Anomalous spin susceptibility and suppressed exchange energy of 2D holes

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Outline

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  - valence-band states in the bulk: Heavy holes & light holes
- Quantum confinement and valence-band structure
  - strong coupling of spin and orbital degrees of freedom
  - valence-band splitting & valence-band mixing
- Physical ramifications of valence-band mixing
  - 2D holes are not like 2D conduction-band electrons!
  - density response, carrier-density controlled anisotropic spin susceptibility, suppression of exchange energy, ...
- Conclusions

Introduction: Semiconductor band structure & bulk-hole states

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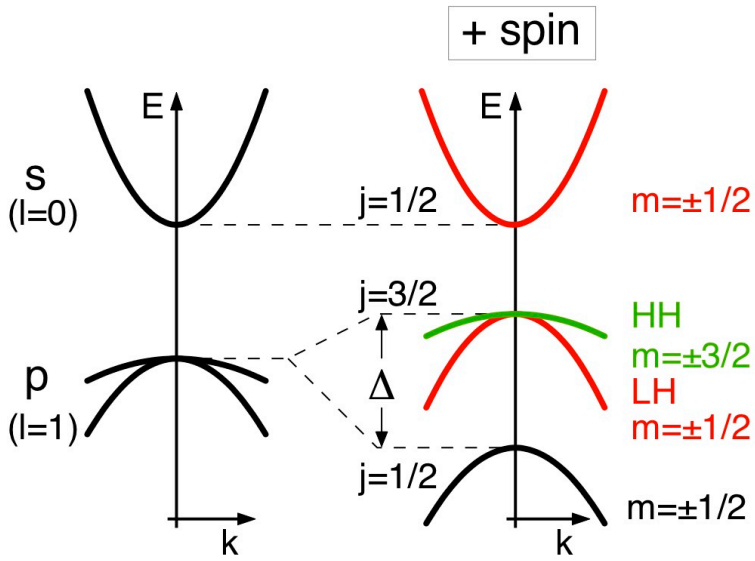
Band structure of common semiconductors

- quasi-free electrons in solids: energy bands
  - conduction band: orbital character of $l = 0$ (atomic s) state
  - valence band: has properties of $l = 1$ (atomic p) state
- spin-orbit coupling has the analogous effect as in atoms:
  - $l = 0$ & $s = ½ \rightarrow j = 1/2$
  - $l = 1$ & $s = ½ \rightarrow j = 3/2, 1/2$
    energy (fine-structure) splitting Δ between valence-band edges



Winkler et al., Sem. Sci. Techn. (2008)

- upper-most valence band: energy-split at finite **k** !

Valence-band states: Heavy & light holes

- effective-mass Hamiltonian for $j = 3/2$ valence band:

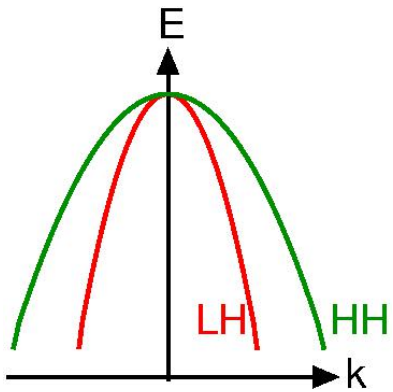
$$\mathcal{H} = -\frac{\hbar^2}{2m_0}\left\{\gamma_1 k^2 - \frac{2\gamma_s}{3}\left[3\left(\mathbf{k}\cdot\hat{\mathbf{J}}\right)^2 - k^2\,\hat{\mathbf{J}}^2\right]\right\}$$

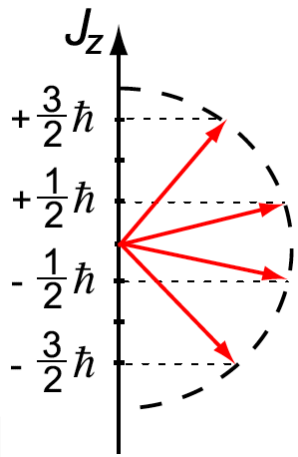
Shockley, Phys. Rev. (1950); Luttinger, Phys. Rev. (1956); Lipari & Baldereschi, PRL (1970)

- choose spin-3/2 quantisation axis || **k** , find energies

$$E_{m_j}(\mathbf{k}) = -\frac{\hbar^2 k^2}{2m_0}\left(\gamma_1 - 2\gamma_s\left[m_j^2 - 5/4\right]\right)$$

- two branches:
  - heavy holes (HHs) having $m_j = \pm 3/2$
  - light holes (LHs) having $m_j = \pm 1/2$





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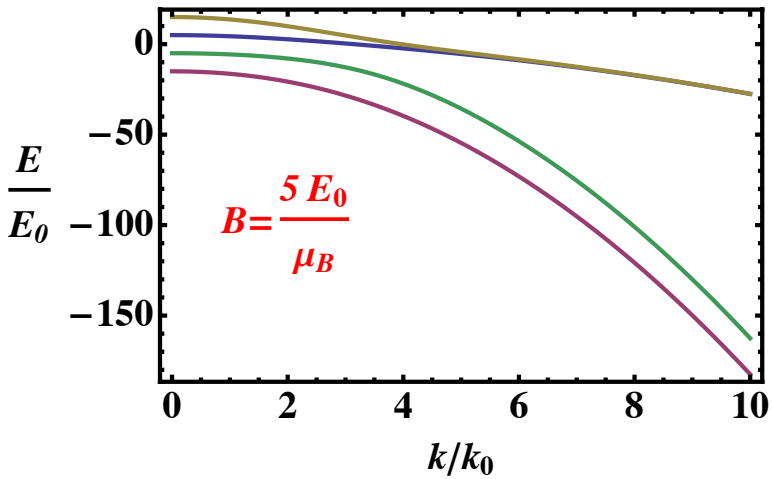
Interdependence of orbital motion and spin

- finite **k**: fixed spin-3/2 projection || **k** and splitting of $m_j = \pm 3/2$ & $m_j = \pm 1/2$ states: HH-LH splitting
- competition with magnetic field **B** ⊥ **k**
  - eg, **B** = B**x**: $B\hat{J}_x \equiv B(\hat{J}_+ + \hat{J}_-)$, hence Zeeman effect couples HH & LH states ⇒ HH-LH mixing
  - unlike spin-1/2 case: a HH-LH mixed state is not a reoriented dipole; spin 3/2 is much richer!

    Winkler, PRB (2004); PRB (2005)
  - unconventional Zeeman effect: HH states not split for large |**k**|

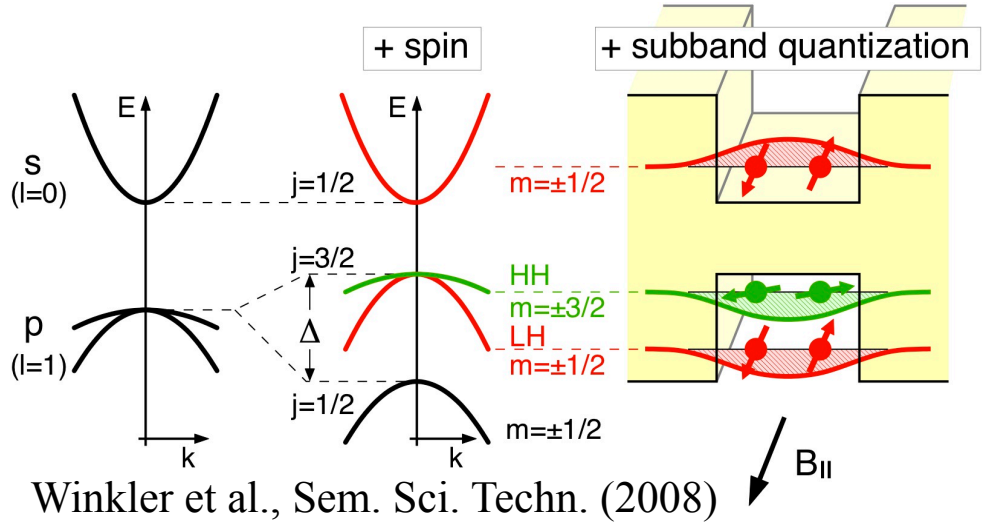


Suzuki & Hensel, PRB (1974); Csontos & UZ, PRB (2007)

Quantum confinement and valence-band structure

Quasi-2D hole systems in quantum wells

- band bending in semiconductor heterostructures realises textbook example of 2D quantum well
  - form 2D bound states in conduction & valence bands
  - different quantisation energy for HH and LH subbands
- HH-LH splitting of quasi-2D subband edges
  - fixes spin quantisation axis || growth direction
  - causes stiff response of the 2D HH states to in-plane **B** fields

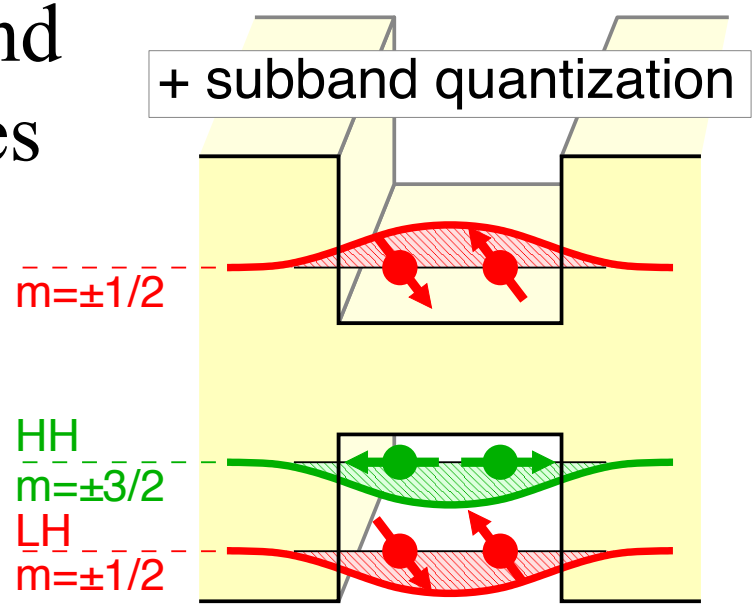


Winkler et al., Sem. Sci. Techn. (2008)

2D holes are different from electrons

- HH-LH splitting: lowest 2D hole subband edge is HH-like and doubly degenerate ($m_j = \pm 3/2$)
  - tempting to represent 2D holes states by pseudospin-½ degree of freedom, equivalent to cond.-band electrons
- but states in lowest 2D hole band have finite $m_j = \pm 1/2$ amplitudes for finite $\mathbf{k}_{\|}$: HH-LH mixing!

$$\Psi_{\rm el}^{(2D)} = \begin{pmatrix} \chi_\uparrow \\ \chi_\downarrow \end{pmatrix} \Leftrightarrow \Psi_{\rm hh}^{(2D)} = \begin{pmatrix} \chi_{\frac{3}{2}} \\ \chi_{\frac{1}{2}} \\ \chi_{-\frac{1}{2}} \\ \chi_{-\frac{3}{2}} \end{pmatrix}$$



2D hole states exhibit a kind of `chirality´

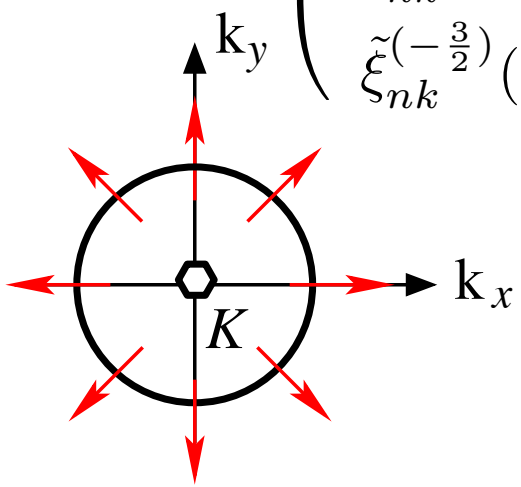
- Luttinger Hamiltonian contains terms that couple intrinsic spin to orbital motion: $\hat{J}_+^2(k_x - ik_y)^2 + \text{H.c.}$
- in axial approx: general state has the form

$$\Psi_{n\mathbf{k}}^{(2\mathrm{D})}(\mathbf{r}) = \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}\,\xi_{n\mathbf{k}}(z) \equiv \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}\;\mathrm{e}^{-i\hat{J}_z\theta_{\mathbf{k}}}\begin{pmatrix} \tilde{\xi}_{nk}^{(\frac{3}{2})}(z) \\ \tilde{\xi}_{nk}^{(\frac{1}{2})}(z) \\ \tilde{\xi}_{nk}^{(-\frac{1}{2})}(z) \\ \tilde{\xi}_{nk}^{(-\frac{3}{2})}(z) \end{pmatrix}$$

- compare with graphene:

$$\mathcal{H} \propto \sigma_+(k_x - ik_y) + \text{H.c.}$$

$$\Psi_{\sigma,\mathbf{k}}^{(\mathrm{g})}(\mathbf{r}) = \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}\;\mathrm{e}^{-i\frac{\sigma_z}{2}\theta_{\mathbf{k}}}\begin{pmatrix} 1 \\ \sigma \end{pmatrix}$$



Physical ramifications of valence-band mixing

I. Warm-up: Density response

Kernreiter, Governale, UZ, New J. Phys. **12**, 093002 (2010)

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Density response of the electron gas

- elementary property of conductors: charge carriers screen impurity potentials

$$\delta n(\mathbf{r}) = \int \frac{d^2q}{(2\pi)^2}\, \mathrm{e}^{i\mathbf{q}\cdot\mathbf{r}}\, \chi(q)\, V_{\mathrm{ext}}(q)$$

- can express general susceptibility $\chi(q)$ in terms of result $\chi_0(q)$ for non-interacting system (Lindhard)

$$\chi(q) = \frac{\chi_0(q)}{1 - v_q[1 - G(q)]\chi_0(q)}$$

Giuliani & Vignale, *Quantum Theory of the Electron Liquid*

- universal $\chi_0(q)$ for parabolic band dispersion

Lindhard function of 2D hole gases

- 2D electron system: universal 2D result applies

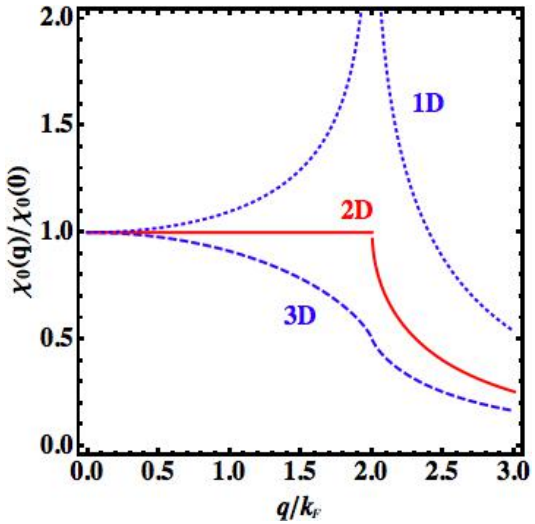
  Giuliani & Vignale, *Quantum Theory of the Electron Liquid*

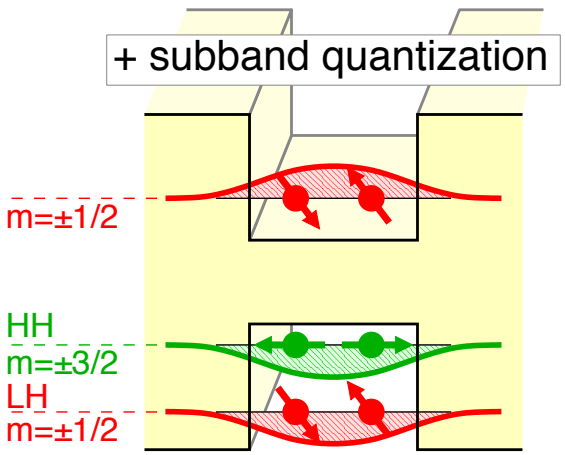
- lowest 2D hole subband: HH-LH mixing matters!

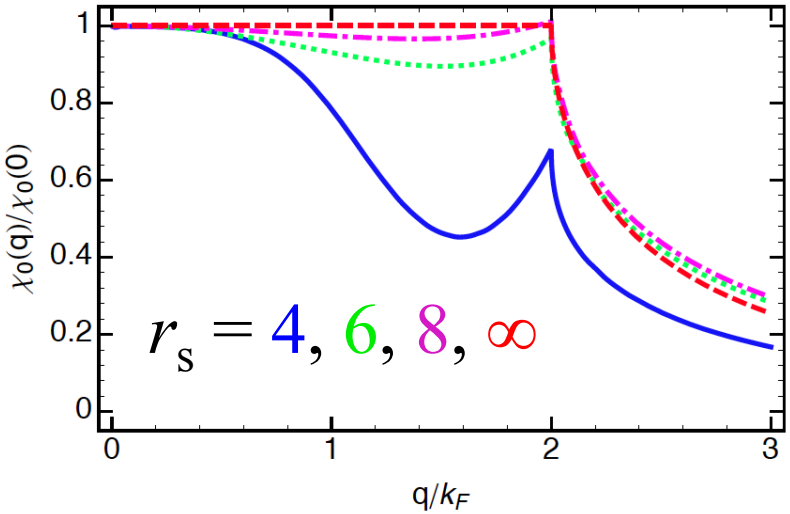
  Kernreiter, Governale & UZ, New J. Phys. (2010); Mat. Sci. Forum (2012)

  – density-dependent 2D-hole Lindhard function!

  – important deviations in the high-density limit







Physical ramifications of valence-band mixing

II. Spin response

Kernreiter, Governale, UZ, Phys. Rev. Lett. **110**, 026803 (2013)

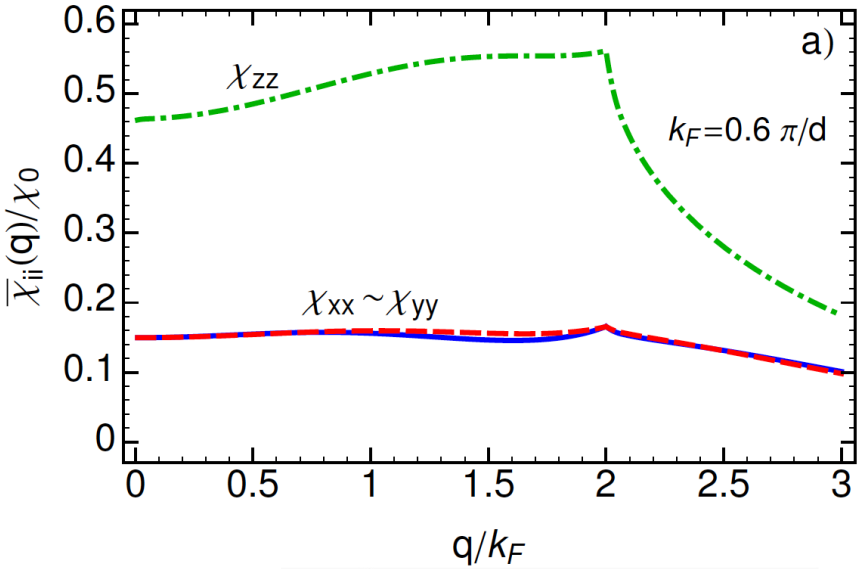
Spin susceptibility of 2D hole gases

- ordinary electron system: spin response tied to the density response (Lindhard): $\chi_{ij}(q) = \chi_0(q)\delta_{ij}$

  Giuliani & Vignale, *Quantum Theory of the Electron Liquid*

- 2D holes: interplay of HH-LH splitting and HH-LH mixing causes **density-dependent** anisotropy!
  - low density: easy axis || z (q-well growth) direction

    Kernreiter, Governale & UZ, PRL (2013)
  - as expected from HH-LH splitting (cf. behaviour of Zeeman splitting/g-factor!)

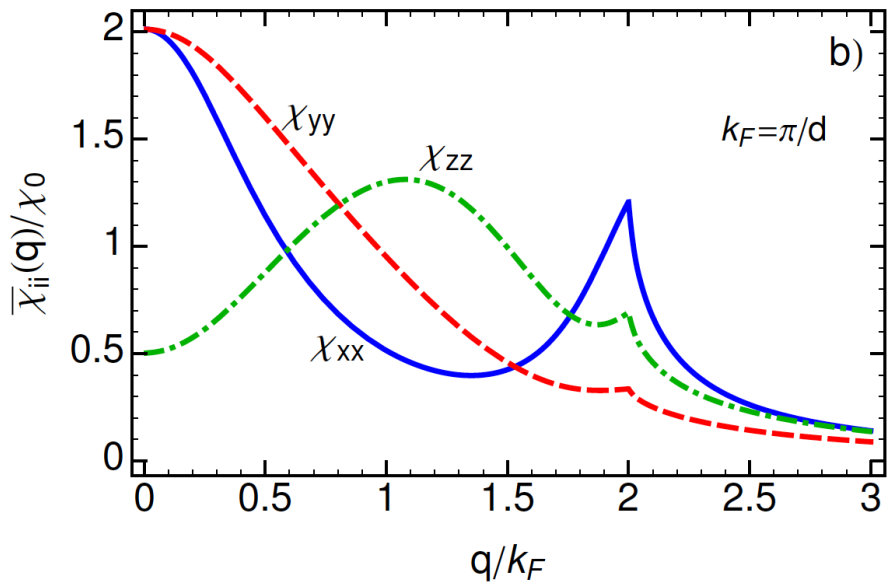


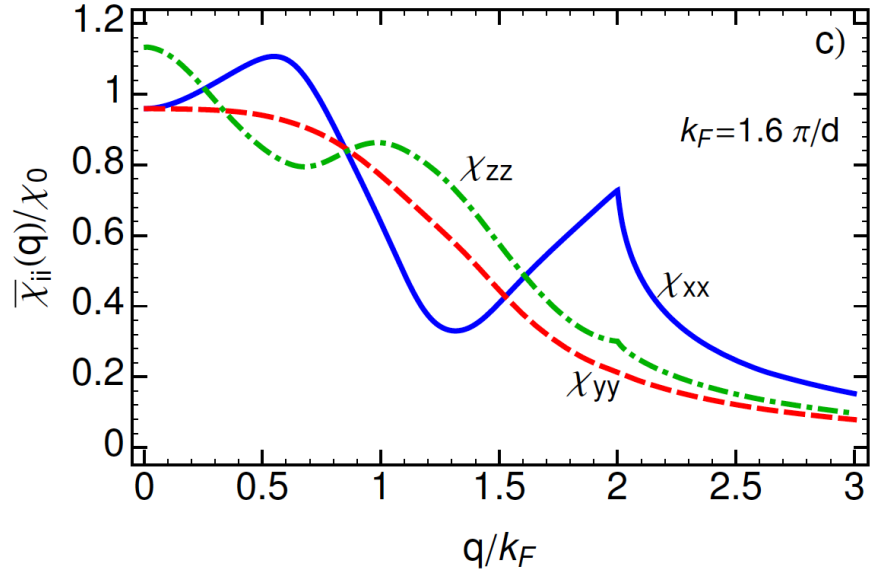
Spin susceptibility of 2D hole gases cont'd

- for higher densities: unexpectedly rich behaviour exhibited by 2D holes due to HH-LH mixing

Kernreiter, Governale & UZ, PRL (2013)

  – easy-plane response dominant at intermediate density
  – easy-axis again at high densities + structure at finite q





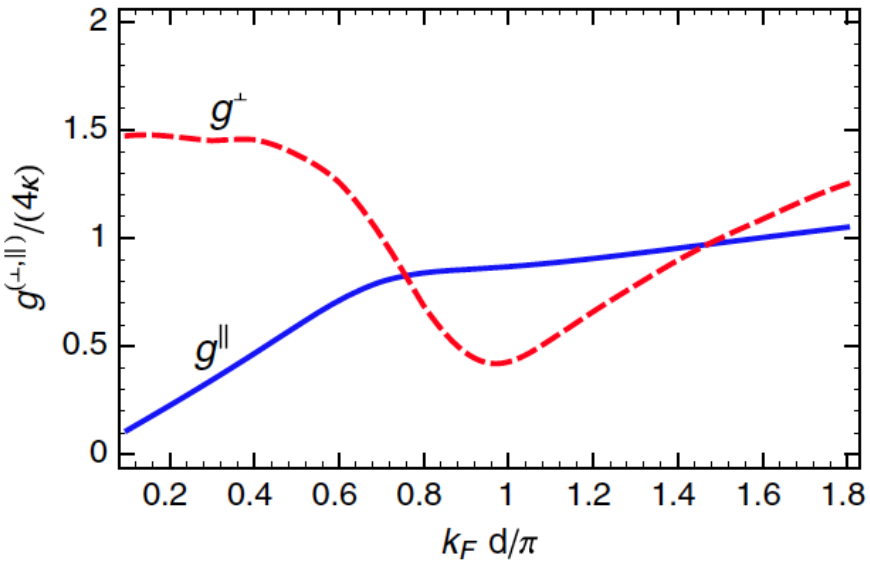
Effective g factor for 2D holes

- hole-spin couples to magnetic field: $\mathcal{H}_{\rm Z} = 2\kappa\,\mu_{\rm B}\,\mathbf{B}\cdot\hat{\mathbf{J}}$
- paramagnetic response: $\chi_{{\rm P},j} = (2\kappa\,\mu_{\rm B})^2\,\chi_{jj}(\mathbf{q}=0)$
- Pauli susceptibility: $\chi_{{\rm P},j} = \left(\frac{g_j\mu_{\rm B}}{2}\right)^2\,\chi_0(\mathbf{q}=0)$
  - $\propto$ Lindhard function/DOS
  - used by experimentalists to extract a 2D-hole g factor

    Chiu et al., PRB (2011)
- this motivates definition

  Kernreiter, Governale & UZ, PRL (2013)

$$g_j = 4\kappa\sqrt{\frac{\chi_{jj}(\mathbf{q}=0)}{\chi_0(\mathbf{q}=0)}}$$

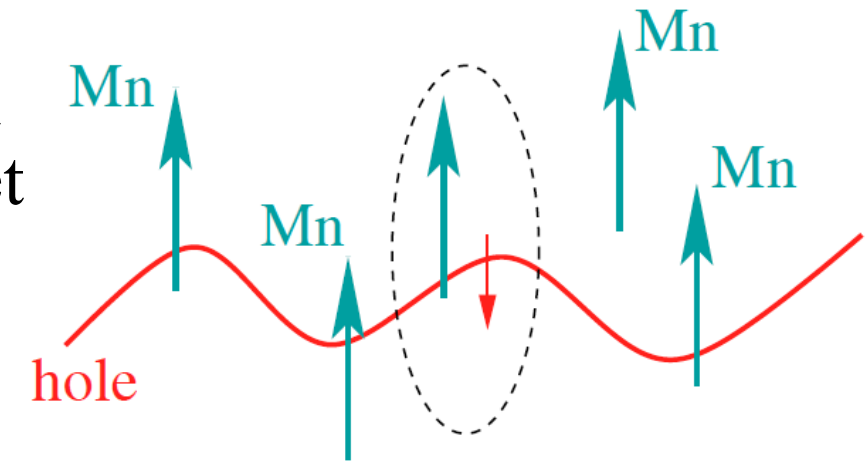


Carrier-mediated magnetic (RKKY) interaction

- charge carriers in conductors can mediate exchange interaction of localised magnetic impurities (spins)
- coupling depends on carriers' spin susceptibility χ_{ij}

$$H_{\mathrm{RKKY}}^{(\alpha\beta)} = -G^2 \sum_{i,j} S_i^{(\alpha)} S_j^{(\beta)} \chi_{ij}\left(\mathbf{R}_\alpha, \mathbf{R}_\beta\right)$$

- electron gas: χ_{ij} is isotropic in spin space; Heisenberg magnet
- 2D hole gas, only HH-LH splitting taken into account: $\chi_{zz} >> \chi_{xx,yy} \approx 0$; Ising magnet



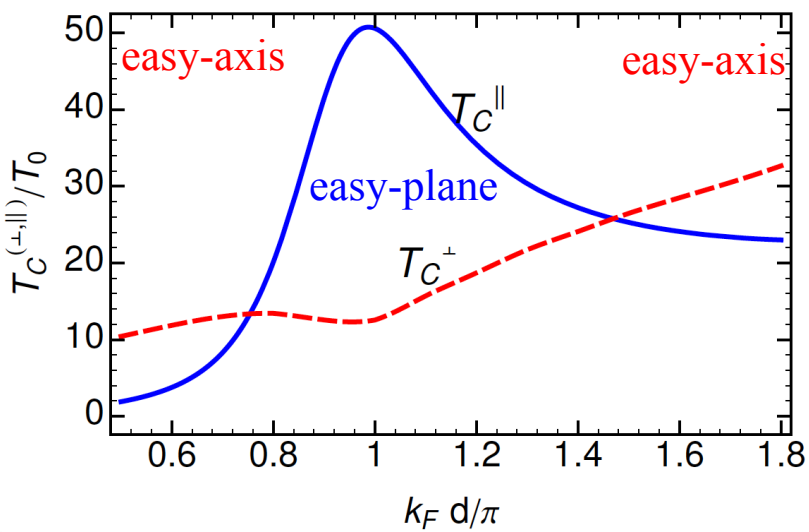
Haury et al., PRL (1997); Wurstbauer et al., Nat. Phys. (2010)

RKKY interaction/magnetism for 2D holes

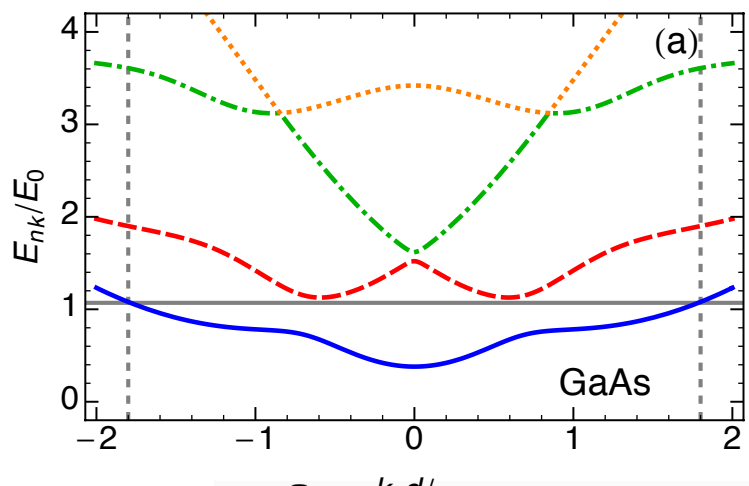
- perform mean-field analysis of RKKY exchange-coupling Hamiltonian ⇒ Curie temperature T_{C}

$$T_{\mathrm{C}}^{(\perp,\parallel)} = \frac{S(S+1)}{3}\,\frac{G^2}{k_{\mathrm{B}}}\,\frac{n_{\mathrm{imp}}}{d}\,|\bar{\chi}_{zz,xx}(\mathbf{q}=0)|$$

- HH-LH mixing: important even for lowest subband
  - easy-axis/easy-plane transitions (+helical magnetism?)



Kernreiter, Governale & UZ, PRL (2013)



Physical ramifications of valence-band mixing

III. Exchange energy

Kernreiter, Governale, Winkler, UZ, Phys. Rev. B **88**, 125309 (2013)

Taking account of Coulomb interactions

- perturbatively for electron gas: change of ground-state energy Giuliani & Vignale, *Quantum Theory of the Electron Liquid*

$$E_{\rm gs} \approx \langle {\rm FS} \,|\, \mathcal{H}_0 + \mathcal{H}_{\rm int} \,|\, {\rm FS} \rangle = E_0 + E_{\rm H} + E_{\rm X}$$

  – consider exchange-energy contribution $E_{\rm X}$

- general expression for 2D system:

$$\frac{E_{\rm X}}{N} = -\frac{1}{2\rho} \sum_{n,n'} \int \frac{d^2k}{(2\pi)^2} \int \frac{d^2k'}{(2\pi)^2} \, V^{(nn')}_{\mathbf{k}\mathbf{k}'} \, n_F(E_{n'\mathbf{k}'}) \, n_F(E_{n\mathbf{k}})$$

  – matrix element of two-particle interaction potential affected by quantum-well state: $V^{(nn')}_{\mathbf{k}\mathbf{k}'} = \frac{e^2/2\epsilon\epsilon_0}{|\mathbf{k}-\mathbf{k}'|} \sum_{\nu,\nu'} F^{(nn')}_{\mathbf{k}\mathbf{k}',\nu\nu'}$

Previously known results & a puzzle

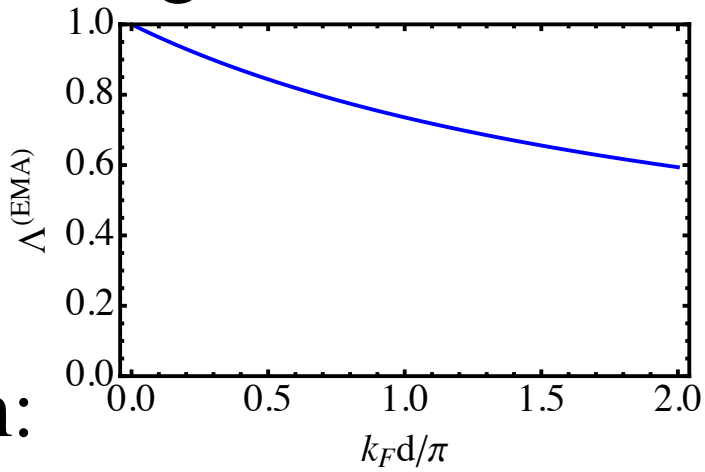
- for strictly 2D electron system: $\frac{E_{\rm X}^{(0)}}{N} = -\frac{e^2}{\epsilon\epsilon_0}\frac{k_{\rm F}}{3\pi^2}$

  Chaplik, JETP (1971); Stern, PRL (1973)

- quasi-2D system with parabolic subbands: finite width *d* results in reduction of exchange effects

  Betbeder-Matibet et al., PRL (1994)

$$\frac{E_{\rm X}^{(\rm EMA)}}{N} = \frac{E_{\rm X}^{(0)}}{N}\Lambda^{(\rm EMA)}(k_{\rm F}d)$$



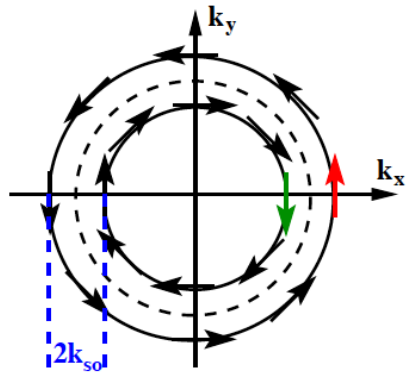
- curious experimental observation: seemingly no exchange effects in 2D hole systems

  Winkler et al., PRB (2005)

Exchange in `spin-orbit-coupled´ systems

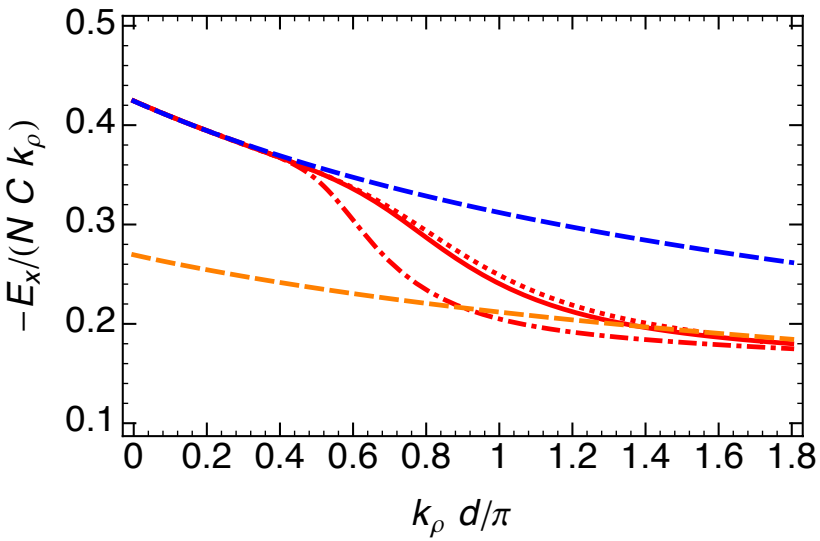
- idea: `chirality´/spinor structure of electronic states affects Coulomb matrix element/form factor
  - spin-rotationally invariant case: can use global quantization axis for spin; exchange matrix element only finite between states with same spin projection
  - in systems with spin-momentum locking (2D system with Rashba spin splitting): no global quantization axis exists; average spin overlap of eigenstates reduced ⇒ exchange reduced (?)

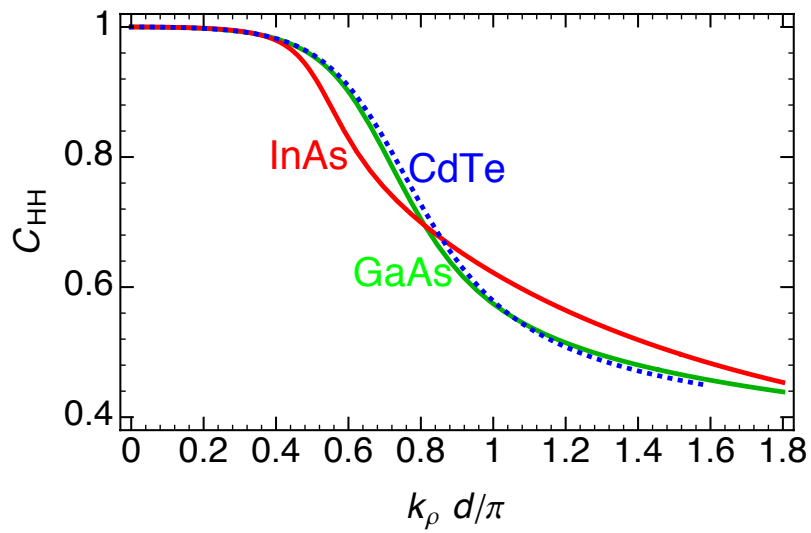
$$V_{\mathbf{k}\mathbf{k}'}^{(nn')} = \frac{e^2/2\epsilon\epsilon_0}{|\mathbf{k}-\mathbf{k}'|} \sum_{\nu,\nu'} F_{\mathbf{k}\mathbf{k}',\nu\nu'}^{(nn')}$$



Exchange reduction in model 2D hole system

- Rashba spin splitting has almost no effect on 2D-electron exchange energy Chesi & Giuliani, PRB (2007, 2011)
- in contrast: HH-LH mixing in 2D hole systems results in significant reduction Kernreiter et al. PRB (2013)
  - exchange suppression correlated w/ spinor mixture





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Conclusions

- quantum confinement of holes introduces HH-LH splitting and HH-LH mixing
  - HH-LH mixing renders properties of confined holes to be much different from conduction electrons
- differences relevant even in single-2D-subband limit and in the absence of magnetic fields
  - carrier-density-controlled anisotropy of spin response
  - significant suppression of exchange energy
- presented work has resulted from collaboration with M. Governale, T. Kernreiter, R. Winkler