Title : Hybrid resonance for sensing applications

1. Overview

The deployment of nanotechnologies in the last decade has led to a proliferation of a totally new generation of micro/nano-resonators, which enables the control of light-matter interaction on nanometer scales for the first time. With an unprecedented creativity, resonator constructs with distinct shapes, compositions and dimension scales are fabricated nowadays and assembled together by combining bottom-up and top-down technologies, see Fig. 1. They play an essential role in current developments in nanophotonics, such as optical metamaterials, integrated photonic circuits, optical sensing, and light tweezers, thereby finding use in many areas of science and technologies, from biology, microfluidic to specific applications such as secure printing documents or DNA nanotechnology. **This PhD project has the potential of resulting in a paradigm change in micro/nano-resonators by enabling a totally new generation of micro/nano-resonators by hybridizing existing technologies.**

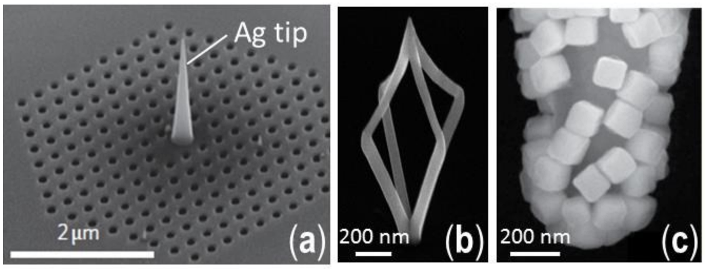

Fig. 1 Some electromagnetic resonators illustrating the diversity of micro-nano-resonator systems. (**a**) Hybrid plasmonic-photonic resonator formed with a silver tapered waveguide on a silicon photonic-crystal micro-cavity [Ang10]. (**b**) plasmonic crown fabricated by real 3D nanoprinting [ACS Nano **10** (2016)]. (**c**) Assembly of Ag-nanocube deposited onto the surface of a commercially available AFM tip [ACS Nano **10** (2016)].

The domain of optical resonance is roughly composed of two completely opposed geometries, plasmonic nano-resonators with $Q\sim 20, V\sim 10^{-4}\lambda^3$, and photonic-crystal cavities with $Q\sim 10^6, V\sim\lambda^3$, which leads research in either of two conflicting directions: either highest Q, or smallest V. So far most researchers aimed for the highest-Q resonators in lossless 'dielectric' optical materials. As confinement in dielectrics needs wave interference, the diffraction limit constrains the mode volume to be at least a cubic wavelength. The opposite strategy is to minimize *V*. A recently developed strategy to shrink photons to 3 orders of magnitude below the diffraction limit is `plasmonics': capturing light in a resonance of free electrons in metal.

Unfortunately, both strategies lead to huge obstacles. The paradigm of ultrahigh Q is very impractical because ultrahigh-Q means ultranarrow bandwidth. It is a huge challenge to exactly tune the cavity resonance to be precisely matched a desired wavelength, since one suffers extreme sensitivity to fluctuations [Arm08]. Further, since the typical response time of any system is limited by the inverse of its Q, ultra-high Qs mean very slow timescales. Conversely, the opposite paradigm of ultimate confinement by plasmonics means detrimental dissipative loss and plasmonic Qs are largely limited to a few tens.

The objective of the thesis is to develop a new nanophotonic strategy "hybrid cavities" that allows any practical Q ($50 < Q < 1000$) while preserving ultra-small V's. Practically, this is reached by hybridizing photonic-crystal cavities with plasmonic antennas. This platform is expected to unite the merits and removes the drawbacks that microcavities and plasmonics separately present. Although hybrid cavities have a huge potential for innovation by considerably enlarging the Q,V parameter space, their study and design is still in their infancy. The topic was initiated in 2008, see Fig. 1a and [Ang08], and then remained dormant during a few years. Several groups are very active since a couple of years [Rue16,Mos16,Con16,Dez16], but the essential question on how to best take benefit from the two extreme characteristics offered by the association remains open. Still nowadays, designs are essentially driven by intuition, extensive brute-force computations that cannot explore the parameter space or approximate and crude models, dipole-dipole models basically, that prevents real breakthrough.

The thesis will propose a symbiotic development of theoretical/numerical tools, approximate models and analytical formulas to establish intuitive clues for initial design of hybrids, to evaluate their ultimate limits in terms of Q's and V's and optimize their performance for resonance optical biosensing.

To push forward the state-of-the-art in micro-nanocavities, the thesis will pursue a two-track strategy:

(1) We will elaborate new theoretical/numerical tools to model resonator hybrids. We expect that the new modal tool will provide considerably much smaller computation times and convey a much clearer physical understanding, than classical approaches.

(2) The core of hybrid-cavity engineering consists in perturbing and hybridizing resonances of different natures, e.g. photonic high-Q, plasmonic dark and bright modes. We will elaborate semi-analytical formulas based on the modal tools developed in (1) to design optimal hybrids for sensing minute changes in the refractive-index around the hybrid.

2. Scientific and technical program

2.1 Development of numerical tools based on QNM expansion

In the vast majority of cases, the resonant interaction of light with resonators is described via continuum (scattering) theory with classical Maxwell's equation solvers, operating either in the frequency-domain with real frequencies, or in the time domain with the FDTD method almost hegemonically used by experimentalist groups.

The problem with existing scattering theory for designing resonators is that the core physical concepts attached to the resonator, the resonance modes, are only indirectly accessed. This has deplorable impacts: (**1**) *Modeling/design tools are ineffective*: in frequency-domain, the *entire* computation should be repeated for each individual frequency, and in time-domain, the *entire* computation should be repeated if one varies the excitation field, e.g. the pulse shape, polarization, incidence angle. (**2**) *Some key complex geometries of contemporary optics*, such as hybrids (Fig. 1a) or multi-resonator systems (Fig. 1c) cannot be modeled practically. (**3**) Interpretation is not straight forward and the high "simplicity" of the physics at hand is often hidden. To better realize the problem, one may imagine what would be integrated optics if optical waveguide theory and concepts were not available nowadays.

We envision a completely different approach, which explicitly relies on the natural resonator modes, also called the quasi-normal modes (QNMs). The latter are the poles of the Green function, and are intrinsic quantities, independent of the excitation field, with complex frequencies. More specifically, we will model the optical response of resonators by expanding the electromagnetic field $\mathbf{E}(\boldsymbol{r},\omega,\check{e})$ scattered by the resonator for an incident excitation $\check{e}(\boldsymbol{r},\omega)$ at (real) frequency ω in the QNM basis

$$\mathbf{E}(\boldsymbol{r},\omega)=\textstyle\sum_m \alpha_m(\omega,\check{e})\,\tilde{\mathbf{E}}_m(\boldsymbol{r}). \tag{1}$$

In Eq. (1), $\tilde{\mathbf{E}}_m(\boldsymbol{r})$ denotes the electric-field map of the m^{th} QNM with quality factor $Q_m=-\frac{1}{2}Re(\widetilde{\omega}_m)/Im(\widetilde{\omega}_m)$ and complex frequency $\widetilde{\omega}_m$; the α_m's are the modal excitation coefficients.

The potential offered by QNM expansions is clear:

- Once the QNM fields are calculated, the optical response (the α_m's) is known analytically as simple spatial-overlap integrals between the excitation field $\check{e}$ and $\widetilde{\mathrm{E}}_m$. Since additionally the frequency-dependence is known analytically, considerable computational-speed improvements, compared to classical Maxwell's equations solvers, are anticipated [Fag16].

- Modal expansions provide key clues towards understanding the physics underlying the optical response of resonators. The required physics is readily available and unambiguous since the resonance modes of the system are explicitly considered, in sharp contrast with classical scattering theories.

QNM solver. First we will develop ground-breaking solvers to compute the QNMs with a high accuracy for the most general case of 3D materials with frequency dispersion. In this case, because the permittivity depends on the frequency, a nonlinear eigenvalue problem has to be solved. We will combine the auxiliary field method [Ram10] with the finite element method to model, for the first time, nano-resonators with complex shapes with a high precision. We will implement and test the formulation on an in-house FEM platform developed at INRIA.

Coupled-resonator theory. An hybrid is nothing else than the association of two resonances. Then in a second step, we will develop a self-consistent coupled-mode theory of non-Hermitian nano-resonators formed by the association of two resonators. Our goal will be to derive semi-analytic expressions for the new complex frequencies and QNM of the coupled system from the sole knowledge of the individual QNMs. Importantly, the expressions will have an analytic dependence on the relative orientation and positioning of the two resonators. The analyticity will be crucial for optimizing the performance of biosensors. We will test the theory by comparing the analytical predictions with exact computations performed with the QNM solver. Preliminary results obtained for the resonant frequency of a plasmonic doublet are shown in Fig. 2.

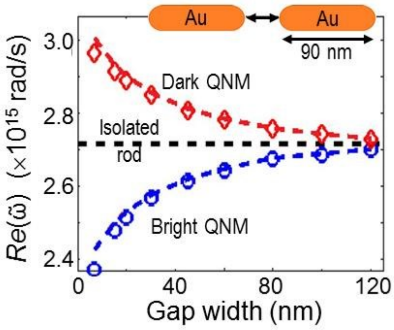

Fig. 2 Preliminary results for the resonant frequency obtained by Kévin Cognée (cotutelle PhD with Dr. F. Koenderink, AMOLF) in the PI lab for the coupling of two identical gold nanorods. Circles are obtained with fully-vectorial computations performed for every gap width. Dashed curves are obtained from the sole knowledge of the electric-dipole QNM of each individual nanorod.

2.2 Hybrid resonances for biosensing

The ability of a sensor to measure minute changes in the refractive-index is directly proportional to bulk refractive-index sensitivity $S_B = \frac{d\lambda_R}{dn_B}$ (λ_R is the resonant wavelength and n_B is the refractive index of the medium in the contact with the sensor surface) and, furthermore, inversely proportional to the width w of the resonant feature (spectral dip or peak in Fig. 2b) being tracked. The combination of these parameters is often referred to as the figure of merit $\mathrm{FOM}_B = S_B/w$ [Špa16], which is widely used, as it allows for the evaluation and comparison of different nanostructures with respect to their sensing potential. Since the width w is inversely proportional to Q and the resonance shift $d\lambda_R$ is proportional to $\Delta n_B \frac{v}{V}$ (v being the tiny volume of the minute analyte to be detected) [Yan15], the FOM_B scales as

$$\mathrm{FOM}_B \propto Q/V. \qquad (2)$$

We immediately understand the importance of creating new resonances with optimized Q/V ratios for sensing, away from the classical strategies that lead research in either of two conflicting directions: either highest Q, or smallest V. The paradigm of ultrahigh Q is very impractical because ultrahigh-Q means ultranarrow bandwidth $\Delta\lambda = \lambda/Q$ [Hom08]. Conversely, the opposite paradigm of ultimate confinements by plasmonics means spectral bandwidths that are so large that plasmon-resonance shifts in and out of the resonance bandwidth $\Delta\lambda = \lambda/Q$ is unthinkable.

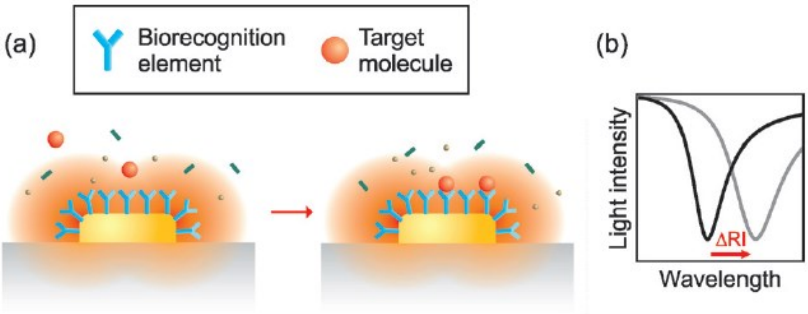

Fig. 3 (a) Principle of a plasmonic affinity biosensor. (b) Change in the spectrum due to the increase of refractive-index in the proximity of a sensor surface induced by captured target molecules. SPR biosensors have been demonstrated to hold vast potential for rapid label-free detection of chemical and biological species in numerous fields, including medical diagnostics, environmental monitoring, and food safety and security [Hom08].

Thanks to the QNM numerical tools developed, we will derive a comprehensive theory that analytically discloses basic rules to design hybrids with enhanced Q/V's, typically targeting intermediate Q's ($50 < Q < 1000$) and V's ($0.01 < V/\lambda^3 < 0.1$). In addition, by combining a recent work of the LP2N group on cavity perturbation theory [Yan15] with the coupled-resonator theory, we will derive analytical formulas for the scattering and absorption cross-section spectra of hybrids perturbed by the presence of an analyte from the sole knowledge of the unperturbed resonance. The capability to analytically predict the scattering cross-section spectra, is likely to be a decisive step to push the state-of-the-art of hybrids, and thanks to the efficient QNM tools developed in the first part of the thesis, we will comprehensively optimize a totally new family of micro/nano-resonators offering sensing capabilities above the state of the art.

Management

The PhD thesis will take place within the framework of a collaboration between the COS group headed by Philippe Lalanne at Laboratoire Photonique Numérique et Nanosciences (CNRS, IOA) in Bordeaux UMR 5298 and the Magique 3D group headed by Hélène Barucq at INRIA in Bordeaux and Pau. COS is composed of physicists specialized in computational electrodynamics and nanophotonics. Magique 3D is composed of applied mathematicians working in scientific computing.

The PhD student will primarily work at Bordeaux, sharing his/her time between INRIA and LP2N (the labs are 300 m distant).

Co-advisors: **Hélène Barucq** (INRIA, Juliette Chabassier (Chercheur INRIA), Marc Durufle (MC-Bordeaux)) and **Philippe Lalanne** (LP2N-Bordeaux, Kevin Vynck (CR))

https://www.lp2n.institutoptique.fr/Membres-Services/Responsables-d-equipe/LALANNE-Philippe

https://team.inria.fr/magique3d/fr/team-members/helene-barucq/publications/

References

[Ang10] F. De Angelis, G. Das, P. Candeloro, M. Patrini, M. Galli, A. Bek, M. Lazzarino′, I. Maksymov, C. Liberale, L.C. Andreani and E. Di Fabrizio, Nat. Nanotech. **5**, 67 (2010).
"Nanoscale chemical mapping using three-dimensional adiabatic compression of surface plasmon polaritons"

[Arm08] A.M. Armani et al., Science **317**, 783 (2007).
"Label-Free, Single-Molecule Detection with Optical Microcavities"

[Ang08] F. De Angelis et al., Nano Lett. 8, 2321–2327 (2008).
"A Hybrid Plasmonic-Photonic Nanodevice for Label-Free Detection of a Few Molecules"

[Rue16] R. Ruesink, H.M. Doeleman, R. Hendrikx, A.F. Koenderink and E. Verhagen, Phys. Rev. Lett. **115**, 203904 (2016).
"Perturbing open cavities: Anomalous resonance frequency shifts in hybrid cavity-nanoantenna systems"

[Mos16] M Mossayebi, A. Parini, A.J. Wright, M.G. Somekh, G. Bellanca and E.C. Larkins, Opt. Express (in press).
"A realizable hybrid photonic-plasmonic nanocavity with high quality factor and confinement of light"

[Con16] D. Conteduca, F. Dell'Olio, T.F. Krauss, C. Ciminelli, PECS XII proceedings.
"Photonic-plasmonic nanotweezer for single particle optical trapping"

[Dez16] M. K. Dezfouli, R. Gordon, and S. Hughes, arXiv:1606.05874v1 (2016).
"Modal theory of modified spontaneous emission for a hybrid plasmonic photonic-crystal cavity system"

[Fag16] R. Faggiani, A. Losquin, J. Yang, E. Mårsell, Anders Mikkelsen, Philippe Lalanne, ACS Photonics (submitted).
"Modal analysis of the ultrafast dynamics of optical nanoresonators"

[Ram10] A. Raman and S. Fan, Phys. Rev. Lett. **104**, 087401 (2010).
"Photonic Band Structure of Dispersive Metamaterials Formulated as a Hermitian Eigenvalue Problem"
Milestone report introducing the auxiliary-field method to simply calculate band diagrams of dispersive periodic media.

[Špa16] B. Špacková, P. Wrobel, M. Bocková, J. Homola, Proc. of the IEEE **104**, 2380-2408 (2016).
"Optical Biosensors Based on Plasmonic Nanostructures: A Review"

[Yan15] J. Yang, H. Giessen, P. Lalanne, Nano Lett. **15**, 3439 (2015).
"Simple Analytical Expression for the Peak-Frequency Shifts of Plasmonic Resonances for Sensing"
On an enduring classic of electromagnetism (perturbation theory of a resonance), the letters makes a significant step forward, by proposing the first consistent perturbation theory of metallic (highly non-hermitian) resonance with important implication for sensing, and also by removing a longstanding error in the standard perturbation theory of photonic resonator.

[Hom08] J. Homola, Chem. Rev. **108**, 462–493 (2008).
"Surface plasmon resonance sensors for detection of chemical and biological species"