Dr. Niclas S. Mueller
Freie Universität Berlin, Physics
(currently: Nanophotonics Centre, University of Cambridge)

3D Plasmonic Supercrystals: Optical Properties and Enhanced Vibrational Spectroscopy

Date: July 5 2021, 15.00 h Hamburg time

Abstract: Plasmonic supercrystals are highly-ordered three-dimensional assemblies of metallic nanoparticles. Light inside these supercrystals gets tightly confined into regularly arranged hotspots for the electric field, because of plasmon resonances of the nanoparticle building blocks. This leads to unique optical properties that enable applications in analytical chemistry, bioimaging, optical coatings and waveguiding. To exploit the full potential of plasmonic supercrystals, we need a better understanding of the optical properties and enhancement mechanisms.

In the first part of my talk, I will show that the optical response of plasmonic supercrystals is determined by 3D plasmon polaritons [1]. These excitations arise from a hybridization of collective plasmonic states with photons. The coupling strength is so large that exotic phenomena of ultra- and deep strong light-matter coupling can be explored [1]. The plasmon polaritons form standing waves inside the crystal (Fig. 1a), which leads to optical resonances across a broad spectral range. To explore these phenomena experimentally, we synthesize fcc crystals of gold nanoparticles via the self-assembly at a liquid subphase and characterize them optically (Fig. 1b, c) [1, 2]. I will show how we extract the polaritonic band structure and demonstrate the decoupling of light and matter in the deep strong coupling regime.

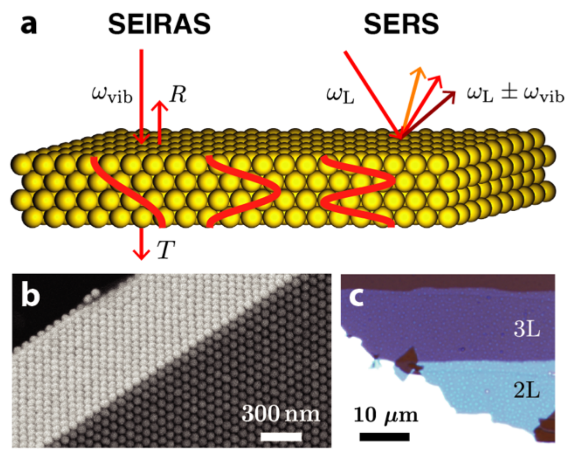

Fig. 1: 3D plasmonic supercrystals. (**a**) Sketch of SERS and SEIRAS with plasmon polaritons. (**b**) SEM and (**c**) optical microscopy image.

In the second part of my talk, I will show how polaritons enhance the vibrational spectra of molecules inside the supercrystals [3]. The polaritons induce near field enhancement across the entire crystal volume, which we probe through the vibrations of polystyrene ligand molecules. Using surface-enhanced Raman spectroscopy (SERS) with tunable excitation wavelength, we measure pronounced polariton resonances in the near infrared that correlate with the transmission and reflection spectra of the supercrystals. The resonances of multilayered crystals extend into the mid-infrared enabling surface-enhanced infrared absorption spectroscopy (SEIRAS). Plasmonic supercrystals therefore offer the possibility to combine SERS and SEIRAS on a single substrate.

References

[1] N.S. Mueller, Y. Okamura, B.G.M. Vieira, S. Juergensen, H. Lange, E.B. Barros, F. Schulz, S. Reich, Deep strong light-matter coupling in plasmonic nanoparticle crystals, *Nature* ***2020***, 583, 780-784.
[2] F. Schulz, O. Pavelka, F. Lehmkühler, F. Westermeier, Y. Okamura, N.S. Mueller, S. Reich, H. Lange Structural order in plasmonic superlattices. *Nat. Commun.* **2020,** 11, 3821.
[3] N.S. Mueller, E. Pfitzner, Y. Okamura, G. Gordeev, P. Kusch, H. Lange, J. Heberle, F. Schulz, S. Reich, Surface-Enhanced Raman Scattering and Surface-Enhanced Infrared Absorption by Plasmon Polaritons in Three-Dimensional Nanoparticle Supercrystals, *ACS Nano*, **2021**, 15, 5523–5533.