Abstract

Antenna-reactor complexes contain optical antennas, which are able to focus light into nanoscale volumes, and transition metal nanoparticles, which interact weakly with light, but exhibit high catalytic activity. Electromagnetic coupling in this system leads to enhanced optical absorption in transition metal nanoparticles, which facilitate prospect applications such as optical monitoring of phenomena accompanying catalysis and plasmon mediated photocatalysis.

In this thesis we provide a comprehensive theoretical study of electromagnetic coupling in antenna-reactor complexes occuring at three length scales present in this system: at the macroscopic level, the single antenna level and at the atomic scale. At the macroscopic scale the optical response of antennas is determined by coupling between them via multiple scattering. In experimental samples nanoantennas are distributed randomly on a dielectric substrate. Thus, we propose a T-matrix method based approach to describe the effective optical properties of such a layer. We show that in addition to antenna properties their minimal center-to-center distance is a key parameter determining electromagnetic coupling. The proposed approach enables formulating simple analytical expressions that may be used to optimize future devices based on amorphous arrays of nanoantennas.

At the single antenna level, we provide accurate models of both the nanoantenna and transition metal nanoparticle layer either by explicit modelling or using a gradient effective medium model developed by the author. This enables us to study absorption enhancement in transition metal nanoparticles and find useful guidelines for maximizing this effect in experimental samples. Also, the results tie changes of the properties of either the nanoantenna or transition metal nanoparticles to changes of the optical response of the system, which might be used to optically monitor the evolution of the system during catalysis.

Realistic modelling of phenomena occuring at atomic scales requires us to use time-dependent density functional theory. At this scale it is possible to credibly model plasmon formation and its subsequent dephasing that results in hot electron generation. We show that the presence of a nanoantenna leads to enhanced hot electron generation in the system compared to an isolated transition metal nanocluster and study the system parameters that determine this enhancement.

The study shows that antenna-reactor complexes form a promising platform for photonic enhancement of interaction of light with transition metal nanoparticles, which might lead to intersting applications in sensing and photocatalysis.