Imaging the Electronic and Vibronic States of Single Semiconductor Nanowires

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Semiconductor Nanowires as Photodetectors
Nanowires as Single Electron Transistors

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Nanowires as single photon emitters
Nanowires as Biosensors

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Core-Shell Nanowire Growth

Pre-growth

Core: GaAs

Shell: AlGaAs

Vapor-Liquid-Solid growth

600°C, 10 min
Desorb surface contaminants and form eutectic alloy.

450°C, 30 min

650°C, 15 min
Wire diameter is determined by Au catalyst and shell growth time

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Motivation

Nanowire diameters $D$ (~50-150 nm) > Bohr exciton’s diameter (~24 nm)

Dielectric “confinement” of EM dipole field ($D<<\lambda$):

- Exciton density: $N_\parallel = N_\perp$
- Photoluminescence intensities: $I_\parallel >> I_\perp$
- Lifetimes: $\tau_\parallel << \tau_\perp$

We are interested in exciton spin dynamics of single nanowires

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Single nanowire studies

Nanowires were removed from the growth substrate into solution and deposited onto a silicon substrate:

- A single nanowire:
  - ~80nm in diameter, ~5-8 μm long

Field-Emission Scanning Electron Microscope (FESEM) image

AFM image:

wire’s diameter > Bohr exciton diameter
=> expect no quantum confinement effects

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Single nanowire studies

Field-Emission Scanning Electron Microscope (FESEM) image: nanowires have tapered shape.

Nanowires were removed from the growth substrate into solution and deposited onto a silicon substrate.

A single nanowire:
~80nm in diameter, ~5-8 μm long

Core diameter > Bohr exciton diameter (24nm) => no quantum confinement effects

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Polarization studies

polarizer = \pi_0 \, ; \, \text{analyzer} = \sigma^+ \, \text{and} \, \text{polarizer} = \sigma^+ \, ; \, \text{analyzer} = \pi_0

PL emission is strongly polarized parallel to the wire, and is strongly enhanced when the laser excitation is polarized parallel to the wire

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Polarization Imaging

Calculate pixel by pixel

\[ P = \frac{I_\parallel - I_\perp}{I_\parallel + I_\perp} \]

\(~82\%~

Strongly polarized due to the large dielectric mismatch between GaAs and air

\((\text{Science 293} \ 1455 \ (2001), \ \text{APL.} \ 89 \ 173126 \ (2006))\)
Resonant Excitation

Tune excitation energy, $E_{\text{Laser}}$, record PL intensity (PLE)

- core GaAs
- shell AlGaAs

AlGaAs

GaAs

resonances

$h\omega_{\text{excitation}}$

$h\omega_{\text{emission}}$

E

r

real space

k-space

E$_{\text{laser}}$

GaAs

AlGaAs

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Clear resonances at 36, 73 and ~133 meV above free exciton energy.
Resonant Excitation

1-LO and 2-LO GaAs phonons

Resonance at ~133 meV:
1. Defect-AlGaAs related.
2. Bottom of AlGaAs band (Low concentration of Al ~10%, instead of growth condition 26%)

How does the polarization depend on excitation energy?
Excitation dependent polarization

Polarization changes with excitation energy!
PL Polarization Imaging

- Excitation laser polarized along nanowire
- Analyze emission polarization

\[ P = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}} \]

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Polarization depends on excitation energy.

- Note that the emission energy does not change.
- Only the energy of excitation changes.
- Changing polarization must result from changing exciton distributions.

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Polarization excitation dependence also depends on wire...

As one comes closer to resonance the relative density of excitons changes.

\[ \frac{n_{\parallel}}{n_{\perp}} \rightarrow 1 \]

\[ \frac{n_{\parallel}}{n_{\perp}} > 1 \]

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Resonant excitation creates non-equilibrium exciton spin distributions

- As excitation comes closer to free exciton energy:
  - Along wire: polarization increases
  - Perpendicular: polarization decreases

- Polarization are different for different wires

- Wire 2: thermal equilibrium
  \[ N_{\parallel} = N_{\perp} \]
Exciton Dynamics

\[ \tau_{x,z} = \tau_y \left( 1 + \varepsilon_s \right)^2 \]

\[ \tau_y = \tau_{\text{vac}} = \frac{3\pi \varepsilon_0 \hbar c_0^3}{\omega_{\text{exc}}^3 D_{\text{exc}}^2} \]

At thermal equilibrium (highest energies) assume:

\[ n_x = n_y \quad \Rightarrow \quad \frac{I_\parallel}{I_\perp} = \frac{\tau_y}{\tau_x} \]

\[ \tau_{x,z} >> \tau_y >> \tau_{nr}, \tau_s \quad \text{and} \quad \frac{I_\parallel}{I_\perp} << 1 \]

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Spin scattering time

Steady state: \( \frac{dn_\alpha}{dt} = 0 \)

\[
\frac{\tau_s}{\tau_{nr}} = \frac{I_\perp(1+P)}{I_\parallel(1-P)} - 1 \quad \text{for} \quad \parallel
\]

\[
\frac{\tau_s}{\tau_{nr}} = \frac{I_\perp(1-P)}{I_\parallel(1+P)} - 1 \quad \text{for} \quad \perp
\]

Spin relaxation time depends on excitation energy

“Non-Equilibrium Exciton Spin Dynamics in Resonantly Pumped Single Core-Shell GaAs-AlGaAs Nanowires”


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Nano Letters - Web release 15 Feb '07
Conclusions

Single GaAs-AlGaAs NWs under resonant excitation:

- Resonances observed at 1-LO and 2-LO and ~133meV (AlGaAs related) above the PL emission line

- Resonant excitation creates non-equilibrium exciton dipole distributions
  - Polarization of PL is strongly enhanced as excitation energy comes closer to resonance with free exciton emission.

- Rate equations: dependent of spin relaxation time on excitation energy

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Rate equations

\[
\begin{align*}
\frac{dn_x}{dt} &= G_x - \frac{n_x}{\tau_x} - \frac{n_x}{\tau_{nr}} - \frac{2 \cdot n_x}{\tau_s} + \frac{n_y}{\tau_s} + \frac{n_z}{\tau_s}, \\
\frac{dn_y}{dt} &= G_y - \frac{n_y}{\tau_y} - \frac{n_y}{\tau_{nr}} - \frac{2 \cdot n_y}{\tau_s} + \frac{n_x}{\tau_s} + \frac{n_z}{\tau_s}, \\
\frac{dn_z}{dt} &= -\frac{n_y}{\tau_z} - \frac{n_y}{\tau_{nr}} - \frac{2 \cdot n_y}{\tau_s} + \frac{n_x}{\tau_s} + \frac{n_z}{\tau_s},
\end{align*}
\]