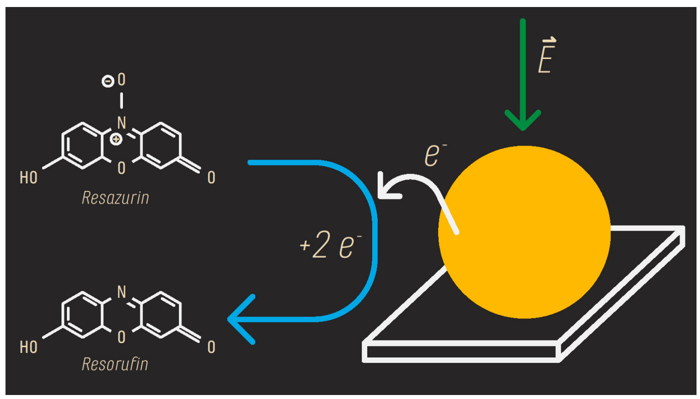
Master's Project
"Super-resolution mapping of plasmonic hot electrons generated in Au and Au@SiO_2 nanorods"

Metal nanoparticles are used in a wide range of applications, from catalysis, to advanced solar cells, photo-thermal cancer therapy, and medical imaging. One of the most striking features of metal nanoparticles is their unique interaction with light, leading to strong absorption and scattering in the UV, visible, and IR ranges[1]. These resonances, commonly called plasmon resonances, are responsible for a variety of interesting effects, from the activation of sub-wavelength electromagnetic field hot spots[1], to the generation of non-equilibrium charge carriers[2] and high temperature gradients at the surface of the nanostructures[3].

This Master's project aims at exploiting plasmon resonances to enhance the catalytic activity of Au nanoparticles and to discriminate between the contributions of non-equilibrium charge carriers, plasmonic heating, and photonic coupling between catalyst and reaction product.



The catalytic reaction that will be studied is the reduction of resazurin (non-fluorescent oxidized molecule) to resorufin (fluorescent reduced molecule)[4]. While this reaction occurs naturally in the presence of a reducing agent (NH_2OH) and a metal nanoparticle catalyst, our working hypothesis is that the generation of hot electrons on the metal nanoparticle catalysts will accelerate the catalytic conversion. Since the reaction product is fluorescent, its formation can be detected in an optical fluorescence microscope. By using sufficiently low reaction rates, each light burst observed on the camera of the microscope corresponds to the emission of one individual reaction product, allowing its position to be determined with sub-diffraction resolution by fitting to a 2D Gaussian[5].

The Au catalysts will be synthesized colloidally[6] and their shape will be tuned such that the plasmon resonance is spectrally separated from the absorption and emission of the reaction products in order to avoid mislocalization effects[7]. The catalyst will then be coated with a mesoporous SiO_2 shell[8-12] and the catalytic behavior of the coated and uncoated particles will be investigated using the abovementioned super-resolution technique. Clarifying the contribution and potential necessity of the shell will be one of the milestones of this project, as it has been proposed that temporary trapping of reaction products in the shell greatly enhances their detection probability[8-12], even though detection on uncoated catalysts has also been reported[4,13,14].

After synthesis of the catalysts, the influence of plasmonics on the catalytic reaction will be studied. By applying controlled illumination with two different light sources, one to excite the plasmon resonance and one to excite the reaction products, the underlying mechanism of plasmon-enhanced catalysis will be investigated.

References

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