Imaging the Electronic and Vibronic States of Single Semiconductor Nanowires

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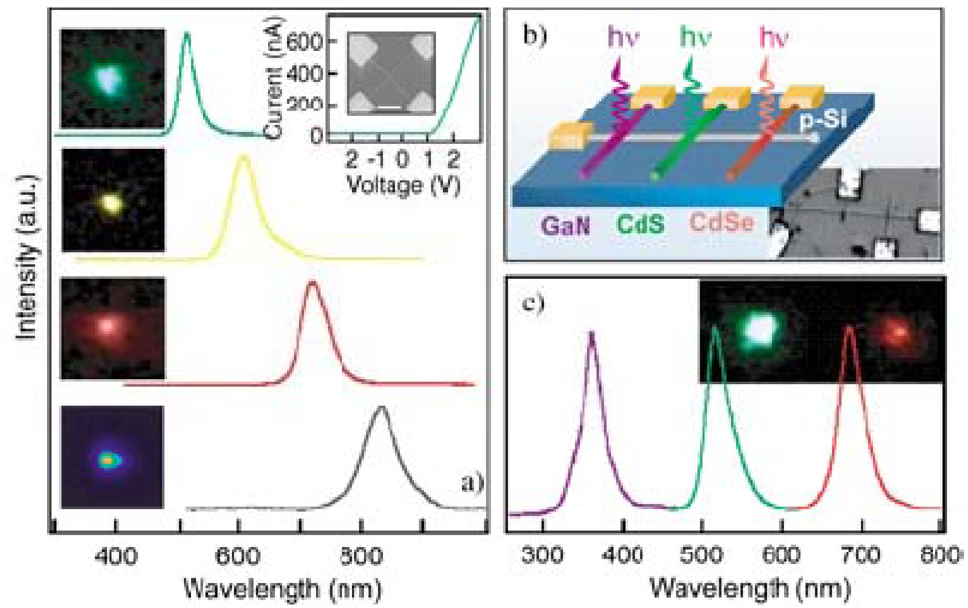
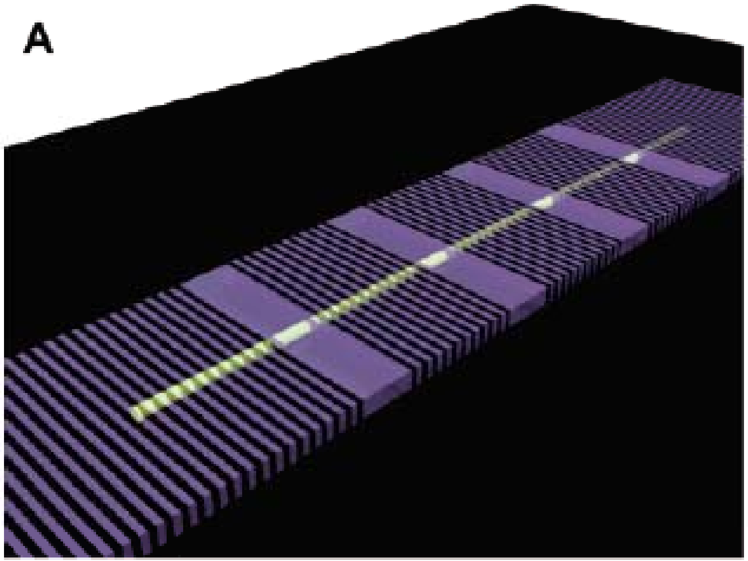
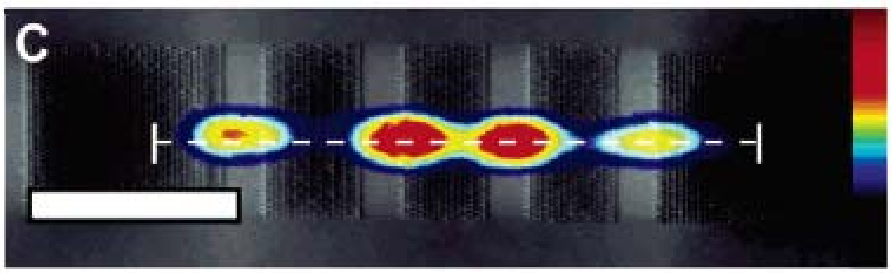
TMS-Orlando, 2007

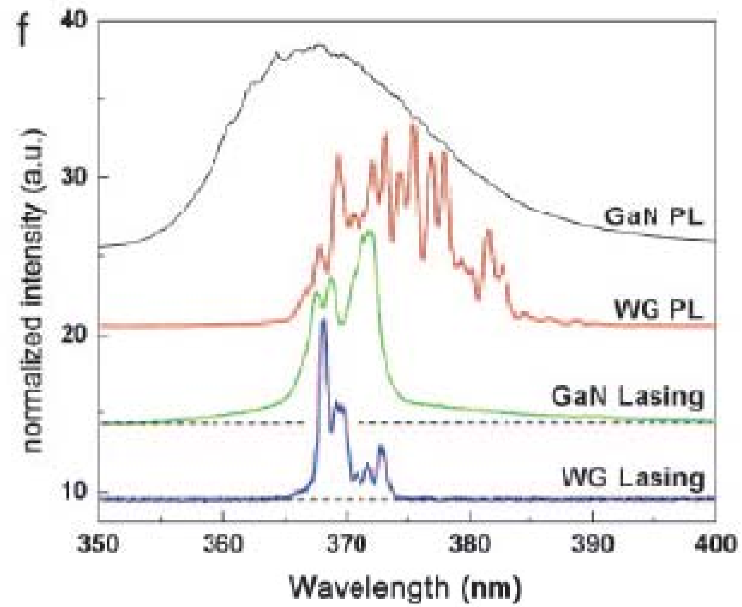
Semiconductor Nanowires as Photodetectors



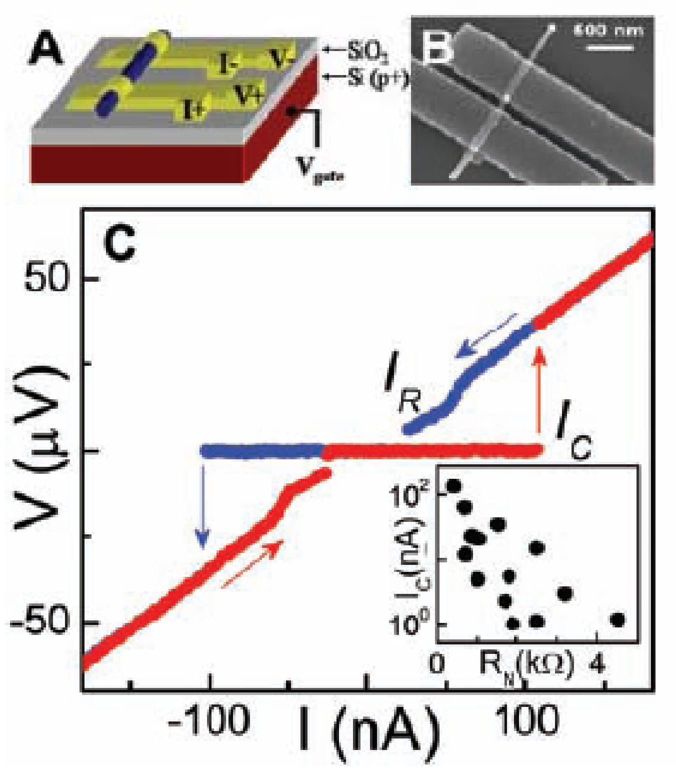
LED and Laser Nanowires

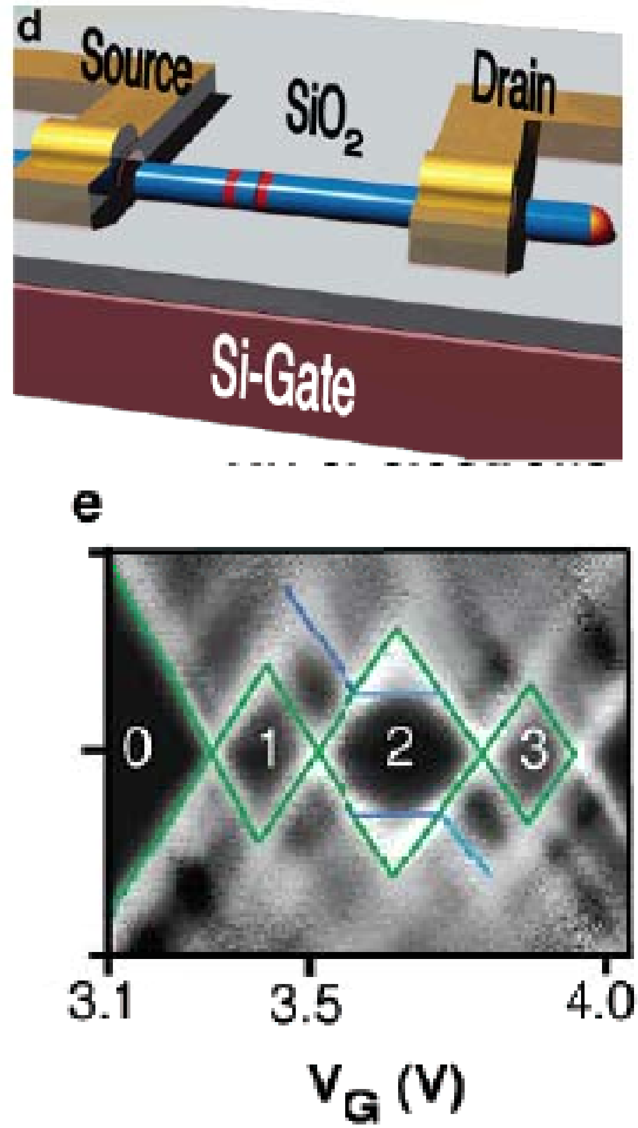




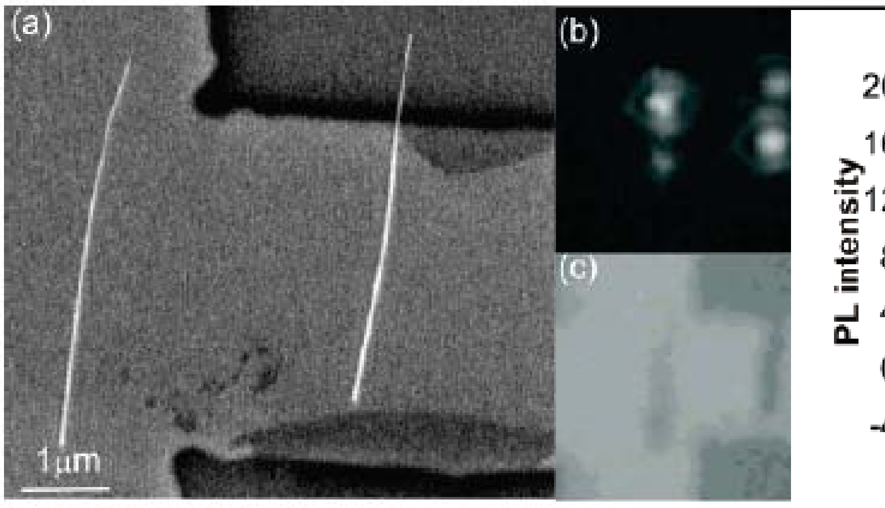


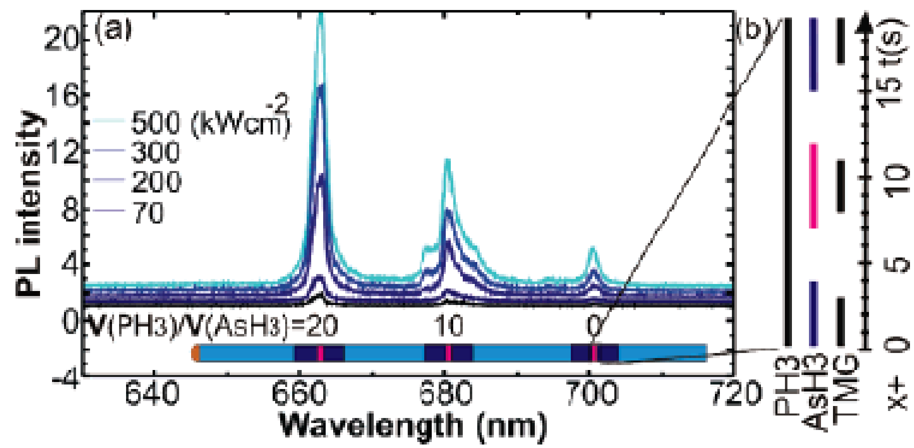
Nanowires as Single Electron Transistors

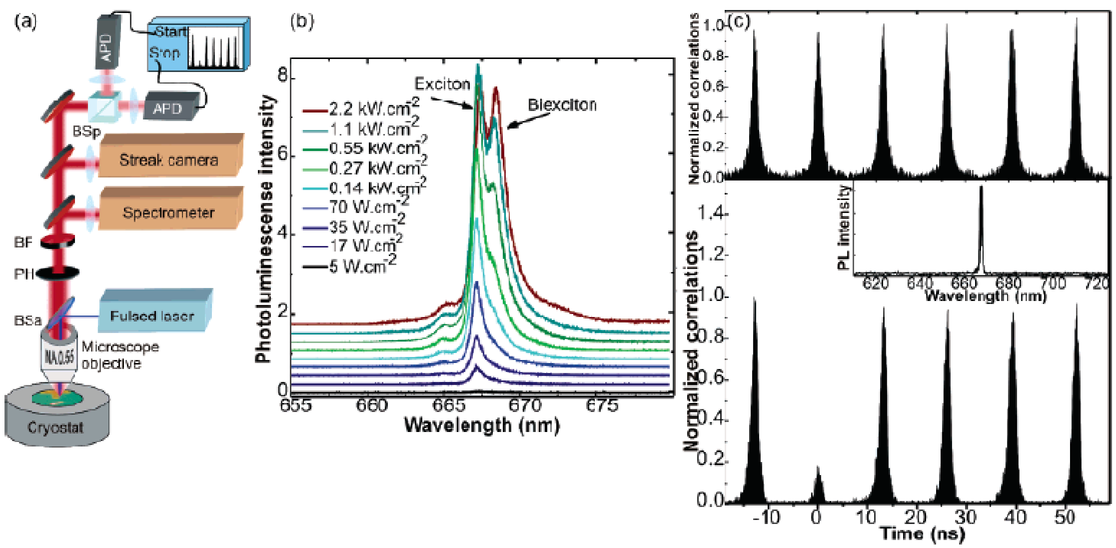




Nanowires as single photon emitters







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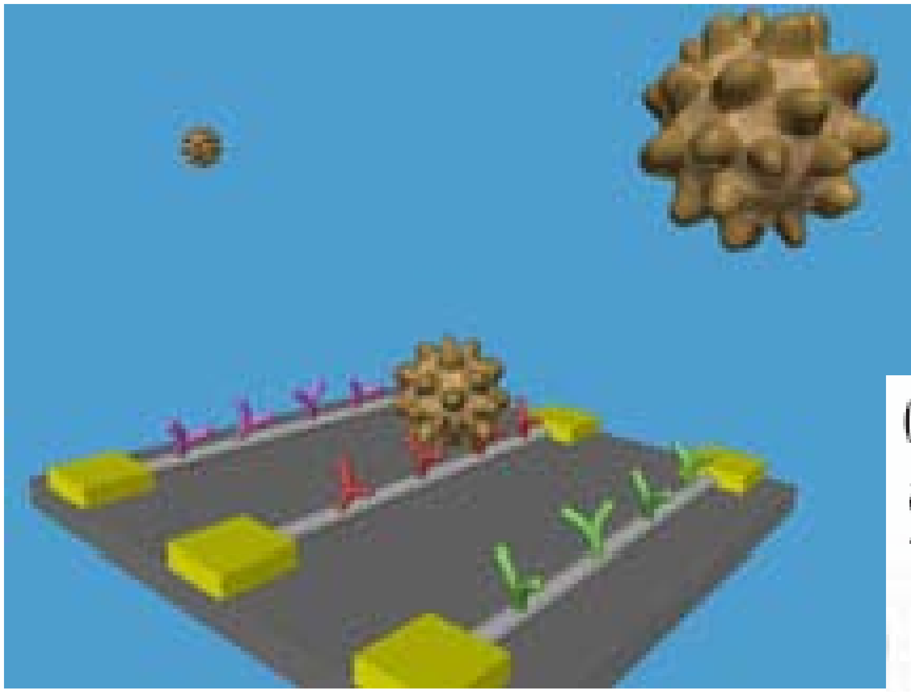
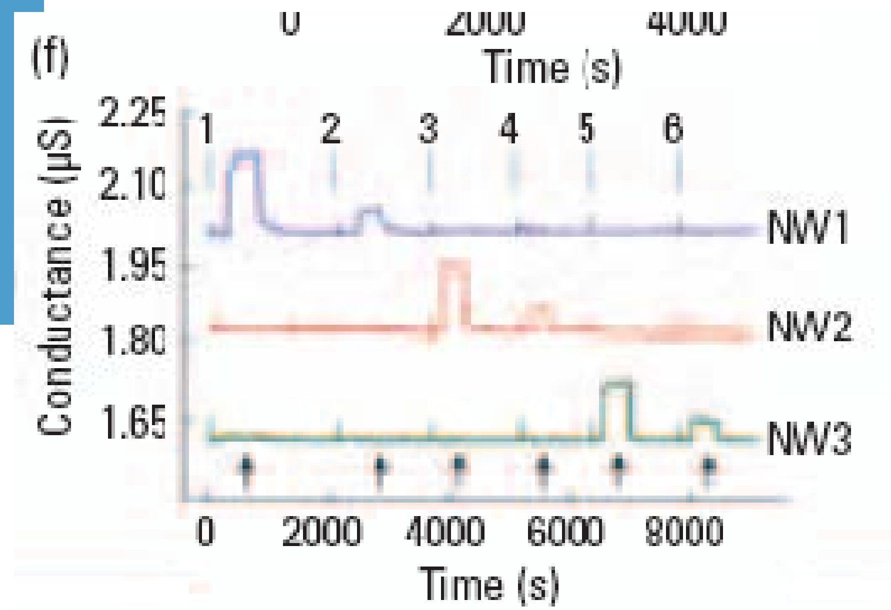
Nanowires as Biosensors





Core-Shell Nanowire Growth

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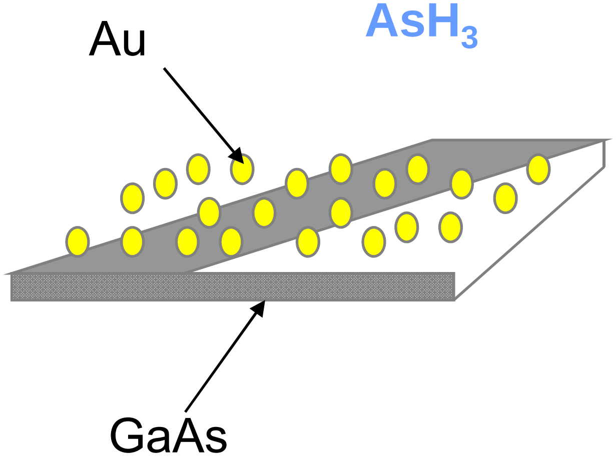
Pre-growth



600°C, 10 min

Desorb surface contaminants
and form eutectic alloy.

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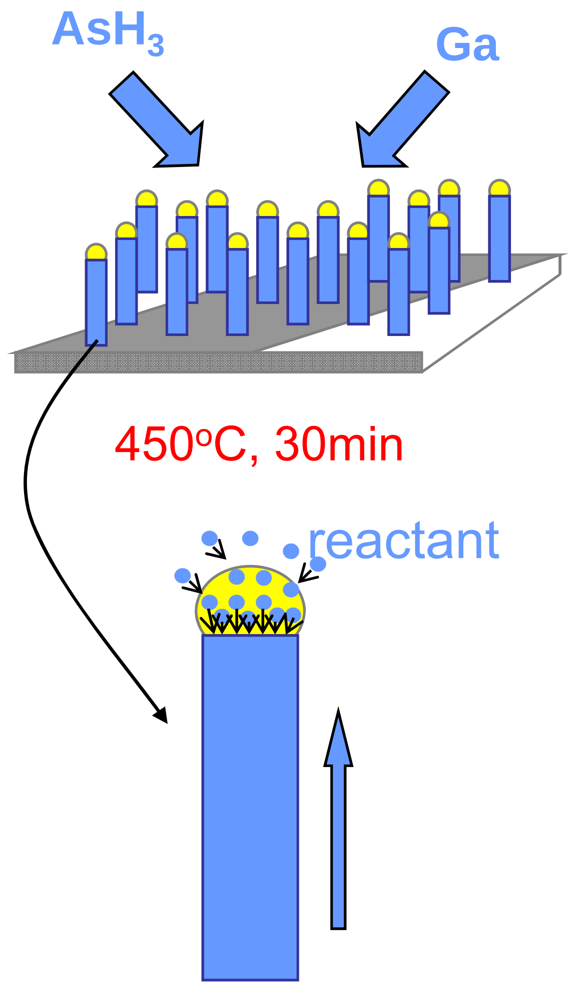
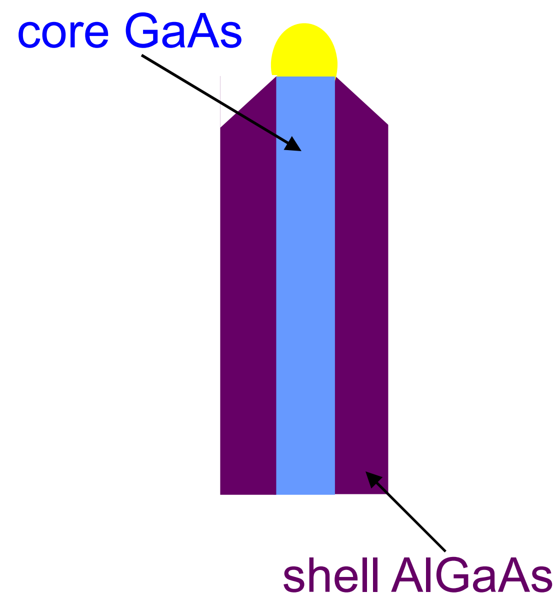
Core: GaAs



Vapor-Liquid-Solid growth

Shell: AlGaAs



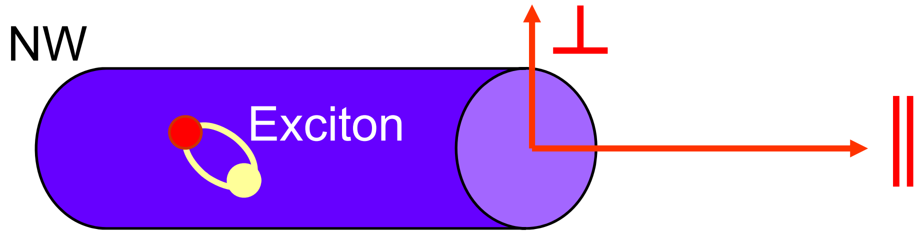
650°C, 15 min

Wire diameter is determined by Au catalyst and shell growth time

Motivation

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

~ bulk exciton



Dielectric "confinement" of EM dipole field (D<<λ):

Exciton density

$$N_{\parallel} = N_{\perp}$$

$\Longrightarrow$

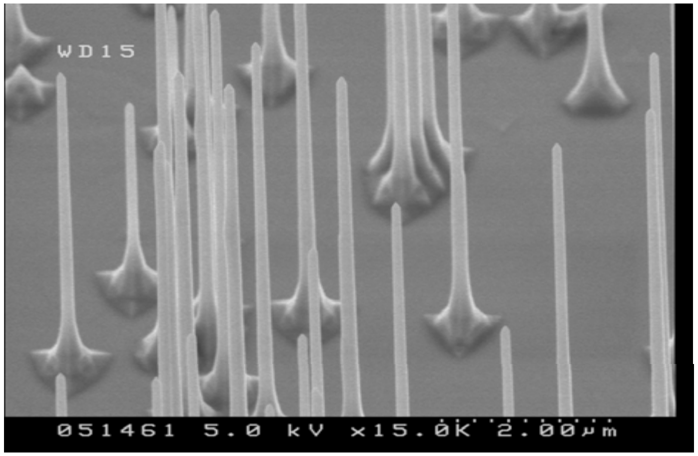
Photoluminescence intensities

$$I_{\parallel} >> I_{\perp}$$

$$\tau_{\parallel} << \tau_{\perp}$$

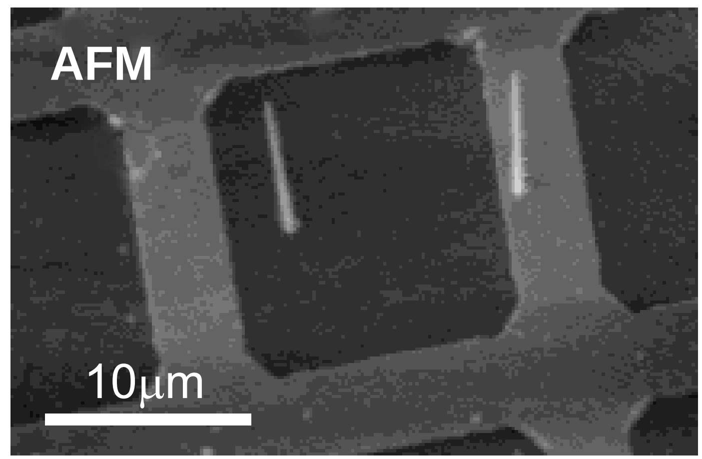
We are interested in exciton spin dynamics of single nanowires

Single nanowire studies



Field-Emission Scanning Electron Microscope (FESEM) image

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



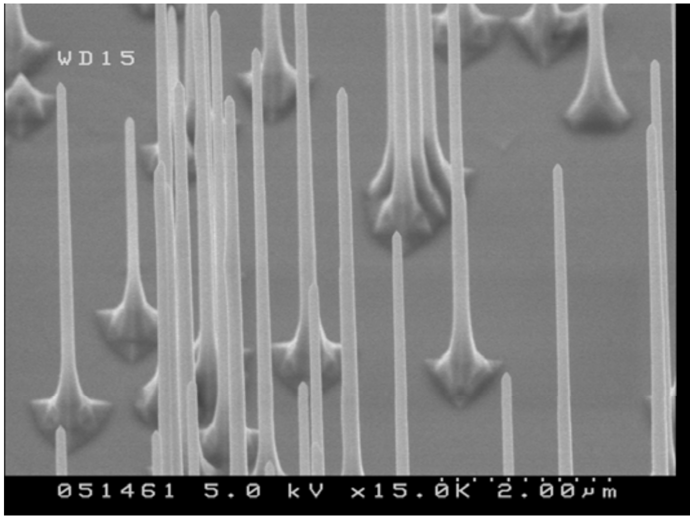
a single nanowire:

~80nm in diameter, ~5-8 µm long

wire's diameter > Bohr exciton diameter

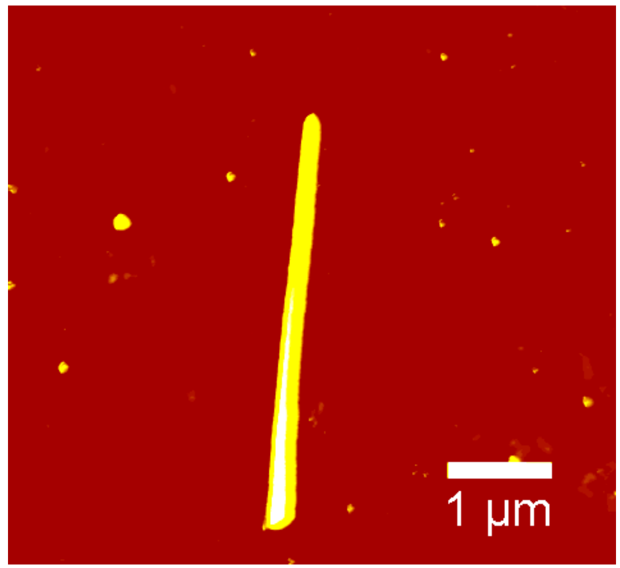
=> expect no quantum confinement effects

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

Photoluminescence Imaging

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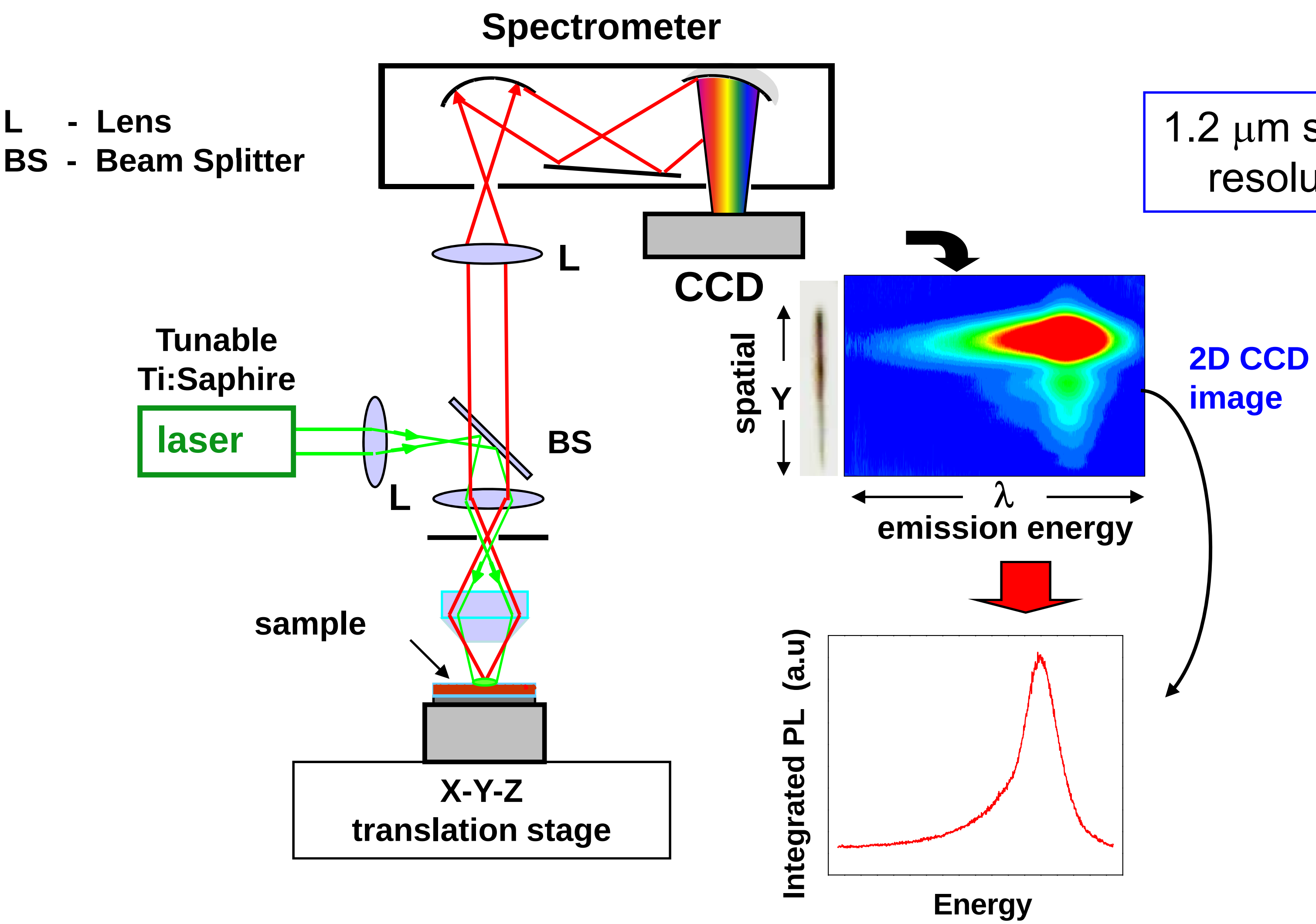

1.2 µm spatial resolution

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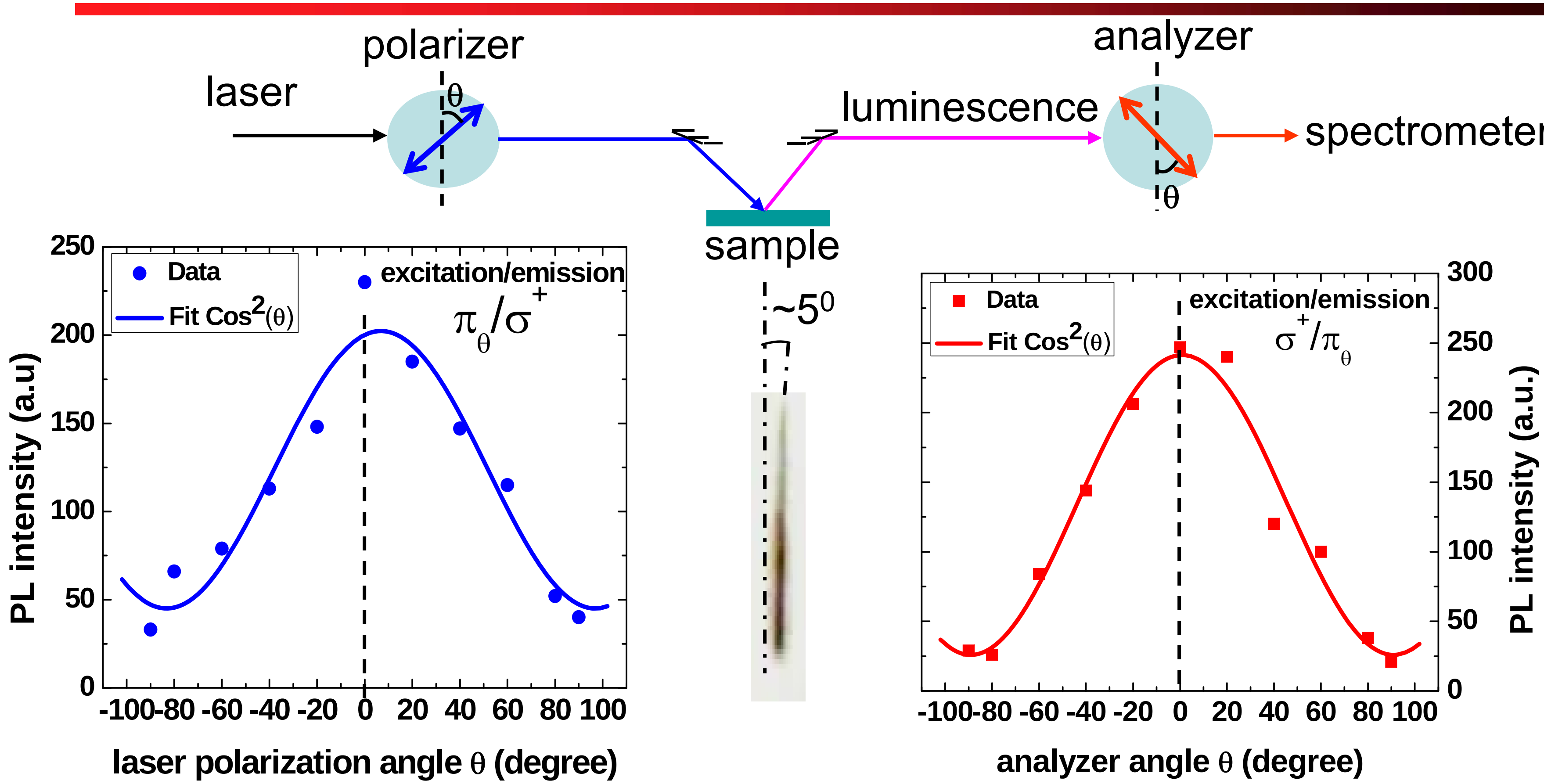
Slit-confocal microscopy

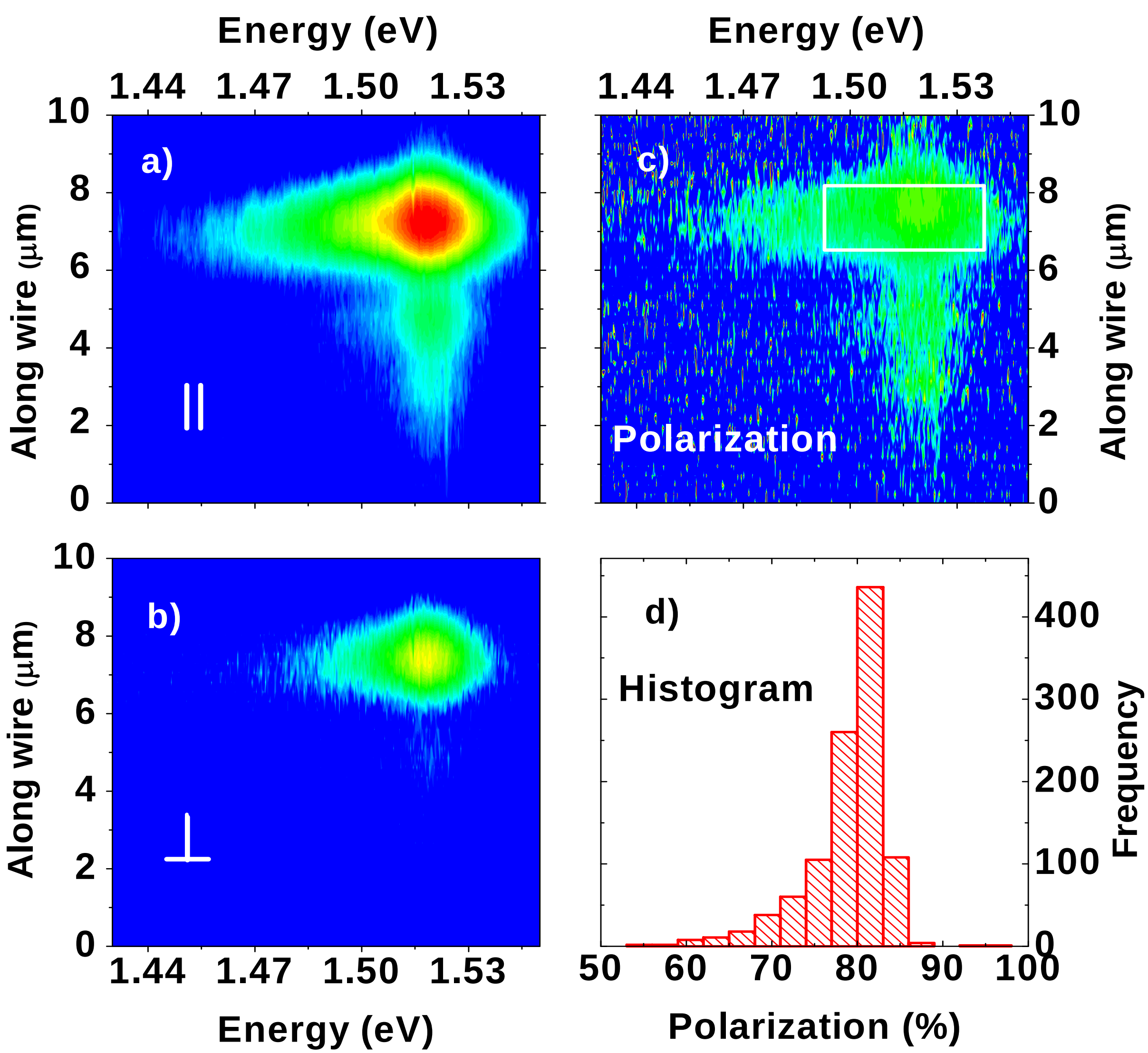
Polarization studies



polarizer = π_θ ; analyzer = σ^+ polarizer = σ^+ ; analyzer = π_θ

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

Polarization Imaging



Calculate pixel by pixel

$$P = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}}$$

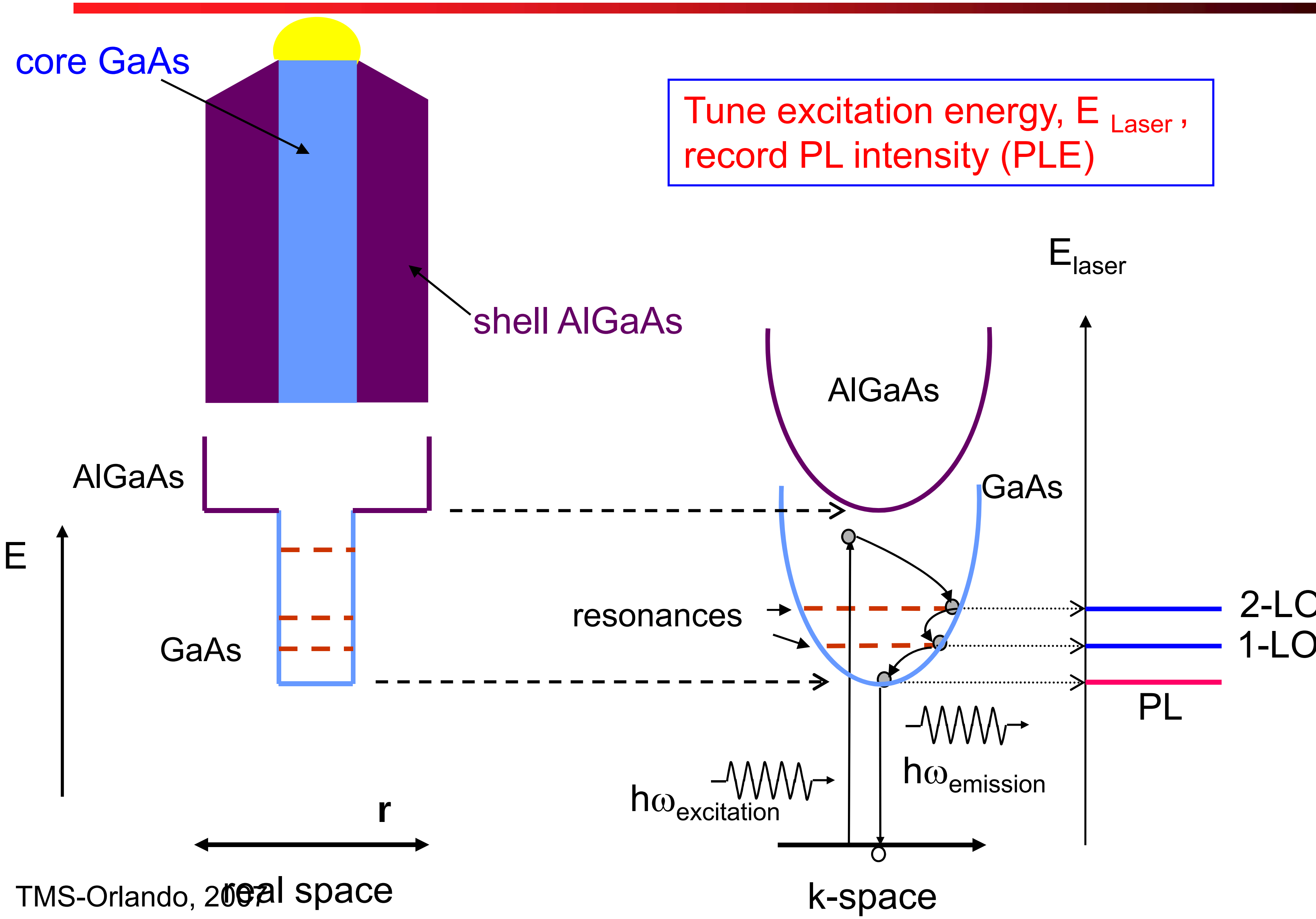
~82%

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

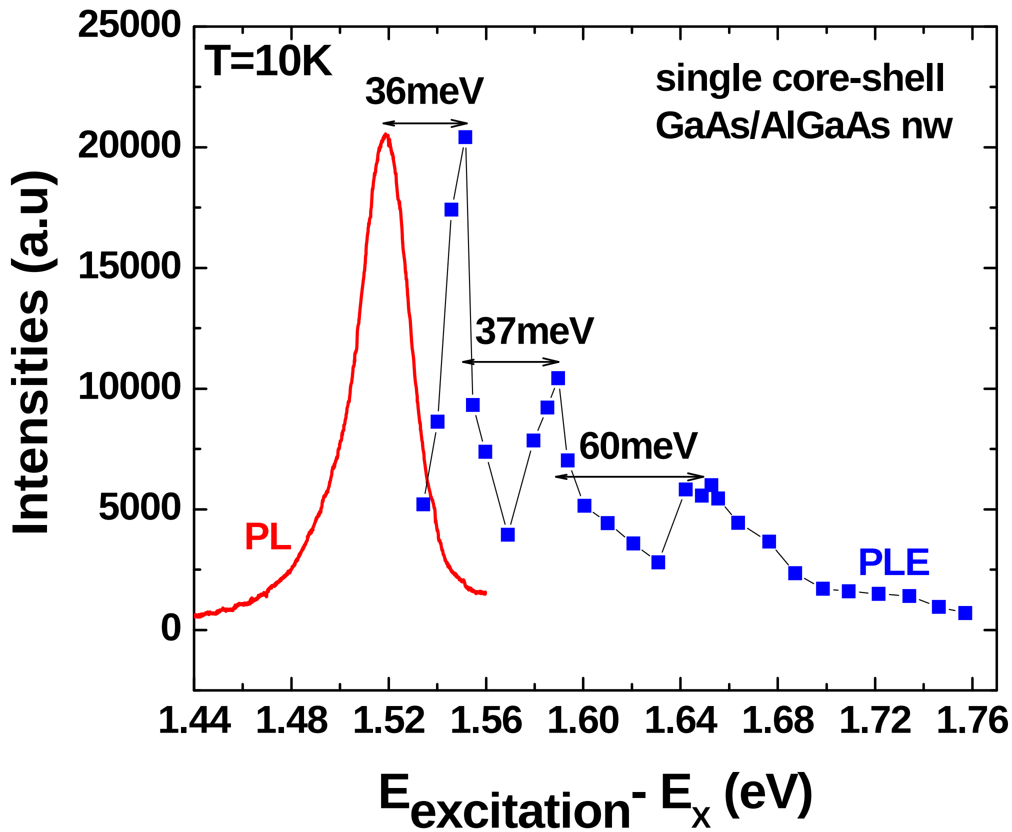
(Science 293 1455 (2001), APL. 89 173126 (2006))

Resonant Excitation

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

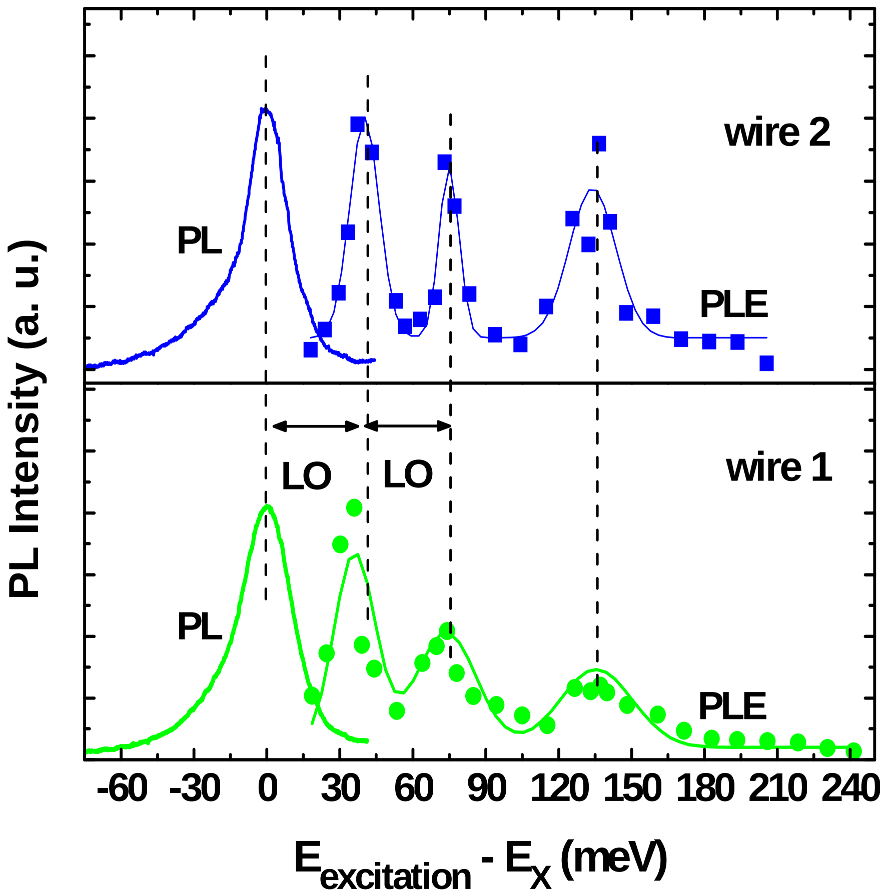


Resonant Excitation



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

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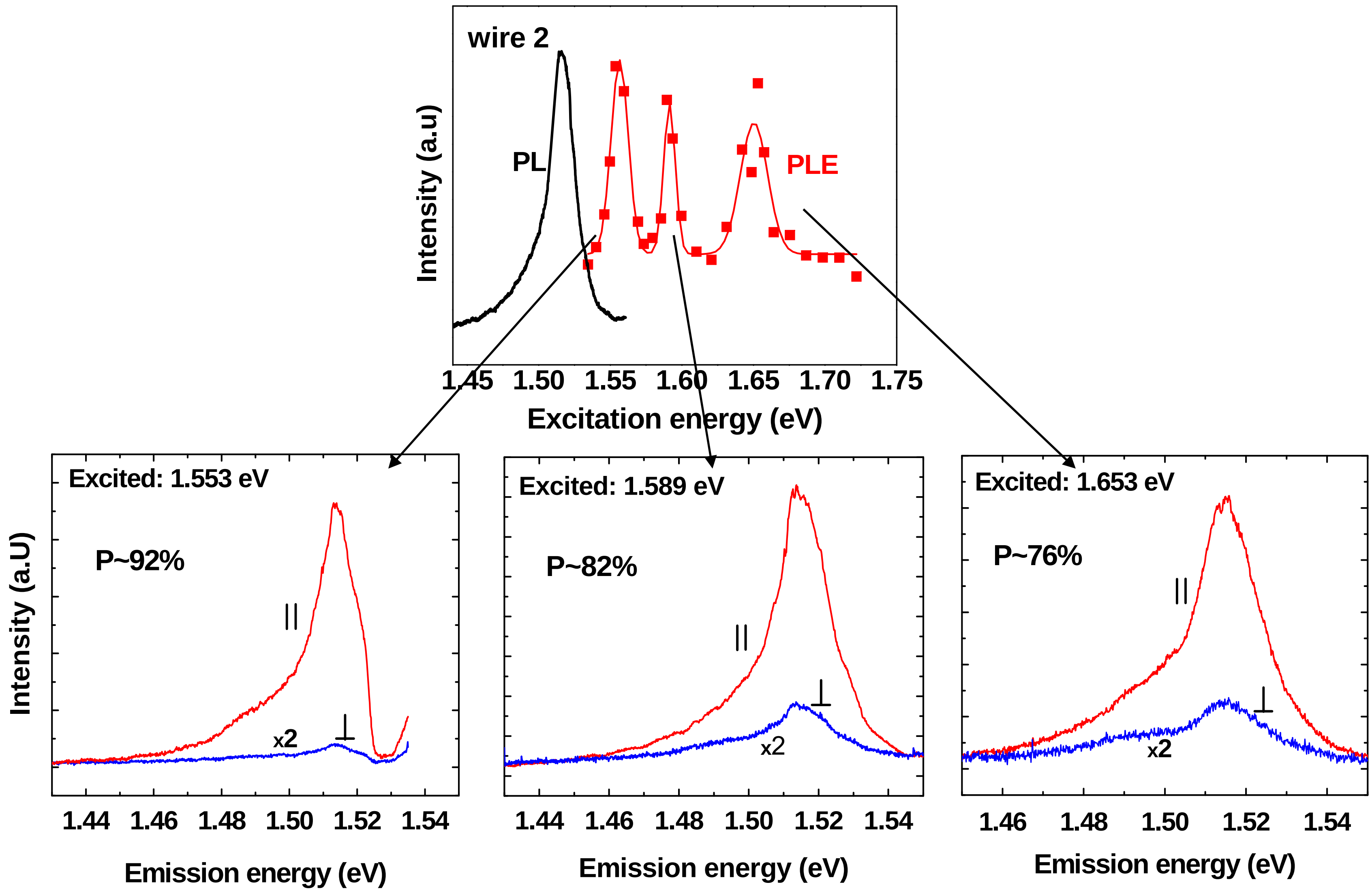

Polarization changes with excitation energy!

PL Polarization Imaging

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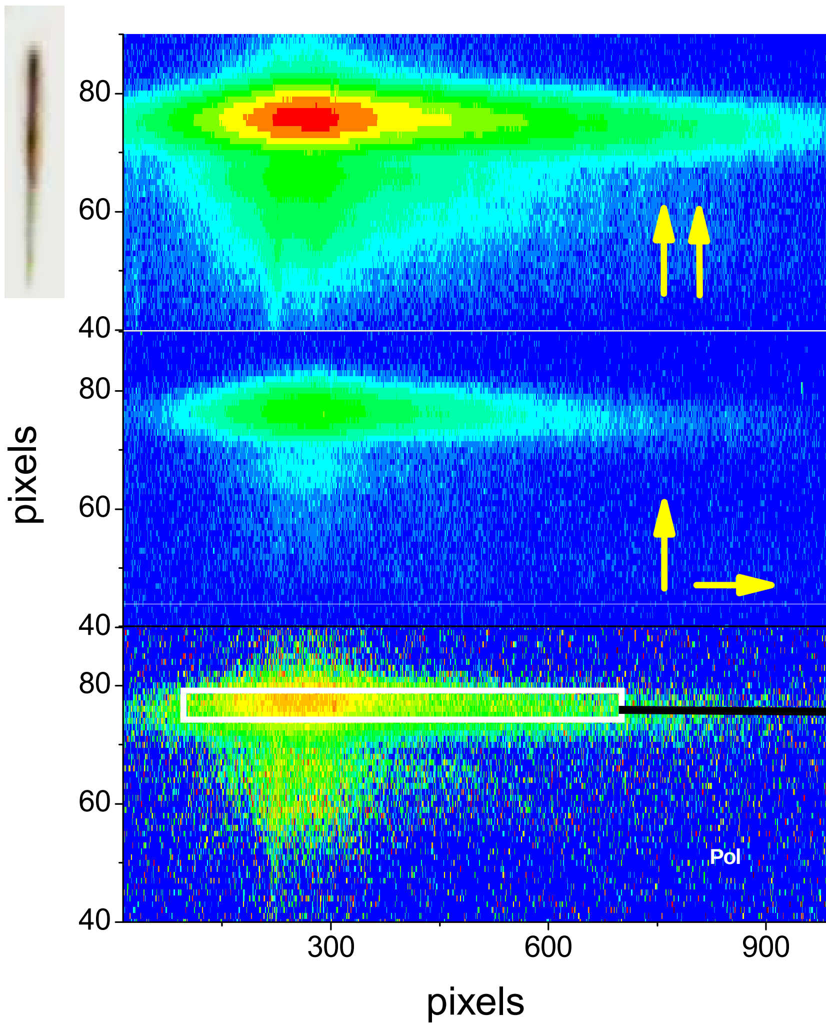
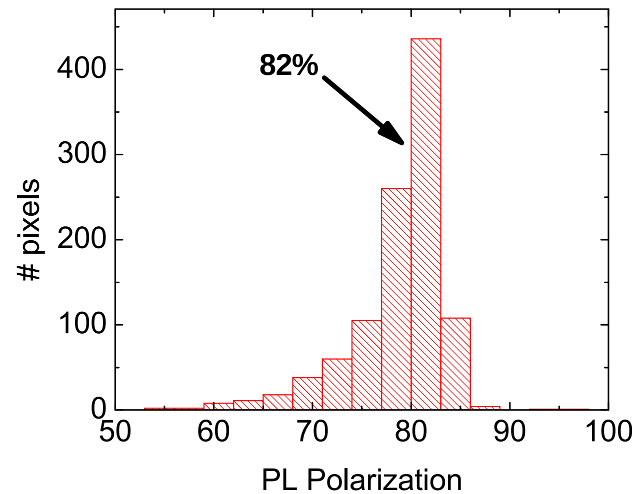


- Excitation laser polarized along nanowire
- Analyze emission polarization

$$P = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}}$$



Polarization depends on excitation energy

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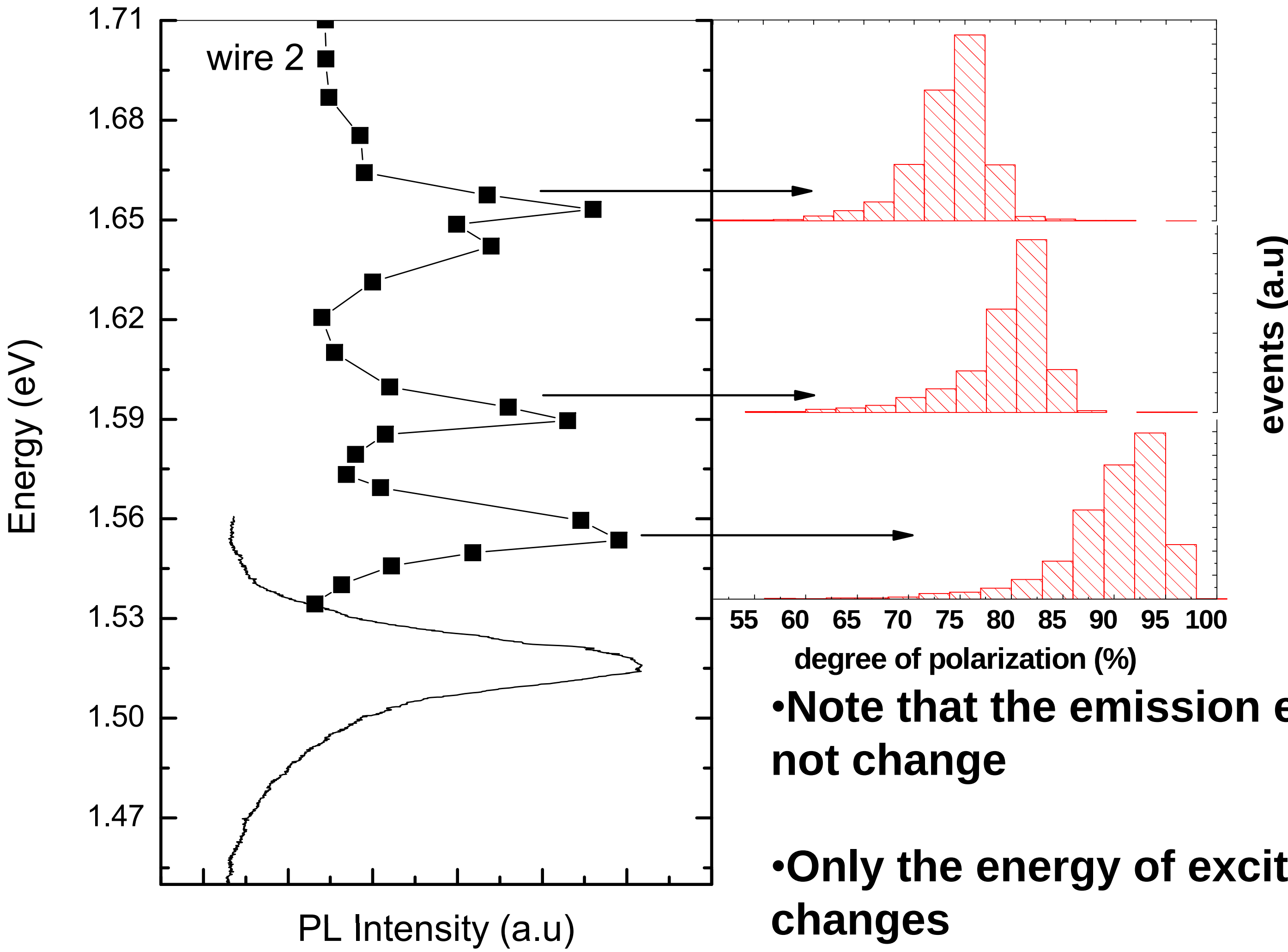
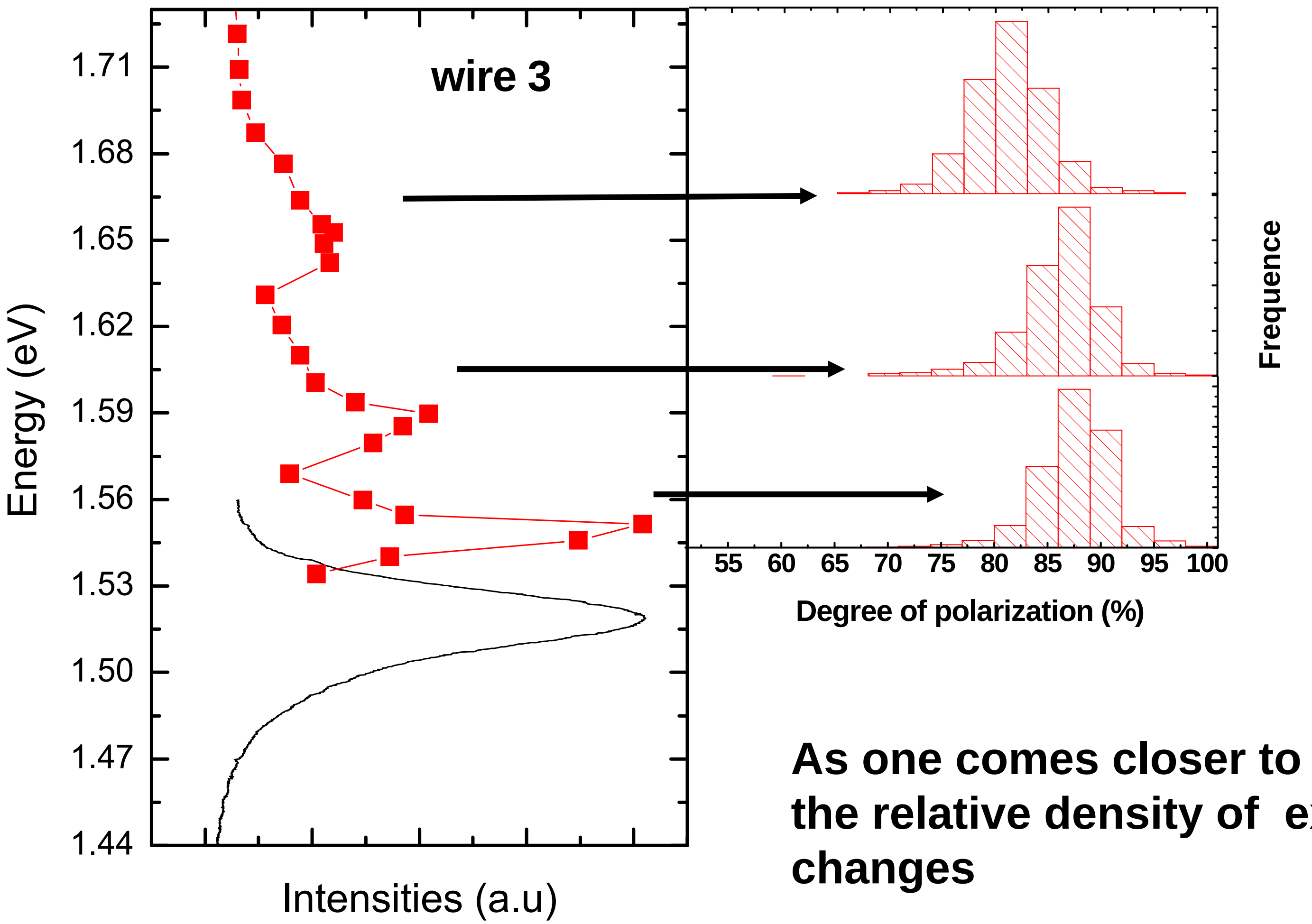


- Note that the emission energy does not change
- Only the energy of excitation changes
- Changing polarization must result from changing exciton distributions

Polarization excitation dependence also depends on wire…



$$\frac{n_{\parallel}}{n_{\perp}} \rightarrow 1$$

$$\frac{n_{\parallel}}{n_{\perp}} > 1$$

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

Resonant excitation creates non-equilibrium exciton spin distributions

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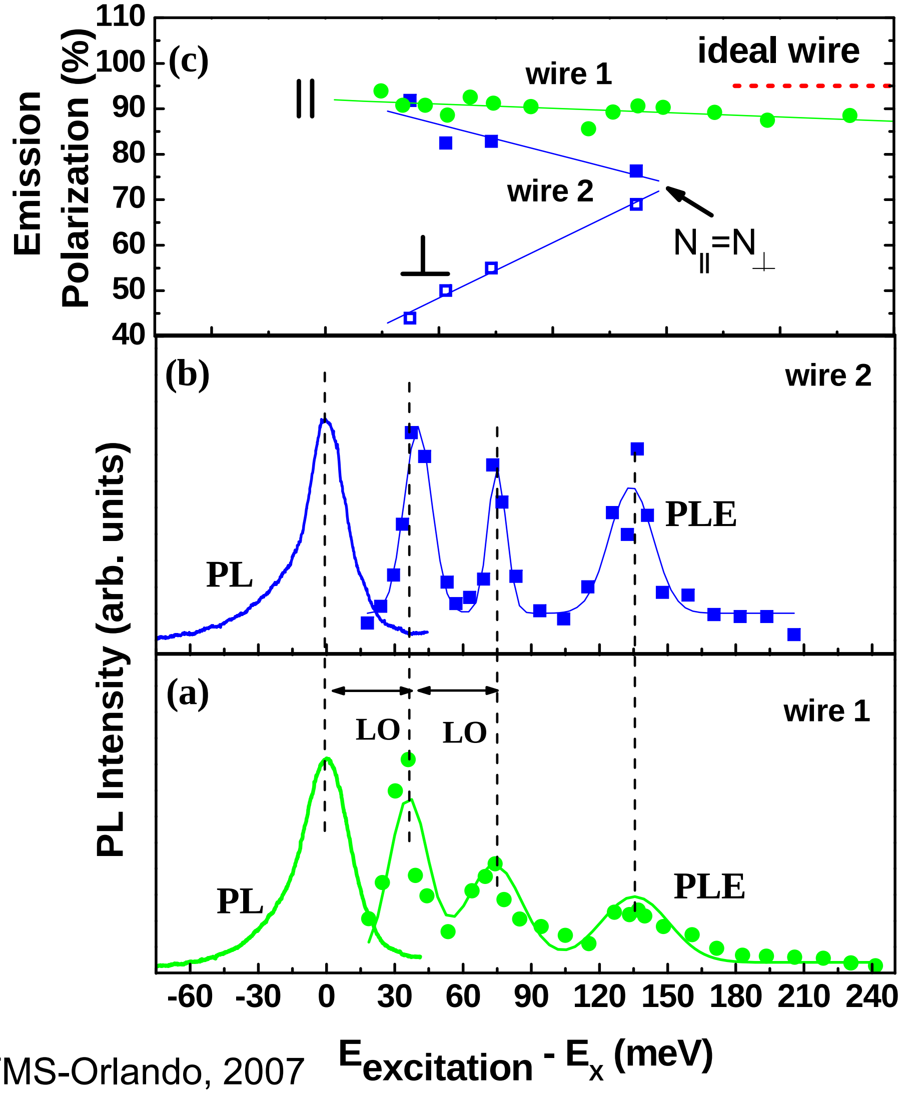
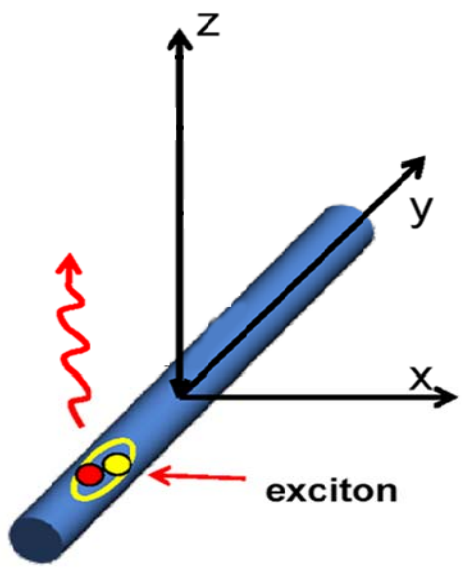
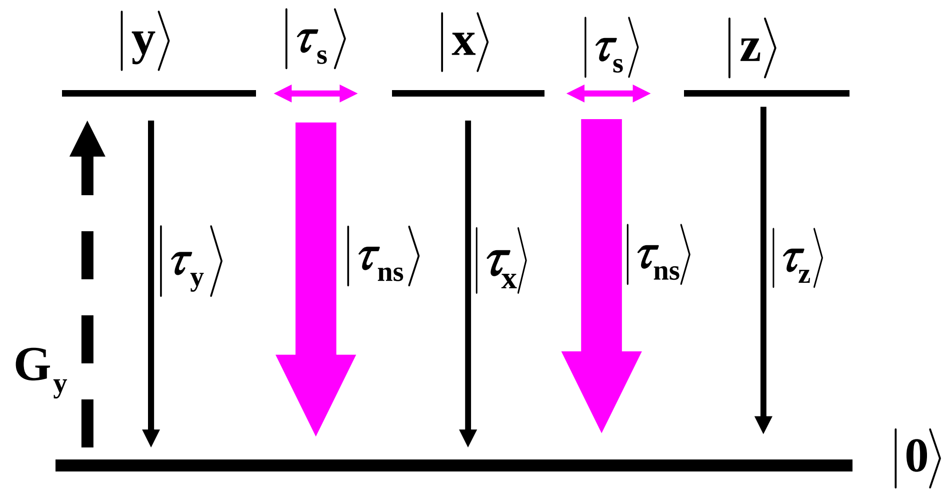


- As excitation comes closer to free exciton energy:
  - Along wire: polarization *increases*
  - Perpendicular: polarization *decreases*
- Polarization are different for different wires
- Wire 2: thermal equilibrium

  $N_{\parallel} = N_{\perp}$

Exciton Dynamics





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

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

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

At thermal equilibrium (highest energies) assume:

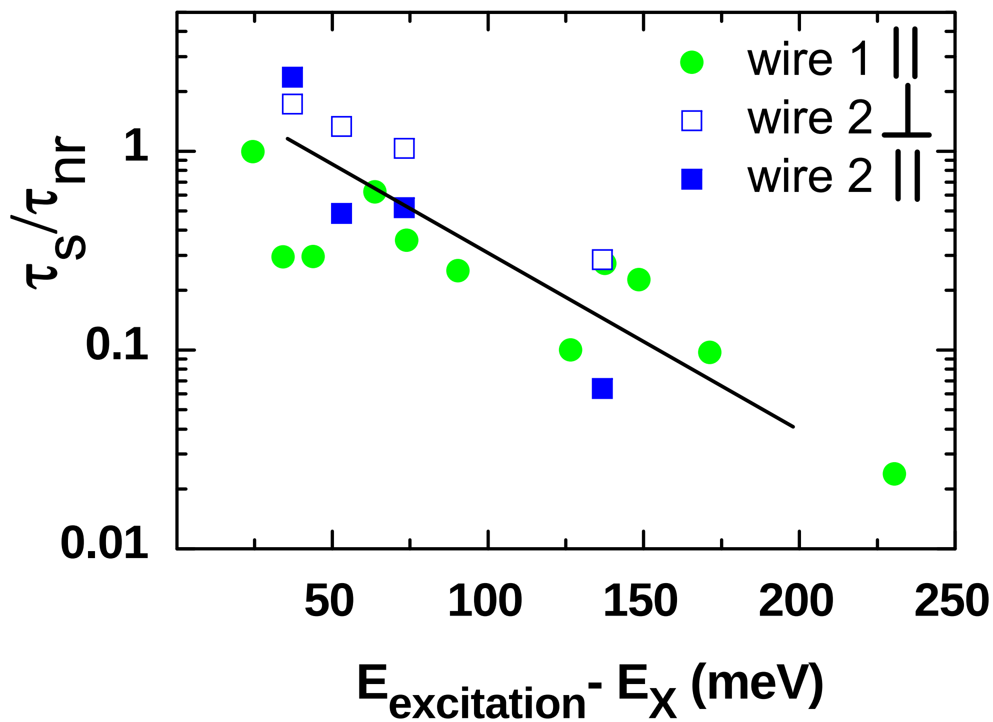
$$n_x = n_y \Longrightarrow \frac{I_\perp}{I_\parallel} = \frac{\tau_y}{\tau_x}$$

Spin scattering time

Steady state: $dn_\alpha / dt = 0$

$$\frac{\tau_s}{\tau_{nr}} = \frac{I_\perp (1+P)}{I_\| (1-P)}) - 1 \quad for \quad \|$$

$$\frac{\tau_s}{\tau_{nr}} = \frac{I_\perp (1-P)}{I_\| (1+P)} - 1 \quad for \quad \perp$$



Spin relaxation time depends on excitation energy

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

Thang. B. Hoang, L.V. Titova, J. M. Yarrison-Rice , H. E. Jackson, , A. O. Govorov, Y. Kim, H. J. Joyce, H. H. Tan, C. Jagadish, L. M. Smith

Nano Letters - Web release 15 Feb '07

Conclusions

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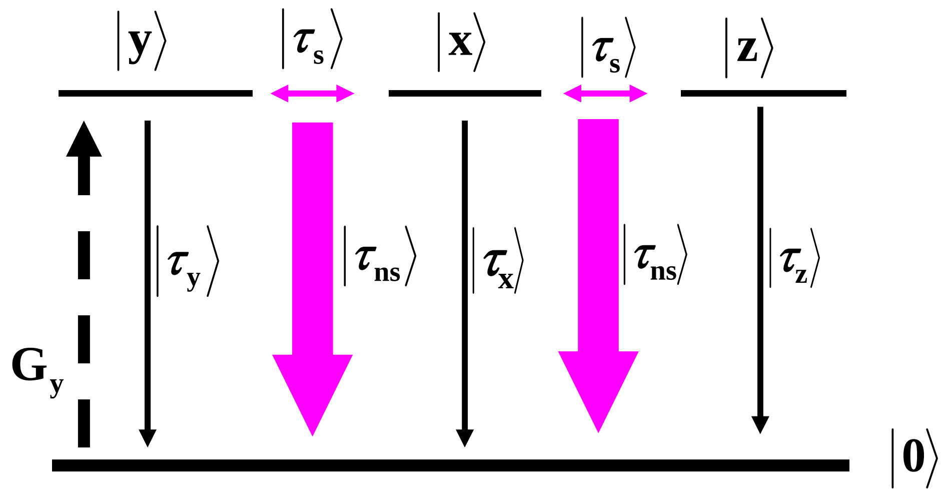
Single GaAs-AlGaAs NWs under resonant excitation:

- **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***

Rate equations



$$\frac{dn_x}{dt} = G_x - \frac{n_x}{\tau_x} - \frac{n_x}{\tau_{nr}} - 2\frac{n_x}{\tau_s} + \frac{n_y}{\tau_s} + \frac{n_z}{\tau_s},$$

$$\frac{dn_y}{dt} = G_y - \frac{n_y}{\tau_y} - \frac{n_y}{\tau_{nr}} - 2\frac{n_y}{\tau_s} + \frac{n_x}{\tau_s} + \frac{n_z}{\tau_s},$$

$$\frac{dn_z}{dt} = -\frac{n_y}{\tau_z} - \frac{n_y}{\tau_{nr}} - 2\frac{n_y}{\tau_s} + \frac{n_x}{\tau_s} + \frac{n_z}{\tau_s},$$