THESIS DEFENSE: Nanocavities and Molecules: Polaritons and their Dynamic Interactions

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11:00

ICFO Auditorium

This thesis investigates light-matter interactions within plasmonic nanocavities, using ultraconfined optical environments to observe and control molecular dynamics at the nanoscale. Plasmonic cavities, formed by metallic nanoparticles in close proximity to metal films, generate highly localized optical fields within nanometer gaps. Such intense confinement allows molecules in these gaps to interact strongly with light at room temperature, producing hybrid states that blend characteristics of both photons and molecules- a phenomenon known as strong coupling. By focusing light into such small volumes, these cavities enable a platform to probe both strong and weak coupling regimes. Various experimental setups were developed to explore these interactions, enabling simultaneous Rayleigh and Raman scattering measurements from individual nanocavities and

introducing new methods for tracking polariton dynamics with femtosecond pulses. Under strong coupling conditions, plasmonic nanoparticles on mirror cavities paired with Methylene Blue molecules demonstrated hybrid light-matter states. Through extensive quantitative measurements, this study identified key parameters that drive strong coupling and distinguished it from the mechanisms underlying Surface-Enhanced Raman Scattering, offering a new understanding of light-molecule interactions. The study also delves into dynamic phenomena like spectral diffusion, observed in host-guest molecular systems within plasmonic nanocavities, revealing time-dependent spectral shifts that shed light on molecular behavior in confined fields. Control over these nanoscale interactions was achieved by tuning the cavity resonance through refractive index means and femtosecond optical pulses, enabling targeted shifts in the plasmonic resonance. Additionally, using femtosecond pulses allowed the investigatio of decay dynamics in strongly coupled systems, advancing optical control methods for polaritons and capturing previously unobserved spectral evolutions in hybrid light-matter states. Finally, a broadband white-light interferometric technique was employed to directly measure the spectral phase of polaritons. This revealed phase shifts across hybrid light-matter states and added to understanding spectral phase dynamics in strongly coupled plasmonic systems.

This thesis contributes a holistic view of light-matter interactions at the nanoscale by bridging the themes of understanding and control. It sets the stage for future advancements in plasmonic nanocavities and their potential applications in chemical sensing and nanoscale spectroscopy.

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Thesis Director: Prof. Dr. Niek van Hulst

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