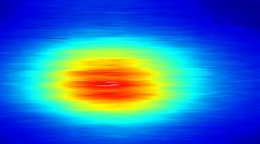


Espectroscopia ótica e a aplicação de ondas acústicas de superfície

Odilon D. D. Couto Jr.



Outline



Introdução

Espectroscopia ótica de nanoestruturas semicondutoras

Resultados

Ondas acústicas de superfície (SAW)

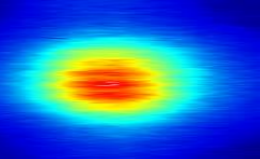
Transporte induzido acusticamente

- ✓ **Portadores (elétrons e buracos)**
- ✓ **Spins**
- ✓ **Injeção de portadores**
- ✓ **Fonte de fótons únicos bombeada acusticamente**

Perspectivas

Conclusão

Going smaller...



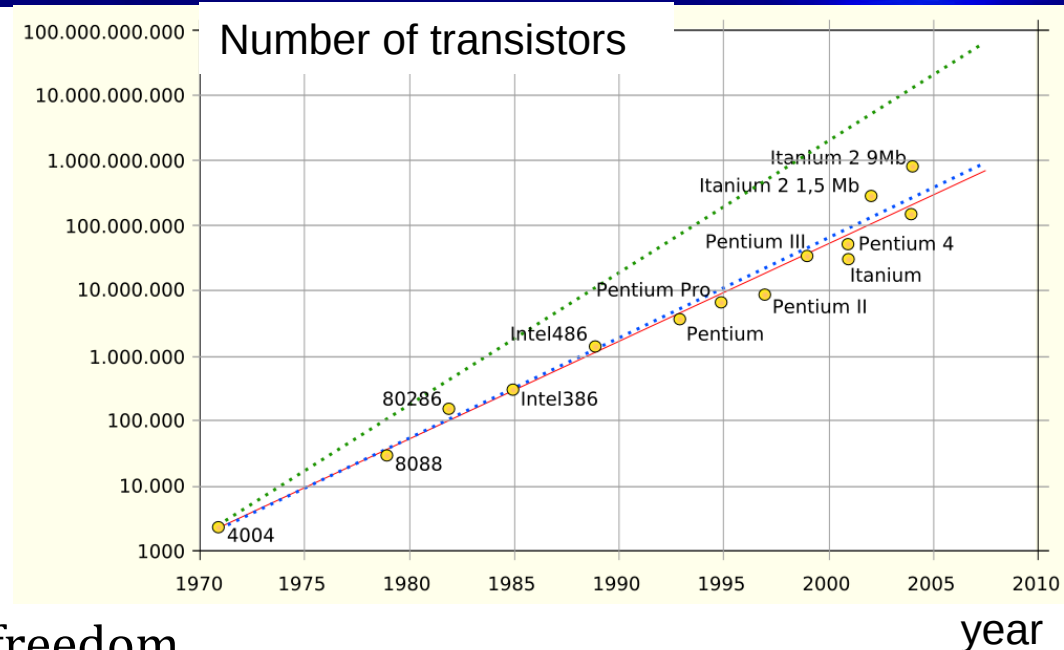
Miniaturization of structures

Transition from micro to nano

- ✓ $1\text{nm} = 10^{-9}\text{m}$
- ✓ Size of an atom $\sim 0.1\text{ nm} = \text{\AA}$
- ✓ Few number of atoms
- ✓ Nano „world“ $\sim$ Quantum „world“
 - ✓ Manipulation of quantum degrees of freedom
 - ✓ Spin – spintronics
 - ✓ Photon – (nano)photonics
 - ✓ Phonon – phononics ... and so on and so forth

Physical properties depend strongly

- ✓ Particle number
- ✓ Dimensionality (3D, 2D, 1D, 0D)



**New functionalities in comparison
to „classical“ devices**

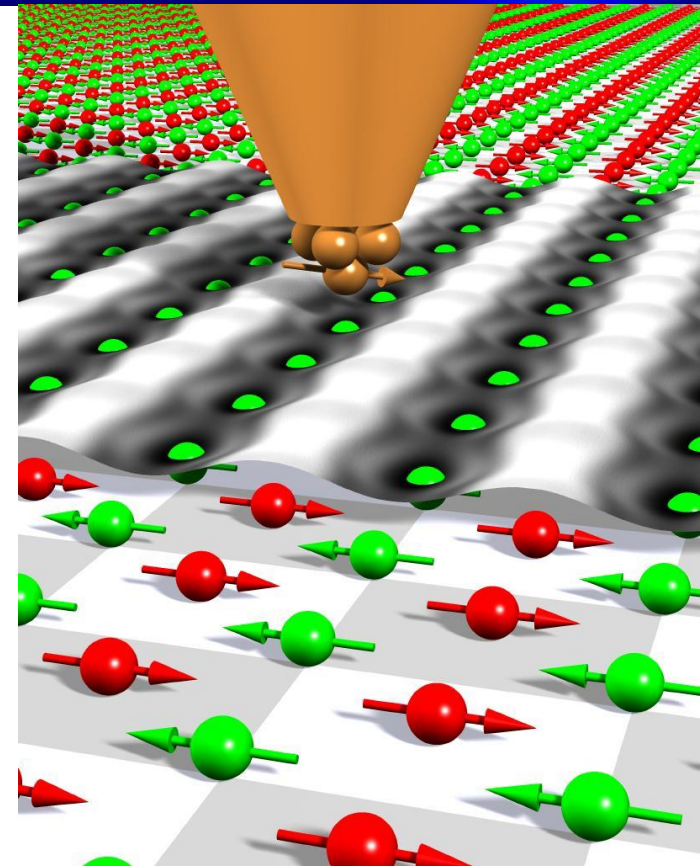
Going more powerful...

For probing nanoscale structures

- ✓ Detection/manipulation have to be modified
 - ✓ Become smaller
 - ✓ More precise
 - ✓ Space, energy, time selectivity

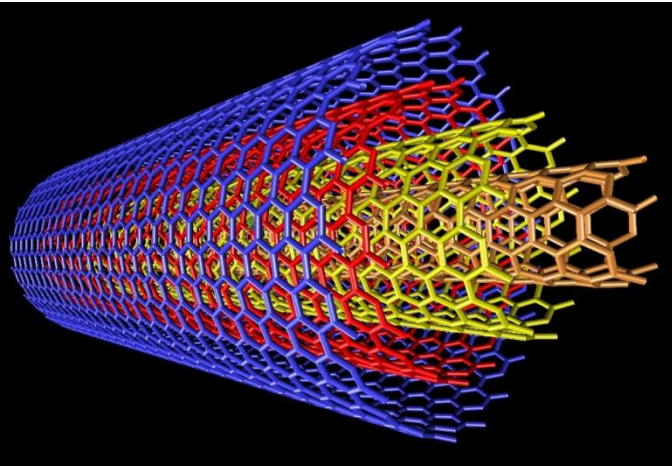
New experimental techniques

- ✓ Sensitive to
 - ✓ Number of particles
 - ✓ Specific interactions
- Quantum information processing

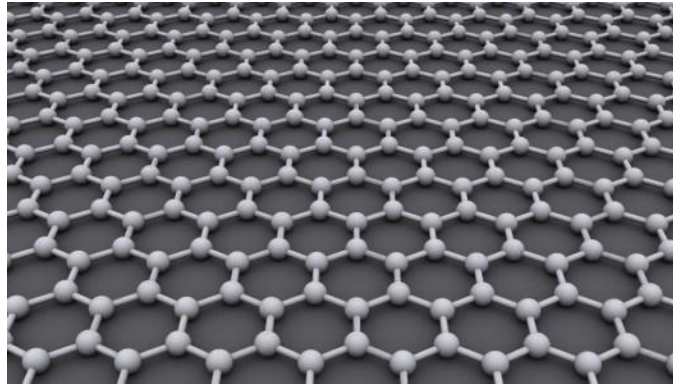


The systems

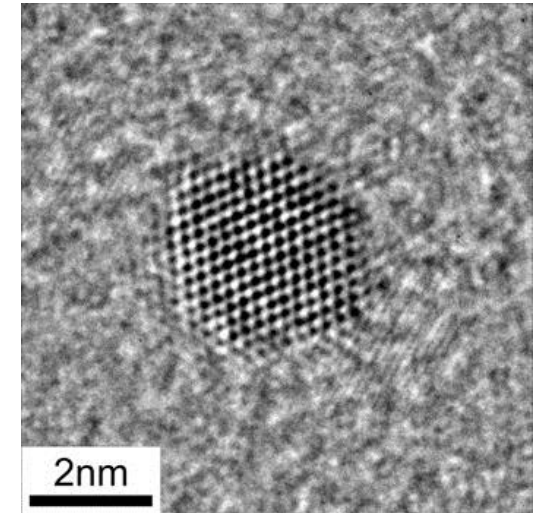
Carbon nanotubes



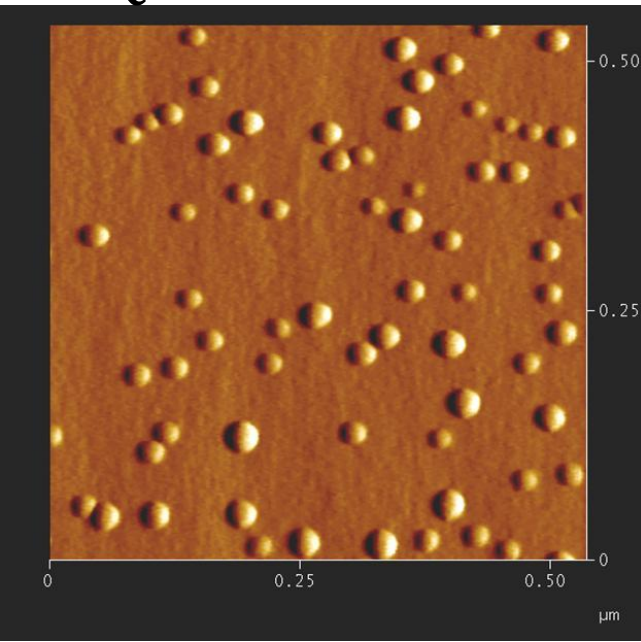
Graphene



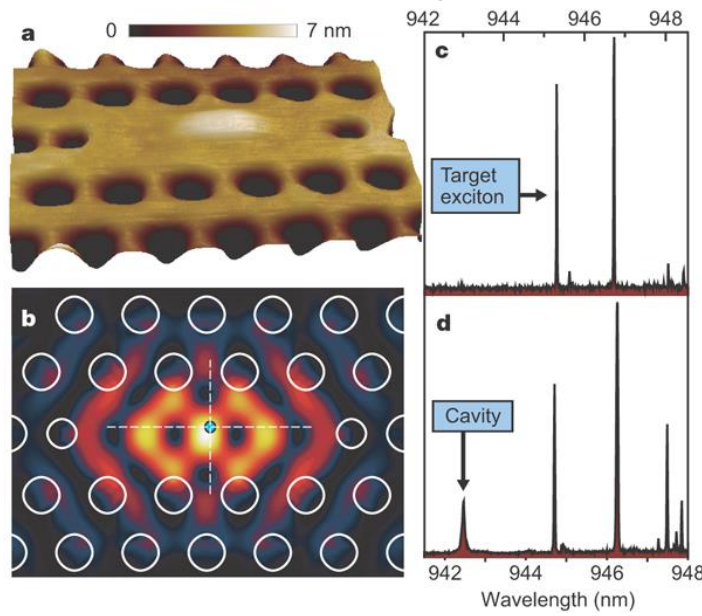
Magnetic nanoparticles



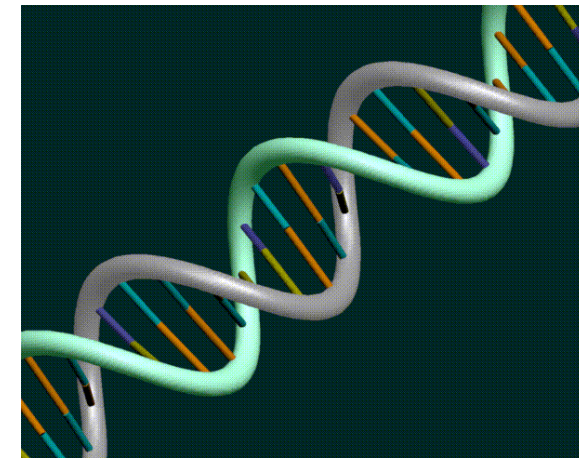
Quantum dots



Photonic crystals



DNA



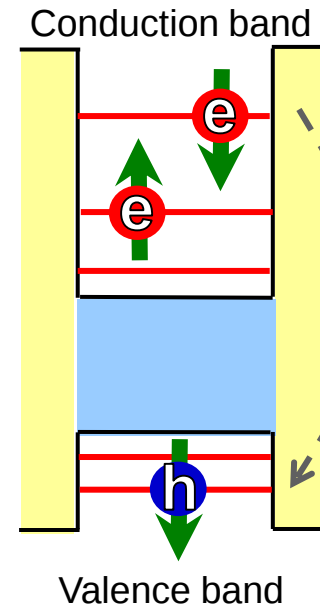
In this talk

Semiconductor nanostructures

Confinement

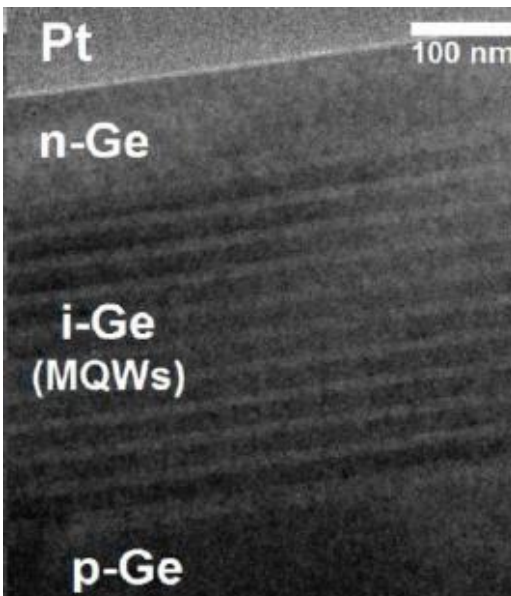
Energy quantization

- **Quantum wells** → (2D)
- **Nanowires** → (1D)
- **Quantum dots** → (0D)

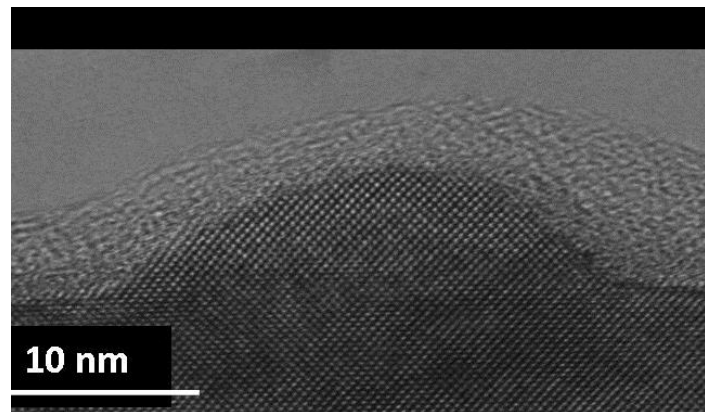


Light emission
↓
**Information about
quantum states**

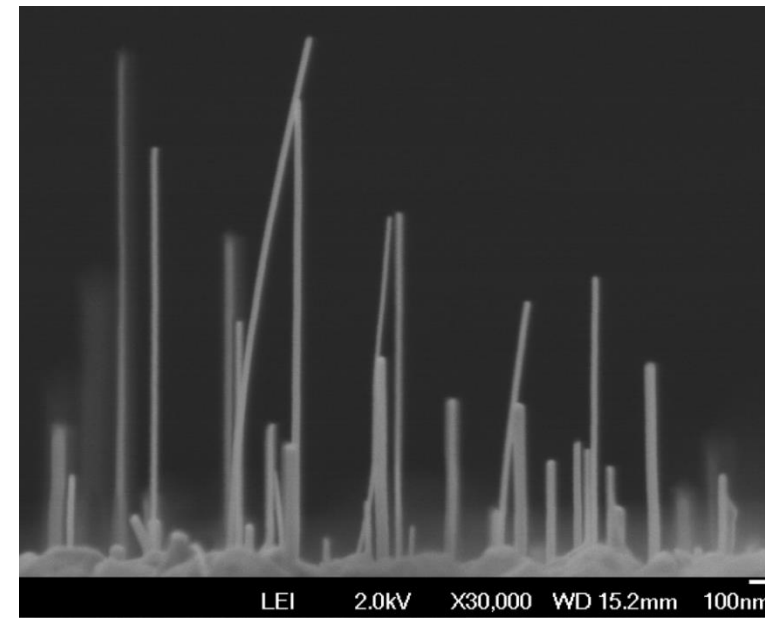
Quantum well (QW)



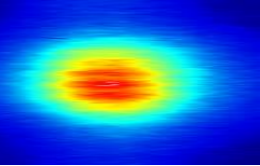
Quantum dot (QD)



Nanowire (NW)



Espectroscopia Ótica



How do we probe the nanostructure energy states?



Optical spectroscopy techniques

Excitação ótica
(laser)



Sistema de interesse
(amostra)



Detecção ótica
(intensidade, energia,
polarização, tempo,
espaço,...)



Técnicas mais usadas em nosso laboratório

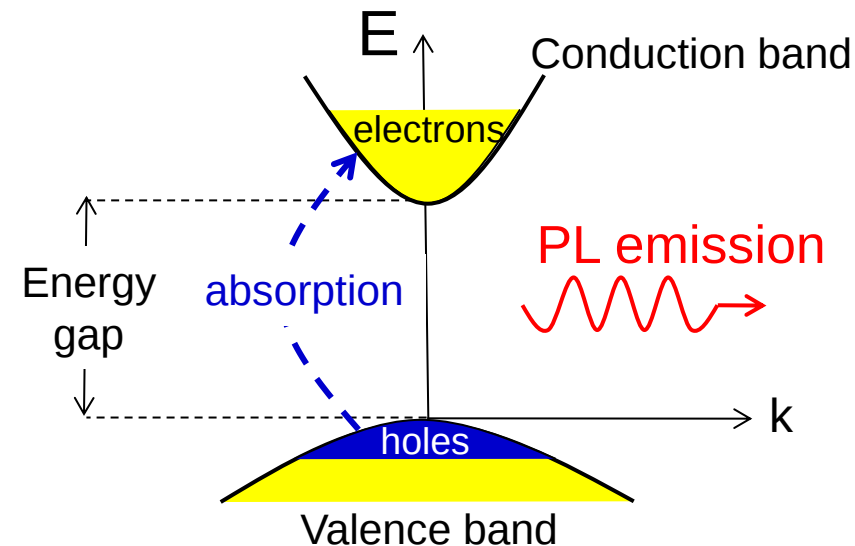
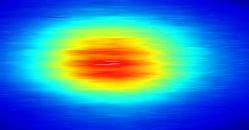
Fotoluminescência

- ✓ Excitação ótica inter ou intra-banda
- ✓ Detecção da recombinação de portadores
- ✓ Informação sobre os níveis de energia
- ✓ Dinâmica de portadores

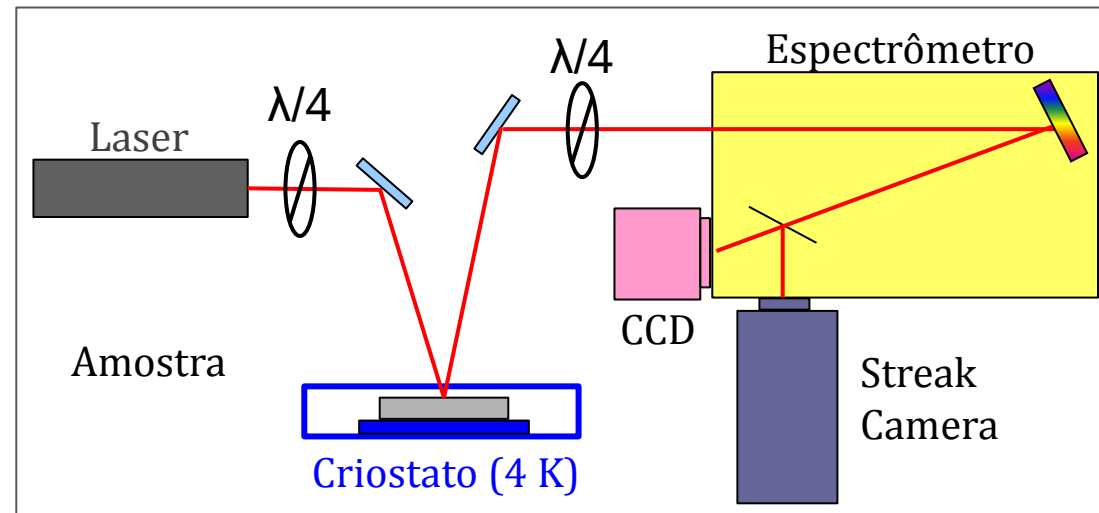
Espalhamento Raman

- ✓ Stoke e anti-Stoke shift
- ✓ Modos de vibração da rede
- ✓ Dispersão de fônons
- ✓ Interação com fônons

Fotoluminescência PL



Simplificadamente



Semiconductor nanostructures

- ✓ Laser excitation → Light absorption
- ✓ Electrons excited to the conduction band
- ✓ Electron-hole recombination
 - ✓ Light emission → Photoluminescence (PL)
 - ✓ Mapping of the energy states

Espectrômetro

- Resolução em energia

Camera CCD

- Intensidade da emissão e resolução espacial

Streak Camera

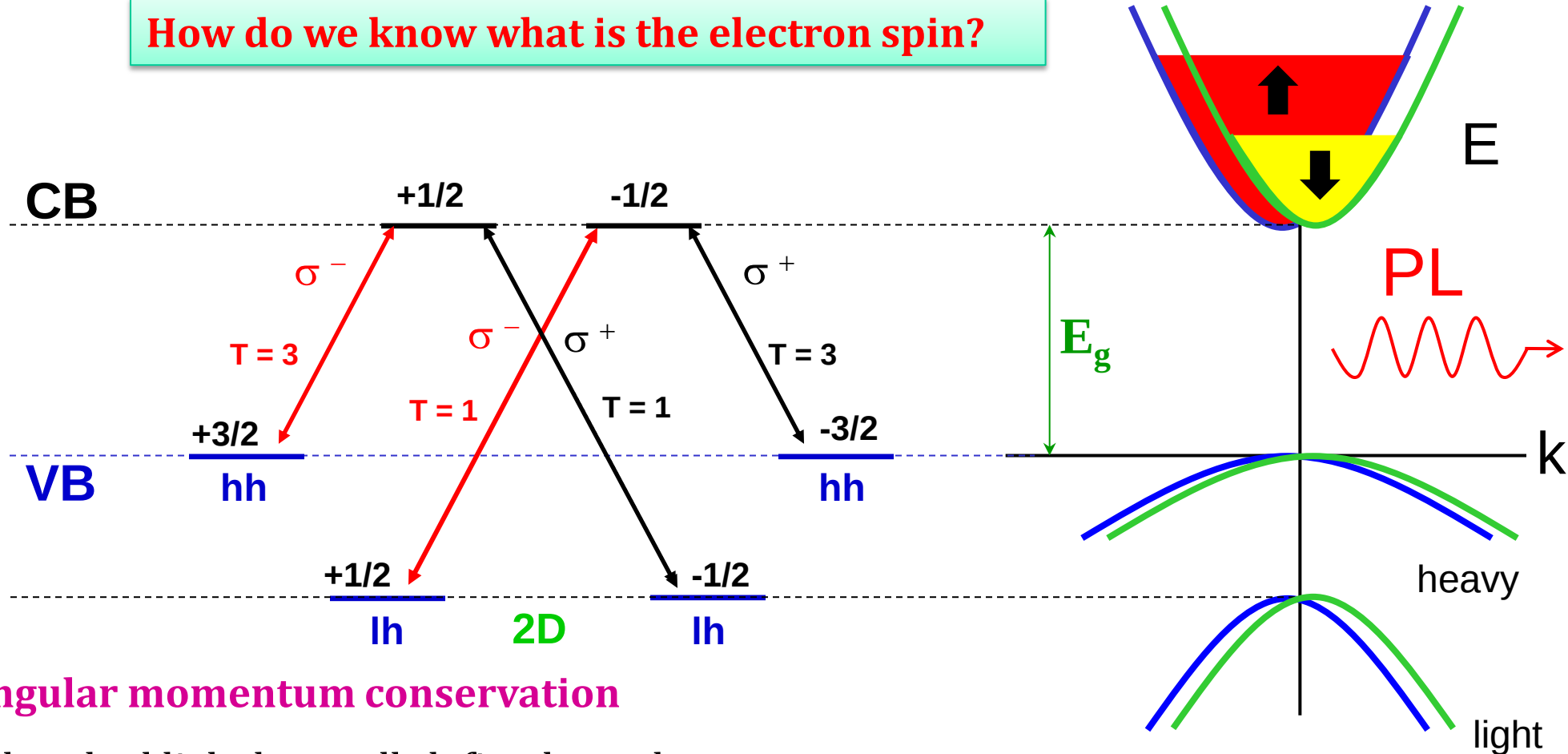
- Resolução temporal ($\sim 2\text{ps}$)

Ótica de polarização

- Elicidade da luz (dinâmica de spins)

Optical orientation

How do we know what is the electron spin?



Angular momentum conservation

- Absorbed light has well-defined angular momentum σ
- $N(s = 1/2) > N(s = -1/2)$ in the CB

✓ PL is polarized

$$\rho_z = \frac{I_+ - I_-}{I_+ + I_-}$$

Time scales

After relaxation to the bottom of CB

Electron dynamics

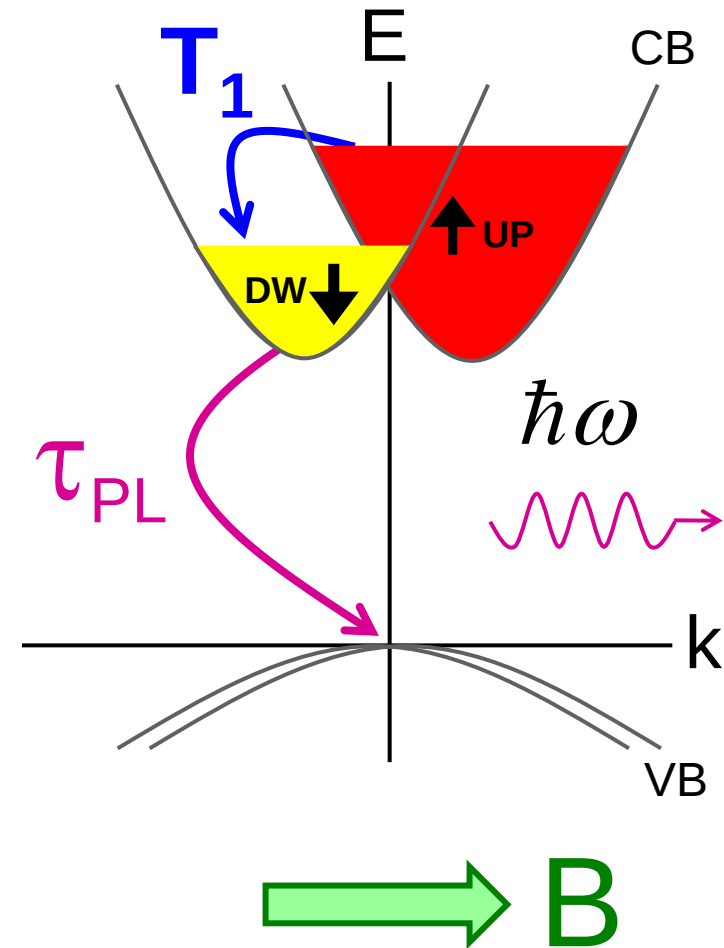
- ✓ Carrier lifetime $\rightarrow \tau_{PL}$
- ✓ Spin relaxation time $\rightarrow T_1$

If there is a transverse magnetic field

- ✓ Spin decoherence time $\rightarrow T_2 > T_2^*$

Spin lifetime

$$\frac{1}{\tau_s} = \frac{1}{T_1} + \frac{1}{T_2^*}$$



Challenges

Under equilibrium conditions

- ✓ Free carriers and spins average to zero (unlike Ferromagnetic metals!)

Out of equilibrium $\rightarrow \rho_c, \rho_s \neq 0$

Strong interaction with the environment

- ✓ Other atoms (crystal lattice)
- ✓ Impurities
- ✓ Temperature (phonons)



Relaxation

For semiconductor QWs

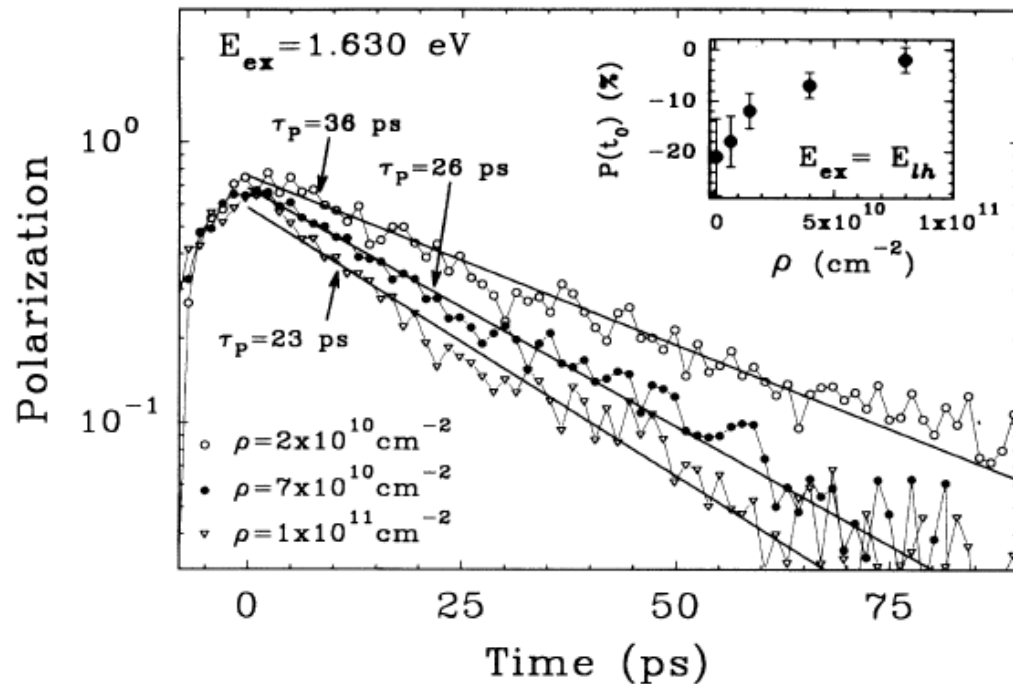
$$\tau_{PL} \rightarrow 1 \text{ ns}$$

$$T_1 \leq \tau_{PL}$$

$$T_2 \leq T_1$$

But it depends on the system!

Munhoz et. al., PRB 51, 4247 (1995)

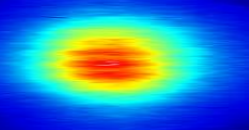


Information process

- Understand carrier/spin dynamics
 - Enhance lifetimes
 - Manipulate particle number/charge/spin
 - Tune energy states
 - Increase temperature ???

Alguns sistemas investigados no Grupo de Propriedades Óticas (GPO)

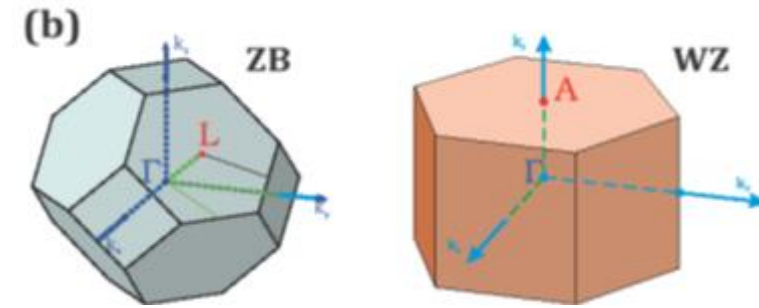
Nanofios semicondutores



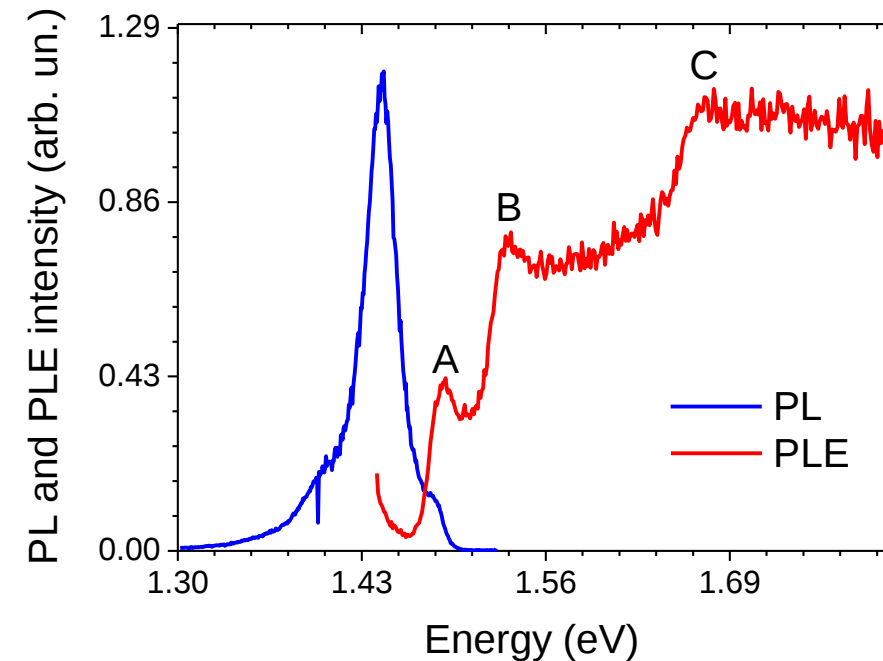
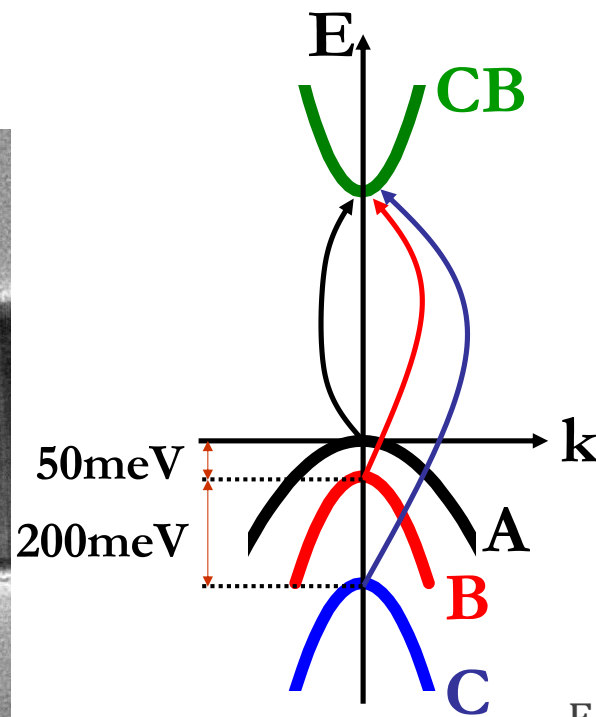
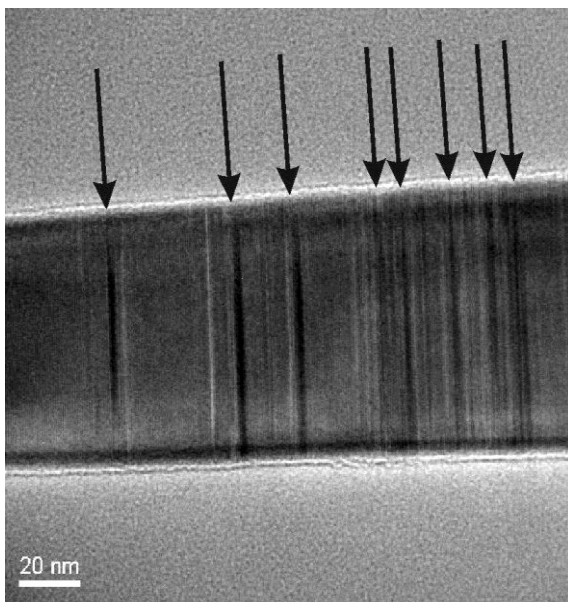
- ✓ Novo tipo de estrutura
- ✓ Novas propriedades estruturais e cristalinas
- ✓ Possíveis aplicações (emissores de luz, sistemas fotovoltaicos)

Nanofios de InP (colaboração Profa. M. A. Cotta)

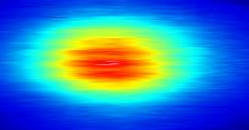
- ✓ Estrutura cristalina → **Wurtzita**



TEM Nanofio de InP

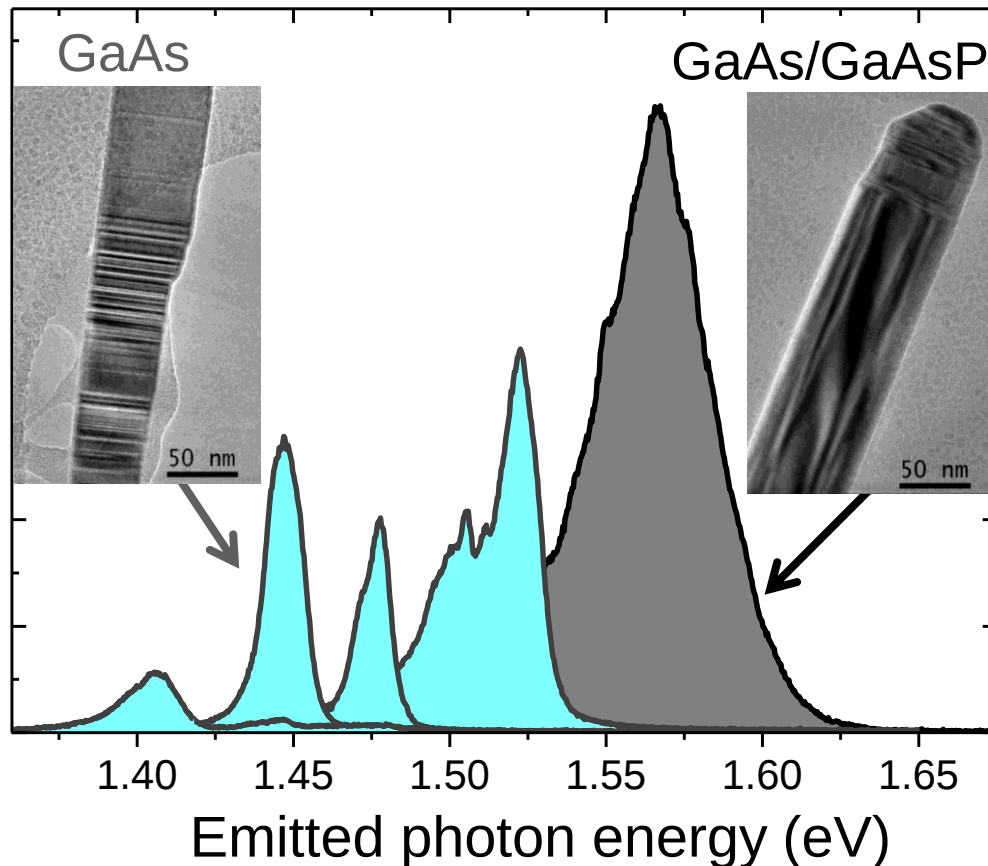


Nanofios semicondutores

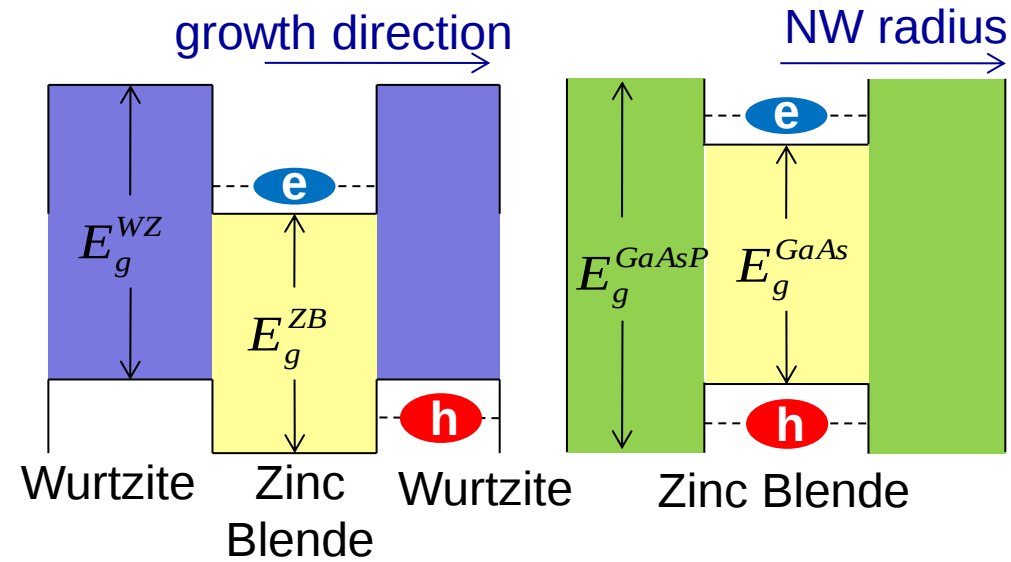
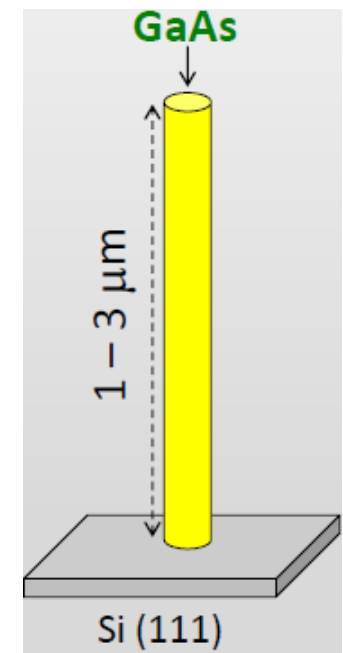
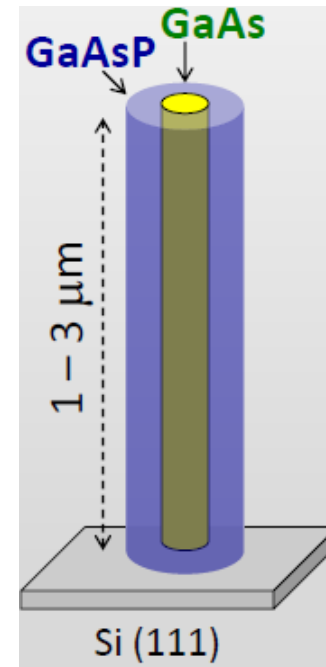


Nanofios de GaAs/AlGaAs

- ✓ Core/shell
- ✓ Alta emissão e baixo nível de defeitos de superfície



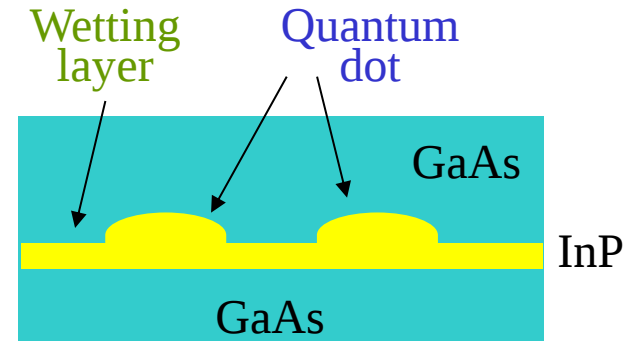
Couto Jr. et. al, *Nano Lett.*, **12**, 5269 (2012)



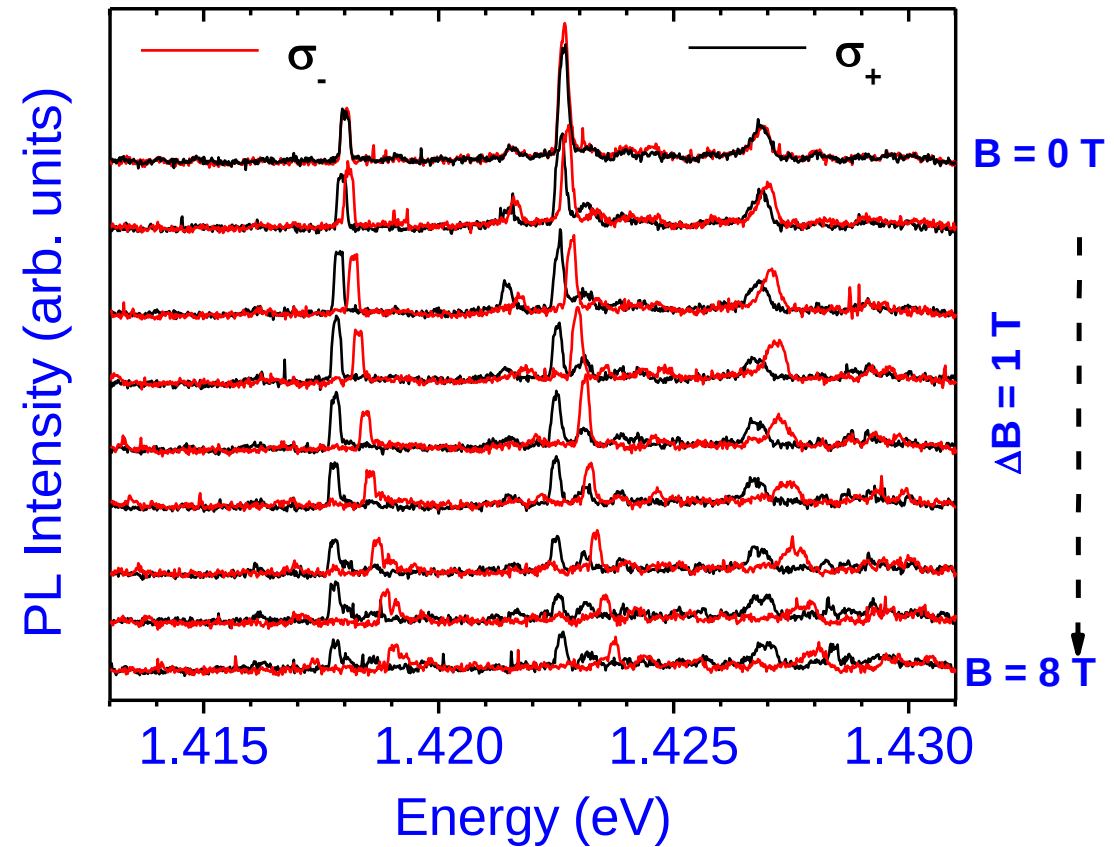
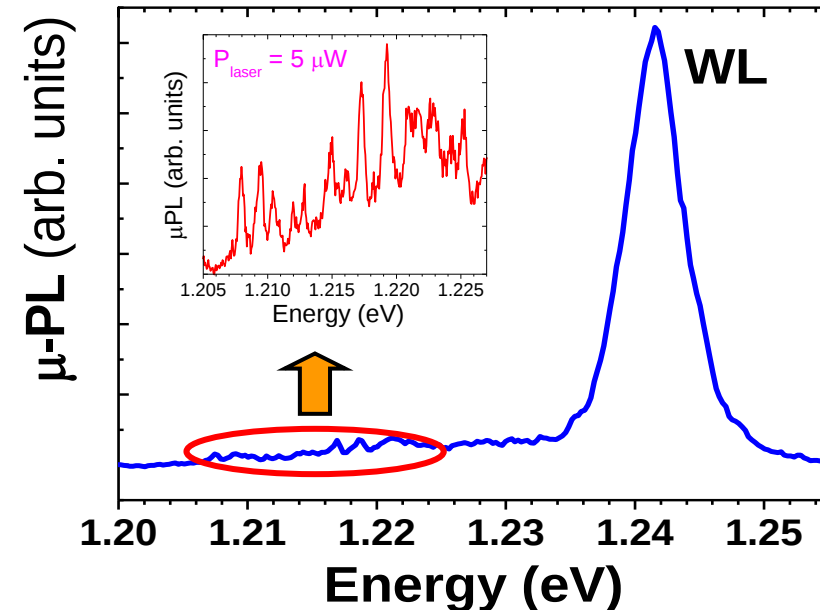
Pontos quânticos

InP/GaAs

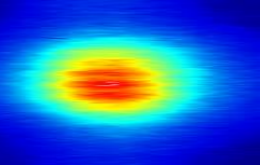
- ✓ Estudo de pontos quânticos individuais
- ✓ Medidas μ -PL com campo magnéticos DC
- ✓ Alinhamento tipo II
- ✓ Estudo da interação elétron-buraco



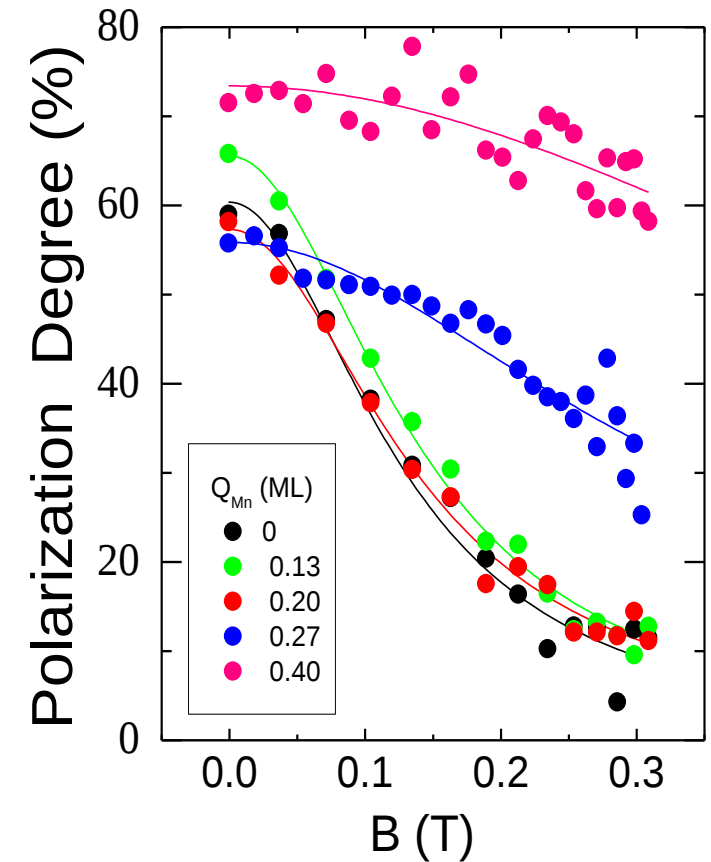
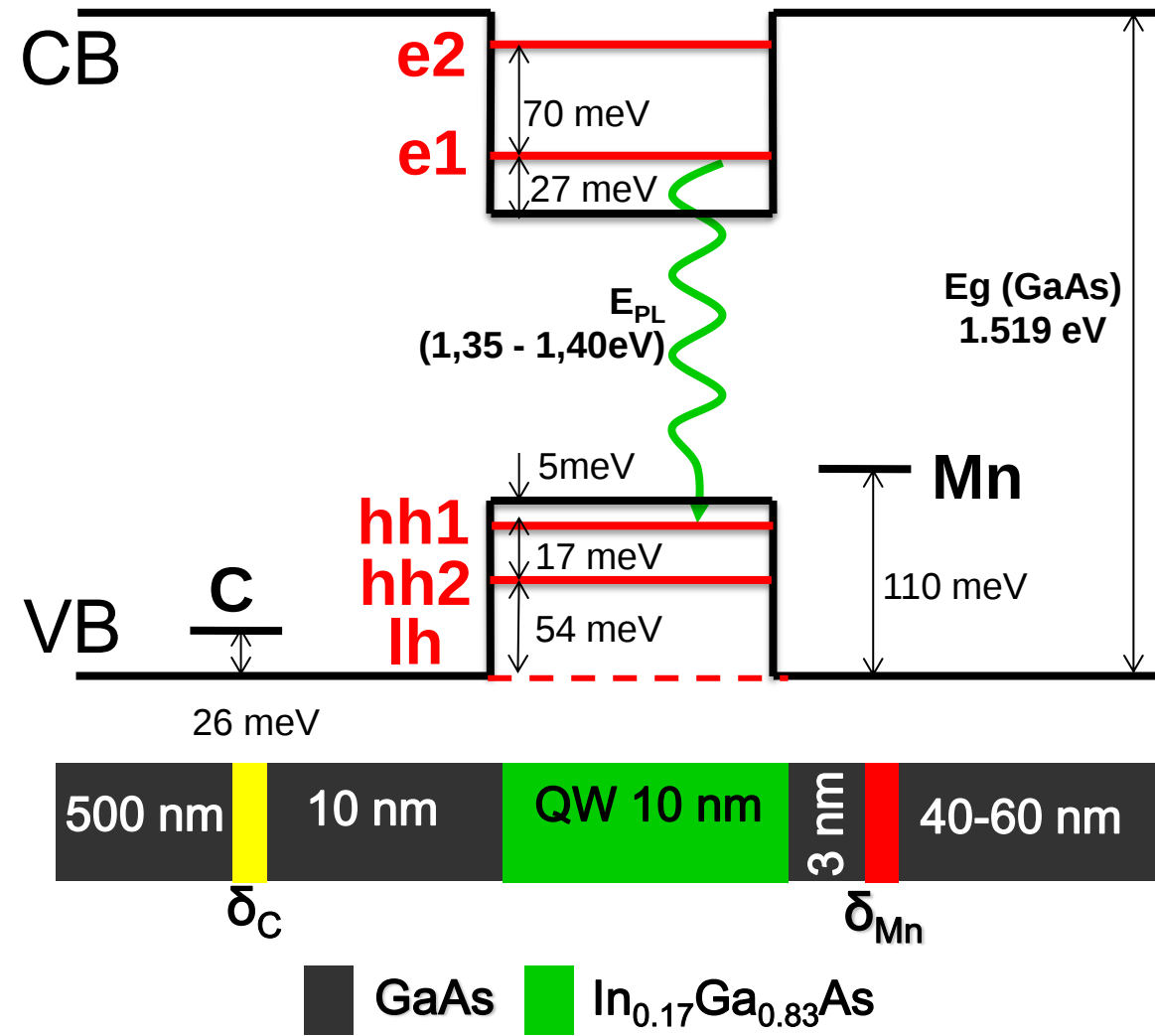
$$E_{\pm} = E_0 \pm \frac{1}{2}g_x\mu_B B + \alpha_d B^2$$



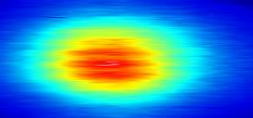
Semicondutores magnéticos



- ✓ Implantação de Mn em GaAs
- ✓ Interação com poço quântico de InGaAs
- ✓ Diminuição no tempo de vida de spins



Nanotubos de carbono

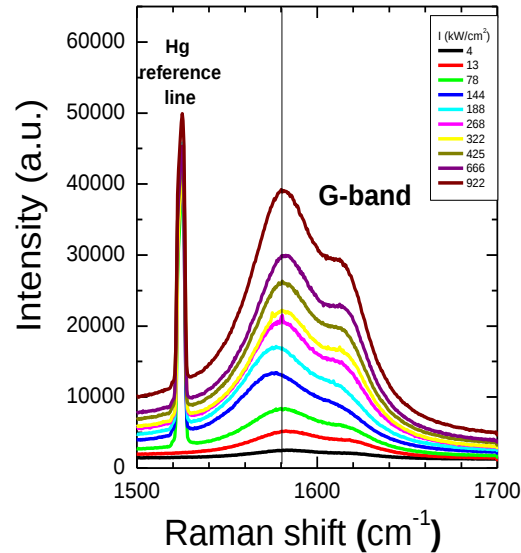


Espalhamento Raman

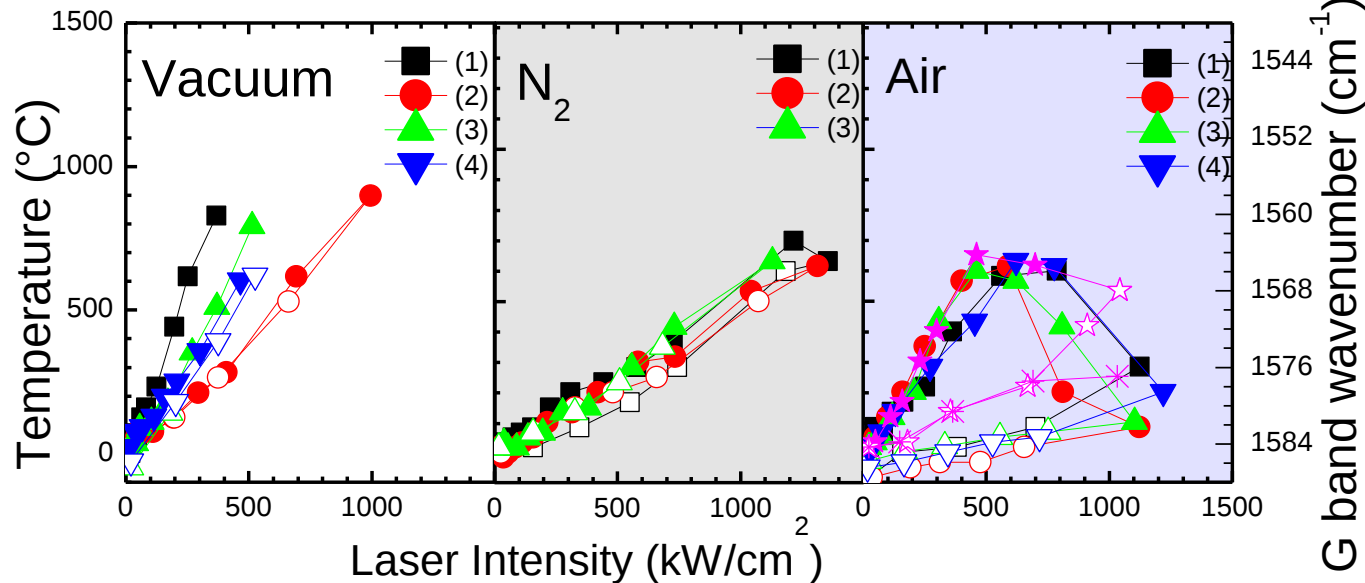
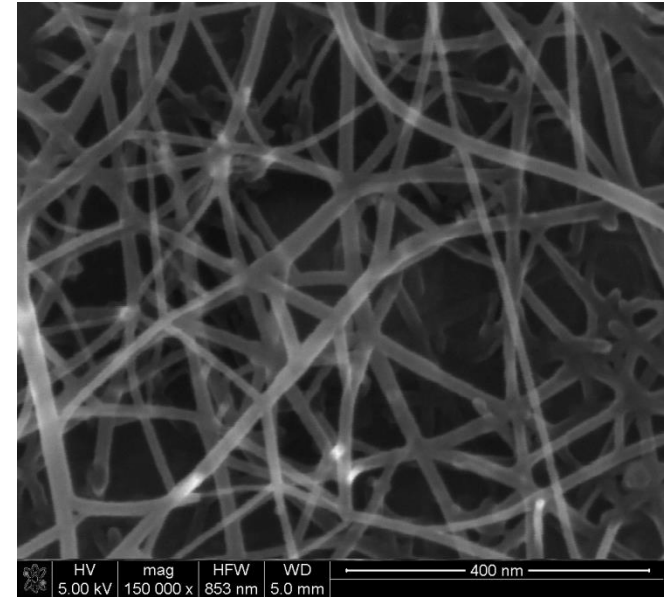
Nanotubos

- ✓ Single Wall
- ✓ Multi Wall (MWCNTs)

- Estudo das propriedades térmicas sob diferentes atmosferas

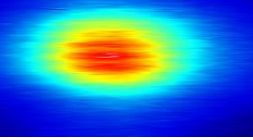


MWCNTs



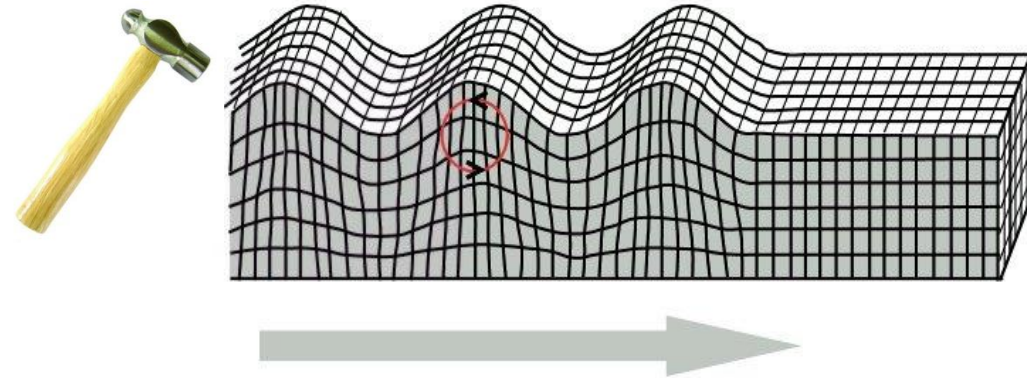
Ondas acústicas de superficie (SAWs)

SAW

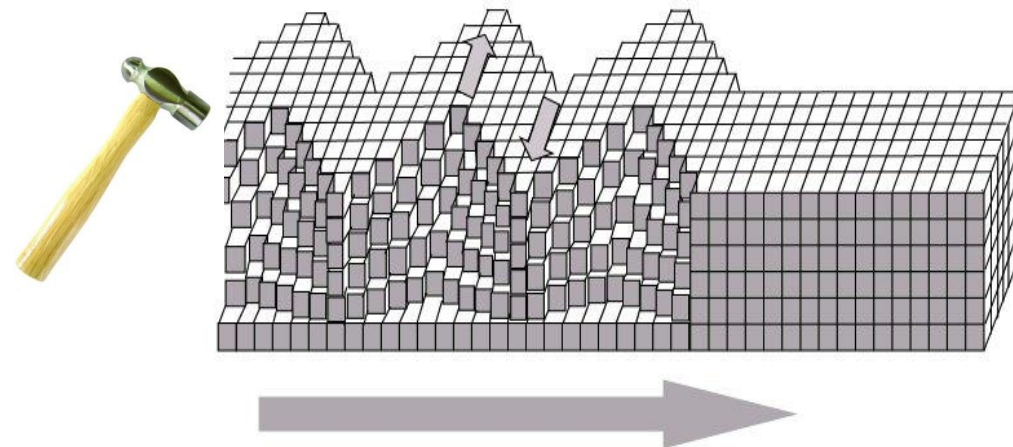


Elastic wave propagating on the surface of solids

Rayleigh Wave



Love Wave



Applications:

- Mobile, Wireless communication
 - Sensors, filters, resonators....

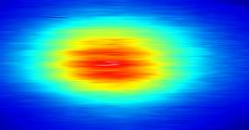
Estimate

- 3 million SAW devices are manufactured **every day!!!**

SAW Touch Screen Technology



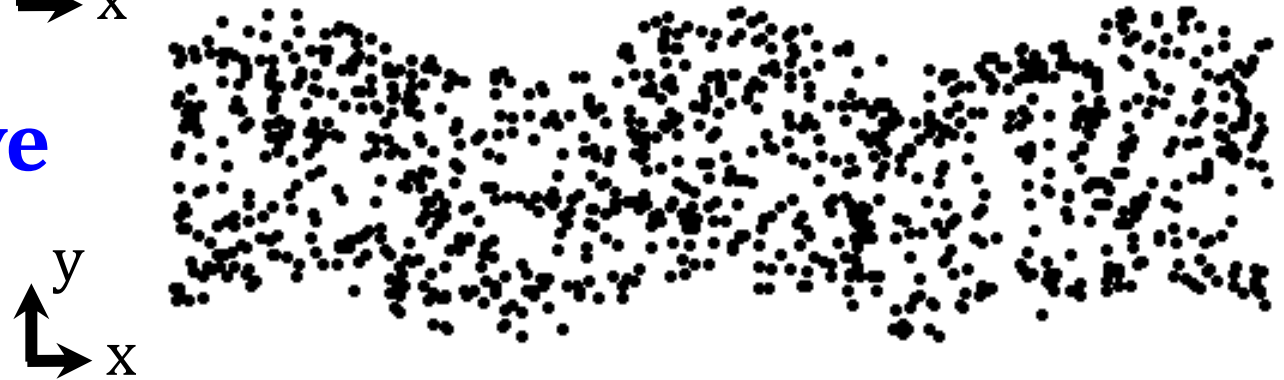
SAW modes



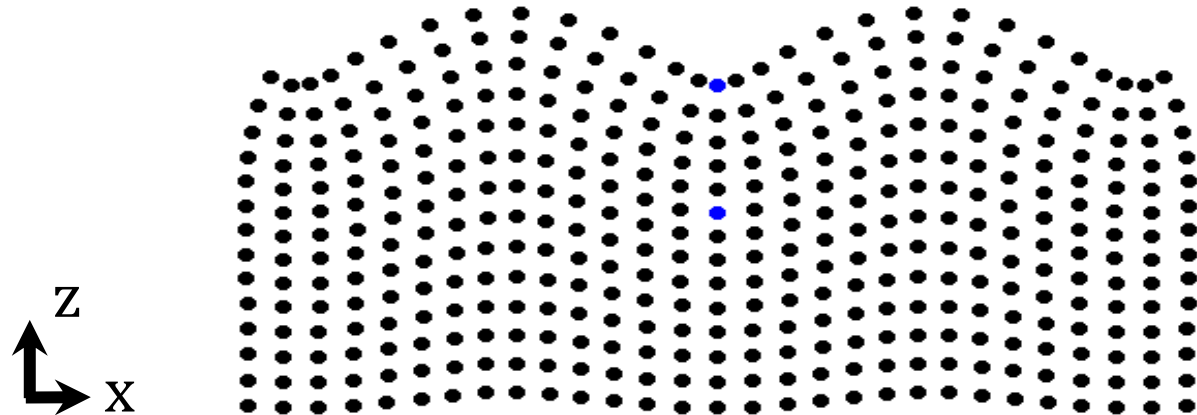
Longitudinal wave



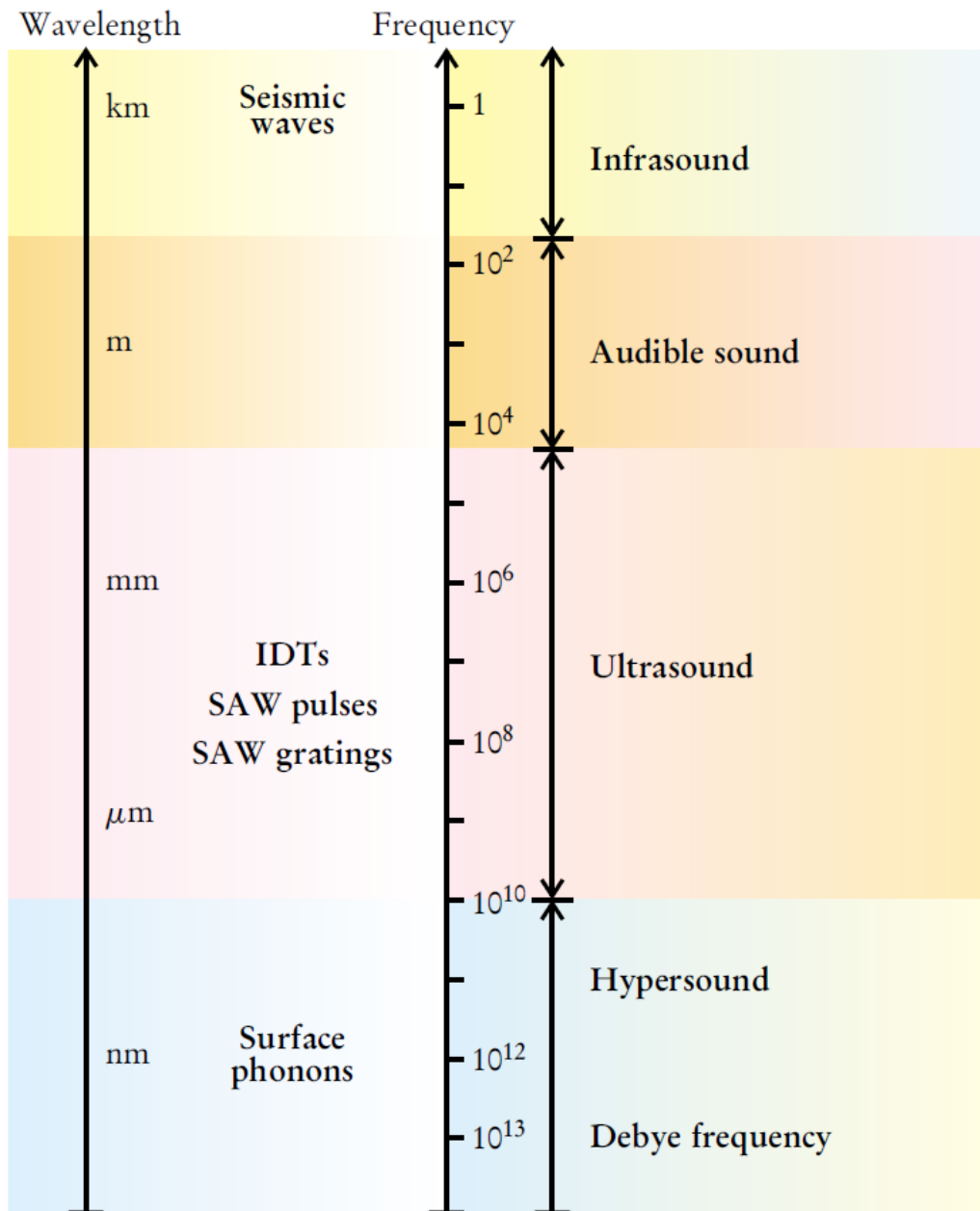
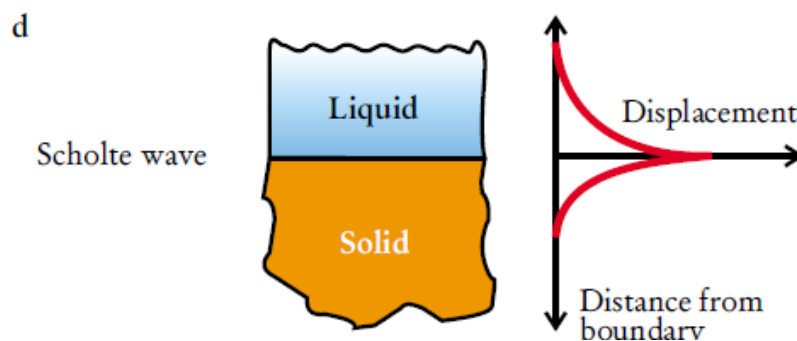
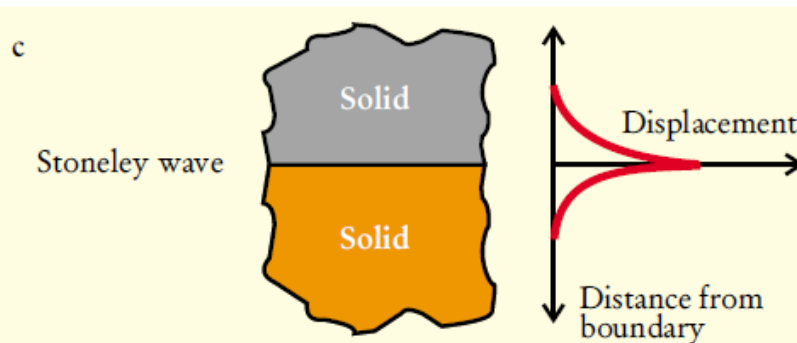
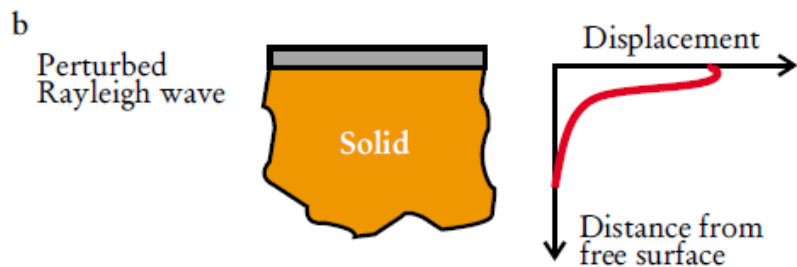
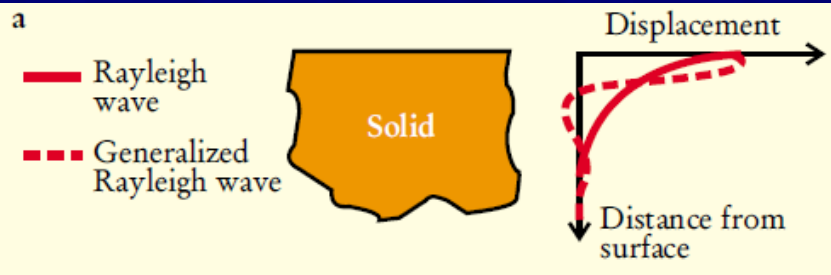
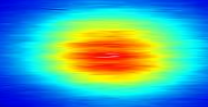
Transverse wave



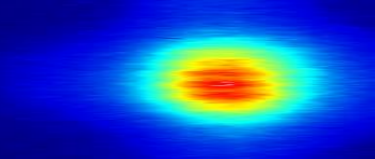
Raleigh wave



SAW



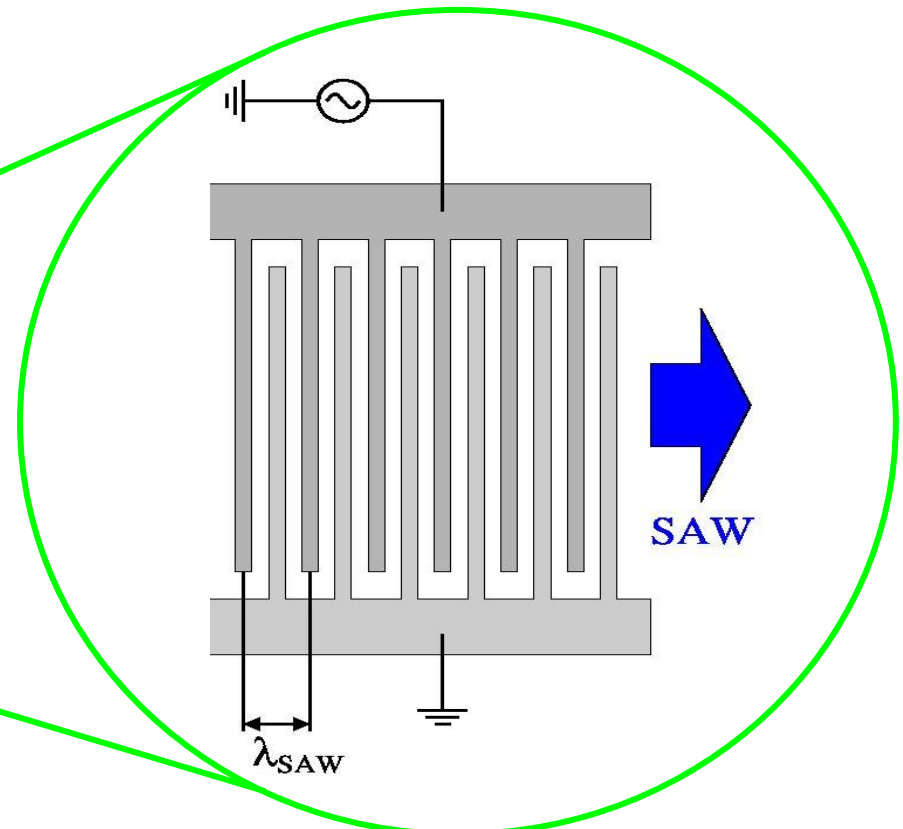
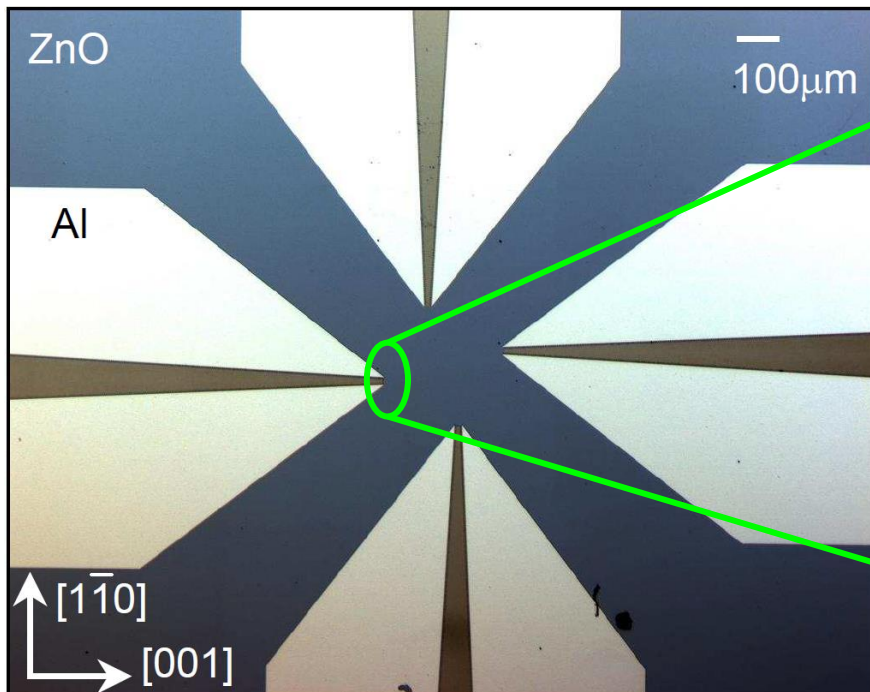
How to generate SAWs



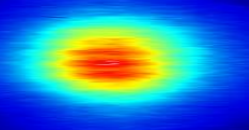
Photolithography

- Thin metal (Al) layer on top
- Interdigital transducers (IDTs)
- Defines the acoustic wavelength λ_{SAW}

Poço Quântico ←



How to generate SAWs



Piezoelectric substrate

- LiNbO_3 , ZnO
- GaAs (in our case)

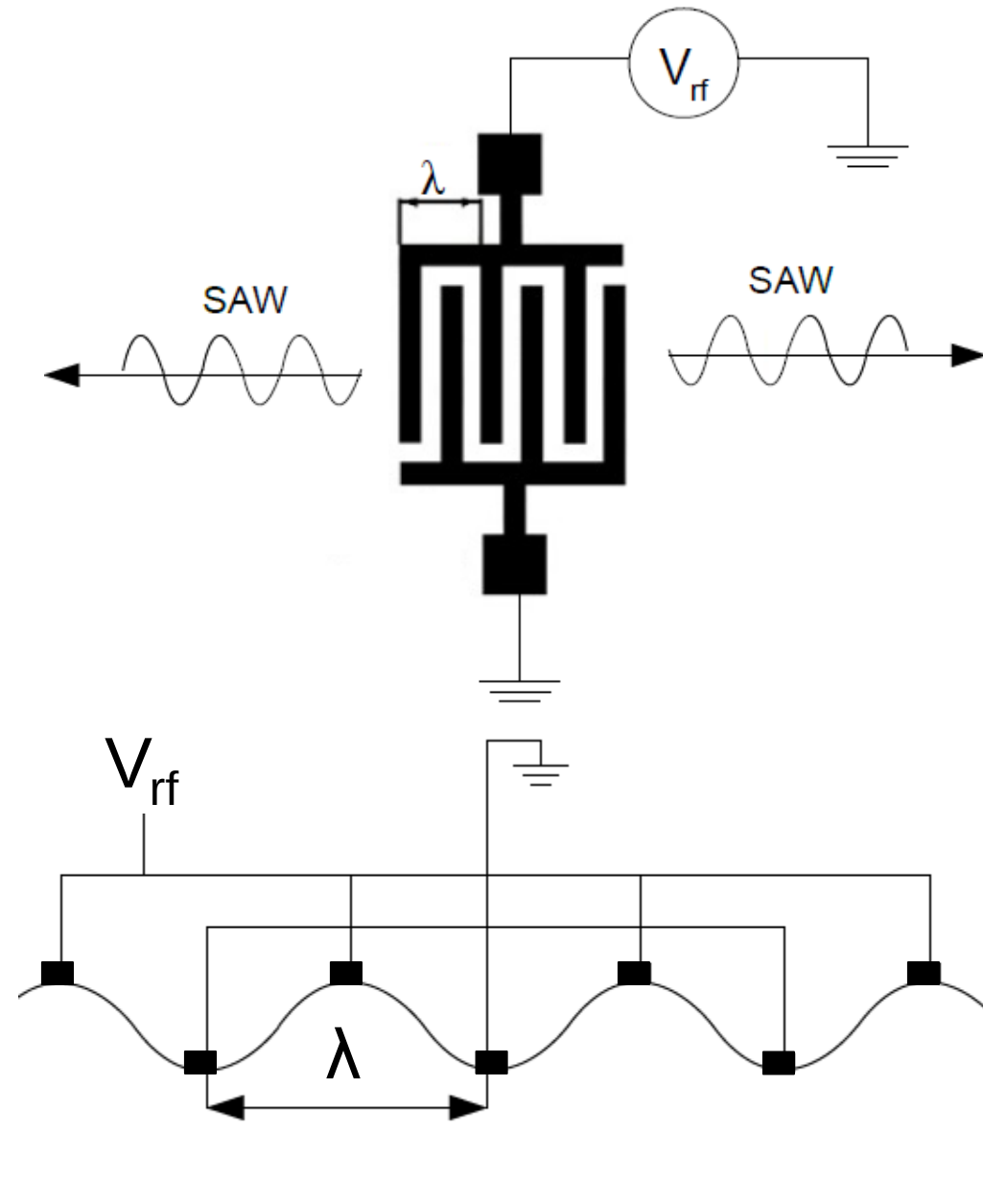
rf-signal

- Efeito piezoelétrico inverso
- Feixe de SAW é lançado

Linear dispersion: $\Rightarrow \omega_{\text{SAW}} \times k_{\text{SAW}}$

Well-defined velocity $f_{\text{SAW}} = \frac{v_{\text{SAW}}}{\lambda_{\text{SAW}}}$

In GaAs $\rightarrow v_{\text{SAW}} \approx 3000 \text{ m/s}$
 $f_{\text{SAW}} \approx \text{MHz} - \text{GHz}$



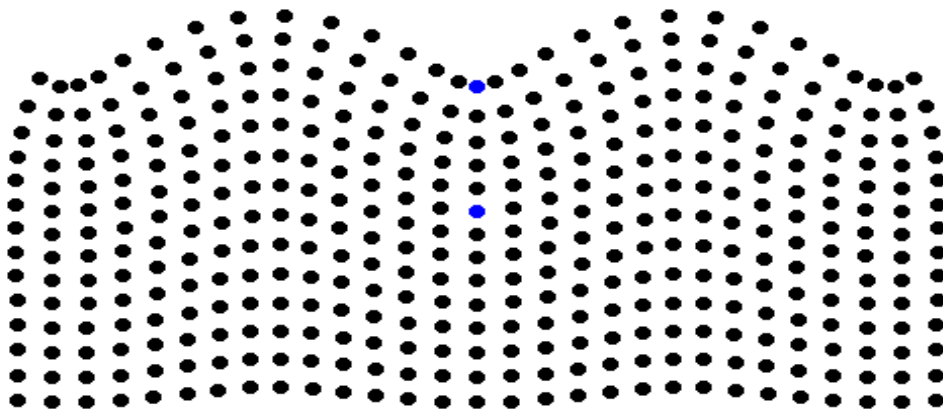
Porque SAWs?

Em ciência de materiais (tipo bulk ou nano)

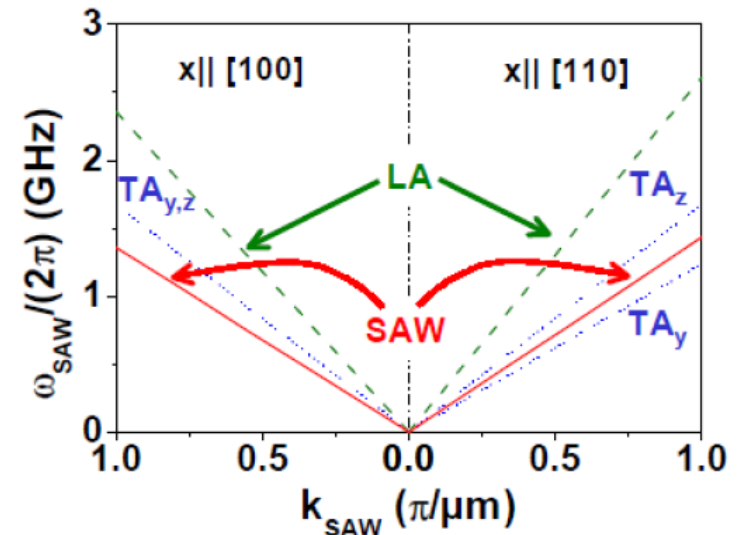
- Novo material → Caracterização (óptica, estrutural, elétrica....)
- Campos externos (elétricos, magnéticos, tensão)
- Aplicações (dispositivos)
- Indústria

Para nanossistemas

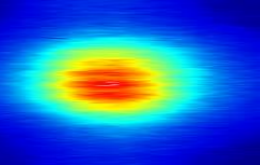
- Método eficaz, local e não destrutivo de aplicação de campos externos
 - SAWs
 - Campo piezoelétrico e de tensão
 - Frequência e velocidade de propagação bem definidas



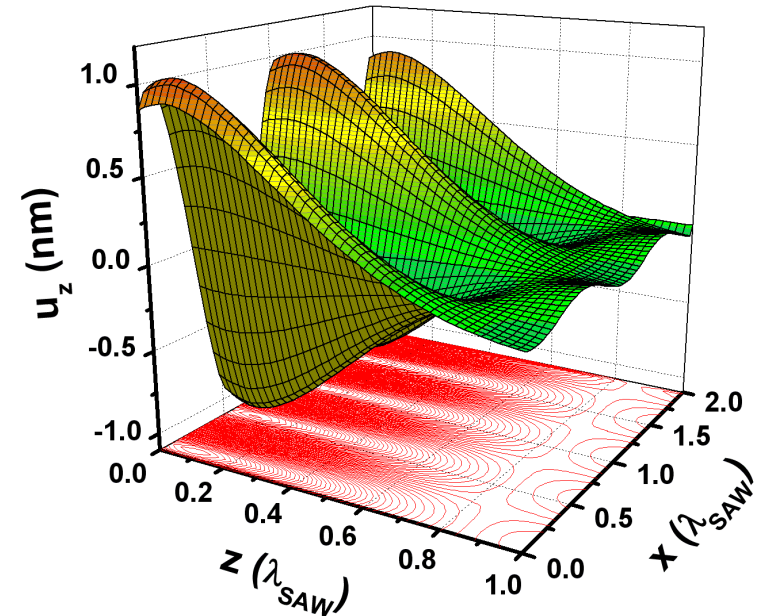
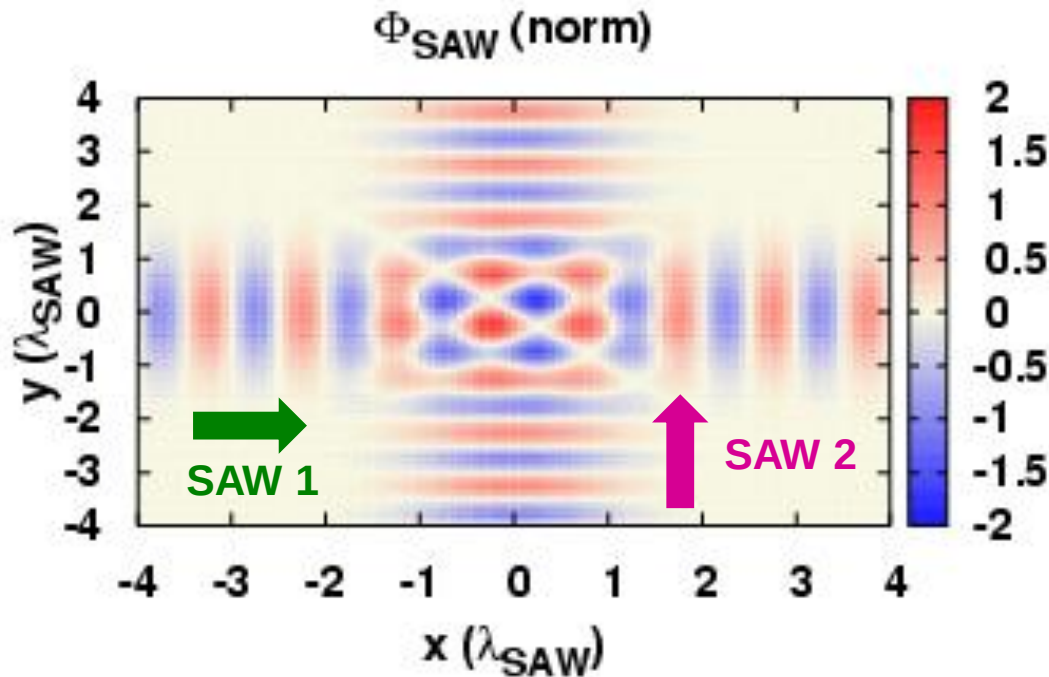
©1999, Daniel A. Russell



Acoustic modulation



Particle displacement (wave amplitude) decays exponentially below the surface



Acoustic Modulation

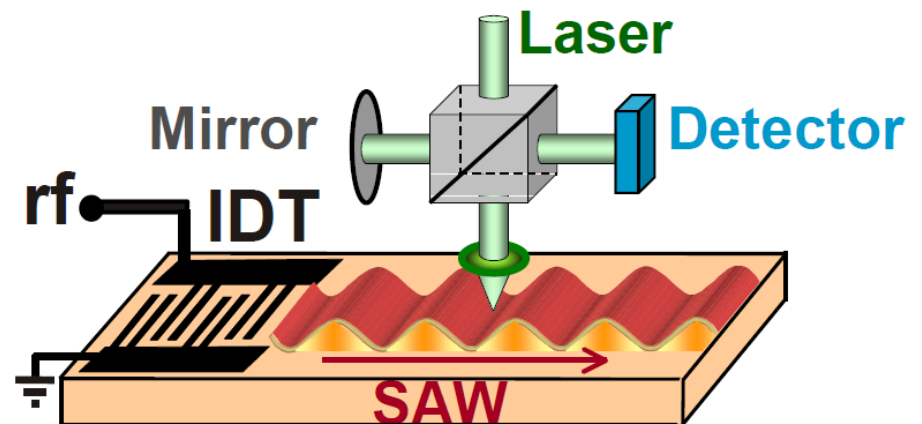
SAWs in Solid-state systems

On the surface

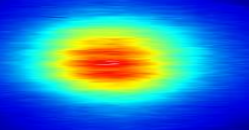
Waveguides, quantum wires

Few nanometers below the surface

Quantum wells, quantum dots



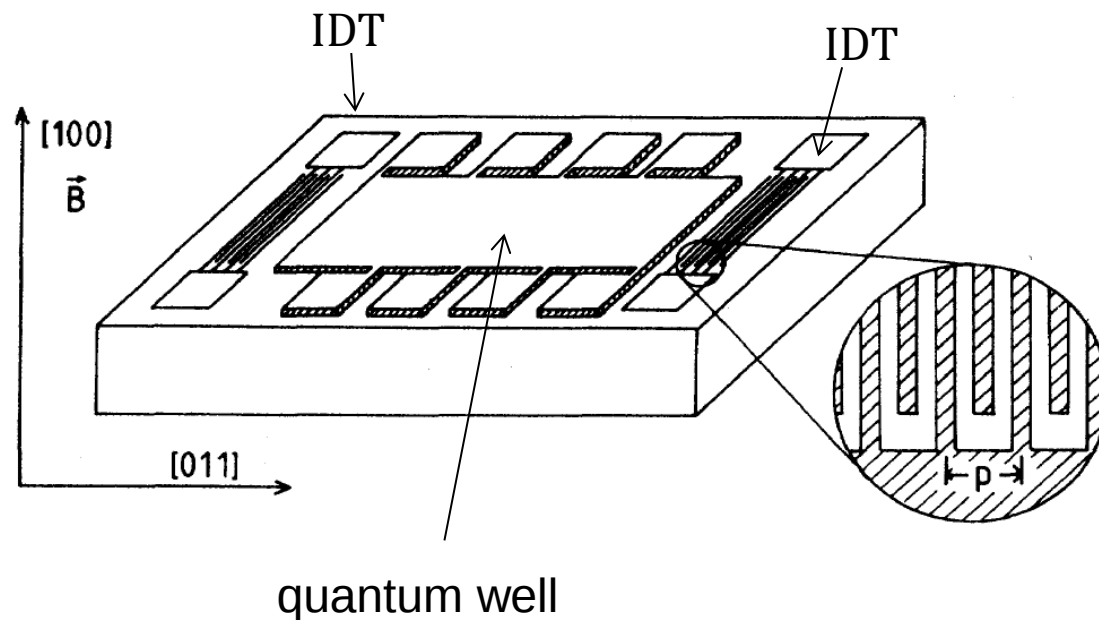
First experiments



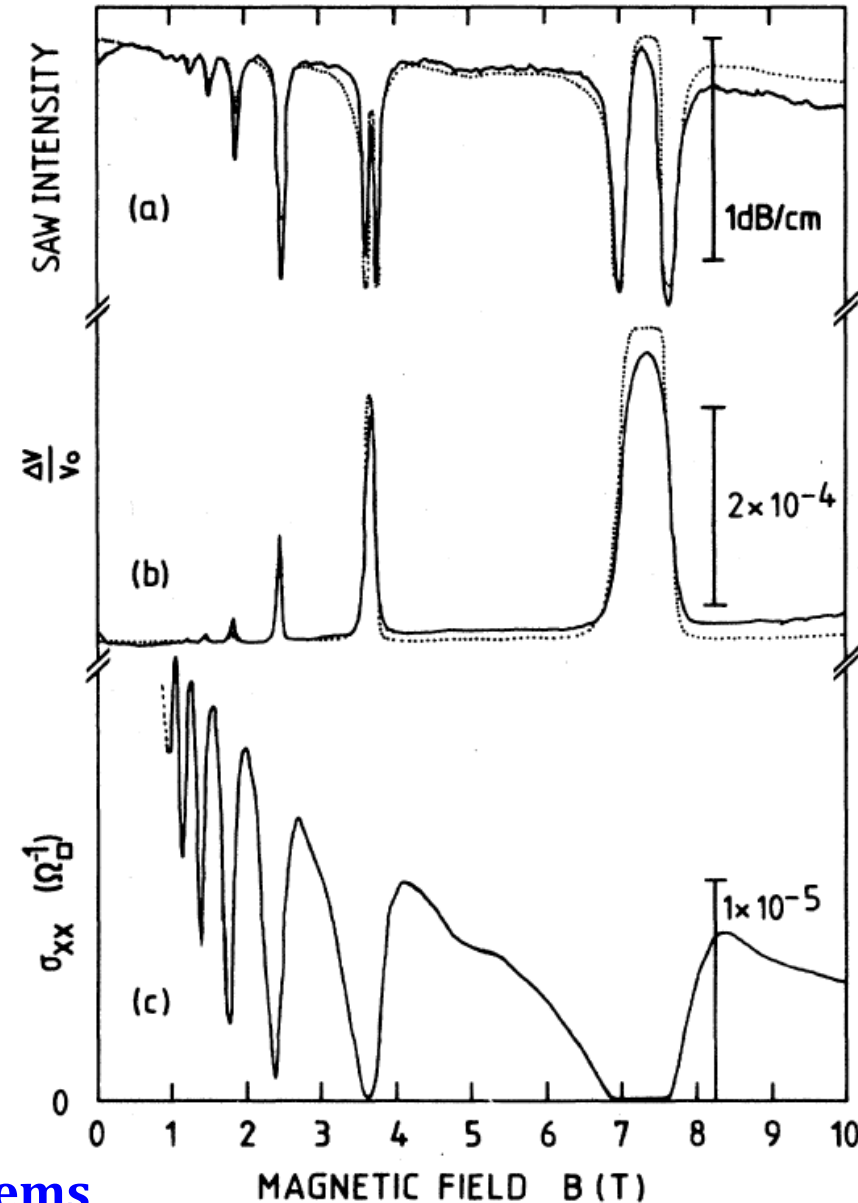
2D electron gases under high magnetic fields

✓ Alternative tool to study the Quantum Hall effect

Wixforth, Phys. Rev. Lett. 40, 7874 (1989)



Untill the end of the 90's this was the major application of SAWs in the studies solid state systems



Other solid state systems

Control of elementary excitations

Carriers

- ✓Rocke *et. al Phys. Rev. Lett.*, 78, 4099
- ✓M. M.de Lima *et. al. Appl. Phys. Lett.* **84**, 2569

Spins

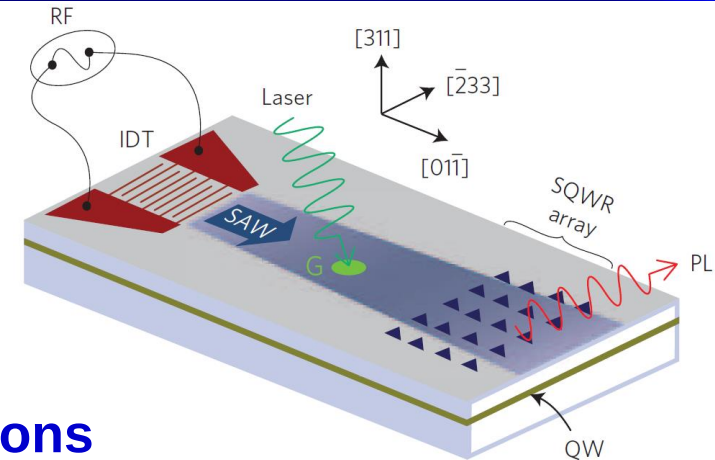
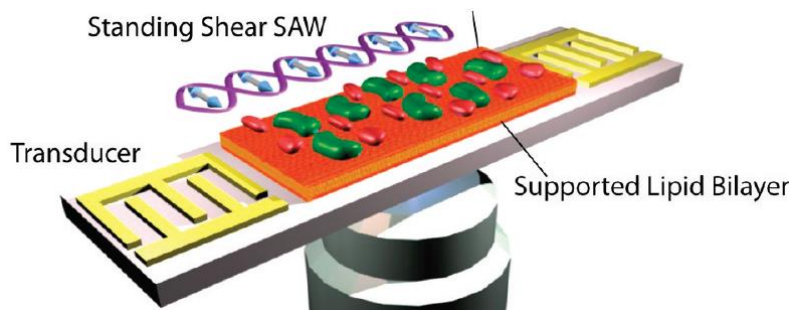
- ✓T. Sogawa *et. al. Phys. Rev. Lett.* **87**, 276601
- ✓J. A. Stotz *et. al. Nature Mat.* **4**, 585
- ✓O. D. D. Couto Jr. *et. al. Phys. Rev. Lett.* **98**, 036603

Excitons

- ✓J. Rudolph *et. al. Phys. Rev. Lett.* **99**, 047602

Bose-Einstein Condensates

- ✓M. M. de Lima *et. al. Phys. Rev. Lett.* **97**, 045501
- ✓E. Cerda-Méndez *et. al. Phys. Rev. Lett.* **105** 116402



Photons

Mach-Zehnder interferometer

- ✓M. M. de Lima *et. al. Appl. Phys. Lett.* **89**, 121104

Single photon sources

- ✓O. D. D. Couto Jr. *et. al. Nature Photon.* **3**, 645
- ✓A. Hernandez-Minguez *et. al. Nanolett.* **12**, 252

Photonic crystal nanocavities modulation

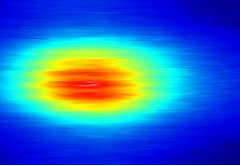
- ✓D. A. Fuhrmann *et. al. Nature Photon.* **5**, 605

Biological systems (new trends)

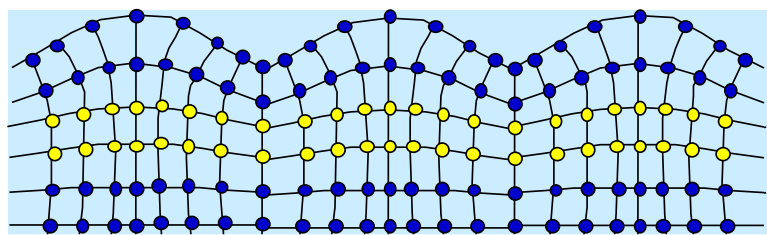
DNA dynamics

- M. Hennig *et. al. Langmuir* **27**, 14721
- J. Neumann *et. al. Nano Lett.* **10**, 2903

Acoustically induced transport

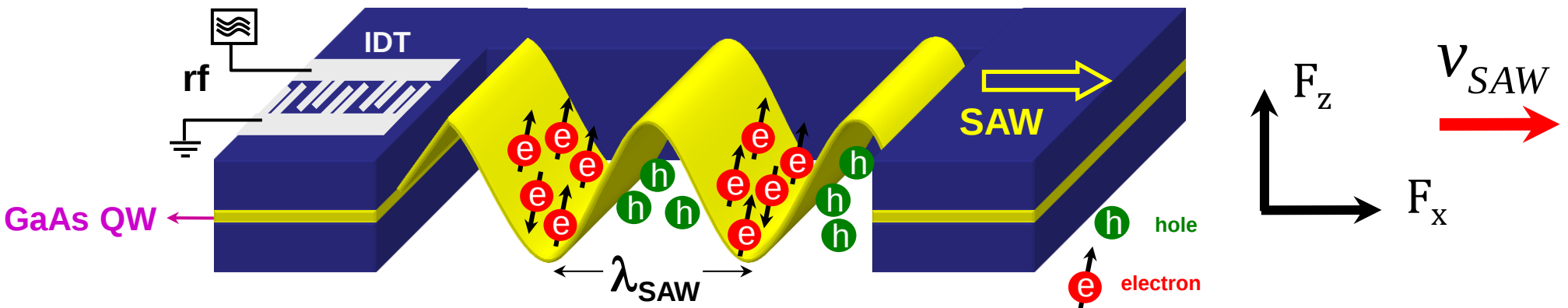


Strain field (S)



Piezoelectric field (E)

- F_x drags carriers along SAW propagation
- F_z modulates confinement potential
 - Electrons and holes confined in the minima of moving potential
 - Longer carrier lifetimes (τ_{PL})



An electron is created at time t_0 , is it possible to know where it will be at $t > t_0$?

Carriers are transported by the SAW with a well-defined velocity

$$m^* v_{SAW} = \hbar \langle k \rangle$$

If $t < \tau_s$: information about the spin state of the particle!!!

Experiments

Semiconductor quantum well GaAs (QW)

Generation (excitation)

- Circularly polarized pulsed laser : σ^-
 - Spins polarized along z

Transport

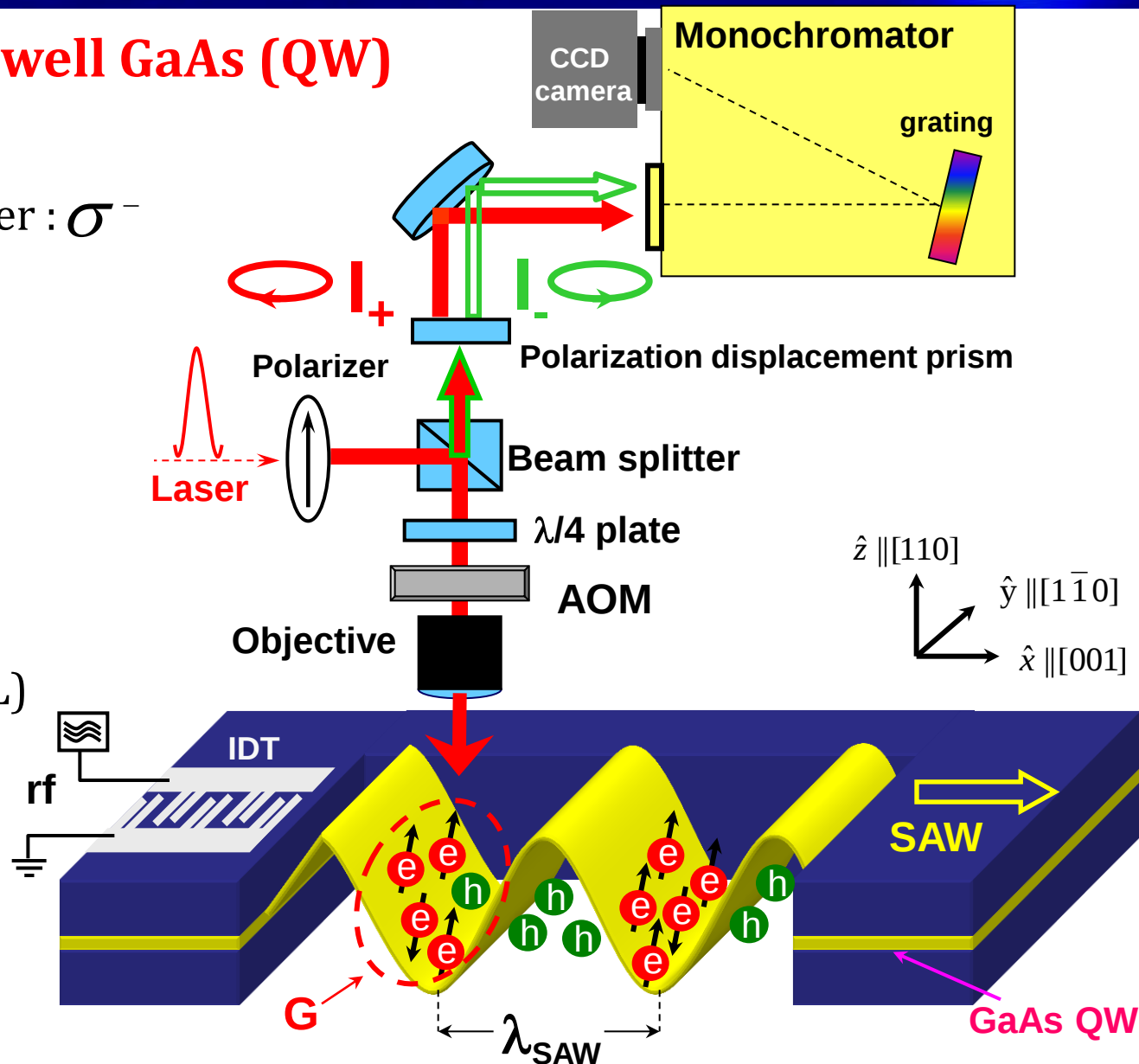
- SAW piezoelectric field (ϕ_{SAW})

Detection

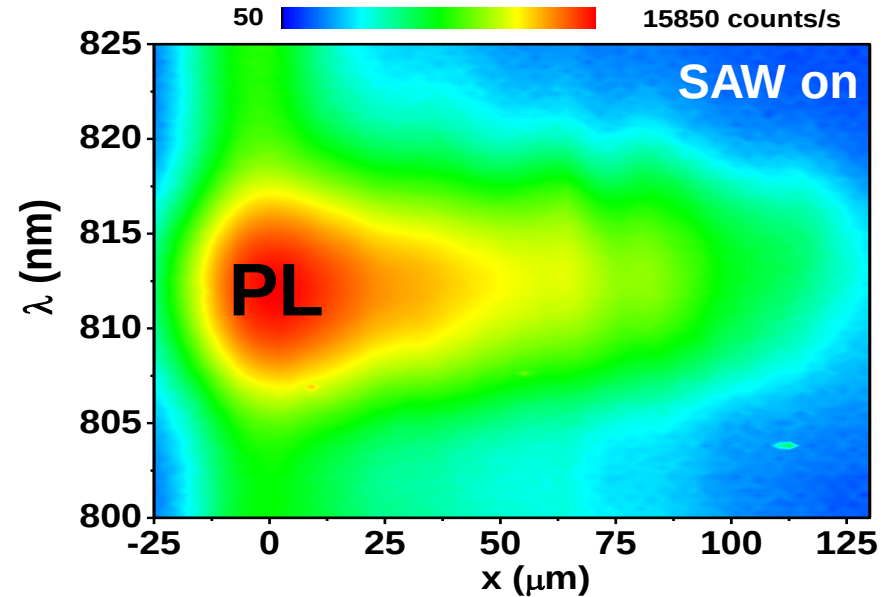
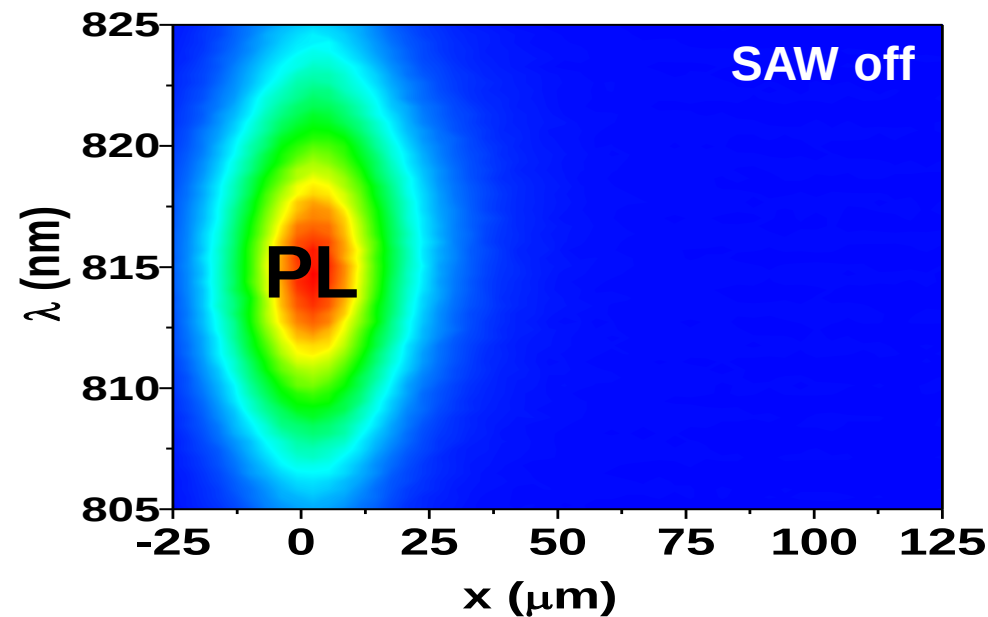
- Time, space, and polarization resolved photoluminescence (PL)

Spin polarization degree

$$\rho_z = \frac{I_+ - I_-}{I_+ + I_-}$$



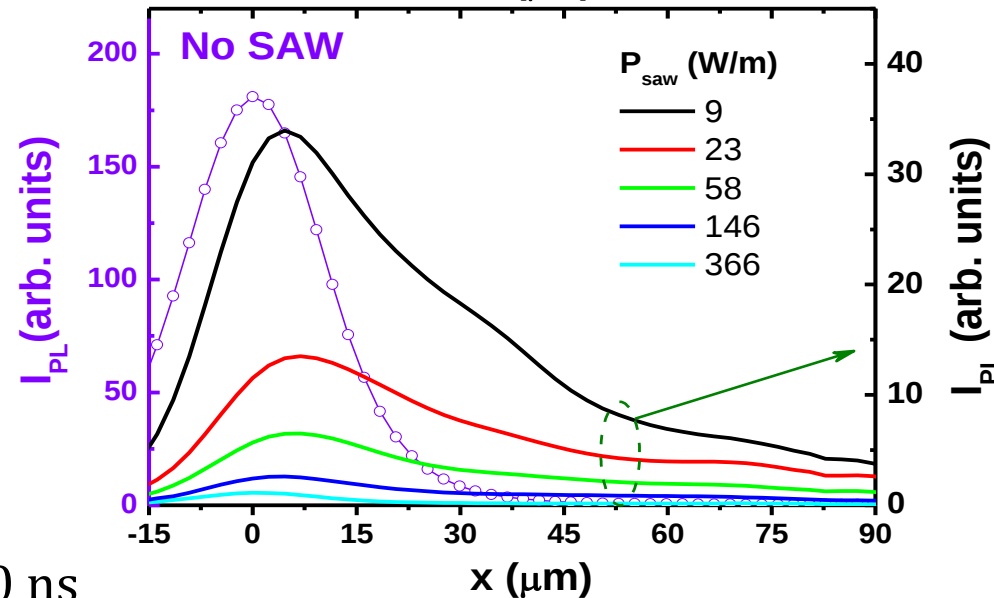
Carrier transport



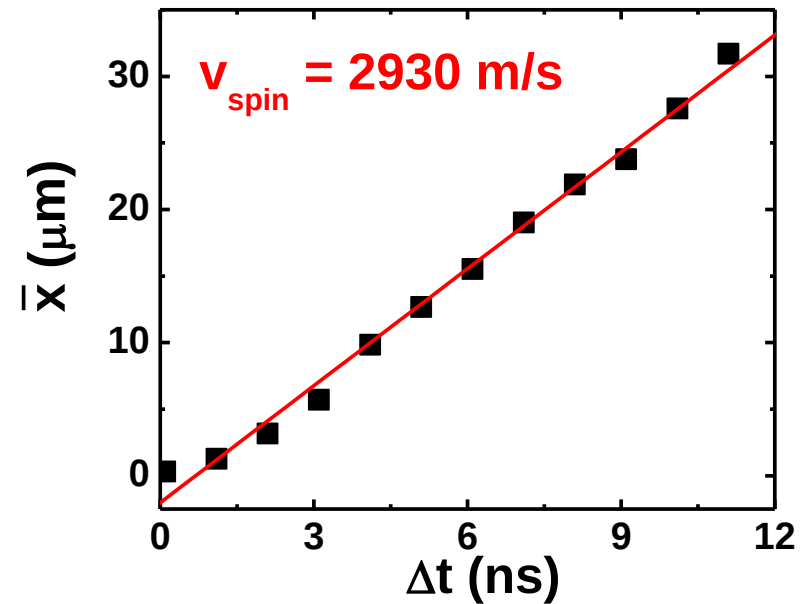
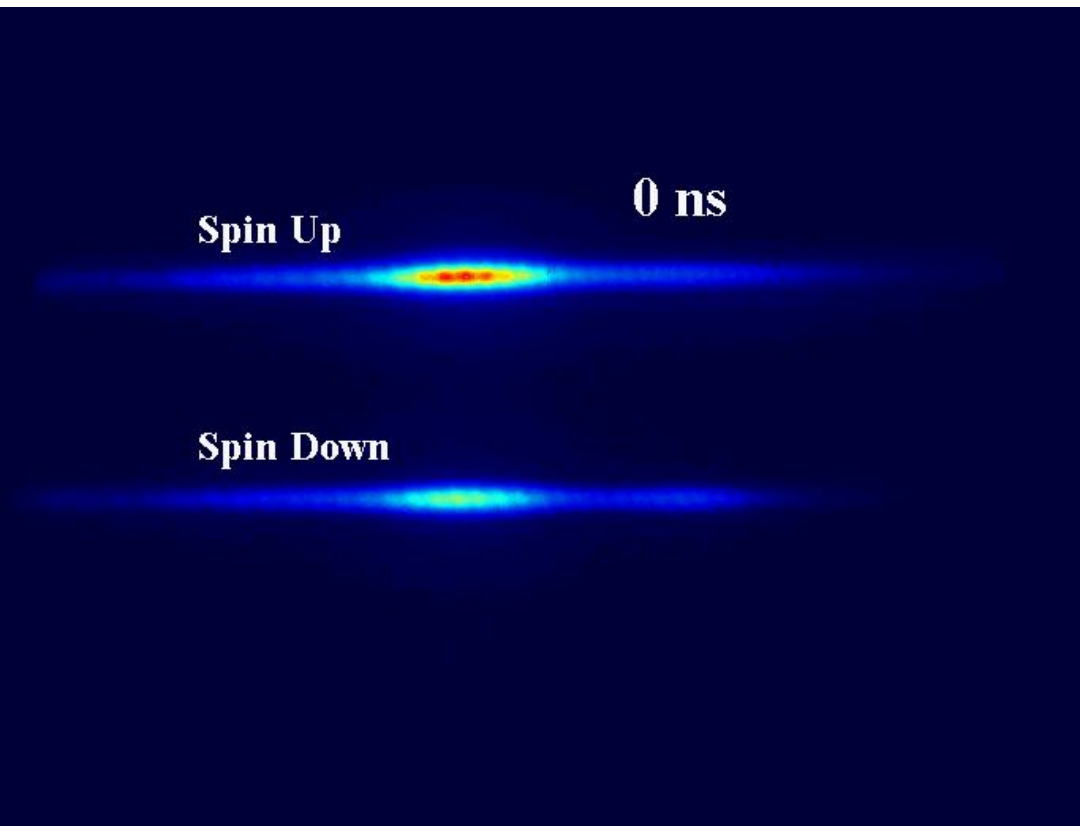
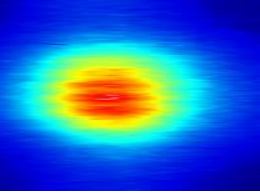
Continuous PL detection

Carrier transport

- ✓ SAW off
 - ✓ Carrier diffusion $\rightarrow \tau_{\text{PL}} \sim 1$ ns
- ✓ SAW on
 - ✓ PL quenching ~ 90 times for high acoustic powers
 - ✓ Efficient carrier transport $\rightarrow \tau_{\text{PL}} > 50$ ns



Time-resolved detection

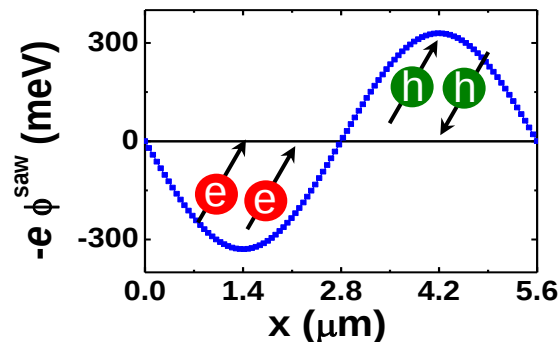


Coherent carrier transport

✓ Well-defined carrier packets

✓ $v_{\text{spin}} = v_{\text{SAW}}$

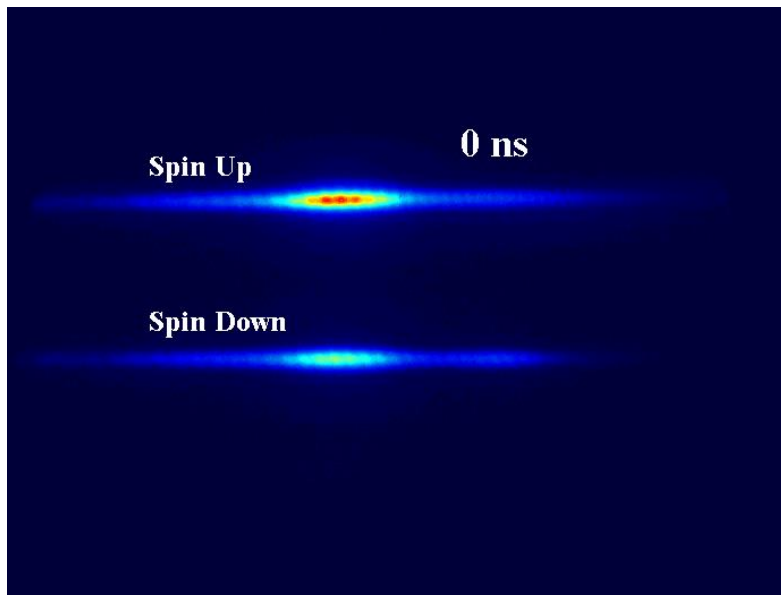
SAW



Spin transport (110) QWs

Time-resolved PL detection

- Electron-heavy hole transition

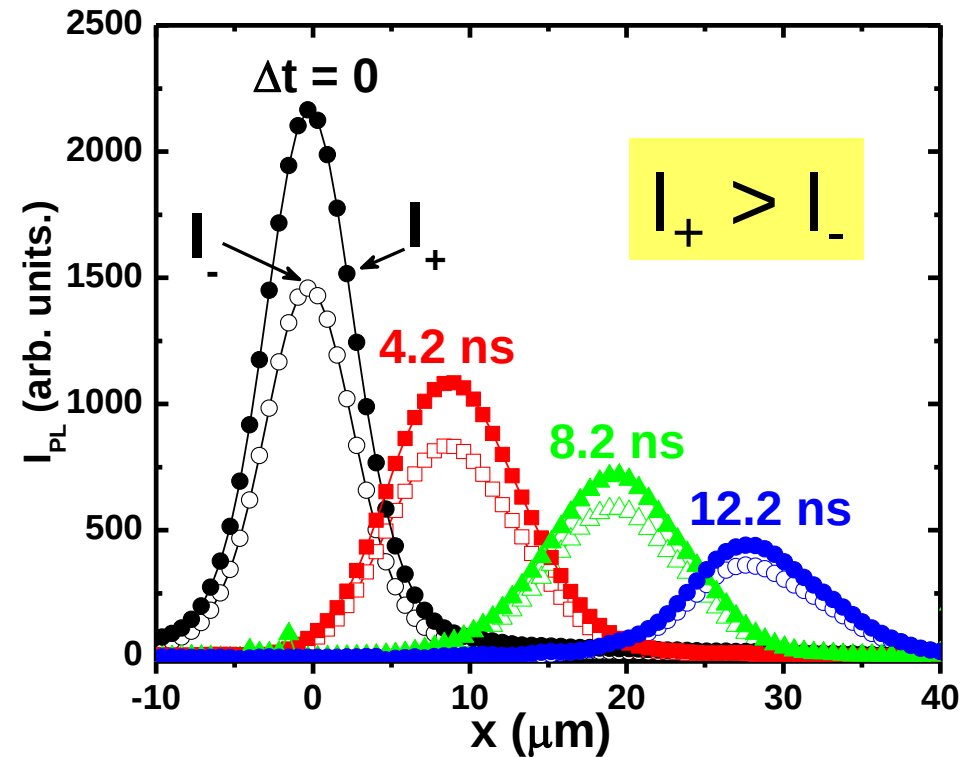


$$I_R \propto n_{\uparrow} \quad \text{and} \quad I_L \propto n_{\downarrow}$$

Spin polarization degree: $\rho_z = \frac{I_R - I_L}{I_R + I_L}$

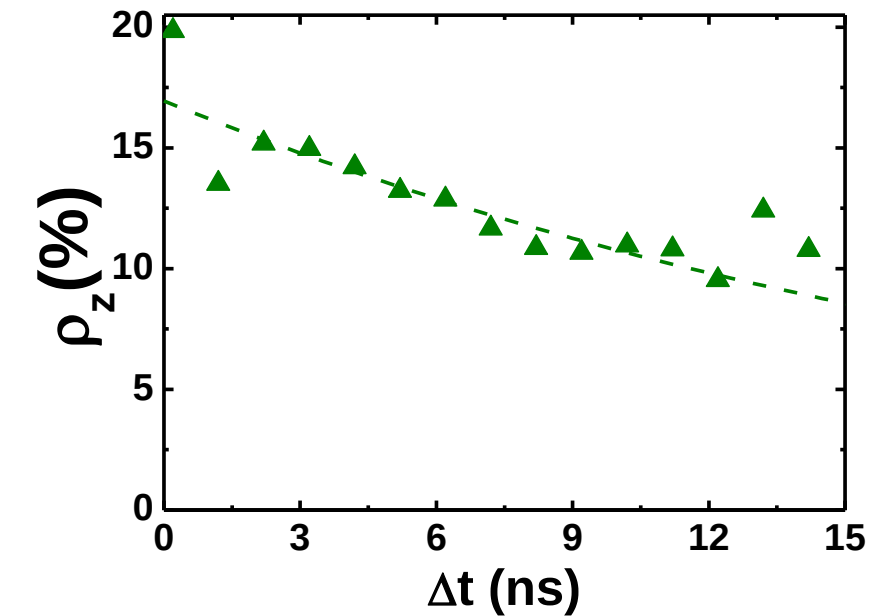
$x \parallel [001]$

SAW



Spin relaxation

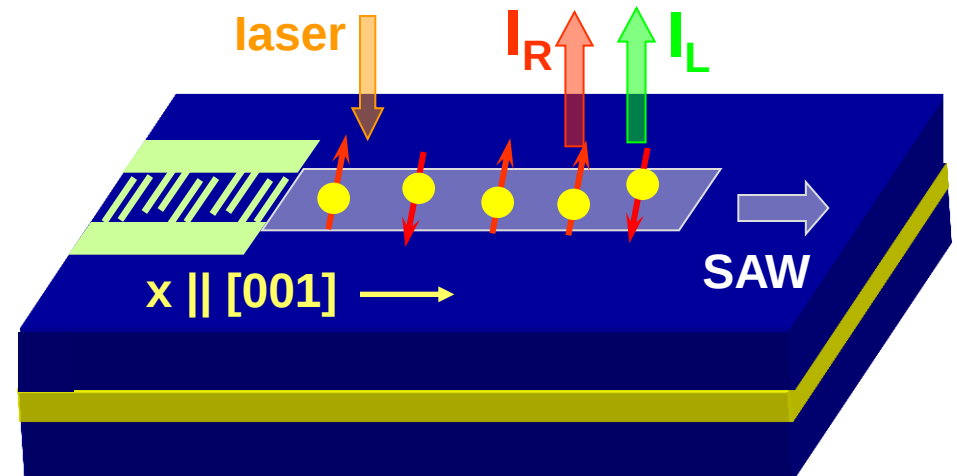
Transport along [001] direction



✓ Exponential decay

✓ Spin lifetime : $T_1 = (22 \pm 2) ns$

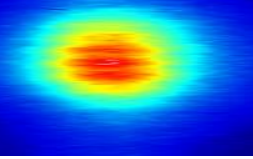
✓ Spin transport length: $L_s = T_1 v_{SAW} = (63 \pm 5) \mu m$



Longest spin lifetime and transport distance for this type of quantum well!!!!

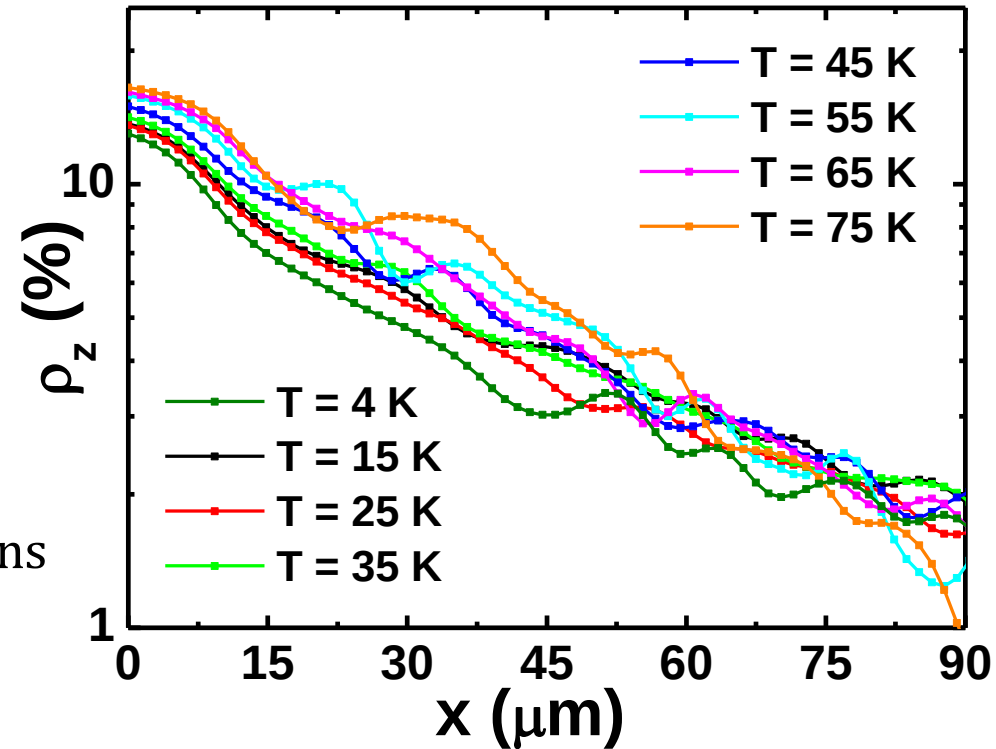
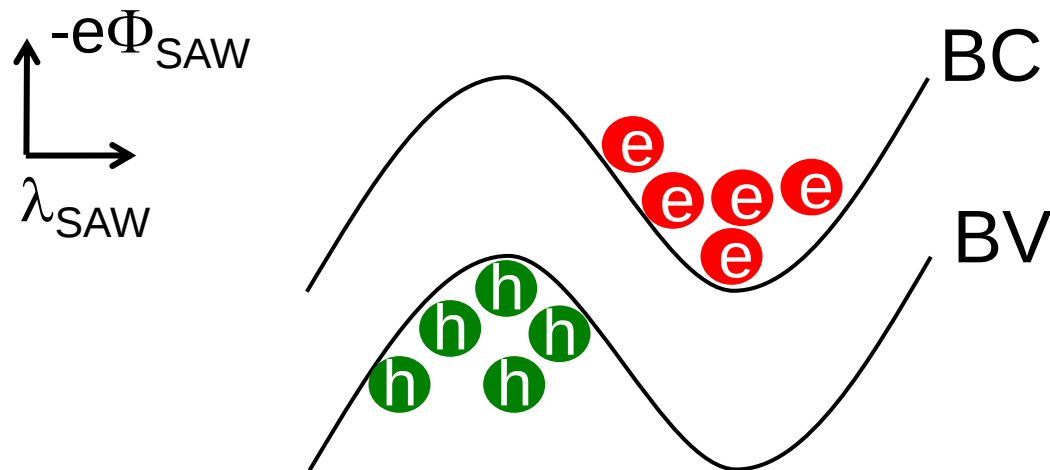
**Suppression of relaxation for
z-oriented spins**

Increasing temperature

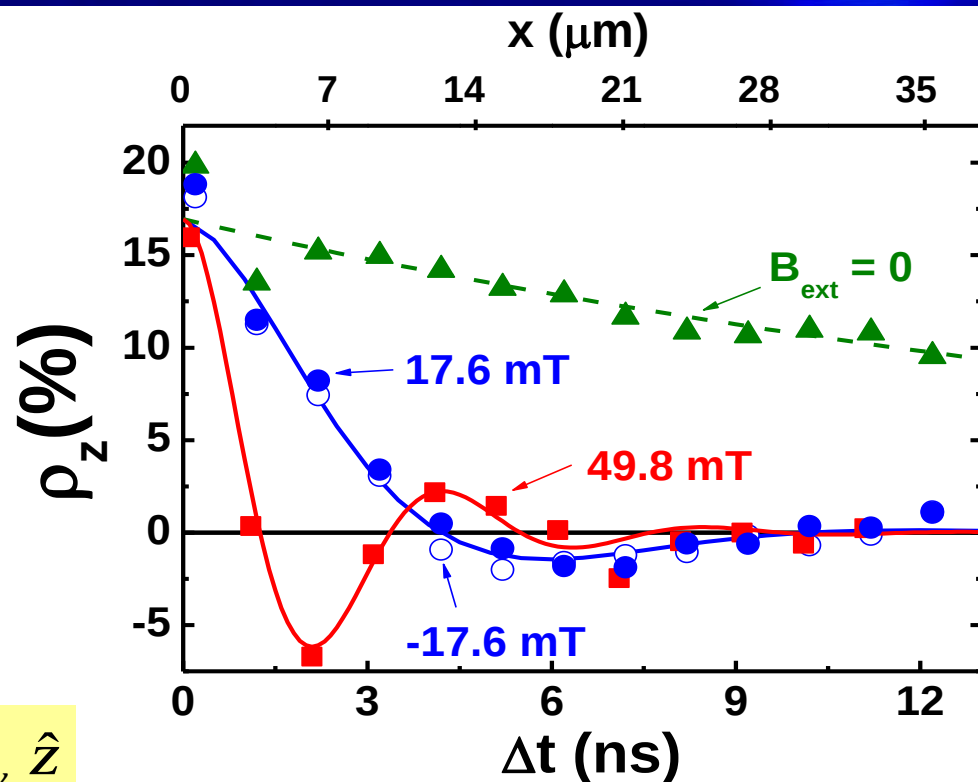
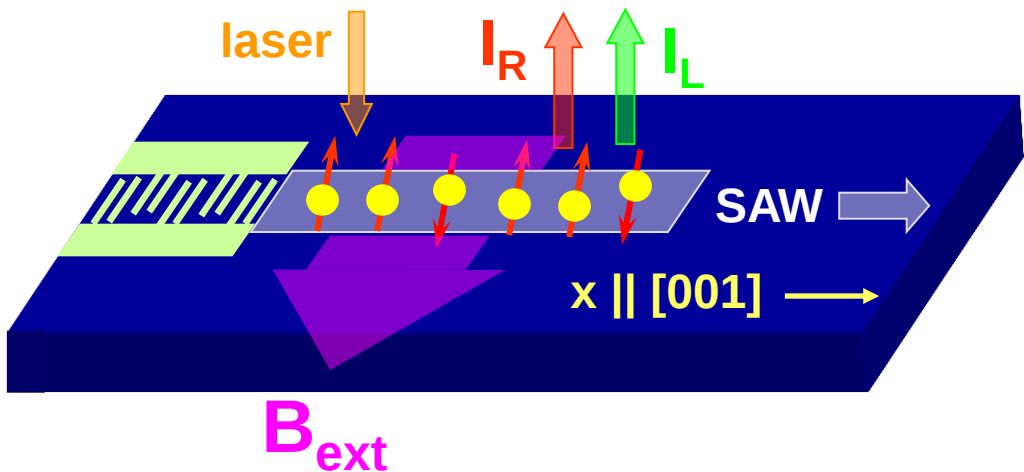
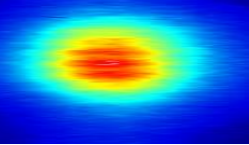


Temperature

- ✓ Spin decay is independent of T up to 75 K
 - ✓ SAW piezoelectric field avoids electron-hole recombination
- ✓ Spin transport at liquid nitrogen temperature
- ✓ Interesting for future applications



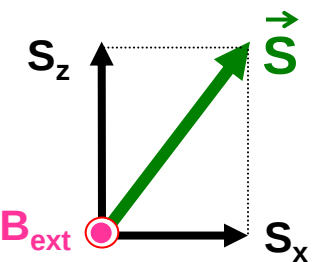
Spin manipulation



- Spin precession around B_{ext}
 - Precession is damped
 - Spin dephasing

$$\vec{B}_{BIA}(k) \propto k_y \hat{z}$$

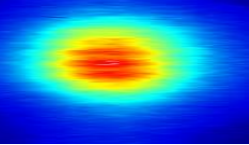
$B_{int} \parallel z$ fluctuates



- High in-plane relaxation rates appear

Effective spin dephasing time: $T_2^* = 2.3$ ns

Spin relaxation dynamics



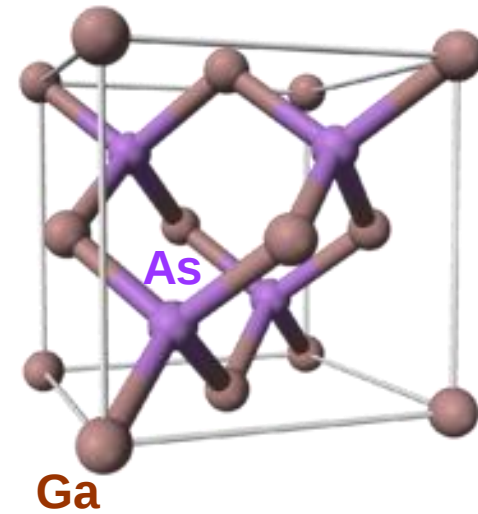
Spin-orbit Coupling

- ✓ Bulk inversion asymmetry
 - ✓ Binary semiconductors: GaAs
 - ✓ Electrons move in the crystal lattice
 - ✓ “Feel” the crystal potential

Effective magnetic field felt by the electron $B_{BIA}(k)$

$$H_{BIA}(k) = \hbar \Omega_{BIA}(k) \cdot \frac{\sigma}{2} = g_e \mu_B B_{BIA}(k)$$

$$mv = \hbar k$$



- Ensemble of $10^3 - 10^4$ spins
- k momentum dependence
- Fast average spin relaxation $\rightarrow T_1 \sim 100 - 300$ ps

(110) quantum wells*

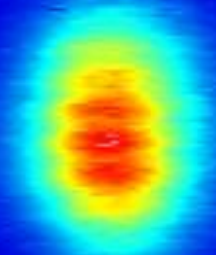
- ✓ Structural symmetry enhances the spin lifetimes $\rightarrow T_1 \sim 1-2$ ns

Acoustic transport

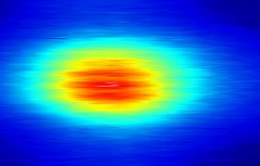
- ✓ SAW confinement potential screens electron spins $\rightarrow T_1 \sim 22$ ns

* Ohno et. al., Phys. Rev. Lett. 83, 4196 (1999)

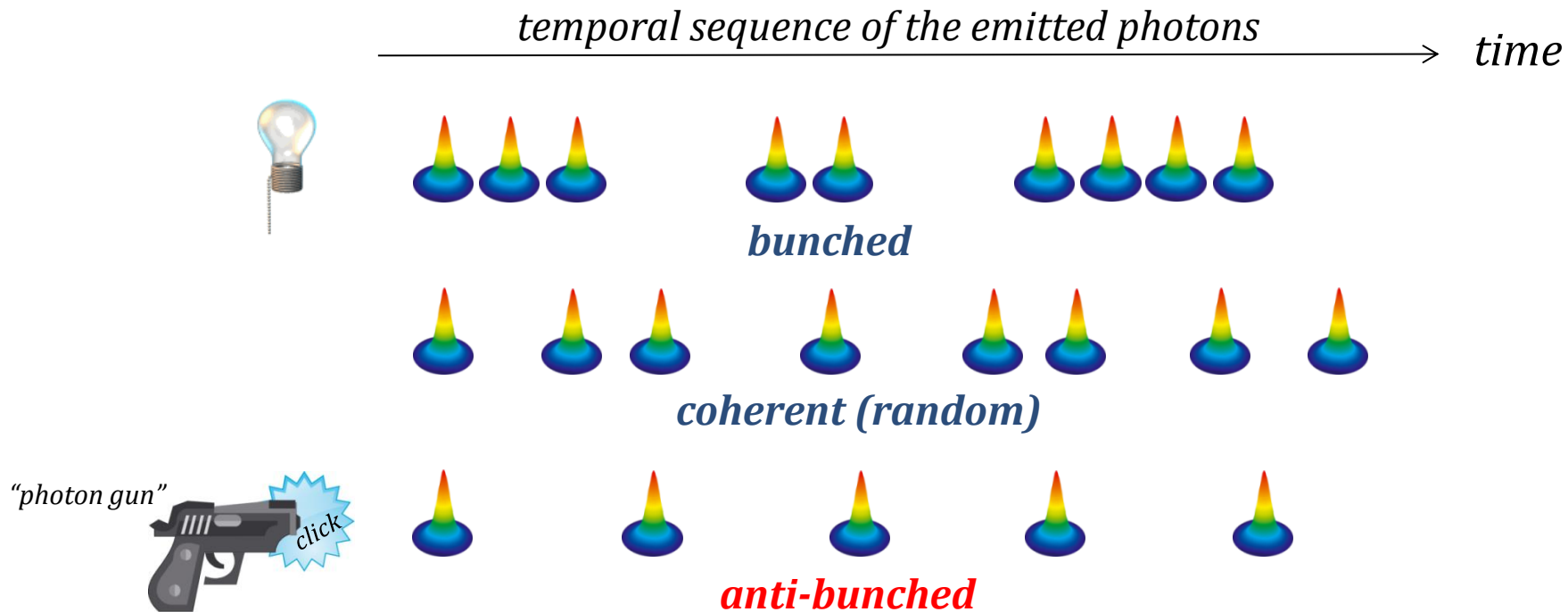
Carrier injection in coupled nanostructures



Single photon sources



What is a single photon source (SPS)



→ low probability of emitting two or more photons at the same time

SPS → source able to emit *single photons* pulses *on demand*...

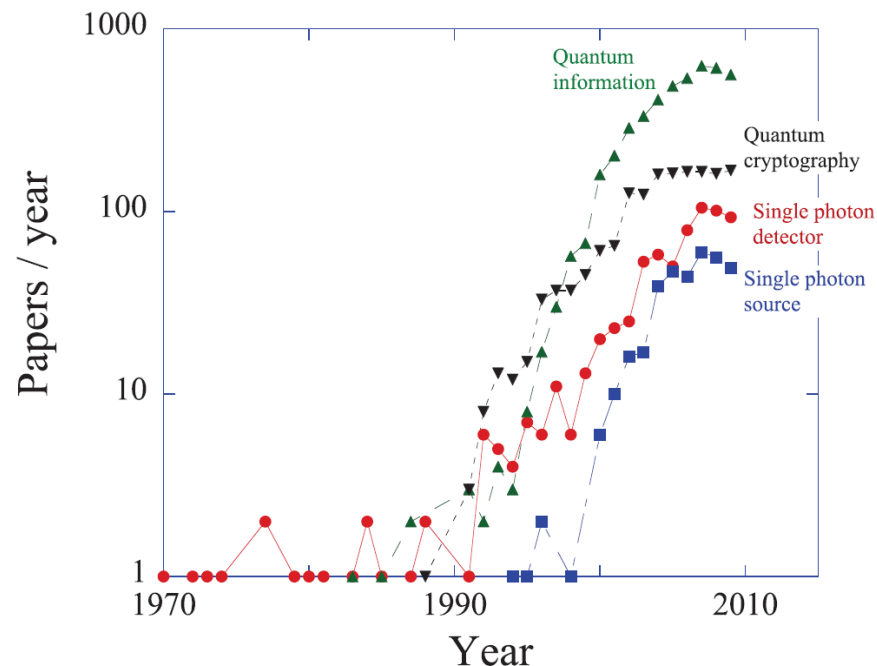
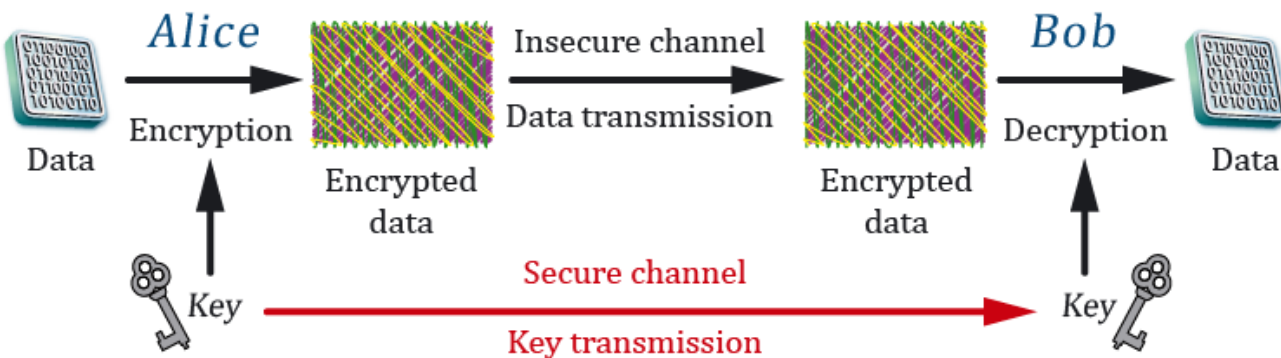
- ✓ regular stream of photons delivered one at a time
- ✓ high emitting probability (ideally, with certainty)

SPS applications

Why do we need SPSs?

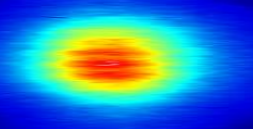
* B. Lounis and M. Orrit, "Single-photon sources", *Rep. Prog. Phys.* **68**, 1129 (2005)

- fundamental tests of quantum mechanics
 - quantum information processing
 - ✓ quantum computation
 - ✓ quantum cryptography
 - secure quantum key distribution
- by single photon pulses



Eisaman et. al. *Rev. Sci. Instr.* **82** 071101

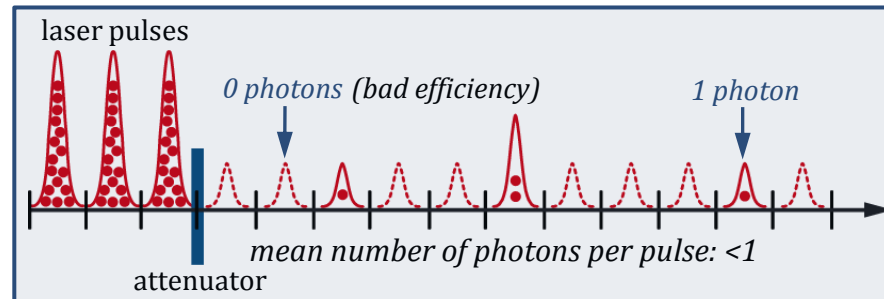
How to make a SPS



→ **elimination of multiple photon events:** *single photon source*

* early and macroscopic

✓ faint laser source



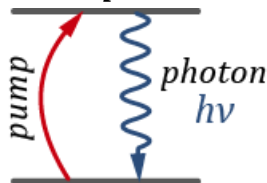
*H.J. Kimble et al., PRL **39**, 691 (1977)

*D.F. Walls Nature **280**, 451 (1979)

→ non-deterministic (Poisson) statistics:
random generation of single photons

* microscopic

✓ single quantum emitters
two-level quantum systems



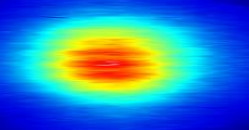
→ deterministic (sub-Poissonian) statistics

(e.g. atoms, organic molecules, defect centers, semiconductor nanocrystals & heterostructures)

SPS emission frequency → f_{pump}

*B. Lounis and M. Orrit, Rep. Prog. Phys. **68**, 1129 (2005)

SAW + SPS

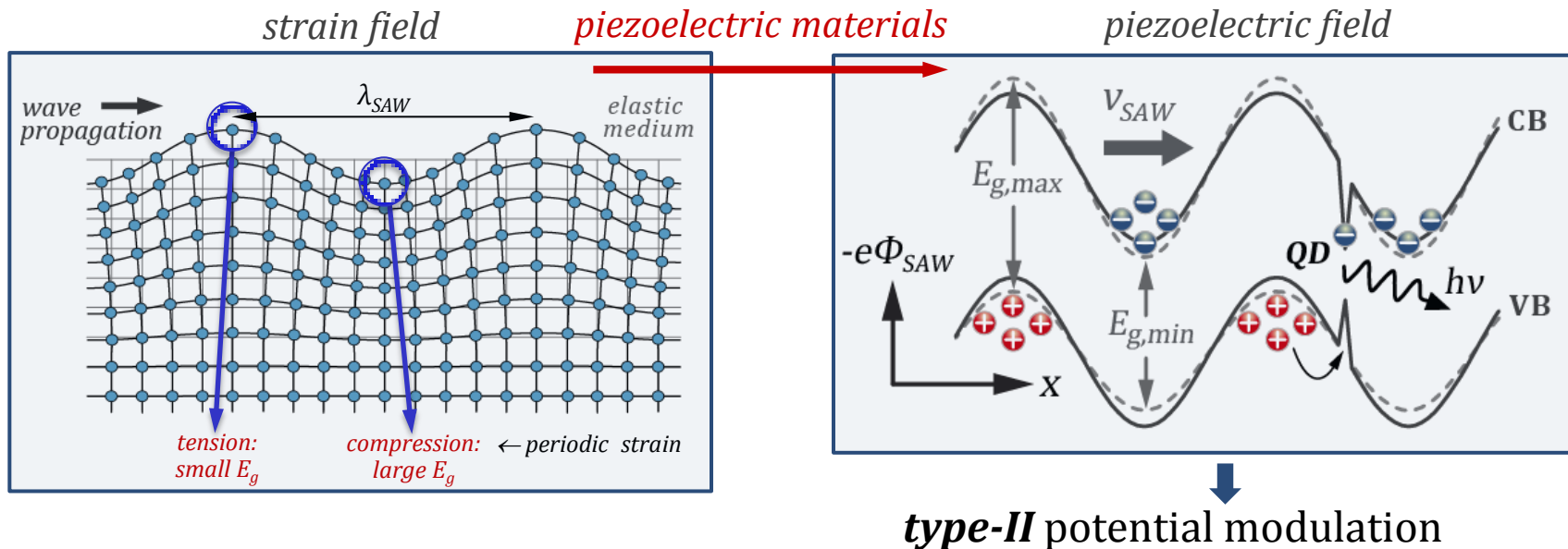


→ alternative way to generate semiconductor-based SPSs

SAW → surface elastic vibrations (acoustic phonons)

✓ SAW frequency: $f_{SAW} \rightarrow 100\text{'s of MHz to a few GHz} \rightarrow$ **high-repetition rate SPS**

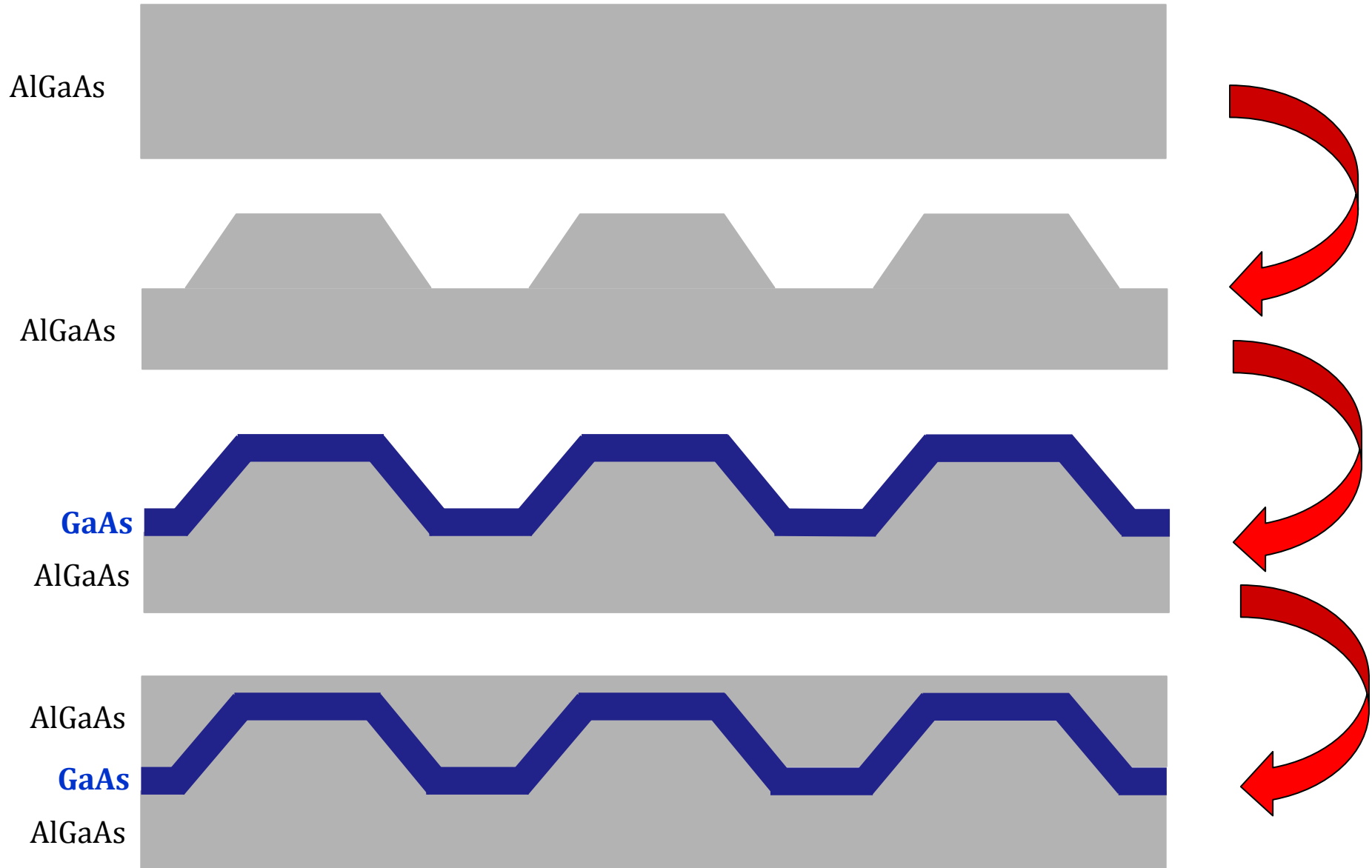
Modulation mechanisms



✓ **controllable transport** of carriers (unipolar or ambipolar)

Como obter o sistema de 2 níveis?

Etching process

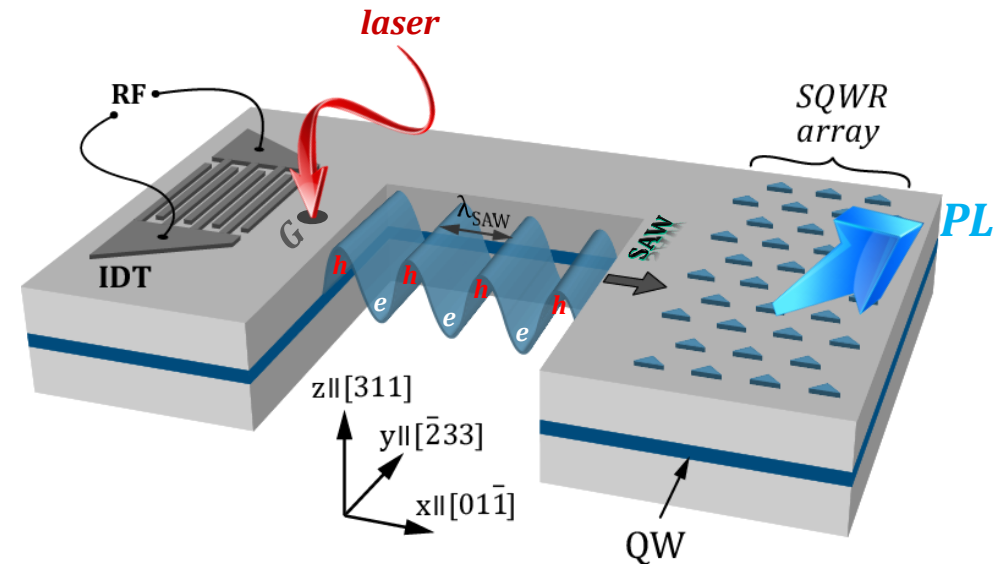


SQWR fabrication

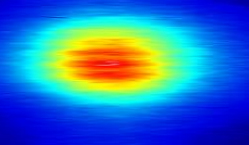
- ## SAW generation

-

*R. Nötzel et al., *Appl. Phys. Lett.* **68**, 1132 (1996)



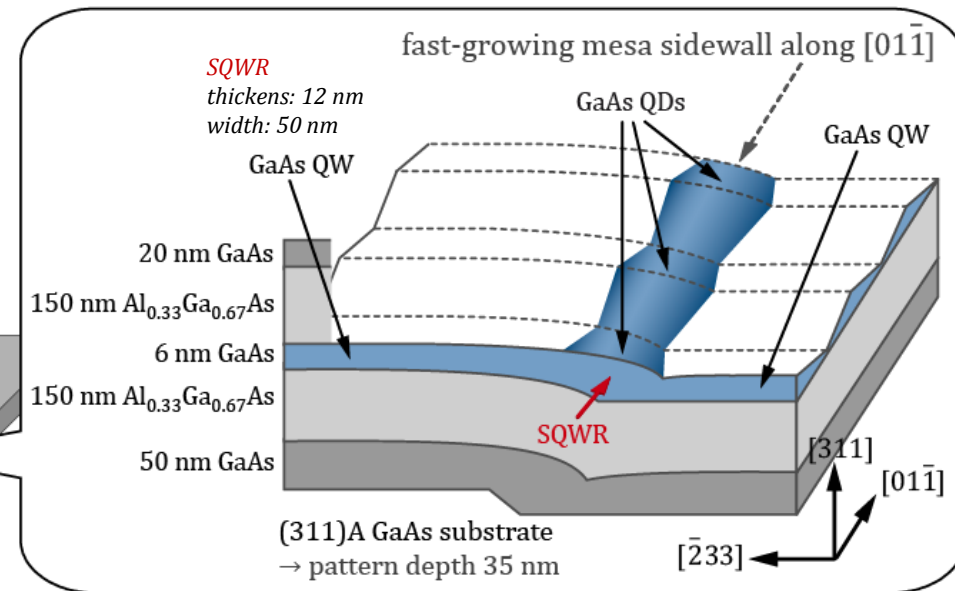
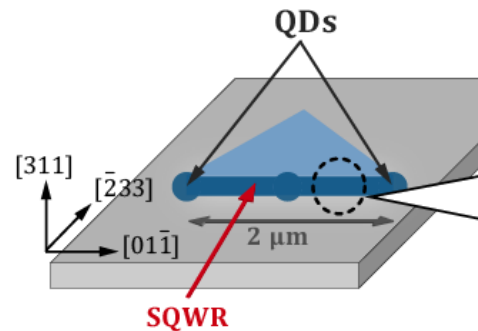
Short Quantum Wires (SQWRs)



Potential fluctuations along the SQWR

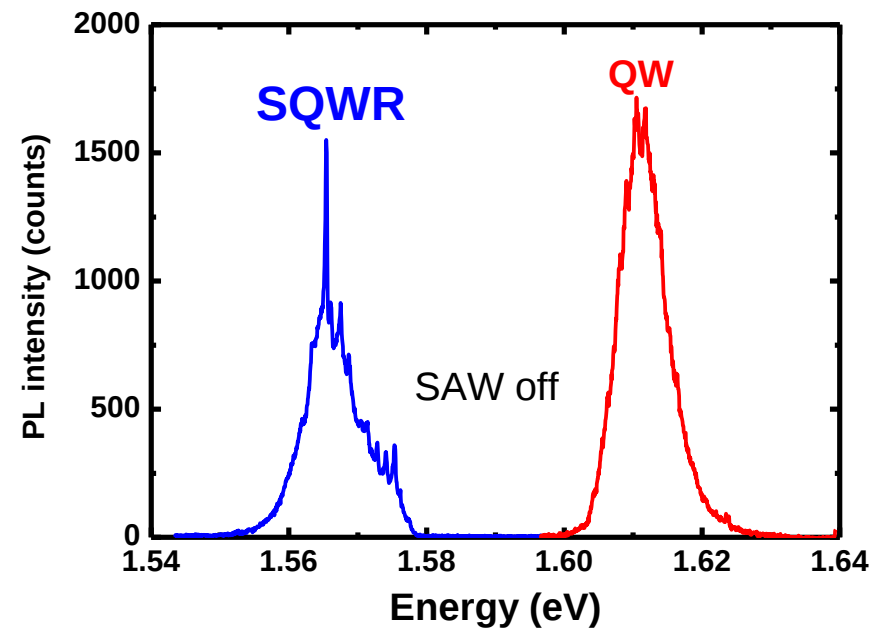
- ✓ Step-bunching
- linear array of QDs

**Intonti et al., Phys. Rev. B 63, 75313 (2001)*

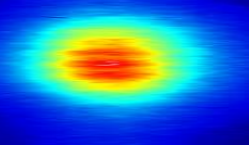


Spectral response from SQWRs

- ✓ Energy separation from the QW
- ✓ Sharp line are QD like states

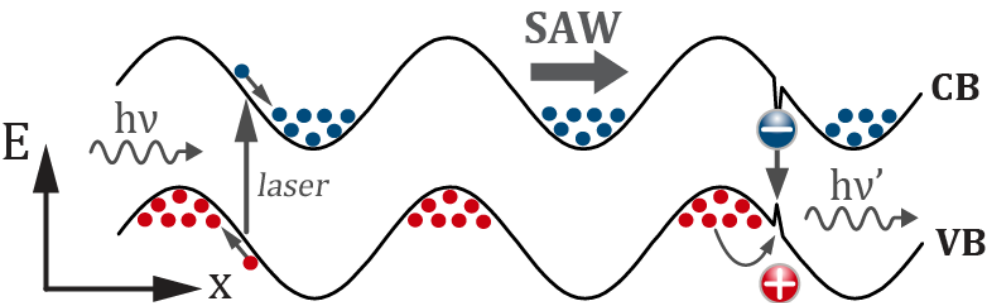


Carrier injection



$$\lambda_{\text{SAW}} = 4 \mu\text{m}$$

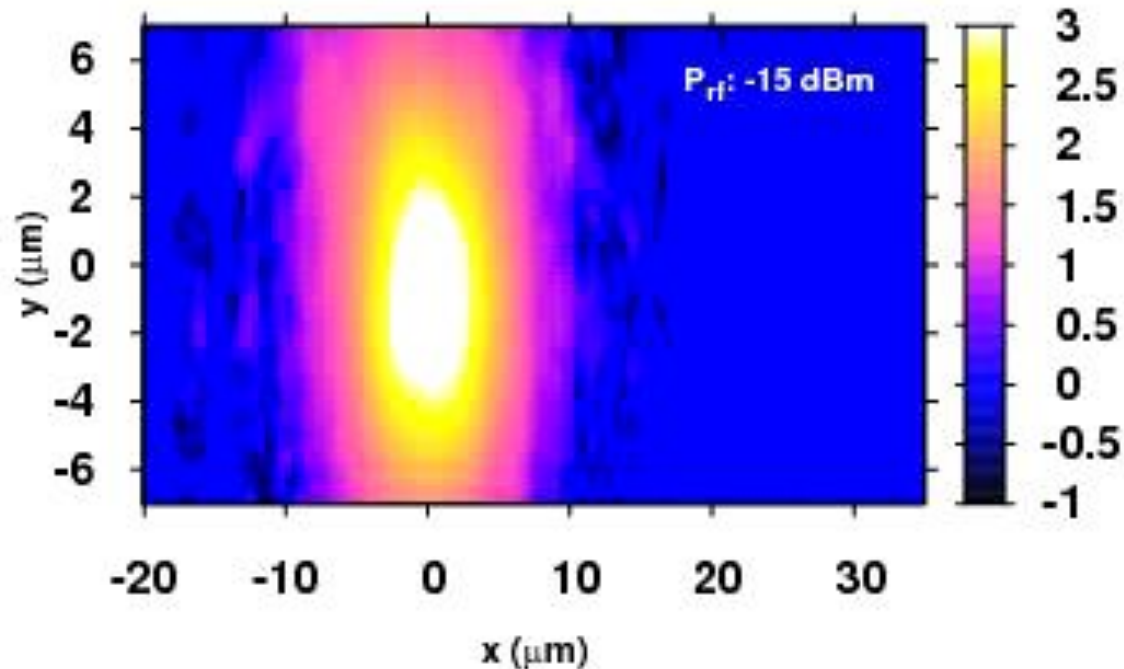
$$f_{\text{SAW}} = 750 \text{ MHz}$$



- ✓ **coupling** of different electronic systems:
carrier transport QW → SQWR
- ✓ recombination dynamics
→ selection of a single QD within a SQWR

Carrier transport and injection

- Optical excitation in the QW
- $\sim 20 \mu\text{m}$ from the SQWRs
- PL detection



PL from a single SQWR

- **low acoustic powers**

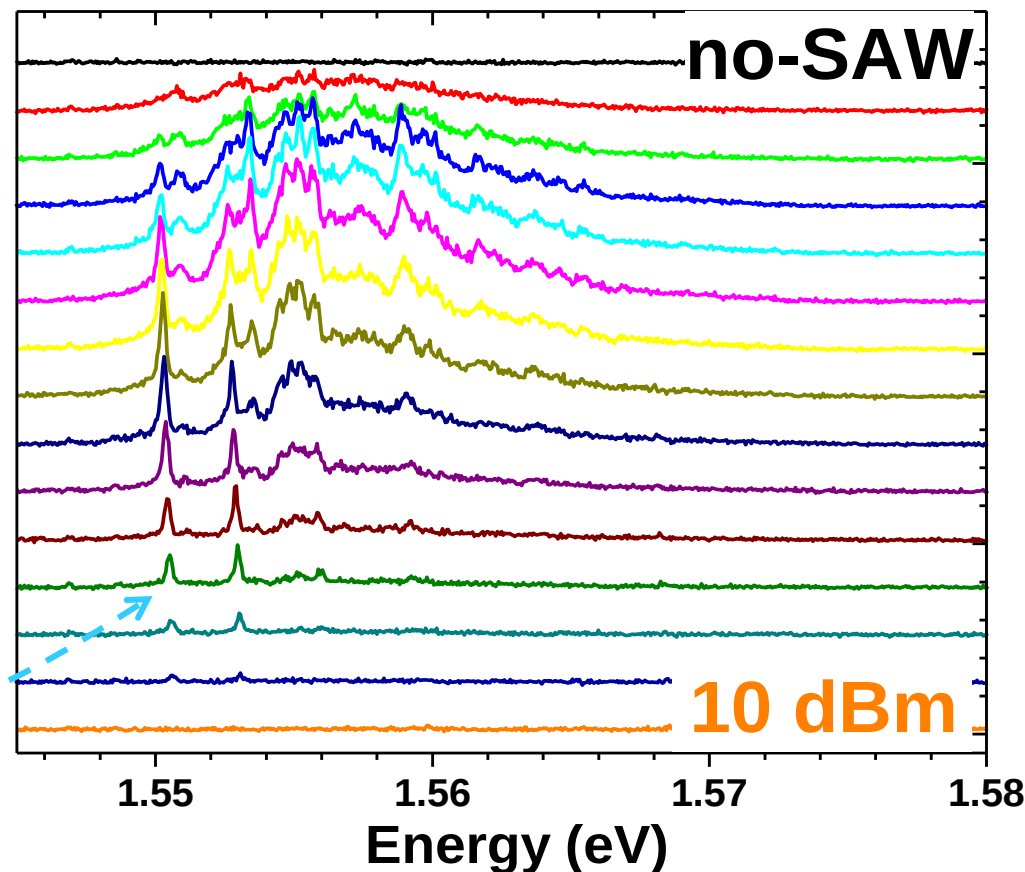
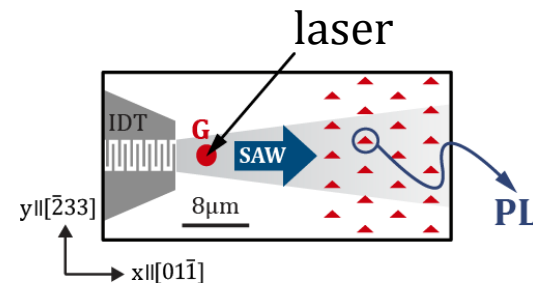
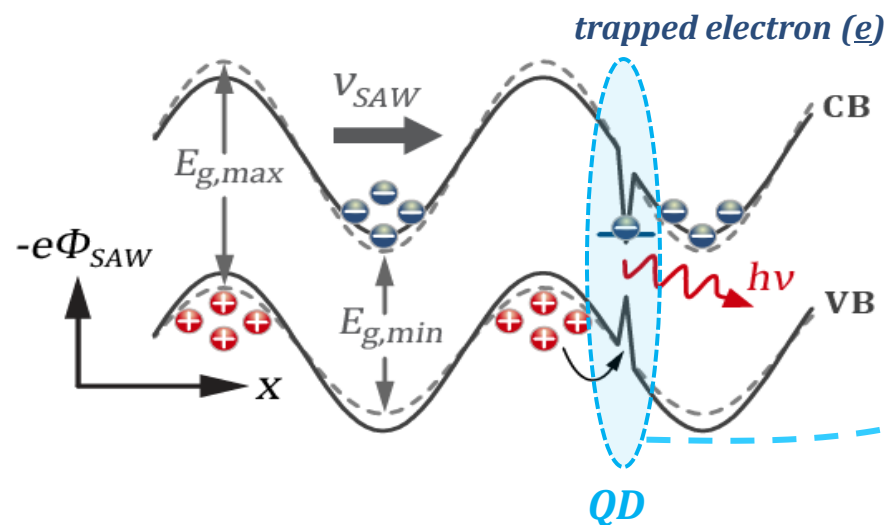
- ✓ large density of trapped electrons

- **high acoustic powers**

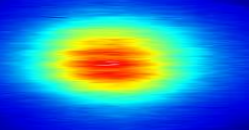
- ✓ low density of trapped electrons
→ fewer recombination events

sharp lines for high P_{SAW}

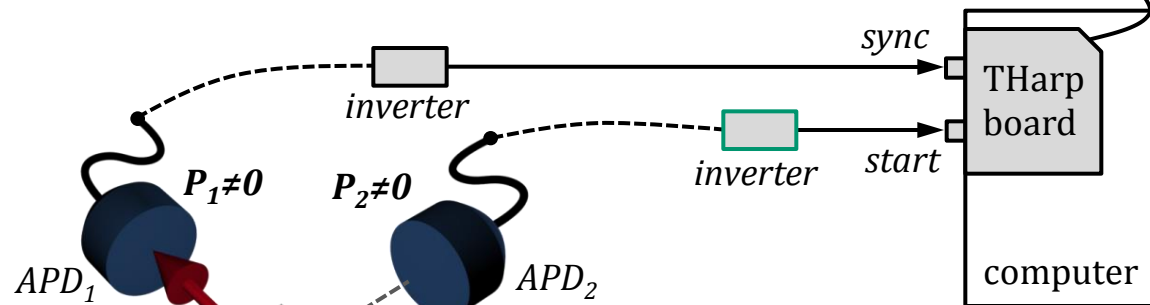
- localized states (QDs) within the SQWRs
✓ SAW amplitude → selection of single line



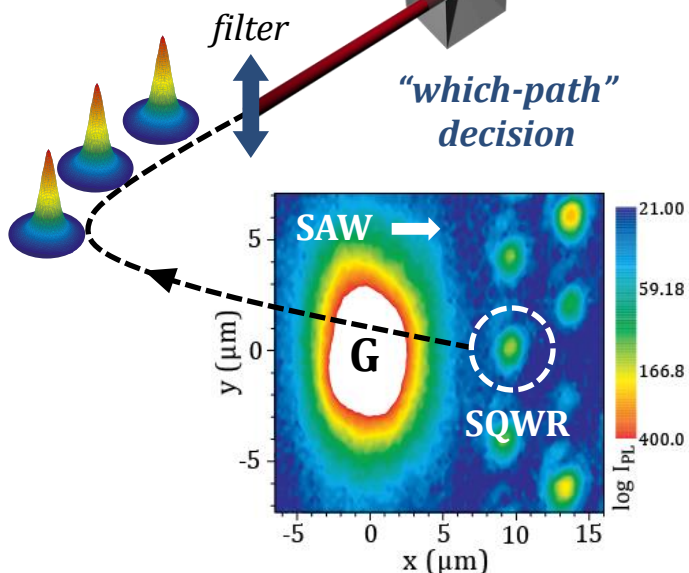
Photon correlation



Hanbury-Brown and Twiss setup



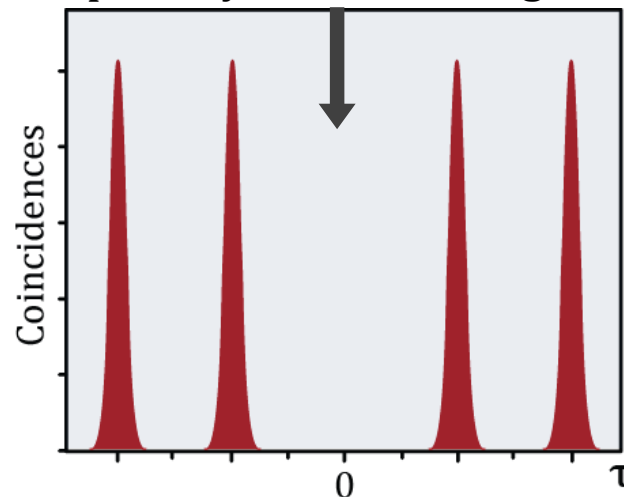
time interval between the successive photon-detection events at two detectors (APD)



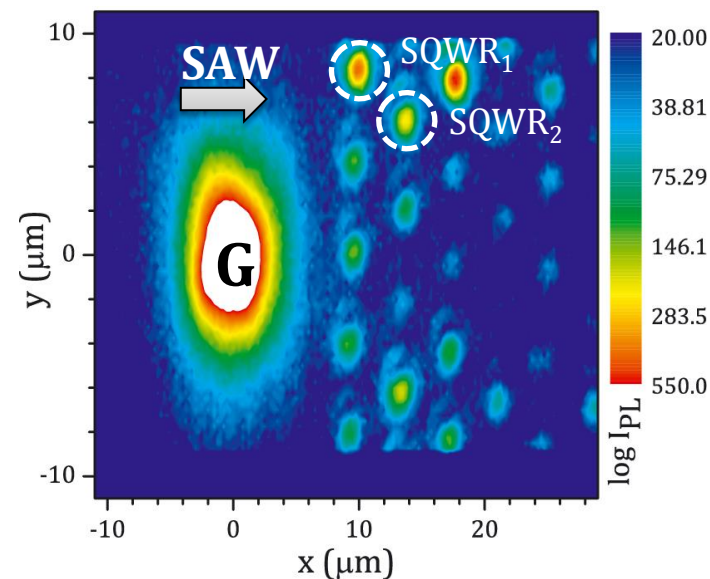
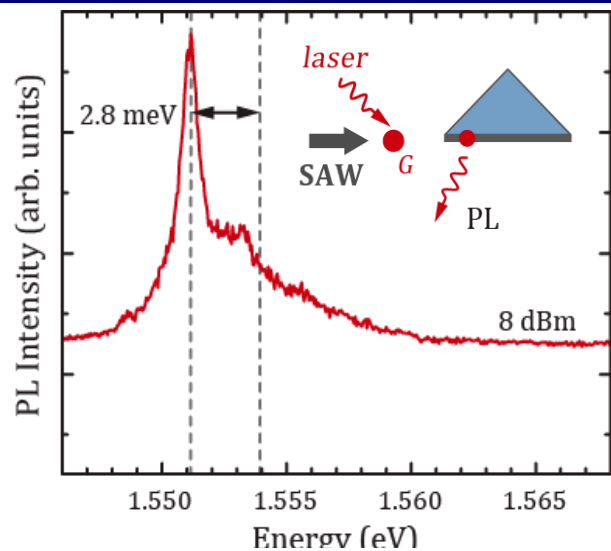
quantum nature of light

✓ joint photo-detection probability: $P = 0$ at $\tau = 0$

suppression of zero-delay peak by anti-bunching

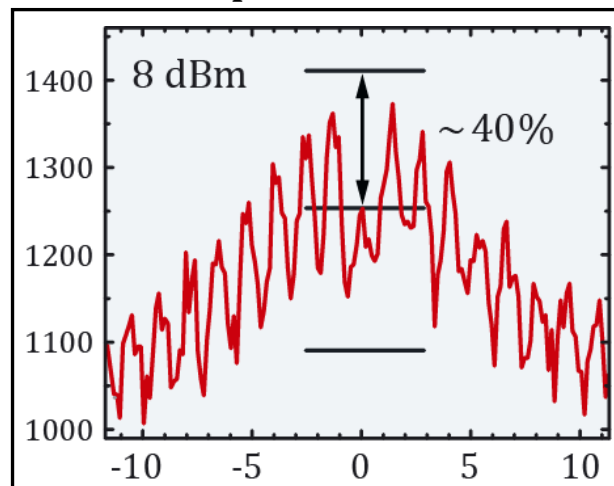
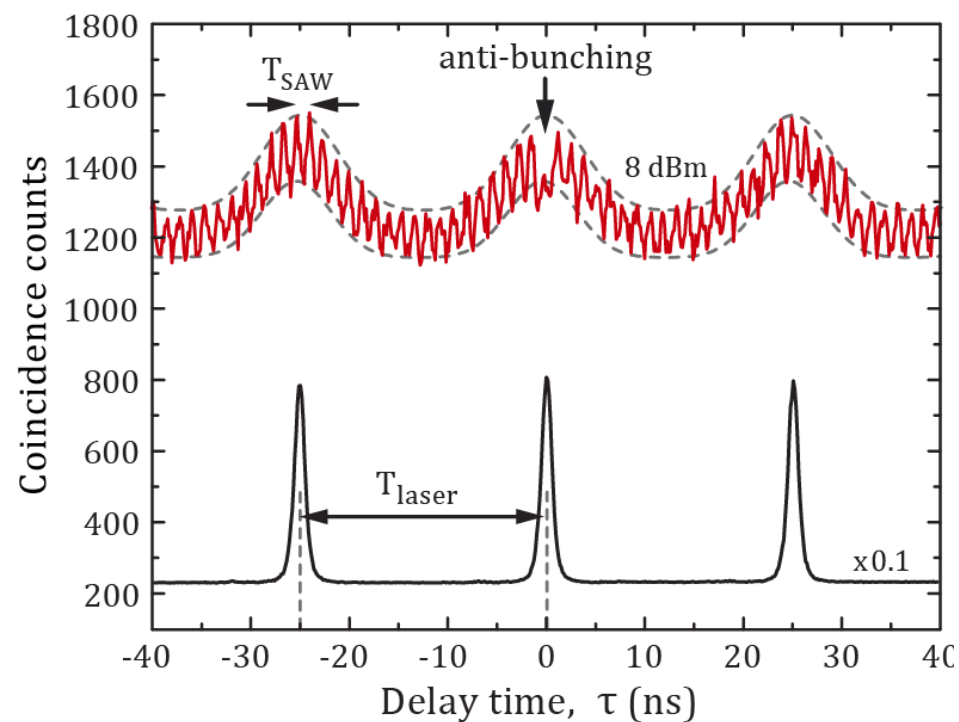


Anti-bunching

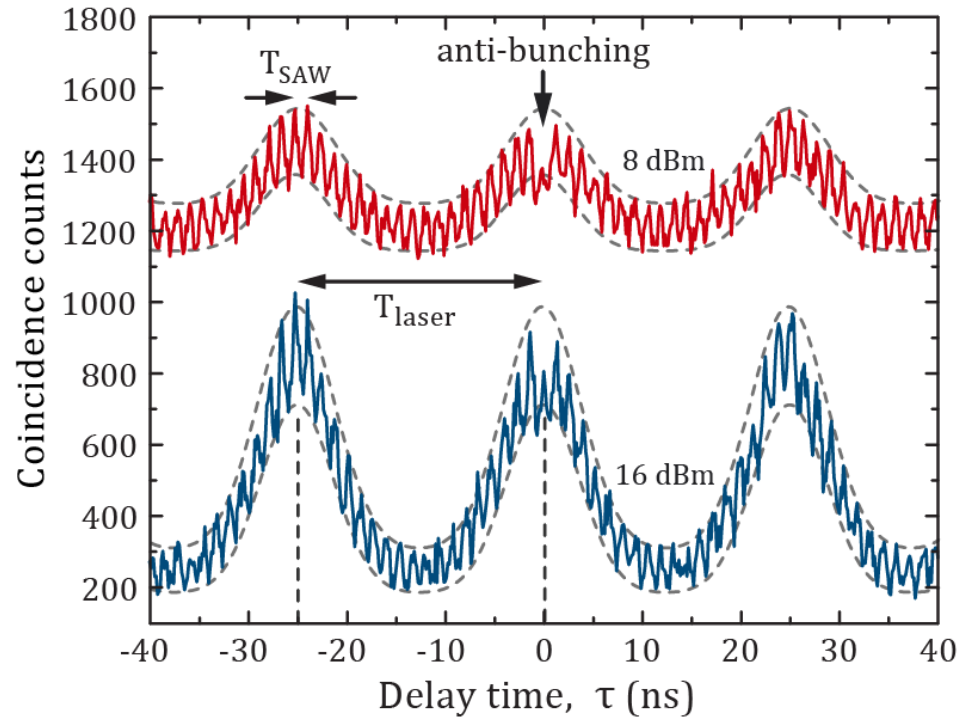
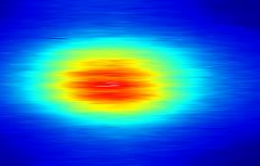


Reduced amplitude at $\tau=0$

→ lower probability for simultaneous emission of two photons

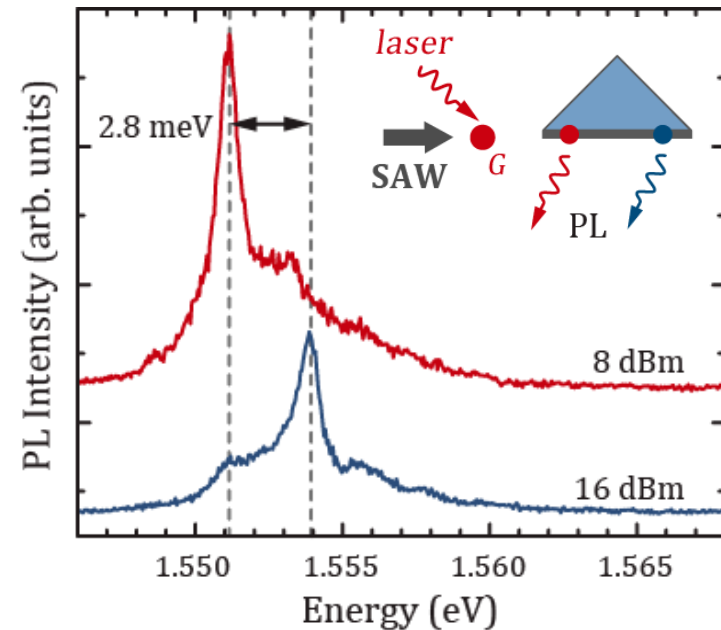


Anti-bunching



Emission energy

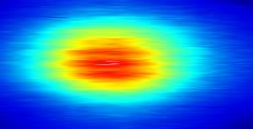
- ✓ Recombination depends on SAW power
- selection of emission centre by controlling P_{rf}



- **SAW based SPS**

- Carrier injection from a QW into individual states of SQWRs
 - ~10 times faster (750 MHz) than optically pumped SPSs
- Adjustable emission energy

Outlook



Surface Acoustic Waves (SAWs)

- ✓ Powerful tool coupled to optical spectroscopy
- ✓ Modulation due to strain and piezoelectric fields
- ✓ **Manipulation** and **transport** of excitations in nanostructures
 - ✓ Carrier transport/injection
 - ✓ Spin transport
 - ✓ Single photon generation
- Basic research
- Applications

Acknowledgments



Optical modulation of semiconductor nanostructures

✓ email: odilon@ifi.unicamp.br
<http://sites.ifi.unicamp.br/odilon>

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Odilon D. D. Couto Jr.

<http://www.ifi.unicamp.br/~iikawa>

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