Strong Light-Matter Coupling and Polariton Lasing in Metallic and Dielectric Metasurfaces

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Motivation

Dipole:
Highly localized source of polarized electromagnetic radiation $\rightarrow$ Highly non-directional source $\delta r \delta p \geq \hbar / 2$

Full control on the emission characteristics of optical sources: Spectrum, efficiency, directionality and polarization

Couple emission to resonant structures $\Rightarrow$ Optical antennas
Antenna + emitter = structure with designed properties
Consider, for example, a piece of material in which we make little coils and condensers (or their solid state analogs) 1,000 or 10,000 angstroms in a circuit, one right next to the other, over a large area, with little antennas sticking out at the other end—a whole series of circuits. Is it possible, for example, to emit light from a whole set of antennas, like we emit radio waves from an organized set of antennas to beam the radio programs to Europe? The same thing would be to beam the light out in a definite direction with very high intensity. (Perhaps such a beam is not very useful technically or economically.)
Optical nanoantennas for SSL

Phosphor-based LEDs

$\lambda = 450 \text{ nm}$
$\lambda > 450 \text{ nm}$
Phosphor ($\eta \approx 1$)

Blue LEDs

Etendue reduction for long range optical communication

The large étendue in wireless optical systems limits a high-bandwidth and sensitivity detection

\[ A_s \Omega_s \leq A_i \Omega_i \]
Collective resonances: surface lattice resonances

Localized resonance
- Localized mode
- High local fields
- High losses: radiative and material (if plasmonic)

Collective resonances (non-local metasurface): Surface Lattice Resonances
- Hybrid modes
- High local fields, small mode volume
- Low losses

Utyushev, Zakomirnyi, Rasskazov, Rev. Phys. (2021)
Surface Lattice Resonances (SLRs)

Gold particles
$W = 85 \text{ nm}, L = 415 \text{ nm}$
$a_x = 500 \text{ nm}, a_y = 300 \text{ nm}$

Quartz
SLRs and in-plane diffraction

Grating equation: \[ \vec{k}_{\text{out}} = \vec{k}_{\text{in}} + m\vec{G}_x + n\vec{G}_y \]

\[ |G_x| = \frac{2\pi}{a} \quad |G_y| = \frac{2\pi}{b} \quad (m, n) \equiv \text{diffraction order} \]

\( a, b \equiv \text{lattice constants} \)

\( k_{\text{out}} = \frac{2\pi}{\lambda} (1,0) \quad (0,\pm1) \quad (-1,0) \)

Localized resonance

Grating equation:

\[ k_{\text{in}} = \frac{2\pi}{\lambda} \sin \theta_{\text{in}} \]

Gold nanorod arrays

S.R.K. Rodriguez et al., PRX, 1, 021019 (2011);
N. Meinzer et al., Nature Photon. 8, 889 (2014)
Surface Lattice Resonances (SLRs)

- **SERS and SLRs**: Carron et al. (JOSA B, 1986), Schatz et al. (J. Chem Phys, 2004)
- **SLRs and extinction**: Kravets (PRL 2008), Crozier et al. (APL 2008), Barnes et al. (PRL 2008)
- **SLRs and spontaneous emission**: Vecchi (PRL 2009), Giannini (PRL 2010), Rodriguez (PRL 2012)
- **SLRs and stimulated emission**: Schatz (Nat. NanoTech. 2013), Schokker (PRB 2014)
- **SLRs and strong coupling**: Rodriguez (Opt. Exp. 2013), Torma (Nano Letters 2014)
- **SLRs and polariton condensation**: Ramezani (Optica 2017)
- **SLRs and photon condensation**: Torma (Nature Physics 2018)
- **SLRs and sensing, non-linear optics, detectors, etc.**
Surface Lattice Resonances (SLRs)

Large array fabrication:
Surface Conformal Imprint Lithography

~1 nm linewidth Fano resonance in Au nanorod array

Lozano et al. Nanoscale (2014); A. Abass et al., ACS Photonics (2014).
Dielectric surface lattice resonances (Mie-SLRs)

Si arrays

Diameter = 120 nm
Height = 90 nm

Reduce or suppress material losses

Cavities for strong light-matter coupling and polariton condensation

Material: poly-c Si

Extinction = 1 - Transmission

Mie SLRs

Single particle Mie resonance

G. Castellanos...JGR, ACS Photonics 7, 5, 1226 (2020);
Heilmann...Törma, Nanophot. 9, 267 (2020); Todisco...Tserkezis, Nanophot. 9, 803 (2020)
Bose Einstein condensates (polariton lasing)

Ground-state accumulation of bosons at high \( n \) & low \( T \)

\[ T = T_c \quad T < T_c \]
\[ T > T_c \]

\[ T_c \propto \frac{1}{m} \]

rubidium-87, sodium-23

Large \( m \)
Low \( T_c \)

Small \( m \)
High \( T_c \)

Excitons – polaritons

Science 269, 198 (1995)
PRL 75, 3969 (1995)
2001
Cornell, Ketterle, Wieman

Imamoglu et al., PRA 53, 4250 (1996)
Exciton-Polaritons (Strong light-matter coupling)

Collective coupling strength:

\[ g = \frac{\hbar \Omega}{2} = \frac{\hbar c n c N}{\sqrt{\lambda \varepsilon \varepsilon_0 V}} \]

\( \mu_m \equiv \) Transition dipole moment

\( N \equiv \) Number of excitons

\( V \equiv \) Mode volume

Strong coupling when Rabi frequency is larger than the cavity loss rate and the exciton decoherence rate

\[ K_{p} \]

\[ \text{Energy (eV)} \]

\( \uparrow \)

\( \text{Rabi energy} \) (\( \hbar \Omega \))


Plumhof et al., Nat. Materials 13, 247 (2014)
Exciton-Polaritons (Strong light-matter coupling)

Bare array

1 layer - TDBC

3 layers - TDBC

6 layers - TDBC
Exciton-Polaritons (Strong light-matter coupling)

- Rylene dye

- Plasmon-exciton-polariton condensation: Plasmonic (Ag) metasurface
- Mie-exciton-polariton condensation: Dielectric (pc-Si) metasurface
- BIC-exciton-polariton condensate
Plasmon-Exciton-Polariton condensation

Ag nanoparticles

Dipole

Multipole

No molecules (Bare modes)

35 wt% Molecules

Polaritons with strong radiative decay (bright)

Polaritons with reduced radiative decay (dark)


Plasmon-Exciton-Polariton condensation

\[ P_{\text{pump}} > P_{\text{th}} \]

\[ P_{\text{pump}} = P_{\text{th}} \]

\[ P_{\text{pump}} < P_{\text{th}} \]
Plasmon-Exciton-Polariton condensation: polarization

M. Ramezani, et. al., Optica, 4(1), 2017
Plasmon-Exciton-Polariton condensation: threshold

Emission quantum efficiency of dye in PMMA

Stimulated scattering of polaritons responsible for condensation in contrast to stimulated emission responsible for lasing

M. Ramezani, et. al., Optica, 4(1), 2017
Plasmon-Exciton-Polariton lasing

Transfer mechanism $\Rightarrow$ Vibronic assisted relaxation

Mazza et al., PRB 88, 2013
Nomaschi et al., APL 99, 2011

M. Ramezani, et. al., Optica, 4(1), 2017
Mie-Exciton-Polariton condensation

Si-nanoparticles

$x$-polarization

$k_{||} = k_x$

Mie-Exciton-Polariton condensation

\[
\frac{dn_R}{dt} = \left(1 - \frac{n_R}{N_0}\right) P(t) - \frac{n_R}{\tau_R} - \frac{n_R^2}{\tau_b} - W_{ep} n_R n_{LP}
\]

\[
\frac{dn_{LP}}{dt} = W_{ep} n_R n_{LP} + \beta \frac{n_R}{\tau_R} - \frac{n_{LP}}{\tau_{LP}}
\]

- Radiative relaxation
- Vibronic assisted relaxation

Mie-Exciton-Polariton condensation

For short periods radiative relaxation is less important
For long periods vibronic assisted relaxation is less important
Lowest threshold for intermediate periods

\[
\frac{dn_{LP}}{dt} = W_{ep} n_R n_{LP} + \beta \frac{n_R}{\tau_R} - \frac{n_{LP}}{\tau_{LP}}
\]

\(\tau_{LP}\) is limited by the cavity lifetime

How good can be a cavity?

- Radiation losses
- Material losses

If both losses are suppressed

Lifetime: \( \tau_{cav} = \infty \)

Linewidth: \( \Delta \omega = 0 \)

Q-factor: \( Q = \frac{\omega}{\Delta \omega} = \infty \)
Bound states in the continuum (BICs)

Hsu, Zhen, Stone, Joannopoulos and Soljacic, Nature Reviews (2016)
BICs in arrays of Si nanoparticles

\[ k_\parallel = k_x \]

Mohamed, Laser Photonics Rev. 16, 2100574 (2022)
BICs in arrays of Si nanoparticles

BICs arise due to a symmetry mismatch between free space radiation and the modes in the metasurface.

Mohamed, Laser Photonics Rev. 16, 2100574 (2022)
BIC- Exciton-Polaritons

Si and no dye

Si + dye (32 %wt)

$E_{\text{energy}} (eV)$

$E_{\text{energy}} (eV)$

$k_{\text{x}} (\text{rad/micrometer})$

$k_{\text{x}} (\text{rad/micrometer})$

$\text{Wavelength (nm)}$

$\text{Quality factor}$

$\text{Angle of incidence (°)}$

M. Berghuis et al., Nano Lett. 23, 5603 (2023)
BIC- Exciton-Polariton condensation

Low threshold of 5 µJ cm$^{-2}$

M. Berghuis et al., Nano Lett. 23, 5603 (2023)
The vortex emission from the condensate shows the quadrupolar character of the mode.

M. Berghuis et al., Nano Lett. 23, 5603 (2023)
BIC- Exciton-Polariton condensate: coherence time

\[
\frac{dn_{LP}}{dt} = W_{ep} n_R n_{LP} + \beta \frac{n_R}{\tau_R} - \frac{n_{LP}}{\tau_{LP}}
\]

\(Q = 2\pi \tau n_0 \sim 15000\)
Conclusion

Nanoparticle arrays (metallic & dielectric) supporting surface lattice resonances offer a versatile platform for several applications: SSL, optical communication, exciton-polariton condensation.

\[ P_{\text{pump}} > P_{\text{th}} \]