Third year PhD report

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Supervisor: Andrea Toma
Academic year: 2024-2025, XXXVIII cycle

Research activity:

I devoted the third year of my PhD activities to the study of transient absorption dynamics in metal and metal oxides nanostructures, combined with quantum emitting materials, such as quantum dots and organic dyes, to further explore the dynamics of plasmon-exciton interaction under strong coupling (SC) regime.

In collaboration with team members, I spent significant efforts in measuring absorption dynamics of aluminum nanodisk arrays sustaining surface lattice resonances (SLRs), i.e. a class of high-quality factor collective modes allowing for more effective SC experiments.

Within this context, J-aggregate molecules with excitonic features at 585 nm have been spin-coated on aluminum nanodisk arrays with periodicities ranging from 315 to 395 nm. In this way, the SLR wavelength is detuned from the exciton resonance position to map the system dispersion. Additionally, the array geometry was varied between three different arrangements, square, hexagonal and honeycomb lattices, for further investigation. The measurements have been performed at low pump fluences (25-100 $\mu J/cm^2$) to reduce J-aggregate damage, showing the formation of upper and lower polariton branches, the fingerprint of strong light-matter interaction, and the photoinduced absorption dynamics as already found for the nanoparticle-on-mirror geometry systems (see second year report – collaboration with Aalborg University, Denmark).

Negative transient signals with less than 1 ps dynamics were observed as well as excited state absorption signals, using a pump wavelength at 605nm in resonance with the J-aggregate excitonic reservoir. Fast dynamics has been also observed pumping at a wavelength of 525nm [1]. As expected, lower polariton state presents a faster decay dynamics with respect to the upper polariton case [2].

Finally, it is worth noticing that the impinging angle of the pump-probe beams play a key role in the polariton dispersion curve and lifetime due to “phase matching” conditions [3]. For these reasons we are optimizing, via computational methods, the array arrangements to compensate for non-zero incidence angles and extending the study to geometry-angle influence on the strong coupling dynamics.

Other activities and collaborations:

In June 2025 I went to the Friedrich-Alexander-Universitat (FAU) in Erlangen, Germany, visiting the laboratories of the research group Quantum materials for quantum spectroscopy (Q4Q) headed by Prof. Daniele Fausti and participating to the Physics Colloquium held by my supervisor Andrea Toma.

Furthermore, I had the opportunity for a longer stay (almost 2 months) in Erlangen to gain confidence with ultrafast spectroscopy systems operating under different configurations and to collaborate on time-resolved experiments on plasmonic structures.

Exams:

- Electronics and data acquisition (exam to be given by the end of September 2025)
- Introduction to the foundations of quantum mechanics and applications (exam to be given by the end of September 2025)
- Optical properties of materials (exam given)

Acknowledgement:

This project received support by the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation program “REPLY ERC-2020-COG Grant agreement No. 101002422”.

References

1) Eizner E., Akulov K., Schwartz T., Ellenbogen T. Temporal Dynamics of Localized Exciton-Polaritons in Composite Organic-Plasmonic Metasurfaces, Nano Lett. 2017, 17, 7675-7683

2) Schwartz T., Hutchison J.A., Léonard J., Genet C., Haacke S., Ebbesen T. W. Polariton Dynamics under Strong Light-Molecule Coupling, ChemPhysChem 2013, 14, 125-131

3) Michail E., Rashidi K., Liu B., He G., Menon V.M., Sfeir M.Y. Addressing the Dark State Problem in Strongly Coupled Organic Exciton-Polariton Systems, Nano Lett. 2024, 24, 557-565