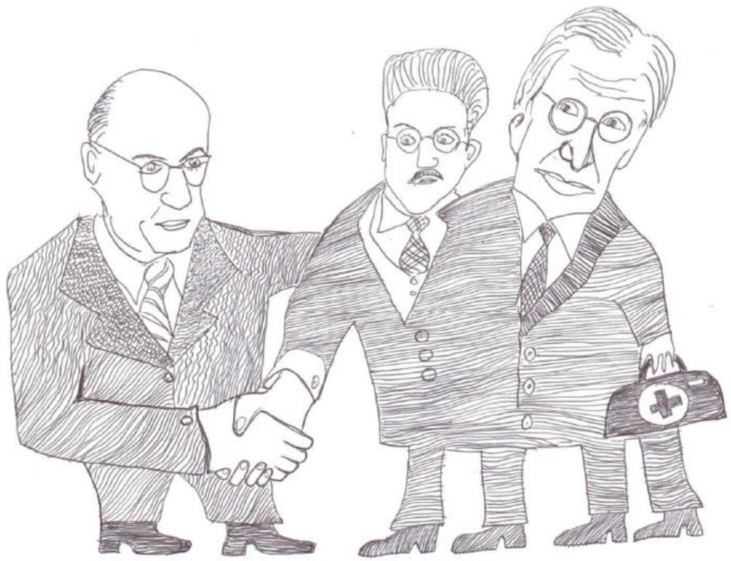
Spin Dynamics of Excitons

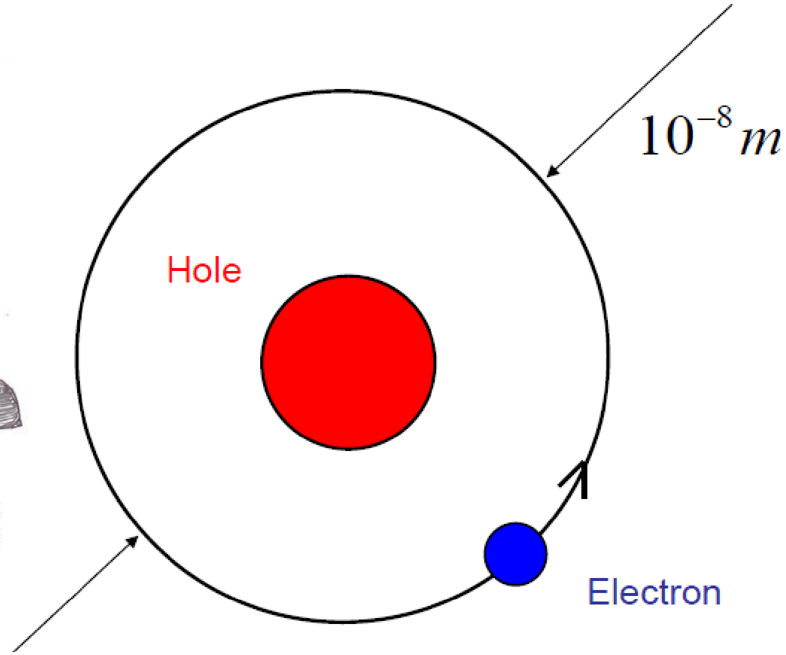
Alexey Kavokin

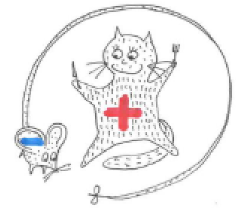
Physics and Astronomy School, University of Southampton, Highfield, Southampton, SO171BJ, UK

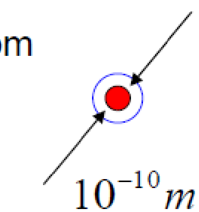
Spin Optics Laboratory, St-Petersburg State University, 1, Ulianovskaya, St-Petersburg, Russia.

Excitons are electron-hole pairs bound by Coulomb interaction







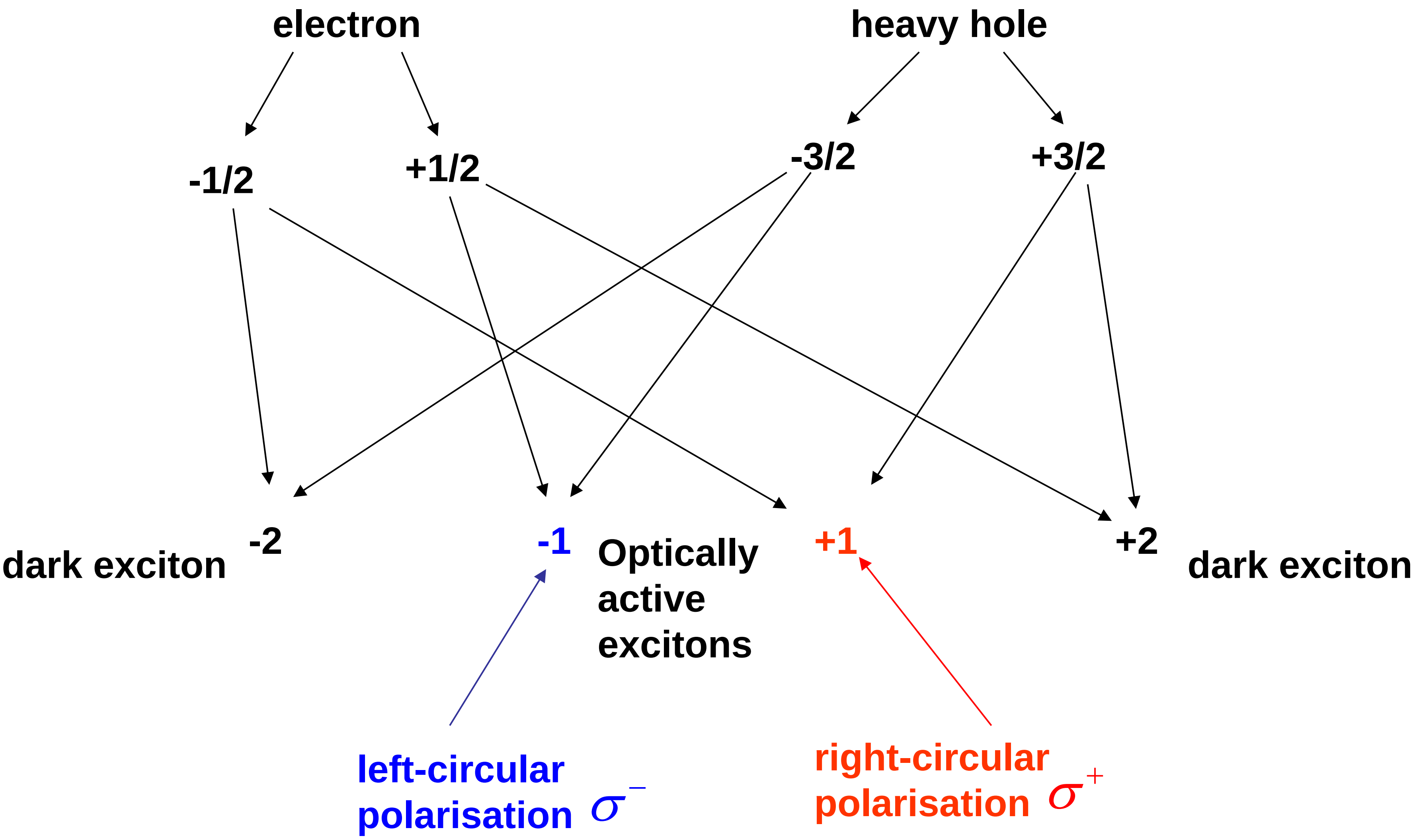


Electron = fermion
Hole = fermion
Exciton = electron + hole = boson

Yakov Il'ich **Frenkel** (1894–1952), Sir Nevill. Francis **Mott** (1905–1996) and Grégory **Wannier** (1911–1983) gave their name to the two main categories of excitons.

Zinc-blend semiconductor quantum wells:

Exciton spin structure



Exciton spin density matrix

Exciton spinor wave-function (no coordinate-dependent part here)

$$\Psi = (\Psi_{+1}, \Psi_{-1}, \Psi_{+2}, \Psi_{-2},) = \left(\Psi_{e,-1/2}\Psi_{h,+3/2}, \Psi_{e,+1/2}\Psi_{h,-3/2}, \Psi_{e,+1/2}\Psi_{h,+3/2}, \Psi_{e,-1/2}\Psi_{h,-3/2}\right)$$

Normalisation condition

$$\Psi_{+1}\Psi^*_{+1} + \Psi_{-1}\Psi^*_{-1} + \Psi_{+2}\Psi^*_{+2} + \Psi_{-2}\Psi^*_{-2} = \Psi_{e,+\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}} + \Psi_{e,-\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}} = \Psi_{h,+\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} + \Psi_{h,-\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} = 1$$

Exciton spin-density matrix:

$$\hat{\rho} = |\Psi\rangle\langle\Psi| = \begin{bmatrix} \Psi_{+1}\Psi^*_{+1} & \Psi_{+1}\Psi^*_{-1} & \Psi_{+1}\Psi^*_{+2} & \Psi_{+1}\Psi^*_{-2} \\ \Psi_{-1}\Psi^*_{+1} & \Psi_{-1}\Psi^*_{-1} & \Psi_{-1}\Psi^*_{+2} & \Psi_{-1}\Psi^*_{-2} \\ \Psi_{+2}\Psi^*_{+1} & \Psi_{+2}\Psi^*_{-1} & \Psi_{+2}\Psi^*_{+2} & \Psi_{+2}\Psi^*_{-2} \\ \Psi_{-2}\Psi^*_{+1} & \Psi_{-2}\Psi^*_{-1} & \Psi_{-2}\Psi^*_{+2} & \Psi_{-2}\Psi^*_{-2} \end{bmatrix}$$

The polarisation degrees are given by:

$$\rho_l = \frac{2S_x}{I} = (\rho_{12} + \rho_{21})/(\rho_{11} + \rho_{22})$$

$$\rho_c = \frac{2S_z}{I} = (\rho_{11} - \rho_{22})/(\rho_{11} + \rho_{22})$$

Relation between electron, hole and exciton spin density matrices

$$\hat{\rho} = |\Psi\rangle\langle\Psi| = \begin{bmatrix} \Psi_{+1}\Psi^*_{+1} & \Psi_{+1}\Psi^*_{-1} & \Psi_{+1}\Psi^*_{+2} & \Psi_{+1}\Psi^*_{-2} \\ \Psi_{-1}\Psi^*_{+1} & \Psi_{-1}\Psi^*_{-1} & \Psi_{-1}\Psi^*_{+2} & \Psi_{-1}\Psi^*_{-2} \\ \Psi_{+2}\Psi^*_{+1} & \Psi_{+2}\Psi^*_{-1} & \Psi_{+2}\Psi^*_{+2} & \Psi_{+2}\Psi^*_{-2} \\ \Psi_{-2}\Psi^*_{+1} & \Psi_{-2}\Psi^*_{-1} & \Psi_{-2}\Psi^*_{+2} & \Psi_{-2}\Psi^*_{-2} \end{bmatrix} =$$

$$= \begin{bmatrix} \Psi_{e,-\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} \\ \Psi_{e,+\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} \\ \Psi_{e,+\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,+\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} \\ \Psi_{e,-\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}}\Psi_{h,-\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} \end{bmatrix}$$

Elements of electron and hole density matrices can be deduced:

$$\hat{\rho}_e = |\Psi_e\rangle\langle\Psi_e| = \begin{bmatrix} \Psi_{e,+\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}} & \Psi_{e,+\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}} \\ \Psi_{e,-\frac{1}{2}}\Psi^*_{e,+\frac{1}{2}} & \Psi_{e,-\frac{1}{2}}\Psi^*_{e,-\frac{1}{2}} \end{bmatrix} = \begin{bmatrix} \rho_{22}+\rho_{33} & \rho_{24}+\rho_{31} \\ \rho_{13}+\rho_{42} & \rho_{11}+\rho_{44} \end{bmatrix}$$

$$\hat{\rho}_h = |\Psi_h\rangle\langle\Psi_h| = \begin{bmatrix} \Psi_{h,+\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{h,+\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} \\ \Psi_{h,-\frac{3}{2}}\Psi^*_{h,+\frac{3}{2}} & \Psi_{h,-\frac{3}{2}}\Psi^*_{h,-\frac{3}{2}} \end{bmatrix} = \begin{bmatrix} \rho_{11}+\rho_{33} & \rho_{14}+\rho_{32} \\ \rho_{23}+\rho_{41} & \rho_{22}+\rho_{44} \end{bmatrix}$$

Here we used the normalisation condition: the trace of each spin density matrix = 1

Electron and hole spins can be obtained from the exciton density matrix elements!

$$\hat{\rho}_e = \begin{bmatrix} \frac{1}{2} + S_{e,z} & S_{e,x} - iS_{e,y} \\ S_{e,x} + iS_{e,y} & \frac{1}{2} - S_{e,z} \end{bmatrix}, \hat{\rho}_h = \begin{bmatrix} \frac{1}{2} + S_{h,z} & S_{h,x} - iS_{h,y} \\ S_{h,x} + iS_{h,y} & \frac{1}{2} - S_{h,z} \end{bmatrix}$$

$$S_{e,z} = (\rho_{22} + \rho_{33} - \rho_{11} - \rho_{44})/2,$$

$$S_{h,z} = (\rho_{11} + \rho_{33} - \rho_{22} - \rho_{44})/2.$$

$$S_{e,x} = (\rho_{13} + \rho_{31} + \rho_{24} + \rho_{42})/2,$$

$$S_{h,x} = (\rho_{14} + \rho_{23} + \rho_{32} + \rho_{41})/2,$$

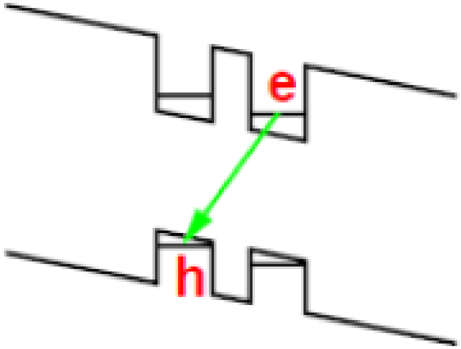
$$S_{e,y} = i(-\rho_{13} + \rho_{31} + \rho_{24} - \rho_{42})/2,$$

$$S_{h,y} = i(\rho_{14} - \rho_{23} + \rho_{32} - \rho_{41})/2.$$

All we know from traditional spintronics can be applied to excitons once we remember that they are composed by electrons and holes.
From exciton polarisation one can deduce electron and hole spins.

Spin dynamics of spatially indirect excitons in coupled quantum wells

L.V. Butov, San Diego



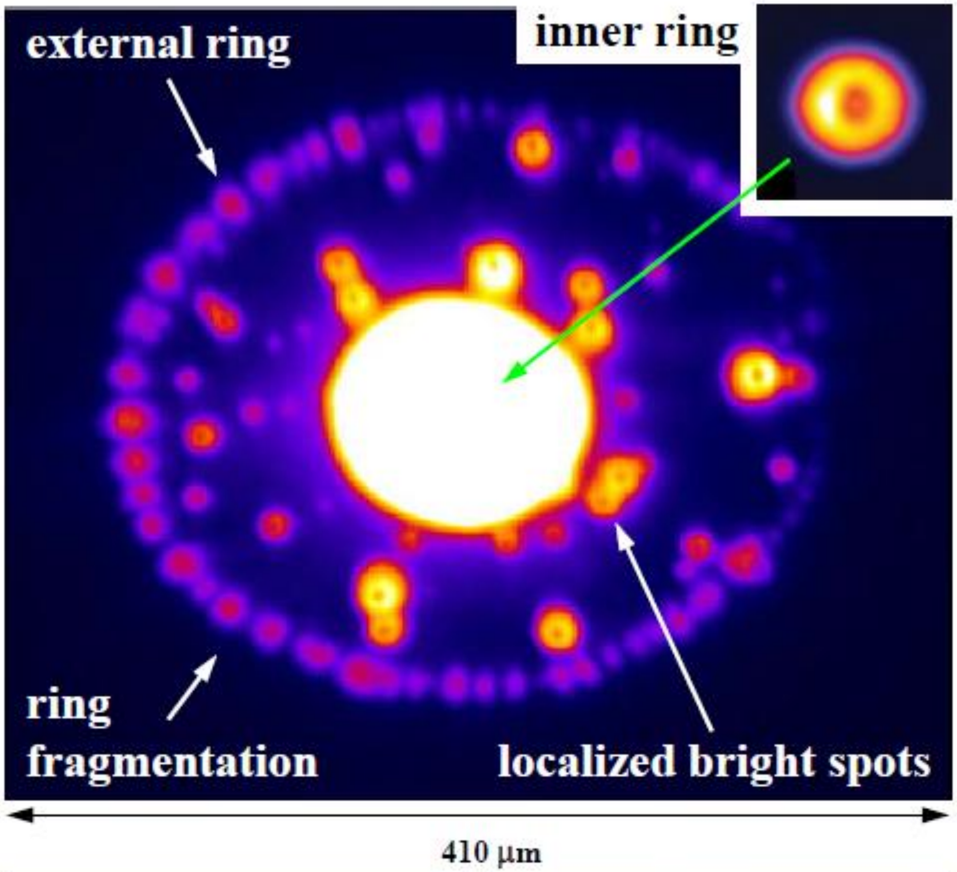
D. Snoke, Pittsburgh

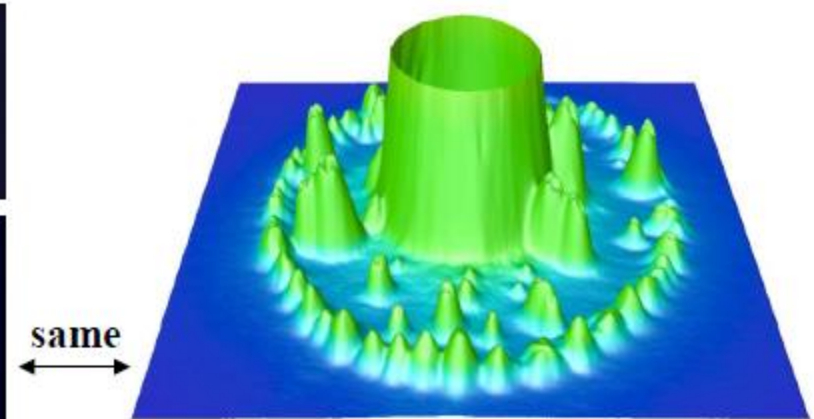
- **Controllable exciton life-time**
- **Controllable exciton interaction strength**
- **Controllable exchange splitting of dark-bright excitons**

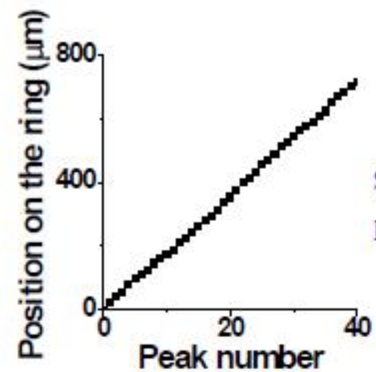
$$k_B T_{BKT} = \frac{\pi \hbar^2 n}{2m} \approx (1-2)K$$

D.R. Nelson and J.M. Kosterlitz, Phys. Rev. Lett., 39, 1977

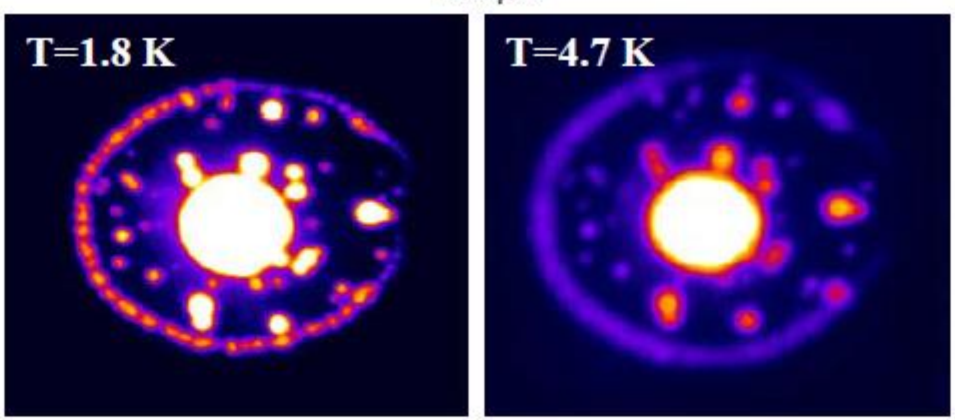
Pattern Formation: Exciton Rings and Macroscopically Ordered Exciton State

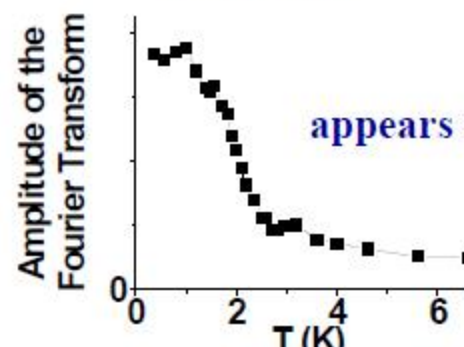






spatial order on macroscopic lengths





appears abruptly at low T

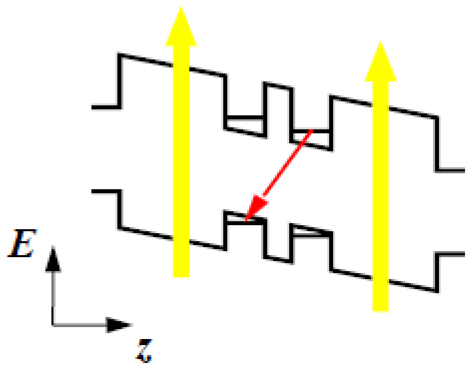
L.V. Butov, A.C. Gossard, D.S. Chemla, Nature 418, 751 (2002)

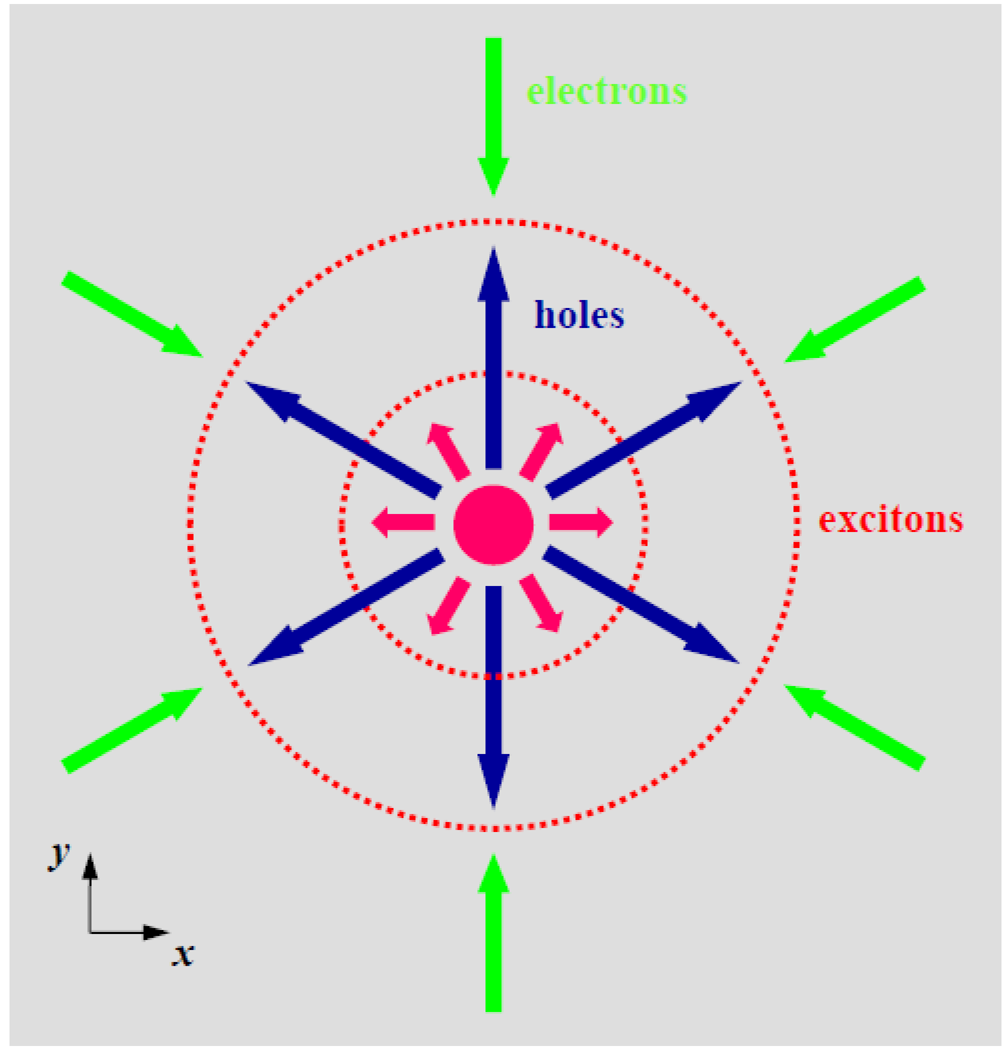
External ring

above barrier laser excitation creates
additional number of holes in CQW

⬆

heavier holes have higher
collection efficiency to CQW



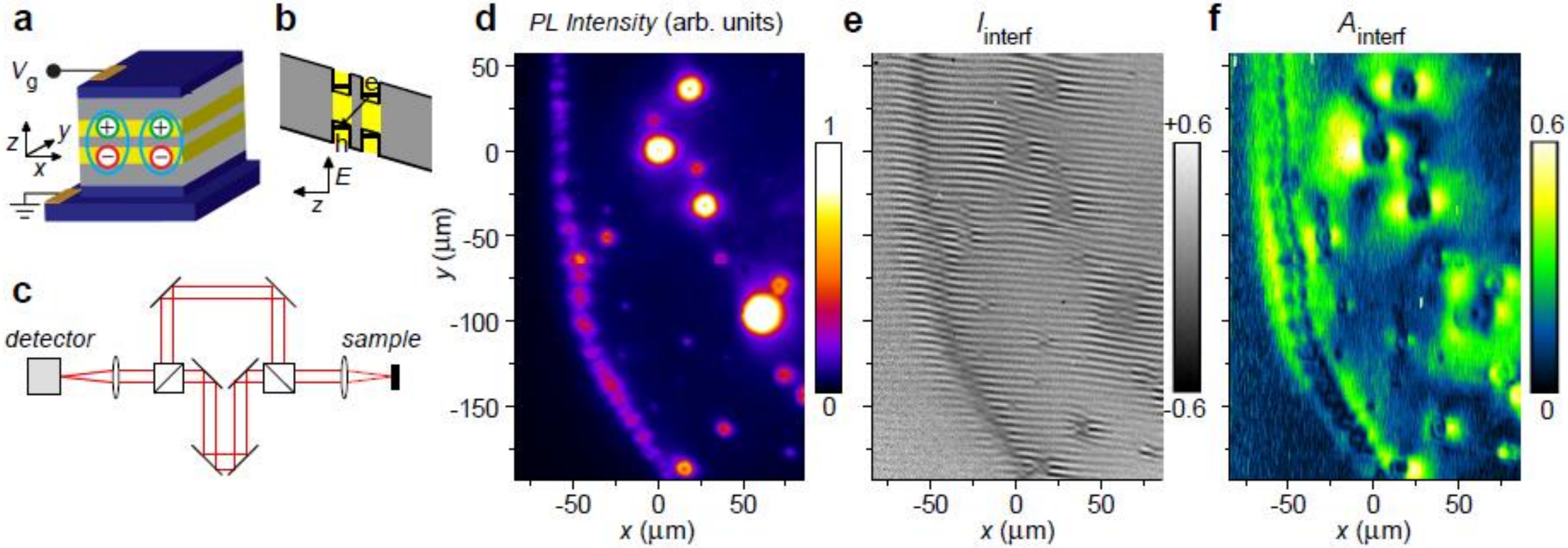


external ring forms at interface between
electron-rich and hole-rich regions

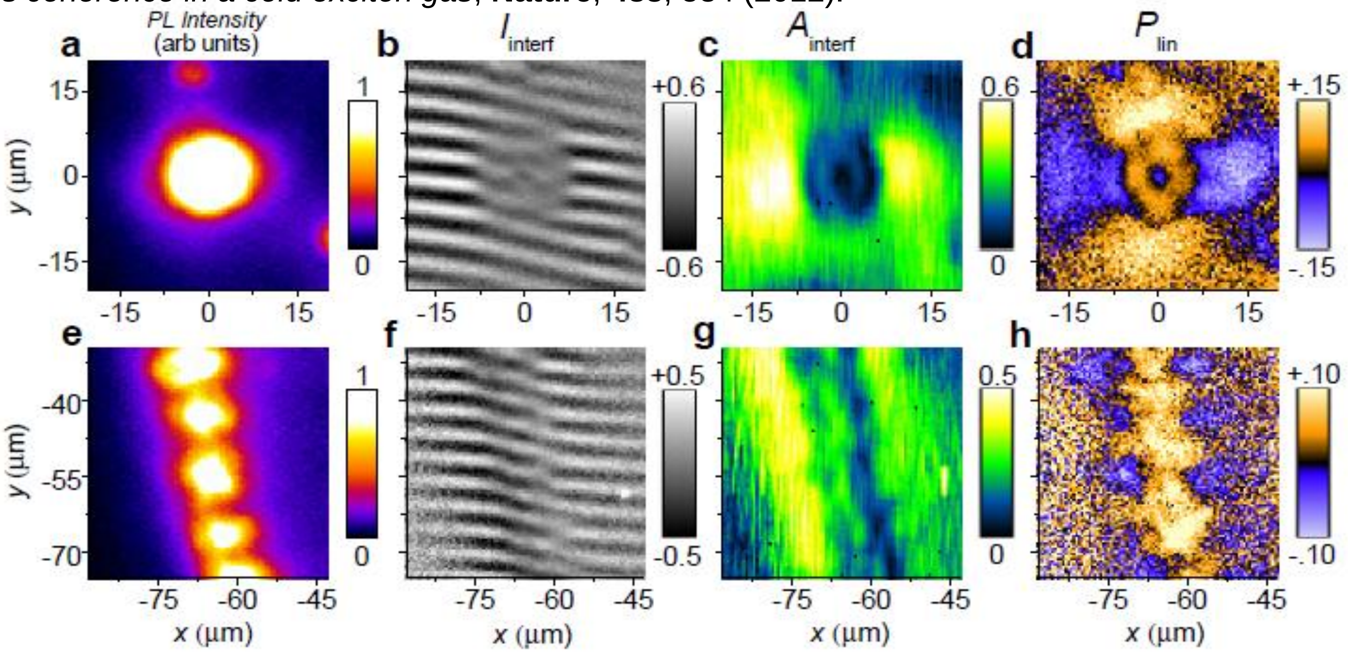
L.V. Butov, L.S. Levitov, B.D. Simons, A.V. Mintsev, A.C. Gossard, D.S. Chemla, PRL 92, 117404 (2004)

R. Rapaport, G. Chen, D. Snoke, S.H. Simon, L. Pfeiffer, K.West, Y.Liu, S.Denev, PRL 92, 117405 (2004)

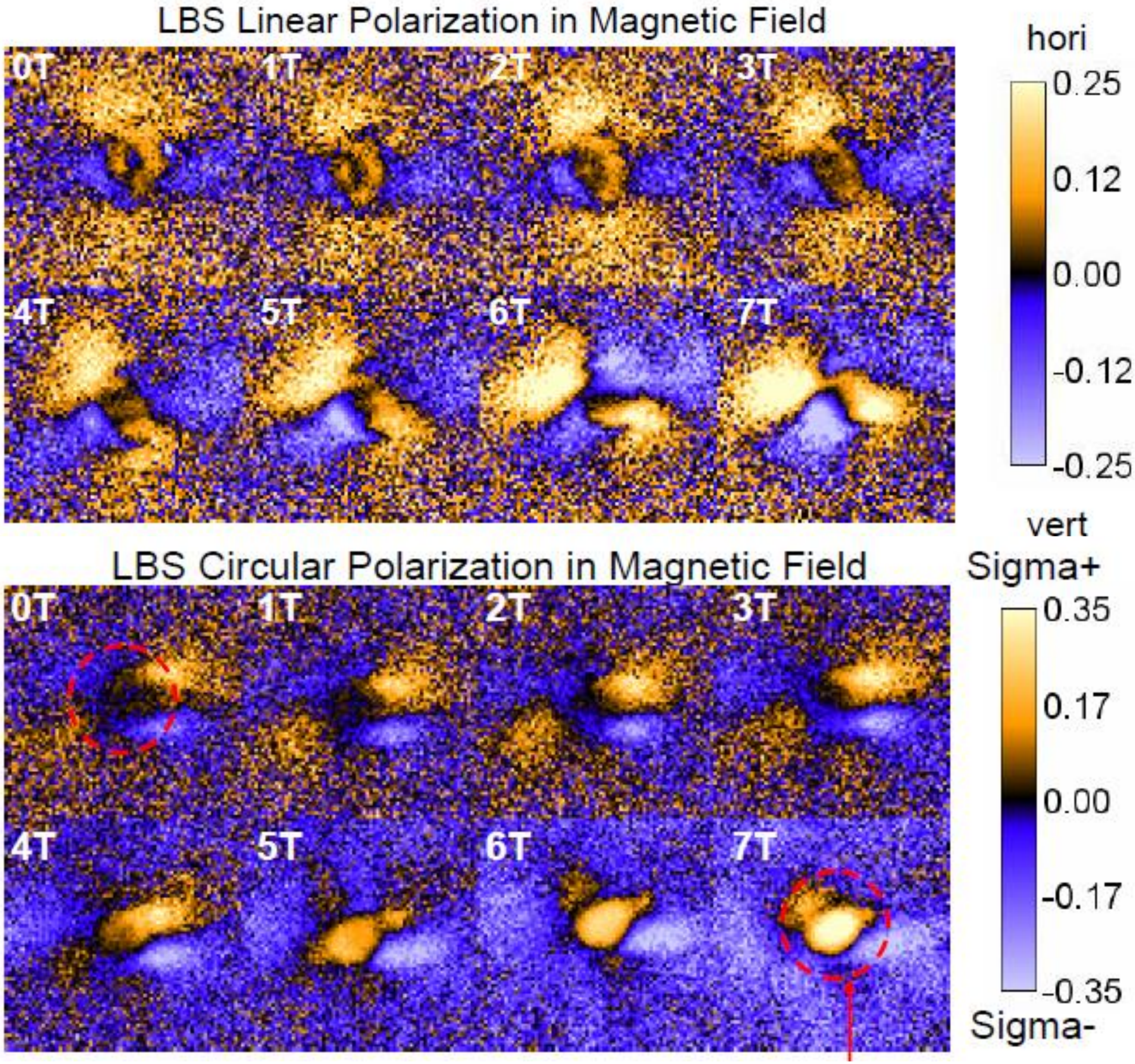
Appearance of polarisation textures correlated with the build up of spatial coherence

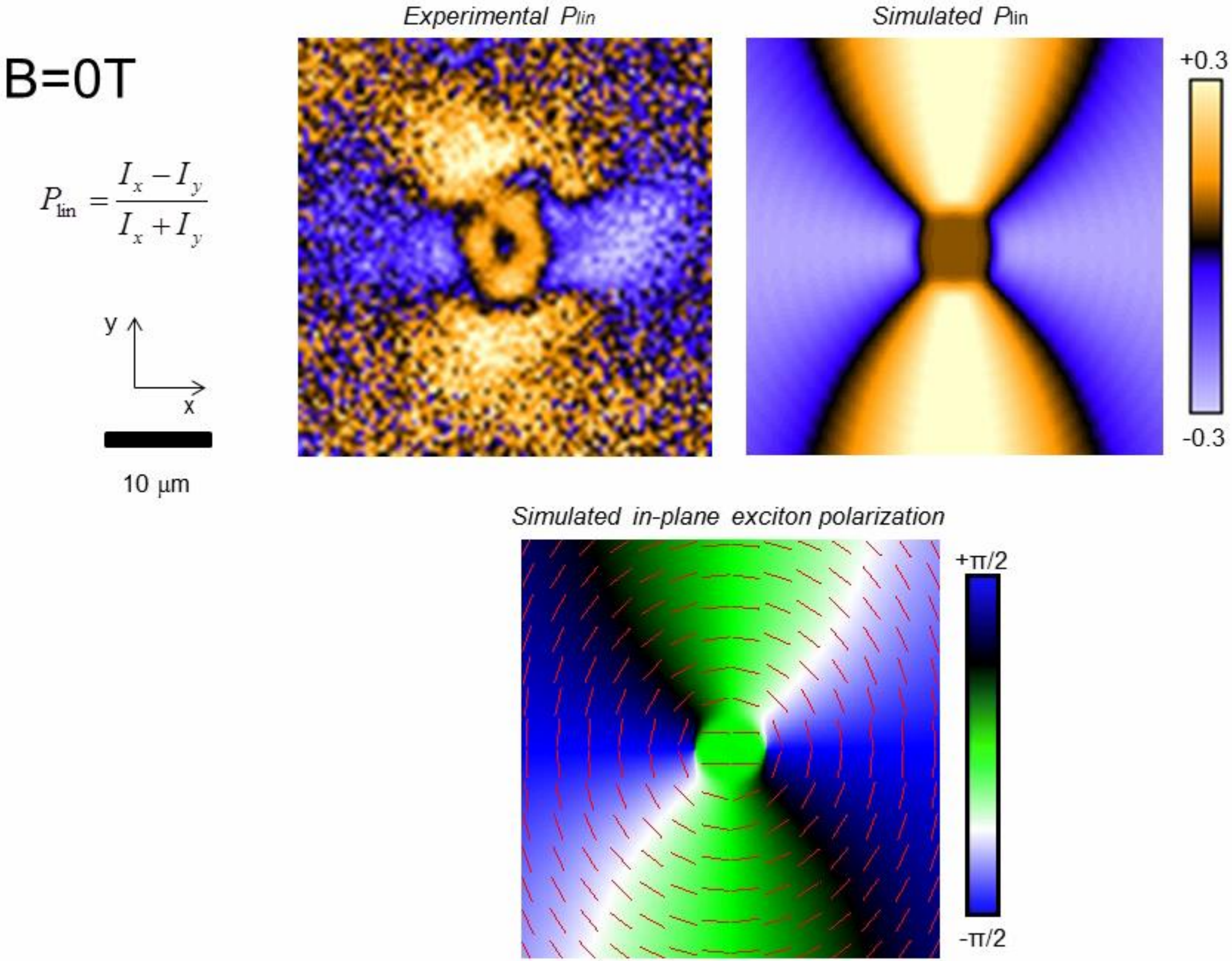


A. A. High, J. R. Leonard, A. T. Hammack, M. M. Fogler, L. V. Butov, A. V. Kavokin, K. L. Campman & A. C. Gossard, *Spontaneous coherence in a cold exciton gas*, **Nature**, **483**, 584 (2012).



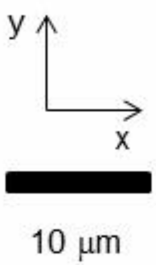
Magnetic field effect (unpublished)

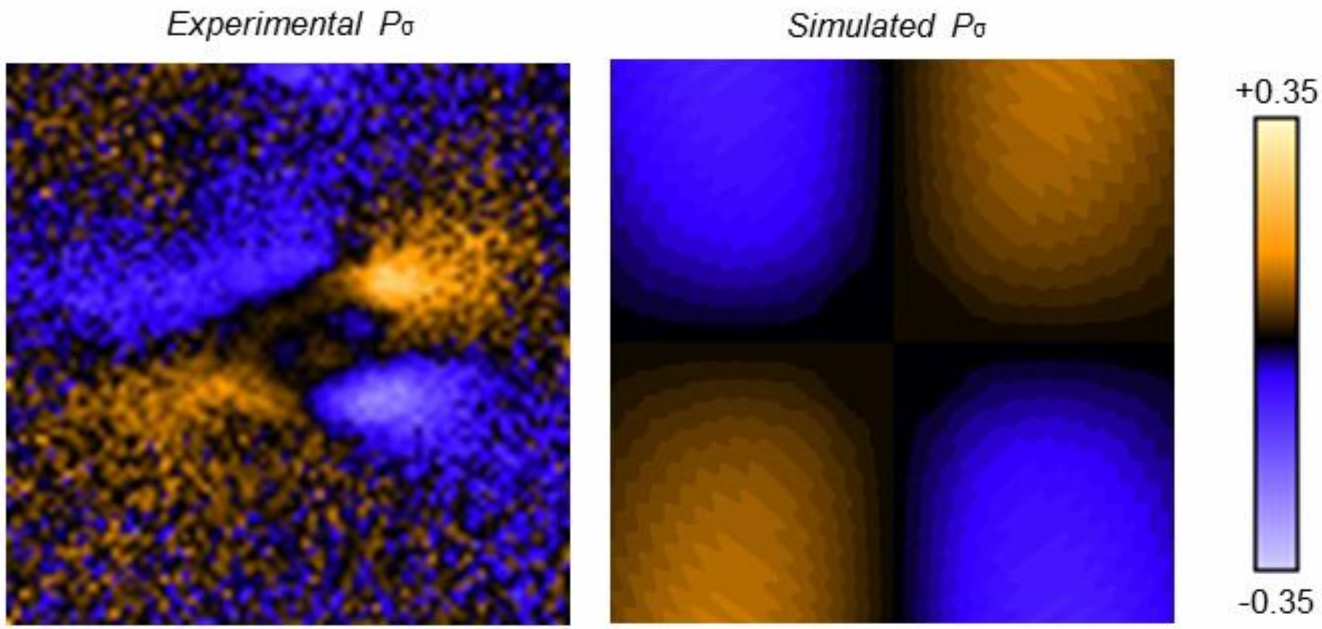



B=0T
$P_{\text{lin}} = \frac{I_x - I_y}{I_x + I_y}$
y
x
10 μm
Experimental P_{lin}
Simulated P_{lin}
+0.3
-0.3
Simulated in-plane exciton polarization
+π/2
-π/2

B=0T

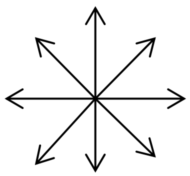
$$P_\sigma = \frac{I_{\sigma^+} - I_{\sigma^-}}{I_{\sigma^+} + I_{\sigma^-}}$$





Linear model of polarisation textures in cold exciton gases:

- Excitons propagate ballistically in radial directions; *r=vt*
- Dark and bright excitons are mixed by spin-orbit interaction.
- Electron and hole spins are rotated by the Dresselhaus field;
- Supplementary beats appear due to linear polarisation splittings;



We solve the Liouville equation for a 4x4 spin density matrix:

$$i\hbar\frac{d\hat{\rho}}{dt} = \left[\hat{\mathcal{H}}_{ex}^{tot}, \hat{\rho}\right] \qquad \hat{\rho} = |\Psi><\Psi| \qquad \Psi = (\Psi_{+1}, \Psi_{-1}, \Psi_{+2}, \Psi_{-2})$$

The polarisation degrees are calculated from:

$$\rho_l = \frac{2S_x}{I} = (\rho_{12} + \rho_{21})/(\rho_{11} + \rho_{22})$$

$$\rho_c = \frac{2S_z}{I} = (\rho_{11} - \rho_{22})/(\rho_{11} + \rho_{22})$$

What contributes to the exciton Hamiltonian?

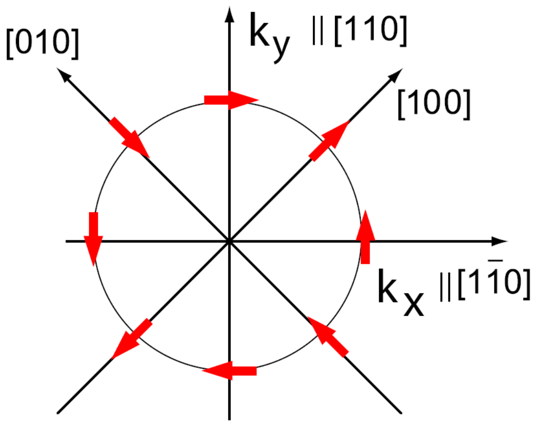
Dresselhaus and Zeeman effects on electrons:

Dresselhaus spin-orbit Hamiltonian

$$H_{eff(e,h)} = -\frac{1}{2} g_{e,h} \mu_B \left(\mathbf{B}_{eff} \hat{\sigma} \right)$$

$$-\frac{1}{2} g_e \mu_B \mathbf{B}_{eff} = \beta_e \left(k_{e,x}, -k_{e,y} \right)$$

$$H_e = \beta_e \left(k_{e,x} \sigma_x - k_{e,y} \sigma_y \right) - \frac{1}{2} g_e \mu_B B \sigma_z$$



Dresselhaus and Zeeman effects on electrons: exciton Hamiltonian

P. Zeeman

$$H_e = \beta_e\left(k_{e,x}\sigma_x - k_{e,y}\sigma_y\right) - \frac{1}{2} g_e \mu_B B \sigma_z$$

Hence:

$$H_e = \begin{bmatrix} -\frac{1}{2} g_e \mu_B B & \beta_e\left(k_{e,x} + i k_{e,y}\right) \\ \beta_e\left(k_{e,x} - i k_{e,y}\right) & \frac{1}{2} g_e \mu_B B \end{bmatrix} = \begin{bmatrix} -\frac{1}{2} g_e \mu_B B & \beta_e k_e e^{i\varphi} \\ \beta_e k_e e^{-i\varphi} & \frac{1}{2} g_e \mu_B B \end{bmatrix}.$$

In the exciton basis:

$$(-1/2,\ +1/2,\ +1/2,\ -1/2) \longrightarrow (+1,-1,+2,-2)$$

$$\widehat{H_e} = \begin{bmatrix} g_e \mu_B B/2 & 0 & k_e \beta_e e^{-i\varphi} & 0 \\ 0 & -g_e \mu_B B/2 & 0 & k_e \beta_e e^{i\varphi} \\ k_e \beta_e e^{i\varphi} & 0 & -g_e \mu_B B/2 & 0 \\ 0 & k_e \beta_e e^{-i\varphi} & 0 & g_e \mu_B B/2 \end{bmatrix}.$$

Dresselhaus and Zeeman effects for holes: $-\frac{1}{2}g_h\mu_B\mathbf{B}_{eff} = \beta_h\left(k_{e,x}, k_{e,y}\right)$

In the (+3/2,-3/2) basis:

$$H_h = \beta_h\left(k_{h,x}\sigma_x + k_{h,y}\sigma_y\right) - \frac{1}{2}g_h\mu_B B\sigma_z.$$

$$H_h = \begin{bmatrix} -\frac{1}{2}g_h\mu_B B & \beta_h\left(k_{h,x} - ik_{h,y}\right) \\ \beta_h\left(k_{h,x} + ik_{h,y}\right) & \frac{1}{2}g_h\mu_B B \end{bmatrix} = \begin{bmatrix} -\frac{1}{2}g_h\mu_B B & \beta_h k_h e^{-i\varphi} \\ \beta_h k_h e^{i\varphi} & \frac{1}{2}g_h\mu_B B \end{bmatrix}$$

In the exciton basis:

(+3/2, -3/2, +3/2, -3/2) $\longrightarrow$ (+1,-1,+2,-2)

$$\widehat{H_h} = \begin{bmatrix} -g_h\mu_B B/2 & 0 & 0 & k_h\beta_h e^{-i\varphi} \\ 0 & g_h\mu_B B/2 & k_h\beta_h e^{i\varphi} & 0 \\ 0 & k_h\beta_h e^{-i\varphi} & -g_h\mu_B B/2 & 0 \\ k_h\beta_h e^{i\varphi} & 0 & 0 & g_h\mu_B B/2 \end{bmatrix}.$$

Long- and short-range exchange splittings of 4 exciton states:

Splitting of bright excitons:

$$H_b = E_b I - \delta_b \sigma_x = \begin{bmatrix} E_b & -\delta_b \\ -\delta_b & E_b \end{bmatrix},$$

S.V. Gupalov, E.L. Ivchenko, A.V. Kavokin, *Fine Structure of Localized Exciton Levels in Quantum Wells*, J. Experim. & Theor. Phys. **86**, 388 (1998).

Splitting of dark excitons:

$$H_d = E_d I - \delta_d \sigma_x = \begin{bmatrix} E_d & -\delta_d \\ -\delta_d & E_d \end{bmatrix}$$

In the exciton basis (+1,-1,+2,-2)

$$H_0 = \begin{bmatrix} E_b & -\delta_b & 0 & 0 \\ -\delta_b & E_b & 0 & 0 \\ 0 & 0 & E_d & -\delta_d \\ 0 & 0 & -\delta_d & E_d \end{bmatrix}.$$

Al. L. Efros and M. Rosen, The Electronic Structure of Semiconductor Nanocrystals, Annual Review of Material Science 30, 475 (2000).

All terms together, we obtain the exciton Hamiltonian:

$$\widehat{H} = \begin{bmatrix} E_b - (g_h - g_e)\mu_B B/2 & -\delta_b & k_e\beta_e e^{-i\varphi} & k_h\beta_h e^{-i\varphi} \\ -\delta_b & E_b + (g_h - g_e)\mu_B B/2 & k_h\beta_h e^{i\varphi} & k_e\beta_e e^{i\varphi} \\ k_e\beta_e e^{i\varphi} & k_h\beta_h e^{-i\varphi} & E_d - (g_h + g_e)\mu_B B/2 & -\delta_d \\ k_h\beta_h e^{i\varphi} & k_e\beta_e e^{-i\varphi} & -\delta_d & E_d + (g_h + g_e)\mu_B B/2 \end{bmatrix}.$$

$k_{ex} = k_h + k_e$ (center of mass wave-vector)

$$k_e = \frac{m_e}{m_e + m_{hh}} k_{ex}, \qquad k_h = \frac{m_{hh}}{m_e + m_{hh}} k_{ex}.$$

Averaging over relative electron-hole motion: fruitful discussion with N. Gippius!

Initial state (t=0) is assumed to be thermal: no external coherence!

---

we take $k_{ex} = 0$

$$\widehat{H_0} = \begin{bmatrix} E_b - (g_h - g_e)\mu_B B/2 & -\delta_b & 0 & 0 \\ -\delta_b & E_b + (g_h - g_e)\mu_B B/2 & 0 & 0 \\ 0 & 0 & E_d - (g_h + g_e)\mu_B B/2 & -\delta_d \\ 0 & 0 & -\delta_d & E_d + (g_h + g_e)\mu_B B/2 \end{bmatrix}.$$

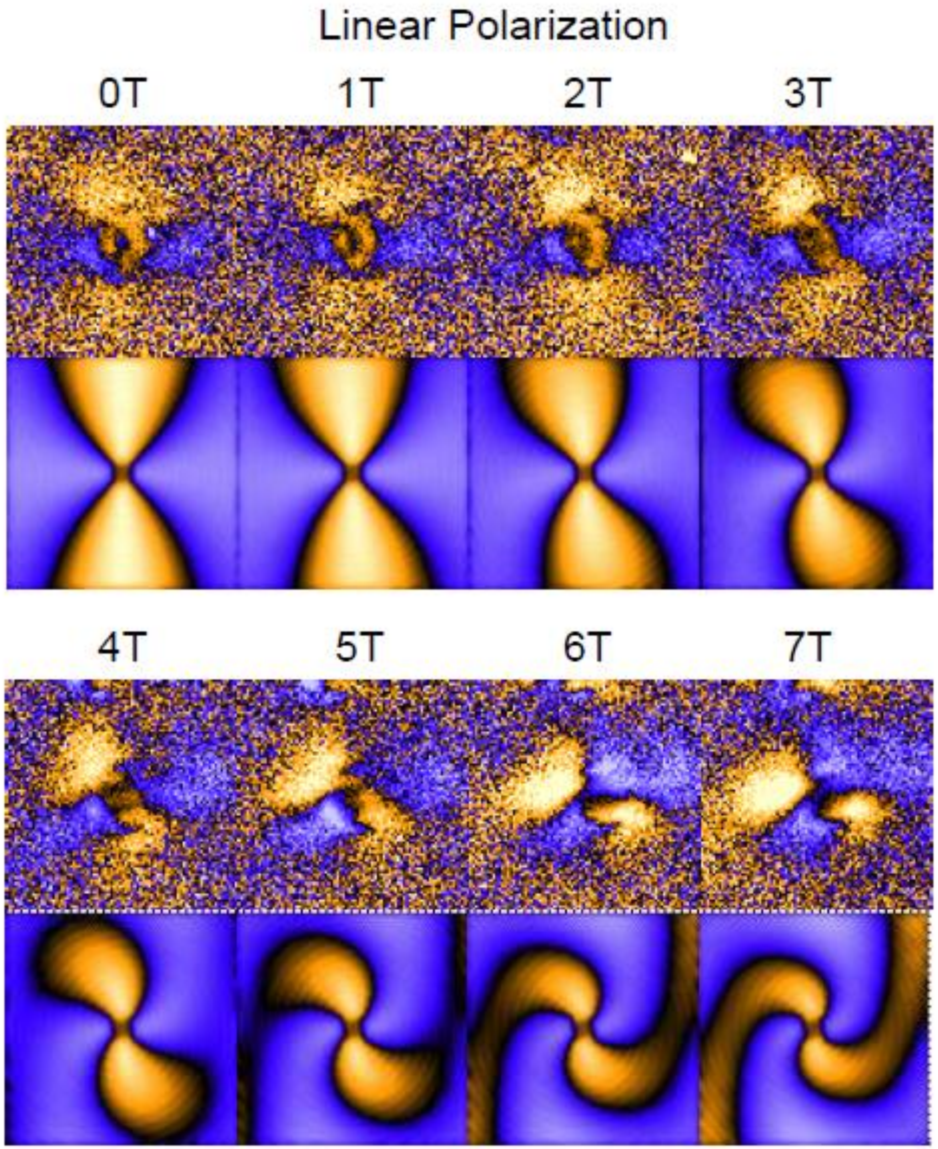
we diagonalise this Hamiltonian and find 4 eigen states

we assume that their populations are related to each other by the Boltzmann law

we assume no coherence in the system (no correlations between 4 states)

Thus we obtain $\hat{\rho}_0$

Theory vs experiment:



$$g_e = 0.010$$

$$g_h = -0.0085$$

$$E_b - E_d = -1\mu eV$$

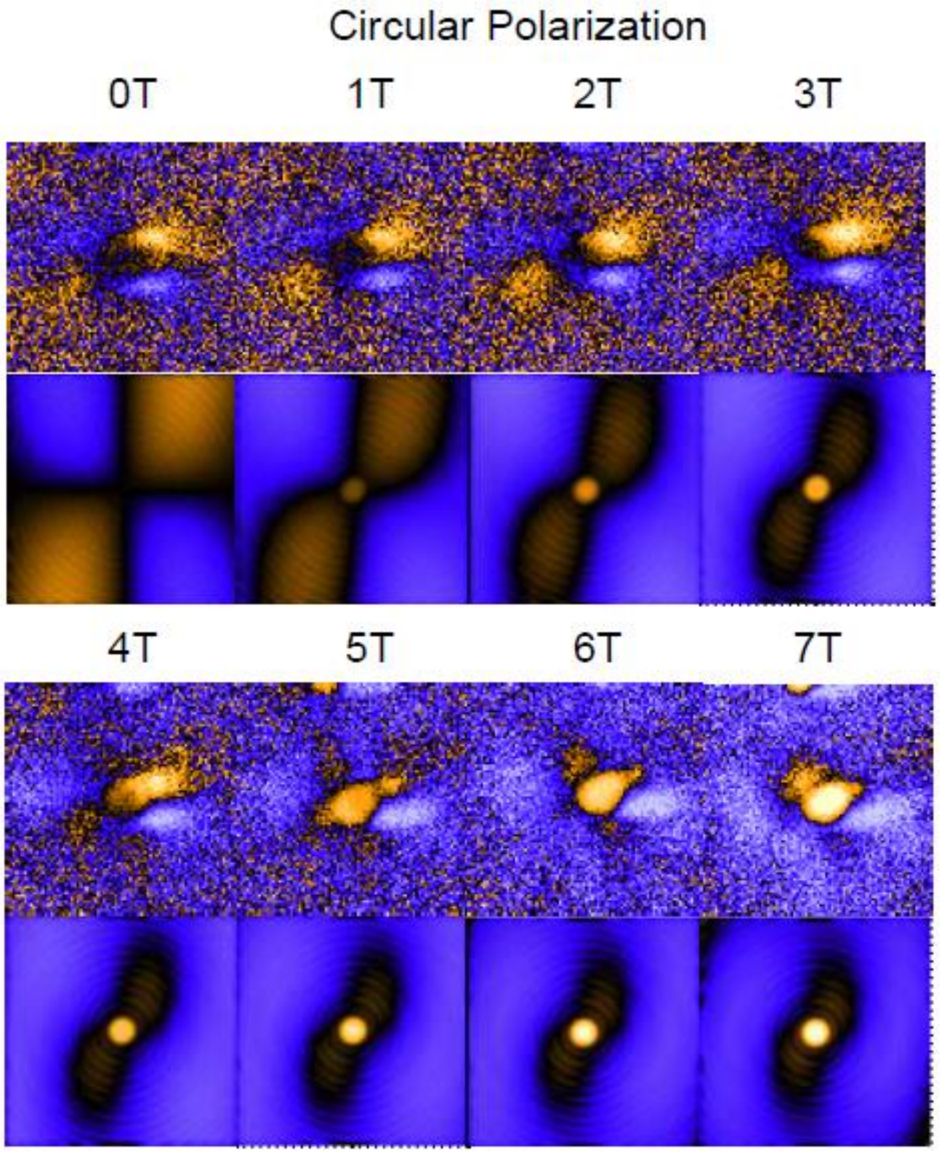
$$\delta_b = 0.55\mu eV$$

$$\delta_d = -13\mu eV$$

$$k = 15.4\mu m^{-1}$$

T=0.1 K

Theory vs experiment:



$$g_e = 0.010$$

$$g_h = -0.0085$$

$$E_b - E_d = -1\mu eV$$

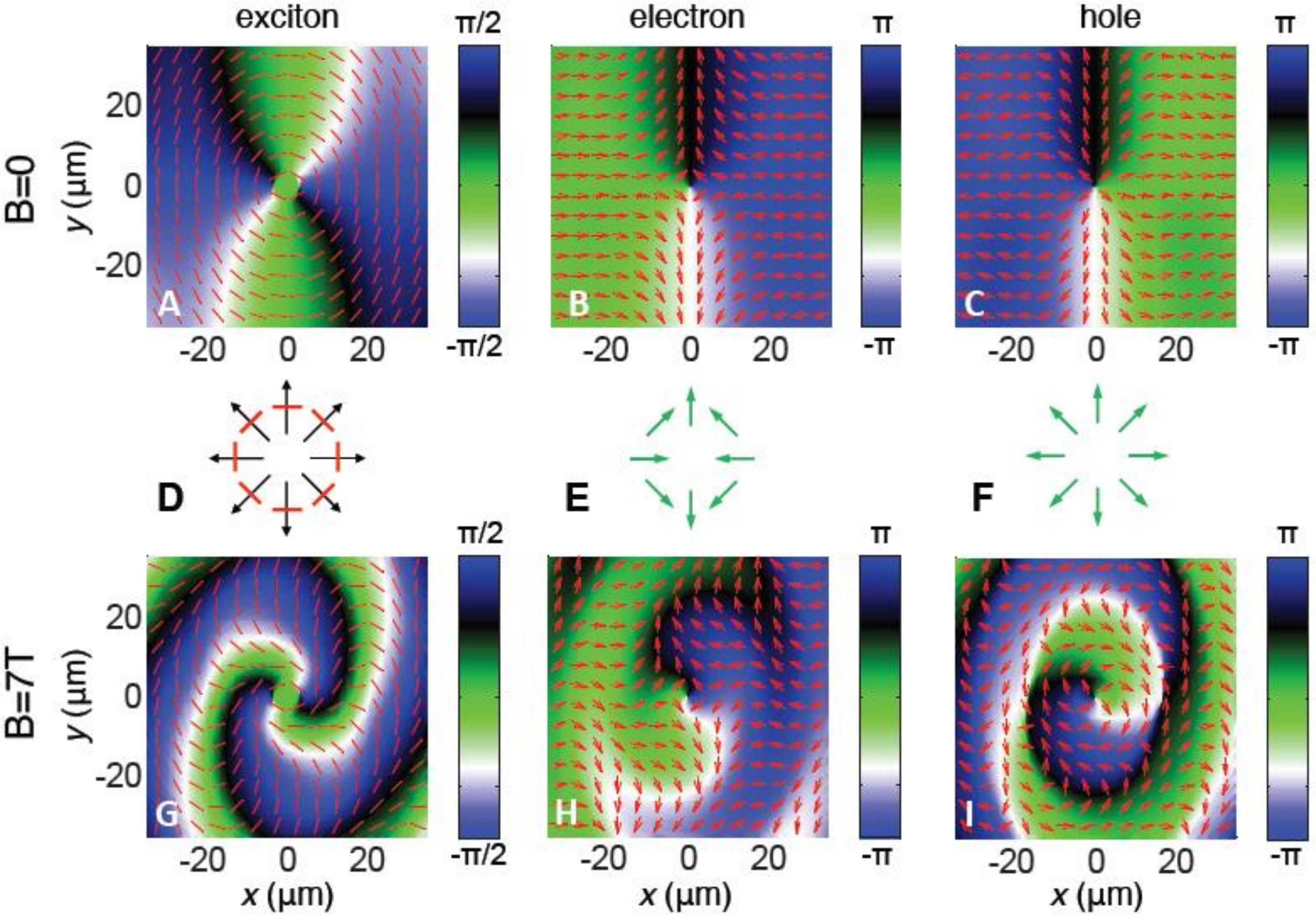
$$\delta_b = 0.55\mu eV$$

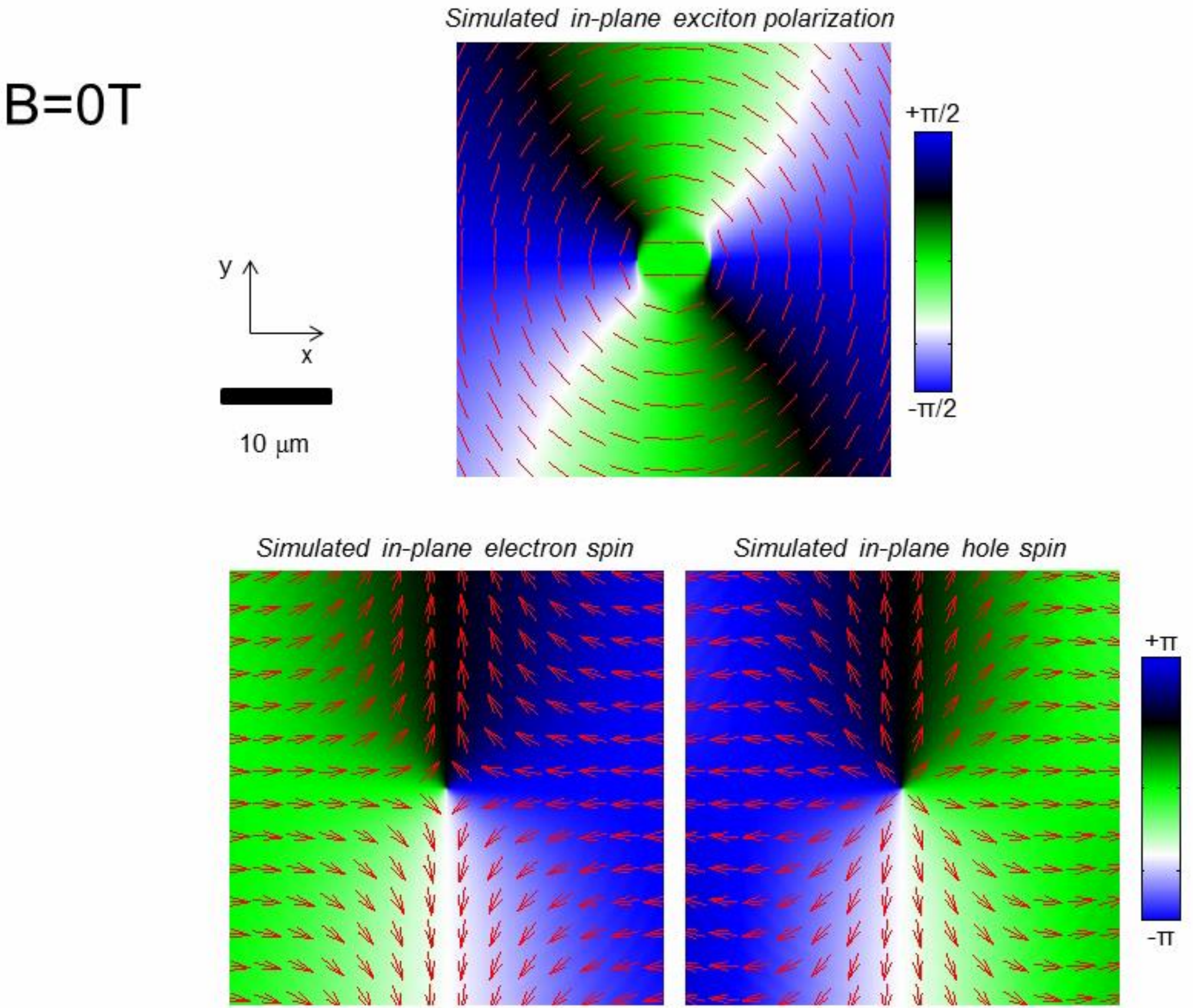
$$\delta_d = -13\mu eV$$

$$k = 15.4\mu m^{-1}$$

T=0.1 K

Excitons carry electron and hole spin currents!





B=0T
Simulated in-plane exciton polarization
+π/2
-π/2
y
x
10 μm
Simulated in-plane electron spin
Simulated in-plane hole spin
+π
-π

Conclusions:

- Electrons and holes carry spins
- When they are bound to excitons, electron and hole spin currents are correlated
- Correlated spin currents manifest themselves by polarisation textures
- In cold exciton gases spin propagates ballistically by about 10 micrometers
- Perspectives for bosonic spintronics