  - complementary to spin polarization $\langle \boldsymbol{\sigma} \rangle$ that signals FM order
- Future work: Broader understanding of magnetoelectricity in AFM

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