Simulation and multivariate statistical analysis of physical characteristics of dispersive ensembles of semiconductor nano-sized objects

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Outline

- Motivation
- Physical response of semiconductor nano-objects
- Dispersive ensembles of semiconductor nano-objects
- Quantum theory of semiconductor nano-objects
- Electromagnetic response of systems of semiconductor nano-objects
- Magnetic response of ensembles of self-assembled semiconductor quantum rings
- Conclusions
Confinement of the electron motion in at least one spatial dimension affects the energy levels and the density of states.

Motivation

Semiconductors in nano-scale

Quantum dots – Artificial Atoms

(Dieter Bimberg. Semiconductor Nanostructures. Springer, 2008)
Advances in the fabrication of semiconductor nano-structures have allowed us to construct nano-scale systems within a wide range of geometries.

**Motivation**

*Semiconductor Nano-Objects (SNOs)*

Advances in the fabrication of semiconductor nano-structures have allowed us to construct nano-scale systems within a wide range of geometries.

- **Self assembled Quantum Dots**
  - InAs/GaS Quantum Rings

- **Self assembled vertical Quantum Dot Molecule**
  - M. Bayer, et al., (2001)
Motivation

Semiconductor Nano-Objects (SNOs)
Physical Response of SNOs

Basic optical processes

Absorption (excitation) and Emission (luminescence)

ZnTe/CdSe core shell QDs
(A.Sitt et al., Proc. of SPIE, 2009)

Optical gap in QDs
(M. Auffan et al., Nature Nanotech., 2009)
Physical Response of SNOs

Different topology – different orbital magnetism

Quantum Dot
“Artificial atom”

Ideal meso-scale ring
“One-dimensional loop”

Quantum Ring

\[ \delta E(B) = E(B) - E(0) \approx C \cdot \left( \rho^2 \right) B^2 + D \cdot l \cdot B \]

\[ E_l(B) = \frac{\hbar^2}{2m_0^2} \left( l + \Phi / \Phi_0 \right)^2; \]
\[ \Phi = S \cdot B, \Phi_0 = h/e \]

\[ E_l(B) \approx P \cdot \left[ l + F(\Phi)/\Phi_0 \right]^2; \]
\[ \Phi = \tilde{S} \cdot B, \Phi_0 = h/e \]

> 50 years of the Aharonov-Bohm effect
Physical Response of SNOs

Para- and dia- magnetism

Constitutive relations:

\[ B = \mu_0(H + M) \quad \text{and} \quad B = \mu H \]

Magnetic permeability:

\[ \mu = \mu_0 \mu_{\text{eff}}; \]

\[ \mu_{\text{eff}} \approx 1 + \mu_0 \chi = 1 + \chi_{\text{eff}}, \left\{ \text{if } |\mu_0 \chi| < 1 \right\} \]

Paramagnetic
Total \( L \neq 0 \) or \( S \neq 0 \),
By magnetic moment polarization:
\( M \) aligns with \( B \)
\[ \chi > 0 \]

Magnetic susceptibility:

\[ \chi(B, T) = \frac{\partial M(B, T)}{\partial B} \]

Diamagnetic
Total \( L = 0 \) and \( S = 0 \),
by the Lenz’s Law:
\( M \) opposes \( B \)
\[ \chi < 0 \]
Physical Response of SNOs

Orbital magnetism in SNOs

Magnetization (magnetic moment) of SNOs

\[ M(B,T) = -\frac{\partial F}{\partial B} = -\frac{1}{Z} \sum_{n,l,s} \frac{\partial E_{n,l,s}(B)}{\partial B} \exp\left(-\frac{E_{n,l,s}(B)}{k_BT}\right) \]

\[ Z = \sum_{n,l,s} \exp\left(-\frac{E_{n,l,s}(B)}{k_BT}\right) \]

\[ \chi_r(B,T) = \frac{\partial M(B,T)}{\partial B} \]

\[ \chi(B,T) = N\chi_r(B,T) \]

Aharonov-Bohn periodic oscillation in one-electron meso-scopic quantum rings

\[ E_l = \frac{\hbar^2}{2m^*R^2} \left(l + \frac{\Phi}{\Phi_0}\right)^2 \]

(V. M. Fomin et al., PRB 2007)

AB oscillation in wobbled InAs/GaAs quantum ring
Physical Response of SNOs

Magnetization of quantum rings

O. Voskoboynikov and C.P. Lee, Physica E, 2004


Magnetic susceptibility (single electron in the ring).

Peak magnitude at 4 K: ~ 7 μB/T

Stable at low temperatures
Dispersive ensembles of SNOs

Individual Response and Collective Response. Multi-Scale Hybrid Approach

Quantum Mechanics of SNO: Electron’s Energy $E_n$, Envelope Functions $F_n$

Polarizability of SNO: Static and Dynamic: $\alpha$ and $\alpha(\omega)$

Interaction Equations: Discrete Dipoles Embedded into Continues Media

Remote Fields: Optics, Magnetic response
Dispersive ensembles of SNOs

Univariate approach (Primitive shapes)

Quantum dot | Wave Function | Light Emission | Univariate Statistics | Broadening in emission spectra

\[ \bar{R} = \int_{r} P(r)R(r)\,dr \]
Dispersive ensembles of SNOs

General description. Multivariate approach.
(Complex shapes, material parameters, random positions, etc.)

Dependence of a physical quantity (response) \( Q \) on discrete \( \{\rho_i\} \) and continuous \( \{\eta_i\} \) parameters

Multidimensional (multivariate) distribution:
\[
P(\{\rho_i;\eta_j\})
\]

Condition: parameters of interest: \( \{x_k\}_C \subseteq \{x_k\} \)

Meaningful multivariate conditional average for a physical quantity \( Q \) of a dispersive ensemble of semiconductor nano-objects:
\[
\bar{Q} = \sum_{i \in C} \int P(\{\rho_i;\eta_j\})Q(\{\rho_i;\eta_j\}) \prod_{j \in C} d\eta_j
\]

For non-correlating distributions:
\[
P(\{x_k\}) = \prod_{k} P_{x_k}(x_k)
\]
Most common choice is to present wave functions in terms of the envelop functions \( u_n \)

\[
\Psi(r) = \sum_n F_n(r) u_n(r)
\]

\[
\begin{align*}
\hat{\Pi}^2 &+ V_{\text{crystal}}(r) + V_{\text{extra}}(r) + \frac{g_0}{2} \mu_B \boldsymbol{\sigma} \cdot \boldsymbol{B} + \frac{\hbar}{4m_0 c^2} \hat{\Pi} \cdot \left[ \boldsymbol{\sigma} \cdot \nabla V_{\text{crystal}}(r) \right] \\
\hat{\Pi} &= -i\hbar \nabla + e \mathbf{A}, \quad \mathbf{B} = \nabla \times \mathbf{A}
\end{align*}
\]
Quantum theory of SNOs

Eight-band approximation (an example)

\[ \hat{H}_{8 \times 8} F = E F; \quad F = \{ F_{1p}, F_{2p}, \ldots, F_{8p} \}^T \]

\[ \hat{H}_{8 \times 8} = \hat{H}_K + \hat{H}_S + \hat{H}_P + V_{\text{conf}} \]

Kinetic part | Strain part | Pauli part
---|---|---
\[ A \quad 0 \quad V^* \quad 0 \quad \sqrt{3}V \quad -\sqrt{2}U \quad -U \quad \sqrt{2}V^* \]
\[ 0 \quad A \quad -\sqrt{2}U \quad -\sqrt{3}V^* \quad 0 \quad -V \quad \sqrt{2}V \quad U \]
\[ V \quad -\sqrt{2}U \quad -P + Q \quad -S^* \quad R \quad 0 \quad \sqrt{3} S \quad -\sqrt{2}Q \]
\[ 0 \quad -\sqrt{3}V \quad -S \quad -P - Q \quad 0 \quad R \quad -\sqrt{2}R \quad \frac{1}{\sqrt{2}} S \]
\[ \sqrt{3}V^* \quad 0 \quad R^* \quad 0 \quad -P - Q \quad S^* \quad \frac{1}{\sqrt{2}} S^* \quad \sqrt{2}R^* \]
\[ -\sqrt{2}U \quad -V^* \quad 0 \quad R^* \quad S \quad -P + Q \quad \sqrt{2}Q \quad \sqrt{3} S \]
\[ -U \quad \sqrt{2}V^* \quad \sqrt{3} S \quad -\sqrt{2}R^* \quad \frac{1}{\sqrt{2}} S \quad \sqrt{2}Q \quad -P - \Delta \quad 0 \]
\[ \sqrt{2}V \quad U \quad -\sqrt{2}Q \quad \frac{1}{\sqrt{2}} S^* \quad \sqrt{2}R \quad \sqrt{3} S \quad 0 \quad -P - \Delta \]

\[ \Delta = \epsilon \frac{\hbar^2}{2m_0} (D_x^2 + D_y^2 + D_z^2), \]
\[ P = -\epsilon \gamma \frac{\hbar^2}{2m_0} (D_x^2 + D_y^2 + D_z^2), \]
\[ Q = -\gamma \frac{\hbar^2}{2m_0} (D_x^2 + D_y^2 + D_z^2), \]
\[ R = \sqrt{3} \frac{\hbar^2}{2m_0} \left[ \gamma_2 (D_x^2 - D_y^2) - 2 \gamma_3 D_x D_y \right], \]
\[ S = -\sqrt{3} \frac{\hbar^2}{m_0} \gamma_3 D_z (D_x - iD_y), \]
\[ U = -\frac{i}{\sqrt{3}} \frac{P_0 D_z}{D_k}, \]
\[ V = -\frac{i}{\sqrt{6}} \frac{P_0 (D_x - iD_y)}{D_k}, \]
\[ D_k = \partial_k + eA_k \]

(C. Pryor (1998) plus magnetic field)
### Quantum theory of SNOs

#### Eight-band approximation. Strain and Pauli

$$
\hat{H}_S = \begin{pmatrix}
    a_v & 0 & -v^* & 0 & -\sqrt{3}v & \sqrt{2}u & u & -\sqrt{2}v^*
    \\
    0 & a_v & \sqrt{2}u & 3v^* & 0 & v & -\sqrt{2}v & -u
    \\
    -v & \sqrt{2}u & -p+q & -s^* & r & 0 & \frac{\sqrt{3}}{2}s & -\sqrt{2}q
    \\
    0 & \sqrt{3}v & -s & -p-q & 0 & r & -\sqrt{2}r & \frac{1}{\sqrt{2}}s
    \\
    -\sqrt{3}v^* & 0 & r^* & 0 & -p-q & s^* & \frac{1}{\sqrt{2}}s^* & \sqrt{2}r^*
    \\
    \sqrt{2}u & v^* & 0 & r^* & s & -p+q & \sqrt{2}q & \frac{\sqrt{3}}{2}s^*
    \\
    u & -\sqrt{2}v^* & \frac{1}{\sqrt{2}}s^* & -\sqrt{2}r^* & \frac{1}{\sqrt{2}s} & \sqrt{2}q & -a_v & 0
    \\
    -\sqrt{2}v & -u & -\sqrt{2}q & \frac{1}{\sqrt{2}}s^* & \sqrt{2}r & \frac{3}{2}s & 0 & -a_v & \end{pmatrix}
$$

$$
\hat{H}_P = \mu_B B \cdot \begin{pmatrix}
    0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
    \\
    0 & 2 & 0 & 0 & 0 & 0 & 0 & 0
    \\
    0 & 0 & 1 & \frac{1}{3} \sigma & 0 & 0 & 0 & 0
    \\
    \end{pmatrix}
$$

- $p = a_v v$
- $q = b \left[ e_{zz} - \frac{1}{2} (e_{xx} + e_{yy}) \right]$
- $r = \frac{\sqrt{3}}{2} b (e_{xx} - e_{yy}) - i e_{xy}$
- $s = -d (e_{xz} - ie_{yz})$
- $u = -\frac{i}{\sqrt{3}} P_0 \sum_k e_{zk} D_k$
- $v = -\frac{i}{\sqrt{6}} P_0 \sum_k (e_{xk} - ie_{yk}) D_k$
- $e = e_{xx} + e_{yy} + e_{zz}$
- $e_{ij} = \frac{1}{2} \partial_i w_j + \partial_j w_i$

Derivatives of the atomic displacements starting from the interfaces

$$
F = \frac{1}{2} \int \frac{\partial}{\partial x_{xx}} \left( e_{xx}^2 + e_{yy}^2 + e_{zz}^2 \right) + \frac{\partial}{\partial x_{xy}} \left( e_{xx} e_{xy} + e_{xx} e_{yz} + e_{xx} e_{zz} \right)
+ 2 \frac{\partial}{\partial x_{yy}} \left( e_{xy}^2 + e_{yy}^2 + e_{yz}^2 \right)
+ \delta F = 0$

with

$$
e_{ij} = \frac{a_{ij}}{a_d}, e_\perp = \frac{a_{im}}{a_d} - \frac{a_{jd}}{a_d} \text{ at the interface}
$$
If we focus on the case of the electrons (holes) we can obtain an effective Hamiltonian for “one component” envelop function.

Multi-band Schrödinger equation

\[ \hat{H} \Psi = \hat{H}_0 \Psi + \hat{V} \Psi = E \Psi \]

Projection onto electron (hole) subspace (Fouldy-Wouthuysen transformation)

\[ \hat{H}_e(P_e \Psi) \approx P_e \hat{H}_0(P_e \Psi) + P_e \hat{V}(P_e \Psi) + P_e \hat{W} \hat{P}_h \frac{1}{\hat{H}_e - \hat{P}_h \hat{H}_0 \hat{P}_h} \hat{P}_h \hat{W}(P_e \Psi) \]

Effective equation for envelop function Hamiltonian with spin-orbit interaction

\[ \hat{H}_{e(h)} = \Pi \frac{1}{m^*(E, r)} \Pi + V_{conf}(r) + V_{strain}(r) + \Phi(r) \]

\[ + \mu_B g(E, r) \hat{\sigma} \cdot \mathbf{B} + \nabla \beta(E, r) \cdot \left[ \hat{\sigma} \times \Pi \right] \]

(de Anrada e Silva et al., 1997, Schäpers et al., 1998, Voskoboynikov et al., 2001)
Quantum theory of SNOs

Effective Hamiltonian

Parameters of the theory

\[
\frac{1}{m_e(E, \mathbf{r})} = \frac{2P^2}{3\hbar^2} \left( \frac{2}{E-V_e(\mathbf{r})+E_g(\mathbf{r})} + \frac{1}{E-V_e(\mathbf{r})+E_g(\mathbf{r})+\Delta(\mathbf{r})} \right);
\]

\[
\beta(E, \mathbf{r}) = \frac{P^2}{3\hbar} \left( \frac{1}{E-V(\mathbf{r})+E_g(\mathbf{r})} - \frac{1}{E-V_e(\mathbf{r})+E_g(\mathbf{r})+\Delta(\mathbf{r})} \right);
\]

\[
g(E, \mathbf{r}) = 2 \left[ 1 - \frac{m_0}{m^*(E, \mathbf{r})} \left( \frac{\Delta(\mathbf{r})}{3(E-V_e(\mathbf{r})+E_g(\mathbf{r}))+2\Delta(\mathbf{r})} \right) \right];
\]

\[m_h(\mathbf{r}); \ g_h(\mathbf{r}); \]

\[V_{e(h)}(\mathbf{r}).\]

For a nano-object of complex geometry parameters to map

\[V_{e(h)}(\mathbf{r}); \ E_g(\mathbf{r}); \ \Delta(\mathbf{r}); \ P(\mathbf{r}); \ m_h(\mathbf{r}); \ g_h(\mathbf{r}) \ldots\]
Quantum theory of SNOs

Mapping procedure starts

Photo Emission Electron Microscopy (PEEM)
Cross-sectional transmission electron microscopy (XTEM)
Atomic Force Microscopy (AFM)
Kelvin Probe Force Microscopy (KPFM)
Cross-sectional scanning tunnel Microscopy (XSTM)

Example: InAs/GaAs nano-ring
(P.Offermans et al. APL 2005 – XSTM)

Nano object’s geometry profile

\( h(x, y) \)

Effective electronic (hole’s) confinement potential

\[
V_{e(h)}(r) = \Phi [x, y, z, h(x, y)]
\]

\[
V_{e(h)}(r) \Rightarrow \begin{cases} 
\Delta E_{C(V)}, & \text{outside} \\
0, & \text{inside} 
\end{cases}
\]

\( \Delta E_{C(V)} \rightarrow \text{conducting (valence) band offset} \)
Quantum theory of SNOs

The mapping function

Unitless mapping function

\[ M_{e(h)}(\mathbf{r}) = 1 - \frac{V_{e(h)}(\mathbf{r})}{\Delta E_{e(h)}} \]

Mapped parameters

\[
\begin{align*}
V_e(\mathbf{r}) &= \Delta E_e - \Delta E_e M(\mathbf{r}); \\
E_g(\mathbf{r}) &= E_g^{in} M(\mathbf{r}) + E_g^{out}[1 - M(\mathbf{r})]; \\
\Delta(\mathbf{r}) &= \Delta^{in} M(\mathbf{r}) + \Delta^{out}[1 - M(\mathbf{r})]; \\
P(\mathbf{r}) &= P^{in} M(\mathbf{r}) + P^{out}[1 - M(\mathbf{r})]; \\
V_h(\mathbf{r}) &= (E_g^{out} - E_g^{in})[1 - M(\mathbf{r})] - V_e(\mathbf{r}); \\
m_h(\mathbf{r}) &= m_h^{in} M(\mathbf{r}) + m_h^{out}[1 - M(\mathbf{r})]; \\
g_h(\mathbf{r}) &= g_h^{in} M(\mathbf{r}) + g_h^{out}[1 - M(\mathbf{r})]; \\
\varepsilon(\mathbf{r}) &= \varepsilon^{in} M(\mathbf{r}) + \varepsilon^{out}[1 - M(\mathbf{r})].
\end{align*}
\]

Parameters of the theory

\[
\begin{align*}
V_{e(h)}(E, \mathbf{r}); \\
m_e(E, \mathbf{r}); \\
m_h(\mathbf{r}); \\
\beta(E, \mathbf{r}); \\
g_e(E, \mathbf{r}); \\
g_h(\mathbf{r}); \\
\varepsilon(\mathbf{r}).
\end{align*}
\]
Electromagnetic response of systems of SNOs

Polarizability of individual semiconductor nano-objects

**Discrete Dipole Approximation** ($\lambda >> a > d$) for SNOs
(Voskoboynikov et al., PRB 2005-2009)

\[ \vec{d} = \alpha_B \vec{E}_A \]

- **Polarizability of SNO**
  \[ \alpha_B(\omega) = \alpha_{BS} + \Delta \alpha_B(\omega) \]
  - Static part
  - Dynamic part

- **Bulk optical matrix element**

- **Excitonic Envelope function**

- **Overlap integral**

- **Frequency dependent factor**

\[ \Delta \alpha_B(\omega) = \frac{e^2}{\hbar} \sum_n r_{eh}^* r_{eh}^T \left| \phi_n^{ex} \right|^2 f_n(\omega) \]

\[ f_n(\omega) = \left( E_n^{ex} - \hbar \omega + i \Gamma_n \right)^{-1} \]
Electromagnetic response of systems of SNOs

**Excitonic polarizability**

\[
\begin{align*}
[H_0^e - \epsilon \Phi_h] \Psi_e &= E_e \Psi_e, \\
[H_0^h - \epsilon \Phi_e] \Psi_h &= E_h \Psi_h, \\
\epsilon_0 \nabla_r \epsilon(r) \nabla_r \Phi_e &= \epsilon |\Phi_e|^2, \\
\epsilon_0 \nabla_r \epsilon(r) \nabla_r \Phi_h &= \epsilon |\Phi_h|^2,
\end{align*}
\]

\[
H_{e(h)}^0 = \frac{1}{2} \Pi_r \left[ \frac{1}{m_{e(h)}(E,r)} \right] \Pi_r + \mu_B g_e(E,r) \sigma \cdot B + V_e(r)
\]

**Excitonic energy**

\[
E_{ex}^0 = E_g^\in + E_e + E_h + \frac{e}{2} \left( \int dr |\Psi_e|^2 \Phi_h + \int dr |\Psi_h|^2 \Phi_e \right)
\]

**Excitonic polarizability**

\[
\Delta \alpha_B(\omega) = \frac{e^2}{\hbar} \frac{r_{eh}^2 |\langle \Phi_0^{ex} \rangle|^2}{E_0^{ex} - \hbar \omega + i \Gamma_0}
\]

**Excitonic emission intensity**

\[
I(\omega) \propto \frac{\Gamma_0}{\left( E_0^{ex} - \hbar \omega \right)^2 + \Gamma_0^2}
\]
Electromagnetic response of systems of SNOs

Polarizability. Bare and Dressed

The dipole strength

\[ \vec{d} = \int_{V_{\text{SNO}}} d \mathbf{r} [ \mathbf{P}(\mathbf{r}) - \mathbf{P}_m ] \]

In the linear theory of the response functions

\[ \vec{d} = \vec{\alpha}_B \vec{E}_A \]

Bare polarizability

Actual Local field
(Spatial average in SNO
\textit{calculated in the theory})

\[ \vec{d} = \vec{\alpha}_D \vec{E}_X \]

Dressed polarizability

External remote field
(\textit{measured in experiments})

\[ \vec{E}_A = \vec{E}_X + \mathbf{t}_i \vec{d} \]

Static intracellular transfer tensor
(depolarization)
Electromagnetic response of systems of SNOs

Static polarizability. Bare and Dressed. Calculation scheme

For a single embedded SNO

\[ \nabla \cdot [\varepsilon(r)\nabla U(r)] = 0 \]

\[ U(L_k) = U_0 \]

\[ U(-L_k) = 0, \quad 2L_k E_{Xk} = U_0 \]

The excess dipole strength of SNO

\[ \bar{d} = \int_{V_{SNO}} dr \left[ P(r) - P_m(r) \right] \]

Spatial average calculated within the SNO

\[ \alpha_{Bkk} = \frac{d_k}{\bar{E}_{Ak}} \]

\[ \alpha_{Dkk} = \frac{d_k}{E_{Xk}} \]

\[ \bar{t}_s = \bar{\alpha}_B^{-1} - \bar{\alpha}_D^{-1} \]

Calculation domain size in the \( k \)-direction, \( L_k \gg d \)

Taken from the boundary conditions
Electromagnetic response of systems of SNOs

Electromagnetic interaction between SNOs

Interacellular transfer tensor
(Dyadic Green Function)

\[ \tilde{G}_{ij} = \tilde{G}(r_i, r_j) = \exp\left(\frac{ik_m}{4\pi \varepsilon_m \varepsilon_0} \Delta r_{ij}\right) \left[ k_m^2 \left( \mathbf{I} - \Delta r_{ij} \Delta r_{ij}^T \right) - \frac{1 - ik_m}{\Delta r_{ij}^2} \left( \mathbf{I} - 3 \Delta r_{ij} \Delta r_{ij}^T \right) \right] \cdot \Delta r_{ij} = r_i - r_j; \quad k_m = \frac{\sqrt{\varepsilon_m \omega}}{c} \]

in the near (static) range:
\[ \lambda >> a > r > d \]

\[ \tilde{G}_{ij} \approx \tilde{T}_{ij} = \frac{3 \Delta r_{ij} \Delta r_{ij}^T - \mathbf{I}}{4\pi \varepsilon_m \varepsilon_0 \Delta r_{ij}^3}, \quad \Delta r_{ij} = r_i - r_j \]

in the far (wave) range:
\[ r >> \lambda >> a > d \]

\[ \tilde{G}(r_i, r) \approx \frac{k_m^2}{4\pi \varepsilon_m \varepsilon_0} \left( \mathbf{I} - \Delta r_i \Delta r_i^T \right) \exp\left(\frac{ik_m}{\Delta r_i} \Delta r_i^2 \right); \quad \Delta r_i = r - r_i \]
Electromagnetic response of systems of SNOs

Electromagnetic interaction between SNOs. Master Equation

The system’s Master equation

\[ d_j = \tilde{\alpha}_{Dj} \left[ E_{Xj} + \sum_{i \neq j} \tilde{T}_{ji} d_i \right] \]

Or

\[ \sum_i \tilde{T}_{ji} d_i = E_{Xj}, \]

where

\[ \tilde{T}_{ji} = (\delta_{ji} - 1) \tilde{T}_{ji} + \delta_{ji} \left( \tilde{\alpha}_{Bj}^{-1} - t_i \right), \]

\[ t_i = \tilde{t}_i + \frac{i \omega^3}{6 \pi \varepsilon_m \varepsilon_0 c^3} \]

Lorentz radiation damping

\[ \tilde{t}_i = \tilde{\alpha}_B^{-1}(\omega) - \tilde{\alpha}_D^{-1}(\omega) \]
Electromagnetic response of systems of SNOs

General flow diagram of the method implementation

Scale of an Individual Nano-Object

Laplace Equation
\[ \nabla_r \left[ \varepsilon(r) \nabla_r \Phi(r) \right] = 0 \]

Shrödinger Equation
\[ \hat{H}_{e(h)} F_{e(h)}(r) = E_{e(h)} F_{e(h)}(r) \]

Static Excess Polarizabilities
\[ \tilde{\alpha}_S^{D}, \tilde{\alpha}_S^{B} \]

Transition Energies and Overlap Integrals
\[ E_{el}, E_{hk}, \Psi_{el}, \Psi_{ek}, \Phi^{ex} \]

Self Interaction Tensor
\[ \tilde{t}_S \]

Overall Embedded Bare Polarizability
\[ \tilde{\alpha}_S^{Bi}(\omega) \]

Scale of Macro-Systems of Nano-Objects

Multi-Dipole Interaction Equation
\[ \sum_j \tilde{T}_{ij} d_j = E_{Xi} \]

Electrostatics
\[ E_R(r) = \sum_i \tilde{G}(r_i, r) d_i \]
\[ E_T(r) = E_X(r) + \sum_i \tilde{G}(r_i, r) d_i \]

\[ C_{abs}(\omega) = \frac{4\pi \sqrt{\varepsilon_m} k}{|E_0|^2} \sum_l \left\{ \text{Im} d_l \cdot \left[ \tilde{\alpha}_{DL}^{-1}(\omega) \right]^* \cdot d_l^* + - \frac{2}{3} k^3 d_l \cdot d_l^* \right\} \]
Electromagnetic response of systems of SNOs
Spectral characteristics ZnTe/CdSe core/shell quantum dots

Type-II ZnTe/CdSe heterostructure inside quantum dots

\[ E_{g_{\text{ZnTe}}} = 2.26 \text{ eV}, \Delta_{\text{ZnTe}} = 0.97 \text{ eV}, \]
\[ m_{e_{\text{ZnTe}}}(E=0) = 0.20m_0, \ m_{h_{\text{ZnTe}}} = 0.35m_0; \]

\[ E_{g_{\text{CdSe}}} = 1.74 \text{ eV}, \Delta_{\text{CdSe}} = 0.42 \text{ eV}, \]
\[ m_{e_{\text{CdSe}}}(E=0) = 0.13m_0, \ m_{h_{\text{CdSe}}} = 0.45m_0 \]

\[ V_{c0} = 1.22 \text{ eV}, \ V_{h0} = 0.7 \text{ eV}, \text{ and } V_{Ce(h)} = 3 \text{ eV} \]
\[ \varepsilon_{\text{core}} = \varepsilon_{\text{ZnTe}} = 7.4, \ \varepsilon_{\text{shell}} = \varepsilon_{\text{CdSe}} = 9.1, \text{ and } \varepsilon_{m} = 3 \]
\[ r_c = 1.2 \text{ nm} \]

Electromagnetic response of systems of SNOs

Spatial disorder and Structural dispersion. Example: ZnTe/CdSe core/shell colloidal quantum dots

ZnTe/CdSe core/shell QDs
(A.Sitt et al., Proc. of SPIE, 2009)

Structural dispersion

Spatial disorder

\[
P{\eta}\left(\{\eta_j\}\right) \Rightarrow A \cdot G\left(\frac{h-h_0}{\Delta h}\right)
\]

\[
P{\rho}\left(\{\rho_j\}\right) \Rightarrow \frac{1}{Z}\bigg|_{N_d=\text{const}}
\]

G(x) – Gaussian function

Z - number of dot re-localizations
Spatial disorder (529 cells and 200 dots)

→ stable behavior of the averaged absorption cross section (density – 7·10^{16} \text{ cm}^{-3})

Structural dispersion (varying only the outer shell width \( h \))

The simulation results we can present with the following fitting functions (\( h =1\div4 \text{ nm} \)):

\[
E_0^{ex}(h) = 1.923 \cdot h^{-0.27} \text{ eV}
\]

\[
\left|\left\langle \Phi_0^{ex}\right\rangle\right|^2 = -0.001 + \frac{0.98}{h + 1.3} - \frac{0.42}{(h + 1.3)^2}
\]

\[
E_0^{ex}(\text{eV})
\]

- theory
- exp. (A.Sitt et al., Proc. of SPIE, 2009)
Electromagnetic response of systems of SNOs

Spectral characteristics of ensembles ZnTe/CdSe core/shell quantum dots

Full Width at Half Maximum (FWHM)

of the peaks of the absorption cross section (structural dispersion)

Uniform relative standard deviations of $h$:
$$\Delta h/h_0 = 0.1 \ (\Gamma = k_B T; \ T=300 \ K)$$

Non-uniform relative standard deviations of $h$:
$$\Delta h/h_0 = 0.1 + 0.3 \times (h_0 - 1.2)$$
$$[h_0] = \text{nm}; \ T=300 \ K$$
Homogeneous broadening vs. inhomogeneous broadening

\[ W(T) = \frac{\text{FWHM}(T)}{\text{FWHM}(T=100 \text{ K})} \]
Electromagnetic response of systems of SNOs

Emission spectra of dispersive ensembles of triple concentric self-assembled semiconductor GaAs/AlGaAs quantum rings

AFM images of GaAs/AlGaAs triple quantum rings
(C. Somaschini et al., Nano Lett. 2009)

\[
h(x, y) = \sum_{k=1}^{3} h_k(x, y),
\]

\[
h_k(x, y) = \begin{cases} 
    h_{mk} \frac{y_k^2 R_k^2}{R_k^2 + (\sqrt{x^2+y^2} - R_k)^2}, & \sqrt{x^2+y^2} \leq R_k \\
    h_{mk} \frac{y_k^2}{(\sqrt{x^2+y^2} - R_k)^2 + y_k^2}, & \sqrt{x^2+y^2} > R_k
\end{cases}
\]

(L. M. Thu et al., PRB, 2011)

\[
V_{e(h)}(r) = \Delta E_{e(h)} \left[ 1 - \frac{1}{4} \left[ 1 + \tanh \left( \frac{z - z_0}{a} \right) \right] \right] \times \left[ 1 - \tanh \left( \frac{z - h(x, y)}{a} \right) \right]
\]

\[
\Delta E_p = E_{g}^{\text{out}} - E_{g}^{\text{in}} - \Delta E_e
\]
Electromagnetic response of systems of SNOs

Emission spectra of dispersive ensembles of triple concentric self-assembled semiconductor GaAs/AlGaAs quantum rings

\[ P_\eta(\{\eta_j\}) \Rightarrow A \cdot G \left( \frac{h_1 - h_1^0}{\Delta h_1} \right) \]

\[ E_{ex}(h_1) = \frac{b}{(h_1)^c} \]

\[ b = 1.66 ; c = 0.03 \text{ (appropriate SI units)} \]

(a) an individual ring with the inner height \( h_1 = 7.9 \text{ nm} \)

(b) Ring ensemble with \( h_1^0 = 7.9 \text{ nm}, \Delta h_1 = 1.2 \text{ nm} \).

\( E_{max} \) is the position of the intensity maximum.
Electromagnetic response of systems of SNOs

Emission spectra of dispersive ensembles of triple concentric self-assembled semiconductor GaAs/AlGaAs quantum rings

Full width at half maximum (FWHM) of the averaged intensity of the optical transitions of the ring ensemble as a function of the inner height for different standard deviations

Normalized averaged intensity of the optical transitions of the ring ensemble as a function of the energy.

$\Delta h_1 = 1.6 \text{ nm}; \Gamma = 1.2 \text{ meV (T =14 K)}.$

$E_{\text{min}}$ and $E_{\text{max}}$ are the positions of the intensities’ minimum and maximum

Experiment: C. Somaschini et al., Nano Lett. 2009
Magnetic response of ensembles of self-assembled semiconductor quantum rings

\textit{InAs/GaAs wobbled rings. Experimental data}

\begin{itemize}
  \item XSTM, InAs/GaAs quantum ring (P. Offermans et al., APL 2005)
  \item Indium distribution in the $y$-$z$ plane ($\text{In}_{c}\text{Ga}_{1-c}\text{As}$) (V.M. Fomin et al., PRB 2007)
  \item Magnetic response of ensembles of self-assembled semiconductor quantum rings. InAs/GaAs wobbled rings. Experimental data (N. Kleemans et al., PRL 2007; PRB 2008)
\end{itemize}
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Mapping the ring’s height

\[ h(x, y) = h_0 + \left[ h_M \left( 1 + \xi \frac{x^2 - y^2}{x^2 + y^2} \right) - h_0 \right] \gamma_0^2 \cdot \frac{R^2 - r^2(x, y)}{r^2(x, y) + \gamma_0^2}, \quad r(x, y) \leq 0; \]

\[ h(x, y) = h_\infty + \left[ h_M \left( 1 + \xi \frac{x^2 - y^2}{x^2 + y^2} \right) - h_\infty \right] \gamma_\infty^2 \cdot \frac{r^2(x, y)}{r^2(x, y) + \gamma_\infty^2}, \quad r(x, y) > 0; \]

\[ r(x, y) = \sqrt{x^2 + y^2} - R. \]

\( R = 11.5 \text{ nm}; \ \xi = 0.2; \)

(L. M. Thu et. al., PRB 2012)
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Effective Potential

\[ V_{\text{min}} = 0, \quad \text{for the electronic band} \]
\[ V_{\text{max}} = \Delta E_c \]

\[ V_e(x, y, z) = \Delta E_c \left\{ 1 - \frac{1}{4} \left[ 1 + \tanh \left( \frac{z}{a} \right) \right] \cdot \left[ 1 - \tanh \left( \frac{z - h(x, y)}{a} \right) \right] \right\} \]

\[ R = 11.5 \text{ nm}; \ \xi = 0.2; \]
\[ a = 0.4 \text{ nm} \]
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Mapping Function

\[ M(\mathbf{r}) = 1 - \frac{V_e(\mathbf{r})}{\Delta E_C} \]

Indium content in the \( y-z \) plane

\[ C_{In}(\mathbf{r}) = C_{In}^{in} \cdot M(\mathbf{r}) \]

- \( R = 11.5 \) nm; \( \xi = 0.2; \)
- \( a = 0.4 \) nm

\( C_{In} \leq 0.15 \)
\( C_{In} \leq 0.2 \)
\( C_{In} \leq 0.3 \)
\( C_{In} \leq 0.4 \)
\( C_{In} \leq 0.6 \)
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Wave functions of an individual ring

\[ \hat{H} F_n(r) = E_n F_n(r) \]

The non-linear iterative method

COMSOL Multiphysics

Electronic wave functions

R = 11.5 nm; \( \xi = 0.2 \)
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Energy states, magnetization of an individual ring

\[ R = 11.5 \text{ nm}; \xi = 0.2 \]

(L. M. Thu et al., PRB 2012)
Magnetic response of ensembles of self-assembled semiconductor quantum rings

An accurate look at the crossing point

\[ E_{0,1}(B) \approx E_0(B_C) + C_{0,1} \cdot (B - B_C) \]

\[ C_{0,1} = \left. \frac{\partial E_{0,1}(B)}{\partial B} \right|_{B = B_C} \]

\[ M(B, T) \approx -\left[ \frac{C_0}{1 + \exp(-\delta)} + \frac{C_1}{1 + \exp(\delta)} \right] \]

\[ \chi(B, T) \approx \frac{(C_1 - C_0)}{2k_B T} \left[ \frac{C_1 \exp(\delta)}{[1 + \exp(\delta)]^2} - \frac{C_0 \exp(-\delta)}{[1 + \exp(-\delta)]^2} \right] \]

\[ \delta = \frac{(C_1 - C_0)}{2k_B T} \cdot (B - B_C) \]
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Near the crossing point

\[ M(B,T) = \int \int M(B,T,R_r,\xi)P(R_r,\xi)dR_r d\xi \]

\[ \chi(B,T) = \int \int \chi(B,T,R_r,\xi)P(R_r,\xi)dR_r d\xi \]

\[ B_c(R_r,\xi) = a_c + b_c \xi + c_c \xi^2 + d_c \xi^3 + e_c R_r^{\beta_c} \]

\[ C_0(R_r,\xi) = a_0 + b_0 \xi + c_0 \xi^2 + d_0 \xi^3 + e_0 R_r^{\beta_0} \]

\[ C_1(R_r,\xi) = a_1 + b_1 \xi + c_1 \xi^2 + d_1 \xi^3 + e_1 R_r^{\beta_1} \]

TABLE I. Fitting parameters.

<table>
<thead>
<tr>
<th>k</th>
<th>0</th>
<th>1</th>
<th>c</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>1.209 \times 10^{-3}</td>
<td>2.131 \times 10^{-3}</td>
<td>0</td>
</tr>
<tr>
<td>b</td>
<td>-9.164 \times 10^{-4}</td>
<td>1.659 \times 10^{-4}</td>
<td>1.058</td>
</tr>
<tr>
<td>c</td>
<td>-4.743 \times 10^{-3}</td>
<td>3.059 \times 10^{-3}</td>
<td>36.96</td>
</tr>
<tr>
<td>d</td>
<td>1.069 \times 10^{-3}</td>
<td>-5.115 \times 10^{-3}</td>
<td>0</td>
</tr>
<tr>
<td>e</td>
<td>-6.544 \times 10^{-5}</td>
<td>-1.247 \times 10^{-3}</td>
<td>5222.2</td>
</tr>
<tr>
<td>\beta</td>
<td>0.722</td>
<td>0.206</td>
<td>-2.491</td>
</tr>
</tbody>
</table>
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Near the crossing point. Fitting
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Averaged magnetic response

Individual ring

\[ R_0 = 11.5 \text{ nm}, \Delta R = 0.5 \text{ nm}; \]
\[ \xi_0 = 0.2, \Delta \xi = 0.05 \]
Magnetic response of ensembles of self-assembled semiconductor quantum rings

Temperature dependence of the averaged magnetic response

\[ R_0 = 11.5 \text{ nm}, \Delta R = 0.5 \text{ nm}; \]
\[ \xi_0 = 0.2, \Delta \xi = 0.05 \]

\[
\begin{array}{|c|c|}
\hline
\text{Individual ring} & \chi_{1.2}/\chi_{4.2} \\
\hline
\text{Ensemble of rings} & 3.5 \\
\hline
\text{Experiment} & 1.4 \\
\hline
\end{array}
\]
We present our method for simulation of multivariate conditional averages of physical responses of ensembles of SNO with dispersion in geometry, material parameters, and positions.

For dispersive ensembles of type-II ZnTe/CdSe core/shell QDs the method theoretically reproduces and explains the homogeneous and inhomogeneous broadenings of excitonic peaks.

We demonstrated the method efficiency for objects with very sophisticated shapes: triple concentric nano-rings. Our simulation results have explained the appearance and properties of the wide asymmetrical excitonic peaks in the photoluminescence spectra of the ring ensembles known from the experiment.

We explain in details why the positive peak in the differential magnetic susceptibility of dispersive ensembles of rings should demonstrate temperature stable behavior (unlike the magnetic response of an individual ring).

Our method allows us efficiently and economically to simulate and study physical properties of dispersive ensembles of SNO of sophisticated geometrical shapes and compositions.
Thank You
For Your Attention!