Coulomb correlations in wide bandgap semiconductors

Ian Galbraith

Physics, School of Engineering and Physical Sciences,
Heriot-Watt University, Edinburgh, U.K.
What do I mean by correlation?

In an excited semiconductor the electrons and holes interact via the Coulomb and exchange interactions.

When this is strong enough their motion is correlated.
Absorption spectrum strongly modified by the Coulomb interaction between electrons and holes.

*Excitons* are Hydrogen-like, bound electron–hole pairs which lower their energy by correlating their motion via the Coulomb attraction.

*Biexcitons* are pairs of excitons likewise bound as a four particle molecule.
Overview

- Non-resonant pumping
  - Experiment & Samples
  - Influence of the light hole band
- Resonant pumping
  - Second Born approximation
  - T-matrix approach
  - Comparison with experiment
- Conclusions
Non-resonant pumping

- The line width and shift of the exciton absorption peak in semiconductor quantum wells are two of the key signatures used to probe Coulomb correlations.

- By pumping well below the HH exciton we can study the influence of the LH on the correlations.
Non-resonant pumping

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Detuning Dependence

Detunings: 36-71 meV, $T=10K$

SCP

OCP

Energy (eV)

Optical density (arb. units)
### ZnSe-based QW samples

<table>
<thead>
<tr>
<th>Structure</th>
<th>width</th>
<th>HH-LH splitting</th>
<th>Detuning</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>A (Zn,Cd)Se/ZnSe</td>
<td>4 nm</td>
<td>36 meV</td>
<td>36-71 meV</td>
<td>1.0-2.0</td>
</tr>
<tr>
<td>B ZnSe/ZnMgSSe</td>
<td>5 nm</td>
<td>33 meV</td>
<td>39-73 meV</td>
<td>1.2-2.2</td>
</tr>
<tr>
<td>C ZnSe/MgS</td>
<td>10 nm</td>
<td>20 meV</td>
<td>41-86 meV</td>
<td>2.0-4.2</td>
</tr>
<tr>
<td>D ZnSe/MgS</td>
<td>11 nm</td>
<td>16 meV</td>
<td>42-79 meV</td>
<td>2.6-4.9</td>
</tr>
</tbody>
</table>
In different samples...

**SCP**: Reduced Pauli blocking for larger detuning

![Diagram showing m_e and m_{LH} configurations for SCP and OCP](image)

**SCP**: Reduced Pauli blocking over-compensated by red shift from LH correlations. (Brick et al PRB 64 75323 2001).

![Graph showing HH exciton shift vs. detuning for samples A and B](image)
In different samples...

SCP: Reduced Pauli blocking for larger detuning
OCP: Reduced Pauli blocking overcompensated by red shift from LH correlations. (Brick et al PRB 64 75323 2001).
Light Hole Stark shifts

Stronger LH Stark shift in OCP in samples with large ratios - selection rules

![Graphs showing LH Stark shift in Samples A, B, and C vs. Detuning (meV)]
Light Hole Stark shifts

- Stronger LH Stark shift in OCP in samples with large ratios - selection rules
- LH Stark shift is identical in samples with small ratios.

![Graphs showing LH Stark shift](image-url)
Light Hole Stark shifts

- Stronger LH Stark shift in OCP in samples with large ratios - selection rules
- LH Stark shift is identical in samples with small ratios.
- This is beyond current theory as HH is not perturbation.
Resonant pumping

The line width and shift of the exciton absorption peak in semiconductor quantum wells are two of the key signatures used to probe Coulomb correlations.

We study the case where a population of excitons has been created by resonant excitation (i.e. they have essentially zero centre-of-mass momentum.)
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If a low density of excitons is present, the exciton line width and shift are dominated by exciton-exciton scattering.
Exciton-Exciton Coulomb Matrix elements

At low exciton momenta, the scattering matrix elements are dominated by scattering events involving electron or hole exchange:

![Graph showing matrix elements (arb. units) versus $k a_B^{2D}$]
Self-consistent Born approximation

The Line width, $\Gamma$, and shift, $\Delta$, are given by (Boldt et al, PSSb 130, 675):

$$\Gamma = N \sum_Q |V(0,0,Q)|^2 \frac{2\Gamma}{\left(\frac{\hbar^2 Q^2}{M}\right)^2 + 4\Gamma^2}$$

$$\Delta = N \left( V(0,0,0) - \frac{1}{2} \sum_Q |V(0,0,Q)|^2 \frac{\hbar^2 Q^2}{M} \left(\frac{\hbar^2 Q^2}{M}\right)^2 + 4\Gamma^2 \right)$$

$V(k, k', q)$ is the scattering matrix element for two excitons of momentum $k, k'$ and momentum transfer $q$ (Ciuti et al, PRB 58, 7926).

$N$ is the number of excitons.

The line width $\Gamma$ is found self-consistently.
Self-consistent Born approximation - results

Line width
First order shift (Schmitt-Rink et al, PRB 32, 6601)
Total shift
Second order shift

Experiment: Wachter et al, PRB 65, 205314
Shift linear with fluence and to the blue
The T-matrix, $T(k, k', q)$, contains the exciton-exciton scattering to infinite order.
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\[
\begin{align*}
T & = \bigg| + \bigg[ \bigg] + \bigg[ \bigg] + \cdots \\
& = \begin{array}{c|c}
   & T \\
\end{array}
\end{align*}
\]
The T-matrix, $T(k, k', q)$, contains the exciton-exciton scattering to infinite order.

\[
T = 1 + \begin{array}{c}
\end{array} + \begin{array}{c}
\end{array} + \cdots
\]

\[
T(k, k', q) = V(k, k', q) + \sum_Q \frac{1}{2} V(k + Q, k' - Q, q - Q) T(k, k', Q)
\]

\[
- \frac{\hbar^2 Q^2}{M} + 2i\Gamma
\]
The T-matrix, \( T(k, k', q) \), contains the exciton-exciton scattering to infinite order.

\[
\Gamma = N \sum_Q |T(0, 0, Q)|^2 \frac{2\Gamma}{\left( \frac{\hbar^2 Q^2}{M} \right)^2 + 4\Gamma^2}
\]

\[
\Delta = N \text{Re}[T(0, 0, 0)]
\]
Self-consistent Born approximation, T-matrix

The dependence of the line width on density is not sublinear in the T-matrix calculation.

The infinite order T-matrix removes the anomalous red shift at low densities.
T-matrix: comparison with experiment

- It is difficult to compare the density dependence with experiment due to uncertainties in the carrier density.
- This may be eliminated by plotting directly shift against line width (Wachter et al, PRB 65, 205314).

![Graph showing comparison between experimental data, T-matrix calculations, and self-consistent Born calculations.](image)

- **Experiment**
- **T-matrix calcs**
- **Self-consistent Born calcs.**
Summary

Non-resonant Pumping
- HH OCP Stark shift depends on HH-LH splitting
- LH Stark shift - theory opportunity

Resonant Pumping
- We have calculated the width and shift of the exciton absorption peak in semiconductor quantum wells due to exciton-exciton scattering.
- An infinite order T-matrix calculation gives no anomalous red shift.
- We have fitted experimental plots of shift against line width for SCP polarisation.
- Calculating the T-matrix for other polarisation configurations is significantly more involved.

... and thanks to ...
Collaborators

- **Non-resonant Pumping**
  - G. Papageorgiou, A. Kar - Expts, HW
  - C. Bradford, K. Prior - samples, HW

- **Resonant Pumping**
  - S. Wachter, H. Kalt - Uni Karlsruhe
  - C. Dent - Theory, HW
  - K. Ohkawa, D. Hommel - samples, Uni Bremen
Temperature dependence

Probe Absorption with Pump
Probe Absorption without Pump

Exciton-biexciton transition

HH LH

Photon Energy (eV)