Active Nanophotonics: From Coherent Control of Quantum Emitters to Plasmonic Nanolasers

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Coherently Driving Quantum Emitters in Micro-cavities

1st CW operation of Plasmonic Nanolaser (SPASER)
Quantum Emitter: A two level electronic system that is optically active

They are also the fundamental quantum information units: Qubits

Examples:

\[ |0\rangle \quad \text{No Exciton (Vacuum)} \]
\[ |1\rangle \quad \text{One Exciton Ground state} \]
\[ |2\rangle \quad \text{One Exciton Excited state} \]
Structural Characterization

Size/Shape/Density/Composition
[Phys Rev Lett 84, 334, (2000)]

- Truncated pyramid exterior
- Non-uniform In-distribution
- Inverted triangle In-rich core

Sample used in this study

Atomic structures
$|\Psi\rangle = \alpha |0\rangle + \beta e^{i\phi} |1\rangle$

$\alpha, \beta$ are real, and $\alpha^2 + \beta^2 = 1$

$S = \begin{pmatrix} S_1 \\ S_2 \\ S_3 \end{pmatrix}$

$S_3 = \alpha^2 - \beta^2 = \cos \theta$

$S_1 = \sin \theta \cos \phi$

$S_2 = \sin \theta \sin \phi$

A quantum states can be represented by pseudo spin vector on the Qubit sphere

Unitary Transformation $\rightarrow$ Qubit Rotation
Bloch Model for a Two-level Quantum System

Bloch Vector

Bloch vector = pseudo spin vector in rotating frame

\[ \frac{d\tilde{\rho}}{dt} = \tilde{G} \times \tilde{\rho} \]

\[ \tilde{G} = \begin{pmatrix} \kappa E(t) \\ 0 \\ \Delta \omega \end{pmatrix} \]

\( \theta \): input pulse area.

For an arbitrary pulse shape, \( \theta(t) = \kappa \int \varepsilon(t) \, dt \)
I- Quantum Coherent Control of Single Quantum Dots

Hall-mark: Rabi Oscillations

Stievater et al., PRL 87, 133603 (2001)
DT of Exciton ground states


• All performed on bare SQDs, all have low fidelity.
• None can be categorized as coherent control of quantum emitters
Cavity QED with QDs as quantum emitters

- Weak coupling – Purcell effect
- Strong coupling – Vacuum Rabi Oscillations

These demonstrated QD-Cavity QED:
No active control using external field
(i.e. Non-resonance excitations)
Resonant Spectroscopy

Photoluminescence (PL) and PL excitation (PLE) spectroscopy:

True resonant excitation:

The main difficulty: Laser scattering
Suppression of laser scattering background

Atoms in an optical cavity

|0⟩\rightarrow hv \rightarrow |1⟩ → Fluorescence

Solid-state Analogy

fluorescence
quantum dots
mirrors
fiber
GaAs
cryostat cold finger
Resonance Fluorescence

\[ T_2 = 470 \text{ ps} \]

\[ \text{Energy (μeV)}: -10 -5 0 5 10 \]

\[ \text{Detuning (μeV):} -18.0 -12.6 -7.3 -1.9 0.4 3.5 8.9 14.2 19.6 \]

\[ \text{Wavelength (nm):} 914.4 \quad 914.8 \]

\[ \text{Intensity (a.u.)} \]

\[ \text{Residual laser line} \]

\[ \sim 0.7 \text{ GHz} \]
Pulsed Control: Rabi Oscillations

Earlier work

\[ |0\rangle \quad \kappa \quad |1\rangle \]

Laser \[ \gamma_1 \]

Photoluminescence intensity \[ |2\rangle \]

\[ \gamma_2 \]

\[ \Delta E = 120 \mu eV \]

\[ 2\pi \]

\[ 0 \]

\[ 1 \]

\[ \frac{\pi}{2} \]

\[ \pi \]

\[ \frac{3\pi}{2} \]

\[ 2\pi \]

\[ \frac{5\pi}{2} \]

Nearly Ideal Rabi Flopping (high Fidelity)

Average intensity\(^{1/2}\) (mW\(^{1/2}\))

PRB 72, 035306 (2005).

\[ T_2(0) = 45 \text{ ps}, \mu = 41 \text{ Debye} \]

\[ \tau_p = 9.3 \text{ ps} \]

\[ \tau_p = 7.2 \text{ ps} \]

\[ \tau_p = 5.4 \text{ ps} \]
Continuously-Driven System

\[ \Omega = \mu E \]

The emission spectrum acquires sidebands!

Emission spectrum:

Strong excitation
$$\Omega = \mu E$$

Single QD ultra-fast optical modulator !!!

Self Interference $\rightarrow$ First order correlation $g^{(1)}(t)$
Instrument resolution determined using ring laser as the source

Longest coherent time that can be resolved $\rightarrow \sim 1.5$ ns
First order correlation function measurements

Müller et al., PRL 99, 187402 (2007)

\[ g^{(1)}(\tau) \]

\[ \text{Fringe contrast} \]

\[ \sqrt{\text{Intensity (a.u.)}} \]

\[ E(\mu eV) \]

\[ |\Omega| = 0.9 \mu eV \]

\[ |\Omega| = 4.8 \mu eV \]

\[ |\Omega| = 6.9 \mu eV \]

\[ |\Omega| = 10.0 \mu eV \]

\[ |\Omega| = 13.3 \mu eV \]

\[ |\Omega| = 16.6 \mu eV \]

\[ \text{Time delay (ps)} \]

\[ \text{Fringe contrast} \]

\[ g^2 \]
Simultaneous measurements of Mollow triplets and 2\textsuperscript{nd} order correlation

Objective

Fiber

Sample

Fabry-Perot

Beamsplitter

APD

APD

Hanbury Brown and Twiss Correlation Measurement

PicoHarp 300\textsuperscript{TM} 4 ps resolution

Spectrometer

Resolution (~300 MHz)
Mollow Triplets in Frequency Domain

With much better resolution (~ 30 MHz)
(Courtesy of A. Muller)
Non-classical photon statistics

Photon statistics: the Hanbury-Brown and Twiss interferometer

\[ g^{(2)}(t) : \text{Probability to detect a second photon} \ t \ \text{seconds after detecting a first photon} \]
Rabi Energies vs Sqrt(Laser Power)

\[ g^2(t) = 1 - e^{-\frac{t}{2\left(\frac{1}{T_1} + \frac{1}{T_2}\right)}} \left\{ \cos(\Omega't) + \frac{(1/T_1 + 1/T_2)/2}{\Omega'} \sin(\Omega't) \right\} \]

Fitting: Using experimentally measured T1 (275 ps), T2 (132 ps)
Summary

• Resonance fluorescence is achieved
• Ideal Rabi Flopping (High Fidelity)
• Mollow fluorescence (i.e. Triplets) is observed
• Rabi oscillations in $g^{(2)}(t)$ → coherent control of quantum emitters
  • Single acoustic phonon mediated inter-level interactions
  • Decoherence limited by single phonon scattering
Coherent Control of Quantum Emitters

Contributors (current and former students/postdocs):
A. Muller, E.N. Flagg, H. Htoon, QQ Wang, P. Bianucci, X. Wang, S. Founta, J. Robertson

Collaborators:
D. Deppe, A. Holmes, G. Salamo

Funding:
NSF, W.M. Keck Foundation, Texas Advanced Research Program
RF as a quantum mechanical tool

- Deterministic Generation of Entangled Photon Pairs

Due to exchange splitting, $\Delta E_{ex}$, the two paths are distinguishable.

Using RF as a QM tool to create indistinguishable paths

A. Muller, PRL 103, 217402 (work done at NIST)
Quantum Interface of Matter Qubits and Photon Qubits → Toward Quantum Network Formation

Photon qubits

Matter qubit

Matter qubit

Resonantly control of quantum emitter in cavity QED regime is the key.

Quantum information of matter qubit truthfully projected to photon qubits

Quantum information of photon qubit is then converted to matter qubits

\[ c_e |e\rangle + c_g |g\rangle \leftrightarrow c_e |\alpha(t)\rangle + c_g |\text{vac}\rangle \]

\[ \alpha(t) = F(\Omega(t)) \]

\[ \Omega(t) = F^{-1}(\alpha(t)) \]
Metallic Nanophotonics (sub-diffraction photonics)

Mark L. Brongersma, and Vladimir M. Shalaev
Science 328, 440 (2010)
Plasmonic Nanolasers using Epitaxial Ag films

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ACT I: Demonstration of 1st CW operation Plasmonic Nanolaser
(SCIENCE 337, 450 (2012))

ACT II: All Color Plasmonic Nanolasers on A Single Platform
Plasmonic Nanolaser
Using Epitaxially Grown Silver Film

[Science 337, 450 (2012)]

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Conventional Laser Cavity

Diffraction-limited cavity size: Smallest $d \approx \lambda/2$
Lasing from Isolated ZnO Nanowires
van Vugt et al., Nano Letters 6, 2707 (2006)

In practice, semiconductor nanowire lasers require at least a few microns in length to overcome the losses.
Size Limits in Semiconductor Nanowire Lasers: Diffraction-Limited Photonic Microcavity


\[ L_{\text{min}}^{(1)} = \frac{\lambda}{2n_{\text{eff}}} \]

\[ L_{\text{min}}^{(2)} = \frac{-\ln(r_1 r_2)}{(-2k'')} = \frac{-\ln(r_1 r_2)}{G_m} \]

Criteria for Nanowire Lasing

- Highly Facetting/Single Crystalline
- Lower limit \( \lambda/2n \): \( r > 60 \) nm
- \( G_{\text{th}} \): 400 – 3000 cm\(^{-1}\)
- High-Q: 500-1500

Surface plasmon polariton excited at planar dielectric/noble metal interface

Metal Optics

Dispersion Relationship

\[ \omega = \frac{\varepsilon'_m + \varepsilon'_d}{\varepsilon_m \varepsilon'_d} c k_x \]

\[ \omega = \omega_p + c^2 k_x^2 \]

\[ \omega = \omega_p \sqrt{\frac{1}{1 + \varepsilon_d}} \]

\[ \omega_s = \omega_p \sqrt{\frac{1}{1 + \varepsilon_d}} \]

\[ \varepsilon' \rightarrow -\varepsilon_d \]

\[ \omega = c k_z \phantom{\frac{\varepsilon'_m + \varepsilon'_d}{\varepsilon_m \varepsilon'_d} c k_x} \]

photons in air

plasmon polariton

surface plasmon polariton
Plammonic Nanolaser (SPASER)
(original concept: Bergman and Stockman, PRL 2003)

Localized Surface Plasmon

Confined Surface Plasmon Polaritons


Spasing: → Needs to contain all Lasing Signatures
   → Coupling between confined SPPs and the Gain medium
Device Platform:
**InGaN@GaN Nanorods on Epitaxial Ag films**

Metal-Oxide-Semiconductor (MOS) Structure

Electric Field Distribution
(Chun-Yuan Wang, NTHU)

3D sub-diffraction semiconductor nanolaser comparable to the size of the MOS transistor

Schematic of the ultralow-threshold plasmonic nanolaser. A single InGaN@GaN core–shell nanorod is placed on a thin SiO₂ covered epitaxial silver film (28 nm thick). The resonant electromagnetic field is concentrated at the 5-nm-thick SiO₂ gap layer sandwiched by the semiconductor nanorod and the atomically smooth smooth silver film.
Epitaxial Growth of Silver Film on Silicon (111)

- “Quantum” growth of silver epifilm by MBE enables low-loss plasmonic cavity.
- Atomically smoothness of Ag epifilm is confirmed by RHEED, STM, STS, and AFM.

RHEED

3D Ag nanoclusters
Smooth Ag film

Quantum well states
Surface state

Growth: Jisun Kim, Charlotte E. Sanders & Chih-Kang Shih (UT-Austin)
Gain Medium: InGaN@GaN Core–Shell Nanorod

✓ High quality, MBE-grown InGaN @GaN core–shell nanorod → gain medium
✓ SEM image of the nanorod placed on the Ag epifilm for lasing measurements
✓ STEM and TEM structural analyses of InGaN@GaN core–shell nanorod
  EDS elemental mapping of In, Ga, and N, confirming the core–shell structure

InGaN@GaN Nanorods: Y. Lu, S. Gwo (NTHU)

TEM: Ming-Yen Lu & Lih-Juann Chen (NTHU)
Lasing Characteristics

Power dependent lasing spectra of a single InGaN@GaN core–shell nanorod placed on the epitaxial Ag film with a 5 nm SiO₂ gap layer, **optically pumped by a CW 405 nm diode laser.**

**Lasing Signatures**
- Pumping power dependence
- Concurrent thresholds of intensity kink & linewidth narrowing
- Mode competition with varying temperature
- Temporal coherence
Lasing Characteristics

The log-log plots of output peak intensity versus the pump power at the main lasing peak (510 nm) are shown with the corresponding linewidth narrowing behavior when the plasmonic laser are measured at 8 K and 78 K, respectively. The spontaneous emission coupling factor ($\beta$) can be estimated to be 0.73 at 8 K.
Temperature dependent lasing behavior, showing mode competition.

Lasing Signatures

- Pumping power dependence
- Concurrent thresholds of intensity kink & linewidth narrowing
- Mode competition with varying temperature
- Temporal coherence
What is the signature for temporal coherence? 
→ Photon-Photon Correlation Function, $G^{(2)}(\tau)$

$$g^{(2)}(\tau) = \frac{\langle I(t+\tau)I(t) \rangle}{\langle I(t+\tau) \rangle \langle I(t) \rangle}$$

- CW laser: $g^{(2)}(\tau) = 1$
- Thermal emission: $g^{(2)}(0) > 1$
- Ideal quantum two-level system: $g^{(2)}(0) = 0$

$g^{(2)}(\tau)$: Probability to detect a second photon $\tau$ seconds after detecting the first photon
A. Muller et al. (on Deppe’s VCSEL sample)

Flagg et al., Nat Phys 5, 203 (2009)
Lasing Characteristics

Lasing Signatures

- Pumping power dependence
- Concurrent thresholds of intensity kink & linewidth narrowing
- Mode competition with varying temperature

Lasing Characteristics

- CW lasing above LN$_2$ temperature
- Ultralow threshold ($\sim$2.1 kW/cm$^2$ at 8K; $\sim$3.7 kW/cm$^2$ at 78K)
- 3D mode volume well below diffraction limit ($\sim$0.03 $\lambda^3$)
- Green semiconductor laser (solving the “green-gap” issue)

Second-order photon correlation function measurement results at 8K.

When pumped just below the lasing threshold, $g^{(2)}(0)$ has a value larger than one, indicating a thermal emission.

In contrast, when pumped slightly above the lasing threshold, we can observe a temporal coherent emission with $g^{(2)}(\tau) = 1$. 

Yu-Jung Lu, Hung-Ying Chen, Wen-Hao Chang, Chih-Kang Shih & Shangjr Gwo
Simulations of Lasing Modes

Using COMSOL, we find:

- Far-field radiation patterns for two experimentally observed lasing modes
- In-plane directional, coherent surface plasmon polaritons (SPPs) are generated at the ends of the GaN nanorod

Chihhui Wu, Nima Dabidian & Gennady Shvets (UT-Austin)
Comparison with the Experimental Results

Dipole mode: \(510\ \text{nm}\)

Quadruple mode: \(522\ \text{nm}\)

\[\lambda = 510\ \text{nm}\]

Comparison with the experimental results:
Both of the measured polarization ratios are in agreement with the simulation results.

Chihhui Wu, Nima Dabidian, & Gennady Shvets

Theory also shows
\begin{itemize}
  \item Moderate Q of 11, and \(F_p\) of 1.8 (exp \(\rightarrow F_p \sim 2\)).
  \item Frequency pulling, etc (also observed experimentally)
  \item Atomic smoothness of the metal film is critical.
\end{itemize}

Polarization ratio becomes as large as 96% when pumped above the lasing threshold!

Yu-Jung Lu
Act II: All Color Plasmonic Nanolasers on A Single Platform
Recent Development: All-Color $\text{In}_x\text{Ga}_{1-x}\text{N}$ Nanorod Emitters

Rod dimension: $d = 30–70\,\text{nm}$
All-Color Plasmonic Nanolasers

Note: No need to tune the cavity length – Auto tuning of cavity
What’s next?  
(Perspectives and Speculations)

- Approaching RT CW operation of Plasmonic Nanolasers
- Toward Electrical Injection Plasmonic Nanolasers
- Ultra-long propagation of SPPs on epitaxial plasmonic platform
- Monolithic integration with Si nano-photonics/nanoelectronics
- Non-linear generation of Coherent Lights: On-chip OPO
- Photonic Topological Insulators
- Others

Applications for Ultrasmall mode volume of Plasmonic Cavities

<table>
<thead>
<tr>
<th>Property</th>
<th>Scaling</th>
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<tr>
<td>Weak coupling cavity-QED Purcell Enhancement</td>
<td>Q/V</td>
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<tr>
<td>Strong Coupling cavity-QED</td>
<td>Q/V^{1/2}</td>
</tr>
<tr>
<td>$\chi^{(3)}$ Nonlinearity (optical)</td>
<td>Q^2/V</td>
</tr>
<tr>
<td>Optical forces/trapping</td>
<td>Q/V</td>
</tr>
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</tr>
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